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 [5]. This coherent process is the matter-wave equivalent of parametric down-conversion. Like its quantum-optics counterpart, when started from the atomic vacuum (molecular BEC) it involves the hyperbolic amplification of the atom-pair number $n = \langle \hat{n} \rangle$ and of its variance $\Delta n = \left(\langle \hat{n}^2 \rangle - \langle \hat{n} \rangle^2\right)^{1/2}$, where $\hat{n}$ is the atomic number operator.

The exponential growth of Δn indicates the formation of a well defined relative-phase φ between the molecular BEC and the emerging atomic condensate, as the conjugate phase variance $\Delta\varphi$ is exponentially decreasing. Also like optical parametric amplification, stimulated dissociation is *phase-sensitive* for atomic states different than the vacuum state. Given a non-vanishing value of n The relative-phase φ 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 [6]. The scheme involves two pulses of atom-molecule coupling, separated by a phase-aquisition 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) or 'two-atom' coherent state (TACS). Our main result is that the $\Delta n \propto n$ atom-number variance of the TACS results in the loss of atom-molecule phase coherence on a short $\tau_{pd} \propto 1/n$ timescale due to collisional phase-diffusion. By contrast, two initially coherent, separated atomic condensates phase-diffuse on a longer $\tau_{pd} \propto 1/\sqrt{n}$ timescale [8], 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,

$$\begin{aligned} H &= E_m \hat{n}_m + E_a \hat{n} + \left(g_{am} \hat{\psi}_m^\dagger \hat{\psi}_a \hat{\psi}_a + H.c. \right) \\ &+ \frac{u_m}{2} \hat{\psi}_m^\dagger \hat{\psi}_m^\dagger \hat{\psi}_m \hat{\psi}_m + \frac{u_a}{2} \hat{\psi}_a^\dagger \hat{\psi}_a^\dagger \hat{\psi}_a \hat{\psi}_a + u_{am} \hat{n}_m \hat{n}, \end{aligned} \quad (1)$$

where $\hat{\psi}_{a,m}$ are the boson annihilation operators for atoms and molecules, $\hat{n} = \hat{\psi}_a^\dagger \hat{\psi}_a$, $\hat{n}_m = \hat{\psi}_m^\dagger \hat{\psi}_m$ are the corresponding particle numbers, and $E_{a,m}$ 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 downconversion. The molecular field operators $\hat{\psi}_m, \hat{\psi}_m^\dagger$ are replaced by the c-numbers $\sqrt{n_m} e^{\pm i\phi_m}$ and Eq. (1) becomes,

$$H = \delta \hat{K}_z + g \hat{K}_x + u \hat{K}_z^2, \quad (2)$$

where c-number terms are omitted. Here $\delta = (E_m - 2E_a + 2u_{am} n_m - 2u_a)$, $g = 4|g_{am}|\sqrt{n_m}$, and $u = 2u_a$. The operators $\hat{K}_+ = (e^{i(\phi_m - \phi)}/2)\psi^\dagger\psi^\dagger$, $\hat{K}_- = (e^{-i(\phi_m-\phi)}/2)\psi\psi$, $\hat{K}_z = \psi^\dagger\psi/2 + 1/4$ are the generators of an SU(1,1) Lie algebra with canonical commutation relations $[\hat{K}_+, \hat{K}_-] = -2\hat{K}_z$, $[\hat{K}_z, \hat{K}_\pm] = \pm\hat{K}_\pm$ 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 $\hat{K}_z$,

$$|k, m\rangle = \sqrt{\frac{\Gamma(2k)}{m!\Gamma(2k+m)}} (\hat{K}_+)^m |k, 0\rangle \quad (3)$$

so that $\hat{C}|k, m\rangle = k(k-1)|k, m\rangle$ and $\hat{K}_z|k, m\rangle = (k + m)|k, m\rangle$, with the Bargmann index $k = 1/4$ and non-

