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Phys. Rev. A **98**, 043609 — Published 5 October 2018

DOI: 10.1103/PhysRevA.98.043609

Effective spin-spin interactions in bilayers of Rydberg atoms and polar molecules

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(Dated: September 17, 2018)

We show that indirect spin-spin interactions between effective spin-1/2 systems can be realized in two parallel 1D optical lattices loaded with polar molecules and/or Rydberg atoms. The effective spin can be encoded into low-energy rotational states of polar molecules or long-lived states of Rydberg atoms, tightly trapped in a deep optical lattice. The spin-spin interactions can be mediated by Rydberg atoms, placed in a parallel shallow optical lattice, interacting with the effective spins by charge-dipole (for polar molecules) or dipole-dipole (for Rydberg atoms) interaction. Indirect XX, Ising and XXZ interactions with interaction coefficients $J^{\perp}$ and J^{zz} sign varying with interspin distance can be realized, in particular, the $J_1 - J_2$ XXZ model with frustrated ferro-(antiferro-)magnetic nearest (next-nearest) neighbor interactions.

I. INTRODUCTION

Polar molecules and Rydberg atoms interact via strong, anisotropic and long-range dipole-dipole intraspecies and charge-dipole interspecies interactions. Both systems have long-lived internal states, which can encode qubits and effective spins, such as low-energy rotational states of the ground electronic and vibrational state of molecules and long-lived high-n states of Rydberg atoms. The long-lived qubit/effective spin states and strong long-range interactions make for highly attractive quantum computation and quantum simulation platforms [1, 2]. In periodic trap arrays, these systems offer the additional advantage of scalability to qubit numbers sufficient for large-scale simulations [3, 4].

Quantum magnetism is particularly amenable to simulations with ultracold atomic and molecular systems because various types of magnetism models can be modelled due to exquisite control over atomic interactions. In particular, polar molecules can efficiently simulate various quantum magnetism models [5], e.g. effective XX spin-exchange has been realized in a 3D lattice of KRb molecules [6]. Rydberg atoms have also been proposed for quantum simulation of magnetism phenomena [7], starting with the seminal work on realization of an Ising model with Rydberg crystals [8], recently demonstrated in [9], and extending to simulation of exotic frustrated magnetic states such as quantum spin-ice [10].

One particularly interesting class of magnetic interactions are indirect, *i.e. mediated*, spin-spin interactions. Examples include superexchange [11, 12], electron-spin mediated interaction between nuclear spins in molecules (J-coupling) [13], and Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between localized magnetic impurities in metals and semiconductors, mediated by coupling to conduction electron spins [14–17]. The RKKY interaction is of special interest in that it has a sign periodically varying with the distance between the impurities, which can lead to frustration and random magnetization, producing non-trivial magnetic phases such as spin glass [18]. Frustrated magnetic systems with sign-changing interactions, in particular, with competing ferromagnetic nearest and antiferromagnetic next nearest neighbor interactions, such as copper oxide spin chains [19–23], have attracted active interest in recent years [24–26] due to unusual magnetic properties of the corresponding materials stemming from large degeneracies of their ground states induced by frustration.

In the present work we consider indirect interaction in a setup comprised of effective spin-1/2 systems, encoded in either rotational molecular or atomic Rydberg states, mediated by their respective interactions with auxiliary spin-1/2 systems, encoded in Rydberg atom states. The effective and mediator spins can be trapped in two 1D parallel optical lattices or trap arrays such that the effective spins are tightly trapped in their sites, while the mediator spins are loosely trapped and because of spatial delocalization of their motional wavefunction can simultaneously interact with several effective spins.

Drawing an analogy with the RKKY interaction the tightly trapped polar molecules/Rydberg atoms act as localized magnetic impurities, and the weakly trapped mediator Rydberg atoms play the role of conduction electrons. We show that this indirect interaction can change sign depending on interspin distance analogous to the one of RKKY. The resulting interaction extends beyond nearest neighbors, e.g. nearest and next-nearest neighbors can interact with comparable strengths, allowing to realize the $J_1 - J_2$ XXZ model [25]. By making the next-nearest neighbor interaction antiferromagnetic, frustrated interactions in the $J_1 - J_2$ model similar to those in 1D copper oxide spin chains, can be realized.

Indirect magnetic interactions such as superexchange have been previously simulated with ultracold atoms in an optical lattice [27]. Past proposals also include simulation of phonon-mediated electron interactions in a hybrid system of trapped ions and ground state atoms [28] and in a bilayer of Rydberg atoms [29]. Spin-spin interactions with distance dependent tunable interaction strengths, giving rise to frustration, were simulated in a linear chain of three ions [30], where the spin-spin interactions were mediated by phonons in the ion chain [31]. A similar approach to realize phonon-mediated spin-spin interactions with distance-dependent interaction strength was considered in Ref.[32] for polar molecules arranged in a dipolar crystal. Spin-spin interactions can also be mediated via interaction with electromagnetic modes of a cavity [33]. Nuclear spins of ultracold atoms of two internal electronic states, tightly and weakly trapped in an optical lattice, were proposed to simulate the RKKY interaction [34]. The atoms interact via short-range s-wave potential, making the corresponding interaction strengths much smaller compared to the long-range dipole-dipole or charge-dipole interactions considered here.

The systems envisioned in this work offer the possibility that a large realizable atomic and molecular parameter set can be exploited to simulate a range of many-body interactions. The paper is organized as follows. In Section II we describe the system and derive the effective Hamiltonian for indirect interaction between effective spins encoded in polar molecules or Rydberg atoms. In Section III two examples of simulation of indirect interaction are discussed: i) XX interaction by encoding spins into low-energy rotational states of polar molecules, interacting via mediator Rydberg atoms; ii) XXZ interaction by spin encoding into states of Rydberg atoms, mediated by a Rydberg atom in a different state. Finally, we conclude in Section IV.

II. MODEL DESCRIPTION

In this section, we will introduce the physical system (cf, Fig. 1), derive its Hamiltonian, and simplify it into the effective Hamiltonian by tracing out and averaging over the degrees of freedom of the mediating Rydberg atoms. It will then be obvious that, like in the RKKY case, this system leads to interspin distance-dependent sign-changing interaction, which constitutes the main result of this article.

A. Effective interaction Hamiltonian

We consider a setup with two parallel 1D optical lattices or trap arrays, one filled with polar molecules or Rydberg atoms representing effective spin-1/2 systems, and another filled with auxiliary Rydberg atoms, mediating the interaction between the effective spins, as illustrated in Fig.1a. The effective spins are assumed to be tightly trapped in their optical lattice such that tunneling between sites is strongly suppressed. They interact with Rydberg atoms trapped in a parallel shallow optical lattice, in which tunneling is significant. In the setup, where the effective spins are comprised of polar molecules, e. g. Fig. 1(b), the interaction is charge-dipole

$$V_{\rm cd} = \frac{e\vec{d}_{\rm spin}\cdot\vec{R}}{R^3} - \frac{e\vec{d}_{\rm spin}\cdot\left(\vec{R}-\vec{r}\right)}{\left|\vec{R}-\vec{r}\right|^3}. \tag{1a}$$

Here, e is the electron charge, $\vec{d}_{\rm spin}$ is the molecule electric dipole moment, $\vec{R}$ is the distance between the mediator Rydberg atom ionic core and the spin-encoding system, and $\vec{r}$ is the distance between the Rydberg electron and the ionic core. In the scheme, where the effective spins are Rydberg atoms, the mediator atoms interact with the spin-encoding species via dipole-dipole interaction

$$V_{\rm dd} = \frac{\vec{d}_{\rm spin}\cdot\vec{d}_{\rm Ryd}}{R^3} - \frac{3\left(\vec{d}_{\rm spin}\cdot\vec{R}\right)\left(\vec{d}_{\rm Ryd}\cdot\vec{R}\right)}{R^5}, \tag{1b}$$

where $\vec{d}_{\rm Ryd}$ is the electric dipole moment of the mediator Rydberg atom. **For distances $\sim\mu$m between the spin and mediator arrays we will consider in the next section the charge-dipole interaction can be approximated by the dipole-dipole one, and in the following we will assume the interaction is dipole-dipole. More details on the charge-dipole interaction mediating the effective interaction can be found in Appendix A.**

The effective spins are assumed to occupy the lowest energy band of the deep optical lattice in both $|\downarrow\rangle$, $|\uparrow\rangle$ spin states, such that the spin-mediator interaction does not excite the spins to higher-energy bands. The mediator atoms, on the other hand, are assumed to be initially prepared in one of the low-energy bands in their lattice in the $|ns, m_j\rangle$ Rydberg state, such that they can be transferred to $\left|np_{j'}, m'_{j'}\right\rangle$, $\left|(n-1)p_{j'}, m'_{j'}\right\rangle$ internal and different motional states by the interaction with the spins. In the following we actually will need to consider only the coupling to $np_{j'}$ states. The mediator states are denoted as $|\mathbf{n}, k_\nu\rangle_q = \phi(X_q, k_{\nu\, q})\,|nl_j, m_j\rangle_q$, describing the motional $\phi(X_q, k_{\nu\, q})$ and internal $|nl_j, m_j\rangle_q$ states of the q$^{\rm th}$ mediator atom, and $\mathbf{n} = \{n, l, j, m_j\}$ is a short-hand notation for its internal quantum numbers. Here we assume for simplicity that the trapping potentials and therefore the motional states for $|ns, m_j\rangle$ and $\left|np_{j'}, m'_j\right\rangle$ internal states are the same, which is not a principal requirement,

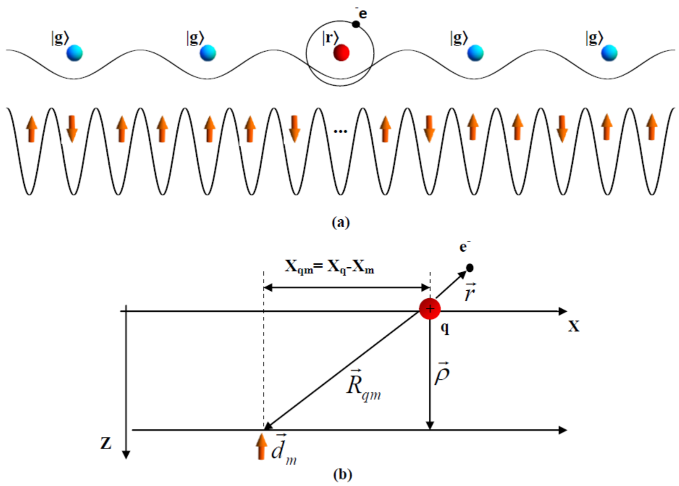

FIG. 1: *Setup schematic.* (a) Illustration of the bilayer setup, in which spin-encoding polar molecules or Rydberg atoms are trapped in a deep optical lattice or trap array, and the mediating Rydberg atom(s) is placed in a shallow optical lattice such that its spatial wave function is delocalized over several sites, and it can simultaneously interact with several spins; (b) Geometry of the setup: an effective m$^{\text{th}}$ spin with a dipole moment $\vec{d}_m$ interacts with a mediator atom via charge-dipole interaction Eq.(1a) in case of the polar molecule spin encoding or dipole-dipole interaction Eq.(1b) in the case of the Rydberg atom spin encoding. The distance between the two parallel lattices is $|\vec{\rho}|$, $\vec{R}_{qm}$ is the vector connecting the ionic core of the q$^{\text{th}}$ mediator atom to the spin, $X_{qm} = X_q - X_m$ is the distance between the mediator atom and the spin along the X axis, $\vec{r}$ is the vector connecting the mediator Rydberg electron and the ionic core.

and will be only used to simplify numerical analysis in the next section.

Rydberg atoms can be trapped in intensity minima in a ponderomotive [35] or a blue-detuned [36] optical lattice. The latter also can also trap atoms in their ground state, which can be used if a superatom or a dressed Rydberg mediator state is used, as will be discussed later. The motional wavefunction of the q$^{\text{th}}$ mediator atom is then given by the Bloch function of a ν^{th} Bloch band, corresponding to the quasimomentum k:

$$\phi(X_q, k_{\nu\, q}) = u_k^{(\nu)}(X_q) e^{ikX_q}, \tag{2}$$

where X_q is the coordinate of the atom along the lattice, $u_k^{(\nu)}(X_q + L_{\text{at}}) = u_k^{(\nu)}(X_q)$ is periodic with the period L_{at} of the mediator atom's lattice. We also assume periodic boundary conditions $N_{\text{latt at}} k L_{\text{at}} = 2\pi n_w$, where $N_{\text{latt at}}$ is the number of sites in the mediator lattice and n_w is the periodicity integer.

The Hamiltonian without the spin-mediator interaction has the form:

$$\hat{H}_0 = \sum_{i=1}^{N} E_{\text{spin}} \left|\uparrow\right\rangle_i \left\langle\uparrow\right|_i + \sum_{q=1}^{N_a} \sum_{\mathcal{M}=\mathbf{m},\mathbf{m}'} \mathcal{E}_{\mathcal{M}} \left|\mathcal{M}\right\rangle_q \left\langle\mathcal{M}\right|_q,$$

where $|\mathbf{m}\rangle_q = |\mathbf{ns}, k_\nu\rangle_q$ with $\mathbf{ns} = \{ns, j = 1/2, m_j = \pm 1/2\}$, $|\mathbf{m}'\rangle_q = |\mathbf{np}, k'_{\nu'}\rangle_q$ with $\mathbf{np} = \{np, j = 3/2, 1/2, m_j = \pm 3/2, \pm 1/2\}$; $E_{\text{spin}} = E_\uparrow - E_\downarrow$ is the spin transition energy and $\mathcal{E}_{\mathcal{M}} = \mathcal{E}_{\mathbf{ns(np)}}(k_\nu(k'_{\nu'}))$ is the energy of the mediator atom, including both internal $E_{\mathbf{ns(np)}}$ and motional energy of the corresponding Bloch states. The summation is over $i = 1, ..., N$ effective spins and $q = 1, .., N_a$ mediator atoms in the first and second optical lattices, respectively; over $nl_j = ns, np_{1/2,3/2}$; $m_j = \pm 1/2, \pm 3/2$ internal states of the mediator atoms, and their quasimomenta k in the first Brillouin zone of $\nu = 1, ..., \infty$ Bloch bands.

The spin-mediator interaction Hamiltonian can be written in the combined basis of spin and mediator states:

$$\hat{V} = \sum_{i=1}^{N} \sum_{q=1}^{N_a} \sum_{\substack{\mathbf{m},\mathbf{m}' \\ \alpha,\beta=\uparrow,\downarrow}} |\mathbf{m}\rangle_q |\alpha_i\rangle \langle\beta_i| \langle\mathbf{m}'|_q \times \times \langle\alpha_i| \langle\mathbf{m}|_q \hat{V} |\mathbf{m}'\rangle_q |\beta_i\rangle + \text{H.c.}, \tag{3}$$

where $\hat{V} = \hat{V}_{\text{dd}}$.

Next we show how the spin-mediator interaction Eq.(3) gives rise to indirect interaction between the effective spins. The interaction Hamiltonian in the basis of two-spin states $|\alpha_i \beta_m\rangle$ is:

$$\hat{V} = \sum_{i,m=1}^{N} \sum_{q=1}^{N_a} \sum_{\substack{\alpha,\beta,\gamma,\delta \\ \mathbf{m},\mathbf{m}'}} \Big[|\mathbf{m}\rangle_q |\alpha_i \beta_m\rangle \Big(V^{iq}_{\mathbf{m},\alpha;\mathbf{m}',\gamma} \delta_{\beta_m,\delta_m} + + V^{mq}_{\mathbf{m},\beta;\mathbf{m}',\delta} \delta_{\alpha_i,\gamma_i} \Big) \langle\gamma_i \delta_m| \langle\mathbf{m}'|_q + \text{H.c.} \Big]. \tag{4}$$

The interaction matrix element between the m$^{\text{th}}$ spin and the q$^{\text{th}}$ mediator atom is $V^{mq}_{\mathbf{m},\xi;\mathbf{m}',\eta} = \langle\mathbf{m}|_q \langle\xi_m| \hat{V} |\eta_m\rangle |\mathbf{m}'\rangle_q$, which describes the process in which the q^{th} mediator atom is transferred from the $|\mathbf{m}\rangle_q$ to the $|\mathbf{m}'\rangle_q$ state, and the m^{th} spin goes from the $|\xi\rangle$ to the $|\eta\rangle$ state.

The interaction Hamiltonian in the limit of weak interaction $|\hat{V}| \ll E_{\text{spin}}, |E_{np_{j'}} - E_{ns}|, |E_{np_{j'}} - E_{ns} \pm E_{\text{spin}}|$ induces energy shifts and couplings among many-body spin states $|\alpha_1 \alpha_2 ... \alpha_N\rangle$, corresponding to the same mediator state $|\mathbf{m}\rangle_q$, which have a form of interaction between the effective spins. This can be shown by allpying the Schrieffer-Wolff transformation to the total Hamiltonian $\hat{H} = \hat{H}_0 + \hat{V}$:

$$e^{\hat{S}} \hat{H} e^{-\hat{S}} = \hat{H} + \left[\hat{S}, \hat{H}\right] + \frac{\left[\hat{S}, \left[\hat{S}, \hat{H}\right]\right]}{2} + O\left(|\hat{S}^2 \hat{V}|\right), \tag{5}$$

in which terms of the first order in $\hat{V}$ are eliminated by setting $\left[\hat{S}, \hat{H}_0\right] = -\hat{V}$, where the corresponding generator $\hat{S}$ is given in Appendix A. The transformed Hamiltonian has the form:

$$e^{\hat{S}}\hat{H}e^{-\hat{S}} = \hat{H}_0 + \frac{\left[\hat{S},\hat{V}\right]}{2} + O\left(|\hat{V}|^3\right), \qquad (6)$$

in which the effective interaction $\hat{V}_{\rm eff} = \left[\hat{S},\hat{V}\right]/2$ terms are now of the second order in $\hat{V}$.

Assuming for concreteness that the mediator atoms are initially prepared in a single or a superposition of $\left|\mathbf{ns},k_\nu\right\rangle_q$ states, we are interested in the projection of the effective interaction on these states:

$$\hat{V}^{ns}_{\rm eff} = \hat{P}_{ns}\hat{V}_{\rm eff}\hat{P}_{ns} = \sum_{q=1}^{N_a}\sum_{\substack{k,\nu\\ m_j=\pm 1/2}} \left|\mathbf{ns},k_\nu\right\rangle_q \left\langle \mathbf{ns},k_\nu\right|_q \times$$
$$\times\left(\sum_{i,m=1}^{N}\sum_{\alpha,\beta,\gamma,\delta=\uparrow,\downarrow} K^{q,k_\nu}_{\alpha_i\beta_m,\gamma_i\delta_m}\left|\alpha_i\beta_m\right\rangle\left\langle\gamma_i\delta_m\right|\right), \qquad (7)$$

where

$$\hat{P}_{ns} = \sum_{q=1}^{N_a}\sum_{\substack{k,\nu\\ m_j=\pm 1/2}} \left|\mathbf{ns},k_\nu\right\rangle_q \left\langle \mathbf{ns},k_\nu\right|_q = \qquad (8)$$
$$= \sum_{q=1}^{N_a}\sum_{\substack{k,\nu\\ m_j=\pm 1/2}} \left|ns_{1/2},m_j,k_\nu\right\rangle_q \left\langle ns_{1/2},m_j,k_\nu\right|_q$$

is the projection operator on the ns state and the $K^{q,k_\nu}_{\alpha\beta,\gamma\delta}$ coefficients are given in Appendix B.

Replacing $\left|\alpha_i\beta_m\right\rangle\left\langle\gamma_i\delta_m\right|$ by $\hat{S}_i^{\pm,z}\hat{S}_m^{\pm,z}$ spin-1/2 operators as shown in Appendix B, we can rewrite Eq.(7) in the following way:

$$\hat{V}^{ns}_{\rm eff} = \sum_{q=1}^{N_a}\sum_{\substack{k,\nu\\ m_j=\pm 1/2}} \left|\mathbf{ns},k_\nu\right\rangle_q \left\langle \mathbf{ns},k_\nu\right|_q \times$$
$$\times\sum_{i,m=1}^{N}\Big[J^{zz\ q,k_\nu}_{im}\hat{S}^z_i\hat{S}^z_m + J^{+-\ q,k_\nu}_{im}\hat{S}^+_i\hat{S}^-_m +$$
$$+\left(J^{+-\ q,k_\nu}_{im}\right)^*\hat{S}^-_i\hat{S}^+_m + J^{++\ q,k_\nu}_{im}\hat{S}^+_i\hat{S}^+_m +$$
$$+\left(J^{++\ q,k_\nu}_{im}\right)^*\hat{S}^-_i\hat{S}^-_m + J^{z+\ q,k_\nu}_{im}\hat{S}^z_i\hat{S}^+_m +$$
$$+\left(J^{z+\ q,k_\nu}_{im}\right)^*\hat{S}^z_i\hat{S}^-_m + b^{z\ q,k_\nu}_{im} n_m\hat{S}^z_i +$$
$$+b^{+\ q,k_\nu}_{im} n_m\hat{S}^+_i + \left(b^{+\ q,k_\nu}_{im}\right)^* n_m\hat{S}^-_i + b^{q,k_\nu}_{0\ im} n_i n_m\Big], \qquad (9)$$

where $n_i(n_m)$ is the number of spins at site $i(m)$, and the interaction coefficients $J^{+-\ q,k_\nu}_{im}$, $J^{zz\ q,k_\nu}_{im}$, $J^{++\ q,k_\nu}_{im}$, $J^{z+\ q,k_\nu}_{im}$ and the coefficients $b^{z\ q,k_\nu}_i$, $b^{+\ q,k_\nu}_i$ and b^{q,k_ν}_0 are given in Appendix C.

