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Phys. Rev. Lett. **129**, 033201 — Published 11 July 2022

DOI: [10.1103/PhysRevLett.129.033201](https://doi.org/10.1103/PhysRevLett.129.033201)

# Coherent reaction between molecular and atomic Bose-Einstein condensates: integrable model

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We solve a model that describes a stimulated conversion between ultracold bosonic atoms and molecules. The reaction is triggered by a linearly time-dependent transition throughout the Feshbach resonance. Our solution predicts a dependence, with a dynamic phase transition, of the reaction efficiency on the transition rate for both atoms-to-molecule pairing and molecular dissociation processes. We find that for the latter process with a linear energy dispersion of atomic modes, the emerging phase can have a thermalized energy distribution of noninteracting bosons with the temperature defined by the rate of the transition. This provides a simple interpretation of the phase transition in terms of the creation of equilibrium Bose-Einstein condensate.

Recently, a coherent conversion between Cs atomic Bose-Einstein condensate (BEC) and the condensate of Cs<sub>2</sub> molecules was demonstrated by varying a magnetic field in time and thus pushing the system across the Feshbach resonance [1]. To suppress irreversible scatterings, BECs of rotating molecules were confined in a quasi-2D trap in [1]. In the created state, possibly up to 50% of molecules formed a condensate, and in the reversed process, about 40% of the molecules coherently dissociated into atoms. Further improvements of the conversion efficiency will enable *the chemistry of coherent BECs* for engineering macroscopic correlated quantum states, with applications in sensing and information-processing.

Until the experiment in [1], a considerable fraction of the molecular condensate, was possible to generate only for the reactions of fermionic atoms [2–6], which had fewer possibilities than bosonic ones to create detrimental excitations. A considerable understanding of fermion-boson reactions was achieved due to the discovery of integrability of the time-dependent Tavis-Cummings model [7, 8], which provided statistical characteristics of the final many-body state. A similarly general approach to the purely bosonic reactions is still missing but desirable for searching new effects and engineering quantum many-body states.

The theory of integrability of explicitly time-dependent quantum systems has recently produced a variety of solvable linearly time-dependent quantum models [9]. Many of their applications have been anticipated [10–14]. However, the bulk of the found models describe interactions of only a few states. Although the conditions for integrability are known, there is still no straightforward path to identify the models with a combinatorially complex phase space that would satisfy such conditions. Hence, the search for such models still relies heavily on intuition and chance.

In this Letter, we identify a new experimentally relevant model that can be solved exactly and that captures the main features of the chemistry of BECs. Its Hamilto-

nian describes a stimulated conversion between bosonic atoms and molecules during a sweep of a linearly time-dependent magnetic field across a Feshbach resonance. Our model captures the main features of the process: many-body interactions, energy dispersion of the atomic states, different initial populations of these states, and an arbitrary sweep rate  $\beta$ :

$$H(t) = -\beta t \hat{\Psi}^\dagger \hat{\Psi} + \sum_k \left\{ \varepsilon_k^a \hat{a}_k^\dagger \hat{a}_k + \varepsilon_k^b \hat{b}_k^\dagger \hat{b}_k + g \left( \hat{\Psi}^\dagger \hat{a}_k \hat{b}_k + \hat{\Psi} \hat{a}_k^\dagger \hat{b}_k^\dagger \right) \right\}. \quad (1)$$

Here,  $\hat{a}_k$ ,  $\hat{b}_k$ ,  $\hat{\Psi}$  are the boson annihilation operators;  $\varepsilon_k^{a,b}$  are the energies of the free atoms;  $\beta$  is proportional to the ramp of the magnetic field that sweeps the system across the Feshbach resonance, and  $g$  is the coupling for the conversion of the atomic pairs into the molecules;  $\hat{\Psi}$  is the molecular operator, and  $\hat{a}_k$  and  $\hat{b}_k$  describe the modes of the ultracold atoms. The atomic modes are generally different [15]. For example, the momentum conservation may force the molecules with zero momentum to split only into atoms with opposite momenta  $\pm \mathbf{p}$ . In this case, the index  $k$  runs over only half of the possible atomic states, for which we associate  $\hat{a}_k$  with  $\hat{a}_{\mathbf{p}}$ , and we associate  $\hat{b}_k$  with  $\hat{a}_{-\mathbf{p}}$ . However, we allow for any reaction channel  $k$  to identify  $\hat{b}_k$  with  $\hat{a}_k$ , which is possible if a pair of emerging atoms has  $\mathbf{p} = 0$ .

