

Observing a coherent superposition of an atom and a molecule

Mark R. Dowling,^{1,*} Stephen D. Bartlett,² Terry Rudolph,^{3,4} and Robert W. Spekkens⁵

¹*School of Physical Sciences, The University of Queensland, St. Lucia, Queensland 4072, Australia*

²*School of Physics, The University of Sydney, Sydney, New South Wales 2006, Australia*

³*Optics Section, Blackett Laboratory, Imperial College London, London SW7 2BW, United Kingdom*

⁴*Institute for Mathematical Sciences, Imperial College London, London SW7 2BW, United Kingdom*

⁵*Department of Applied Mathematics and Theoretical Physics, University of Cambridge, Cambridge CB3 0WA, United Kingdom*

(Received 1 July 2006; published 29 November 2006)

We demonstrate that it is possible, in principle, to perform a Ramsey-type interference experiment to exhibit a coherent superposition of a single atom and a diatomic molecule. This gedanken experiment, based on the techniques of Aharonov and Susskind [Phys. Rev. **155**, 1428 (1967)], explicitly violates the commonly accepted superselection rule that forbids coherent superpositions of eigenstates of differing atom number. A Bose-Einstein condensate plays the role of a reference frame that allows for coherent operations analogous to Ramsey pulses. We also investigate an analogous gedanken experiment to exhibit a coherent superposition of a single boson and a fermion, violating the commonly accepted superselection rule forbidding coherent superpositions of states of differing particle statistics. In this case, the reference frame is realized by a multimode state of many fermions. This latter case reproduces all of the relevant features of Ramsey interferometry, including Ramsey fringes over many repetitions of the experiment. However, the apparent inability of this proposed experiment to produce well-defined relative phases between two distinct systems each described by a coherent superposition of a boson and a fermion demonstrates that there are additional, outstanding requirements to fully “lift” the univalence superselection rule.

DOI: [10.1103/PhysRevA.74.052113](https://doi.org/10.1103/PhysRevA.74.052113)

PACS number(s): 03.65.Ta, 03.75.Dg, 03.75.Gg

I. INTRODUCTION

Part of the dogma of orthodox quantum mechanics is the presumed existence of *superselection rules* [1,2] for certain quantities. For instance, it is often stated that one cannot create or observe quantum coherence between eigenstates of differing charge, or of differing mass, or of number eigenstates of particles with differing statistics (e.g., a superposition of a boson and a fermion).¹ Originally, superselection rules were introduced to enforce additional constraints to quantum theory beyond the well-studied constraints of selection rules (conservation laws). In a classic paper, Aharonov and Susskind [4] challenged the necessity of such superselection rules, and outlined a gedanken experiment for exhibiting a coherent superposition of charge eigenstates as an example of how superselection rules can be obviated in practice.

The gedanken experiment of Aharonov and Susskind, and subsequent investigations [5–9], highlighted the requirement of an appropriate *reference frame* in order to exhibit coherence between eigenstates of superselected quantities. For example, while a reference frame for spatial orientation is required to exhibit coherence between states of differing angular momentum (in some direction), a more exotic form

of reference frame is required to exhibit coherence between states of differing charge. Arguably, it is the ubiquity of reference frames for some quantities (such as spatial orientation or phase) and not for others (such as the type of frame needed to exhibit superpositions of charge eigenstates) that has led to the proposed superselection rules for some quantities and not others.

While this gedanken experiment served to illustrate a concept, recent advances in the preparation and manipulation of exotic quantum states of matter may offer the opportunity to demonstrate these concepts in experiment. In this paper, we discuss the principles of an experiment that may be performed with ultracold atoms and molecules where the superselection rule in question is for atom number, or equivalently, baryon number. (The existence of such a superselection rule is commonly assumed; cf. [1,10–15].) The experiment we present aims to exhibit quantum coherence between states corresponding to a single atom and a diatomic molecule; the reference frame in this case is a Bose-Einstein condensate (BEC).

We advance this concept further by outlining a gedanken experiment to exhibit coherence between a boson and a fermion. We demonstrate that, using a reference frame consisting of many fermions in many modes, the essential features in this experiment are similar to those of the fully bosonic one, and that the commonly accepted superselection rule disallowing superpositions of a boson and a fermion (the univalence superselection rule [16]) can be violated in principle.

Aharonov and Susskind used an operational approach to identify coherence between eigenstates of some superselected quantity: the observation of Ramsey-type fringes in an interference experiment. The reference frame allows for the implementation of Ramsey pulses to create and subsequently measure the superposition states. We follow this approach

*Electronic address: dowling@physics.uq.edu.au

¹In Lorentz-invariant quantum field theories, it has been argued that some superselection rules can be derived within the theory [3]. However, such arguments do not apply to nonrelativistic quantum theory, and in particular do not apply when classical reference frames (i.e., measurement apparatuses) are employed within the theory.

here, and extend their results by identifying several salient features of the reference frame that allow for high-visibility Ramsey fringes for many repetitions of the experiment. We demonstrate that such a reference frame can be treated as either a classical or a quantum system, with both descriptions leading to equivalent predictions. In addition, we identify where in the mathematical formalism the coherent superpositions arise: if the reference frame is treated as a quantum system, this coherence arises in the relational degrees of freedom.

Finally, our gedanken experiment to exhibit a coherent superposition of a boson and a fermion suggests that there must be an *additional* requirement beyond the ability to repeatedly observe high-visibility interference fringes for one to say that a superselection rule has been “lifted.” We show that this gedanken experiment, despite possessing all of the salient features of a Ramsey interferometry experiment, cannot induce a well-defined *relative* phase between two distinct systems each described by a coherent superposition of a boson and a fermion. We therefore demonstrate that the ability to repeatedly violate a superselection rule is not equivalent to lifting it.

II. EXHIBITING QUANTUM COHERENCE WITH INTERFEROMETRY

Consider a two-level atom, defined by two energy eigenstates $|g\rangle$ and $|e\rangle$. How would one demonstrate coherence between these two states? That is, how does one discriminate the coherent superposition $\frac{1}{\sqrt{2}}(|g\rangle + |e\rangle)$ from the incoherent mixture $\frac{1}{2}(|g\rangle\langle g| + |e\rangle\langle e|)$? A direct method would be to measure many identically prepared atoms in the basis $|\pm\rangle = \frac{1}{\sqrt{2}}(|g\rangle \pm |e\rangle)$ and observe the statistics. However, in practice, preparations and measurements are restricted to the basis $\{|g\rangle, |e\rangle\}$. The standard method, then, is to perform an interference experiment, in the form of a Ramsey interferometer, which we now outline.

Between the preparation and the measurement, the interference experiment makes use of two types of evolutions. The first evolution is described by a Hamiltonian of the form

$$\hat{H}_{\text{Ram}} = \frac{\hbar\Omega}{2}(|g\rangle\langle e| + |e\rangle\langle g|), \quad (1)$$

where Ω is a real number. This evolution is implemented by a laser tuned to the energy difference of these two levels, and this description is adequate if the laser pulse can be treated as an external potential (i.e., with a well-defined amplitude and phase). Application of this Hamiltonian for some finite time is called a *Ramsey pulse*, and if this Hamiltonian is applied for a time $t = \pi/(2\Omega)$, the resulting unitary operation is called a $\pi/2$ pulse. The second type of evolution is governed by the free Hamiltonian

$$\hat{H}_{\text{free}} = \Delta|e\rangle\langle e|, \quad (2)$$

with Δ the detuning between the energy splitting between $|e\rangle$ and $|g\rangle$ and the laser frequency when the laser is tuned off resonance.

The interference experiment proceeds as follows. The system is prepared in the initial state $|g\rangle$, and then subjected to a $\pi/2$ pulse. The relative phase shift ϕ is then applied using the second type of evolution for a time $\tau = \phi/\Delta$, followed by another $\pi/2$ pulse. At the time of the preparation, and subsequent to each of the three interactions, the state of the atom is, respectively,

$$|\Psi_0\rangle = |g\rangle, \quad (3)$$

$$|\Psi_1\rangle = \frac{1}{\sqrt{2}}(|g\rangle - i|e\rangle), \quad (4)$$

$$|\Psi_2\rangle = \frac{1}{\sqrt{2}}(|g\rangle - ie^{-i\phi}|e\rangle), \quad (5)$$

$$|\Psi_3\rangle = \sin(\phi/2)|g\rangle - \cos(\phi/2)|e\rangle. \quad (6)$$

Finally, the atom is measured in the $\{|g\rangle, |e\rangle\}$ basis. By controlling the magnitude of the phase shift ϕ between the pulses over many runs of the experiment, one observes so-called *Ramsey fringes*—oscillations in the probability of measuring the outcome corresponding to $|g\rangle$ (or $|e\rangle$) as a function of ϕ . Such Ramsey fringes are the signature of coherent operation, i.e., that the description of the system for the time period between the two $\pi/2$ pulses is given by a coherent superposition of $|g\rangle$ and $|e\rangle$. If instead the system was described at this intermediate time by an incoherent superposition, then the resulting statistics would be independent of ϕ , i.e., no fringes would be observed. See the experimental paper of Bertet *et al.* [17] for a presentation of a Ramsey interferometry experiment that takes a similar perspective to the one adopted in this paper.

The basic structure of this Ramsey interference experiment has analogies in many other quantum systems. An experiment using a single photon and a Mach-Zender interferometer is formally equivalent, with beam splitters acting in the role of the Ramsey pulses; see Fig. 1. Other analogous experiments are commonplace in atomic, molecular, and nuclear systems, and have more recently been demonstrated in artificial structures such as semiconductor quantum dots [18] and superconducting qubits [19]. Because the same abstract structure can be realized in a wide variety of physical systems, the identification between these different realizations of the same basic interference experiment has been called the “quantum Rosetta stone” [20]. We seek to add another realization to this list.