If the effective interaction is weak such that $|J_{im}| \ll E_{\rm spin}, |E_{np_{j'}} - E_{ns}|, |E_{np_{j'}} - E_{ns} \pm E_{\rm spin}|$, the non-resonant terms $J^{++\ q,k_\nu}_{im}\hat{S}^+_i\hat{S}^+_m$, $\left(J^{++\ q,k_\nu}_{im}\right)^*\hat{S}^-_i\hat{S}^-_m$, $J^{z+\ q,k_\nu}_{im}\hat{S}^z_i\hat{S}^+_m$, $\left(J^{z+\ q,k_\nu}_{im}\right)^*\hat{S}^z_i\hat{S}^-_m$, $b^{z\ q,k_\nu}_i\hat{S}^+_i$ and $\left(b^{z\ q,k_\nu}_i\right)^*\hat{S}^-_i$, coupling collective spin states with energies differing by the spin transition energy or twice this energy, can be neglected. As a result, we are left with the effective interaction Hamiltonian:

$$\hat{V}^{ns}_{\rm eff} = \sum_{i,m=1}^{N}\sum_{q=1}^{N_a}\sum_{\substack{k,\nu\\ m_j=\pm 1/2}} \left|\mathbf{ns},k_\nu\right\rangle_q \left\langle \mathbf{ns},k_\nu\right|_q \times$$
$$\times\left(J^{zz\ q,k_\nu}_{im}\hat{S}^z_i\hat{S}^z_m + \frac{J^{\perp\ q,k_\nu}_{im}}{2}\left(\hat{S}^+_i\hat{S}^-_m + \hat{S}^-_i\hat{S}^+_m\right) + \right.$$
$$\left. + b^{z\ q,k_\nu}_{im}\hat{S}^z_i\right), \qquad (10)$$

where we assumed $\left(J^{+-\ q,k_\nu}_{im}\right)^* = J^{+-\ q,k_\nu}_{im} = J^{\perp\ q,k_\nu}_{im}/2$ and a unity filling of the spin lattice with $n_i = n_m = 1$ and omitted constant terms $\sim b^{q,k_\nu}_{0\ im}$.

The Hamiltonian, acting only on effective spins can be obtained by taking the expectation value of Eq.(10) with respect to an unperturbed initial state of the mediator atoms. We consider two examples:

1) the mediator atoms prepared in a Rydberg superatom state:

$$|\Psi\rangle_{\rm sat} = \sum_{q=1}^{N_a}\sum_{\substack{k',k_0\\ \nu',\nu_0}}\prod_{q'\neq q}\frac{c_{k'_{\nu'},k_{0\ \nu_0}}}{\sqrt{N_a}}\phi_{g_{q'}}\left(X_{q'},k'_{\nu'\ q'}\right)$$
$$\times\,\Phi_{ns_q}\left(X_q,k_{0\ \nu_0\ q}\right)\left|g_1,...\mathbf{ns}_q,...,g_{N_a}\right\rangle, \qquad (11)$$

in which the $q^{\rm th}$ mediator atom is in the $\left|\mathbf{ns},k_{0\ \nu_0}\right\rangle_q$ Rydberg state and $q' \neq q$ atoms are in the ground state $\left|g,k'_{\nu'}\right\rangle_{q'}$,

or

2) the Rydberg dressed state:

$$|\Psi\rangle_{\rm dress} = \prod_{q=1}^{N_a}\sum_{\substack{k',k_0\\ \nu',\nu_0}} c_{k'_{\nu'},k_{0\ \nu_0}}\left(c_g\phi_{g_q}(X_q,k'_{\nu'\ q})\left|g\right\rangle_q + \right.$$
$$\left. + c_{ns}\Phi_{ns_q}(X_q,k_{0\ \nu_0\ q})\left|\mathbf{ns}\right\rangle_q\right), \qquad (12)$$

created when all mediator atoms interact with a dressing laser field of Rabi frequency Ω and detuning Δ from the Rydberg state, and $c_g = \sqrt{\sqrt{\Delta^2/4+\Omega^2}+\Delta/2}/[\sqrt{2}\left(\Delta^2/4+\Omega^2\right)^{1/4}]$, $c_{ns} = \sqrt{\sqrt{\Delta^2/4+\Omega^2}-\Delta/2}/[\sqrt{2}\left(\Delta^2/4+\Omega^2\right)^{1/4}]$.

In both cases $\phi_{g_{q'}}\left(X_{q'},k'_{\nu'\ q'}\right)$ is the spatial wave function of a q'$^{\rm th}$ atom in the ground state; $\Phi_{ns_q}\left(X_q,k_{0\ \nu_0\ q}\right)$ is the spatial wave function of the q$^{\rm th}$ atom in the $\left|\mathbf{ns}\right\rangle_q$ Rydberg state. In the general case the atoms in the ground and Rydberg states are assumed to be prepared in a wave packet of Bloch states with quasimomenta $k'_{\nu'}$ and $k_{0\ \nu_0}$, respectively, weighted by the coefficients $c_{k'_{\nu'},k_{0\ \nu_0}}$. The spin Hamiltonian averaged over the mediator states takes the form:

$$\hat{V}^{ns}_{\text{eff spin}} = {}_{\substack{\text{sat}\\ \text{(dress)}}}\langle\Psi|\,\hat{V}^{ns}_{\text{eff}}\,|\Psi\rangle_{\substack{\text{sat}\\ \text{(dress)}}} = \tag{13}$$
$$= \sum_{i,m=1}^{N}\left(J^{zz}_{im}\hat{S}^z_i\hat{S}^z_m + \frac{J^{\perp}_{im}}{2}(\hat{S}^+_i\hat{S}^-_m + \hat{S}^-_i\hat{S}^+_m) + \right.$$
$$\left. + b^z_{im}\hat{S}^z_i\right).$$

As shown in Appendix D, using the assumption that initially the mediator atoms are prepared in a superposition of Bloch states the averaged interaction coefficients (same for b^z_{im}) can be written in the following form for the superatom initial mediator state:

$$J^{zz(\perp)}_{im} = \frac{1}{N_a}\sum_{q=1}^{N_a}\sum_{\substack{k',k_0\\ \nu',\nu_0}}|c_{k'_{\nu'},k_{0\ \nu_0}}|^2 J^{zz(\perp)\ q,k_0\ \nu_0}_{im} =$$
$$= \frac{1}{N_a}\sum_{q=1}^{N_a}\sum_{k_0,\nu_0}|c_{k_0\ \nu_0}|^2 J^{zz(\perp)\ q,k_0\ \nu_0}_{im}, \tag{14}$$

and the dressed initial state:

$$J^{zz(\perp)}_{im} = |c_{ns}|^2\sum_{q=1}^{N_a}\sum_{\substack{k',k_0\\ \nu',\nu_0}}|c_{k'_{\nu'},k_{0\ \nu_0}}|^2 J^{zz(\perp)\ q,k_0\ \nu_0}_{im}$$
$$= |c_{ns}|^2\sum_{q=1}^{N_a}\sum_{k_0,\nu_0}|c_{k_0\ \nu_0}|^2 J^{zz(\perp)\ q,k_0\ \nu_0}_{im}, \tag{15}$$

where $|c_{k_0\ \nu_0}|^2 = \sum_{k',\nu'}|c_{k'_{\nu'},k_{0\ \nu_0}}|^2$. In particular, for mediator atoms initially prepared in a stationary BEC with $k_0 = 0$, $\nu_0 = 1$ [37] the averaged interaction coefficients are $J^{zz(\perp)}_{im} = \frac{1}{N_a}\sum_{q=1}^{N_a} J^{zz(\perp)\ q,k_0=0_{\nu_0=1}}_{im}$ for the superatom and $J^{zz(\perp)}_{im} = |c_{ns}|^2\sum_{q=1}^{N_a} J^{zz(\perp)\ q,k_0=0_{\nu_0=1}}_{im}$ for the dressed initial states. In a more general case the initial mediator state is a superposition of Bloch states with quasimomenta k_0 in Bloch bands ν_0 determined by the distribution $|c_{k_0\ \nu_0}|^2$. Assuming for simplicity that the only dependence of the $J^{zz(\perp)\ q,k_0\ \nu_0}_{im}$ coefficients on the initial quasimomentum k_0 is given by the prefactor $J^{zz(\perp)\ q,k_0\ \nu_0}_{im} \sim e^{-ik_0(X_i-X_m)}$, the averaged interaction coefficients for the superatom initial state (similar for the dressed state) have the form:

$$J^{zz(\perp)}_{im} \sim \frac{1}{N_a}\sum_{q=1}^{N_a} J^{zz(\perp)\ q}_{im}\sum_{k_0,\nu_0}$$
$$|c_{k_0\ \nu_0}|^2 e^{-ik_0(X_i-X_m)}. \tag{16}$$

A Gaussian distribution $|c_{k_0\ \nu_0}|^2 \sim e^{-k_0^2/\kappa_0^2}$ will give $\sum_{k_0,\nu_0}|c_{k_0\ \nu_0}|^2 e^{-ik_0(X_i-X_m)} \sim e^{-(X_i-X_m)^2\kappa_0^2/4}$ for a narrow wave packet with $\kappa_0 \ll \pi/L_{\text{at}}$, resulting in an additional factor of decay of interaction coefficients with an interspin distance, controlled by the wave packet distribution width κ_0. The κ_0 therefore can allow to control the relative strengths of e.g. nearest neighbor to next nearest neighbor and more distant interactions.

The total effective Hamiltonian will thus take the form:

$$\hat{H}_{\text{eff}} = \hat{H}_{0\text{ spin}} + \hat{V}^{ns}_{\text{eff spin}} =$$
$$= \sum_{i,m=1}^{N}\left(J^{zz}_{im}\hat{S}^z_i\hat{S}^z_m + \frac{J^{\perp}_{im}}{2}\left(\hat{S}^+_i\hat{S}^-_m + \hat{S}^-_i\hat{S}^+_m\right)\right) +$$
$$+ \sum_{i=1}^{N}\left(E_{\text{spin}} + b^z_i\right)\hat{S}^z_i, \tag{17}$$

where the effective magnetic field $b^z_i = \sum_{m\neq i} b^z_{im}$ at site i was introduced. The effective Hamiltonian (17) with averaged interaction coefficients from Eqs. (14), and (15) is the main result of our work. In the case b^z_i do not depend on i, the Hamiltonian couples collective spin states with the same z component of the total spin $\hat{S}^z = \sum_{i=1}^{N}\hat{S}^z_i$, and describes the XXZ model of magnetism in the presence of a longitudinal magnetic field. As will be shown below, due to mediator atoms being spatially delocalized in their lattice, both the magnitude and the sign of the interaction coefficients J^{zz}_{im}, $J^{\perp}_{im}$ can depend on the distance between the spins.

We note that since in the XXZ model the total magnetization $\sum_{i=1}^{N}\hat{S}^z_i$ is conserved, the effective Zeeman energy (last) term in the Hamiltonian (17) is needed to neglect the magnetization non-conserving terms $\hat{S}^+_i\hat{S}^+_j$, $\hat{S}^-_i\hat{S}^-_j$, $\hat{S}^+_i\hat{S}^z_j$, $\hat{S}^-_i\hat{S}^z_j$, etc. in Eq.(9). It dominates over interactions since $E_{\text{spin}} \gg |J^{\perp}_{im}|, |J^{zz}_{im}|$, and the ground state corresponds to all $|\downarrow\rangle$ spins. However, if the effective spins are initially prepared in a superposition of states, corresponding to zero total magnetization (setting the Zeeman term to zero), the system can have non-trivial non-magnetic ground states [26]. Any zero magnetization state, i.e. a state with an equal number of spins in the $|\uparrow\rangle$ and $|\downarrow\rangle$ states can be prepared e.g. by individually manipulating the spins and setting each spin in the $|\uparrow\rangle$ or $|\downarrow\rangle$ state with MW or radiofrequency fields. Next, this state can be coupled to other zero magnetization states by e.g. adiabatic passages using chirped or STIRAP microwave or radiofrequency pulse sequencies via states with ± 1 magnetization, differing from the zero magnetization ones by one flip of a spin. In order to selectively couple particular zero magnetization states by the adiabatic passage pulses the transition frequencies of the spins can be individually controlled using e.g. electric or magnetic field induced gradients of the spin frequency along the spin chain.

The XXZ model in the presence of the Zeeman term can also be used to study the dynamics of out-of-equilibrium systems, when the spin system is initially prepared in a superposition of its eigenstates, which can be considered as an analog of a quantum quench. Its evolution will involve growth and spread of quantum correlations and entanglement between spins, which is known to lead to an equlibrium state, closely resembling a thermal one on a local scale [38]. It was theoretically predicted [39] and experimentally shown in a BEC of magnetic Cr atoms [40] and a trapped ions system [41] (XX model in the presence of a large longitudinal magnetic field similar to the one of Section IIIa below was simulated), that the system dynamics strongly depends on the initial averaged magnetization $\langle \hat{S}^z \rangle = \langle \sum_{i=1}^{N} \hat{S}_i^z \rangle$. In particular, it was shown that the closer the total averaged magnetization to zero, the more important role quantum correlations play and the faster the entanglement between the spins grows.

B. Sign varying interactions

Below we show that the interaction coefficients $J_{im}^{\perp}$, J_{im}^{zz} can change sign depending on the distance between the spins. From Eq.(4) one can see that the coefficients depend on the interaction matrix elements, given by the expression:

$$V^{mq}_{\mathbf{m_0},\alpha;\mathbf{m}',\alpha'} = \int dX_q \Phi^{*}_{ns_q}(X_q, k_{0\ \nu_0\ q}) \times \\ \times \langle \mathbf{ns}| \langle \alpha_m | \hat{V}(\vec{R}_{qm}) | \alpha'_m \rangle | \mathbf{np}' \rangle \, \phi(X_q, k_{\nu\ q}), \quad (18)$$

where the initial mediator state is denoted as $|\mathbf{m_0}\rangle_q = |\mathbf{ns}, k_{0\ \nu_0}\rangle_q$. Let us approximate the Bloch functions of the mediator motional state by plane waves as $\phi(X_q, k_{\nu\ q}) = e^{ikX_q}/\sqrt{N_{\text{latt at}} L_{\text{at}}}$, $\Phi_{ns_q}(X_q, k_{0\ \nu_0}) = e^{ik_0 X_q}/\sqrt{N_{\text{latt at}} L_{\text{at}}}$. The matrix elements then have the form:

$$V^{mq}_{\mathbf{m_0},\alpha;\mathbf{m}'\alpha'} = \frac{1}{N_{\text{latt at}} L_{\text{at}}} e^{i(k-k_0)X_m} \times \quad (19) \\ \times \int dX_{qm} \langle \mathbf{ns}| \langle \alpha_m | \hat{V}(\vec{R}_{qm}) | \alpha'_m \rangle | \mathbf{np}' \rangle \, e^{i(k-k_0)X_{qm}},$$

where the x coordinate X_m of the m^{th} spin and the separation between the q^{th} mediator atom and the m^{th} spin along the x axis $X_{qm} = X_q - X_m$ are introduced (see Fig.1b). The integral in the above expression no longer depends on X_m for sufficiently long spin and mediator atom arrays. As a result, the matrix element can be written as

$$V^{mq}_{\mathbf{m_0},\alpha;\mathbf{m}',\alpha'} = c^{mq}_{\mathbf{m_0},\alpha;\mathbf{m}'\alpha'} e^{i(k-k_0)X_m}. \quad (20)$$

with $c^{qm}_{\mathbf{m_0},\alpha;\mathbf{m}',\alpha'}$ independent of X_m. The interaction coefficients will be proportional to a sum over quasimomenta k of the first Brillouin zone of the product of the matrix elements corresponding to the interaction of the q^{th} atom with i^{th} and m^{th} spins:

$$J^{\perp(zz)\ q}_{im} \sim \sum_{k,\nu} \frac{V^{iq}_{\mathbf{m_0},\alpha;\mathbf{m}',\alpha'} \left(V^{mq}_{\mathbf{m_0},\beta;\mathbf{m}',\beta'} \right)^*}{\mathcal{E}_{\mathbf{m}'} - \mathcal{E}_{\mathbf{m_0}}} \\ \sim \sum_{\nu} \frac{c^{iq}_{\mathbf{m_0},\alpha;\mathbf{m}'\alpha'} \left(c^{mq}_{\mathbf{m_0},\beta;\mathbf{m}',\beta'} \right)^*}{\mathcal{E}_{\mathbf{m}'} - \mathcal{E}_{\mathbf{m_0}}} \times \\ \times \sum_k e^{i(k-k_0)(X_i - X_m)},$$

where it is assumed for simplicity that the c^{qm} terms and the total energies of the mediator states weakly depend on the quasimomentum k. For high ν Bloch bands the motional energy will eventually become comparable to the internal energies, but for these bands the overlap integral between the interaction potential and Bloch functions in Eq.(18) will already be negligible. The summation over the quasimomenta of the first Brillouin zone will give the factor

$$\sum_k e^{ik(X_i - X_m)} = \frac{\sin\left[\frac{\pi(X_i - X_m)(1+N_{\text{latt at}})}{N_{\text{latt at}} L_{\text{at}}}\right]}{\sin\left[\frac{\pi(X_i - X_m)}{N_{\text{latt at}} L_{\text{at}}}\right]} = (-1)^p$$

for a spin lattice having the same period as the mediator lattice with $X_i - X_m = pL_{\text{at}}$. In addition to varying the sign the interaction coefficients also fall off with the distance between the spins. The c^{qm} factors have approximate dependence $\sim 1/R^3_{qm}$, resulting in $J^{\perp(zz)\ q}_{im} \sim 1/(R^3_{qi} R^3_{qm}) \sim 1/|X_i - X_m|^6$ for distant spins.

This simplified derivation qualitatively shows that the interaction coefficients can change sign with an interspin distance, analogous to the RKKY effect. In the next section two examples of spin encoding in polar molecules and Rydberg atoms will be considered and the corresponding interaction coefficients $J^{\perp(zz)}_{im}$ will be numerically calculated.

III. MODELLING XX, ISING, XXZ INTERACTIONS

In this section effective spin-spin interactions that can be realized in the bilayer system are discussed by analyzing the interaction coefficients $J^{\perp}_{im}$ and J^{zz}_{im}, given in Appendix C. Examples of i) XX interaction using spin encoding in polar molecule states and ii) XXZ interaction using Rydberg atom spin encoding are considered.

A. XX interaction with NaCs effective spins and Rb Rydberg mediator atoms

The XX interaction $\sum_{i,m=1}^{N} J^{\perp}_{im} \left(\hat{S}^+_i \hat{S}^-_m + \hat{S}^-_i \hat{S}^+_m \right)/2 + \sum_{i=1}^{N} (E_{\text{spin}} + b^z_i) \hat{S}^z_i$ can be realized if the spin states have zero dipole moments $\langle\uparrow|\vec{d}_{\rm spin}|\uparrow\rangle = \langle\downarrow|\vec{d}_{\rm spin}|\downarrow\rangle = 0$, non-zero spin transition dipole moment $\langle\uparrow|\vec{d}_{\rm spin}|\downarrow\rangle \neq 0$, and spin $|\uparrow\rangle \leftrightarrow |\downarrow\rangle$ and mediator $|ns\rangle \leftrightarrow |np_{j'}\rangle$ transitions are close in energy such that their energy difference $\Delta E = E_{np_{j'}} - E_{ns} \pm E_{\rm spin}$ is $|\Delta E| \ll E_{\rm spin}, |E_{np_{j'}} - E_{ns}|$ (but $|\Delta E| \gg |\hat{V}|$) (see Fig.2a,b). In this case from Eqs.(C.2)-(C.6) one can see that $|J_{im}^{\perp\ q,k_\nu}|, |b_i^{z\ q,k_\nu}| \neq 0$, $|J_{im}^{zz\ q,k_\nu}| = 0$, i.e. only the spin flipping terms $J_{im}^{\perp}\hat{S}_i^{\pm}\hat{S}_m^{\mp}$ will be present in the effective interaction.

In the polar molecules setup the spin-exchange interaction between a polar molecule and a mediator Rydberg atom can be realized if a rotational molecular transition is close in energy to a Rydberg transition (atom-molecule Forster resonance). The Forster resonances between rotational states of a polar molecule and atomic Rydberg states have been studied recently in [42], where the resonant exchange between a NH_3 molecule and a He atom has been experimentally observed. For effective spins, encoded in Rydberg states, atom-atom Forster resonances can be used to realize the spin-mediator interaction. In this case e.g. different atomic species can be used to encode the spin and mediate the interaction, such as Rb and Cs. For this atomic pair there are several interspecies Forster resonances available such as $\left|\text{Rb}59s_{1/2}, \text{Cs}57s_{1/2}\right\rangle \leftrightarrow \left|\text{Rb}58p_{1/2}, \text{Cs}57p_{1/2}\right\rangle$ with an energy defect $\Delta E = -16.6$ MHz, $\left|\text{Rb}81s_{1/2}, \text{Cs}78s_{1/2}\right\rangle \leftrightarrow \left|\text{Rb}80p_{1/2}, \text{Cs}78p_{1/2}\right\rangle$ with $\Delta E = 6.31$ MHz, $\left|\text{Rb}82s_{1/2}, \text{Cs}79s_{1/2}\right\rangle \leftrightarrow \left|\text{Rb}81p_{1/2}, \text{Cs}79p_{1/2}\right\rangle$ with $\Delta E = -6.41$ MHz, $\left|\text{Rb}84s_{1/2}, \text{Cs}89s_{1/2}\right\rangle \leftrightarrow \left|\text{Rb}84p_{1/2}, \text{Cs}88p_{1/2}\right\rangle$ with $\Delta E = -2.43$ MHz [43]. In fact, recently signs of indirect interaction between two Rydberg atoms, mediated by a third one, have been observed in [44]. An advantage of using different species for spin encoding and mediating the interaction is that they can be spectrally addressed using laser fields of different frequencies, allowing to separately initialize, control and read out their states. Finally, a spin can be encoded in two ground state sublevels of neutral atoms, which can be coupled to two Rydberg states such as $|ns\rangle$ and $|np_j\rangle$ to form Rydberg dressed states. In this case two dressed atoms encoding spins can indirectly interact via spin-exchange with a mediator Rydberg atom.