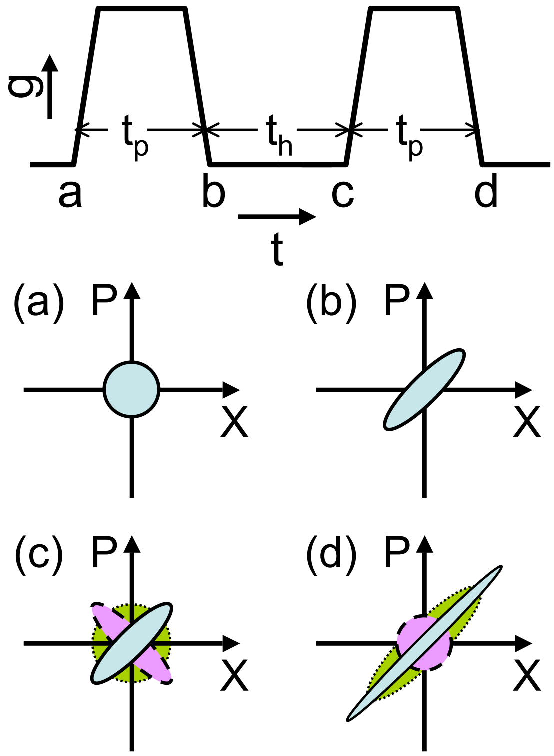

FIG. 1: (Color online) Atom-molecule SU(1,1) interferometer. The quadrature phase-amplitude distribution is shown at the time points marked on the upper $g(t)$ plot. Note that the polar angle in the X, P plot is $\varphi/2$, not φ. Starting from the atomic vacuum (a) the first Lorentzian boost results in the squeezing of the atom-molecule phase around $\varphi = \pi/2$ (b), which is allowed to evolve during the hold time (c). The atom number and its variance after the second pulse (d) depend on the value of φ acquired during the hold time. When φ remains $\pi/2$ (solid line) the second pulse yields further squeezing with exponentially increasing n whereas if $\varphi = -\pi/2$ (dashed line) the atomic vacuum is recovered. Dotted circles correspond to the loss of coherence due to φ phase-diffusion.

negative integer m. The states $|k, m\rangle$ are atom number states with $n = 2m$.

The SU(1,1) interferometer [6] for probing the atom-molecule phase coherence, is illustrated in Fig. 1 by snapshots of the quadrature plane $\hat{X} = \psi + \psi^\dagger$, $\hat{P} = (\psi - \psi^\dagger)/i$. Starting from the coherent atomic vacuum state $|k, 0\rangle$ (Fig. 1(a)), the first step is the dissociation of a small fraction of the molecular BEC into atoms, by setting $g \gg \delta, un$. This is attained for Feshbach-coupling, by magnetic control of the atom-molecule detuning and for the optical resonant Raman coupling, by switching the photodissociation lasers. The atomic state following this Lorentzian boost of duration t_p, is an SU(1,1) TACS [6, 7],

$$
\begin{aligned}
|\theta, \varphi\rangle_s &= \exp(z\hat{K}_+ - z^*\hat{K}_-)|k, 0\rangle \qquad (4) \\
&= \left[1 - \zeta^2\right]^k \sum_m \left[\zeta e^{-i\varphi}\right]^m \sqrt{\frac{\Gamma(2k+m)}{m!\Gamma(2k)}}|k, m\rangle \ ,
\end{aligned}
$$

with $z = e^{-i\varphi}\theta/2$ and $\zeta = \tanh(\theta/2)$. The obtained squeeze parameter is $\theta = \theta_p \equiv gt$, and the atom-molecule relative phase is $\varphi = \phi - \phi_m + 2\phi_a = \pi/2$ (corresponding to quadrature phase of $\pi/4$, see Fig. 1(b)). The average atom number of $|\theta, \varphi\rangle$ is $n = 2k(\cosh\theta - 1)$ and its variance is $\Delta n = \sqrt{2k}\sinh\theta$ [7], corresponding to the amplification of vacuum fluctuations in stimulated dissociation [5].

