Superposition of macroscopic numbers of atoms and molecules

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We theoretically examine photoassociation of a non-ideal Bose-Einstein condensate, focusing on evidence for a macroscopic superposition of atoms and molecules. This problem raises an interest because, rather than two states of a given object, an atom-molecule system is a seemingly impossible macroscopic superposition of different objects. Nevertheless, photoassociation enables coherent intraparticle conversion, and we thereby propose a viable scheme for creating a superposition of a macroscopic number of atoms with a macroscopic number of molecules.

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In a now famous gedanken experiment, Schrödinger highlighted the absurdity of applying quantum theory to macroscopic objects by coupling the fate of a cat to the decay of a radioactive atom, thereby forcing the animal into a superposition of alive and dead. Over the years, this nefarious situation has come to mark the fundamental difference of principle between microscopic quantum mechanics and macroscopic realism: whereas the latter asserts that a system with two (or more) macroscopically distinct states must be in one or the other of these states, the former of course allows for superpositions of different states. The technology of superconducting quantum interference devices (SQUIDs) opened the door to the first proposed macroscopic superposition, and the latest SQUID experiments have taken a first step towards demonstrating such a state. Other candidates for creating macroscopic superpositions include nanoscale magnets, trapped ions, photons in a high-Q cavity, second-harmonic generated photons and amplitude-dispersed photons, C₆₀ molecules, and two-component Bose-Einstein condensates (BECs). While it is a matter of contention as to whether or not these states are correctly termed a Schrödinger cat, their unambiguous observation would nevertheless invalidate the concept of macroscopic realism.

Even more provocative, we propose coherent photoassociation of a BEC as a means of creating a seemingly impossible macroscopic superposition of two different objects: atoms and molecules. Photoassociation occurs when two atoms absorb a photon, thereby forming a bound molecule. Besides the strongly enhanced efficiency of molecule formation, which actually relies on an order-unity phase-space density rather than coherence, recent theoretical investigations into photoassociation of a BEC have led to a variety of phenomena involving macroscopic quantum coherence. For example, there are predictions of coherent molecular solitons, rapid and Raman adiabatic passage from an atomic to a molecular condensate, Josephson-like BEC-MBEC oscillations, and squeezed atomic condensates formed from an initial MBEC. It is, however, a somewhat subtle point that marks the difference between macroscopic coherence and a macroscopic superposition. Given a collection of N two-level systems, the macroscopic coherence that leads, e.g., to Josephson-like oscillations, where all N systems tunnel in tandem between the two states, requires only that each system is in a coherent superposition of the two levels, whereas a macroscopic superposition with all of the systems in one or the other of the two states necessarily exhibits an N-body coherence. It is exactly this distinction that warrants the present investigation.

Beyond the usual macroscopic superposition of two states of a given object, photoassociation actually leads to a more counterintuitive situation since, like (say) protons and quarks, molecules and atoms are different objects. To the best of our knowledge, the only contemporary systems with a similar potential are a BEC tuned to a Feshbach resonance, which would also give a macroscopic atom-molecule superposition, and second-harmonic generation (SHG), which would create, for example, a macroscopic superposition of red and blue photons. These three systems are of course mathematically identical, and the subsequent intramode superposition lies in the possibility for coherent atom-molecule (red-blue photon) conversion. So far, SHG has only yielded intramode phase superpositions between coherent states, but these results are somewhat ambiguous as they do not differentiate between a coherent superposition and a statistical mixture. In contrast, the present work, based on unitary evolution of a Fock state, clearly demonstrates an intramode superposition of a macroscopic number of atoms with a macroscopic number of molecules. With photoassociation currently on the verge of coherence, this state should be realizable in the next generation of experiments.

Nonetheless, one-color photoassociation generally involves an excited molecular state, and the ensuing spontaneous decay will inevitably introduce decoherence. Moreover, rogue photodissociation, i.e., irreversible bound-free transitions to non-condensate modes, will destroy coherence as well. In this Letter, we therefore consider an atom-molecule superposition formed via two-color free-bound-bound photoasso-
cation of an interacting BEC, where the primary photoassociation molecules are transferred with another laser field to a stable molecular state. Adiabatic elimination [30] of the intermediate molecular state enables an effective two-mode model involving only atoms and stable molecules [21,27], a sufficiently large intermediate detuning should eliminate the harmful effects of irreversible losses of primarily photoassociated molecules, rogue [28] or otherwise [27], and full inclusion of particle collisions—its a novel aspect—sets the table for the appearance of the desired superposition.