To keep the model (1) integrable, we disregarded the energy dispersion of molecular states, which can be justified by the fact that the molecular condensate dominates the coherent reactions with atoms. We also do not include other than cubic interactions of the particles, which means that the molecular-atomic interactions near the resonance dominate.

We assume that as  $t \rightarrow -\infty$  the system is close to its ground state. Hence, the model (1) for  $\beta > 0$  describes the driven transition from atoms as  $t \rightarrow -\infty$  into an initially empty molecular mode [Fig. 1(a)]. We will call

this a *forward process*. The *reverse process* describes the dissociation of the molecular condensate in Fig. 1(b). It corresponds to  $\beta < 0$ , so that the state with a molecular condensate is at  $t = -\infty$ , and it is then coherently converted into atomic pairs. Our goal is to find the final state as  $t \rightarrow +\infty$ .

Prior theories of the BEC chemistry were usually restricted by considering steady conditions [16–20]. They revealed a possibility of a phase transition [16, 17] but the effect of the nonadiabatic dynamics during the stimulated transition through the Feshbach resonance was missing. On the other hand, the explicitly time-dependent evolution could be studied only with considerable approximations, for example, using the mean field quadratic Hamiltonians [21] or applying semiclassical and diagrammatic techniques that disregarded the atomic energy dispersion [22, 23] and could be justified only for proper limits of the model's parameters.

The integrability of the model (1) thus reveals the most nontrivial behavior that can be tested for BEC reactions: the emergence of dynamic phase transition, the effect of competition between different reaction channels, and the properties of quantum correlations that are induced by cubic interactions in  $H(t)$ .

To solve the model, we note that  $Q_k = \hat{a}_k^\dagger \hat{a}_k - \hat{b}_k^\dagger \hat{b}_k$  commutes with  $H(t)$ , and introduce operators  $K_k^+ \equiv \hat{a}_k^\dagger \hat{b}_k^\dagger$  and  $K_k^- \equiv \hat{a}_k \hat{b}_k$ , which satisfy

$$[\hat{q}_k, K_k^\pm] = \pm 2K_k^\pm, \quad [K_k^-, K_k^+] = (\hat{q}_k + 1), \quad (2)$$

where  $\hat{q}_k \equiv \hat{a}_k^\dagger \hat{a}_k + \hat{b}_k^\dagger \hat{b}_k$ . We also introduce parameters  $\varepsilon_k = (\varepsilon_k^a + \varepsilon_k^b)$ , and  $\tau$  to rewrite  $H(t)$  as

$$H(t) = -\beta t \hat{\Psi}^\dagger \hat{\Psi} + \sum_k \left\{ \tau \varepsilon_k \hat{q}_k / 2 + g \left[ \hat{\Psi}^\dagger K_k^- + \hat{\Psi} K_k^+ \right] \right\}. \quad (3)$$

At  $\tau = 1$ , we reproduce Eq. (1) up to  $Q_k$ -dependent terms that do not change the dynamics. Our main observation is that  $H(t)$  in Eq. (3) commutes with

$$H_1(t) = \sum_k \left\{ \varepsilon_k \left( t + \frac{\tau \varepsilon_k}{\beta} \right) \frac{\hat{q}_k}{2} + \frac{g \varepsilon_k}{\beta} \left[ \hat{\Psi}^\dagger K_k^- + \hat{\Psi} K_k^+ \right] \right\} + \frac{g^2}{\beta \tau} \sum_{i,j; i \neq j} (K_i^+ K_j^- - (\hat{q}_i + 1)(\hat{q}_j + 1)/4), \quad (4)$$

and these two operators satisfy the relation

$$\partial H / \partial \tau = \partial H_1 / \partial t. \quad (5)$$

According to [9, 25], this renders our model an integrable multistate Landau-Zener model, for which any rescaling of  $\tau$  in Eq. (3) does not change the scattering probabilities between any eigenstates of  $H(t)$ .

