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Phys. Rev. Lett. **108**, 193006 — Published 11 May 2012

DOI: [10.1103/PhysRevLett.108.193006](https://doi.org/10.1103/PhysRevLett.108.193006)

Dipole Interaction Mediated Laser Cooling of Polar Molecules to Ultra-cold Temperatures

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We present a method to design a finite decay rate for excited rotational states in polar molecules. The setup is based on a hybrid system of polar molecules with atoms driven into a Rydberg state. The atoms and molecules are coupled via the strong dipolar exchange interaction between two rotation levels of the polar molecule and two Rydberg states. Such a controllable decay rate opens the way to optically pump the hyperfine levels of polar molecules and it enables the application of conventional laser cooling techniques for cooling polar molecules into quantum degeneracy.

The fast decay of excited electronic state of atoms is a paradigm of interactions between light and matter. This decay is the main building block for several technological applications like laser cooling or optical pumping. In turn, these technologies open the way to the potential implementation of quantum simulators and scalable quantum computers. In contrast to the atomic case, the decay of electronically excited states in polar molecules generally ends up in an unwanted excited rotational or vibrational internal state. This proves to be a major obstacle in combining the tools for controlling and cooling atoms with the strong electronic dipole-dipole interactions present in polar molecules. In this letter, we demonstrate the possibility to design an adjustable finite life time for an excited *rotational level* in a polar molecule, which overcomes the obstacles on the way towards optical pumping and laser cooling of polar molecules.

Several alternative methods are experimentally developed in order to cool polar molecules which reside in the rotational and vibrational ground state to cold temperatures. For example, buffer gas cooling [1] and stark deceleration [2]; see Ref. [3, 4] for a review. However, currently the most successful approach is to use ultra cold clouds of atoms from which molecules are created via photo association or coherent formation [5–7]. This technique has recently been perfectionized to create molecules in a well defined hyperfine state [8, 9]. Removing the remaining kinetic energy turns out to be extremely challenging due to the complex internal structure of the polar molecules [10]. Given these complications it may be profitable to use a hybrid setup of dipolar molecules with another system. A gas of Rydberg atoms is a promising candidate as they have both a large dipole moment and can be cooled to ultra cold temperatures [11–14].

In this letter, we demonstrate that a hybrid system of polar molecules with Rydberg atoms [15] allows for the design of a fast and controllable decay for an internal rotational state. The main idea is based on the resonant and strong dipole-dipole interaction between two rotational states and two Rydberg levels: The polar molecules make a transition into the ground state, while at the same time the exchange of a virtual microwave

photon excites the Rydberg atom into a higher level, see Fig. 1. Then, the spontaneous decay of the Rydberg level induces a finite effective decay rate for the rotational level of the polar molecules.

We show that such a decay opens up the way for optical pumping of hyperfine levels and laser cooling of polar molecules using standard techniques. Our setup has two main advantages. First, the decay rate is tunable to arbitrary low values and, second, the decay takes place with very low recoil momentum. Consequently, the Doppler temperature is much lower than in their atomic counterparts and allows one to cool polar molecules into a regime where interactions and particle statistics play an important role. We demonstrate that the method exhibits the potential for laser cooling of strongly interacting many-body system into strongly correlated phases, such as the crystalline phase of polar molecules. We envisage, that our scheme is most suitable for polar molecules created by a coherent formation [8, 9], where unpaired atoms are naturally present, and the polar molecules are already at rather low temperatures.