As an example of how indirect XX interaction can be modelled we consider a system of spin-encoding polar molecules and mediator Rydberg atoms in a superatom state Eq.(11). In the case interactions between a Rydberg atom and ground state atoms are present in the superatom state [45], a single mediator atom can be used. Each molecule is assumed to be in the ground electronic and vibrational state; two low-energy rotational states are used to encode the spin states, e.g. $|\uparrow\rangle = |J=1, m_J=0\rangle$, $|\downarrow\rangle = |J=0, m_J=0\rangle$. We consider the case shown in Fig.2a, where the spin $|\uparrow\rangle \leftrightarrow |\downarrow\rangle$ and mediator $|ns\rangle \leftrightarrow \left|np_{j'}, m'_{j'}\right\rangle$ transitions are close in energy such that $|\Delta E = E_{np_{j'}} - E_{ns} - E_{\rm spin}| \ll E_{\rm spin}, |E_{np_{j'}} - E_{ns}|$ and the states $|ns, m_j\rangle|\uparrow\rangle$ and $\left|np_{j'}, m'_{j'}\right\rangle|\downarrow\rangle$ are almost degenerate. In this case the atomic and molecular basis sets can be limited to only the $|ns, m_j\rangle$, $\left|np_{j'}, m'_{j'}\right\rangle$ and $|\downarrow\rangle$, $|\uparrow\rangle$ states. The coefficients $J_{im}^{\perp\ q,k_0\ \nu_0}$ and $b_i^{z\ q,k_0\ \nu_0}$ will have the form (see Eqs.(C.3),(C.6)):

$$
J_{im}^{\perp\ q,k_0\ \nu_0} \approx -\sum_{\mathbf{m}'} \frac{2V^{iq}_{\mathbf{m_0},\uparrow;\mathbf{m}',\downarrow}\left(V^{mq}_{\mathbf{m_0},\uparrow;\mathbf{m}',\downarrow}\right)^*}{\mathcal{E}_{\mathbf{m}'} - \mathcal{E}_{\mathbf{m_0}} - E_{\rm spin}},
$$

$$
b_i^{z\ q,k_0\ \nu_0} = -\sum_{\substack{m\neq i\\ \mathbf{m}'}} \frac{2\left|V^{iq}_{\mathbf{m_0},\uparrow;\mathbf{m}',\downarrow}\right|^2}{\mathcal{E}_{\mathbf{m}'} - \mathcal{E}_{\mathbf{m_0}} - E_{\rm spin}}. \tag{21}
$$

As a concrete example, we consider a 1D array of spin-encoding NaCs molecules and mediator Rb($99s_{1/2}$) Rydberg atoms in a superatom state (or a single mediator atom in the Rydberg state), placed in two parallel 1D optical lattices (see Fig.1a). NaCs has the permanent dipole moment $d = 4.607$ D [46] and the rotational constant $B = 0.06047$ cm^{-1} $= 1.813$ GHz [47], and is actively studied for formation of ground state ultracold molecules [48]. The molecular rotational transition with $E_{\rm spin}/\hbar = 2B \approx 3.62569$ GHz is nearly resonant with the Rb($99p_{3/2} - 99s_{1/2}$) transition, $(E_{99p_{3/2}} - E_{99s_{1/2}})/\hbar \approx 3.62745$ GHz, where $E_{nlj} = -\frac{1}{2(n-\mu_{lj})^2}$ (in a.u.) and $\mu_s = 3.1311804$, $\mu_{p_{1/2}} = 2.6548849$ [49], with a frequency defect of only $(\Delta E = E_{99p_{3/2}} - E_{99s_{1/2}} - E_{\rm spin})/\hbar \approx 1.76$ MHz. The frequency defect between the Rb($99p_{1/2} - 99s_{1/2}$) and the molecule spin transition is ≈ -95.4 MHz, and spin flips involving this transition can be neglected. Other examples of near resonant molecular rotational $J=1 \leftrightarrow J=0$ and Rydberg transitions are listed in Table I.

Details of the calculations of the matrix elements of the spin-mediator charge-dipole interaction and the coefficients Eqs.(21) are given in Appendix E. Fig.3 shows the numerically calculated $J_{im}^{\perp} = \sum_{q=1}^{N_a} J_{im}^{\perp\ q,k_0=0_{\nu_0=1}}/N_a$ and $b_i^z = \sum_{q=1}^{N_a} b_i^{z\ q,k_0=0_{\nu_0=1}}/N_a$ coefficients for an $m^{\rm th}$ spin at the center ($m=0$) of the array interacting with an $i^{\rm th}$ spin depending on their spatial separation assuming that initially the mediator atoms are prepared in a BEC state $k_0 = 0$, $\nu_0 = 1$ and in the $\left|99s_{1/2}, m_j = 1/2\right\rangle$ internal state. The following parameters were used in the calculations: the spin-mediator arrays distance $\rho = 1.5$ μm; the spin and mediator lattices periods $L_{\rm spin} = L_{\rm at} = 1.5$ μm; number of spins and mediator atoms $N = N_a = 100$; the mediator atoms lattice depth $V_0 = -E_{\rm rec}$, where $E_{\rm rec} = \hbar^2\left(\pi/L_{\rm at}\right)^2/2m_{\rm Rb}$ is the recoil energy of mediator atoms, m_{Rb} is the mass of Rb atom; and $\nu = 1, ..., 5$ Bloch bands of the mediator lattice were taken into account. One can see that i) the $J_{im}^{\perp}$ changes sign with an interspin distance. The nearest neighbor and the next nearest neighbor interactions can be ferro- and antiferromagnetic, respectively; ii) the interaction extends beyond

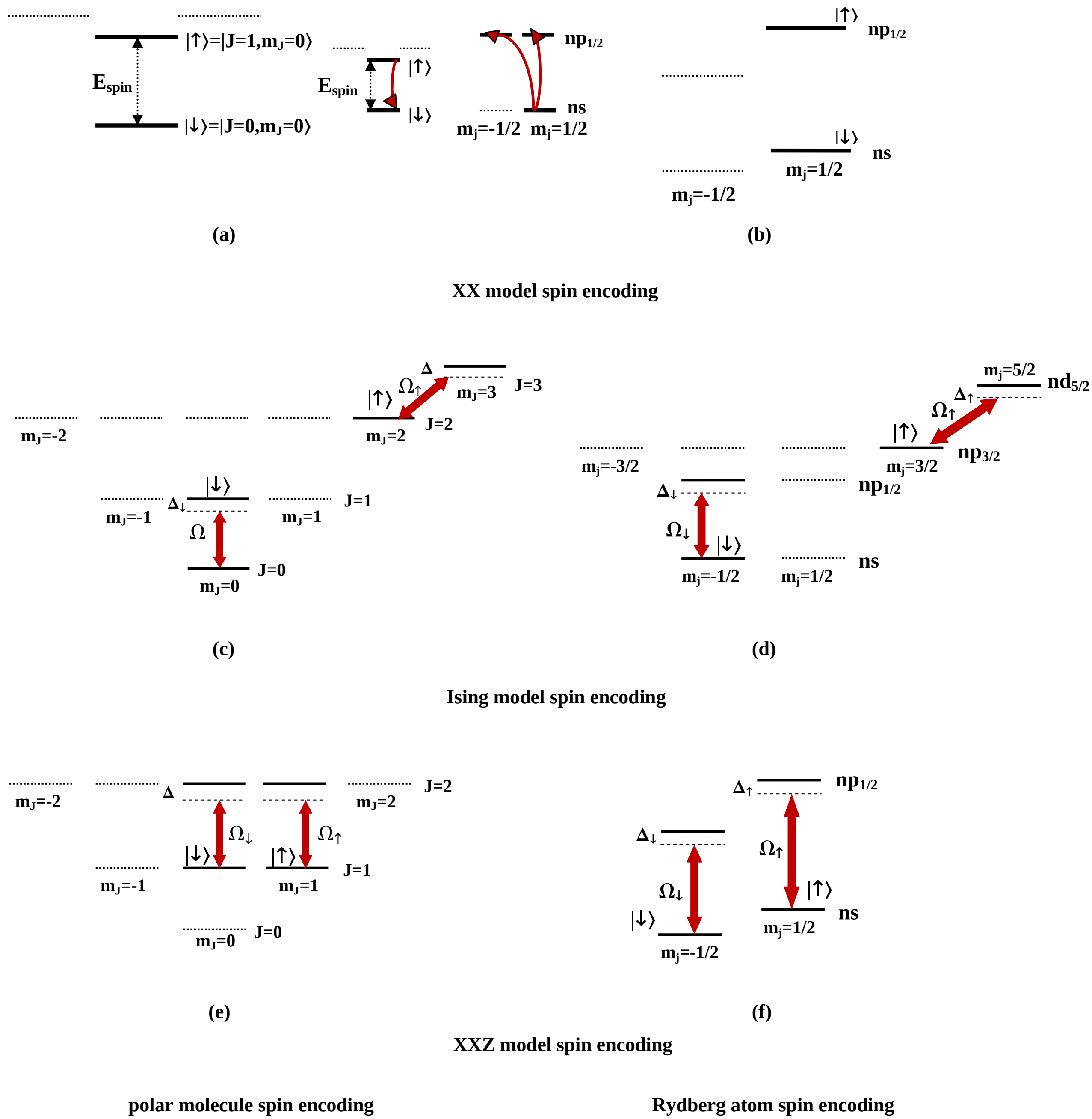

FIG. 2: *Schemes of different types of spin encoding into low-energy rotational states of polar molecules (left side) or long-lived states of Rydberg atoms (right side), allowing to model indirect interactions in a 1D effective spin chain*: (a) spin encoding into low-energy rotational states of a polar molecule and near resonant interaction between molecular rotational and mediator Rydberg atomic states, allowing to model XX type of interaction; (b) example of spin encoding into long-lived states of a Rydberg atom; (c) polar molecule rotational states not connected by a dipole-allowed transition can be used as spin states to realize effective Ising-type spin-spin interaction. Spin state dipole moments can be induced by near-resonant coupling to opposite parity rotational states by MW fields; (d) the same as in (c) for spins encoded in Rydberg states; (e) spin encoding in nearly degenerate rotational sublevels of a polar molecule such that $|E_{\rm spin}| \ll |\mathcal{E}_{np_{j'}} - \mathcal{E}_{ns}|$ is satisfied allows to realize an effective XXZ interaction; (f) the same as (e) for Rydberg atom spin encoding. The Rydberg encoded spin states can be initially additionally split by an external magnetic field.

TABLE I: Examples of Forster resonances between $J = 1 \leftrightarrow J = 0$ rotational transitions of alkali dimer polar molecules and Rydberg transitions of alkali atoms

Species	B, cm^{-1} ($E_{\rm spin}/\hbar = 2B$, GHz)	$nl'_{j'} - nl_j$	$(\Delta E = E_{nl'_{j'}} - E_{nl_j} - E_{\rm spin})/\hbar$, MHz
LiCs+Na	0.218 (13.071)	$64p_{3/2(1/2)} - 64s_{1/2}$	-26.4 (-42.7)
LiCs+Rb	0.218 (13.071)	$65p_{1/2} - 65s_{1/2}$	11
LiRb+Rb	0.254 (15.229)	$62p_{1/2} - 62s_{1/2}$	-52
LiNa+Rb	0.425 (25.482)	$53p_{3/2} - 53s_{1/2}$	111
LiK+Rb	0.293 (17.568)	$59p_{1/2} - 59s_{1/2}$	176

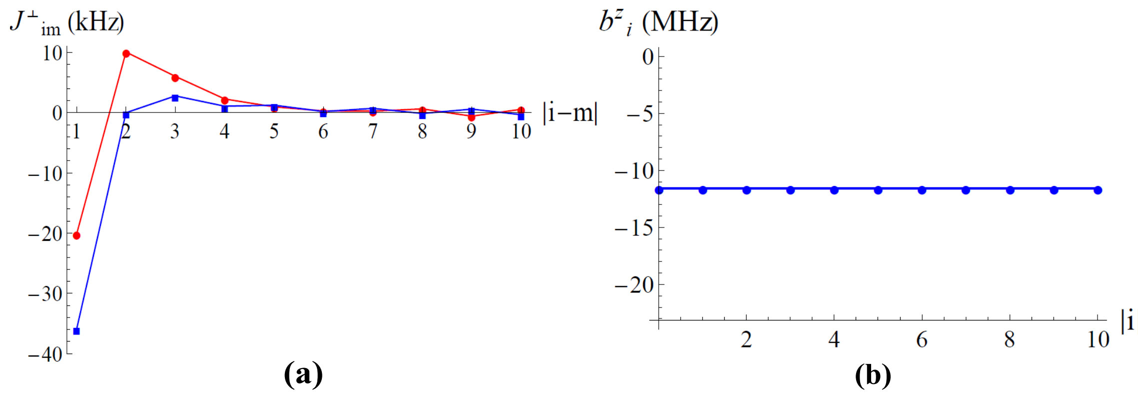

FIG. 3: *Spin encoding in low-energy rotational states of polar molecules near resonantly interacting with a mediator atom Rydberg transition allows to model XX interaction.* Numerically calculated from Eqs.(21): (a) Interaction coefficient $J^{\perp}_{im}$ and (b) effective magnetic field b^z_i for spins encoded in rotational states of NaCs $|\downarrow\rangle = |J = 0, m_J = 0\rangle$, $|\uparrow\rangle = |J = 1, m_J = 0\rangle$, near resonant with the Rb transition $|99p_{3/2}, m_j = \pm 1/2, 3/2\rangle - |99s_{1/2}, m_j = 1/2\rangle$. The mediator atoms are assumed to be initially prepared in the superatom state Eq.(11) in the $|99s_{1/2}, m_j = 1/2\rangle$ internal and in a BEC motional state with $k_0 = 0$, $\nu_0 = 1$. In Eqs.(21), summation over quasimomenta in the first Brillouin zone for $\nu = 1, ..., 5$ lowest Bloch bands of the mediator lattice with the depth $V_0 = -E_{\rm rec}$ was performed. The red (circles) and blue (squares) curves in (a) correspond to $B_M = 0$ G and $B_M = 0.92$ G, respectively; in (b) the effective magnetic field was calculated for $B_M = 0$ G. Setup dimensions: $\rho = 1.5$ μm, spin and mediator lattice periods $L_{\rm spin} = L_{\rm at} = 1.5$ μm, $N = N_{\rm at} = 100$. The calculations were done for a spin in the center of the array $m = 0$.

nearest neighbors and falls off at about $|i - m| \sim 5$; iii) the interaction strengths $\sim$ ten kHz can be realized; iv) b^z_i do not depend on i and only change the initial spin transition frequency. Two consequencies follow: first, the term $\sum_{i=1}^{N}(E_{\rm spin} + b^z_i)\hat{S}^z_i$ corresponds to a Zeeman interaction with a homogeneous effective magnetic field; cecond, the effective spin-spin interaction strength $\sim$ ten kHz is more than an order of magnitude larger than the strength of the direct dipole-dipole interaction between NaCs molecules $V_{dd} \sim d^2_{\rm spin\ z}/L^3_{\rm spin} \sim 300$ Hz.

From Fig.3a one can see that if the mediator atoms are initially prepared in a BEC state the interaction is significant between spins separated by up to five lattice sites. The interaction strength between the next-nearest and more distant neighbors Eq.(14) can be controlled by preparing the initial mediator superatom state as a superposition of Bloch states Eqs.(11). For example, if the mediator atoms are initially prepared in a superposition of Bloch states in the second Bloch band $\nu_0 = 2$ with a Gaussian distribution of quasimomenta $|c_{k_0\ \nu_0=2}|^2 \sim e^{-k_0^2/\kappa_0^2}$, the ratio of the next-nearest and more distant to the nearest neighbor interaction strengths can be controlled by the quasimomenta distribution width κ_0, as shown in Fig.4a. **At $\kappa_0/(\pi/L_{\rm at}) \sim 0.2$, the ratios $J^{\perp}_{im}/|J^{\perp}_{m+1,m}| \leq 0.4$ and $J^{\perp}_{im}/J^{\perp}_{m+2,m} \leq 0.3$ for $|m - i| \geq 3$ and the interaction can be approximated as the $J_1 - J_2$ XX model $\sum_{l=1,2} J_l \left(\hat{S}^x_i \hat{S}^x_{i+l} + \hat{S}^y_i \hat{S}^y_{i+l}\right)$. The phase diagram for ferromagnetic nearest neighbour $J_1 = J^{\perp}_{m+1,m} < 0$ and antiferromagnetic next-nearest neighbor $J_2 = J^{\perp}_{m+2,m} > 0$ interactions were analyzed in [26], where it was found that for $J_2/|J_1| \approx 1/1.4 = 0.71$ realized at $\kappa_0/(\pi/L_{\rm at}) \sim 0.2$ in our case, the system is in a vector chiral phase [50]. Another way to control the strength of the interaction be-**

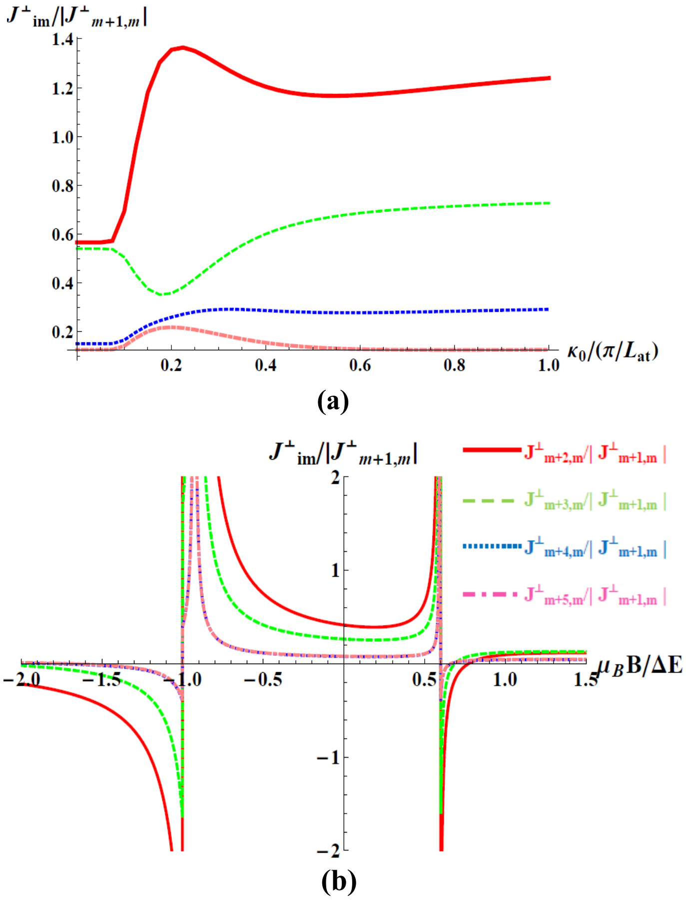

FIG. 4: *In the XX model next-nearest and more distant interaction strengths relative to the nearest neighbor interaction strength can be controlled by (a) the width of the quasimomenta distribution of the initial superposition of Bloch states of mediator atoms in Eq.(14); (b) external DC magnetic field.* (a) Assuming a Gaussian initial distribution of the quasimomenta in the second Bloch band $|c_{k_0\,\nu_0=2}|^2 \sim e^{-k_0^2/\kappa_0^2}$, the ratios of interaction coefficients $J^\perp_{im}/|J^\perp_{m+1,m}|$ can be controlled by the distribution width κ_0: $J^\perp_{m+2,m}/|J^\perp_{m+1,m}|$ (red solid curve), $J^\perp_{m+3,m}/|J^\perp_{m+1,m}|$ (green dashed curve), $J^\perp_{m+4,m}/|J^\perp_{m+1,m}|$ (blue dotted curve), $J^\perp_{m+5,m}/|J^\perp_{m+1,m}|$ (pink dashed-dotted curve). (b) the external magnetic field can tune the energies of the initial $|ns_{1/2}, m_j = \pm 1/2\rangle$ and virtually excited $|np_{3/2}, m_j = \pm 1/2, 3/2\rangle$ mediator states, which can be used to tune the ratios of interaction coefficients $J^\perp_{im}/|J^\perp_{m+1,m}|$.

tween distant neighbors is to tune the energies of $|ns_{1/2}, m_j = \pm 1/2\rangle$ and $|np_{3/2}, m_j = \pm 1/2, 3/2\rangle$ states with a magnetic field B_M. In particular, for $\mu_B B_M/\Delta E \approx 0.77$, corresponding to $B_M \approx 0.92$ G, the $J^\perp_{im} \ll |J^\perp_{m+1,m}|$, as shown by the blue squares curve in Fig.3a, resulting in the exactly solvable XX model with only nearest neighbor interactions [51]. Fig. 4b shows how the sign of interaction coefficients between distant neighbors and their strengths relative to the nearest neighbor one can be controlled more extensively by tuning the magnetic field.

The mediator atoms have to stay in the ns state long enough for the indirect interaction to take place, i.e. the effective interaction strength should be larger than the mediator Rydberg state decay rate. For Rb$(99s_{1/2})$, the lifetimes, including contributions from spontaneous emission and interaction with black-body radiation, are $\tau_{99s_{1/2}} = 1.168$ ms at $T = 4.2$ K and $\tau_{99s_{1/2}} = 330$ μs at $T = 300$ K [52]. The $J^\perp_{im} \sim 10$ kHz corresponds to interactions times ~ 10 μs, which are two orders of magnitude shorter than the mediator Rydberg state decay times at cryogenic temperatures, making the interaction observable.