Hence, without affecting the final result, we can set  $\tau \rightarrow \infty$ , which renders all atomic energies,  $\tau \varepsilon_k$  in  $H(t)$ , well-separated by the time of the corresponding resonances, at which  $\tau \varepsilon_k = -\beta t$ . Near each such resonance, we can safely disregard all other reaction channels. After

finding the scattering amplitudes at each channel (resonance), we can then treat the effects of different resonances sequentially in their chronological order to find the state after the passage through all resonances.

The Hamiltonian restricted to a single reaction channel,  $\hat{\Psi} \leftrightarrow \hat{a}\hat{b}$ , is given by

$$H(t) = -\beta t \hat{\Psi}^\dagger \hat{\Psi} + g \left( \hat{\Psi}^\dagger \hat{a}\hat{b} + \hat{\Psi} \hat{a}^\dagger \hat{b}^\dagger \right), \quad (6)$$

where we shifted the timescale to set the resonance at  $t = 0$ . The dynamics with Eq. (6) conserves two quantities:

$$N = \hat{\Psi}^\dagger \hat{\Psi} + \hat{b}^\dagger \hat{b}, \quad Q = \hat{a}^\dagger \hat{a} - \hat{b}^\dagger \hat{b}. \quad (7)$$

This allows us to express the microstates only via the number of molecules  $m$ :  $|m\rangle \equiv |m; N - m + Q, N - m\rangle$ , where  $N - m + Q$  and  $N - m$  are the numbers of atoms in  $a$  and  $b$  modes, respectively. Note also that, at  $Q = 1$ , Eq. (6) has the same matrix elements as the Hamiltonian with a single atomic mode:  $\hat{a} \equiv \hat{b}$ .

Following [22], Eq. (6) can be mapped to the *driven Tavis-Cummings model* (DTCM) [7, 8]:

$$H_{TC} = \beta t (\hat{\psi}^\dagger \hat{\psi} - N) + g (\hat{\psi}^\dagger \hat{S}^- + \hat{\psi} \hat{S}^+), \quad (8)$$

where  $\hat{\psi}$  is a boson annihilation operator, and  $\hat{S}^\pm$  are raising/lowering operators of a spin with size  $S = (N + Q)/2$ . The DTCM conserves  $N = \hat{\psi}^\dagger \hat{\psi} + (S + \hat{S}_z)$ . Hence, we can mark its states as  $|m\rangle$  where  $m = S + S_z$ . Then,

$$\langle m+1 | H_{TC} | m \rangle = \sqrt{(N-m)(N+Q-m)(m+1)}. \quad (9)$$

Comparing with (6), we find

$$\langle m | H | m' \rangle = \langle m | H_{TC} | m' \rangle, \quad \forall m, m'.$$

This map is not intuitive in the sense that the number of molecules  $m$  in Eq. (6) is not the same as the number of bosons in the DTCM. Instead,  $\langle m | \hat{\psi}^\dagger \hat{\psi} | m \rangle = N - m$ .

*Reaction efficiency.* The DTCM has been solved previously [7]. Its transition probabilities between any microstates can be found in Eqs. (23-24) in [8], where the limits of large  $N$  are also described. For the forward process, there is a negative feedback because the net number of bosons in the system decreases with emergence of molecules. Using the map to the Tavis-Cummings model [24], we find that if all atoms are initially in the ground state with some number of pairs  $N \gg 1$  and  $Q = O(1)$  then the probability distribution of finding  $m$ -molecules, for  $\langle m \rangle \gg 1$ , at the end is nearly Gaussian and sharply peaked near the average value

$$\langle m \rangle \equiv \langle \hat{\Psi}^\dagger \hat{\Psi} \rangle_{t \rightarrow \infty} = N + \frac{\log(2 - x^{N+Q})}{\log x}, \quad x = e^{-\frac{2\pi g^2}{\beta}}. \quad (10)$$