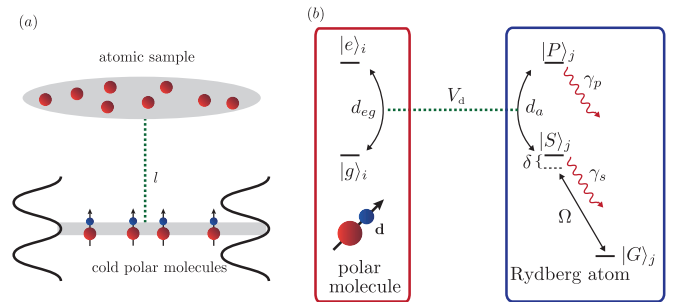


FIG. 1: (a) Hybrid system: trapped polar molecules are in proximity to a cloud of a cold atomic gas with a separation l . (b) Relevant level structure: two rotational states for the polar molecule $|e\rangle_i$ and $|g\rangle_i$ are coupled via dipole-dipole interaction to two Rydberg levels $|S\rangle_j$ and $|P\rangle_j$ of an atom. In addition, the atom is driven from the ground state $|G\rangle_j$ into a Rydberg level. The rotational level $|e\rangle_i$ acquires a finite decay time due to the resonant coupling and the finite life time of Rydberg levels.

We start with the description of the setup for the en-

gineering of a controlled decay rate for an internal rotational state of polar molecules. The decay takes place from the excited state denoted as $|e\rangle_i$ into the ground state $|g\rangle_i$ for the i^{th} polar molecule, see Fig. 1. The transition between the two states is characterized by a dipole matrix element $\mathbf{d}_{eg} = \langle g|\mathbf{d}|e\rangle$ with \mathbf{d} the dipole operator. The internal structure of the polar molecule is described by an effective spin 1/2 system with the Hamiltonian $H_m^{(i)} = \hbar\omega_0\sigma_i^z/2$. The energy difference between the excited and the ground state is $\hbar\omega_0$, and its typical value is in the range of the rotational splitting $2B$. The polar molecule is coupled via the dipole-dipole interaction to the Rydberg levels of a close by atom. The relevant levels for the atom are the ground state $|G\rangle_j$ and the two Rydberg states $|S\rangle_j$ and $|P\rangle_j$. The ground state $|G\rangle_j$ is coupled via a driving laser field to the Rydberg state $|S\rangle_j$. Within the rotating frame and using the rotating wave approximation, the Hamiltonian for the atom reduces to

$$H_a^{(j)} = \sum_{\alpha \in \{G, S, P\}} E_\alpha |\alpha\rangle\langle\alpha|_j + \frac{\Omega}{2} (|G\rangle\langle S|_j + |S\rangle\langle G|_j), \quad (1)$$

where Ω is the Rabi frequency of the driving laser field with the detuning $\hbar\delta = E_S - E_G$. In addition, the Rydberg states are characterized by a dipole transition matrix element $\mathbf{d}_a = \langle S|\mathbf{d}|P\rangle$.

The dipole-dipole interaction gives rise to a resonant exchange interaction between the polar molecules and the Rydberg levels $|e\rangle_i|S\rangle_j \rightarrow |g\rangle_i|P\rangle_j$. The setup is designed such that the exchange takes place near resonant with the detuning $\hbar\Delta = E_P - E_S - \hbar\omega_0$. The interaction between the atom and the molecule reduces to

$$H_c = \sum_{i,j} V_d(\mathbf{r}_i - \mathbf{R}_j) [S_i^- T_j^+ + S_i^+ T_j^-], \quad (2)$$

where $V_d(\mathbf{r}) = \mathbf{d}_{eg} \cdot \mathbf{d}_a / r^3 - 3(\mathbf{d}_{eg} \cdot \mathbf{r})(\mathbf{d}_a \cdot \mathbf{r}) / r^5$ denotes the dipole-dipole interaction with \mathbf{r}_i the positions of the polar molecules and \mathbf{R}_j the coordinates of the atoms, respectively. The operators $S_i^- = |g\rangle\langle e|_i$ account for the transition within the polar molecule, while $T_j^+ = |P\rangle\langle S|_j$ is the corresponding operator acting on the Rydberg levels.