Finally, we note that in order to selectively address the spin transition the degeneracy between the $|\uparrow\rangle = |J = 1, m_J = 0\rangle$ state and the $|J = 1, m_J = \pm 1\rangle$ rotational states should be lifted such that their energy difference exceeds the effective interaction strength. The degeneracy can be lifted by a DC electric field or by MW fields, coupling the $|J = 1, m_J = 0, \pm 1\rangle$ to $|J = 2, m_J = 0, \pm 1, \pm 2\rangle$ states. For example, for a σ^+ polarized MW field the ratio of the dipole moments for the transitions $|1, -1\rangle \leftrightarrow |2, 0\rangle$, $|1, 0\rangle \leftrightarrow |2, 1\rangle$ and $|1, 1\rangle \leftrightarrow |2, 2\rangle$ is $|d_{1,-1;2,0}|/|d_{1,0;2,1}|/|d_{1,1;2,2}| = (1/\sqrt{6})/(\sqrt{2})/(2)$ and the states will be shifted differently by the MW field. For a MW field with detuning $\tilde{\Delta} \sim 10$ MHz and Rabi frequency $\tilde{\Omega} \sim 3$ MHz shifts $|\tilde{\Omega}|^2/\tilde{\Delta} \sim 1$ MHz will be induced, exceeding the effective interaction strength by more than an order of magnitude. There is a subtle point, however, that the coupling of $|\uparrow\rangle$ to the $|J = 2, m_J = 1\rangle$ state will induce the state dipole moment $\langle\uparrow|\vec{d}_{\text{spin}}|\uparrow\rangle \sim d_{\text{spin}}\tilde{\Omega}/\tilde{\Delta}$. In turn, the non-zero dipole moment of the $|\uparrow\rangle$ states will give rise to non-zero matrix elements of the charge-dipole interaction $V^{mq}_{\mathbf{m_0},\uparrow;\tilde{\mathbf{m}},\uparrow} = V^{mq}_{\mathbf{ns},k_0\,\nu_0,\uparrow;\mathbf{ns},k_\nu,\uparrow}$, which in turn will give rise to terms $\sim V^{iq}_{\mathbf{m_0},\uparrow;\tilde{\mathbf{m}},\uparrow}\left(V^{mq}_{\mathbf{m_0},\uparrow;\tilde{\mathbf{m}},\uparrow}\right)^*/(\mathcal{E}_{\tilde{\mathbf{m}}} - \mathcal{E}_{\mathbf{m_0}})$ and $\sim \left|V^{iq}_{\mathbf{m_0},\uparrow;\tilde{\mathbf{m}},\uparrow}\right|^2/(\mathcal{E}_{\tilde{\mathbf{m}}} - \mathcal{E}_{\mathbf{m_0}})$ in the coefficients $J^{zz\ q,k_0\,\nu_0}_{im}$ and $b^{z\ q,k_0\,\nu_0}_i$, respectively, where $\tilde{\mathbf{m}} = |\mathbf{ns}, k'_{\nu'}\rangle$ and $\mathcal{E}_{\tilde{\mathbf{m}}} - \mathcal{E}_{\mathbf{m_0}} \sim \hbar^2(2\pi)^2/2m_{\text{Rb}}(N_{\text{at latt}}L_{\text{at}})^2 = 4E_{\text{rec}}/N^2_{\text{at latt}} \approx 0.87$ Hz for a Rb mediator atom. It shows that 1) the $V^{iq}_{\mathbf{m_0},\uparrow;\tilde{\mathbf{m}},\uparrow}$ matrix elements should be much smaller than the latter energy difference for the Schrieffer-Wolff expansion to be valid and 2) the resulting $J^{zz\ q,k_0\,\nu_0}_{im}$ and $b^{z\ q,k_0\,\nu_0}_i$ terms should be much smaller than $J^{\perp\ q,k_0\,\nu_0}_{im}$ in order to be neglected. Both of these requirements are indeed satisfied due to small values of the $V^{iq}_{\mathbf{m_0},\uparrow;\tilde{\mathbf{m}},\uparrow}$ matrix elements, as discussed in Appendix E, end of part A.

B. Ising interaction

Ising interaction can be realized in the bilayer system if the spin states $|\uparrow\rangle$, $|\downarrow\rangle$ are not coupled by a dipole-allowed transition, i.e. $\langle\uparrow|\vec{d}_{\text{spin}}|\downarrow\rangle = 0$, but at the same time have non-zero dipole moments $\langle\uparrow|\vec{d}_{\rm spin}|\uparrow\rangle \neq 0$, $\langle\downarrow|\vec{d}_{\rm spin}|\downarrow\rangle \neq 0$ (see Fig.2c,d). One can see from Eqs.(C.2)-(C.6) that in this case $J_{im}^{\perp\, q,k_\nu} = 0$ and $J_{im}^{zz\, q,k_\nu} \neq 0$, $b_i^{z\, q,k_\nu} \neq 0$, which describes the Ising interaction in the presence of a longitudinal magnetic field. Fig.2c shows an example of spin encoding in the polar molecule case, where $|\downarrow\rangle = |J=1, m_J=0\rangle$, $|\uparrow\rangle = |J=2, m_J=2\rangle$ spin states are not coupled by a dipole-allowed transition. The dipole moments in the states $|\uparrow\rangle$, $|\downarrow\rangle$ can be induced by MW fields, e.g. by coupling the $|\downarrow\rangle = |J=1, m_J=0\rangle$ to $|J=0, m_J=0\rangle$ and $|\uparrow\rangle = |J=2, m_J=2\rangle$ to $|J=3, m_J=3\rangle$ state by near-resonant MW fields (Fig.2c). An additional advantage of the MW dressing is that it shifts the energies of the dressed states with respect to near energy states, allowing for the $|\uparrow\rangle \leftrightarrow |\downarrow\rangle$ transition to be addressed spectroscopically. In the Rydberg atom spin encoding case the MW dressing can also induce non-zero spin state dipole moments, which can be done in the way, shown in Fig.2d.

C. XXZ interaction with Rb ($\tilde{n} = 50$) effective spins and Rb ($n = 100$) mediator atoms

There is a growing number of theoretical proposals and experimental demonstrations on simulation of many-body interacting systems using Rydberg atoms [7]. Below we discuss how the XXZ interaction can be realized in the case of effective spins encoded into long-lived states of Rydberg atoms, which interact indirectly via mediator Rydberg atoms. The XXZ interaction, in which both $J_{im}^{\perp}$ and J_{im}^{zz} are non-zero, can be realized if both transitional $\langle\uparrow|\vec{d}_{\rm spin}|\downarrow\rangle$ and state $\langle\uparrow|\vec{d}_{\rm spin}|\uparrow\rangle$ and/or $\langle\downarrow|\vec{d}_{\rm spin}|\downarrow\rangle$ dipole moments are non-zero and of comparable strength and the spin transition frequency is smaller or comparable to the mediator transition frequencies $E_{\rm spin} \lesssim |\mathcal{E}_{np_{j'}} - \mathcal{E}_{ns}|$. It can be seen then from Eqs.(C.2)-(C.6) that in this case $J_{im}^{\perp\, q,k_\nu} \sim J_{im}^{zz\, q,k_\nu}$. These requirements can be met by choosing the spin states to be nearly degenerate such as e.g. $|\uparrow\rangle = |J=1, m_J=1\rangle$, $|\downarrow\rangle = |J=1, m_J=0\rangle$ in the polar molecule case and $|\uparrow\rangle = \left|ns_{1/2}, m_j = 1/2\right\rangle$, $|\downarrow\rangle = \left|ns_{1/2}, m_j = -1/2\right\rangle$ in the Rydberg atom case, where the latter can be additionally split by a DC magnetic field (see Fig.2e,f). The spin states can acquire dipole moments if dressed with MW fields, nearly resonantly coupling them to states of opposite parity, e.g. in the way, shown in Fig.5a:

$$|\pm\rangle_\uparrow = a_\uparrow^\pm \left|\tilde{n}p_{1/2}, m_j = \frac{1}{2}\right\rangle + b_\uparrow^\pm \left|\tilde{n}s_{1/2}, m_j = \frac{1}{2}\right\rangle,$$
$$|\pm\rangle_\downarrow = a_\downarrow^\pm \left|\tilde{n}p_{1/2}, m_j = -\frac{1}{2}\right\rangle + b_\downarrow^\pm \left|\tilde{n}s_{1/2}, m_j = -\frac{1}{2}\right\rangle,$$

with

$$a_{\uparrow(\downarrow)}^\pm = \frac{\sqrt{\sqrt{\left(\frac{\Delta_{\uparrow(\downarrow)}}{2}\right)^2 + \Omega_{\uparrow(\downarrow)}^2} \pm \frac{\Delta_{\uparrow(\downarrow)}}{2}}}{\sqrt{2}\left(\left(\frac{\Delta_{\uparrow(\downarrow)}}{2}\right)^2 + \Omega_{\uparrow(\downarrow)}^2\right)^{1/4}},$$
$$b_{\uparrow(\downarrow)}^\pm = \pm\frac{\sqrt{\sqrt{\left(\frac{\Delta_{\uparrow(\downarrow)}}{2}\right)^2 + \Omega_{\uparrow(\downarrow)}^2} \mp \frac{\Delta_{\uparrow(\downarrow)}}{2}}}{\sqrt{2}\left(\left(\frac{\Delta_{\uparrow(\downarrow)}}{2}\right)^2 + \Omega_{\uparrow(\downarrow)}^2\right)^{1/4}}.$$

where $\Omega_{\uparrow(\downarrow)}$ and $\Delta_{\uparrow(\downarrow)}$ are the MW dressing fields Rabi frequencies and detunings. The $|+\rangle_\uparrow$ and $|+\rangle_\downarrow$ or $|-\rangle_\uparrow$ and $|-\rangle_\downarrow$ dressed states can be chosen as spin states. This encoding makes both the spin transition and spin states dipole moments to be non-zero and, by tuning the $a_{\uparrow,\downarrow}^\pm$, $b_{\uparrow,\downarrow}^\pm$ coefficients, of comparable strength: $\langle\uparrow|\vec{d}_{\rm spin}|\uparrow\rangle = -2a_\uparrow^\pm b_\uparrow^\pm \vec{e}_z d_{\tilde{n}p,\tilde{n}s}/3$, $\langle\downarrow|\vec{d}_{\rm spin}|\downarrow\rangle = 2a_\downarrow^\pm b_\downarrow^\pm d_{\tilde{n}p,\tilde{n}s}\vec{e}_z/3$ and $\langle\uparrow|\vec{d}_{\rm spin}|\downarrow\rangle = -(a_\uparrow^\pm b_\downarrow^\pm + b_\uparrow^\pm a_\downarrow^\pm)(\vec{e}_x - i\vec{e}_y)d_{\tilde{n}p,\tilde{n}s}/3$. Here $d_{\tilde{n}p,\tilde{n}s} = e\int_0^\infty R_{\tilde{n}p}(r)R_{\tilde{n}s}(r)r^3dr$ is the radial part of the dipole moment between the $\tilde{n}p$ and $\tilde{n}s$ states. We also assume that the $\tilde{n}s_{1/2}, m_j = \pm 1/2$ states are split by a DC magnetic field such that their splitting is much larger than the energy differences between the $|\pm\rangle_\uparrow$, $|\pm\rangle_\downarrow$ dressed states. Additionally, the effective interaction strength should exceed the decoherence rate of the system, given mainly by the lifetimes of the Rydberg states. The interaction can be made $\sim$ hundred kHz and faster than the decay, if the mediator atom transition frequency $(|\mathcal{E}_{n'p_{j'}} - \mathcal{E}_{ns}| \sim E_{\rm spin})/\hbar \lesssim$ hundreds MHz. These requirements can be met by using dressed mediator states instead of bare ones in the way shown in Fig.5b. One can initially prepare the mediator atom in the $\left|ns_{1/2}, m_j = -1/2\right\rangle$ state and then the closest in energy virtual states, to which it can be transferred from the initial state by the dipole-dipole interaction with a spin will be

$$|+\rangle_{\rm med} = c_+ \left|ns_{1/2}, m_j = \frac{1}{2}\right\rangle + d_+ \left|np_{1/2}, m_j = -\frac{1}{2}\right\rangle,$$
$$|-\rangle_{\rm med} = c_- \left|ns_{1/2}, m_j = \frac{1}{2}\right\rangle + d_- \left|np_{1/2}, m_j = -\frac{1}{2}\right\rangle.$$

Other possible virtually excited mediator states are separated by ~ 3.5 GHz and can be therefore neglected.

The $J_{im}^{\perp(zz)\, q,k_0\, \nu_0}$ coefficients and the effective magnetic field $b_i^{z\, q,k_0\, \nu_0}$ in this case will be given by the following expressions:

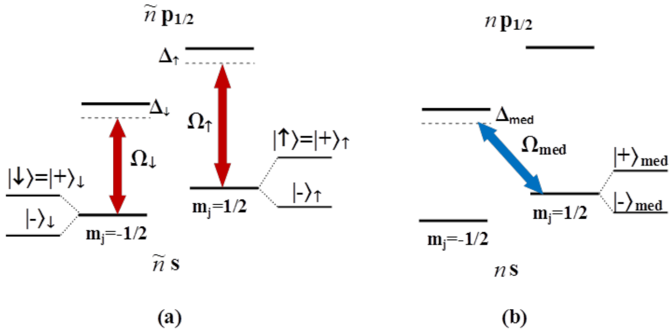

FIG. 5: *Spin encoding in MW dressed states of a Rydberg atom interacting with a MW dressed mediator Rydberg atom allows to realize the XXZ interaction.* (a) Spin-1/2 system can be encoded in long-lived $\left|\tilde{n}s_{1/2}, m_j = \pm 1/2\right\rangle$ Rydberg states dressed by MW fields near resonantly coupling them to $\left|\tilde{n}p_{1/2}, m_j = \pm 1/2\right\rangle$ states. The sublevels of the $\tilde{n}p_{1/2}$ and $\tilde{n}s_{1/2}$ states can be additionally split by a DC magnetic field to make the spin transition frequency different from frequencies between other dressed states; (b) The $J_{im}^{\perp}$ and J_{im}^{zz} interaction coefficients can be made $\sim$ hundred kHz by using mediator virtual transitions between the initial $\left|ns_{1/2}, m_j = -1/2\right\rangle$ and dressed by a MW field states $|\pm\rangle_{\rm med}$, which can be separated by $\lesssim$ hundreds MHz.

$$
\begin{aligned}
J_{im}^{\perp\; q,k_0\;\nu_0} &= \sum_{\mathbf{med}} -2\left(\frac{V^{iq}_{\mathbf{m_0}\uparrow;\mathbf{med},\downarrow}\left(V^{mq}_{\mathbf{m_0},\uparrow;\mathbf{med},\downarrow}\right)^*}{\mathcal{E}_{\mathbf{med}}-\mathcal{E}_{\mathbf{m_0}}-E_{\rm spin}} + \frac{V^{mq}_{\mathbf{m_0},\downarrow;\mathbf{med},\uparrow}\left(V^{iq}_{\mathbf{m_0},\downarrow;\mathbf{med},\uparrow}\right)^*}{\mathcal{E}_{\mathbf{med}}-\mathcal{E}_{\mathbf{m_0}}+E_{\rm spin}}\right),\\
J_{im}^{zz\; q,k_0\;\nu_0} &= \sum_{\mathbf{med}} -\frac{\left(V^{iq}_{\mathbf{m_0},\uparrow;\mathbf{med},\uparrow}-V^{iq}_{\mathbf{m_0},\downarrow;\mathbf{med},\downarrow}\right)\left(\left(V^{mq}_{\mathbf{m_0},\uparrow;\mathbf{med},\uparrow}\right)^*-\left(V^{mq}_{\mathbf{m_0},\downarrow;\mathbf{med},\downarrow}\right)^*\right)}{\mathcal{E}_{\mathbf{med}}-\mathcal{E}_{\mathbf{m_0}}} + \text{c.c.},\\
b_i^{z\; q,k_0\;\nu_0} &= \sum_{\substack{m\neq i\\ \mathbf{med}}} \frac{2\left(\left|V^{iq}_{\mathbf{m_0},\downarrow;\mathbf{med},\downarrow}\right|^2-\left|V^{iq}_{\mathbf{m_0},\uparrow;\mathbf{med},\uparrow}\right|^2\right)}{\mathcal{E}_{\mathbf{med}}-\mathcal{E}_{\mathbf{m_0}}}\\
&\quad -\frac{2\left|V^{iq}_{\mathbf{m_0},\uparrow;\mathbf{med},\downarrow}\right|^2}{\mathcal{E}_{\mathbf{med}}-\mathcal{E}_{\mathbf{m_0}}-E_{\rm spin}}+\frac{2\left|V^{iq}_{\mathbf{m_0},\downarrow;\mathbf{med},\uparrow}\right|^2}{\mathcal{E}_{\mathbf{med}}-\mathcal{E}_{\mathbf{m_0}}+E_{\rm spin}}+\\
&\quad +\left[\frac{\left(V^{mq}_{\mathbf{m_0},\uparrow;\mathbf{med},\uparrow}\left(V^{iq}_{\mathbf{m_0},\downarrow;\mathbf{med},\downarrow}\right)^*-V^{mq}_{\mathbf{m_0},\downarrow;\mathbf{med},\downarrow}\left(V^{iq}_{\mathbf{m_0},\uparrow;\mathbf{med},\uparrow}\right)^*\right)}{\mathcal{E}_{\mathbf{med}}-\mathcal{E}_{\mathbf{m_0}}}+\text{c.c.}\right].
\end{aligned}
\tag{22}
$$

where $|\mathbf{med}\rangle_q = \left|\text{med}, k'_{\nu'}\right\rangle_q$ and $|\text{med}\rangle = |\pm\rangle$.

For spins encoded in Rydberg states an additional complication arises from the fact that the direct dipole-dipole interaction will be larger than the effective one, even for not high $\tilde{n}$, and should be canceled. The direct interaction between the i$^{\rm th}$ and m$^{\rm th}$ spins placed in the x-z plane has the form:

$$
\begin{aligned}
\hat{V}_{\rm dd} &= \frac{\hat{\vec{d}}_i\hat{\vec{d}}_m - 3\left(\hat{\vec{d}}_i\vec{R}_{im}\right)\left(\hat{\vec{d}}_m\vec{R}_{im}\right)/R_{im}^2}{R_{im}^3} =\\
&= \frac{1}{2R_{im}^3}\left(\left(1-3\cos^2\theta\right)\left(\hat{d}_{i+}\hat{d}_{m-}+\hat{d}_{i-}\hat{d}_{m+}+2\hat{d}_{iz}\hat{d}_{mz}\right)+\right.\\
&\quad +\frac{3}{\sqrt{2}}\sin\theta\cos\theta\left(\hat{d}_{i+}\hat{d}_{mz}-\hat{d}_{i-}\hat{d}_{mz}+\hat{d}_{iz}\hat{d}_{m+}-\hat{d}_{iz}\hat{d}_{m-}\right)\\
&\quad \left. -\frac{3}{2}\sin^2\theta\left(\hat{d}_{i+}\hat{d}_{m+}+\hat{d}_{i-}\hat{d}_{m-}\right)\right),
\end{aligned}
$$

where θ is the angle between the vector $\vec{R}_{im}$ connecting the two dipoles and their quantization axis Z, $\hat{d}_{i\pm} = \mp(\hat{d}_{i\,x} \mp i\hat{d}_{i\,y})/\sqrt{2}$. This expression shows that the resonant interaction terms $\hat{d}_{i+}\hat{d}_{m-}$, $\hat{d}_{i-}\hat{d}_{m+}$, $\hat{d}_{iz}\hat{d}_{mz}$, connecting states with the same energies, can be cancelled by tilting the dipoles such that $\cos^2\theta = 1/3$, which corresponds to $\theta \approx 54.73^o$. The dipoles quantization axis can be set by applying a magnetic field along the Z axis, as shown in Fig.6.

As a concrete example we consider a 1D bi-layer setup shown in Fig.1, in which the effective spins are encoded in the dressed $|\uparrow\rangle = |+\rangle_\uparrow$, $|\downarrow\rangle = |+\rangle_\downarrow$ states of $\tilde{n} = 50$ of Rb, and a Rb superatom mediator state (or a single mediator Rb atom if interaction between Rydberg and ground state atoms is to be avoided [45]) initially prepared in the state Eq.(11) with the Rydberg atoms in the $\left|ns_{1/2}, m_j = -1/2\right\rangle$ state of $n = 100$. Assuming also that initially the mediator atoms are prepared in a BEC state with $k_0 = 0$, $\nu_0 = 1$, the $J_{im}^{\perp,zz} = \sum_{q=1}^{N_a} J_{im}^{\perp,zz\;q,k_0=0_{\nu_0=1}}/N_a$ and $b_i^z = \sum_{q=1}^{N_a} b_i^{z\;q,k_0=0_{\nu_0=1}}/N_a$ coefficients were calculated from Eqs.(22) using the $V_{\mathbf{m_0},\alpha;\mathbf{med},\beta}^{mq}$ matrix elements given in Appendix E, part B. The b_i^z coefficients can be minimized and the J_{im}^{zz} coefficients can be maximized at the same time by setting $a_{\uparrow,\downarrow}^+ = b_{\uparrow,\downarrow}^+ = c_\pm = d_\pm = 1/\sqrt{2}$, which can be realized by using dressing microwave fields resonant to the corresponding transitions. In this case $V_{\mathbf{m_0},\uparrow;\mathbf{med},\uparrow}^{mq} = -V_{\mathbf{m_0},\downarrow;\mathbf{med},\downarrow}^{mq}$, and only the second and third terms in $b_i^{z\;q,k_0\,\nu_0}$ are non-zero.

The numerically calculated $J_{im}^{\perp(zz)}$ and b_i^z coefficients are shown in Fig.7. One can see that similar to the case of XX interaction considered in the previous subsection, the XXZ interaction i) changes sign with the interspin distance. The nearest neighbor interaction is ferro- and the next nearest neighbor and more distant ones are antiferromagnetic; ii) extends beyond nearest neighbors and falls off at the distances $|i-m| \sim 5$; (iii) interaction strengths $|J_{im}^\perp|, |J_{im}^{zz}| \sim$ hundred kHz can be realized; iv) b_i^z do not depend on spin position i. In the general case the interaction is of XXZ type, but tuning the spin and mediator transition frequencies by adjusting the corresponding MW dressing fields Rabi frequencies it can be made of symmetric Heisenberg type with $J_{im}^\perp = J_{im}^{zz}$ for chosen im spin neighbors. In particular, in Fig.7 the nearest neighbor interaction with $|i-m| = 1$ is of the Heisenberg type, and the next-nearest-neighbor and more distant interaction terms are of XXZ type. This case can be realized with the following parameters: spin and mediator lattice periods $L_{\rm spin} = L_{\rm at} = 7\ \mu$m, $\rho = 7\ \mu$m, $E_{ns_{1/2},m_j=1/2} - E_{ns_{1/2},m_j=-1/2} = E_{\tilde{n}s_{1/2},m_j=1/2} - E_{\tilde{n}s_{1/2},m_j=-1/2} = 148.5$ MHz, $E_{\rm spin} = E_\uparrow - E_\downarrow = 150$ MHz, the energy splittings of the mediator states $E_+ - E_{ns_{1/2},m_j=-1/2} = 151.155$ MHz, $E_- - E_{ns_{1/2},m_j=-1/2} = 145.845$ MHz, the spin and mediator states dressing fields Rabi frequencies and detunings are $\Omega_\uparrow = 2.5$ MHz, $\Omega_\downarrow = 1$ MHz, $\Omega_{\rm med} = 2.655$ MHz, and $\Delta_\uparrow = \Delta_\downarrow = \Delta_{\rm med} = 0$, respectively.