For the reverse sweep, starting with  $N$  molecules and no atoms, after the passage through one resonance, the probability to produce  $n$  atomic pairs is given by [24]

$$P_n = x^{N-n} (x^{N-n+1}, x)_n, \quad x = e^{-2\pi g^2/\beta}, \quad (11)$$

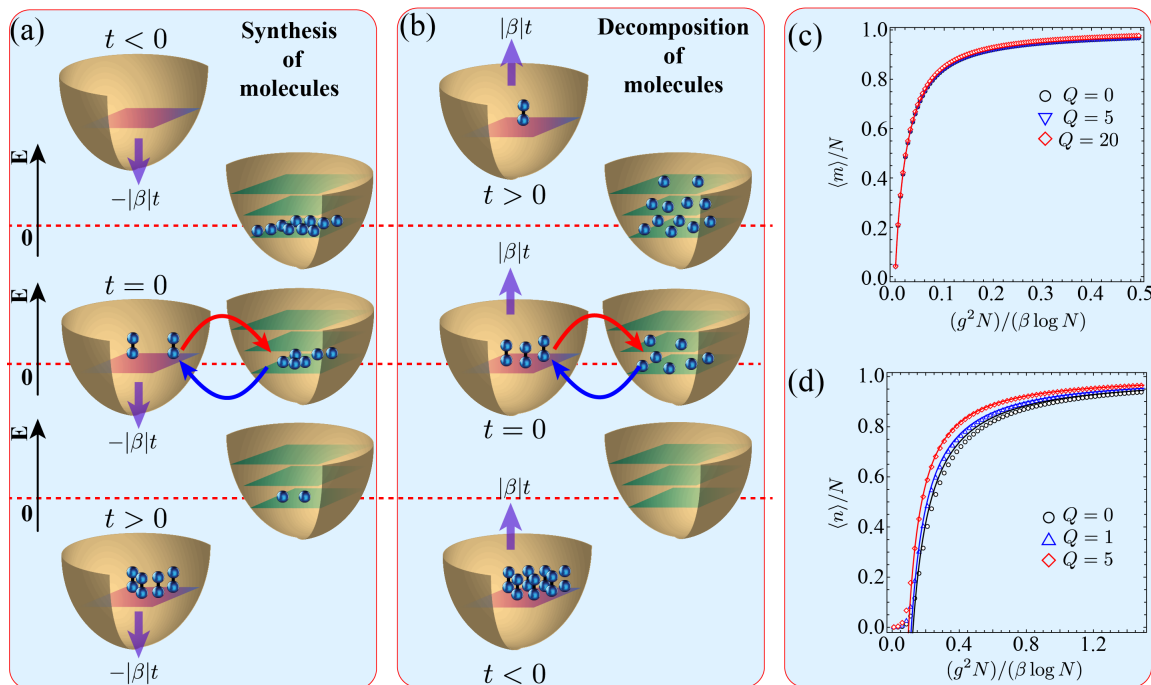


FIG. 1. The reaction between the atomic and the molecular condensate in (a) the forward and (b) the reverse process. The  $\mathbf{E}$ -axis is energy; purple arrows show the direction of the transition throughout the resonance. Molecular and atomic modes in an arbitrary trap are shown as pink and green surfaces, respectively. (c) and (d) characterize the average number of molecules  $\langle m \rangle$  and of atomic pairs  $\langle n \rangle = N - \langle m \rangle$  that are found for different values of the inverse sweep rate,  $1/\beta$ , at  $g = 1$ , after the transition starting from the ground state in the forward and the backward processes in Eq. (6), respectively. The discrete points are the exact predictions for the initial number of particles  $N = 10^4$  [24], and the solid curves in (c) and (d) are the large- $N$  predictions of, respectively, Eq. (10) and Eq. (13) from [24].

where

$$(a, x)_n \equiv \prod_{k=0}^{n-1} (1 - ax^k) = (1 - a)(1 - ax) \cdots (1 - ax^{n-1}) \quad (12)$$

is the  $q$ -Pochhammer symbol. According to [26], this distribution is broad due to the positive feedback that also leads to a dynamic phase transition. If the number of molecules  $N$  is initially macroscopically large, then the fraction of molecules converted to atoms,  $\langle n \rangle / N = 1 - \langle m \rangle / N$ , behaves discontinuously as a function of  $g^2 / \beta$ . Let

$$f = \frac{2\pi g^2}{\beta} \frac{N}{\log_e N}, \quad (13)$$

then the distribution (11) has the property

$$\begin{aligned} \langle n \rangle / N &= 0 \quad \text{for } f < 1, \quad N \rightarrow \infty, \\ \langle n \rangle / N &= \frac{f - 1}{f} \quad \text{for } f \geq 1, \quad N \rightarrow \infty. \end{aligned} \quad (14)$$