The excited electronic states of the atom $|S\rangle_j$ and $|P\rangle_j$ are characterized by a finite life time with the decay rates γ_s and γ_P , respectively. The time evolution of the full system is then described by the master equation for the density matrix ρ

$$\partial_t \rho = \frac{i}{\hbar} [H, \rho] + \sum_{\alpha \in \{S, P\}} \frac{\gamma_\alpha}{2} [2c_\alpha \rho c_\alpha^\dagger - c_\alpha^\dagger c_\alpha \rho - \rho c_\alpha^\dagger c_\alpha], \quad (3)$$

with the jump operators $c_\alpha = |G\rangle\langle\alpha|$ with $\alpha \in \{S, P\}$ accounting for the decay from the Rydberg states into the ground state $|G\rangle_j$. In addition, the Hamiltonian H contains the polar molecules as well as the driven atomic system, i.e., $H = \sum_i H_m^{(i)} + \sum_j H_a^{(j)} + H_c$.

The master equation Eq. (3) implies that any initial state ends up with all polar molecules in the rotational ground state $|g\rangle_i$, as can be seen by a numerical simulation of the time evolution. For weak couplings, it follows that the dynamics is well described by an effective decay rate for the excited rotational state of the polar molecules, which reduces to

$$\Gamma_i = \sum_j \frac{|M_{ij}|^2}{\hbar^2} \frac{\gamma_P}{\gamma_P^2/4 + (\Delta - \delta)^2} \frac{\Omega^2}{\gamma_s^2/4 + \delta^2}. \quad (4)$$

The last factor in the product $p_s = \Omega^2/(\gamma_s^2/4 + \delta^2)$ describes the probability for an atom to be in the excited Rydberg state $|S\rangle_j$ under the external drive. The second factor accounts for the finite line width of the Rydberg states and the first factor is the coupling matrix elements determined by the dipole-dipole interaction

$$|M_{ij}|^2 = \sum_{n'm'} |\langle \psi_{in'} \phi_{jm'} | V(\mathbf{r}_i - \mathbf{R}_j) | \psi_{in} \phi_{jm} \rangle|^2. \quad (5)$$

Here, $\psi_{in}(\phi_{jm})$ are the initial spatial wave function and $\psi_{in'}(\phi_{jm'})$ the final spatial wave function for the polar molecule (atom), respectively. The decay rate in Eq. (4) is the main result of this paper. In the following we provide (i) a discussion of Eq. (4) and the limits of its applicability, (ii) numerical values relevant for concrete experiments, and (iii) a possible application in the form of Doppler cooling.

Discussion of Eq. (4) – We start with analyzing the matrix elements M_{ij} for two different setups: First, we consider molecules tightly confined in a three dimensional harmonic trap with oscillator length a_0 . If a_0 is much smaller than the distance between the atoms and the polar molecules l , we can express the dipole-dipole interaction in terms of the harmonic oscillator wave functions. The excitation from the oscillator ground state is then suppressed by $(a_0/l)^2$. Consequently, in leading order the polar molecule remains within the spatial ground state. However, the resulting average energy transfer by $2\hbar^2\pi^2/ml^2$ corresponds to a recoil momentum of $q_d = 2\pi\hbar/l$.

Alternatively, for polar molecules tightly confined in only two dimensions, see Fig. 1, a transfer of in-plane momentum \mathbf{K} can appear. Its contribution is described by the in-plane Fourier transformation of the interaction potential $V_d(l, \mathbf{K}) = \int d\mathbf{r}_\parallel V_d(l, \mathbf{r}_\parallel) e^{i\mathbf{K}\mathbf{r}_\parallel}$. It is peaked for $K \sim 1/l$ and decays exponentially for larger momenta. Consequently, also such a setup gives rise to a characteristic momentum recoil $q_d = 2\pi\hbar/l$.

The above analysis reveals an important aspect of our setup: The recoil of the spontaneously emitted photon from the Rydberg state only acts on the atom, while the recoil for the polar molecule q_d in turn is completely determined by the separation l and can be strongly suppressed compared to the photon recoil. Note, that this discussion for the recoil energy is limited to the regime,

where Fermi's Golden rule is applicable. Hence, we need to assess the validity of Eq. (4).

