Rapid phase-diffusion between atomic and molecular Bose-Einstein condensates

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We study the collisional loss of atom-molecule coherence after coherently dissociating a small fraction of a molecular Bose-Einstein condensate into atoms. The obtained n-atoms states are two-atom (SU(1,1)) coherent states with number variance \( \Delta n \propto n \) compared to \( \Delta n \propto \sqrt{n} \), for the spin (SU(2)) coherent states formed by coherent splitting of an atomic condensate. Consequently, the Lorentzian atom-molecule phase-diffusion is faster than the Gaussian phase-diffusion between separated atomic condensates, by a \( \sqrt{n} \) factor.

Atom-molecule coherence in a Bose-Einstein condensate (BEC) was first demonstrated experimentally by observing coherent oscillations in a Ramsey-like interferometer [1]. Its existence paves the way to a wealth of novel phenomena, including large-amplitude atom-molecule Rabi oscillations [2], Atom-Molecule dark states [3], and 'super-chemistry' [4] characterized by collective, Bose-enhanced and ultraselective dynamics.

One important implication of atom-molecule coherence, is the stimulated dissociation of a molecular BEC into its constituent boson atoms [2]. This coherent process is the matter-wave equivalent of parametric down-conversion. Like its quantum-optics counterpart, the stimulated dissociation, starting from two pulses of atom-molecule coupling, separated by a phase-acquisition period, similar to the Ramsey procedure in [1] but starting from a molecular BEC instead of an atomic one, the stimulated dissociation is phase-sensitive for atomic states different than a molecular BEC and the emerging atomic condensate, as the conjugate phase variance \( \Delta \varphi \) is exponentially decreasing. Also like optical parametric amplification, the stimulated dissociation induces a state transition between the two coherent states.

The exponential growth of \( \Delta n \) indicates the formation of a well defined relative-phase \( \varphi \) between the molecular BEC and the emerging atomic condensate, as the conjugate phase variance \( \Delta \varphi \) is exponentially decreasing. This relative-phase \( \varphi \) between molecules and atoms, determines whether it will be amplified or attenuated.

In this work we propose to use the phase-sensitivity of the stimulated dissociation of a molecular BEC, to implement a sub-shot-noise SU(1,1) interferometer [3]. The scheme involves two pulses of atom-molecule coupling, separated by a phase-acquisition period, similar to the Ramsey procedure in [1] but starting from a molecular BEC instead of an atomic one. In the limit where the dissociation does not deplete the molecular BEC, the atomic state will be an SU(1,1) coherent state (TACS). Our main result is that the \( \Delta n \propto n \) atom-number variance of the TACS results of the loss of atom-molecule phase coherence on a short \( \tau_{pd} \propto 1/\sqrt{n} \) timescale due to collisional phase-diffusion. By contrast, two initially coherent, separated atom-molecule condensates phase-diffuse on a longer \( \tau_{pd} \propto 1/\sqrt{n} \) timescale [3], since their initial state is an SU(2) or 'spin' coherent state (SCS) with \( \Delta n \propto \sqrt{n} \). Moreover, we find that for \( n \gg 1 \) the phase-diffusion of the TACS is Lorentzian in time, as compared to the familiar Gaussian phase-diffusion of the SCS, due to the difference in atom-number distributions between the two coherent states.

We consider the atom-molecule model Hamiltonian, where interacting atoms and molecules are coupled by means of either a Feshbach resonance or a resonant Raman transition,

\[
H = E_m \hat{n}_m + E_a \hat{n} + \left( g_{am} \hat{\psi}_m^\dagger \hat{\psi}_a + H.c. \right) + \frac{u_m}{2} \hat{\psi}_m^\dagger \hat{\psi}_m^\dagger \hat{\psi}_m \hat{\psi}_m + \frac{u_a}{2} \hat{\psi}_a^\dagger \hat{\psi}_a^\dagger \hat{\psi}_a \hat{\psi}_a + u_{am} \hat{n}_m \hat{n}_a,
\]

where \( \hat{\psi}_{am} \) are the boson annihilation operators for atoms and molecules, \( \hat{n}_m = \hat{\psi}_m^\dagger \hat{\psi}_m \) are the corresponding particle numbers, and \( E_{am} \) are the respective mode energies. The atom-molecule coupling is \( g_{am} = |g_{am}| e^{i\phi} \) whereas \( u_m, u_a \), and \( u_{am} = u_{ma} \) are the collisional interaction strengths for molecule-molecule, atom-atom, and atom-molecule scattering, respectively.