III. COHERENT SUPERPOSITIONS OF AN ATOM AND A MOLECULE

We now consider an analog of the Ramsey experiment that aims to exhibit coherence between a single atom and a molecule. Consider a bosonic atom; we denote the species of this atom as type 1. Also, consider a diatomic bosonic molecule, denoted M , which consists of one atom of type 1 and one other bosonic atom of another type, 2. We now define a two-level system, which will serve as the analog in our ex-

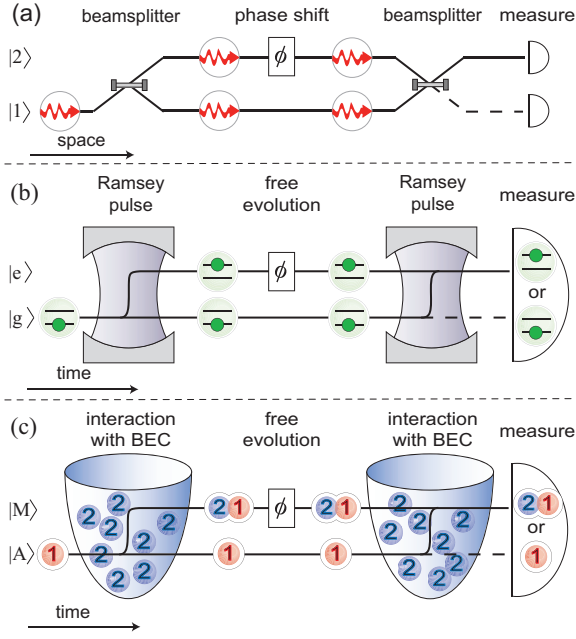


FIG. 1. (Color online) Schematic of the demonstration of quantum coherence in different physical systems: (a) Mach-Zender interferometer, (b) Ramsey interferometry, and (c) the ultracold atom-molecule system considered in this paper.

periment of the two-level atom in the Ramsey experiment. This two-level system is spanned by the following basis states:

$$|A\rangle = |0\rangle_M |1\rangle_A, \quad |M\rangle = |1\rangle_M |0\rangle_A, \quad (7)$$

where $|i\rangle_A$ is a Fock state for an atomic mode of type 1 and $|i\rangle_M$ is a Fock state for a diatomic molecule mode. One may consider the single-atom state $|A\rangle$ as the analog of the ground state of the two-level atom in the Ramsey experiment, and the molecule state $|M\rangle$ as the analog of the excited state of this two-level atom. The aim is to demonstrate coherence in this two-level system, i.e., coherence between $|A\rangle$ and $|M\rangle$. Such a demonstration of coherence violates a superselection rule for atom number.²

We will use a Bose-Einstein condensate of atoms of type 2 to serve as our reference frame, i.e., as the analog of the electromagnetic field that constitutes the Ramsey pulse. (The use of a BEC as a phase reference has been discussed in [24].) The analog of the Ramsey pulse in our experiment is given by an interaction of the two-level system with the reference frame (the BEC). The relative phase shift will be implemented by free evolution of the system. Using these two basic evolutions we will describe an experiment that is formally equivalent to Ramsey interferometry.

²We note that such an experiment is distinct from ones that aim to exhibit coherence between two atoms and a diatomic molecule, or between a BEC of N atoms and a BEC of $N/2$ diatomic molecules, as considered in [21–23].

A. Using a quantum reference frame

In the Ramsey interference experiment presented in Sec. II, the laser pulse which served as a reference frame was treated as an external potential. However, another description of the same experiment could be presented wherein the laser pulse was treated as a dynamical quantum system. In general, different descriptions of the same interference experiment are possible depending on whether the reference frame is treated as a nondynamical macroscopic apparatus or as a dynamical quantum system. In the language of [25] to treat the reference frame as part of the apparatus is to use it as an *external* reference frame, while to treat it as a dynamical quantum object is to use it as an *internal* reference frame.

We begin by treating the reference frame—in this case, the BEC—as a fully dynamical quantum system. At the end of Sec. III B, we will demonstrate that this experiment is equivalent to the Ramsey interference experiment described in the previous section when the reference frame (the BEC) is treated externally.

1. Quantum state of the BEC

For simplicity, we adopt a single-mode description of the BEC. The single mode corresponds to the Gross-Pitaevskii ground state, which has had great success in describing BEC dynamics [13,26]. Following the terminology of [4] we refer to the three modes—the mode of atom type 1, the molecular mode, and the BEC mode—together as the *laboratory*.³ States of the laboratory are most generally defined on the Fock space $\mathcal{F}_M \otimes \mathcal{F}_A \otimes \mathcal{F}_2$ spanned by the basis of Fock states $|n_M\rangle_M |n_A\rangle_A |n_2\rangle_2$, where n_M , n_A , and n_2 are occupation numbers for the modes. In our experiment, $\hat{n}_M + \hat{n}_A$ will be a constant of the motion. Initially, it has eigenvalue 1, as we consider an initial state consisting of precisely one atom of type 1 and zero molecules. Thus, we can restrict our attention to the two-dimensional subspace of $\mathcal{F}_M \otimes \mathcal{F}_A$ spanned by the two states $|A\rangle$ and $|M\rangle$ of Eq. (7). We refer to this two-level system as the *system*, and the Hilbert space associated with it (spanned by $|A\rangle$ and $|M\rangle$) is denoted \mathcal{H}_S . The *reference frame* is the remaining mode describing the BEC of atomic species 2, with infinite-dimensional Hilbert space $\mathcal{H}_R = \mathcal{F}_2$. States of the laboratory (system+reference frame) are thus defined on $\mathcal{H}_S \otimes \mathcal{H}_R$. We will use the modified Dirac notation $|\cdot\rangle$ for the state of the reference frame, to emphasize the special role it plays.

The BEC consists of atoms of type 2 in a single mode (the Gross-Pitaevskii ground state mentioned above). In simplistic treatments, it is common to treat the condensate as a coherent state, i.e., to assign a state $|\beta\rangle$, defined in the number basis as $|\beta\rangle = \sum_{n=0}^{\infty} c_n |n\rangle$ with $c_n = \exp(-|\beta|^2/2) \beta^n / \sqrt{n!}$, as the quantum state of the BEC. However, as argued in [25], the coherence properties of a state assigned to a bosonic mode are defined relative to a classical reference frame for a

³We will describe our proposed experiment in terms of the dynamics of these three modes. There are, of course, many details that are not captured by this simple model, for example the many vibrational modes of the molecule, which would need to be taken into consideration in an experiment.

phase that is held in the background. If the phase of the BEC is uncorrelated with any classical phase reference in the background, as we will assume in our experiment, then it is instead appropriate to assign it a quantum state that is incoherently averaged over all possible orientations with respect to the background phase reference. Such a state is given by

$$\rho_0^{\text{rf}} = \int_0^{2\pi} \frac{d\theta}{2\pi} e^{-i\hat{n}_2\theta} |\beta\rangle\langle\beta| e^{i\hat{n}_2\theta}. \quad (8)$$

It is straightforward to show that this state is equal to a Poissonian mixture of number states

$$\rho_0^{\text{rf}} = \sum_{n=0}^{\infty} p_n(\bar{n}) |n\rangle\langle n|, \quad (9)$$

where $p_n(\bar{n}) = e^{-\bar{n}} \bar{n}^n / n!$ is a Poisson distribution with $\bar{n} = |\beta|^2$. It is this state, ρ_0^{rf} , that we choose to describe the BEC.⁴ The treatment of the BEC as an incoherent state is consistent with the arguments of Refs. [10,11,28].

Equation (8) is particularly useful as a pedagogical tool for understanding interference experiments such as the one described here: the state ρ_0^{rf} can be intuitively viewed as a coherent state $|\beta\rangle$, the phase of which ($\arg \beta$) is uncorrelated with any classical phase reference used to describe the experiment.

If the BEC mode is initially described by this mixed state ρ_0^{rf} , and the system starts in the state $|A\rangle$, then the initial state of the laboratory may be expressed as

$$W_{0L} = |A\rangle\langle A| \otimes \rho_0^{\text{rf}} = \int_0^{2\pi} \frac{d\theta}{2\pi} e^{-i\hat{N}_{\text{tot}}\theta} |\Psi_0\rangle_L \langle\Psi_0| e^{i\hat{N}_{\text{tot}}\theta}, \quad (10)$$

where

$$|\Psi_0\rangle_L \equiv |A\rangle|\beta\rangle, \quad (11)$$

and where the total atom number operator \hat{N}_{tot} is defined as

$$\hat{N}_{\text{tot}} \equiv 2\hat{n}_M + \hat{n}_A + \hat{n}_2. \quad (12)$$

We define the *twirling operator* acting on density operators on the laboratory as

$$\mathcal{T}[\rho_L] = \int_0^{2\pi} \frac{d\theta}{2\pi} e^{-i\hat{N}_{\text{tot}}\theta} \rho_L e^{i\hat{N}_{\text{tot}}\theta}. \quad (13)$$

Then $W_{0L} = \mathcal{T}[|\Psi_0\rangle_L \langle\Psi_0|]$. It will be illustrative to follow the evolution of the $\theta=0$ element $|\Psi_0\rangle_L$ of the ensemble in Eq. (10) through the experiment, bearing in mind that the global phase θ is not physically significant, and the quantum state of

⁴The intuition that motivates using a coherent state for the BEC is the belief that coherence is required to explain interference experiments involving BECs such as [27]. However, coherent states are not required for such an explanation; it has been demonstrated that number states, or incoherent mixtures of number states such as Eq. (9), can also interfere [28]. See [29–31] for further discussion of this fact.

the laboratory is obtained by averaging over this phase (implemented by the twirling operation \mathcal{T}).

2. Interactions with the quantum reference frame

We now outline the Hamiltonians that will be used to induce the required evolutions in our gedanken experiment. The first is an interaction between the system and the quantum reference frame. At a Feshbach resonance, which occurs when an external magnetic field is tuned so that the energy of two free atoms is equal to that of a bound molecular state, coherent tunneling occurs between pairs of atoms and molecules. A simple model Hamiltonian for this phenomenon, where the two atoms are of different species, is (cf. [32])

$$\hat{H}_{\text{int}} = \frac{\hbar\kappa}{2} (\hat{b}_M^\dagger \hat{b}_A \hat{b}_2 + \hat{b}_M \hat{b}_A^\dagger \hat{b}_2^\dagger), \quad (14)$$

where \hat{b}_M is the annihilation operator for the bound molecular state, and \hat{b}_A and \hat{b}_2 are annihilation operators for the modes containing the two distinct atomic species. This Hamiltonian can be reexpressed in terms of an operator on the Hilbert space $\mathcal{H}_S \otimes \mathcal{H}_R$ of the laboratory as

$$\hat{H}'_{\text{int}} = \frac{\hbar\kappa}{2} (|M\rangle\langle A| \otimes \hat{b}_2 + |A\rangle\langle M| \otimes \hat{b}_2^\dagger). \quad (15)$$

We will also make use of a Hamiltonian that induces a relative phase shift on the system. Such a Hamiltonian is provided by moving off of the Feshbach resonance and allowing free evolution under the Hamiltonian

$$\hat{H}_{\text{free}} = \hbar\omega_M \hat{n}_M + \hbar\omega_A \hat{n}_A + \hbar\omega_2 \hat{n}_2, \quad (16)$$

where $\hbar\omega_i$, $i=M,A,2$, are the internal energies of the two atomic species and the molecule away from the Feshbach resonance. There will be some internal energy difference between the bound molecular state and the sum of the two free atomic states when off resonance, given by

$$\Delta_{\text{int}} \equiv \omega_M - \omega_A - \omega_2. \quad (17)$$

We note that both \hat{N}_{tot} and $\hat{n}_M + \hat{n}_A$ are constants of the motion, and thus we can move to an interaction picture in which the free-evolution Hamiltonian on \mathcal{H}_S is simply

$$\hat{H}'_{\text{free}} = \hbar\Delta_{\text{int}} |M\rangle\langle M|, \quad (18)$$

up to addition of a constant.