Let us return to the evaluation of Eq. (4). The decay rate is enhanced by the number of atoms within range of the dipole-dipole interaction, and can be written as $\sum_j |M_{ij}|^2 = N_i |V_d(\mathbf{l})|^2$. The number of atoms within interaction range N_i strongly depends on the spatial distribution of the atomic sample and reduces to $N_i = \int d\mathbf{R} |V_d(\mathbf{r}_i - \mathbf{R})|^2 \rho(\mathbf{R})$. Here, $\rho(\mathbf{R})$ denotes the density of Rydberg atoms. For weak Rabi frequencies $\Omega/\delta \ll 1$ this density is determined by the atomic density reduced by a factor p_s accounting for the probability to find an atom in the Rydberg state. For strong drives the Rydberg-blockade phenomena starts to play a role and limits the Rydberg density; see [16] for a detailed discussion. In either case, this analysis provides us with an effective coupling strength $N_i |V_d(\mathbf{l})|^2$ between a single molecule and the atomic cloud.

We can now answer the question on the validity of Fermi's Golden rule which is based on weak coupling. In the presence of the enhancement by the different decay channels, this condition requires $\sqrt{N_i} |V_d(\mathbf{l})| \lesssim \gamma_p$. As a consequence, the induced decay rate Γ_i is limited by the decay rate of the Rydberg level. However, the latter can be strongly enhanced and controlled by dressing the Rydberg level with a lower lying electronic state [15]. Denoting the decay rate of the additional electronic state by γ_d , the far-detuned coupling of the Rydberg level $|P\rangle_j$ with this electronic state with Rabi frequency Ω_d and detuning Δ_d provides a total decay rate for the dressed Rydberg level $\gamma'_p \approx \gamma_p + \gamma_d \Omega_d^2 / 4\Delta_d^2$. When providing numerical values for the obtainable decay rates for the rotational levels with take this enhancement into account.

Numerical values – In the following, we provide experimental parameters for LiCs polar molecules with the atomic reservoir given by Cs atoms excited into the Rydberg state. First, we ignore the weak hyper fine interaction for the polar molecules, and describe the characteristic energy scale; the incorporation of the nuclear spin is then straightforward. We choose for the excited states of the polar molecule the first excited rotational states with angular momentum one $|e\rangle = |1, 0\rangle$; here $|j, m\rangle$ denote angular momentum eigenstates of the rotational degree of freedom. The dipole moment to the ground state reduces to $|\mathbf{d}_{eg}| = d/\sqrt{3}$ with $d = 5.5$ Debye $\approx 2.14ea_0$ the permanent electric dipole moment of the polar molecule. With a rotational splitting $B \approx 5.84$ GHz, the transition $|1, 0\rangle \rightarrow |g\rangle = |0, 0\rangle$ becomes nearly resonant for the Rydberg transition between states with principle quantum number $n = 69$ and (electronic) angular momentum $|s\rangle \rightarrow |p_{3/2}\rangle$. According to the well established quantum defect theory [17] they have an energy difference of $\Delta \approx 40$ MHz and a dipole moment $|\mathbf{d}_a| \approx 10500$ Debye. With these large dipole moments the resonant dipole-dipole interaction reduces to $V_d(\mathbf{l}) \approx 1.7$ kHz even at a distance of $l = 25\mu\text{m}$ between the polar molecules and

the Rydberg atoms (separated along the z-direction).

This energy scale should be compared with the decay rate from the Rydberg state γ_p , which is dominated by black body radiation and reduces to $\gamma_p \sim 840$ Hz. In the following, we will enhance this decay rate to $\gamma'_p \approx 0.3$ MHz as discussed above. Consequently, we obtain a decay rate ≈ 39 Hz for each atom excited into the Rydberg level $|S\rangle$.

Next, we need to estimate the enhancement N_i due to the coupling to a whole ensemble of Rydberg atoms. The characteristic blockade radius is in the range of $\approx 3\mu\text{m}$ [16], and consequently, the Rydberg density is limited by $\approx 10^{11}\text{cm}^{-3}$; this density is much lower than the characteristic atomic density for a cold sample of Cs atoms. For an atomic sample within an oblate trap with transverse confining along the z-axis with $10\mu\text{m}$, the summation over the different Rydberg atoms provides an enhancement $N_i \approx 52000$. Then, the condition $\sqrt{N_i} V_d(\mathbf{l}) \lesssim \gamma'_p$ is satisfied and the application of Fermi's Golden rule for the estimation of the induced decay rate is valid. We finally find a induced decay rate for the rotational level of the polar molecules $\Gamma_i \approx 0.25$ MHz.