The development herein is outlined as follows. First, we review the reduction of the problem from three to two modes, followed by an estimate of the fixed parameters for which the neglect of irreversible decay is reasonable. We then focus on a dynamical approach to creating a macroscopic superposition, which begins with a joint atom-molecule condensate formed by the photoassociative equivalent of a phase-tailored, strong \( \pi/2 \) pulse. Switching from a laser-dominated to a collision-dominated system, the symmetric BEC-MBEC is then shown to evolve into a superposition comprised of macroscopic numbers of atoms and molecules, with a “size” determined by the ratio of the free-bound and intraparticle interactions. Finally, we discuss a possible experimental signature, as well as the effects of any additional decoherence.

Turning to the situation of Fig. 1, we assume that \( N \) atoms have Bose-condensed into the same one-particle state, e.g., a plane wave with wave vector \( \mathbf{k} = 0 \). Photoassociation then removes two atoms from this state [1], creating a molecule in the excited state [2]. Including a second laser, bound-bound transitions remove excited molecules from state [2] and create stable molecules in state [3]. In second quantized notation, boson annihilation operators for atoms, primarily photoassociated molecules, and stable molecules are denoted, respectively, by \( a, b, \) and \( g \). The laser-matter interactions that drive the atom-molecule and molecule-molecule transitions are determined by the ratio of the free-bound and intraparticle interactions.

\[
\rho = |\psi\rangle\langle \psi| = |\sigma\rangle\langle \sigma| + |\tau\rangle\langle \tau| + |\rho\rangle\langle \rho|
\]

where \( a_{ij} \) is the particle-particle scattering length, \( V_{ij}^{-1} = \int d^3r |\psi_i(\mathbf{r})|^2|\psi_j(\mathbf{r})|^2 \) is the inverse effective volume of the single particle wavefunctions \( (i,j = 1,2) \), \( m = \sum M \) is the mass of the atom (molecule), and \( \mu = 2m/3 \) is the reduced mass of the atom-molecule pair. Note that, due to the photoassociating laser [31,21,27], the atom-atom scattering length in Eq. (3a) has also acquired a complex light shift.

Based generally on ratios of parameters, a quantum optics formalism is a definite plus here since the molecular-molecule and atom-molecule scattering lengths are unknown. Hence, we specify the magnitude of the intermediate detuning as \( |\delta| \gg \Gamma \), allowing for neglect of the imaginary terms in \( \Delta' \) and \( \lambda_1 \). To reduce the number of parameters we also set \( \lambda_2 = 4 \lambda_1 \), where the factor of four arises from the conserved particle number being equal to the number of atoms plus twice the number of molecules (see below). Exploiting the light-shifted scattering length, this equality could in practice be achieved by setting \( \Delta = \Delta' + \lambda_1 \). Choosing \( \Delta = \Delta' + \lambda_1 \) for the BEC volume, \( \Delta = \pi \lambda \) is the wavelength of the photoassociating light, \( \epsilon_R = (2m \lambda)^{-1} \) is the photon recoil frequency, and we have used [27] \( \kappa/\epsilon_R = (\lambda^3/V)(I_1/I_0)^{1/2} \). The obscure numerical factor comes from the definition of the characteristic intensity \( I_0 \) for a given photoassociation transition [27]. Focusing on \(^{87}\)Rb, we have [21,27] \( I_0 = 0.07 \text{mW/cm}^2 \), \( \Delta = 780 \text{nm} \), \( \epsilon_R = 3.77 \times 10^3 \text{kHz} \), \( \Gamma = 12 \times 2\pi \text{MHz} \), and \( a_{11} = 5 \text{nm} \).
Estimating $V[a_{11}V_{11}^{-1} - 2a_{22}V_{22}^{-1}] = 10 a_{11}$, the fixed parameters are $|\delta| \approx 10^{3} \times 2\pi$ MHz and $I_1 \approx 10$ W/cm².

Within the two-mode approximation, the total number of particles $N = a_1^\dagger a_1 + 2g_1^\dagger g_1$ is conserved and, without altering the physics, we may subtract the term $\lambda (N^2 - N)$ from $H_2$, resulting in the simplified form

$$H_2 = H_0 - \frac{1}{2} \chi (e^{-i\varphi} a_1^\dagger a_1 g + e^{i\varphi} g_1^\dagger a g) - 2\lambda a_1^\dagger ag_1^\dagger g,$$

(6)

where $H_0 = -\Delta g_1^\dagger g_1$, $\lambda = 2\lambda_1 - \lambda_2$, and a factor of $\lambda_1$ has been absorbed into the detuning. Furthermores, $\chi$ is hereafter a real quantity, and $\varphi$ specifies the relative phase between the free-bound and detuning. The main difference from the dual atomic condensate problem [13,14] is that the photoassociative coupling is trilinear, and the “tunneling rate”, as opposed to being $N$-independent, therefore scales as $\sqrt{N}$. Comparing $\sqrt{N} \chi$ to $N \lambda$, the qualitative condition that determines a collision-dominated system is $\chi / \lambda \ll \sqrt{N}$, where the ratio $\chi / \lambda$ is readily adjustable according to bound-bound intensity $I_2$.

