

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 ultrasensitive 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 $\Delta n = (\langle \hat{n}^2 \rangle - \langle \hat{n} \rangle^2)^{1/2}$, 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 from the vacuum state. Given a nonvanishing 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 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 “two-atom” 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 phase-acquisition 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 $n \gg 1$ 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,

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

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_α , μ_α are the s -wave scattering length and reduced mass for atom-atom, 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 $\hat{n}_m + \hat{n} = N$ 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

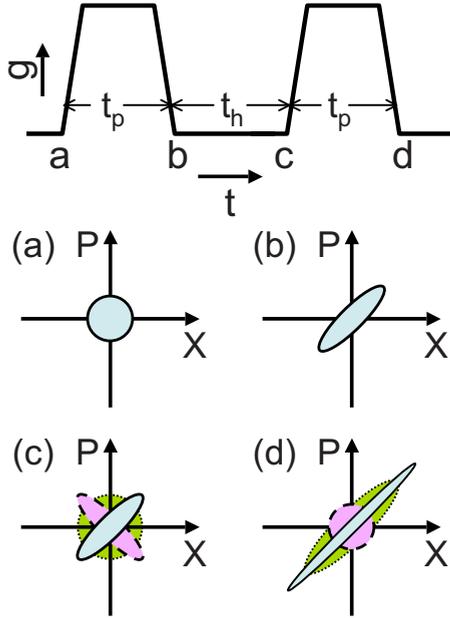


FIG. 1. (Color online) Atom-molecule SU(1,1) interferometer. The quadrature phase-amplitude distribution is shown at the time points marked on the upper $g(t)$ plot. Note that the polar angle in the X, P plot is $\varphi/2$, not φ . Starting from the atomic vacuum (a) the first Lorentzian boost results 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.

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

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 ,

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

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-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 $g \gg \delta, un$. As shown below, this condition is easily attained in current experimen-

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],

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

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 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, the final number of atoms n_f and its variance Δn_f are given by

$$n_f = 2k(\cosh\theta_f - 1) = \frac{1 + \cos\varphi_h}{2} \sinh^2\theta_p, \\ (\Delta n_f)^2 = 2k \sinh^2\theta_f = \frac{\sinh^2\theta_p}{2} [\sin^2\varphi_h + (1 + \cos\varphi_h)^2 \cosh^2\theta_p]. \quad (5)$$

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 $(\Delta\varphi_f)^2 = [(\Delta n_f)^2 / |\partial n_f / \partial \varphi_h|^2]_{\varphi_h = \pi} = (2 \sinh^2\theta_p)^{-1} = [8n(n+1)]^{-1}$ 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

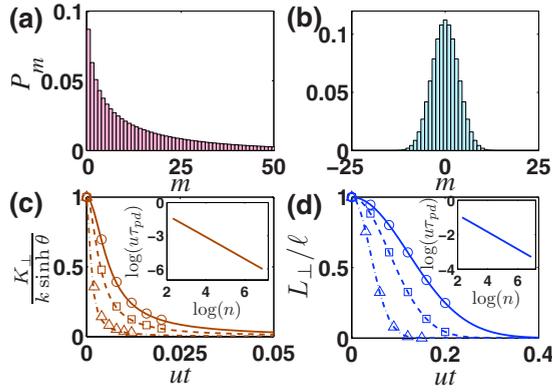


FIG. 2. (Color online) Comparison of atom-molecule phase diffusion with the collisional dephasing of separated atomic condensates: (a) number distribution of a TACS $|\theta, \varphi\rangle$ with $\theta=4.8$, corresponding to $n=30$ dissociated atoms; (b) same for a SCS $|\pi/2, \varphi\rangle_s$ with $\ell=n/2=25$; (c) phase diffusion of TACS with $n=100$ (solid, \circ), 167 (dashed, \square), and 500 (dashed-dotted, \triangle), symbols mark numerical results with $n+2n_m=5000$; (d) same for SCS with $n=70$ (solid, \circ), 156 (dashed, \square), 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 φ_h independent [dotted ellipse in Fig. 1(d)].

Atom-molecule coherence may be quantified by defining the SU(1,1) purity $K^2 \equiv \langle \hat{K}_z^2 \rangle - \langle \hat{K}_x \rangle^2 - \langle \hat{K}_y \rangle^2$. 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

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

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 $\sin(ut) \sim 1/(2n)$ 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

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],

$$|\theta, \varphi\rangle_s = \exp(z\hat{L}_+ - z^*\hat{L}_-)|\ell, -\ell\rangle = [1 + \xi^2]^{-\ell} \sum_{m=-\ell}^{\ell} (\xi e^{-i\varphi})^{\ell+m} \binom{2\ell}{\ell+m}^{1/2} |\ell, m\rangle, \quad (7)$$

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{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

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

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 $\tau_{pd} = (u \sin \theta \sqrt{n/2})^{-1}$ [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,

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

where $\Phi_h = \varphi_h + (2k+1) \arctan[\cosh \theta_p \tan(ut_h)]$. An exact form is also found for 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} 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 phase-diffusion 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

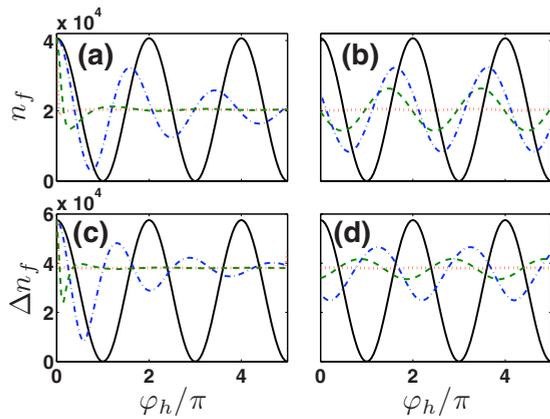


FIG. 3. (Color online) Final number of atoms n_f [(a) and (b)] and its variance Δn_f [(c) and (d)] as a function of $\varphi_h = \delta t_h$ in an SU(1,1) interferometer with $n = 2k(\cosh \theta_p - 1) = 100$. Time domain fringes (fixed δ and varying t_h) are shown in (a) and (c) with $unt_h = 0$ (solid), 0.1 (dashed-dotted), 1 (dashed), and 10 (dotted). Frequency domain fringes (fixed t_h and varying δ) 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 ^{85}Rb to 1.3 ms for ^{23}Na . Assuming a molecular BEC of 5×10^4 mol-

ecules (2% dissociation) in the same trap, the dephasing of the molecular field takes place on a much longer $1/(u_m \sqrt{n_m}) \sim 10\text{--}100$ ms time scale. Thus, the rapid phase diffusion of the atomic seed will be the limiting factor in atom-molecule interferometry. By contrast, the atom-molecule Feshbach conversion frequency $|g_{am} \sqrt{n_m/V}|$ with $n_m/V \sim 10^{15} \text{ cm}^{-3}$ 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-Zehnder 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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