The contribution of the next-nearest and more distant neighbors can be controlled by preparing initial motional state of the mediator atoms as a superposition of Bloch states Eq.(11). Assuming the atoms prepared in a Gaussian distribution of quasimomenta of initial Bloch states in the lowest Bloch band with $|c_{k_0\;\nu_0=1}|^2 \sim e^{-k_0^2/\kappa_0^2}$, the strength of the next-nearest and more distant relative to the nearest neighbor interaction one can be controlled by the quasimomenta distribution width κ_0, as shown in Fig.8. In particular, for $\kappa_0/(\pi/L_{\rm at}) \approx 0.65$ the $J_{im}^{\perp,zz} \ll |J_{m+1,m}^{\perp,zz}|$ for $2 \le |i-m| \le 5$, resulting in the Heisenberg model with nearest neighbor interactions. In the range $0.3 \lesssim \kappa_0/(\pi/L_{\rm at}) \lesssim 0.65$ the $J_{m+3,m}^\perp/J_{m+2,m}^\perp \lesssim 0.2$ and $J_{m+2,m}^\perp/|J_{m+1,m}^\perp| \le 1$ and the interaction can be approximated as the $J_1 - J_2$ XXZ model $\sum_{l=1,2} J_l\left(\hat{S}_i^x\hat{S}_{i+l}^x + \hat{S}_i^y\hat{S}_{i+l}^y + \Delta_l\hat{S}_i^z\hat{S}_{i+l}^z\right)$ with $J_1 = J_{m+1,m}^\perp < 0$, $J_2 = J_{m+2,m}^\perp > 0$, $\Delta_1 = 1$ and $\Delta_2 \approx 0.27$. The $J_1 - J_2$ XXZ model with both $\Delta_1 = \Delta_2$ and $\Delta_1 \ne \Delta_2$ attracts significant interest due to its relevance for description of spin-1/2 frustrated antiferromagnetic copper oxide spin chain compounds such as $LiCu_2O_2$ [19], $NaCu_2O_2$ [20], $PbCuSO_4(OH)_2$ [21], $LiCuSbO_4$ [22], etc. Numerical analysis of the phase diagram of this model has shown [26] that in the case $\Delta_1 = \Delta_2$ for $J_1/J_2 < -4$ the system is in the ferromagnetic state, for $-4 < J_1/J_2 < -2.5$ it is in the vector chiral phase, and for $-2.5 < J_1/J_2 < -0.5$ in the Haldane dimer state, i.e. there are two critical points denoting the transitions between the phases. In the 1D copper oxide spin chains the ratios J_1/J_2 are fixed at certain values, while in our setup the ratio J_1/J_2 can be set between ≈ -1 to $-\infty$ (keeping the J_3 and J_4 small) by initially preparing the mediator atoms in the superposition of Bloch states with a certain width κ_0 of the quasimomenta distribution. The $J_1 - J_2$ XXZ model does not have an exact solution yet, and the bi-layer setup could potentially allow to study its phases, in particular, near quantum critical points.

The effective spins encoded into Rydberg states have finite lifetimes due to decay by spontaneous emission and interaction with black-body radiation. For the effective interactions to be observable their magnitude should exceed the spin states decay rates. The $50s_{1/2}$ state of Rb has lifetimes $\tau_{50s_{1/2}} = 141.3\ \mu$s at $T = 4.2$ K and $65.2\ \mu$s at $T = 300$ K, and the $50p_{1/2}$ state has lifetimes $\tau_{50p_{1/2}} = 257.4\ \mu$s at $T = 4.2$ K and $86.5\ \mu$s at $T = 300$ K [52]. Since the spin states are equal superpositions of the $50s_{1/2}$ and $50p_{1/2}$ states their decay rates will be an average of the s and p decay rates: $(1/\tau_{50s_{1/2}} + 1/\tau_{50p_{1/2}})/2$, giving the averaged spin states lifetimes $\approx 182.4\ \mu$s at $T = 4.2$ K and $\approx 74.4\ \mu$s at room temperature. The effective XXZ interaction strengths ~ 50 kHz correspond to the interaction times $\sim 2\ \mu$s, which is more than an order of magnitude smaller than the averaged spin state decay times, making the effective interaction observable.

Higher-order truncated terms in the expansion

Eq.(5) will result in an error of the phase accumulated by the effective interaction, which needs to be estimated. The nearest higher-order interaction which will bring the mediator atom back to the initial ns state is $\sim \mathcal{O}(\hat{S}^3\hat{V}) \sim |\hat{V}|^4/|\Delta E|^3$ because as shown in Eq.(A.5) $|\hat{S}| \sim |\hat{V}|/|\Delta E|$. The effective interaction coefficients (21), (22) are $J_{im}^{\perp,zz} \sim |\hat{V}|^2/|\Delta E|$, which gives the order of $\sim \mathcal{O}(J_{im}^{\perp,zz}|\hat{V}|^2/|\Delta E|^2)$ for the truncated terms. As was discussed in the previous paragraph, the finite lifetimes of ~ 100 μs of the mediator Rydberg states will allow to realize $\sim 10^2$ effective interaction cycles. It means that the decoherence rate due to the higher-order truncated terms will be smaller than the one due to mediator decay if $|\hat{V}|/|\Delta E| \leq 0.1$. This condition is satisfied in the examples analyzed above. In particular, for the NaCs+Rb(99s$_{1/2}$) system the interaction matrix elements $|V^{iq}_{\mathbf{m_0},\alpha;\mathbf{m}',\beta}| \sim$ 10 kHz, and the ratio $|V|/|\Delta E| \sim 10^{-2}$ for the energy defect $\Delta E = 1.76$ MHz; for the spin encoding in Rydberg states considered in this section the interaction matrix elements $|V^{iq}_{\mathbf{m_0},\alpha;\mathbf{med},\beta}| \sim$ 100 kHz, giving $|V|/|\Delta E| \sim 0.1$ for $|\Delta E| \sim$ 1 MHz. The finite $|\hat{V}|/|\Delta E|$ will also result in the readout error, when the state of the combined system will be measured at the end of the simulation. The readout will be assuming that the mediator is in the initial ns state, but there will be small population on the order of $\sim |\hat{V}|^2/|\Delta E|^2$ in the virtually excited np state. Both the Hamiltonian truncation induced decoherence rate and the readout error can be reduced by increasing the energy defect $|\Delta E|$ or reducing the spin-mediator interaction strength $|\hat{V}|$ by using a larger spin-mediator array distance ρ.

Finally, we come back to the direct dipole-dipole interaction between spin states. Tilting of the spin dipoles allows one to cancel the resonant direct dipole-dipole interaction which, for the case of $\theta = 0$, when the quantization axis is perpendicular to $\vec{R}_{im}$, would be of the order $V_{\rm dd} \sim 2d^2_{\tilde{n}p,\tilde{n}s}/9R^3_{im} \sim 11.5$ MHz, which is three orders of magnitude larger than the effective interaction with strengths $\sim$ hundred kHz. The non-resonant parts of the direct dipole-dipole interaction $\sim d_{i\pm}d_{mz}, d_{iz}d_{m\pm}$ and $\sim d_{i\pm}d_{m\pm}$ are, however, not cancelled and are of the order of $d^2_{\tilde{n}p,\tilde{n}s}/9\sqrt{2}R^3_{im} \sim 4.1$ MHz and $d^2_{\tilde{n}p,\tilde{n}s}/9R^3_{im} \sim 5.7$ MHz, respectively, but they are more than an order of magnitude smaller compared to the spin transition frequency $E_{\rm spin}/\hbar = 100$ MHz, leading to the probability of the spin changing its state due to the direct dipole-dipole interaction being $< 10^{-2}$.

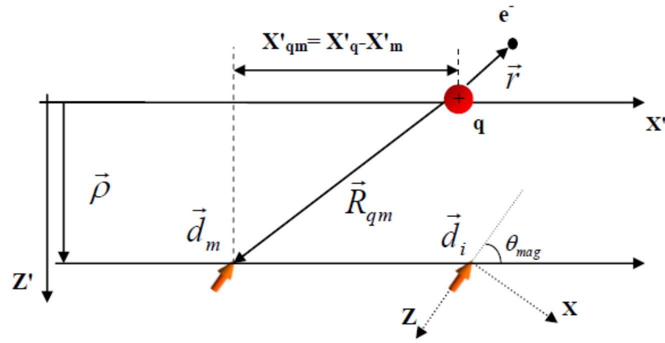

FIG. 6: *Tilting the effective spins with respect to the axis of the spin array allows to cancel direct dipole-dipole interaction in the case Rydberg atom spin encoding.* In the case of spin encoding into long-lived Rydberg states the direct dipole-dipole interaction between the effective spins can be canceled by tilting the spin quantization axis Z with respect to the line, connecting the spins. For the angle $\theta = \arccos(1/\sqrt{3})$ between the $-Z$ and X' axes the resonant part of the direct dipole-dipole interaction is zero.

IV. CONCLUSIONS

We propose a platform for simulating indirect spin-spin interactions based on polar molecules and/or Rydberg atoms trapped in two parallel 1D optical lattices. The effective spin-1/2 systems are encoded in rotational states of polar molecules or long-lived Rydberg states of ultracold atoms, which are tightly trapped. The interaction between effective spins is mediated by Rydberg atoms in a parallel shallow lattice to allow for mediator atom motional state to be delocalized and interact simultaneously with several effective spins. The effective interaction is therefore realized via direct charge-dipole (dipole-dipole) spin-mediator interactions with polar molecule (Rydberg atom) spin encoding. By a particular choice of spin-encoding states XX, Ising and XXZ spin-spin interaction types are realized, with $J^{\perp}$, J^{zz} interaction coefficients sign changing with interspin distance analogous to the RKKY interaction. The interactions extend beyond nearest neighbors and can reach magnitudes $\sim$ 100's kHz, limited by the trapping frequency of the spins optical lattice.

The bi-layer setup allows to control the strengths of the next nearest and more distant neighbor relative to the nearest neighbor interaction by initially preparing the mediator atoms in a superposition of motional Bloch states with a specific distribution (e.g. a Gaussian) of quasimomenta. Additionally, the Rabi frequencies and detuning of spin and mediator MW dressing fields, can be tuned to engineer symmetric Heisenberg interactions for selected pairs of neighbors, e.g. for nearest neighbors. In the XX model with spin encoding into rotational states of polar molecules an external DC magnetic field can be used to control the relative strengths of interaction between distant neighbors with respect to the

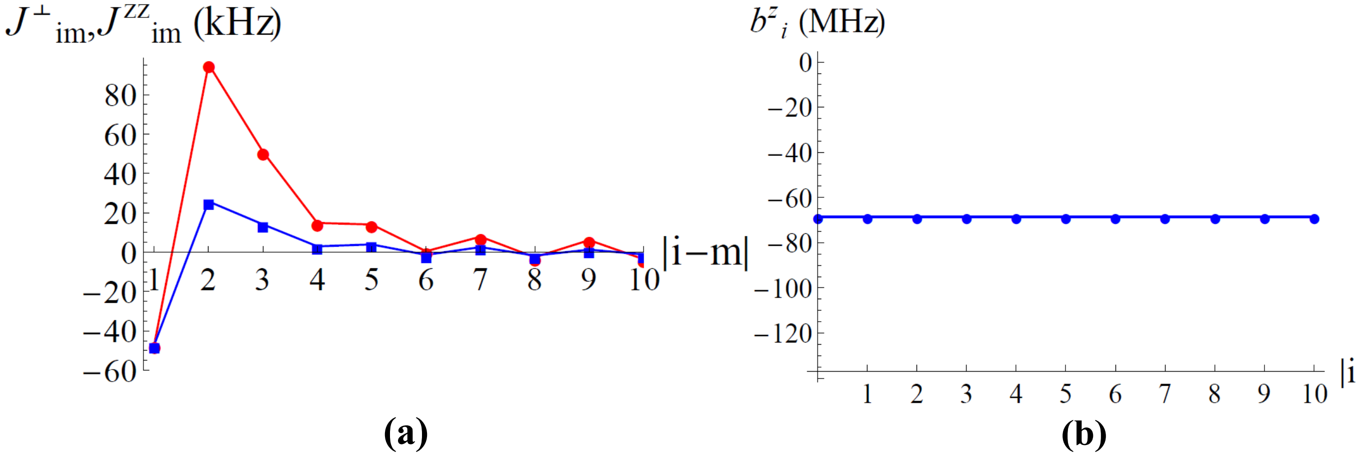

FIG. 7: *Interaction coefficients in the case of spin encoding into long-lived Rydberg states, allowing to realize XXZ interaction*: (a) $J^{\perp}_{im}$ (circles, red curve), J^{zz}_{im} (squares, blue curve) and (b) effective magnetic field b^z_i for spins encoded in $|\uparrow\rangle = |+\rangle_{\uparrow} = (|\tilde{n}p_{1/2}, m_j = 1/2\rangle + |\tilde{n}s_{1/2}, m_j = 1/2\rangle)/\sqrt{2}$, $|\downarrow\rangle = |+\rangle_{\downarrow} = (|\tilde{n}p_{1/2}, m_j = -1/2\rangle + |\tilde{n}s_{1/2}, m_j = -1/2\rangle)/\sqrt{2}$ states of Rb with $\tilde{n} = 50$. The mediator atoms are initially prepared in a superatom state (11) in the $|100s_{1/2}, m_j = -1/2\rangle$ internal state and a BEC motional state with $k_0 = 0$, $\nu_0 = 1$. In calculations of $J^{\perp,zz}_{im}$, b^z_i of Eqs.(22) quasimomenta in the first Brillouin zone were summed for $\nu = 1, ..., 5$ lowest Bloch bands of the mediator atom lattice. Setup dimensions: inter-layer distance $\rho = 7$ μm, spin and mediator lattice periods $L_{\mathrm{spin}} = L_{\mathrm{at}} = 7$ μm. The calculations were done for a spin in the center of the array $m = 0$. Other parameters were as follows: $E_{ns_{1/2},m_j=1/2} - E_{ns_{1/2},m_j=-1/2} = E_{\tilde{n}s_{1/2},m_j=1/2} - E_{\tilde{n}s_{1/2},m_j=-1/2} = 148.5$ MHz, $\Omega_{\uparrow} = 2.5$ MHz, $\Omega_{\downarrow} = 1$ MHz, $\Omega_{\mathrm{med}} = 2.655$ MHz, $\Delta_{\uparrow} = \Delta_{\downarrow} = \Delta_{\mathrm{med}} = 0$, resulting in $E_{+\ \mathrm{med}} - E_{ns_{1/2},m_j=-1/2} = 151.155$ MHz, $E_{-\ \mathrm{med}} - E_{ns_{1/2},m_j=-1/2} = 145.845$ MHz.

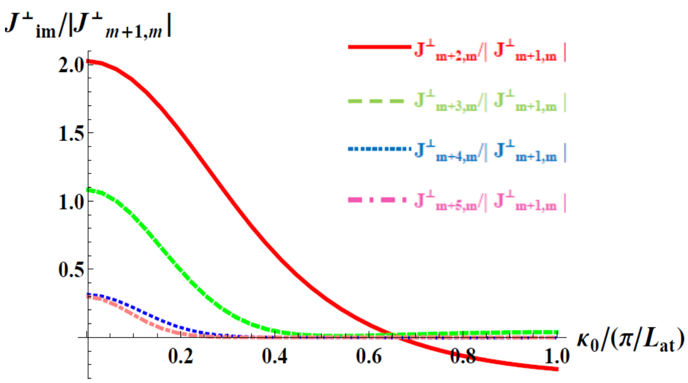

FIG. 8: *In the XXZ model with interaction coefficients Eqs.(14),(22) the strengths of next-nearest and more distant neighbors relative to the nearest neighbor interaction can be controlled by the width of the quasimomenta distribution of the initial superposition of Bloch states of mediator atoms.* Assuming the atoms initially prepared in the lowest Bloch band with the Gaussian initial distribution $|c_{k_0\ \nu_0=1}|^2 \sim e^{-k_0^2/\kappa_0^2}$ of quasimomenta, the ratios of interaction coefficients $J^{\perp}_{im}/|J^{\perp}_{m+1,m}|$ can be controlled by the distribution width κ_0: $J^{\perp}_{m+2,m}/|J^{\perp}_{m+1,m}|$ (red solid curve), $J^{\perp}_{m+3,m}/|J^{\perp}_{m+1,m}|$ (green dashed curve), $J^{\perp}_{m+4,m}/|J^{\perp}_{m+1,m}|$ (blue dotted curve), $J^{\perp}_{m+5,m}/|J^{\perp}_{m+1,m}|$ (pink dashed-dotted curve). The interaction coefficients shown in Fig.7 correspond to $\kappa_0 = 0$.

nearest neighbor one.

The bi-layer system can be extended to 2D geometries to simulate not only the Heisenberg/XXZ models, but also the indirect Dzyaloshinskii-Moriya (DM) anisotropic spin-spin interaction, which can also be mediated by conduction electrons [53, 54], provided spin-orbit interaction between internal and motional states of mediator atoms can be incorporated. In this case the DM vector $\vec{D}$ has both the magnitude and the orientation oscillating with an interspin distance, which can produce chiral magnetic states with spatially oscillating chirality.

We note an interesting analogy between the XX model, considered in Section III, with the interaction coefficients given by Eqs.(21), with the Cook model [55], which is the generalized Hopfield model of associative memory [56], describing a system of N interacting neurons, encoding p different patterns. The Cook model is predicted to have a phase transition at a certain storage capacity p/N between a self-organized phase, when the stored patterns can be reliably retrieved, and a spin glass.

V. ACKNOWLEDGMENTS

The authors are grateful for fruitful discussions with S. Gopalakrishnan, D. Podolsky and R. Mukherjee. E.K.

and I.B. thank the Russian Science Foundation (Grant No. 16-12-00028) in the part of derivation of indirect spin-spin interaction in the bi-layer of Rydberg atoms. E.K. is grateful for financial support from the Office of Naval Research (Award No. N00014-16-1-3054) and Robert A. Welch Foundation (Grant No. A-1261). S.F.Y. would like to thank the National Science Foundation through the CUA Center grant. S.T.R. acknowledges support from NSF Grant No. PHY-1516421 and funding from the Research Corporation for Science Advancement. H.R.S. was supported by a grant from the NSF to ITAMP.

Appendix A

Effective spins encoded in rotational states of polar molecules will interact with the mediator Rydberg atoms via charge-dipole interaction [57]. The spin-mediator interaction Hamiltonian in this case can be written in the combined basis of spin and mediator states:

$$\begin{aligned}\hat{V} = \sum_{i=1}^{N}\sum_{q=1}^{N_a}\sum_{\substack{\mathbf{m},\mathbf{m}',\tilde{\mathbf{m}},\mathbf{m}''\\ \alpha,\beta=\uparrow,\downarrow}} &|\mathbf{m}\rangle_q |\alpha_i\rangle\langle\beta_i| \langle\mathbf{m}'|_q \times \\ &\times \langle\alpha_i| \langle\mathbf{m}|_q \hat{V} |\mathbf{m}'\rangle_q |\beta_i\rangle + \\ &+ |\mathbf{m}\rangle_q |\alpha_i\rangle\langle\beta_i| \langle\tilde{\mathbf{m}}|_q \times \\ &\times \langle\alpha_i| \langle\mathbf{m}|_q \hat{V} |\tilde{\mathbf{m}}\rangle_q |\beta_i\rangle + \\ &+ |\mathbf{m}'\rangle_q |\alpha_i\rangle\langle\beta_i| \langle\mathbf{m}''|_q \times \\ &\times \langle\alpha_i| \langle\mathbf{m}'|_q \hat{V} |\mathbf{m}''\rangle_q |\beta_i\rangle + \text{H.c.},\end{aligned} \tag{A.1}$$

where $\hat{V} = \hat{V}_{\text{cd}}$, and $|\mathbf{m}\rangle_q = |\mathbf{ns}, k_\nu\rangle_q$, $|\tilde{\mathbf{m}}\rangle_q = |\mathbf{ns}, k'_{\nu'}\rangle_q$, and $|\mathbf{m}'\rangle_q = |\mathbf{np}', k'_{\nu'}\rangle_q$, $|\mathbf{m}''\rangle_q = |\mathbf{np}'', k''_{\nu''}\rangle_q$ and the sums over the index vectors are restricted to $\mathbf{ns} = \{ns, j = 1/2, m_j = \pm 1/2\}$, $\mathbf{np}' = \{np, j' = 3/2, 1/2, m'_{j'} = \pm 3/2, \pm 1/2\}$,and $\mathbf{np}'' = \{np, j'' = 3/2, 1/2, m''_{j''} = \pm 3/2, \pm 1/2\}$ for a fixed radial quantum number n. The summation is over j, m_j, j', m'_j and j'', m''_j quantum numbers, and over k, k', k'' quasimomenta of ν, ν' and ν'' Bloch bands. At spin-mediator distances comparable to the mediator Rydberg electron orbit radius the charge-dipole interaction can couple electronic states of the same parity, i.e. the interaction matrix elements $\langle\mathbf{m}|_q \hat{V} |\tilde{\mathbf{m}}\rangle_q$ and $\langle\mathbf{m}'|_q \hat{V} |\mathbf{m}''\rangle_q$ can be nonzero.