Alternatively, it is experimentally interesting to work in a parameter regime with strong electric fields in combination with a strong confinement of the molecules into a two-dimensional setup [18]. However, the strong electric field requires to work with much lower main principal number n of the Rydberg levels to prevent field ionization.

In this setup the condition of resonant dipole-dipole interaction is conveniently obtained by selecting two Rydberg states which undergo a true level crossing within the stark map, but are coupled by exchanging a quantum of angular momentum. For Cs Rydberg atoms, the states with $n = 17$ and $|S\rangle = |d_{5/2}, \pm 3/2\rangle$ and $|P\rangle = |d_{5/2}, \pm 5/2\rangle$ exhibit such a level crossing [19] at an electric field $E \approx 3\text{kV/cm}$. Moreover, they allow for a resonant dipole exchange with the two states $|e\rangle = |1, \pm\rangle$ and $|g\rangle = |0, 0\rangle$ of the polar molecule [20]. On a distance with $l = 3\mu\text{m}$ the dipole exchange energy reduces to $V_d(\mathbf{l}) \approx 3100$ Hz. With the decay rate for the Rydberg atoms $\gamma_s \approx \gamma_p \approx 72$ kHz, the induced decay rate with a single Rydberg atom reduces to ≈ 530 Hz. Again, this decay rate is enhanced by the number independent decay channels N_i . Assuming the atomic system confined into a 2D layer above the polar molecules with a Rydberg density $\approx 10^{12}\text{cm}^{-3}$, the total decay rate reduces to $\Gamma_i \approx 11$ kHz.

Applications – An immediate application capitalizing on the finite life-time of a rotational level in polar molecules is the optical pumping of the hyper fine structure. The main mechanism is the electric quadrupole interaction, which couples nuclear spins to rotational excitations, and has been successfully used for the coherent transfer of population between different hyperfine states [8, 9]. Note, that the optical pumping may give rise to coherence between polar molecules separated on distances

smaller than l ; it is this coherence, which allows to apply conventional laser cooling techniques [21–23] to temperatures below quantum degeneracy.

In the following, we describe the peculiarities of our setup for Doppler cooling. However, we would like to stress, that the induced decay rate can also be used for more sophisticated cooling techniques, such as Sisyphus cooling, which in general will give rise to more efficient cooling of the system. In order to achieve Doppler cooling, the transition into the excited rotational state is driven via a Raman transition with Rabi frequency Ω , the detuning from resonance Δ , and a momentum transfer \mathbf{q} . In contrast to conventional laser cooling, here, the spontaneous decay from the excited rotational state transfers a different momentum to the polar molecule. Consequently, we have to distinguish between two momenta. The momentum \mathbf{q} absorbed in the Raman process and the small momentum q_d from the spontaneous decay; see above for a discussion of this momentum.

The standard laser cooling techniques are characterized by two energy scales: the higher energy/temperature $T_c = m\Gamma^2/(2\mathbf{q}^2)$ determines the kinetic energy, where laser cooling becomes effective; here Γ denotes the induced decay rate from the excited rotational state. Second, the equilibrium state induced by the laser cooling is characterized by the Doppler temperature $T_d = \hbar\Gamma$.

A major advantage of the present scheme is the possibility to tune the recoil momentum \mathbf{q} independently of the decay rate Γ . As a consequence, the capturing temperature T_c and the Doppler temperature can be adjusted for optimal cooling. The Doppler temperature of the equilibrium state can be continuously decreased by reducing the decay rate Γ . The lowest temperature is therefore only limited by the small recoil momentum q_d from the spontaneous decay, i.e., $T_{\text{limit}} = \hbar^2 q_d^2 / 2m$. This temperature is in general much lower than the Doppler temperature for conventional laser cooling of alkali atoms [23], and the present scheme allows to laser cool polar molecules to temperatures comparable to quantum degeneracy.

