## Atom-molecule dephasing in an SU(1,1) interferometer based on the stimulated dissociation of a molecular Bose-Einstein condensate

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We propose to implement a sub-shot-noise matter-wave interferometer via the stimulated dissociation of a molecular Bose-Einstein condensate and study the collisional loss of atom-molecule coherence during its phase-acquisition time. The obtained *n*-atom 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.

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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 "superchemistry" [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 matterwave 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  $\Delta n = (\langle \hat{n}^2 \rangle - \langle \hat{n} \rangle^2)^{1/2}$ , where  $\hat{n}$  is the atomic number operator.

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. Also like optical parametric amplification, stimulated dissociation is *phase* sensitive for atomic states different from the vacuum state. Given a nonvanishing value of *n* the 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 construct a sub-shot-noise SU(1,1) interferometer [6]. 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) or "twoatom" coherent state (TACS). The phase squeezing inherent to the TACS allows the SU(1,1) interferometer to reach Heisenberg-limited accuracy without the preparation of a number-squeezed input, required to go beyond the shot-noise limit in Mach-Zendher atom interferometry [7]. Since experiments in optical-lattice and double-well potentials currently attain relatively moderate squeezing factors [8-10], SU(1,1) interferometry with phase squeezing embedded in its beam splitter offers a significant advantage.

Unlike its optical counterpart, matter-wave interferometry is limited by particle interactions. Our main result in this respect is that similar to the spin-squeezed state in the phaseacquisition stage of a Mach-Zendher atom interferometer, the  $\Delta n \propto n$  atom-number variance of the TACS results in the loss of atom-molecule phase coherence on a short  $\tau_{pd} \propto 1/n$  time scale due to collisional phase diffusion. By contrast, two initially coherent separated atomic condensates phase diffuse on a longer  $\tau_{pd} \propto 1/\sqrt{n}$  time scale [12] since their initial state is an SU(2) or "spin" coherent state (SCS) with  $\Delta n \propto \sqrt{n}$ . Moreover, we find that for  $n \ge 1$  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,

$$H = E_m \hat{n}_m + E_a \hat{n} + (g_{am} \hat{\psi}_m^{\dagger} \hat{\psi}_a \hat{\psi}_a + \text{H.c.}) + \frac{u_m}{2} \hat{\psi}_m^{\dagger} \hat{\psi}_m^{\dagger} \hat{\psi}_m \hat{\psi}_m$$
$$+ \frac{u_a}{2} \hat{\psi}_a^{\dagger} \hat{\psi}_a^{\dagger} \hat{\psi}_a \hat{\psi}_a + u_{am} \hat{n}_m \hat{n}, \qquad (1)$$

where  $\hat{\psi}_{a,m}$  are 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  $g_{am} = |g_{am}| e^{i\phi}$ . The interaction strengths are  $u_{\alpha} = 2\pi\hbar^2 a_{\alpha}/(\mu_{\alpha}V)$ ,  $\alpha = a, m, am$ , where  $a_{\alpha}, \mu_{\alpha}$ are the *s*-wave scattering length and reduced mass for atomatom, molecule-molecule, and atom-molecule scatterings (i.e.,  $\mu_a = m/2$ ,  $\mu_m = m$ , and  $\mu_{am} = 2m/3$ , where *m* is the atomic mass) and *V* denotes the quantized volume.

Using total pair number conservation  $\hat{n}_m + \hat{n}/2 = N$  to eliminate *c*-number terms and assuming in what follows that the molecular condensate remains large and is never significantly depleted by the conversion of a small number of molecules into atoms  $n_m \approx N/2$ , 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



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  $\varphi$ . 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  $\varphi$  acquired during the hold time. When  $\varphi$  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  $\varphi$  phase diffusion.

$$H = \delta \hat{K}_z + g \hat{K}_x + u \hat{K}_z^2. \tag{2}$$

Here  $\delta = 2(E_a + u_{am}n_m - u_a) - (E_m + u_m n_m)$ ,  $g = 4|g_{am}|\sqrt{n_m}$ , and  $u = 2u_a + u_m/2 - 2u_{am}$ . 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  $\hat{K}_z$ ,

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

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

The SU(1,1) interferometer [6] for probing the atommolecule 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  $g \ge \delta$ , un. As shown below, this condition is easily attained in current experimen-

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tal setups by magnetic control of the atom-molecule detuning for Feshbach-resonance coupling and by switching the photodissociation lasers for the optical resonant Raman coupling. The atomic state following this Lorentzian boost of duration  $t_p$  is an SU(1,1) TACS [6,11],

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

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  $\Delta n=\sqrt{2k} \sinh \theta$  [11], 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 neglected (u=0), coherence is maintained may be state at the end of the hold time is and the  $\exp(-i\delta \tilde{K}_{z}t_{h})|\theta_{p},\pi/2\rangle = |\theta_{p},\pi/2+\varphi_{h}\rangle$ with  $\varphi_h \equiv \delta t_h$ [Fig. 1(c)]. The accumulated atom-molecule phase  $\varphi_h$  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 boostrotation-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, the final number of atoms  $n_f$  and its variance  $\Delta n_f$  are given by

$$n_{f} = 2k(\cosh \theta_{f} - 1) = \frac{1 + \cos \varphi_{h}}{2} \sinh^{2} \theta_{p},$$
$$(\Delta n_{f})^{2} = 2k \sinh^{2} \theta_{f} = \frac{\sinh^{2} \theta_{p}}{2} [\sin^{2} \varphi_{h} + (1 + \cos \varphi_{h})^{2} \cosh^{2} \theta_{p}].$$
(5)