3. The interference experiment

We initiate the experiment with the laboratory in the state W_{0L} of Eq. (10). The system and reference frame first interact for a time $t = \pi / (2\kappa\sqrt{\bar{n}})$ at the Feshbach resonance according to (15). This interaction plays the role of a Ramsey pulse. As a result of this interaction, the state of the laboratory evolves (in the Schrödinger picture) to $W_{1L} = \mathcal{T}[|\Psi_1\rangle_L \langle\Psi_1|]$, with

$$|\Psi_1\rangle_L = |A\rangle|\beta_A^1\rangle + |M\rangle|\beta_M^1\rangle, \quad (19)$$

where we have defined unnormalized states

$$|\beta_A^1\rangle \equiv \sum_{n=0}^{\infty} c_n \cos\left(\sqrt{\frac{n}{\bar{n}}}\frac{\pi}{4}\right)|n\rangle, \quad (20)$$

$$|\beta_M^1\rangle \equiv -i \sum_{n=0}^{\infty} c_n \sin\left(\sqrt{\frac{n}{\bar{n}}}\frac{\pi}{4}\right)|n-1\rangle. \quad (21)$$

We can interpret these states as (unnormalized) coherent states that have undergone a disturbance due to the interaction with the system.

Next, we allow the laboratory to freely evolve for time τ away from the Feshbach resonance according to (18). This free evolution induces a relative phase between the atom and molecule terms. The result is a state $W_{2L} = \mathcal{T}[|\Psi_2\rangle_L \langle \Psi_2|]$, with

$$|\Psi_2\rangle_L = |A\rangle|\beta_A^1\rangle + e^{-i\phi}|M\rangle|\beta_M^1\rangle, \quad (22)$$

and where we have defined

$$\phi \equiv \Delta_{\text{int}}\tau \quad (23)$$

as the net relative phase shift between the atom and molecule terms acquired during this stage.

Finally we implement the analog of the second Ramsey pulse by allowing the system and reference frame to interact again at the Feshbach resonance for time $t = \pi/(2\kappa\sqrt{\bar{n}})$, giving the final state $W_{3L} = \mathcal{T}[|\Psi_3\rangle_L \langle \Psi_3|]$, with

$$|\Psi_3\rangle_L = |A\rangle|\beta_A^3\rangle + |M\rangle|\beta_M^3\rangle, \quad (24)$$

where we have defined unnormalized states

$$|\beta_A^3\rangle \equiv \sin(\phi/2)|\beta\rangle - i \cos(\phi/2) \sum_{n=0}^{\infty} c_n \cos\left(\sqrt{\frac{n}{\bar{n}}}\frac{\pi}{2}\right)|n\rangle, \quad (25)$$

$$|\beta_M^3\rangle \equiv -\cos(\phi/2) \sum_{n=0}^{\infty} c_n \sin\left(\sqrt{\frac{n}{\bar{n}}}\frac{\pi}{2}\right)|n-1\rangle, \quad (26)$$

and where we have ignored the global phase ($ie^{-i\Delta_{\text{int}}\tau/2}$) in the final states. Finally, the system is measured in the $\{|A\rangle, |M\rangle\}$ basis.

4. A large-amplitude limit of the BEC

We now examine the limit $\bar{n} \rightarrow \infty$. The Poisson distribution of the state (9) has two key properties that will be useful for our purposes. First, in this limit, the atom number distribution $p_n(\bar{n})$ of the BEC becomes highly peaked about the mean atom number in the sense that the standard deviation in atom number, $\Delta n = \sqrt{\bar{n}}$, goes to zero relative to the mean. This property ensures that the interaction of the system with the BEC yields precisely a $\pi/2$ Ramsey pulse in this limit.⁵ The second property is that, in absolute terms, the standard de-

viation grows unbounded and so intuitively we might think that the state of the BEC is negligibly disturbed by the loss of a single atom in this limit. Formally, the states of the reference frame after the first and second interactions approach the following limits⁶ as $\bar{n} \rightarrow \infty$:

$$|\beta_A^1\rangle \rightarrow \frac{1}{\sqrt{2}}|\beta\rangle, \quad |\beta_A^3\rangle \rightarrow \sin(\phi/2)|\beta\rangle, \quad (27)$$

$$|\beta_M^1\rangle \rightarrow -\frac{ie^{i \arg \beta}}{\sqrt{2}}|\beta\rangle, \quad |\beta_M^3\rangle \rightarrow -e^{i \arg \beta} \cos(\phi/2)|\beta\rangle. \quad (28)$$

Therefore, in the limit $\bar{n} \rightarrow \infty$, the evolution of the $\theta=0$ term of Eq. (10), that is, the evolution of $|\Psi_0\rangle_L$ given by Eqs. (11), (19), (22), and (24), reduces to

$$|\Psi_0\rangle_L = |A\rangle|\beta\rangle, \quad (29)$$

$$|\Psi_1\rangle_L \rightarrow \frac{1}{\sqrt{2}}(|A\rangle - ie^{i \arg \beta}|M\rangle)|\beta\rangle, \quad (30)$$

$$|\Psi_2\rangle_L \rightarrow \frac{1}{\sqrt{2}}(|A\rangle - ie^{i(\arg \beta - \phi)}|M\rangle)|\beta\rangle, \quad (31)$$

$$|\Psi_3\rangle_L \rightarrow [\sin(\phi/2)|A\rangle - e^{i \arg \beta} \cos(\phi/2)|M\rangle]|\beta\rangle. \quad (32)$$

We see that the $\theta=0$ term (in fact, any term) remains a product on $\mathcal{H}_S \otimes \mathcal{H}_R$ for the entire experiment. Moreover, applying the twirling operator \mathcal{T} to such a product yields a separable state, which involves classical correlations between the system and the reference frame but no entanglement. In addition, we note that in this limit the reduced density operator of the reference frame remains ρ_0^{rf} of Eq. (8) at each stage; i.e., the reference frame is left undisturbed throughout the experiment.

The probabilities for detecting an atom or molecule at the end display Ramsey oscillations

$$p_A = \sin^2(\phi/2), \quad p_M = \cos^2(\phi/2). \quad (33)$$

This interference pattern has perfect visibility.

5. Interpreting the results

We see therefore that the probabilities for detecting an atom or molecule exhibit the familiar Ramsey oscillations, identified in Sec. II as the operational signature of coherence. So it would seem that we can safely conclude that this experiment demonstrates the possibility of a coherent superposition of an atom and a molecule.

However, the careful reader might have noted the following peculiar fact. From Eqs. (30) and (31), we find that the reduced density operators on \mathcal{H}_S of $|\Psi_2\rangle_L$ or $|\Psi_3\rangle_L$ are

⁵Each Fock state $|n\rangle$ component of the reference-frame state induces a Rabi oscillation between $|A\rangle$ and $|M\rangle$ at frequency $\kappa\sqrt{n+1}$. In the limit $\bar{n} \rightarrow \infty$, the fact that the uncertainty in n goes to zero relative to the mean ensures that all relevant oscillations occur at the same frequency.

⁶These limits follow from the fact that the inner products of the BEC states with the appropriate coherent states approach unity in the limit $\bar{n} \rightarrow \infty$.

$$\frac{1}{\sqrt{2}}(|A\rangle - ie^{i \arg \beta}|M\rangle), \quad (34)$$

$$\frac{1}{\sqrt{2}}(|A\rangle - ie^{i(\arg \beta - \phi)}|M\rangle), \quad (35)$$

respectively. The reduced density operators on \mathcal{H}_S of W_{2L} and W_{3L} are obtained from these by averaging over $\arg \beta$ through the use of the twirling operator \mathcal{T} . In both cases, this state is found to be the completely mixed state $\frac{1}{2}(|A\rangle\langle A| + |M\rangle\langle M|)$, i.e., an *incoherent* mixture of an atom and a molecule. Thus, we have a puzzle: we have clearly predicted the standard operational signature of coherence, namely, Ramsey-type fringes, but the coherence is not present in our mathematical description of the system.

Before suggesting the resolution of this puzzle, we shall demonstrate that it is not unique to the atom-molecule example we are considering. We find precisely the same peculiarity in the context of the original Ramsey experiment, as follows.

Within the analysis of the Ramsey experiment presented in Sec. II, the electromagnetic (EM) field constituting the Ramsey pulses was treated as an external potential. As is well known, this description is perfectly adequate if the fields have large amplitude. But note that one could have also chosen to treat this EM field *within* the quantum formalism, as a dynamical system interacting with the atom. We emphasize that whether the reference frame (the EM field) is treated internally or externally is merely a choice of the physicist. As long as the field has large amplitude, either description is adequate to the task of making accurate predictions about what will be observed by the experimenter.

Treating the reference frame internally, we can take the state of the field to be a coherent state of a single bosonic mode associated with a bosonic annihilation operator \hat{a} . The phase is unimportant (i.e., the demonstration of Ramsey oscillations does not depend on the phase of the coherent state), so one obtains the same experimental predictions if this state is averaged over all phases [29,30], i.e., if one uses a quantum state for the EM field of the same form as the BEC state of Eq. (8).

The relevant Hamiltonian for interacting the two-level atom with the EM field in this internalized description is the Jaynes-Cummings Hamiltonian

$$\hat{H}_{\text{JC}} = \hbar\chi(|g\rangle\langle e| \otimes \hat{a} + |e\rangle\langle g| \otimes \hat{a}^\dagger). \quad (36)$$

Starting with the coherent state averaged over all phases for the EM field and the Jaynes-Cummings interaction Hamiltonian, one is led to a description of the original Ramsey experiment that is precisely equivalent to the one we have just provided for the atom-molecule system.