In order to show how the spin-mediator interaction Eq.(A.1) gives rise to indirect interaction between the effective spins, the interaction Hamiltonian can be written in the basis of two-spin states $|\alpha_i\beta_m\rangle$ as follows:

$$\begin{aligned}\hat{V} = \sum_{i,m=1}^{N}\sum_{q=1}^{N_a}\sum_{\substack{\alpha,\beta,\gamma,\delta\\ \mathbf{m},\mathbf{m}',\tilde{\mathbf{m}},\mathbf{m}''}} &\Big[|\mathbf{m}\rangle_q |\alpha_i\beta_m\rangle \\ &\times \Big(V^{iq}_{\mathbf{m},\alpha;\mathbf{m}',\gamma}\delta_{\beta_m,\delta_m} + \\ &+ V^{mq}_{\mathbf{m}\beta;\mathbf{m}',\delta}\delta_{\alpha_i,\gamma_i}\Big) \langle\gamma_i\delta_m| \langle\mathbf{m}'|_q + \text{H.c.}\Big] + \\ &+ \Big[|\mathbf{m}\rangle_q |\alpha_i\beta_m\rangle \Big(V^{iq}_{\mathbf{m},\alpha;\tilde{\mathbf{m}},\gamma}\delta_{\beta_m,\delta_m} + \\ &+ V^{mq}_{\mathbf{m},\beta;\tilde{\mathbf{m}},\delta}\delta_{\alpha_i,\gamma_i}\Big) \langle\gamma_i\delta_m| \langle\tilde{\mathbf{m}}|_q + \text{H.c.}\Big] + \\ &+ \Big[|\mathbf{m}'\rangle_q |\alpha_i\beta_m\rangle \Big(V^{iq}\end{aligned} \tag{A.2}$$

Terms in the first square bracket involve the interaction matrix elements between the m$^{\text{th}}$ spin and the q$^{\text{th}}$ mediator atom $V^{mq}_{\mathbf{m},\xi;\mathbf{m}',\eta} = \langle\mathbf{m}|_q \langle\xi_m| \hat{V} |\eta_m\rangle |\mathbf{m}'\rangle_q$, which describes the process in which the q^{th} mediator atom is transferred from the $|\mathbf{m}\rangle$ to the $|\mathbf{m}'\rangle$ state, and the m^{th} spin goes from the $|\xi\rangle$ to the $|\eta\rangle$ state, i.e. it describes the interaction in which a mediator atom changes parity of its electronic state $|ns, m_j\rangle \leftrightarrow |np_{j'}, m'_{j'}\rangle$. The terms in the second and third square brackets involve the interaction matrix elements $V^{mq}_{\mathbf{m},\xi;\tilde{\mathbf{m}},\eta} = \langle\mathbf{m}|_q \langle\xi_m| \hat{V} |\eta_m\rangle |\tilde{\mathbf{m}}\rangle_q$ and $V^{mq}_{\mathbf{m}',\xi;\mathbf{m}'',\eta} = \langle\mathbf{m}'|_q \langle\xi_m| \hat{V} |\eta_m\rangle |\mathbf{m}''\rangle_q$ and describe the mediator being transferred to electronic states of the same parity $|ns, m_j\rangle \leftrightarrow |ns, m_j\rangle$ and $|np_j, m_j\rangle \leftrightarrow |np_{j''}, m''_{j''}\rangle$, respectively, which is allowed by the charge-dipole interaction. The parity conserving interaction terms are present only in the polar molecule spin encoding setup and can be neglected for sufficiently large spin-mediator distances, allowing for the charge-dipole interaction to be approximated by the dipole-dipole one, because at these distances the matrix elements of the charge-dipole and dipole-dipole interactions differ by a very small amount, as is discussed in Appendix E, end of part A.

The $\hat{V}$ terms can give rise to second order energy shifts, having a form of an indirect interaction between the effective spins. This can be shown via the Schrieffer-Wolff transformation of the total Hamiltonian $\hat{H} = \hat{H}_0 + \hat{V}$:

$$e^{\hat{S}}\hat{H}e^{-\hat{S}} = \hat{H} + \left[\hat{S}, \hat{H}\right] + \frac{\left[\hat{S}, \left[\hat{S}, \hat{H}\right]\right]}{2} + O\left(\hat{S}^2\hat{V}\right)$$

in such a way that $\left[\hat{S}, \hat{H}_0\right] = -\hat{V}$, giving as a result the transformed Hamiltonian

$$e^{\hat{S}}\hat{H}e^{-\hat{S}} = \hat{H}_0 + \frac{\left[\hat{S}, \hat{V}\right]}{2} + O\left(|\hat{V}|^3\right), \tag{A.4}$$

where the generator $\hat{S}$ has the form:

$$\hat{S}=\sum_{i,m=1}^{N}\sum_{q=1}^{N_a}\sum_{\substack{\alpha,\beta,\gamma,\delta=\uparrow,\downarrow\\ \mathbf{m},\mathbf{m}',\tilde{\mathbf{m}},\mathbf{m}''}}\Bigg[-\frac{|\mathbf{m}\rangle_q|\alpha_i\beta_m\rangle V^{iq}_{\mathbf{m},\alpha;\mathbf{m}',\gamma}\delta_{\beta_m,\delta_m}\langle\gamma_i\delta_m|\langle\mathbf{m}'|_q}{\mathcal{E}_{\mathbf{m}'}-\mathcal{E}_{\mathbf{m}}+E_\gamma-E_\alpha}-\frac{|\mathbf{m}\rangle_q|\alpha_i\beta_m\rangle V^{mq}_{\mathbf{m},\beta;\mathbf{m}',\delta}\delta_{\alpha_i,\gamma_i}\langle\gamma_i\delta_m|\langle\mathbf{m}'|_q}{\mathcal{E}_{\mathbf{m}'}-\mathcal{E}_{\mathbf{m}}+E_\delta-E_\beta}+$$
$$+\frac{|\mathbf{m}\rangle_q|\alpha_i\beta_m\rangle V^{iq}_{\mathbf{m},\alpha;\tilde{\mathbf{m}},\gamma}\delta_{\beta_m,\delta_m}\langle\gamma_i\delta_m|\langle\tilde{\mathbf{m}}|_q}{\mathcal{E}_{\tilde{\mathbf{m}}}-\mathcal{E}_{\mathbf{m}}+E_\gamma-E_\alpha}+\frac{|\mathbf{m}\rangle_q|\alpha_i\beta_m\rangle V^{mq}_{\mathbf{m},\beta;\tilde{\mathbf{m}},\delta}\delta_{\alpha_i,\gamma_i}\langle\gamma_i\delta_m|\langle\tilde{\mathbf{m}}|_q}{\mathcal{E}_{\tilde{\mathbf{m}}}-\mathcal{E}_{\mathbf{m}}+E_\delta-E_\beta}+$$
$$+\frac{|\mathbf{m}'\rangle_q|\alpha_i\beta_m\rangle V^{iq}_{\mathbf{m}',\alpha;\mathbf{m}'',\gamma}\delta_{\beta_m,\delta_m}\langle\gamma_i\delta_m|\langle\mathbf{m}''|_q}{\mathcal{E}_{\mathbf{m}''}-\mathcal{E}_{\mathbf{m}'}+E_\gamma-E_\alpha}+\frac{|\mathbf{m}'\rangle_q|\alpha_i\beta_m\rangle V^{mq}_{\mathbf{m}',\beta;\mathbf{m}'',\delta}\delta_{\alpha_i,\gamma_i}\langle\gamma_i\delta_m|\langle\mathbf{m}''|_q}{\mathcal{E}_{\mathbf{m}''}-\mathcal{E}_{\mathbf{m}'}+E_\delta-E_\beta}\Bigg]-\text{H.c..}\tag{A.5}$$

TABLE II: Notations for internal and motional mediator states

$\lvert\mathbf{m}\rangle_q=\lvert\mathbf{ns},k_\nu\rangle_q$
$\lvert\tilde{\mathbf{m}}\rangle_q=\lvert\mathbf{ns},k'_{\nu'}\rangle_q$
$\lvert\mathbf{m}'\rangle_q=\lvert\mathbf{np}',k'_{\nu'}\rangle_q$
$\lvert\mathbf{m}''\rangle_q=\lvert\mathbf{np}'',k''_{\nu''}\rangle_q$
$\lvert\mathbf{med}\rangle_q=\lvert\text{med},\text{k}'_{\nu'}\rangle_q$
$\mathbf{ns}=\{ns,j=1/2,m_j=\pm1/2\}$
$\mathbf{np}'=\{np,j'=1/2,3/2,m'_{j'}=\pm1/2,\pm3/2\}$
$\mathbf{np}''=\{np,j''=1/2,3/2,m''_{j''}=\pm1/2,\pm3/2\}$

Below we list notations for all internal and motional states of the mediator atoms used in the paper:

Appendix B

In this section we give the $K^{q,k_\nu}_{\alpha_i,\beta_m;\gamma_i,\delta_m}$ coefficients of Eq.(7):

$$K^{q,k_\nu}_{\uparrow\uparrow,\uparrow\uparrow\ im}=\sum_{\xi,\eta=i,m}\sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'}(-1)^l\left[\frac{V^{\xi q}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}\left(V^{\eta q}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}\right)^*}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}}+\frac{\left|V^{\xi q}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow}\right|^2}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}-E_\text{spin}}\right],\tag{B.1}$$

$$K^{q,k_\nu}_{\downarrow\downarrow,\downarrow\downarrow\ im}=\sum_{\xi,\eta=i,m}\sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'}(-1)^l\left[\frac{V^{\xi q}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\left(V^{\eta q}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\right)^*}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}}+\frac{\left|V^{\xi q}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow}\right|^2}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}+E_\text{spin}}\right],\tag{B.2}$$

$$K^{q,k_\nu}_{\uparrow\downarrow,\uparrow\downarrow\ im}=\sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'}(-1)^l\left[\frac{\left|V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}+V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\right|^2}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}}+\frac{\left|V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow}\right|^2}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}-E_\text{spin}}+\frac{\left|V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow}\right|^2}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}+E_\text{spin}}\right],\tag{B.3}$$

$$K^{q,k_\nu}_{\downarrow\uparrow,\downarrow\uparrow\ im}=\sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'}(-1)^l\left[\frac{\left|V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}+V^{mq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}\right|^2}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}}+\frac{\left|V^{mq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow}\right|^2}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}-E_\text{spin}}+\frac{\left|V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow}\right|^2}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}+E_\text{spin}}\right],\tag{B.4}$$

$$K^{q,k_\nu}_{\uparrow\downarrow,\downarrow\uparrow\ im}=\sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'}(-1)^l\left[\frac{V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow}\left(V^{mq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow}\right)^*}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}-E_\text{spin}}+\frac{V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow}\left(V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow}\right)^*}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}+E_\text{spin}}\right],\tag{B.5}$$

$$K^{q,k_\nu}_{\uparrow\uparrow,\uparrow\downarrow\;im} = \sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'} (-1)^l \left[\frac{\left(V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow} + V^{mq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}\right)}{2} \left(V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow}\right)^* \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} + E_\text{spin}} \right) + \right.$$
$$\left. + \frac{V^{mq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow} \left(\left(V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}\right)^* + \left(V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\right)^* \right)}{2} \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} - E_\text{spin}} \right) \right]$$
$$- \frac{V^{mq}_{\mathbf{m},\uparrow;\mathbf{m},\downarrow} \left(\left(V^{iq}_{\mathbf{m},\uparrow;\mathbf{m},\uparrow}\right)^* + \left(V^{mq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow}\right)^* \right)}{E_\text{spin}} + \frac{\left(V^{iq}_{\mathbf{m},\uparrow;\mathbf{m},\uparrow} + V^{mq}_{\mathbf{m},\uparrow;\mathbf{m},\uparrow}\right) \left(V^{mq}_{\mathbf{m},\downarrow;\mathbf{m},\uparrow}\right)^*}{E_\text{spin}}, \tag{B.6}$$

$$K^{q,k_\nu}_{\uparrow\uparrow,\downarrow\uparrow\;im} = \sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'} (-1)^l \left[\frac{\left(V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow} + V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\right)}{2} \left(V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow}\right)^* \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} - E_\text{spin}} \right) + \right.$$
$$\left. + \frac{V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow} \left(\left(V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}\right)^* + \left(V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\right)^* \right)}{2} \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} + E_\text{spin}} \right) \right] +$$
$$+ \frac{V^{iq}_{\mathbf{m},\downarrow;\mathbf{m},\uparrow} \left(\left(V^{iq}_{\mathbf{m},\uparrow;\mathbf{m},\uparrow}\right)^* + \left(V^{mq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow}\right)^* \right)}{E_\text{spin}} - \frac{\left(V^{iq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow} + V^{mq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow}\right) \left(V^{iq}_{\mathbf{m},\uparrow;\mathbf{m},\downarrow}\right)^*}{E_\text{spin}}, \tag{B.7}$$

$$K^{q,k_\nu}_{\downarrow\downarrow,\uparrow\downarrow\;im} = \sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'} (-1)^l \left[\frac{\left(V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow} + V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\right)}{2} \left(V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow}\right)^* \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} - E_\text{spin}} \right) + \right.$$
$$\left. + \frac{V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow} \left(\left(V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}\right)^* + \left(V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\right)^* \right)}{2} \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} + E_\text{spin}} \right) \right] +$$
$$+ \frac{V^{iq}_{\mathbf{m},\downarrow;\mathbf{m},\uparrow} \left(\left(V^{iq}_{\mathbf{m},\uparrow;\mathbf{m},\uparrow}\right)^* + \left(V^{mq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow}\right)^* \right)}{E_\text{spin}} - \frac{\left(V^{iq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow} + V^{mq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow}\right) \left(V^{iq}_{\mathbf{m},\uparrow;\mathbf{m},\downarrow}\right)^*}{E_\text{spin}}, \tag{B.8}$$

$$K^{q,k_\nu}_{\downarrow\downarrow,\downarrow\uparrow\;im} = \sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'} (-1)^l \left[\frac{\left(V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow} + V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\right)}{2} \left(V^{mq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow}\right)^* \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} - E_\text{spin}} \right) + \right.$$
$$\left. + \frac{V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow} \left(\left(V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\right)^* + \left(V^{mq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}\right)^* \right)}{2} \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} + E_\text{spin}} \right) \right] +$$
$$+ \frac{V^{mq}_{\mathbf{m},\downarrow;\mathbf{m},\uparrow} \left(\left(V^{iq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow}\right)^* + \left(V^{mq}_{\mathbf{m},\uparrow;\mathbf{m},\uparrow}\right)^* \right)}{E_\text{spin}} - \frac{\left(V^{iq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow} + V^{mq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow}\right) \left(V^{mq}_{\mathbf{m},\uparrow;\mathbf{m},\downarrow}\right)^*}{E_\text{spin}}, \tag{B.9}$$

$$K^{q,k_\nu}_{\uparrow\uparrow,\downarrow\downarrow\;im} = \sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'} (-1)^l \left[\frac{V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow} \left(V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow}\right)^*}{2} \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} - E_\text{spin}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} + E_\text{spin}} \right) + \right.$$
$$\left. + \frac{V^{mq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow} \left(V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow}\right)^*}{2} \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} - E_\text{spin}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} + E_\text{spin}} \right) \right], \tag{B.10}$$

$$K^{q,k_\nu}_{\alpha\beta,\gamma\delta;im} = \left(K^{q,k_\nu}_{\gamma\delta,\alpha\beta;im}\right)^*. \tag{B.11}$$

where l is the orbital quantum number of the $\mathcal{M}$ state.

Appendix C

The $|\alpha_i\beta_m\rangle\langle\gamma_i\delta_m|$ can be expressed via the two spin-1/2 variables $\hat{S}_i^{\pm,z}\hat{S}_m^{\pm,z}$ using relations

$$\begin{aligned}
|{\uparrow_i}{\uparrow_m}\rangle\langle{\uparrow_i}{\uparrow_m}| &= \left(\frac{1}{2}+\hat{S}_i^z\right)\left(\frac{1}{2}+\hat{S}_m^z\right) = \frac{1}{4}+\frac{1}{2}\left(\hat{S}_i^z+\hat{S}_m^z\right)+\hat{S}_i^z\hat{S}_m^z,\\
|{\downarrow_i}{\downarrow_m}\rangle\langle{\downarrow_i}{\downarrow_m}| &= \left(\frac{1}{2}-\hat{S}_i^z\right)\left(\frac{1}{2}-\hat{S}_m^z\right) = \frac{1}{4}-\frac{1}{2}\left(\hat{S}_i^z+\hat{S}_m^z\right)+\hat{S}_i^z\hat{S}_m^z,\\
|{\downarrow_i}{\uparrow_m}\rangle\langle{\downarrow_i}{\uparrow_m}| &= \left(\frac{1}{2}-\hat{S}_i^z\right)\left(\frac{1}{2}+\hat{S}_m^z\right) = \frac{1}{4}-\frac{1}{2}\left(\hat{S}_i^z-\hat{S}_m^z\right)-\hat{S}_i^z\hat{S}_m^z,\\
|{\uparrow_i}{\downarrow_m}\rangle\langle{\uparrow_i}{\downarrow_m}| &= \left(\frac{1}{2}+\hat{S}_i^z\right)\left(\frac{1}{2}-\hat{S}_m^z\right) = \frac{1}{4}+\frac{1}{2}\left(\hat{S}_i^z-\hat{S}_m^z\right)-\hat{S}_i^z\hat{S}_m^z,\\
|{\uparrow_i}{\downarrow_m}\rangle\langle{\downarrow_i}{\uparrow_m}| &= \hat{S}_i^+\hat{S}_m^-,\\
|{\uparrow_i}{\uparrow_m}\rangle\langle{\uparrow_i}{\downarrow_m}| &= \left(\frac{1}{2}+\hat{S}_i^z\right)\hat{S}_m^+,\\
|{\uparrow_i}{\uparrow_m}\rangle\langle{\downarrow_i}{\uparrow_m}| &= \hat{S}_i^+\left(\frac{1}{2}+\hat{S}_m^z\right),\\
|{\downarrow_i}{\downarrow_m}\rangle\langle{\uparrow_i}{\downarrow_m}| &= \hat{S}_i^-\left(\frac{1}{2}-\hat{S}_m^z\right),\\
|{\downarrow_i}{\downarrow_m}\rangle\langle{\downarrow_i}{\uparrow_m}| &= \left(\frac{1}{2}-\hat{S}_i^z\right)\hat{S}_m^-,\\
|{\uparrow_i}{\uparrow_m}\rangle\langle{\downarrow_i}{\downarrow_m}| &= \hat{S}_i^+\hat{S}_m^+,
\end{aligned} \tag{C.1}$$

and other states can be obtained using $|\gamma_i\delta_m\rangle\langle\alpha_i\beta_m| = (|\alpha_i\beta_m\rangle\langle\gamma_i\delta_m|)^\dagger$.

The interaction coefficients are given by the following expressions:

$$\begin{aligned}
J_{im}^{zz\ q,k_\nu} &= K^{q,k_\nu}_{\uparrow\uparrow,\uparrow\uparrow\ im}+K^{q,k_\nu}_{\downarrow\downarrow,\downarrow\downarrow\ im}-K^{q,k_\nu}_{\downarrow\uparrow,\downarrow\uparrow\ im}-K^{q,k_\nu}_{\uparrow\downarrow,\uparrow\downarrow\ im}=\\
&=\sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'}(-1)^l\left[\frac{\left(V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}-V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\right)\left(\left(V^{mq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}\right)^*-\left(V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\right)^*\right)}{\mathcal{E}_{\mathcal{M}}-\mathcal{E}_{\mathbf{m}}}\right]+\text{c.c.},
\end{aligned} \tag{C.2}$$

$$J_{im}^{+-\ q,k_\nu} = K^{q,k_\nu}_{\uparrow\downarrow,\downarrow\uparrow\ im} = -\sum_{\mathbf{m}'}\left(\frac{V^{iq}_{\mathbf{m},\uparrow;\mathbf{m}',\downarrow}\left(V^{mq}_{\mathbf{m},\uparrow;\mathbf{m}',\downarrow}\right)^*}{\mathcal{E}_{\mathbf{m}'}-\mathcal{E}_{\mathbf{m}}-E_{\text{spin}}}+\frac{V^{mq}_{\mathbf{m},\downarrow;\mathbf{m}',\uparrow}\left(V^{iq}_{\mathbf{m},\downarrow;\mathbf{m}',\uparrow}\right)^*}{\mathcal{E}_{\mathbf{m}'}-\mathcal{E}_{\mathbf{m}}+E_{\text{spin}}}\right), \tag{C.3}$$

$$
\begin{aligned}
J_{im}^{z+\ q,k_\nu} = K^{q,k_\nu}_{\uparrow\uparrow,\uparrow\downarrow\ im} - K^{q,k_\nu}_{\downarrow\uparrow,\downarrow\downarrow\ im} = \sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'} (-1)^l \Bigg[V^{mq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow} \times \\
\times \left(\left(V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow} \right)^* - \left(V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow} \right) \right) \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} - E_\text{spin}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} \right) + \\
+ \left(V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow} - V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow} \right) \left(V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow} \right)^* \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} + E_\text{spin}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} \right) \Bigg] \\
- \frac{V^{mq}_{\mathbf{m},\uparrow;\mathbf{m},\downarrow} \left(\left(V^{iq}_{\mathbf{m},\uparrow;\mathbf{m},\uparrow} \right)^* - \left(V^{iq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow} \right)^* \right)}{E_\text{spin}} + \frac{\left(V^{iq}_{\mathbf{m},\uparrow;\mathbf{m},\uparrow} - V^{iq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow} \right) \left(V^{mq}_{\mathbf{m},\downarrow;\mathbf{m},\uparrow} \right)^*}{E_\text{spin}} \\
J_{im}^{++\ q,k_\nu} = K^{q,k_\nu}_{\uparrow\uparrow,\downarrow\downarrow\ im}, \\
J_{im}^{z-\ q,k_\nu} = \left(J_{im}^{z+\ q,k_\nu} \right)^*, \\
J_{im}^{+z\ q,k_\nu} = J_{mi}^{z+\ q,k_\nu}, \\
J_{im}^{-z\ q,k_\nu} = J_{mi}^{z-\ q,k_\nu},
\end{aligned}
\tag{C.4}
$$

$$
\begin{aligned}
b_{im}^{z\ q,k_\nu} = K^{q,k_\nu}_{\uparrow\uparrow,\uparrow\uparrow\ im} - K^{q,k_\nu}_{\downarrow\downarrow,\downarrow\downarrow\ im} + K^{q,k_\nu}_{\uparrow\downarrow,\uparrow\downarrow\ im} - K^{q,k_\nu}_{\downarrow\uparrow,\downarrow\uparrow\ im} = \\
= \sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'} (-1)^l \left[\frac{2\left| V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow} \right|^2}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} - E_\text{spin}} - \frac{2\left| V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow} \right|^2}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} + E_\text{spin}} + \frac{2\left(\left| V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow} \right|^2 - \left| V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow} \right|^2 \right)}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} + \right. \\
\left. + \frac{\left(V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow} - V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow} \right) \left(\left(V^{mq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow} \right)^* + \left(V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow} \right)^* \right)}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} + \text{c.c.} \right],
\end{aligned}
\tag{C.5}
$$

$$
\begin{aligned}
b_{im}^{+\ q,k_\nu} = K^{q,k_\nu}_{\uparrow\uparrow,\downarrow\uparrow\ im} + K^{q,k_\nu}_{\uparrow\downarrow,\downarrow\downarrow\ im} = \sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'} (-1)^l \left[\frac{\left(V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow} + V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow} \right)}{2} \times \right. \\
\times \left(V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow} \right)^* \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} - E_\text{spin}} \right) + \\
\left. + \frac{V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow} \left(\left(V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow} \right)^* + \left(V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow} \right)^* \right)}{2} \left(\frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m}} + \frac{1}{\mathcal{E}_\mathcal{M} - \mathcal{E}_\mathbf{m} + E_\text{spin}} \right) + \text{c.c.} \right] + \\
+ \left[\frac{V^{iq}_{\mathbf{m},\downarrow;\mathbf{m},\uparrow} \left(\left(V^{iq}_{\mathbf{m},\uparrow;\mathbf{m},\uparrow} \right)^* + \left(V^{mq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow} \right)^* \right)}{E_\text{spin}} - \frac{\left(V^{iq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow} + V^{mq}_{\mathbf{m},\downarrow;\mathbf{m},\downarrow} \right) \left(V^{iq}_{\mathbf{m},\uparrow;\mathbf{m},\downarrow} \right)^*}{E_\text{spin}} + \text{c.c.} \right],
\end{aligned}
\tag{C.6}
$$

$$b_{0\ im}^{q,k_\nu} = \frac{1}{4}\left(K^{q,k_\nu}_{\uparrow\uparrow,\uparrow\uparrow\ im} + K^{q,k_\nu}_{\downarrow\downarrow,\downarrow\downarrow\ im} + K^{q,k_\nu}_{\uparrow\downarrow,\uparrow\downarrow\ im} + K^{q,k_\nu}_{\downarrow\uparrow,\downarrow\uparrow\ im}\right) =$$

$$= \sum_{\xi,\eta=i,m}\sum_{\mathcal{M}=\tilde{\mathbf{m}},\mathbf{m}'}\frac{(-1)^l}{4}\left[\frac{V^{\xi q}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}\left(V^{\eta q}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}\right)^*}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}} + \right.$$

$$+\frac{V^{\xi q}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\left(V^{\eta q}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\right)^*}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}} + \frac{\left|V^{\xi q}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow}\right|^2}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}-E_{\text{spin}}} + \frac{\left|V^{\xi q}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow}\right|^2}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}+E_{\text{spin}}} +$$

$$+\frac{\left|V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}+V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}\right|^2}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}} + \frac{\left|V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\downarrow}+V^{mq}_{\mathbf{m},\uparrow;\mathcal{M},\uparrow}\right|^2}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}} +$$

$$\left.+\frac{\left(\left|V^{iq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow}\right|^2+\left|V^{mq}_{\mathbf{m},\uparrow;\mathcal{M},\downarrow}\right|^2\right)}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}-E_{\text{spin}}} + \frac{\left(\left|V^{iq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow}\right|^2+\left|V^{mq}_{\mathbf{m},\downarrow;\mathcal{M},\uparrow}\right|^2\right)}{\mathcal{E}_\mathcal{M}-\mathcal{E}_\mathbf{m}+E_{\text{spin}}}\right], \quad \text{(C.7)}$$

where l is the orbital quantum number of the $\mathcal{M}$ state.