Next, the coupling g is turned off and the atom-molecule phase is allowed to evolve for a hold-time t_h. In the limit where atom-atom and atom-molecule collisions may be neglected ($u = 0$), coherence is maintained and the state at the end of the hold time is $\exp(-i\delta\hat{K}_z t_h)|\theta_p, \pi/2\rangle = |\theta_p, \pi/2 + \varphi_h\rangle$ with $\varphi_h \equiv \delta t_h$ (Fig. 1(c)). The accumulated atom-molecule phase φ_h may be determined by a second strong coupling pulse of duration t_p (Fig. 1(d)) because the fraction of reassociated atoms is phase-sensitive [6]. For example, if $\varphi_h = 0$ the second pulse will further dissociate the molecular BEC, 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 $e^{-i\theta_p\hat{K}_x}e^{-i\varphi_h\hat{K}_z}e^{-i\theta_p\hat{K}_x}$ 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,

$$
\begin{aligned}
n_f &= 2k(\cosh\theta_f - 1) = \frac{1 + \cos\varphi_h}{2}\sinh^2\theta_p \ , \\
(\Delta n_f)^2 &= 2k\sinh^2\theta_f \qquad (5) \\
&= \frac{\sinh^2\theta_p}{2}\left[\sin^2\varphi_h + (1 + \cos\varphi_h)^2\cosh^2\theta_p\right] \ .
\end{aligned}
$$

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 φ_h-independent (dotted ellipse in Fig. 1(d)).

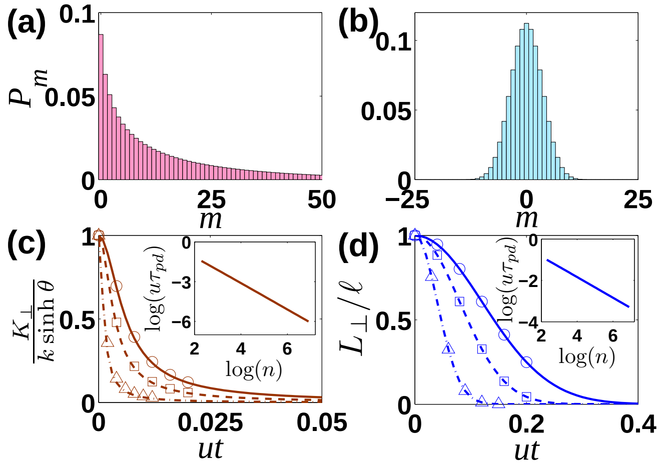

FIG. 2: (Color online) Comparison of atom-molecule phase-diffusion with the collisional dephasing of separated atomic condensates: (a) number distribution of a TACS $|\theta,\varphi\rangle$ with $\theta = 4.8$, corresponding to $n = 30$ dissociated atoms; (b) same for a SCS $|\pi/2,\varphi\rangle_s$ with $\ell = n/2 = 25$; (c) phase-diffusion of TACS with $n = 100$ (solid, ○), 167 (dashed, □), and 500 (dash-dotted, △), symbols mark numerical results with $n + 2n_m = 5000$; (d) same for SCS with $n = 70$ (solid, ○), 156 (dashed, □), and 626 (dash-dotted, △), symbols mark numerical results. Insets in (b) and (c) show the decay half-times $\tau_{pd} \propto (un)^{-1}$ for TACS and $\tau_{pd} \propto (u\sqrt{n})^{-1}$ 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 t_h hold time where $g = 0$ and hence $\langle\hat{K}_z\rangle$ is fixed, we may use $K_\perp^2 \equiv \langle\hat{K}_x\rangle^2 + \langle\hat{K}_y\rangle^2$ 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 φ , δ and decays as,

$$K_\perp(t) = \frac{k\sinh\theta}{\left[1+\sin^2(ut)\sinh^2\theta\right]^{k+1/2}} \; . \tag{6}$$