Along with being formally equivalent to our atom-molecule experiment, this description of the Ramsey experiment with an internalized reference frame is operationally equivalent to the one presented in Sec. II, and necessarily makes the same predictions. In particular, it agrees with the model of Sec. II in predicting the presence of Ramsey fringes. Given that such a model is formally equivalent to the

atom-molecule experiment presented in the previous section, we see that that the latter necessarily predicted the presence of such fringes.

We now turn to the resolution of this puzzle, which applies to both our proposed atom-molecule experiment as well as the Ramsey experiment. We denote the system by S and the reference frame by R . In the case of the Ramsey experiment, S is the atom and R is the EM field constituting the Ramsey pulse, while in the case of the atom-molecule interference experiment, S is the mode pair of atom mode and molecule mode and R is the BEC constituting the analog of the Ramsey pulse.

In such an experiment, if the reference frame is treated externally, the total Hilbert space is denoted \mathcal{H}_S and the quantum state on this Hilbert space describes *the relation between S and R* . As demonstrated in Sec. II, the observation of fringes in this experiment implies coherence between states $|g\rangle$ and $|e\rangle$ of this particular relational degree of freedom. Alternately, if the reference frame is treated internally, the total Hilbert space is denoted by $\mathcal{H}_S \otimes \mathcal{H}_R$; however, in this description the quantum state on \mathcal{H}_S describes the relation between S and a *background* reference frame, distinct from R . Thus although it is standard practice to use a common notation, the Hilbert space we denote by \mathcal{H}_S when R is treated externally and the Hilbert space we denote by \mathcal{H}_S when R is treated internally describe *distinct* degrees of freedom.

Thus, it is a mistake to think that a coherent superposition of states $|g\rangle$ and $|e\rangle$ on \mathcal{H}_S when R is treated externally necessarily implies a coherent superposition of states $|g\rangle$ and $|e\rangle$ on \mathcal{H}_S when R is treated internally. Specifically, if one investigates only the reduced density matrix on \mathcal{H}_S when R is treated internally, no coherence will be found because the reference frame R relative to which these coherences are defined has been discarded in taking the partial trace. To find the relation between S and R when the latter is treated internally, one should not look to the reduced density operator on \mathcal{H}_S but rather to the reduced density operator on a different Hilbert space: one for which its degrees of freedom are the *relation* between S and R . In the following section, we identify this relational Hilbert space and demonstrate that when Ramsey fringes are observed, states in this Hilbert space are indeed coherent.

B. A relational description

We now provide the details of the relational description. This description could be viewed as an example of quantum coherence in the presence of unobservable quantities, in this case the overall phase, discussed in [33]. The averaging over all phases, given by the twirling operator \mathcal{T} , ensures that the density matrix of the laboratory is, at all times in this experiment, block diagonal in the eigenspaces of total type-2 atom number $\hat{N}_2 = \hat{n}_2 + \hat{n}_M$. (Note that this total number operator also counts atoms of type 2 that are bound in molecules.) Thus, we can express the state of the laboratory at each stage as

$$W_{iL} = \sum_{N=0}^{\infty} p_N(\bar{n}) |\Psi_i^{(N)}\rangle_L \langle \Psi_i^{(N)}|, \quad i=0,1,2,3, \quad (37)$$

where

$$|\Psi_i^{(N)}\rangle_L = \Pi_N |\Psi_i\rangle_L, \quad (38)$$

with Π_N the projector onto the eigenspace of \hat{N}_2 with eigenvalue N , spanned by $|A\rangle|N\rangle$ and $|M\rangle|N-1\rangle$.

In the limit $\bar{n} \rightarrow \infty$, the states W_{iL} have most of their support on the subspaces for which $\bar{n} - \sqrt{\bar{n}} \leq N \leq \bar{n} + \sqrt{\bar{n}}$, and using the same approximations as in Sec. III A 4 we have, for N in this range,

$$|\Psi_0^{(N)}\rangle_L \rightarrow |A\rangle|N\rangle, \quad (39)$$

$$|\Psi_1^{(N)}\rangle_L \rightarrow \frac{1}{\sqrt{2}}(|A\rangle|N\rangle - i|M\rangle|N-1\rangle), \quad (40)$$

$$|\Psi_2^{(N)}\rangle_L \rightarrow \frac{1}{\sqrt{2}}(|A\rangle|N\rangle - ie^{-i\phi}|M\rangle|N-1\rangle), \quad (41)$$

$$|\Psi_3^{(N)}\rangle_L \rightarrow \sin(\phi/2)|A\rangle|N\rangle - \cos(\phi/2)|M\rangle|N-1\rangle. \quad (42)$$

The fact that the coefficients in these superpositions are independent of N suggests that, in this limit, we can express these states on an alternate Hilbert space, as we now demonstrate.

We define a new two-dimensional Hilbert space \mathcal{H}_{rel} with an orthonormal basis denoted by $|A\rangle_{\text{rel}}$ and $|M\rangle_{\text{rel}}$, corresponding respectively to $n_M=0$ (no molecules) and $n_M=1$ (one molecule). We call this the *relational* Hilbert space. We also define a new Hilbert space \mathcal{H}_{gl} which has an orthonormal basis labeled by N and defined for $N \geq 1$. We call this the *global* Hilbert space. Define the subspace \mathcal{H}'_L of \mathcal{H}_L as the orthogonal complement to the vector $|A\rangle|0\rangle$. We can define a linear map from the subspace \mathcal{H}'_L of $\mathcal{H}_S \otimes \mathcal{H}_R$ to this new tensor product Hilbert space $\mathcal{H}_{\text{rel}} \otimes \mathcal{H}_{\text{gl}}$ by its action on basis vectors as

$$|A\rangle|N\rangle \mapsto |A\rangle_{\text{rel}}|N\rangle_{\text{gl}}, \quad (43)$$

$$|M\rangle|N-1\rangle \mapsto |M\rangle_{\text{rel}}|N\rangle_{\text{gl}}, \quad (44)$$

for all $N \geq 1$.

It is illustrative to construct this alternate Hilbert space and the associated map (43) and (44) by simultaneously diagonalizing two commuting operators. Note that the states $|A\rangle|N\rangle$ and $|M\rangle|N\rangle$ on $\mathcal{H}_S \otimes \mathcal{H}_R$ are simultaneous eigenstates of the operators \hat{n}_M (or equivalently, \hat{n}_1) and \hat{n}_2 ; the former labels states on \mathcal{H}_S , and the latter labels states on \mathcal{H}_R . Specifically,

$$|A\rangle|N\rangle = |n_M=0, n_2=N\rangle, \quad (45)$$

$$|M\rangle|N\rangle = |n_M=1, n_2=N\rangle. \quad (46)$$

We can instead choose a different set of commuting operators to achieve an alternate tensor product structure for the laboratory Hilbert space. We choose the commuting operators \hat{n}_M and $\hat{N}_2 = \hat{n}_2 + \hat{n}_M$, the latter being the total number operator for atoms of type 2. We note that the states $|A\rangle|N\rangle$ and $|M\rangle|N\rangle$ are also joint eigenstates of \hat{n}_M and \hat{N}_2 , so that we may write

$$|A\rangle|N\rangle = |n_M=0, N_2=N\rangle, \quad (47)$$

$$|M\rangle|N\rangle = |n_M=1, N_2=N+1\rangle. \quad (48)$$

We note that in the limit $\bar{n} \rightarrow \infty$, the states we consider have no support on the vector $|A\rangle|0\rangle = |n_M=0, N_2=0\rangle$ and thus we can focus our attention on the subspace \mathcal{H}'_L of \mathcal{H}_L that is orthogonal to this vector. The states on \mathcal{H}'_L are of the form $|n_M, N_2\rangle$ with $N_2 \geq 1$.

Because the spectra of \hat{n}_M and \hat{N}_2 are independent, we can introduce a new tensor product structure $\mathcal{H}_{\text{rel}} \otimes \mathcal{H}_{\text{gl}}$ on \mathcal{H}'_L which is made by identifying

$$|A\rangle_{\text{rel}}|N\rangle_{\text{gl}} \equiv |n_M=0, N_2=N\rangle, \quad (49)$$

$$|M\rangle_{\text{rel}}|N\rangle_{\text{gl}} \equiv |n_M=1, N_2=N\rangle, \quad (50)$$

for all $N \geq 1$. We then have a vector space isomorphism

$$\mathcal{H}'_L \cong \mathcal{H}_{\text{rel}} \otimes \mathcal{H}_{\text{gl}}. \quad (51)$$

This identification recovers the map of Eqs. (43) and (44).

Note that, under the map of Eqs. (43) and (44), we have

$$\alpha|A\rangle|N\rangle + \beta|M\rangle|N-1\rangle \mapsto (\alpha|A\rangle_{\text{rel}} + \beta|M\rangle_{\text{rel}})|N\rangle_{\text{gl}}, \quad (52)$$

so that while the reduced density operator for this state on the system \mathcal{H}_S is an *incoherent* mixture of $|A\rangle$ and $|M\rangle$, the reduced density operator on the new subsystem \mathcal{H}_{rel} is a *coherent* superposition of $|A\rangle_{\text{rel}}$ and $|M\rangle_{\text{rel}}$.

This fact implies that the states W_{iL} of the laboratory (in the limit $\bar{n} \rightarrow \infty$) map to product states

$$W_{iL} = |\Psi_i\rangle_{\text{rel}} \langle \Psi_i| \otimes \rho_{\text{gl}}, \quad (53)$$

where

$$|\Psi_0\rangle_{\text{rel}} = |A\rangle_{\text{rel}}, \quad (54)$$

$$|\Psi_1\rangle_{\text{rel}} = \frac{1}{\sqrt{2}}(|A\rangle_{\text{rel}} - i|M\rangle_{\text{rel}}), \quad (55)$$

$$|\Psi_2\rangle_{\text{rel}} = \frac{1}{\sqrt{2}}(|A\rangle_{\text{rel}} - ie^{-i\phi}|M\rangle_{\text{rel}}), \quad (56)$$

$$|\Psi_3\rangle_{\text{rel}} = \sin(\phi/2)|A\rangle_{\text{rel}} - \cos(\phi/2)|M\rangle_{\text{rel}}, \quad (57)$$

and

$$\rho_{\text{gl}} = \sum_{N=1}^{\infty} p_N(\bar{n}) |N\rangle_{\text{gl}} \langle N|. \quad (58)$$

This new tensor product structure demonstrates explicitly how we resolve the puzzle posed in the previous section. Both the states of \mathcal{H}_S and the states of \mathcal{H}_{rel} are labeled by the number of molecules, and consequently describe whether the system is an atom or a molecule. Thus, the question “can one have a coherent superposition of an atom and a molecule?” is seen to be ambiguous as stated. Does it refer to a coherent superposition of $|A\rangle$ and $|M\rangle$ on \mathcal{H}_S or to a coherent superposition of $|A\rangle_{\text{rel}}$ and $|M\rangle_{\text{rel}}$ on \mathcal{H}_{rel} ? To resolve the ambiguity, we take an operational stance. By arguing in analogy with the Ramsey interference experiment, we have proposed that an operational signature of coherence is the appearance

of Ramsey fringes, and we have shown that this coincides with having a coherent superposition of $|A\rangle_{\text{rel}}$ and $|M\rangle_{\text{rel}}$ on \mathcal{H}_{rel} .