Appendix D

The Hamiltonian, acting only on effective spins can be obtained by taking the expectation value of Eq.(10) with respect to an unperturbed initial state of the mediator atoms. As a first example we consider the mediator atoms prepared in a Rydberg $|\mathbf{ns}, k_{0\ \nu_0}\rangle$ superatom state:

$$|\Psi\rangle_{\text{sat}} = \sum_{q=1}^{N_a}\sum_{\substack{k',k_0\\ \nu',\nu_0}}\prod_{q'\neq q}\frac{c_{k'_{\nu'},k_{0\ \nu_0}}}{\sqrt{N_a}}\phi_{g_{q'}}\left(X_{q'},k'_{\nu'\ q'}\right)$$
$$\times\,\Phi_{ns_q}\left(X_q,k_{0\ \nu_0\ q}\right)\left|g_1,...(ns_{1/2},m_j)_q,...,g_{N_a}\right\rangle, \quad \text{(D.1)}$$

where $\phi_{g_{q'}}\left(X_{q'},k'_{\nu'\ q'}\right)$ is the spatial wave function of a q'$^{\text{th}}$ atom in the ground state; $\Phi_{ns_q}\left(X_q,k_{0\ \nu_0\ q}\right)$ is the spatial wave function of the q$^{\text{th}}$ atom in the $|ns\rangle$ state; in the general case the atoms in the ground and Rydberg states are assumed to be prepared in a wave packet of Bloch states with quasimomenta $k'_{\nu'}$ and $k_{0\ \nu_0}$, respectively, weighted by the coefficients $c_{k'_{\nu'},k_{0\ \nu_0}}$. In this case the spin Hamiltonian takes the form:

$$\hat{V}^{ns}_{\text{eff spin}} = \langle\Psi_{\text{sat}}|\,\hat{V}^{ns}_{\text{eff}}\,|\Psi_{\text{sat}}\rangle = \quad \text{(D.2)}$$
$$= \sum_{i,m=1}^{N}\left(J^{zz}_{im}\hat{S}^z_i\hat{S}^z_m + \frac{J^\perp_{im}}{2}\left(\hat{S}^+_i\hat{S}^-_m+\hat{S}^-_i\hat{S}^+_m\right) + \right.$$
$$\left. + b^z_{im}n_m\hat{S}^z_i + b_{0\ im}n_in_m\right),$$

with the averaged interaction coefficients:

$$J^{zz(\perp)}_{im} = \frac{1}{N_a}\sum_{q=1}^{N_a}\sum_{\substack{k,k',k_0,k'_0\\ \nu,\nu',\nu_0,\nu'_0}} c_{k'_{\nu'},k'_{0\ \nu'_0}}\left(c_{k'_{\nu'},k_{0\ \nu_0}}\right)^*\times$$
$$\times J^{zz(\perp)\ q,k_\nu}_{im}\int dX_q\Phi^*_{ns_q}(X_q,k_{0\ \nu_0\ q})\phi(X_q,k_{\nu\ q})\times$$
$$\times\int dX_q\Phi_{ns_q}(X_q,k'_{0\ \nu'_0\ q})\phi^*(X_q,k_{\nu\ q}), \quad \text{(D.3)}$$

where $\phi(X_q,k_{\nu\ q})$ is the spatial part of the mediator atom wave function in the $\hat{P}_{ns}$ projector Eq.(8). The averaged effective magnetic field b^z_{im} and $b_{0\ im}$ satisfy the same relation.

Next, we use the assumption that initially the mediator atoms are prepared in a superposition of Bloch states and write explicitly Bloch functions as the spatial parts of the mediator atom wavefunction $\Phi_{ns_q}(X_q,k_{0\ \nu_0\ q}) = u^{(\nu_0)}_{k_0}e^{ik_0X_q}$, $\phi(X_q,k_{\nu\ q}) = u^{(\nu)}_k e^{ikX_q}$. In this case $\int\Phi^*_{ns_q}(X_q,k_{0\ \nu_0\ q})u^{(\nu)}_k e^{ikX_q} = \delta_{k,k_0}\delta_{\nu,\nu_0}$, giving the averaged interaction coefficients (same for b^z_{im}, $b_{0\ im}$):

$$J^{zz(\perp)}_{im} = \frac{1}{N_a}\sum_{q=1}^{N_a}\sum_{\substack{k',k_0\\ \nu',\nu_0}}|c_{k'_{\nu'},k_{0\ \nu_0}}|^2 J^{zz(\perp)\ q,k_{0\ \nu_0}}_{im} =$$
$$= \frac{1}{N_a}\sum_{q=1}^{N_a}\sum_{k_0,\nu_0}|c_{k_{0\ \nu_0}}|^2 J^{zz(\perp)\ q,k_{0\ \nu_0}}_{im}, \quad \text{(D.4)}$$

where $|c_{k_{0\ \nu_0}}|^2 = \sum_{k',\nu'}|c_{k'_{\nu'},k_{0\ \nu_0}}|^2$. In particular, for mediator atoms initially prepared in a stationary BEC $k_0 = 0$, $\nu_0 = 1$ the averaged interaction coefficients are $J^{zz(\perp)}_{im} = \frac{1}{N_a}\sum_{q=1}^{N_a}J^{zz(\perp)\ q,k_0=0_{\nu_0=1}}_{im}$ [37]. In a more general case the initial superatom state is a superposition of Bloch states with quasimomenta k_0 and Bloch bands ν_0, determined by the distribution $|c_{k_{0\ \nu_0}}|^2$.

Another possible initial mediator state is the Rydberg dressed state

$$|\Psi\rangle_{\rm dress} = \prod_{q=1}^{N_a}\sum_{\substack{k',k_0\\ \nu',\nu_0}} c_{k'_{\nu'},k_{0\ \nu_0}}\Big(c_g\phi_{g_q}(X_q,k'_{\nu'\ q})\,|g\rangle_q + +c_{ns}\Phi_{ns_q}(X_q,k_{0\ \nu_0\ q})\,|\mathbf{ns}\rangle_q\Big), \tag{D.5}$$

created when all mediator atoms interact with a dressing laser field of Rabi frequency Ω and detuning Δ from the Rydberg state, and $c_g = \sqrt{\sqrt{\Delta^2/4+\Omega^2}+\Delta/2}/[\sqrt{2}\left(\Delta^2/4+\Omega^2\right)^{1/4}]$, $c_{ns} = \sqrt{\sqrt{\Delta^2/4+\Omega^2}-\Delta/2}/[\sqrt{2}\left(\Delta^2/4+\Omega^2\right)^{1/4}]$. Here $\phi_{g_q}(X_q,k_{\nu\ q})$ is the spatial wave function of a q$^{\rm th}$ atom in the ground state; $\Phi_{ns_q}(X_q,k_{0\ \nu_0\ q})$ is the spatial wave function of the q$^{\rm th}$ atom in the Rydberg ns state.

In this case the spin Hamiltonian takes the same form as Eq.(D.2) with the averaged interaction coefficients (the same for b^z_{im}, $b_{0\ im}$):

$$J^{zz(\perp)}_{im} = |c_{ns}|^2\sum_{q=1}^{N_a}\sum_{\substack{k,k',k_0,k'_0\\ \nu,\nu',\nu_0,\nu'_0}} c_{k'_{\nu'},k'_{0\ \nu'_0}}(c_{k'_{\nu'},k_{0\ \nu_0}})^*\times \times J^{zz(\perp)\ q,k_\nu}_{im}\int dX_q\Phi^*_{ns_q}(X_q,k_{0\ \nu_0\ q})\phi(X_q,k_{\nu\ q})\times \times\int dX_q\Phi_{ns_q}(X_q,k'_{0\ \nu'_0\ q})\phi^*(X_q,k_{\nu\ q}), \tag{D.6}$$

where we again consider a case when initially the ground and Rydberg state atoms are prepared in a superposition of Bloch states determined by the weights $|c_{k'_{\nu'},k_{0\ \nu_0}}|^2$. Plugging the Bloch functions for the Rydberg and ground states motional wavefunctions, their averages will be given by the following expression:

$$J^{zz(\perp)}_{im} = |c_{ns}|^2\sum_{q=1}^{N_a}\sum_{\substack{k',k_0\\ \nu',\nu_0}}|c_{k'_{\nu'},k_{0\ \nu_0}}|^2 J^{zz(\perp)\ q,k_{0\ \nu_0}}_{im} = |c_{ns}|^2\sum_{q=1}^{N_a}\sum_{k_0,\nu_0}|c_{k_{0\ \nu_0}}|^2 J^{zz(\perp)\ q,k_{0\ \nu_0}}_{im}. \tag{D.7}$$

In particular, for the case of an initial BEC with $k_0=0$, $\nu_0=1$ we have

$$J^{zz(\perp)}_{im} = |c_{ns}|^2\sum_{q=1}^{N_a} J^{zz(\perp)\ q,k_0=0_{\nu_0=1}}_{im}.$$

The expressions (D.3), (D.6) are also valid in the case of a single mediator atom corresponding to $N_a=1$.

Appendix E

A. Calculation of $V^{mq}_{\mathbf{m_0},\alpha;\mathbf{m'},\beta}$ matrix elements for charge-dipole interaction

The interaction matrix elements for the charge-dipole interaction between an $m^{\rm th}$ spin, encoded in polar molecule rotational states, and a $q^{\rm th}$ mediator Rydberg atom have the form

$$V^{mq}_{\mathbf{m_0},\alpha;\mathbf{m'},\beta} = \left\langle ns_{1/2},m_j,k_{0\ \nu_0}\right|_q\langle\alpha|_m\hat{V}_{\rm cd}\,|\beta\rangle_m\left|np_{j'},m'_{j'},k_\nu\right\rangle_q = = -e\,\langle\alpha|_m\,\vec{d}_{\rm spin}\,|\beta\rangle_m\int dX_q\Phi^*_{ns\ q}(X_q,k_{0\ \nu_0\ q})\,\langle ns,m_j|\,\frac{\vec{R}_{qm}-\vec{r}}{\left|\vec{R}_{qm}-\vec{r}\right|^3}\,\left|np_{j'},m'_{j'}\right\rangle\phi(X_q,k_{\nu\ q}) = = -e\,\langle\alpha|_m\,\vec{d}_{\rm spin}\,|\beta\rangle_m\,e^{-i(k-k_0)X_m}\int dX_{qm}u^{(\nu_0)*}_{k_0}(X_m+X_{qm})\,\langle ns,m_j|\,\frac{\vec{R}_{qm}-\vec{r}}{\left|\vec{R}_{qm}-\vec{r}\right|^3}\,\left|np_{j'},m'_{j'}\right\rangle u^{(\nu)}_k(X_m+X_{qm})e^{-i(k-k_0)X_{qm}} = = c^{mq}_{\mathbf{m_0},\alpha;\mathbf{m'}\beta}e^{-i(k-k_0)X_m}. \tag{E.1}$$

Assuming for concreteness that the mediator atom is initially excited to the $\left|ns_{1/2},m_j=1/2\right\rangle$ state, the matrix elements Eq.(E.1) will be non-zero for virtual excitations only to the $m'_j=\pm1/2,3/2$ sublevels of the $np_{j'}=99p_{3/2}$ state, which can be expanded in terms of the l,m_l states as follows:

$$
\begin{aligned}
\left|np_{3/2}, m_j = 3/2\right\rangle &= \left|n, l=1, m_l=1; s=\frac{1}{2}, m_s=\frac{1}{2}\right\rangle, \\
\left|np_{3/2}, m_j = \frac{1}{2}\right\rangle &= \sqrt{\frac{2}{3}}\left|n, l=1, m_l=0; s=\frac{1}{2}, m_s=\frac{1}{2}\right\rangle + \sqrt{\frac{1}{3}}\left|n, l=1, m_l=1; s=\frac{1}{2}, m_s=-\frac{1}{2}\right\rangle, \\
\left|np_{3/2}, m_j = -\frac{1}{2}\right\rangle &= \sqrt{\frac{1}{3}}\left|n, l=1, m_l=-1; s=\frac{1}{2}, m_s=\frac{1}{2}\right\rangle + \sqrt{\frac{2}{3}}\left|n, l=1, m_l=0; s=\frac{1}{2}, m_s=-\frac{1}{2}\right\rangle,
\end{aligned} \tag{E.2}
$$

which can be used to calculate $c^{qm}_{\mathbf{m_0},\alpha;\mathbf{m'},\beta}$ coefficients.

The interaction coefficient corresponding to the resonance between the $\left|99p_{3/2}, m_j = 1/2\right\rangle \leftrightarrow \left|99s_{1/2}, m_j = 1/2\right\rangle$ transition of the q$^{\text{th}}$ atom and the spin transition of the m$^{\text{th}}$ molecule are given by the expression

$$
\begin{aligned}
c^{mq}_{\mathbf{m_0},\uparrow;\mathbf{m'},\downarrow} &= \\
= \frac{e\sqrt{2}\left\langle\uparrow\right|\vec{d}_{\text{spin}}\left|\downarrow\right\rangle_m}{\sqrt{3}} &\int dX_{qm}\left\langle ns\right|\frac{\vec{R}_{qm}-\vec{r}}{|\vec{R}_{qm}-\vec{r}|^3}\left|np, m_l=0\right\rangle \times \\
&\times u^{(\nu_0)*}_{k_0}\left(X_m+X_{qm}\right)u^{(\nu)}_{k}\left(X_m+X_{qm}\right)e^{-i(k-k_0)X_{qm}},
\end{aligned} \tag{E.3}
$$

where the Bloch functions are normalized as $\int\phi^*(X_q,k_{\nu\ q})\phi(X_q,k'_{\nu'\ q})dX_q = \delta_{\nu,\nu'}\delta_{k,k'}$. Let us first analyze the integral over Rydberg electron's coordinates:

$$
I^{mq}_{ns;np,0} = -e\left\langle\uparrow\right|\vec{d}_{\text{spin}}\left|\downarrow\right\rangle_m\left\langle ns\right|\frac{\vec{R}_{qm}-\vec{r}}{\left|\vec{R}_{qm}-\vec{r}\right|^3}\left|np, m_l=0\right\rangle.
$$

For the effective spin states $\left|\downarrow\right\rangle = \left|J=0, m_J=0\right\rangle$ and $\left|\uparrow\right\rangle = \left|J=1, m_J=0\right\rangle$ the spin dipole moment has only the z component $\left\langle\uparrow\right|\vec{d}_{\text{spin}}\left|\downarrow\right\rangle = \vec{e}_z\left\langle\uparrow\right|d_{\text{z spin}}\left|\downarrow\right\rangle$. One then can use the following expression [58]:

$$
\begin{aligned}
-e\left\langle\uparrow\right|d_{\text{z spin}}\left|\downarrow\right\rangle_m\frac{\left(\vec{R}_{qm}-\vec{r}\right)_z}{\left|\vec{R}_{qm}-\vec{r}\right|^3} &= 4\pi e\left\langle\uparrow\right|d_{\text{z spin}}\left|\downarrow\right\rangle_m\cos\eta\times \\
\times\begin{cases}\sum_{l''=0}^{\infty}-\frac{l''+1}{2l''+1}\frac{r^{l''}}{R_{qm}^{l''+2}}\sum_{m''=-l''}^{l''}Y_{l''}^{m''}(\theta,\phi)Y_{l''}^{m''\ *}(\eta,\chi) & \text{for } r<R_{qm}\\ \sum_{l''=0}^{\infty}\frac{l''}{2l''+1}\frac{R_{qm}^{l''-1}}{r^{l''+1}}\sum_{m''=-l''}^{l''}Y_{l''}^{m''}(\theta,\phi)Y_{l''}^{m''\ *}(\eta,\chi) & \text{for } r>R_{qm}\end{cases} \\
-4\pi e\left\langle\uparrow\right|d_{\text{z spin}}\left|\downarrow\right\rangle_m\frac{\sin\eta}{R_{qm}}\times \\
\times\begin{cases}\sum_{l''=0}^{\infty}\frac{1}{2l''+1}\frac{r^{l''}}{R_{qm}^{l''+1}}\sum_{m''=-l''}^{l''}Y_{l''}^{m''}(\theta,\phi)\frac{\partial Y_{l''}^{m''\ *}(\eta,\chi)}{\partial\eta} & \text{for } r<R_{qm}\\ \sum_{l''=0}^{\infty}\frac{1}{2l''+1}\frac{R_{qm}^{l''}}{r^{l''+1}}\sum_{m''=-l''}^{l''}Y_{l''}^{m''}(\theta,\phi)\frac{\partial Y_{l''}^{m''\ *}(\eta,\chi)}{\partial\eta} & \text{for } r>R_{qm}\end{cases},
\end{aligned} \tag{E.4}
$$

where θ and ϕ are the Rydberg electron's angular coordinates with respect to the ionic core, η and χ are the angular coordinates of the core-molecule vector $\vec{R}_{qm}$ with respect to the quantization axis, chosen to be perpendicular to the spin and mediator lattices and parallel to $\vec{\rho}$ (see Fig.9). When calculating the matrix element $I^{mq}_{ns;np,0}$ between the $\left|ns\right\rangle$ and $\left|np, m_l=0\right\rangle$ states, there will be integrals over three spherical harmonics, expressed via 3j-symbols

$$
\begin{aligned}
\int_0^{2\pi}d\phi\int_0^{\pi}\sin\theta d\theta Y_0^{0\ *}Y_{l''}^{m''}Y_1^0 &= \\
= \sqrt{\frac{3(2l''+1)}{4\pi}}\begin{pmatrix}0 & l'' & 1\\ 0 & 0 & 0\end{pmatrix}&\begin{pmatrix}0 & l'' & 1\\ 0 & m'' & 0\end{pmatrix},
\end{aligned} \tag{E.5}
$$

which are non-zero only for $l''=1, m''=0$, with the corresponding integral given by $\int_0^{2\pi}d\phi\int_0^{\pi}\sin\theta d\theta Y_0^{0\ *}Y_1^0Y_1^0 = 1/\sqrt{4\pi}$.

As a result,

$$I^{mq}_{ns;np,0} = 4\pi e \langle\uparrow| d_{\text{z spin}} |\downarrow\rangle_m \cos\eta Y_1^{0\,*}(\eta,\chi)\left(-\frac{2}{3\sqrt{4\pi}R_{qm}^3}\int_0^{R_{qm}} r^3 R_{ns}R_{np}dr + \frac{1}{3\sqrt{4\pi}}\int_{R_{qm}}^{\infty} R_{ns}R_{np}dr\right)$$
$$-4\pi e \langle\uparrow| d_{\text{z spin}} |\downarrow\rangle_m \frac{\sin\eta}{R_{qm}}\frac{\partial Y_1^{0\,*}}{\partial\eta}\left(\frac{1}{3\sqrt{4\pi}R_{qm}^2}\int_0^{R_{qm}} r^3 R_{ns}R_{np}dr + \frac{R_{qm}}{3\sqrt{4\pi}}\int_{R_{qm}}^{\infty} R_{ns}R_{np}dr\right),$$

where $\frac{\partial Y_1^{0\,*}}{\partial\eta} = -\frac{1}{2}\left(\sqrt{2}\left(Y_1^{-1}\right)^* e^{-i\chi} - \sqrt{2}\left(Y_1^{1}\right)^* e^{i\chi}\right) = -\frac{1}{2}\sqrt{\frac{3}{\pi}}\sin\eta$, and $Y_1^0 = \frac{1}{2}\sqrt{\frac{3}{\pi}}\cos\eta$, $Y_1^{\pm 1} = \mp\frac{1}{2}\sqrt{\frac{3}{2\pi}}\sin\eta e^{\pm i\chi}$. After rearrangement,

$$I^{mq}_{ns;np,0} = \frac{e \langle\uparrow| d_{\text{z spin}} |\downarrow\rangle_m}{\sqrt{3}}\times$$
$$\times\left(\frac{\sin^2\eta - 2\cos^2\eta}{R_{qm}^3}\int_0^{R_{qm}} r^3 R_{ns}R_{np}dr + \int_{R_{qm}}^{\infty} R_{ns}R_{np}dr\right), \tag{E.6}$$

where $\cos^2\eta = \frac{\rho^2}{R_{qm}^2}$, $\sin^2\eta = \frac{(X_{qm})^2}{R_{qm}^2}$.