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 $\sin(ut) \sim 1/(2n)$ timescale. Thus we replace $\sinh\theta \approx 2n$, $\sin(ut) \approx ut$ to obtain Lorentzian dephasing $K_\perp = (n/2)[1+(2nut)^2]^{-3/4}$ which reflects the exponential form of P_m and agrees well with numerical simulations (Fig. 2(c)). The phase-diffusion time $\tau_{pd} = 1/(2un)$ 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 [8, 9, 10]. The pertinent Hamiltolian is the two-site Bose-Hubbard model (sometimes referred to as the Bosonic Josephson junction) [11] and the initial coherent states are the SU(2) SCS [7],

$$\begin{aligned} |\theta,\varphi\rangle_s &= \exp(z\hat{L}_+ - z^*\hat{L}_-)|\ell,-\ell\rangle \\ &= \left[1+\xi^2\right]^{-\ell} \sum_{m=-\ell}^{\ell} \left(\xi e^{-i\varphi}\right)^{\ell+m} \begin{pmatrix} 2\ell \\ \ell+m \end{pmatrix}^{1/2} |\ell,m\rangle, \end{aligned} \tag{7}$$

where $\xi = \tan(\theta/2)$. The SU(2) generators $\hat{L}_x = (\hat{\psi}_1^\dagger\hat{\psi}_2 + \hat{\psi}_2^\dagger\hat{\psi}_1)/2$, $\hat{L}_y = (\hat{\psi}_1^\dagger\hat{\psi}_2 - \hat{\psi}_2^\dagger\hat{\psi}_1)/(2i)$, 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 $|\ell,m\rangle$ are the standard $\hat{\mathbf{L}}^2$, $\hat{L}_z$ eigenstates. Experimentally, such states are prepared either by coherently splitting an atomic BEC or by controlling optical or magnetic double-well potentials confining it [9, 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 $(L_\perp)^2 \equiv \langle\hat{L}_x\rangle^2 + \langle\hat{L}_y\rangle^2$ under a collisional $\delta\hat{L}_z + u\hat{L}_z^2$ 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 $L_\perp = (n/2)\sin\theta e^{-n(\sin\theta ut)^2/2}$ with phase-diffusion time $\tau_{pd} = (u\sin\theta\sqrt{n/2})^{-1}$ [8] (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-disffusion present during the hold time,

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

where $\Phi_h = \varphi_h + (2k+1)\arctan[\cosh\theta_p\tan(ut_h)]$. An exact form is also found for Δn_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 τ_{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. 1(d)). It is also evident from Eq. (6) and Eq. (9) that

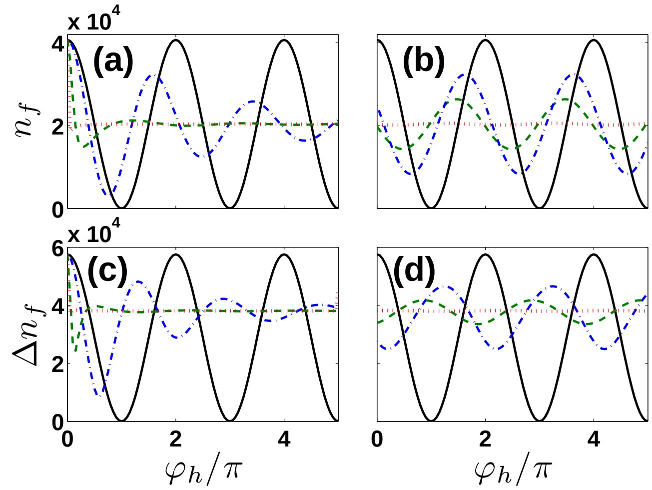

FIG. 3: (Color online) Final number of atoms n_f (a,b) and its variance Δn_f (c,d) as a function of $\varphi_h = \delta t_h$ in an SU(1,1) interferometer with $n = 2k(\cosh\theta_p - 1) = 100$. Time domain fringes (fixed δ and varying t_h) are shown in (a,c) with $un/\delta = 0$ (solid), 0.1 (dash-dotted), 1 (dashed), and 10 (dotted). Frequency domain fringes (fixed t_h and varying δ) are plotted in (b,d) with $unt_h = 0$ (solid), 0.5 (dash-dotted), 1 (dashed), and 10 (dotted).

coherence revives on a very long $\tau_r = \pi/u$ timescale, similarly to the SU(2) case [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-Zendher atom interferometry [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 [4, 5].

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