In Figs. 1(c,d) we confirm the predictions (10), (14) using numerically exact transition probabilities [24] for  $N = 10^4$ , and also verify robustness of such predictions against an initial asymmetry in atomic population due to nonzero  $Q$ . Here we note similarity of Fig. 1(d) with

the experimentally obtained red curve in Fig.4(a) in [1]. Both figures describe the number of produced atoms from molecules: in our case as a function of  $1/\beta$  but as a function of time after the molecules are placed at resonance in [1]. We attribute this to the fact that the time-scale  $\tau_{\text{eff}} = g/\beta$  in our model characterizes the effective time that the system spends sufficiently close to the resonance to dissociate the molecules during the sweep. Thus, both figures describe similar physics: it takes initially certain critical time for the process to produce  $\sim \log N$  atomic pairs before they induce the positive feedback that makes  $O(N)$  of the remaining molecules dissociate due to super-radiance [26]. This indicates a possibility that our dynamic phase transition can be confirmed with the setup that was used in [1].

*Coherent thermalization.* Consider now the effect of the dispersion  $\varepsilon_k$  of atomic modes in the general model Eq. (1). For the forward-sweep, the integrability means that only higher energy atomic pairs can influence the lower energy ones. Hence, if the initial state is the atomic ground state then the higher energy states remain empty at the end, and Eq. (10) applies to this case as well. According to it, we predict even for multi-channel reactions an exponential dependence of the reaction efficiency on  $1/\beta$  for fast sweeps and a power-law tail,  $N - \langle m \rangle \sim 1/\beta$  in the limit of slow transitions.

For the reverse process, if the initial state has  $N$  molecules, then Eq. (11) applies to the first encountered resonance. It also applies to the following resonances but the number of entering molecules must be reduced by the amount that has already dissociated. If there are many resonances, all molecules will dissociate.

The final multi-mode population distribution has a very simple structure. To demonstrate this, we write the joint transition probability to produce  $n_1$  atomic pairs in the first and  $n_2$  pairs in the second resonance:

$$P_{n_1, n_2} = P_{n_1} x^{N-n_1-n_2} (x^{N-n_1-n_2+1}, x)_{n_2}, \quad (15)$$

and compare two probabilities of the populations that are different by moving one atomic pair from the lower energy mode to the higher energy one. Taking the ratio of such probabilities, we find

$$\frac{P_{n_1-1, n_2+1}}{P_{n_1, n_2}} = e^{-2\pi g^2/\beta}, \quad (16)$$

which does not depend on  $n_1$  and  $n_2$ . The same is true for any pair of the nearest energy atomic modes. This means that all probabilities satisfy the detailed balance conditions that are found in the Gibbs distribution:

$$P_{\{n_s\}} = \frac{1}{R} e^{-\frac{2\pi g^2}{\beta} \sum_{s=1}^{\infty} s n_s} \delta \left( N - \sum_s n_s \right), \quad (17)$$

where  $s$  enumerates the atomic states according to their increasing energy ordering  $\varepsilon_s$  in Eq. (3),  $\{n_s\}$  is the vector of the final atomic  $a_s b_s$ -mode populations, and  $R$  is a normalization factor; the delta function follows from the particle conservation.