We finish our analysis by considering the dynamics on the laboratory in terms of the new tensor product structure. Using the map of Eq. (43), we find that the free Hamiltonian for the evolution between Ramsey pulses, defined by Eq. (18), becomes simply

$$\hat{H}'_{\text{free}} = \hbar \Delta_{\text{int}} |M\rangle_{\text{rel}} \langle M|, \quad (59)$$

and the Hamiltonian governing the interaction between the system and the reference frame, Eq. (15), becomes

$$\hat{H}'_{\text{int}} = \frac{\hbar \kappa}{2} (|M\rangle_{\text{rel}} \langle A| + |A\rangle_{\text{rel}} \langle M|) \otimes \sqrt{\hat{N}}_{\text{gl}}, \quad (60)$$

The effective map for the interaction with the BEC is determined as follows. Noting that the interaction occurs for a time $t = \pi/2\kappa\sqrt{\bar{n}}$ and that the initial state on \mathcal{H}_{gl} is ρ_{gl} of Eq. (58), the effective evolution on \mathcal{H}_{rel} is represented by a completely positive trace-preserving map \mathcal{E} of the form

$$\begin{aligned} \mathcal{E}(\rho_{\text{rel}}) &= \text{Tr}_{\text{gl}}(U_{\text{int}}(\rho_{\text{rel}} \otimes \rho_{\text{gl}})U_{\text{int}}^\dagger) \\ &= \sum_N p_N(\bar{n})_{\text{gl}} \langle N|U_{\text{int}}\rho_{\text{rel}}U_{\text{int}}^\dagger|N\rangle_{\text{gl}} \\ &= \sum_N p_N(\bar{n}) e^{i\hat{H}'_{\text{int}}(N)t} \rho_{\text{rel}} e^{-i\hat{H}'_{\text{int}}(N)t}, \end{aligned} \quad (61)$$

where $\hat{H}'_{\text{int}}(N) = \langle N|\hat{H}'_{\text{int}}|N\rangle_{\text{gl}}$. But given that in the limit of large \bar{n} the distribution $p_N(\bar{n})$ is only significant in the range $\bar{n} - \sqrt{\bar{n}} \leq N \leq \bar{n} + \sqrt{\bar{n}}$ and given that $\lim_{\bar{n} \rightarrow \infty} [(\bar{n} \pm \sqrt{\bar{n}})t/\bar{n}t] = 1$, we have that in this limit

$$\mathcal{E}(\rho_{\text{rel}}) = e^{i\hat{H}_{\text{Ram}}t} \rho_{\text{rel}} e^{-i\hat{H}_{\text{Ram}}t}, \quad (62)$$

where

$$\hat{H}_{\text{Ram}} = \frac{\hbar \kappa \sqrt{\bar{n}}}{2} (|M\rangle_{\text{rel}} \langle A| + |A\rangle_{\text{rel}} \langle M|). \quad (63)$$

This has a natural interpretation as the analog of a Ramsey pulse where the pulse is implemented by a BEC that is treated as a classical external field.

We can now make a complete comparison of our relational description of the atom-molecule interference experiment with the original description of the Ramsey experiment in Sec. II (where the fields corresponding to the Ramsey pulses were treated as external potentials). The Hamiltonians \hat{H}'_{free} of Eq. (59) and \hat{H}_{Ram} of Eq. (63) governing the relational degree of freedom are precisely analogous to the Hamiltonians of Eq. (2) and Eq. (1) governing the internal state of the atom in the Ramsey experiment, and the states on \mathcal{H}_{rel} at the four stages of the experiment, given by Eqs. (54)–(57), are precisely analogous to the states for Eqs. (3)–(6) for the internal states of the atom in the Ramsey experiment. We conclude that \mathcal{H}_{rel} can be understood as describing either (i) the relation between the system (atom and molecule modes) and the reference frame formed by the BEC when the latter is treated internally, or (ii) the system

(atom and molecule modes) when the reference frame formed by the BEC is treated as an external potential.

IV. COHERENT SUPERPOSITIONS OF A BOSON AND A FERMION

We now repeat this analysis for the case when the atoms are fermions, and demonstrate that it is possible in principle to exhibit coherence between a fermion (a single atom) and a boson (a molecule). This result is quite surprising; it is commonly accepted that there exists a superselection rule preventing a coherent superposition of a boson and a fermion. We emphasize that we are considering a superposition of a single fermionic atom with a single bosonic molecule, not a superposition of two fermionic atoms and a composite bosonic molecule as considered in [34,35].

Consider our laboratory to consist of two types of atomic species, type 1 and type 2, which are fermions, along with a bosonic diatomic molecule consisting of one of each type of fermion. As with the previous discussion, we will consider creating a superposition of an atom of type 1 and a molecule, using atoms of type 2 as a reference frame.

As we are using fermions, the natural Hilbert space for states of the laboratory will be a Fock space. However, we will want to make use of a *tensor-product structure* of the laboratory Hilbert space which divides it into a system and a reference frame (and, subsequently, into a relational and a global Hilbert space). To do this, we will make use of the natural mapping between the Fock space \mathcal{F}^N of N fermionic modes and the tensor-product Hilbert space $(\mathbb{C}^2)^{\otimes N}$ of N qubits, given by Bravyi and Kitaev [36] in the Fock basis as

$$|n_1, n_2, \dots, n_N\rangle \mapsto |n_1\rangle \otimes |n_2\rangle \otimes \dots \otimes |n_N\rangle \quad (64)$$

for $n_i \in \{0, 1\}$. This identification implies a nontrivial relation between operations on \mathcal{F}^N and operations on $(\mathbb{C}^2)^{\otimes N}$, as a result of phases acquired by commuting operations through occupied modes. Fortunately, due to the highly incoherent nature of the states that we will make use of, and by working with mixed rather than pure states, we will find that this nontrivial identification does not add much additional complication.

Again, we use the notation

$$|A\rangle = |0\rangle_M |1\rangle_A, \quad |M\rangle = |1\rangle_M |0\rangle_A, \quad (65)$$

where the first mode $|\cdot\rangle_M$ is bosonic (the molecule), and the second mode $|\cdot\rangle_A$ is fermionic (atom type 1). We define a system Hilbert space \mathcal{H}_S spanned by these two states $|A\rangle$ and $|M\rangle$.

A. Using a quantum reference frame consisting of fermions

1. Generalizing the previous experiment

In order to construct an interference experiment that exhibits coherence between a boson and a fermion, we must identify an appropriate reference frame consisting of fermions. This is nontrivial, given the difficulties of defining a fermionic coherent state that has analogous properties to the

standard (bosonic) coherent state [37–39]. Again, we meet these challenges by being operational.

We first examine some of the properties of the state of the BEC in the experiment discussed in Sec. III A which allowed it to serve as a good reference frame. To obtain good visibility in the experiment, we required that all relevant Rabi oscillations corresponding to different Fock state components $|n\rangle$ in the state of the reference frame occur at the same frequency, which is obtained by requiring the variance in n to be small compared to the mean, \bar{n} . This requirement could be satisfied by a state with a modest value of \bar{n} , and we could still have predicted good fringe visibility. For example, a single Fock state $|n\rangle$ with $n \geq 1$ satisfies this requirement.

In addition, though, we noted that the reduced density operators for the reference frame at each state of the experiment were undisturbed in the limit $\bar{n} \rightarrow \infty$, which would allow for the experiment to be repeated many times using the same reference frame. For this additional condition to be satisfied, a large absolute variance in total atom number is required (thus implying a large mean total atom number).

We now consider an analogous situation in the fermion case. First, consider a reference frame of fermionic atoms that consists of a single mode. Due to the Pauli exclusion principle, the mean atom number of this reference frame can be at most one. Consider using a Hamiltonian of the form

$$\hat{H} = \frac{\hbar\kappa}{2} (\hat{b}_M^\dagger \hat{f}_2 \hat{f}_A + \hat{f}_A^\dagger \hat{f}_2^\dagger \hat{b}_M), \quad (66)$$

where \hat{b}_M is the boson annihilation operator for the molecular mode, \hat{f}_A is the fermion annihilation operator for the mode of atom type 1, and \hat{f}_2 is the annihilation operator for the reference-frame mode of atom type 2. It is clear that if the initial state of the system is $|A\rangle$ and of the reference frame is $|1\rangle$, then it is possible to perform an interference experiment yielding maximum visibility.

However, in this experiment, the state of the reference frame is highly disturbed. One might (rightly) argue that it is essentially just a Rabi oscillation. We now consider what conditions on the state of the reference frame must be satisfied in order for it to be undisturbed throughout the experiment, thereby making it analogous to the bosonic case in this regard as well. First, we require that the mean number of fermions in the reference frame must be large. To achieve this, because of the Pauli exclusion principle, the reference frame must be multimode. Second, we cannot allow every new system to interact with the same mode of the reference frame. The reason for the latter is that a single mode can have at most one fermion, and if we happen to find a molecule at the end of the Ramsey experiment (and we should assume the worst and say that we do find a molecule) then this fermion has been depleted from that mode. The next system that interacts with this mode will therefore only interact with the vacuum.

We now define a multimode reference frame with a large mean number of atoms, and an associated multimode interaction between the system and the reference frame. The experiment we describe yields high visibility and also leaves the state of the reference frame undisturbed.