In what follows we shall assume that the molecular condensate remains large and is never significantly depleted by the conversion of a small number of molecules into atoms. This approximation is equivalent to the undepleted pump approximation in parametric down-conversion. The molecular field operators \( \hat{\psi}_m, \hat{\psi}_m^\dagger \) are replaced by the c-numbers \( \sqrt{n_m} e^{i\phi} \) and Eq. (1) becomes,

\[
H = \delta \hat{K}_z + g \hat{K}_x + u \hat{K}_z^2,
\]

where c-number terms are omitted. Here \( \delta = (E_m - 2E_a + 2u_m n_m - 2u_a), \ g = 4 |g_{am}| \sqrt{n_m}, \) and \( u = 2u_a \). The operators \( \hat{K}_z = (e^{i(\phi_m - \phi)})/2) \hat{\psi}_m^\dagger \hat{\psi}_m^\dagger, \) \( \hat{K}_x = (e^{-i(\phi_m - \phi)})/2) \psi^\dagger \psi/2 + 1/4 \) are the generators of an SU(1,1) Lie algebra with canonical commutation relations \( [\hat{K}_+, \hat{K}_-] = -2\hat{K}_z, \) \( [\hat{K}_z, \hat{K}_z] = \pm \hat{K}_z \) and we define the usual Hermitian operators \( \hat{K}_x = (\hat{K}_+ + \hat{K}_-)/2, \) \( \hat{K}_y = (\hat{K}_+ - \hat{K}_-)/2i. \) Since the Casimir operator of SU(1,1) is \( \hat{C} = \hat{K}_z^2 - \hat{K}_x^2 - \hat{K}_y^2, \) we will use for representation the joint eigenstates of \( \hat{C} \) and \( \hat{K}_z, \)

\[
|k, m\rangle = \sqrt{\frac{\Gamma(2k)}{m!\Gamma(2k + m)}} (\hat{K}_z)^m |0, 0\rangle
\]

so that \( \hat{C}|k, m\rangle = (k + m - 1)|k, m\rangle \) and \( \hat{K}_z |k, m\rangle = (k + m)|k, m\rangle, \) with the Bargmann index \( k = 1/4 \) and non-
Lorentzian boost of duration photodissociation lasers. The atomic state following this the optical resonant Raman coupling, by switching the magnetic control of the atom-molecule detuning and for 

$$g$$ fraction of the molecular BEC into atoms, by setting (Fig. 1(a)), the first step is the dissociation of a small shots of the quadrature plane \(\hat{\phi} \). The quadrature phase-amplitude distribution is shown at the polar angle in the

$$\theta, \phi$$ , whereas if \(\varphi_{h} = \pi\) it will reassociate all atoms into it. The final number of atoms is obtained by noting that the combined boost-rotation-boost sequence \(e^{-i\varphi_{h}\hat{K}_{z}}e^{-i\varphi_{m}\hat{K}_{z}}e^{-i\varphi_{p}\hat{K}_{z}}\) preserves coherence and transforms the vacuum into the final TACS \(|\theta_{f}, \varphi_{f}\rangle\) with 

$$\cosh \theta_{f} = 1 + \cos \varphi_{h}\cosh^{2} \theta_{p} - \cos(\varphi_{h})$$. Hence in the absence of collisions,

$$\Delta n_{f} = 2k(\cos \theta_{f} - 1) = \frac{1 + \cos \varphi_{h}}{2} \sinh^{2} \theta_{p},$$

$$\Delta n_{f}^2 = 2k \sinh \theta_{f} \Delta n_{f}^2 = \frac{\sinh \theta_{p}}{2} \left[ \sinh^{2} \varphi_{h} + (1 + \cos \varphi_{h})^{2} \cosh^{2} \theta_{p} \right].$$

Note these expressions are slightly different than in Ref. [6] because the proposed scheme uses two identical, equal phase pulses, as opposed to the reversed Lorentzian boosts of the two degenerate parametric amplifiers in [6].

From Eqs. (5) it is clear that an accumulated phase \(\varphi_{h} = \pi\) may be determined within \((\Delta \varphi_{h})^2 = \left((\Delta n_{f})^2 / |\partial n_{f} / \partial \varphi_{h}|^2 \right)_{|\varphi_{h}=\pi} = (2 \sinh^{2} \theta_{p})^{-1} = [8n(n + 1)]^{-1}\) accuracy. Thus due to the squeezing inherent in coherent dissociation, \(\Delta \varphi_{h}\) around \(\varphi_{h} = \pi\) goes below the \(1/\sqrt{n}\) standard quantum limit (a.k.a. shot-noise limit) and approaches the Heisenberg \(1/n\) uncertainty, where \(n\) is the number of atoms dissociated by the first pulse [6].