Physically, Eq. (17) would be thermal only for a linear energy dispersion,  $s = D\varepsilon_s$ , where  $D$  is the density of states. For a 2-dimensional trap geometry the atomic energy dispersion, indeed, is expected to be linear because  $D = \mathcal{S} \iint \frac{dp_x dp_y}{(2\pi)^2} \delta(E - p^2/(2m_a)) = \mathcal{S} \int_0^\infty \frac{pdp}{(2\pi)} \delta(E - p^2/(2m_a)) = \mathcal{S} m_a/(2\pi)$ , where  $m_a$  is the atomic mass,  $p_{x,y}$  are the momentum components, and  $\mathcal{S}$  is the area of the trap. Hence, for the time-linear sweep through all resonances, we predict that the atomic distribution after the molecular dissociation may be thermalized at temperature

$$k_B T = \frac{\beta}{2\pi g^2 D} = \beta / (\mathcal{S} m_a g^2). \quad (18)$$

The physical coupling  $g$  decays with  $\mathcal{S}$ , so only the linear dependence on  $\beta$  is our testable prediction. The dynamic phase transition in Fig. 1(d) now has a new interpretation: the final atomic distribution coincides with the equilibrium one for free bosons, which form a BEC below a critical temperature. Indeed, the first encountered resonance is macroscopically populated if according to Eq. (14) we have  $f > 1$ , which corresponds to the condensation of atoms in this mode.

We also mention an intriguing similarity of this thermalization with simulations of Unruh temperature

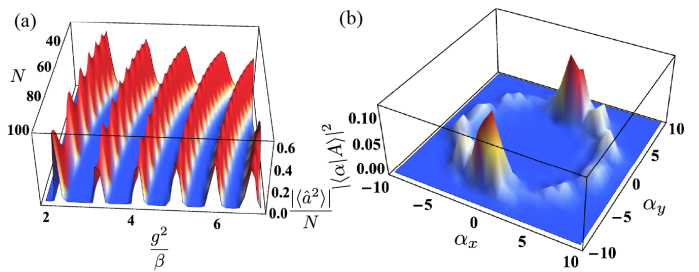


FIG. 2. (a) The ratio  $|\langle A|\hat{a}^2|A\rangle|/N$  oscillates for varying  $\beta$  and  $N$ . (b) The overlap of  $|A\rangle$  with Glauber states  $|\alpha\rangle$ ,  $\alpha = \alpha_x + i\alpha_y$ , reveals two peaks. Here, the parameters for  $|A\rangle$  are:  $\phi_M = 0$ ,  $|\alpha_m| = g^2/\beta = 5$ , which corresponds, on average, to 50 atoms in the final state.

viewed in an accelerating reference frame by a parametrically modulated BEC [27]. However, unlike [27], our thermalization is found for our system globally, without thermalizing out the state of unobserved atoms.

Finally, we comment on the role of degenerate channels, e.g., with  $\varepsilon_s = \varepsilon_{s+1}$  for some  $s$ . The probability

$$P(n) \equiv \sum_{n_s + n_{s+1} = n} (P_{n_s} + P_{n_{s+1}}) \quad (19)$$

is a continuous function of  $\delta\varepsilon \equiv \varepsilon_{s+1} - \varepsilon_s$  in the vicinity of  $\delta\varepsilon = 0$ . Indeed, if  $\delta\varepsilon$  does not change sign, this follows from integrability, and changing sign of  $\delta\varepsilon$  merely corresponds to changing channel indices  $s$  and  $s+1$ , which does not affect  $P(n)$ . Hence, the net number of atoms produced in both modes at  $\delta\varepsilon = 0$  is the same as for  $\delta\varepsilon \neq 0$ . Inductively, we find that the probability to find  $n$  atomic pairs in an arbitrarily degenerate atomic mode is the same as when such modes are non-degenerate. This means that if we coarse-grain energy into sufficiently large intervals the effect of degeneracy on the atomic energy distribution will be suppressed after the characteristic time  $\tau_{\text{eff}}$  of the passage through the resonance.

*Phase coherence.* The system after the reaction is still described by a coherent state vector with possibly non-trivial quantum correlations. This can be revealed by measuring the relative phases of the final states, which can also be found exactly. In [24], we show how such phases can be derived but we postpone the detailed analysis to a follow-up work, and consider here only the adiabatic limit in order to illustrate possible future research directions.