2. The quantum state of the reference frame

Consider a K -mode fermionic reference frame of atoms of type 2, using the tensor product structure of Eq. (64), initially prepared in the state

$$\rho_0^{\text{rf}} = \sigma^{\otimes K}, \quad (67)$$

where σ is given by

$$\sigma = \epsilon|0\rangle\langle 0| + (1 - \epsilon)|1\rangle\langle 1|. \quad (68)$$

The distribution of total atom number in the state ρ_0^{rf} is given by the binomial distribution $c_n^K(1 - \epsilon)$, where we define the binomial coefficient $c_n^K(p)$ of n successes in K trials where the probability of success is p as

$$c_n^K(p) = \binom{K}{n} p^n (1 - p)^{K-n}. \quad (69)$$

As we will argue below, in the limit $K \rightarrow \infty$ this atom number distribution has the property that the loss of a single particle leaves the distribution indistinguishable from the original.

3. The interference experiment

We initiate the experiment with the system prepared in the state $|A\rangle$ and the reference frame in the state ρ_0^{rf} of Eq. (67). The initial state of the laboratory is thus

$$W_{0L} = |A\rangle\langle A| \otimes \rho_0^{\text{rf}}. \quad (70)$$

To perform the operation that is analogous to a Ramsey $\pi/2$ pulse in this experiment, we use the Hamiltonian

$$\hat{H}^{(j)} = \frac{\hbar\kappa}{2} (\hat{b}_M^\dagger \hat{f}_2^{(j)} \hat{f}_A + \hat{f}_A^\dagger \hat{f}_2^{(j)\dagger} \hat{b}_M), \quad (71)$$

where \hat{b}_M is the boson annihilation operator for the molecular mode, \hat{f}_A is the fermion annihilation operator for the mode of atom type 1, and $\hat{f}_2^{(j)}$ is the annihilation operator for the j th mode ($j=1, 2, \dots, K$) of the reference frame for atom type 2. Again, this Hamiltonian describes the evolution at a Feshbach resonance, at which the energy of two free fermions (one each of types 1 and 2) is equal to the energy of a bound molecular state.

One might naturally consider implementing the $\pi/2$ pulses by using the unitary operation generated by

$$\hat{H}_{\text{coh}} = \sum_j \hat{H}^{(j)}. \quad (72)$$

The problem with such a Hamiltonian is that it only couples the system with a *single* reference-frame mode, specifically, the mode associated with the fermionic annihilation operator $\hat{F} = \frac{1}{\sqrt{M}} \sum_j \hat{f}_k$. As discussed above, such a coupling is unsatisfactory.

Consider instead the following evolution: the system interacts with a *random* mode j of the reference frame via the interaction $\hat{H}^{(j)}$. We can formalize such an evolution by using the language of quantum operations [40]. Let $U_{\pi/2}^{(j)}$ be the unitary operation that describes $\pi/2$ pulse obtained by cou-

pling the system and the j th reference-frame mode via the Hamiltonian $\hat{H}^{(j)}$ for a time $t = \pi/(2\kappa)$ ⁷. Let U_ϕ be the unitary operation that applies a phase shift $\phi = \Delta_{\text{int}}\tau$ to the $|M\rangle$ component of the system, i.e., evolution for time τ according to the Hamiltonian $\hat{H}_{\text{int}} = \Delta_{\text{int}}|M\rangle\langle M|$.

If the laboratory is initially in the state W_{0L} , and if the system interacts with a *known* reference-frame mode j , the state at each stage of the experiment is given by

$$W_{1L}^{(j)} = U_{\pi/2}^{(j)} W_{0L} (U_{\pi/2}^{(j)})^{-1}, \quad (73)$$

$$W_{2L}^{(j)} = U_\phi U_{\pi/2}^{(j)} W_{0L} (U_\phi U_{\pi/2}^{(j)})^{-1}, \quad (74)$$

$$W_{3L}^{(j)} = U_{\pi/2}^{(j)} U_\phi U_{\pi/2}^{(j)} W_{0L} (U_{\pi/2}^{(j)} U_\phi U_{\pi/2}^{(j)})^{-1}, \quad (75)$$

where the unitaries $U^{(j)}$ are taken to act as the identity on all modes other than the j th mode.

To describe an evolution where, with probability $1/K$, the system interacts with the j th reference-frame mode, we make use of generalized quantum operations, which have the form of completely positive trace-preserving maps given, in this instance, by convex combinations of unitary transformations. We describe the evolution that takes the laboratory from the initial state W_{0L} to a state W_{iL} for steps $i=1,2,3$ as

$$W_{1L} = \mathcal{E}_1(W_{0L}) = \frac{1}{K} \sum_{j=1}^K U_{\pi/2}^{(j)} W_{0L} (U_{\pi/2}^{(j)})^{-1}, \quad (76)$$

$$W_{2L} = \mathcal{E}_2(W_{0L}) = \frac{1}{K} \sum_{j=1}^K U_\phi U_{\pi/2}^{(j)} W_{0L} (U_\phi U_{\pi/2}^{(j)})^{-1}, \quad (77)$$

$$W_{3L} = \mathcal{E}_3(W_{0L}) = \frac{1}{K} \sum_{j=1}^K U_{\pi/2}^{(j)} U_\phi U_{\pi/2}^{(j)} W_{0L} (U_{\pi/2}^{(j)} U_\phi U_{\pi/2}^{(j)})^{-1}. \quad (78)$$

As noted above, there is a nontrivial relation between a unitary $U_{\pi/2}^{(j)}$ coupling two modes together in the tensor-product Hilbert space, and the same coupling on the Fock space, due to the phases acquired by commuting through modes $j' < j$; in general, this mapping can be determined using the techniques of [36]. However, this mapping does not exhibit any nontrivial consequences for the interaction presented here, for the following simple reason. Note that each sequence of unitary operations (either $U_{\pi/2}^{(j)}$, $U_\phi U_{\pi/2}^{(j)}$, or $U_{\pi/2}^{(j)} U_\phi U_{\pi/2}^{(j)}$) only couples the system with a single reference-frame mode j ; such coupling will thus lead to a nontrivial phase due to the modes $j' < j$. However, the expressions above sum *incoherently* over the different possibilities j , and thus the phases acquired for each term in this sum do not interfere.

⁷Note that the interaction times do not scale inversely with the square root of the mean atom number as only one reference-frame mode containing at most one fermion interacts. Essentially, the system undergoes a type of Rabi oscillation with a single mode of the reference frame.

For clarity, it will be useful to follow the evolution associated with the $j=1$ element of the above operation, remembering at each step that the state of the laboratory is described by interacting the system with a random mode. To do this, we define the *shuffling operation* \mathcal{S} , which is the incoherent symmetrizer acting on states of the reference frame as

$$\mathcal{S}[\rho^{\text{rf}}] = \frac{1}{K!} \sum_{\pi} S_{\pi} \rho^{\text{rf}} S_{\pi}^{\dagger}, \quad (79)$$

where the sum is over all permutations π of K indices and S_{π} is the unitary representation of the symmetric group on K fermion modes. We determine the evolution for the system interacting with the $j=1$ mode of the reference frame at each step, and then apply the shuffling operator to the state of the reference frame to obtain the state corresponding to an interaction with a random mode as in Eqs. (76)–(78).

We first apply a $\pi/2$ pulse by evolving with the Hamiltonian $\hat{H}^{(j=1)}$ for time $t = \pi/(2\kappa)$ (step 1), then freely evolving off resonance for time τ (step 2), followed by another $\pi/2$ pulse (step 3), yielding at each step $i=0,1,2,3$

$$[\epsilon|A\rangle\langle A| \otimes |0\rangle\langle 0| + (1-\epsilon)|\Psi_i\rangle_L\langle\Psi_i|] \otimes \sigma^{\otimes K-1}, \quad (80)$$

where

$$|\Psi_0\rangle_L = |A\rangle|1\rangle, \quad (81)$$

$$|\Psi_1\rangle_L = \frac{1}{\sqrt{2}}(|A\rangle|1\rangle - i|M\rangle|0\rangle), \quad (82)$$

$$|\Psi_2\rangle_L = \frac{1}{\sqrt{2}}(|A\rangle|1\rangle - ie^{-i\phi}|M\rangle|0\rangle), \quad (83)$$

$$|\Psi_3\rangle_L = \cos(\phi/2)|A\rangle|1\rangle - \sin(\phi/2)|M\rangle|0\rangle. \quad (84)$$

Because the mode with which the system interacts is unknown, the state of the laboratory at any particular stage i of the experiment may be written

$$W_{iL} = \mathcal{S}[(\epsilon|A\rangle\langle A| \otimes |0\rangle\langle 0| + (1-\epsilon)|\Psi_i\rangle_L\langle\Psi_i|) \otimes \sigma^{\otimes K-1}], \quad (85)$$

where, in a slight abuse of notation, it is understood that \mathcal{S} acts only on the K reference-frame modes and as identity on the system. It is straightforward to show that these states are equivalent to those presented in Eqs. (76)–(78).

The probabilities for measuring an atom or a molecule after the final step are, respectively,

$$P_A = \text{Tr}[W_{3L}|A\rangle\langle A|] = \epsilon + (1-\epsilon)\cos^2(\phi/2), \quad (86)$$

$$P_M = \text{Tr}[W_{3L}|M\rangle\langle M|] = (1-\epsilon)\sin^2(\phi/2), \quad (87)$$

yielding a visibility of $V=(1-\epsilon)$. Thus, for small ϵ , we can achieve high visibility for the Ramsey fringes.

Along with achieving a high visibility in the limit $\epsilon \rightarrow 0$, this result possesses another analogy with the bosonic experiment of Sec. III: in the limit $K \rightarrow \infty$, the state of the reference frame is left undisturbed throughout the experiment. Specifically, the reduced density operator of the refer-

ence frame remains ρ_0^{rf} of Eq. (67) after the experiment, regardless of what measurement outcome (atom or molecule) is obtained. We provide a proof of this fact in the Appendix.

4. Interpreting the results

In the limit discussed above, the state of the reference frame described above is undisturbed throughout the interference experiment. Specifically, if the system is prepared in the state $|A\rangle$ and the measurement result $|M\rangle$ is obtained, the state of the reference frame is undisturbed even though a fermionic atom has been removed from the reference frame. This property is shared with the single-mode bosonic atom state of Eq. (8).

It is illustrative to compare the single-mode bosonic atom state of Eq. (8) and the multimode fermionic atom state of Eq. (67), and to address possible concerns of a skeptic who questions whether the latter is a good generalization of the former for the purposes of a Ramsey experiment.