This gives the following expression for the c^{qm} coefficient for the $\left|99p_{3/2}, m_j = 1/2\right\rangle \leftrightarrow \left|99s_{1/2}, m_j = 1/2\right\rangle$ transition

$$c^{qm}_{\mathbf{m_0},\uparrow;\mathbf{m}',\downarrow} = \frac{e\sqrt{2}\langle\uparrow| d_{\text{z spin}} |\downarrow\rangle_m}{3}\int dX_{qm}\left(\frac{\sin^2\eta - 2\cos^2\eta}{R_{qm}^3}\int_0^{R_{qm}} r^3 R_{ns}R_{np}dr + \right.$$
$$\left. + \int_{R_{qm}}^{\infty} R_{ns}R_{np}dr\right) u_{k_0}^{(\nu_0)*}\left(X_m + X_{qm}\right) u_k^{(\nu)}\left(X_m + X_{qm}\right) e^{-i(k-k_0)X_{qm}}. \tag{E.7}$$

For the distances between spin and mediator arrays $\rho \sim 1.5\ \mu$m, which we consider, the $\frac{1}{R_{qm}^3}\int_0^{R_{qm}} r^3 R_{ns}R_{np}dr$ term will be much larger than the $\int_{R_{qm}}^{\infty} R_{ns}R_{np}dr$ term, such that the latter can be neglected. At these distances one can also approximate $\int_0^{R_{qm}} r^3 R_{ns}R_{np}dr \approx \int_0^{\infty} r^3 R_{ns}R_{np}dr$. As a result, the dependence on the mediator position of the c^{qm} coefficients will be given by the factor $(\sin^2\eta - 2\cos^2\eta)/R_{qm}^3$, which shows that at such distances the charge-dipole interaction can be approximated by the dipole-dipole one.

Next, we calculate the c^{qm} coefficients for the spin-mediator interaction involving the $|\downarrow\rangle - |\uparrow\rangle$ and the $\left|99p_{3/2}, m_j = -1/2\right\rangle \leftrightarrow \left|99s_{1/2}, m_j = 1/2\right\rangle$ transitions

$$c^{mq}_{\mathbf{m_0},\uparrow;\mathbf{m}',\downarrow} = \frac{e\langle\uparrow| \vec{d}_{\text{spin}} |\downarrow\rangle_m}{\sqrt{3}}\int dX_{qm}\langle ns|\frac{\vec{R}_{qm} - \vec{r}}{|\vec{R}_{qm} - \vec{r}|^3}|np, m_l = -1\rangle\times$$
$$\times u_{k_0}^{(\nu_0)*}\left(X_m + X_{qm}\right) u_k^{(\nu)}\left(X_m + X_{qm}\right) e^{-i(k-k_0)X_{qm}}. \tag{E.8}$$

When averaging the $-\frac{4\pi e\langle\uparrow| d_{\text{z spin}}|\downarrow\rangle_m\left(\vec{R}_{qm}-\vec{r}\right)_z}{\left|\vec{R}_{qm}-\vec{r}\right|^3}$ function over $|ns\rangle$, $|np, m_l = -1\rangle$ states the integral over three spherical harmonics Eq.(E.5) will have the form $\int_0^{2\pi} d\phi\int_0^{\pi}\sin\theta d\theta Y_0^{0\,*} Y_{l''}^{m''} Y_1^{-1}$, which is non-zero only for $l'' = 1, m'' = 1$. Following the same steps as above one obtains

$$I^{mq}_{ns;np,-1} = -e\langle\uparrow| d_{\text{z spin}} |\downarrow\rangle_m \langle ns|\frac{\left(\vec{R}_{qm} - \vec{r}\right)_z}{\left|\vec{R}_{qm} - \vec{r}\right|^3}|np, m_l = -1\rangle =$$
$$= -e\langle\uparrow| d_{\text{z spin}} |\downarrow\rangle_m \cos\eta\sin\eta e^{-i\chi}\frac{1}{R_{qm}^3}\sqrt{\frac{3}{2}}\int_0^{R_{qm}} r^3 R_{ns}R_{np}dr, \tag{E.9}$$

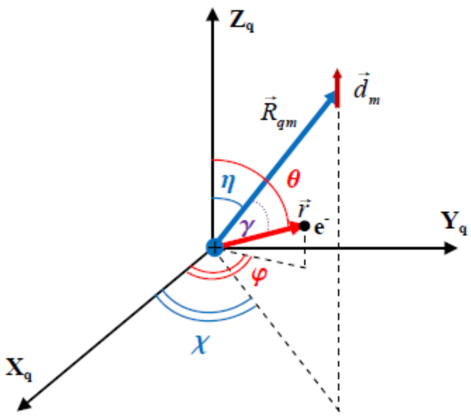

FIG. 9: Angles of the vectors $\vec{R}_{qm}$ and $\vec{r}$ in the case of a general orientation of the Rydberg atom with respect to the molecule.

resulting in

$$c^{mq}_{\mathbf{m_0},\uparrow;\mathbf{m'},\downarrow} = -\frac{e\left\langle\uparrow\right| d_{\text{z spin}} \left|\downarrow\right\rangle_m}{\sqrt{2}} \times$$
$$\times \int dX_{qm} \left(\sin\eta\cos\eta e^{-i\chi}\frac{1}{R^3_{qm}}\int_0^{R_{qm}} r^3 R_{ns}R_{np}dr\right) u^{(\nu_0)*}_{k_0}\left(X_m+X_{qm}\right) u^{(\nu)}_{k}\left(X_m+X_{qm}\right) e^{-i(k-k_9)X_{qm}} \tag{E.10}$$

for the $\left|99p_{3/2}, m_j=-1/2\right\rangle \leftrightarrow \left|99s_{1/2}, m_j=1/2\right\rangle$ transition.

Similarly, the coefficients for the interaction involving the spin $\left|\downarrow\right\rangle - \left|\uparrow\right\rangle$ and the mediator $\left|99p_{3/2}, m_j=3/2\right\rangle \leftrightarrow \left|99s_{1/2}, m_j=1/2\right\rangle$ transitions will have the form:

$$c^{mq}_{\mathbf{m_0},\uparrow;\mathbf{m'},\downarrow} = e\left\langle\uparrow\right| \vec{d}_{\text{spin}} \left|\downarrow\right\rangle_m \int dX_{qm} \left\langle ns\right| \frac{\vec{R}_{qm}-\vec{r}}{|\vec{R}_{qm}-\vec{r}|^3}\left|np, m_l=1\right\rangle \times$$
$$\times u^{(\nu_0)*}_{k_0}\left(X_m+X_{qm}\right) u^{(\nu)}_{k}\left(X_m+X_{qm}\right) e^{-i(k-k_0)X_{qm}} \tag{E.11}$$

with the corresponding

$$I^{mq}_{ns;np,1} = -e\left\langle\uparrow\right| d_{\text{z spin}} \left|\downarrow\right\rangle_m \left\langle ns\right| \frac{\left(\vec{R}_{qm}-\vec{r}\right)_z}{\left|\vec{R}_{qm}-\vec{r}\right|^3}\left|np, m_l=1\right\rangle =$$
$$= e\left\langle\uparrow\right| d_{\text{z spin}} \left|\downarrow\right\rangle_m \cos\eta\sin\eta e^{i\chi}\frac{1}{R^3_{qm}}\sqrt{\frac{3}{2}}\int_0^{R_{qm}} r^3 R_{ns}R_{np}dr, \tag{E.12}$$

where in the average of the $-\frac{4\pi e\left\langle\uparrow\right| d_{\text{z spin}} \left|\downarrow\right\rangle_m \left(\vec{R}_{qm}-\vec{r}\right)_z}{\left|\vec{R}_{qm}-\vec{r}\right|^3}$ factor over $\left|ns\right\rangle$ and $\left|np, m_l=1\right\rangle$ states the integral over three spherical harmonics Eq.(E.5) will be non-zero only for $l''=1$, $m''=-1$. This gives the coefficient:

$$c^{mq}_{\mathbf{m_0},\uparrow;\mathbf{m'},\downarrow} = \frac{e\sqrt{3}\left\langle\uparrow\right| d_{\mathrm{z\ spin}} \left|\downarrow\right\rangle_m}{\sqrt{2}} \times$$

$$\times \int dX_{qm} \left(\sin\eta\cos\eta e^{i\chi} \frac{1}{R^3_{qm}} \int_0^{R_{qm}} r^3 R_{ns} R_{np} dr \right) u^{(\nu_0)*}_{k_0} \left(X_m + X_{qm}\right) u^{(\nu)}_{k} \left(X_m + X_{qm}\right) e^{-i(k-k_9)X_{qm}}. \tag{E.13}$$

In the 1D case $\chi = 0$ and $c^{mq}_{ns,1/2,\uparrow,k_0\ \nu_0;np_{3/2},3/2,\downarrow,k_\nu} = -\sqrt{3}c^{mq}_{ns,1/2,\uparrow,k_0\ \nu_0;np_{3/2},-1/2,\downarrow,k_\nu}$.

Here again one can approximate $\int_0^{R_{qm}} r^3 R_{ns} R_{np} dr \approx \int_0^{\infty} r^3 R_{ns} R_{np} dr$, which will give the dependence on the mediator position under the integral in the form of $\sin\eta\cos\eta/R^3_{qm}$.

Finally, we discuss the interaction matrix elements for transition in which the mediator does not change its internal state, allowed by the change-dipole interaction, $V^{mq}_{\mathbf{m_0},\uparrow;\tilde{\mathbf{m}},\uparrow} = V^{mq}_{\mathbf{ns},k_0\ \nu_0,\uparrow;\mathbf{ns},k_\nu,\uparrow} = c^{mq}_{\mathbf{ns},k_0\ \nu_0,\uparrow;\mathbf{ns},k_\nu,\uparrow} e^{-i(k-k_0)X_m}$ matrix elements, where

$$c^{mq}_{\mathbf{ns},k_0\ \nu_0,\uparrow;\mathbf{ns},k_\nu,\uparrow} =$$

$$= e\left\langle\uparrow\right| \vec{d}_{\mathrm{spin}} \left|\uparrow\right\rangle_m \int dX_{qm} \left\langle ns\right| \frac{\vec{R}_{qm}}{R^3_{qm}} - \frac{\vec{R}_{qm} - \vec{r}}{\left|\vec{R}_{qm} - \vec{r}\right|^3} \left|ns\right\rangle \times$$

$$\times u^{(\nu_0)*}_{k_0} \left(X_m + X_{qm}\right) u^{(\nu)}_{k} \left(X_m + X_{qm}\right) e^{-i(k-k_0)X_{qm}}. \tag{E.14}$$

The integral over Rydberg electron's coordinates

$$I^{mq}_{ns;ns} = e\left\langle\uparrow\right| \vec{d}_{\mathrm{spin}} \left|\uparrow\right\rangle_m \left\langle ns\right| \frac{\vec{R}_{qm}}{R^3_{qm}} - \frac{\vec{R}_{qm} - \vec{r}}{\left|\vec{R}_{qm} - \vec{r}\right|^3} \left|ns\right\rangle \tag{E.15}$$

can be calculated using the expansion (E.4) since $\left\langle\uparrow\right| \vec{d}_{\mathrm{spin}} \left|\uparrow\right\rangle = \vec{e}_z \left\langle\uparrow\right| d_{\mathrm{z\ spin}} \left|\uparrow\right\rangle$ also has only the z component. As a result,

$$I^{mq}_{ns;ns} = \frac{e\left\langle\uparrow\right| d_{\mathrm{z\ spin}} \left|\uparrow\right\rangle \rho}{R^3_{qm}} \left(1 - \int_0^{R_{qm}} R^2_{ns}(r) r^2 dr\right). \tag{E.16}$$

We can estimate the term $\int_0^{R_{qm}} R^2_{ns}(r) r^2 dr > \int_0^{\rho} R^2_{ns}(r) r^2 dr \approx 0.999999999$ for $99s$ of Rb and $\rho = 1.5$ μm, resulting in $I^{mq}_{ns;ns} \approx 10^{-10} e\rho \left\langle\uparrow\right| d_{\mathrm{z\ spin}} \left|\uparrow\right\rangle / R^3_{qm}$. This shows that the effective dipole moment of the ns state is $e\rho\left(1 - \int_0^{\rho} R^2_{ns}(r) r^2 dr\right) \approx 10^{-10} e\rho \approx 3 \cdot 10^{-6}$ a.u. If the $\left|\uparrow\right\rangle$ state is coupled to a $\left|J = 2, m_J\right\rangle$ state by a detuned MW field with a Rabi frequency $\tilde{\Omega}$ and detuning $\tilde{\Delta}$, the dipole moment of the $\left|\uparrow\right\rangle$ state induced by the MW field is $\left\langle\uparrow\right| d_{\mathrm{z\ spin}} \left|\uparrow\right\rangle \sim \left(\tilde{\Omega}/\tilde{\Delta}\right) d_{\mathrm{spin}}$. The interaction coefficients can then be estimated as $c^{mq}_{ns,1/2,k_0\ \nu_0,\uparrow;ns,1/2,k_\nu,\uparrow} \lesssim 10^{-10} e\rho \left\langle\uparrow\right| d_{\mathrm{z\ spin}} \left|\uparrow\right\rangle / \rho^3 \sim 10^{-10} e\rho \left(\tilde{\Omega}/\tilde{\Delta}\right) d_{\mathrm{spin}}/\rho^3 \sim 10^{-3}$ Hz for $\tilde{\Omega}/\tilde{\Delta} \sim 0.1$. It shows that the $|V^{mq}_{ns,1/2,k_0,\uparrow;ns,1/2,k_\nu,\uparrow}| = |c^{mq}_{ns,1/2,k_0\ \nu_0,\uparrow;ns,1/2,k_\nu,\uparrow}| \ll (\mathcal{E}_{\mathbf{ns}}(k_\nu) - \mathcal{E}_{\mathbf{ns}}(k_{0\ \nu_0=1})) \sim 4E_{\mathrm{rec}}/N^2_{\mathrm{at\ latt}} \approx 0.87$ Hz and $|J^{zz\ q,k_0\ \nu_0}_{im}| \sim |V^{mq}_{\mathbf{ns},k_0\ \nu_0,\uparrow;\mathbf{ns},k_\nu,\uparrow}|^2/(4E_{\mathrm{rec}}/N^2_{\mathrm{at\ latt}}) \sim 10^{-8}$ Hz, which is much less than the $|J^{\perp\ q}_{im}|$, $|b^{z\ q}_i|$ terms in Eq.(21).

B. $c^{qm}_{\mathbf{m_0},\alpha;\mathbf{med},\beta}$ coefficients for the Rydberg spin encoding

$$c^{mq}_{\mathbf{m_0},\uparrow;\mathbf{med},\downarrow} = \frac{a_\uparrow b_\downarrow + b_\uparrow a_\downarrow}{3} d_{np,ns} d_{\tilde{n}p,\tilde{n}s} d_\pm \int dX_{qm} u^{(\nu_0)*}_{k_0} \left(X_m + X_{qm}\right) \frac{X_{qm}\rho}{R^5_{qm}} u^{(\nu)}_{k} \left(X_m + X_{qm}\right) e^{ikX_{qm}},$$

$$c^{mq}_{\mathbf{m_0},\downarrow;\mathbf{med},\uparrow} = c^{mq}_{\mathbf{m_0},\uparrow;\mathbf{med},\downarrow},$$

$$c^{mq}_{\mathbf{m_0},\uparrow;\mathbf{med},\uparrow} = \frac{2a_\uparrow b_\uparrow}{9} d_{np,ns} d_{\tilde{n}p,\tilde{n}s} d_\pm \int dX_{qm} u^{(\nu_0)*}_{k_0} \left(X_m + X_{qm}\right) \frac{1 - 3\rho^2/R^2_{qm}}{R^3_{qm}} u^{(\nu)}_{k} \left(X_m + X_{qm}\right) e^{ikX_{qm}},$$

$$c^{mq}_{\mathbf{m_0},\downarrow;\mathbf{med},\downarrow} = -\frac{2a_\downarrow b_\downarrow}{9} d_{np,ns} d_{\tilde{n}p,\tilde{n}s} d_\pm \int dX_{qm} u^{(\nu_0)*}_{k_0} \left(X_m + X_{qm}\right) \frac{1 - 3\rho^2/R^2_{qm}}{R^3_{qm}} u^{(\nu)}_{k} \left(X_m + X_{qm}\right) e^{ikX_{qm}},$$

with the corresponding dipole moments between the $\left|ns_{1/2}, m_j = 1/2\right\rangle$ and the $|\pm\rangle_{\rm med}$ states:

$$\left\langle ns_{1/2}, m_j = \frac{1}{2}\right| \vec{d}_{\rm Rydb}\,|+\rangle_{\rm med} = -\frac{d_+ d_{np,ns}\vec{e}_z}{3},$$
$$\left\langle ns_{1/2}, m_j = \frac{1}{2}\right| \vec{d}_{\rm Rydb}\,|-\rangle_{\rm med} = -\frac{d_- d_{np,ns}\vec{e}_z}{3}, \quad \text{(E.17)}$$

C. Averaging over atomic motional states

In order to calculate the $c^{mq}_{\mathbf{m_0},\alpha;\mathbf{m}',\beta}$ and $c^{mq}_{\mathbf{m_0},\alpha;\mathbf{med},\beta}$ coefficients energies and wavefunctions of mediator atoms Bloch states are needed. In this section we numerically calculate the Bloch functions by analyzing atomic motion in a 1D optical lattice described by the trapping potential $V(X_q) = V_0\cos^2 K_{\rm at}X_q$ with the lattice momentum $K_{\rm at} = \pi/L_{\rm at}$ and $V_0 = -E_{\rm rec}$, where $E_{\rm rec} = \hbar^2(K_{\rm at})^2/2M_{\rm at}$ is the atomic recoil energy in the lattice, $M_{\rm at}$ is the atomic mass, $L_{\rm at}$ is the mediator atom lattice period. The Bloch states and energies can be found by solving the Schrodinger equation for atomic motion in the 1D lattice:

$$-\frac{\hbar^2}{2M_{\rm at}}\frac{d^2\phi(X_q,k_{\nu\,q})}{dX_q^2} + V_0\cos^2(K_{\rm at}X_q)\phi(X_q,k_{\nu\,q}) = = E^{(\nu)}(k)\phi(X_q,k_{\nu\,q}), \quad \text{(E.18)}$$

where the wavefunction corresponding to the $\nu^{\rm th}$ Bloch band and the quasimomentum k is $\phi(X_q,k_{\nu\,q}) = u_k^{(\nu)}(X_q)e^{ikX_q}$. The periodic function u_k can be expanded in terms of the harmonics of the lattice momentum: $u_k^{(\nu)}(X_q) = \sum_{s=-S_{\rm max}}^{S_{\rm max}} c_s^{(\nu)}(k)e^{2isK_{\rm at}X_q}$ with the expansion truncated at some $S_{\rm max}$. The periodic boundary conditions $\phi(X_q,k_{\nu\,q}) = \phi(X_q + N_{\rm latt\ at}L_{\rm at}, k_{\nu\,q})$ require $k = 2\pi\kappa/L_{\rm at}N_{\rm latt\ at}$ and $\kappa = -N_{\rm latt\ at}/2, ..., N_{\rm latt\ at}/2$, where $N_{\rm latt\ at}$ is the number of unitary cells in the mediator atoms lattice. The coefficients $c_s^{(\nu)}(k)$ and Bloch energies $E^{(\nu)}$ can be calculated by numerically solving Eq.(E.18):

$$\left(\frac{\kappa}{N_{\rm at}} + s\right)^2 c_s^{(\nu)} + \frac{V_0}{4E_{\rm rec}}\left(c_{s-1}^{(\nu)} + c_{s+1}^{(\nu)}\right) = \frac{E^{(\nu)} - V_0/2}{E_{\rm rec}}c_s^{(\nu)}. \quad \text{(E.19)}$$

Fig.10 shows energies of the five lowest Bloch bands in the case $V_0 = -E_{\rm rec}$ assuming that the lattice has $N_{\rm latt\ at} = 100$ unitary cells. The expansion of u_k was truncated at $S_{\rm max} = 10$.

We numerically calculated the $u_k^{(\nu)}(X_q)$ functions for five lowest Bloch bands and used them for obtaining the c^{mq} coefficients discussed in the subsections A, B.

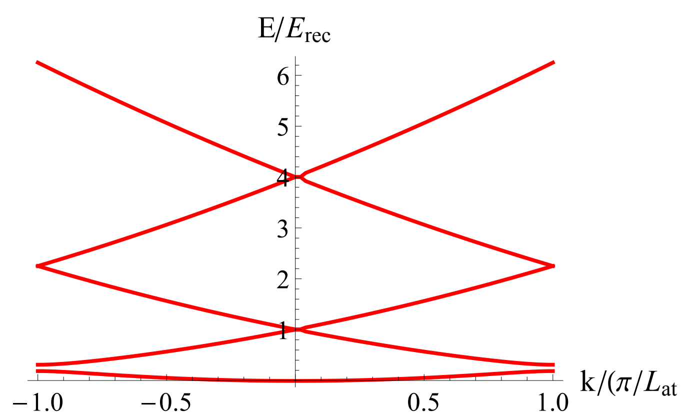

FIG. 10: Bloch energies of the lowest five bands of the 1D optical lattice described by the trapping potential $V(X_q) = V_0\cos^2(K_{\rm at}X_q)$ with $V_0 = -E_{\rm rec}$.

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