Let the molecular condensate be in a coherent state, which is a good approximation for an equilibrated bosonic system [28]:

$$|\alpha_m\rangle = e^{-|\alpha_m|^2/2} \sum_{n=0}^{\infty} \frac{\alpha_m^n}{\sqrt{n!}} |n\rangle. \quad (20)$$

The condensate phase  $\phi_M$  is then defined from  $\langle \alpha_m | \hat{\Psi} | \alpha_m \rangle \sim \sqrt{N} e^{i\arg(\alpha_m)}$ , where  $N = |\alpha_m|^2$ , so  $\phi_M =$

$\arg(\alpha_m)$ . Consider the adiabatic dissociation of this condensate into a single atomic mode:  $H = -\beta t \hat{\Psi}^\dagger \hat{\Psi} + g[\hat{\Psi}^\dagger \hat{a}^2 + \hat{\Psi}(\hat{a}^\dagger)^2]$ . Each  $|n\rangle$  in (20) is then converted to  $e^{i\phi_n}|2n\rangle_a$ , where  $a$  marks the atomic states. In [24] we derive an exact formula for the scattering phase of the complete dissociation amplitude:

$$\phi_n = 3n\pi/4 - \sum_{k=1}^n \arg\Gamma[i(g^2/\beta)(k+1)]. \quad (21)$$

For  $g^2/\beta \gg 1$  it is simplified because, for any  $x \gg 1$ ,  $\pi/4 + \arg\Gamma[ix] \approx x(\log_e x - 1)$ . As  $t \rightarrow +\infty$ , the atomic state becomes

$$|A\rangle = e^{-|\alpha_m|^2/2} \sum_{n=0}^{\infty} \frac{\alpha_m^n}{\sqrt{n!}} e^{i\phi_n} |2n\rangle_a. \quad (22)$$

This state contains information about the original molecular state. Thus, imagine that the molecular condensate has a vortex, such that  $\phi_M$  winds by  $2\pi$  around some spatial point. Let us also assume, phenomenologically, that locally the dynamics in a linear field ramp is described by our model. The topological property is then preserved after the reaction because the circulation

$$-i \int_0^{2\pi} \langle A | \frac{d}{d\phi_M} | A \rangle d\phi_M = 2\pi N \quad (23)$$

remains the same as it would be for the initial state  $|\alpha_m\rangle$ . Hence, the stimulated reaction should preserve vortex-like spatial distributions of particles and currents but the emerging atomic state has nontrivial correlations.

The state  $|A\rangle$  is far from a coherent state because  $\langle A | \hat{a} | A \rangle = 0$ . To test whether it can be close to a squeezed state, in Fig. 2(a) we plot a numerically calculated ratio  $|\langle A | \hat{a}^2 | A \rangle|/N$  as a function of  $g^2/\beta$ . Its

values close to 1 at large  $N$  indicate the emergence of the squeezed state. We find that this ratio is generally small except at narrow resonant values of width  $\sim 1/\sqrt{N}$  near  $g^2 \log_e(g^2 N/\beta)/\beta = 2\pi n$ ,  $n = 1, 2, \dots$ , where it exceeds 0.5. Figure 2(b) shows that the nonclassical correlations are illustrated better by the overlap of a typical  $|A\rangle$  (at non-resonant values of the parameters) with coherent states  $|\alpha\rangle$ , revealing two peaks with opposite signs of the corresponding  $\alpha$ . This makes  $|A\rangle$  akin to a macroscopic cat-state, whose origin can be traced to the even parity of all states  $|2n\rangle_a$  that contribute to  $|A\rangle$ . As  $\phi_M$  changes from 0 to  $2\pi$ , the axis connecting the two peaks rotates by angle  $\pi$  so that the peaks exchange their positions at the end. This is a topological consequence of the geometric phase Eq. (23). This cat-state must be fragile against decoherence. Breaking it down would be in agreement with breaking  $Z_2$  symmetry in the atomic phase that was predicted in [17]. This process, however, is beyond the scope of our model, so we refer to [17] for further discussion.

### ACKNOWLEDGEMENTS

The authors thank Avadh Saxena and Junyu Lin for useful discussions. This work was supported (R.K.M., V.Y.C., N.A.S.) by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division, Condensed Matter Theory Program. The completion of this work was also supported (V.Y.C and N.A.S.) by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, under Award Number DE-SC0022134. R.K.M. was partly supported by LANL Center for Nonlinear Studies. C.S. contributed to the discussion that lead to the idea of this article. He was supported by NSFC (No. 12105094) and by the Fundamental Research Funds for the Central Universities from China.

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