Bosons admit coherent states that are eigenstates of the annihilation operator; such coherent states yield good visibility in an interference experiment, and are also undisturbed by the interactions. Thus, one way to explain the fact that the state (8) is undisturbed by the interference experiment is to note that every coherent state would be undisturbed, so the convex sum of them will also be undisturbed. One might naively expect that these properties *cannot* be generalized to fermionic states, because it is not possible to define pure states of (single -or multimode) fermionic systems that are eigenstates of an annihilation operator.

However, no convex decomposition should be preferred over others. We can also view the mixed bosonic state of Eq. (8) as a Poissonian distribution of number states, as in Eq. (9). In that case, it is the large uncertainty in total number that explains why the mixed state is undisturbed. One can appeal to this same sort of explanation in the fermionic case. A skeptic might still claim that there is a difference, namely, that “really” the bosonic system is in a coherent state, and it is the nondisturbance to this real state that is significant. However, to make this statement is to commit the partition ensemble fallacy [41].

Another possible distinction between these two states that a skeptic might claim to be important is that, in the fermionic case, there is classical information (the integer j) indicating the mode of the reference frame with which the system interacted. If one considers the state of this mode, then it is highly disturbed by the interaction. However, the classical uncertainty about which mode was the mode of interaction ensures that the reference frame, as a whole, is undisturbed. This classical uncertainty ensures that, if the experiment was repeated many times, there is only a vanishing probability that the system will interact with the same mode more than once; thus, visibility is maintained for many runs of the experiment.

There are indeed significant differences between the bosonic and fermionic states described here, in terms of how they can be used as reference frames. We discuss one such difference in Sec. V B. However, we emphasize that, if one takes an operational view of these Ramsey-type interference experiments, then the bosonic and fermionic examples are

completely equivalent in that they produce high-visibility fringes for potentially many repetitions of the experiments.

B. A relational description

This experiment exhibits high-visibility Ramsey fringes, demonstrating coherence between a fermionic atom and a bosonic molecule. However, as with the experiment illustrated in Sec. III A, the reduced density operator for the system at all times during the experiment is diagonal in the $|A\rangle, |M\rangle$ basis. To observe the coherence, one must instead look to a relational description, which we now develop in analogy to that presented in Sec. III B.

First, we introduce some simplifying notation. Let \vec{n} be a vector in $(\mathbb{Z}_2)^{\otimes K}$, i.e., a K -dimensional vector where each element $n^{(i)}$ is either zero or one. Then $|\vec{n}\rangle = |n^{(1)}, n^{(2)}, \dots, n^{(K)}\rangle$ is a Fock state of K fermionic modes.

Now, note that the states W_{iL} of Eq. (85) are block diagonal in the eigenspaces of total number of atoms of type 2 (counting any atoms of type 2 which are bound into molecules as well). Defining a total type-2 atom number operator

$$\hat{N}_2 = \hat{n}_M + \sum_{j=1}^K \hat{n}_2^{(j)}, \quad (88)$$

where $\hat{n}_M = \hat{b}_M^\dagger \hat{b}_M$ and $\hat{n}_2^{(j)} = \hat{f}_2^{(j)\dagger} \hat{f}_2^{(j)}$, we can express

$$W_{iL} = \sum_{N=0}^K c_N^{(K)} (1 - \epsilon) W_{iL}^{(N)}, \quad (89)$$

where $c_N^{(K)}(p)$ is defined in Eq. (68) and where the states $W_{iL}^{(N)}$ are eigenstates of \hat{N}_2 with eigenvalue N . In the limit $K \rightarrow \infty$, $\epsilon \rightarrow 0$ with $K\epsilon$ fixed, this expression becomes

$$\lim W_{iL} = \sum_{N=0}^{\infty} p_N(\bar{n}) W_{iL}^{(N)}, \quad (90)$$

with $\bar{n} = K(1 - \epsilon)$, with the states $W_{iL}^{(N)}$ given by

$$W_{iL}^{(N)} = \mathcal{S}[|\Psi_i\rangle_L \langle \Psi_i| \otimes |\vec{n}_{N-1}\rangle \langle \vec{n}_{N-1}|], \quad (91)$$

where \vec{n}_{N-1} is the vector $(1, 1, \dots, 1, 0, \dots, 0)$ with $N-1$ ones and $K-N$ zeros, \mathcal{S} is the shuffling operator of Eq. (79), and the states $|\Psi_i\rangle_L$ are given by Eqs. (81)–(84). The fact that the states $|\Psi_i\rangle_L$ are independent of N suggests an alternate partitioning of the Hilbert space, as follows.

First note that our current tensor product structure for the Hilbert space, in terms of modes, is associated with the eigenvalues of the number operators

$$\hat{n}_M, \hat{n}_2^{(1)}, \hat{n}_2^{(2)}, \dots, \hat{n}_2^{(K)}. \quad (92)$$

We choose the following different set of commuting operators:

$$\hat{n}_M, \hat{N}_2, \hat{r}_2^{(i)} \equiv \hat{n}_2^{(i)} - \hat{n}_2^{(i-1)}, \quad i = 2, \dots, K. \quad (93)$$

We note that the states $|A\rangle|\vec{n}\rangle$ and $|M\rangle|\vec{n}\rangle$ are also eigenstates of this new set of operators, specifically,

$$\hat{n}_M |A\rangle|\vec{n}\rangle = 0, \quad (94)$$

$$\hat{n}_M |M\rangle |\vec{n}\rangle = |M\rangle |\vec{n}\rangle, \quad (95)$$

$$\hat{N}_2 |A\rangle |\vec{n}\rangle = \left(\sum_i n^{(i)} \right) |A\rangle |\vec{n}\rangle, \quad (96)$$

$$\hat{N}_2 |M\rangle |\vec{n}\rangle = \left(1 + \sum_i n^{(i)} \right) |M\rangle |\vec{n}\rangle, \quad (97)$$

$$\hat{r}_2^{(i)} |A\rangle |\vec{n}\rangle = (n^{(i)} - n^{(i-1)}) |A\rangle |\vec{n}\rangle, \quad (98)$$

$$\hat{r}_2^{(i)} |M\rangle |\vec{n}\rangle = (n^{(i)} - n^{(i-1)}) |M\rangle |\vec{n}\rangle. \quad (99)$$

We note that in the limit $\epsilon \rightarrow 0$, the states we consider have no support on the vector $|A\rangle |0, 0, \dots, 0\rangle$ and thus we can focus our attention on the subspace \mathcal{H}'_L of \mathcal{H}_L that is orthogonal to this vector.

We introduce a new tensor-product structure on \mathcal{H}'_L as follows. We define a two-dimensional Hilbert space \mathcal{H}_{rel} with an orthonormal basis denoted by $|A\rangle_{\text{rel}}$ and $|M\rangle_{\text{rel}}$, corresponding respectively to $n_M=0$ (no molecules) and $n_M=1$ (one molecule). We call this the *relational* Hilbert space. We also define a Hilbert space \mathcal{H}_{gl} which has an orthonormal basis labeled by (N_2, \vec{r}_2) , where \vec{r}_2 is the vector consisting of the eigenvalues of the operators $\hat{r}_2^{(i)}$ for $i=2, 3, \dots, K$. These labels are defined for $N_2 \geq 1$, and \vec{r}_2 consistent with this total type-2 atom number N_2 . We call this the *global* Hilbert space. Then, because the spectra of \hat{n}_M and that of the operators \hat{N}_2 and $\hat{r}_2^{(i)}$ are independent, we have a virtual tensor-product structure

$$\mathcal{H}'_L \simeq \mathcal{H}_{\text{rel}} \otimes \mathcal{H}_{\text{gl}}, \quad (100)$$

which is defined in terms of a linear map from $\mathcal{H}_S \otimes \mathcal{H}_R$ to $\mathcal{H}_{\text{rel}} \otimes \mathcal{H}_{\text{gl}}$ in terms of their respective basis states as

$$|A\rangle |\vec{n}\rangle \mapsto |A\rangle_{\text{rel}} \left| N_2 = \left(\sum_i n_2^{(i)} \right), \vec{r}_2 \right\rangle_{\text{gl}}, \quad (101)$$

$$|M\rangle |\vec{n}\rangle \mapsto |M\rangle_{\text{rel}} \left| N_2 = \left(1 + \sum_i n_2^{(i)} \right), \vec{r}_2 \right\rangle_{\text{gl}}, \quad (102)$$

where $r_2^{(i)} = n_2^{(i)} - n_2^{(i-1)}$.

The states W_{iL} of Eq. (90) are expressed in terms of this new tensor-product structure as

$$W_{iL} = |\Psi_i\rangle_{\text{rel}} \langle \Psi_i| \otimes \rho_{\text{gl}}, \quad (103)$$

where the states on the relational Hilbert space are pure, given by

$$|\Psi_0\rangle_{\text{rel}} = |A\rangle_{\text{rel}}, \quad (104)$$

$$|\Psi_1\rangle_{\text{rel}} = \frac{1}{\sqrt{2}} (|A\rangle_{\text{rel}} - i|M\rangle_{\text{rel}}), \quad (105)$$

$$|\Psi_2\rangle_{\text{rel}} = \frac{1}{\sqrt{2}} (|A\rangle_{\text{rel}} - ie^{-i\phi}|M\rangle_{\text{rel}}), \quad (106)$$

$$|\Psi_3\rangle_{\text{rel}} = \cos(\phi/2)|A\rangle_{\text{rel}} - \sin(\phi/2)|M\rangle_{\text{rel}}. \quad (107)$$

The state ρ_{gl} on \mathcal{H}_{gl} is identified via the following observations about the shuffling operation \mathcal{S} . First, if a state ρ has support entirely within an eigenspace of the operators \hat{N}_2 and \hat{n}_M with eigenvalues N and n_m , respectively, then $\mathcal{S}[\rho]$ also has support entirely within this same eigenspace; intuitively, this is because symmetrization does not alter the total type-2 atom number, or whether a type-2 atom is bound into a molecule or not. (Recall that the shuffling operation acts only on the state of the reference frame.) Second, the shuffling operation completely randomizes the eigenvalues of the operators $\hat{r}^{(i)}$, i.e., if ρ on \mathcal{H}'_L has support entirely within an eigenspace of the operators \hat{N}_2 and \hat{n}_M with eigenvalues N and n_m , respectively, then $\mathcal{S}[\rho]$ is a uniform mixture of all states on \mathcal{H}'_L that are eigenstates \hat{N}_2 and \hat{n}_M with the same eigenvalues.