Note that these expressions are slightly different from those 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  $(\Delta \varphi_h)^2 = [(\Delta n_f)^2 / |\partial n_f / \partial \varphi_h|^2]_{\varphi_h = \pi} = (2 \sinh^2 \theta_p)^{-1} = [8n(n+1)]^{-1}$  accuracy. Thus, unlike sub-shot-noise atom interferometry which requires the preparation of an initial number-squeezed state, the squeezing inherent in coherent dissociation  $\Delta \varphi_h$  around  $\varphi_h = \pi$  goes below the  $1/\sqrt{n}$  standard quantum limit (also known as 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



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,  $\bigcirc$ ), 167 (dashed,  $\Box$ ), and 500 (dashed-dotted,  $\triangle$ ), symbols mark numerical results with n+ $2n_m$ =5000; (d) same for SCS with n=70 (solid,  $\bigcirc$ ), 156 (dashed,  $\Box$ ), and 626 (dashed-dotted,  $\triangle$ ), 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.

scenario. Atom-atom and molecule-atom collisions will degrade atom-molecule coherence during the phase-acquisition time since for nonvanishing *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)].

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  $\varphi$ ,  $\delta$  and decays as

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

Noting that  $\sinh^2 \theta = (n/2k)[(n/2k)+2] = 4n(n+1)$  we obtain that for a moderately large  $n \ge 1$ , coherence decays on a  $\sin(ut) \sim 1/(2n)$  time scale. 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

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dephasing with phase diffusion between two initially coherent atomic BECs [9,12,13]. The pertinent Hamiltonian is the two-site Bose-Hubbard model (sometimes referred to as the bosonic Josephson junction) [14] and the initial coherent states are the SU(2) SCS [11],

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

where  $\xi = \tan(\theta/2).$ The SU(2)generators  $\hat{L}_v \!=\! (\hat{\psi}_1^\dagger \hat{\psi}_2 \!-\! \hat{\psi}_2^\dagger \hat{\psi}_1)/(2i),$  $\hat{L}_{r} = (\hat{\psi}_{1}^{\dagger}\hat{\psi}_{2} + \hat{\psi}_{2}^{\dagger}\hat{\psi}_{1})/2,$ 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 [9,13]. Most common are states with equal population of the two condensates, i.e.,  $\theta = \pi/2$ .

The binomial or 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

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

approaching for  $n \ge 1$  the Gaussian decay  $L_{\perp} = (n/2)\sin \theta e^{-n(\sin \theta u t)^{2}/2}$  with phase-diffusion time  $\tau_{pd} = (u \sin \theta \sqrt{n/2})^{-1}$  [12] [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. [9].

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,

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

where  $\Phi_h = \varphi_h + (2k+1) \arctan[\cosh \theta_p \tan(ut_h)]$ . 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  $\tau_{pd}$  time scale, 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 Eqs. (6) and (9) that coherence revives on a very long  $\tau_r = \pi/u$  time scale, similarly to the SU(2) case [12,13].

Before closing, we evaluate the anticipated phasediffusion time under current experimental conditions, comparing it to the phase diffusion of the Poissonian molecular field and the characteristic atom-molecule frequency. Molecule-molecule and atom-molecule *s*-wave scattering



FIG. 3. (Color online) Final number of atoms  $n_f$  [(a) and (b)] and its variance  $\Delta n_f$  [(c) and (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  $\delta$  and varying  $t_h$ ) are shown in (a) and (c) with  $un/\delta=0$  (solid), 0.1 (dashed-dotted), 1 (dashed), and 10 (dotted). Frequency domain fringes (fixed  $t_h$  and varying  $\delta$ ) are plotted in (b) and (d) with  $unt_h=0$  (solid), 0.5 (dashed-dotted), 1 (dashed), and 10 (dotted).

lengths are approximated by the zero-range single-channel scattering approximation [15] as  $a_m = 0.6a_a$ ,  $a_{am} = 1.2a_a$ , giving  $u = 0.35u_a$ . For 2000 dissociated atoms in a 100 Hz spherical trap  $(n/V \sim 10^{13} \text{ cm}^{-3})$ , the obtained phase-diffusion times range from 0.1 ms for <sup>85</sup>Rb to 1.3 ms for <sup>23</sup>Na. Assuming a molecular BEC of  $5 \times 10^4$  mol-

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ecules (2% dissociation) in the same trap, the dephasing of the molecular field takes place on a much longer  $1/(u_m\sqrt{n_m}) \sim 10-100$  ms time scale. Thus, the rapid phase diffusion of the atomic seed will be the limiting factor in atom-molecule interferometry. By contrast, the atommolecule Feshbach conversion frequency  $|g_{am}\sqrt{n_m/V}|$  with  $n_m/V \sim 10^{15}$  cm<sup>-3</sup> is of order 1 MHz [16], justifying the neglect of phase diffusion during the coupling pulses.

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 [6,7]. 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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