Our goal here is to study the effect of interactions on this scenario. Atom-atom and molecule-atom collisions will degrade atom-molecule coherence during the phase acquisition time since for non-vanishing \(u\) the pertinent \(|k, m\rangle\) eigenstates are not equally spaced. This collisional dephasing drives the quadrature variances to 

\((\Delta X)^2 = (\Delta P)^2 = 2n + 1\), while keeping 

\((\Delta X)^2 + (\Delta P)^2 = 2(2n + 1)\) fixed, as depicted by the dotted circle in Fig. 1(c). Phase information is lost and the final atom number on invoking the second pulse is \(\varphi_{h}\)-independent (dotted ellipse in Fig. 1(d)).
**FIG. 2**: (Color online) Comparison of atom-molecule phase-diffusion with the collisional dephasing of separated atomic condensates: (a) number distribution of a TACS $|\theta,\varphi\rangle$ with $\theta = 4.8$, corresponding to $n = 30$ dissociated atoms; (b) same for a SCS $|\pi/2,\varphi\rangle_s$, with $\ell = n/2 = 25$; (c) phase-diffusion of TACS for $n = 100$ (solid, $\circ$), 167 (dashed, $\square$), and 500 (dash-dotted, $\triangle$), symbols mark numerical results with $n \pm 2n_{\text{m}} = 5000$; (d) same for SCS with $n = 70$ (solid, $\circ$), 156 (dashed, $\square$), and 626 (dash-dotted, $\triangle$), symbols mark numerical results. Insets in (b) and (c) show the decay half-times $\tau_{\text{pd}} \propto (\text{un})^{-1}$ for TACS and $\tau_{\text{pd}} \propto (\text{un})^{-1}$ for SCS.

Atom-molecule coherence may be quantified by defining the SU(1,1) purity $K^2 \equiv \langle \hat{K}_z \rangle^2 - \langle \hat{K}_x \rangle^2 - \langle \hat{K}_y \rangle^2$. For an SU(1,1) coherent state we have $K = k$ whereas dephasing is characterized by going inside the upper sheet of the hyperboloid $K^2 = k^2$, so that $K > k$. Thus, during the time $t_0$ hold time where $g = 0$ and hence $\langle \hat{K}_z \rangle$ is fixed, we may use $K^2 = \langle \hat{K}_z \rangle^2 + \langle \hat{K}_y \rangle^2$ as a measure of coherence. The time dependence of $K_\perp$ is related to the Fourier transform of the initial number distribution. Starting from the TACS $|\theta,\varphi\rangle$ with the number distribution $P_m = |\langle k, m | \theta, \varphi \rangle|^2$ shown in Fig. 2(a), we find the exact result that in the presence of interactions, $K_\perp$ is independent of $\varphi$, $\delta$ and decays as

$$K_\perp(t) = \frac{k \sinh \theta}{1 + \sin^2(ut) \sin^2 \theta} e^{-\frac{\theta}{k}}. \tag{6}$$

Noting that $\sinh^2 \theta = (n/2k)[(n/2k) + 2] = 4n(n + 1)$ we obtain that for a moderately large $n \gg 1$, coherence decays on a $\text{sin}(\text{ut}) \sim 1/(2n)$ timescale. Thus we replace $\sinh \theta \approx 2n$, $\sin(\text{ut}) \approx \text{ut}$ to obtain Lorentzian dephasing $K_\perp = (n/2)[1 + (2\text{ut})^2]^{-3/4}$ which reflects the exponential form of $P_m$ and agrees well with numerical simulations (Fig. 2(c)). The phase-diffusion time $\tau_{\text{pd}} = 1/(2\text{un})$ reciprocates the super-Poissonian $\Delta n \propto n$ variance of the TACS.