Considering the formation of a macroscopic superposition of atoms and molecules, we strongly suspect that a superposition as a ground state [14] is absent due to the lack of particle-exchange symmetry for the photoassociation Hamiltonian (6). Hence, we take a dynamical approach [14], which begins with the production of a joint atom-molecule condensate having an “evenly” split population (i.e., $N/2$ atoms and $N/4$ molecules), and a real-amplitude. Setting the relative phase between the lasers to $\varphi = \pi / 2$, this state [shown in Fig. 2(a)] is formed by a strong ($\chi / \lambda \gg \sqrt{N}$) two-photon resonant photoassociation pulse lasting a duration $t$ determined by the solution to the semiclassical approximation [22] for the number of molecules: $N/4 = (N/2) \tanh^2 (\sqrt{2N} \chi t)$. Interestingly enough, rather than the binomial distribution of particles obtained for dual atomic condensates [14], the trilinear free-bound interaction leads to a squeezed atom-molecule condensate. Such squeezing occurs exactly as in SHG [8], and is a source of many-particle entanglement [22,22].

If, upon preparation of the dual atom-molecule condensate, the lasers are adjusted so that $\varphi = 0$, $\Delta' \approx \sqrt{N} \chi$, and $\chi / \lambda \ll \sqrt{N}$, the system will, as shown in Fig. 2(b), evolve into a macroscopic atom-molecule superposition over a timescale set by the Bose-enhanced two-photon Rabi coupling $\sqrt{N} \chi$. The lack of smoothness in the probability distribution increases with particle number, and occurs due to the nonlinear nature of the system. Furthermore, the “size” of the superposition, defined by the separation between the main peaks, depends on the ratio $\chi / \lambda$, and for a large number of particles is a maximum $\sim N/2$ when $\chi / \lambda \approx \frac{1}{2} \sqrt{N}$. Consistent with the lack of particle-exchange symmetry, the formation of this state depends on the relative sign of $\chi$ and $\lambda$, which may be flipped if needed by setting $\varphi = \pi$.

Referring to Fig. 2(c), because the system returns approximately to the initial dual condensate, one could, for example, imprint a phase on the molecular component of the system, leading to a signature interference effect. Crucially, the rate of phase variations in the amplitude $C_n(t)$, dictated by the collisional interference term, is large only near $n = N/4$ molecules, so that the subsequent decoherence from a temporally imprecise imprinting of the phase should not effect a large superposition. The main obstacle to realization is then external decoherence arising from interactions with the ubiquitous thermal cloud of atoms; however, because the BEC-decoherence arising from interactions with the ubiquitous thermal cloud of atoms; however, because the BEC-decoherence arising from interactions with the ubiquitous thermal cloud of atoms; however, because the BEC-decoherence arising from interactions with the ubiquitous thermal cloud of atoms; however, because the BEC-decoherence arising from interactions with the ubiquitous thermal cloud of atoms.

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FIG. 1. Three-level illustration of coherent free-bound-bound photoassociation, where $N$ atoms have assumedly Bose-condensed into state $|1\rangle$. The free-bound and bound-bound Rabi frequencies are $\kappa$ and $\Omega$, respectively. Similarly, the two-photon and intermediate detunings are $\Delta$ and $\delta$. The wavy line denotes the irreversible losses that a large $|\delta|$ is expected to combat.

FIG. 2. Creation of a macroscopic superposition of atoms and molecules by photoassociating a BEC of atoms. The frequency scale is defined by $\lambda = 1$, the dimensionless time is $\tau = \sqrt{N}\chi(t - \bar{t})$, and the initial state is $|\psi_i\rangle = |N, 0\rangle$ with $N = 5000$ atoms and $n = 0$ molecules. (a) For $\varphi = \pi/2$, a strong and two-photon resonant photoassociation pulse forms a symmetric joint atom-molecule condensate with a real amplitude: $|\psi(\tau = 0)\rangle = \sum_{n=0}^{N/2} C_n(0) |N - 2n, n\rangle$, $\text{Im}[C_n(0)] = 0$. Switching to $[\varphi, \chi, \Delta'] = [0, 0.1\sqrt{N}, 0.1N]$, the system evolves (b) into a coherent superposition of atoms and molecules and (c) back to a symmetric atom-molecule condensate.