Note, that for conventional Doppler cooling, the re-absorption of emitted photon provides a decrease in cooling efficiency for dense atomic samples. The analogue effect in the present scheme would be the re-excitation of a polar molecule by a resonant dipolar exchange. However, this rate is strongly suppressed for a fast decay of the Rydberg level and a proper balance between the number of atoms N and polar molecules M with the requirement $M \ll N$.

Close to quantum degeneracy, the statistics of the particles starts to play an important role. The Doppler cooling scheme described above remains valid also in this regime. This allows one in principal to cool directly into a Fermi sea for fermions or a Bose-Einstein condensate for bosons. However, the Doppler cooling scheme breaks down at low temperatures and high particle densities,

where interactions between the molecules become relevant.

There are two main effects due to interactions: first, the interaction potential between two molecules in state $|g\rangle$, and the interaction potential between the states $|g\rangle$ and $|e\rangle$ are in general different; especially, as for the two states $|g\rangle$ and $|e\rangle$ also a resonant exchange of a microwave photon can appear. During the Doppler cooling process, each quantum jump with the excitation of a polar molecule into the higher rotational state provides a fluctuating force to the surrounding polar molecules and eventually provides a source of heating. Second, the difference of the interaction potentials leads to a level broadening of the excited rotational state, and the desired energy selectivity for the Doppler cooling breaks down as soon as the level broadening reaches the induced decay rate.

However, one can envisage a strategy that overcomes this limitation and potentially allows cooling directly into a strongly interacting many-body phase such as the crystalline state [18]. The trick is to adiabatically eliminate the excited rotational states by irradiating a far-detuned Raman laser. Consequently, the many-body states acquire a finite lifetime inherited from the small admixture of the excited state. In the following, we denote by $|\alpha\rangle$ and $|\beta\rangle$ eigenstates of the fully interacting many-body system with energies E_α and E_β , respectively. Using Fermi's Golden rule, we obtain transition rates $\Gamma_{\alpha\rightarrow\beta}$ from the initial state $|\alpha\rangle$ into the final state $|\beta\rangle$ due to the coupling of the system to the bath

$$\Gamma_{\alpha\rightarrow\beta} = \frac{\Omega^2}{4\Delta^2} |M_{\alpha\beta}|^2 \frac{\Gamma}{(\tilde{\Delta} - E_\alpha + E_\beta)^2 + \Gamma^2/4}. \quad (6)$$

Here, Γ denotes the decay rate of the bath, while the matrix elements for the many-body system reduces to

$$M_{\alpha\beta} = \int d\mathbf{x} \langle \alpha | \psi^\dagger(\mathbf{x}) \psi(\mathbf{x}) e^{i\mathbf{q}\mathbf{x}} | \beta \rangle. \quad (7)$$

It is important to point out, that the energy dependence is in analogy to conventional laser cooling techniques, i.e., for $E_\alpha - E_\beta > 0$ and blue detuning $\Delta > 0$, the transitions $|\alpha\rangle \rightarrow |\beta\rangle$ take place with a higher rate than the reversed transitions $|\beta\rangle \rightarrow |\alpha\rangle$, and consequently leads to a cooling of the system.

Conclusion. In summary, we proposed a scheme to induce a controlled decay rate for rotational levels of polar molecules. We showed how one can harness this finite lifetime to perform Doppler cooling into quantum degeneracy. Finally, we commented on how to implement a direct cooling into a strongly interacting many-body phase.

Acknowledgment: We thank T. Pfau and P. Zoller for fruitful discussions. Support by the Deutsche Forschungsgemeinschaft (DFG) within SFB / TRR 21 and National Science Foundation under Grant No. NSF

PHY05-51164 is acknowledged. SDH acknowledges support by the Swiss Society of Friends of the Weizmann Institute of Science.

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