It is instructive to compare atom-molecule collisional dephasing with phase diffusion between two initially coherent atomic BECs $|\theta,\varphi\rangle_s$. The pertinent Hamiltonian is the two-site Bose-Hubbard model (sometimes referred to as the Bosonic Josephson junction $|11\rangle$) and the initial coherent states are the SU(2) SCS $|\theta,\varphi\rangle_s$,

$$|\theta,\varphi\rangle_s = \exp(z\hat{L}_+ - z^*\hat{L}_-)(\ell, -\ell)$$

$$= \left[1 + \xi^2\right]^{-\ell} \sum_{\ell = m = -m} \left(\xi e^{-i\varphi}\right)^{\ell + m} \left(\frac{2\ell}{\ell + m}\right)^{1/2}(\ell, m), \tag{7}$$

where $\xi = \tan(\theta/2)$. The SU(2) generators $\hat{L}_\pm = (\hat{\psi}_1^\dagger \hat{\psi}_2 + \hat{\psi}_2^\dagger \hat{\psi}_1)/\sqrt{2}$, $\hat{L}_y = (\hat{\psi}_1^\dagger \hat{\psi}_2 - \hat{\psi}_2^\dagger \hat{\psi}_1)/(2i)$, and $\hat{L}_z = (\hat{n}_1 - \hat{n}_2)/2$, are defined in terms of the boson annihilation and creation operators $\hat{\psi}_i$, $\hat{\psi}_i^\dagger$ for particles in condensate $i = 1, 2$ with the number operators $\hat{n}_i = \hat{\psi}_i^\dagger \hat{\psi}_i$. The total particle number $\hat{n} = \hat{n}_1 + \hat{n}_2 = 2\ell$ is conserved and the Fock states $(\ell, m)$ are the standard $L^2$, $L^2$ eigenstates. Experimentally, such states are prepared either by coherently splitting an atomic BEC or by controlling optical or magnetic double-well potentials confining it $[11,10]$. Most common are states with equal population of the two condensates, i.e. $\theta = \pi/2$.

The binomial/Poissonian number distribution of the SCS $|\theta,\varphi\rangle_s$ (Fig. 2(b)) results in the loss of relative-phase coherence $(\langle L_\perp \rangle)_s \equiv (\langle L_x \rangle^2 + (\langle L_y \rangle)_s^2)$ under a collisional $\Delta L_z + u\Delta L_z$ Hamiltonian, as

$$L_\perp(t) = \ell \sin \theta \left(1 - \sin^2(ut) \sin^2 \theta\right)^{\ell - 1/2}, \tag{8}$$

approaching for $n \gg 1$, the Gaussian decay $L_\perp = (n/2) \sin \theta e^{-n(\sin \theta \text{ut})^2/2}$ with phase-diffusion time $\tau_{\text{pd}} = (u \sin \theta \sqrt{n/2})^{-1}$ (Fig. 2(d)). For equal $n$, the loss of atom-molecule coherence is thus typically $\sqrt{n}$ times faster than the phase-diffusion between atomic BECs. We note that the accelerated decay of the super-Poissonian, phase-squeezed SU(1,1) coherent state, is the counterpart of the decelerated phase-diffusion of a sub-Poissonian SU(2) number-squeezed states, observed experimentally in Ref. 10.

To demonstrate the effect of interactions on the SU(1,1) interferometer, we find the final atom number $n_f(\varphi_h)$ with phase-diffusion present during the hold time,

$$n_f = 2k \left\{1 + \frac{\cos \Phi_h}{[1 + \sin^2(\text{ut}) \sin^2 \theta_p]^{1/2}}\right\} \sin^2 \theta_p, \tag{9}$$

where $\Phi_h = \varphi_h + (2k+1) \arctan[\cosh \theta_p \tan(\text{ut})]$. An exact form is also found for $\Delta n_f$. The Ramsey-like fringes are thus shifted due to the collisional shift in the atomic energy, and attenuated due to the loss of atom-molecule coherence (Fig. 3). They vanish on a $\tau_{\text{pd}}$ timescale, approaching the fixed value $n_f = 2k \sinh^2 \theta_p$ (which corresponds to the state depicted by a dotted ellipse in Fig. 3(d)). It is also evident from Eq. 9 and Eq. 9 that
coherence revives on a very long $\tau_r = \pi/u$ timescale, similarly to the SU(2) case \cite{8,9}.

To conclude, the dissociation of molecular BECs holds great potential for the construction of Heisenberg limited SU(1,1) interferometers, due to the inherent phase-squeezing of the TACS. However, phase-squeezing comes at the price of a super-Poissonian $\Delta n \sim n$ number distribution, making the TACS very sensitive to collisional phase-diffusion. The same observation holds true for the SU(2) phase-squeezed states produced by rotation of number-squeezed inputs, in proposals for sub-shot-noise Mach-Zender atom interferometry \cite{6,12}. Controlling this dephasing process will pose a major challenge to the implementation of precise atom interferometers, as well as to the realization of coherent superchemistry \cite{4,5}.

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