Thus, the state ρ_{gl} is given by

$$\rho_{\text{gl}} = \sum_{N=0}^{\infty} p_N(\vec{n}) \Sigma_N, \quad (108)$$

where Σ_N is the completely mixed state on the eigenspace of \hat{N}_2 in \mathcal{H}_{gl} with eigenvalue N .

Thus we see that having fermionic atoms does not present any new difficulties compared to the case of bosonic atoms, so that one may interpret the fermionic version of this interference experiment in precisely the same way that we interpreted the bosonic version, as described in Secs. III A 5 and III B.

V. DISCUSSION

A. Experimental considerations

The above descriptions of our proposed experiments are clearly idealized and intended to illustrate the essential physics. We now address some of the issues that may arise in attempting to perform our proposed experiments in real systems of ultracold bosonic or fermionic atoms and molecules.

Both the bosonic and fermionic versions of our proposed experiment require creation of mixtures of degenerate atoms of two different species. Such mixtures have now been created with a number of different atomic species via the process of sympathetic cooling [42–46]. In [42] a mixture of two BECs of different bosonic species ^{41}K and ^{87}Rb was created. Furthermore the location of Feshbach resonances in these two atomic species was estimated in [47]. We therefore consider these two species as good candidates for implementing the bosonic version of our experiment described in Sec. III. For the fermionic version of the experiment described in Sec. IV we require a mixture of two different fermionic species. While mixtures of bosonic and fermionic species are common [43–46], to our knowledge simultaneous degeneracy of two different fermionic atom species has not yet been achieved experimentally. However, a degenerate mixture of two different spin states of the same fermionic atom has been created [48], which would suffice as the two distinguishable fermionic species for our purposes.

Clearly one of the issues in performing the experiment, once appropriate atomic species have been chosen, is detec-

tion. We seem to require the ability to perform a projective measurement of a single atom or molecule in order to observe the interference pattern, Eq. (33). The issue of detecting single atoms also arises in the context of quantum-information processing (QIP) with neutral atoms in optical lattices and magnetic microtraps (see [49] for a review of experimental progress, and references therein). It may be possible to perform the projective measurement of atom or molecule using the fiber-based Fabry-Pérot resonators described in [49].

Alternatively, one may attempt to perform many copies of the experiment simultaneously by beginning with an optical lattice containing atoms of the first species in the Mott-insulator regime at unit filling, so that atoms in different lattice sites are essentially noninteracting. Again, the creation of optical lattices containing precisely one atom per site has been considered in the context of QIP with neutral atoms [49]. We would then seem to require a separate BEC of the second atomic species at each site to create distinct copies of the same experiment. However, this could be challenging experimentally and given that we are working in the classical limit it may suffice to use a single BEC with a spatial profile that overlaps the entire lattice.⁸ The interactions could be implemented by tuning an external magnetic field (uniform over the lattice) onto the Feshbach resonance. Free evolution could be implemented by switching off the external magnetic field for the desired period of time. At the end of the experiment standard techniques should suffice to detect the number of atoms of the first species present in the lattice (molecules being typically much more difficult to detect), and we would expect to see an interference pattern in this number as a function of the free-evolution time.

Next, from a theoretical perspective, one may question whether the single-mode description used in Sec. III is adequate for the BEC. For small BECs there is some “quantum depletion” whereby some atoms do not occupy the condensate, even at zero temperature, due to interactions between the atoms [26]. However this effect is small (a few percent or less) in most current experiments, and in any case we are concerned with the limit of a large BEC—precisely the limit in which the mean-field dynamics given by the Gross-Pitaevskii equation become exact.

For the experiment involving fermions described in Sec. IV both preparation of the initial state and implementation of the interaction are likely to be far more difficult. We do not have any concrete suggestions for how this experiment might be performed, however we note that the initial state Eq. (67) may be well approximated by fermions near the Fermi level at a small but nonzero temperature, i.e., the different fermion modes are the momentum modes close to the Fermi momentum. Alternatively, it may be more convenient to use spatially isolated modes, such as the lattice sites in an optical lattice, as the multimode fermionic reference state. Fermionic atoms were confined to a three dimensional optical lattice in [48], so the reference state (67) could perhaps be

created by trapping slightly fewer atoms than the number of lattice sites, so that the vacant sites are randomly distributed. One must also address the issue of implementing the nonunitary coupling described by Eqs. (76)–(78). It may be possible to implement such a coupling using the Hamiltonian of Eq. (72) if the reference frame is kept in thermodynamic equilibrium with a particle heat bath that can replenish an exhausted fermionic mode.

Finally, another question that must be addressed concerns the time scales and mechanisms for decoherence. One might expect interactions with background atoms and molecules and with uncondensed thermal atoms to cause the superpositions to decohere, but whether or not this would occur on a time scale shorter than is necessary to observe the interference fringes has not been studied, to our knowledge.

Thus there are clearly many experimental challenges to be overcome. However many of the required elements have been demonstrated individually. With the rapid pace of progress in the field of ultracold atomic physics, where molecular condensates, degenerate Fermi gases, and Bose-Fermi mixtures are all topics of much current interest, it is plausible that experiments of this type may be performed in the near future. This would open up the possibility of experimentally investigating the role of superselection rules in these systems.

B. Lifting superselection rules

If a superselection rule is completely lifted by the existence of an appropriate reference frame, meaning that one can in principle perform any quantum operation as if the superselection rule did not exist, then one would expect to be able to perform an experiment on *two* systems that exhibits a relative phase between these systems, independent of the reference frame. Specifically, one could generalize the methods we presented here to define a two-system relational Hilbert space for the composite; states on this relational Hilbert space would describe the two systems relative to the single reference frame. One would expect that some degrees of freedom in this relational Hilbert space describe the relation between the two systems themselves, independent of the reference frame. For example, one could perform measurements of observables defined on the relational Hilbert space that provide information about the relative phase of the two systems.

Consider the following experiment. Two two-level systems such as those described in this paper are initially prepared as single atoms. A $\pi/2$ pulse is performed on the first system followed by a $\pi/2$ pulse on the second system (both pulses being implemented by interaction with the reference frame), and then a phase shift ϕ is applied to the second system. One would expect, then, that a measurement of the relative phase on the relational Hilbert space could yield information about ϕ . (One such measurement would be the two-outcome projection onto the symmetric and antisymmetric subspaces of the relational Hilbert space. If the two systems are in phase, the symmetric outcome is obtained with certainty.) For the bosonic reference frames explored in Sec. III, it is straightforward to show that this is indeed the case.

⁸The variation in phase over the profile of the BEC is unlikely to be relevant because, as we discussed above, the absolute phase of the BEC is irrelevant to the interference pattern.

However, for the fermion reference frame introduced in Sec. IV, such an experiment would be completely insensitive to ϕ . The reason for this insensitivity is that, for the interactions describing the $\pi/2$ pulses, each of the two systems would interact with a *different* random mode of the reference frame. As the individual reference-frame modes are uncorrelated in phase, the two systems would also be uncorrelated.

Thus, although our second example of a Ramsey interference experiment exhibits a coherent superposition of a single fermionic atom and a bosonic molecule, we do not currently foresee how such an experiment can be directly generalized to create arbitrary superposition states of multiple systems. The form of reference frame used in Sec. IV does not lift the superselection rule, but provides only a demonstration of a violation of this rule in a single system.

VI. CONCLUSIONS

We conclude by responding to some anticipated objections by a skeptic who questions that our gedanken experiments would exhibit a coherent superposition of a single atom and a molecule. Suppose a skeptic asserted that the only adequate description of the atom-molecule interference experiments is the one wherein the BEC is internalized, that is, treated quantum mechanically. She could appeal to the fact that there is no coherence between the states $|A\rangle$ and $|M\rangle$ on \mathcal{H}_S in this description to argue that a coherent superposition of atom and molecule had actually not been generated. We would of course disagree with the assessment that internalizing the reference frame is the only way to obtain an adequate description of the experiment, but leaving this aside, one can respond to such a skeptic by noting that this argument would also apply to the Ramsey experiment outlined in Sec. II. The reason is that when the EM field in the Ramsey experiment is treated quantum mechanically, then the reduced density operator on the Hilbert space of the atom has no coherence between the internal states $|A\rangle$ and $|B\rangle$.

Thus, a skeptic could deny that we have demonstrated the possibility of having a coherent superposition of atom and molecule, but then she would also have to deny that the Ramsey interference experiment demonstrates coherence between two internal states of an atom. In fact, the sort of argument the skeptic is presenting can be applied to essentially *any* degree of freedom that requires a reference frame for its definition.

For instance, suppose one internalized the reference frame for spatial orientation or for the spatial position of a system. Then by exactly analogous arguments to those presented here, the reduced density operator on the Hilbert space of the system would be found to be an *incoherent* sum of angular momentum or linear momentum eigenstates. Thus, a dogmatic insistence on the necessity of internalizing reference frames would lead one in this context to conclude that it is impossible to prepare coherent superpositions of angular momentum or linear momentum eigenstates. But although the

latter quantities are conserved, no one feels that it is fruitful to insist on a superselection rule for them.⁹

The only real difference we can see between reference frames for spatial orientation, position, or the phase of the internal state of an atom, on the one hand, and for the phase conjugate to charge, atom number, or univalence on the other, is that reference frames of the first sort are ubiquitous while those of the latter sort are difficult to prepare. Any rigid object can act as a reference frame for spatial position, whereas a reference for the phase conjugate to atom number presumably requires one to have succeeded in the experimentally challenging task of achieving and maintaining Bose-Einstein condensation for that atomic species.

Along these lines, we note that the recent demonstrations of Ramsey fringes in so-called superconducting qubits in a single-Cooper-pair box configuration [19] is analogous in many ways to the experiments we propose here (see [50] for related theory). This experiment demonstrates coherence between states of differing charge, i.e., of states of a superconducting island which differ by charge $2e$ (the charge of a single Cooper pair). The reference frame for this system, which is used to implement the Ramsey pulses, is a nearby superconductor, the state of which—a BCS ground state—has an accurate and successful description as a classical field. This superconducting qubit experiment can be interpreted as violating a superselection rule for charge [4,51], in direct analogy with the way that the interference experiments proposed here can be interpreted as violating a superselection rule for atom number.

ACKNOWLEDGMENTS

The authors gratefully acknowledge Howard Wiseman for pointing out that the univalence superselection rule has not been lifted, Dominic Berry for suggesting the method used to prove Eq. (A14), and Andrew Doherty and David Poulin for helpful discussions. S.D.B. acknowledges support from the Australian Research Council. T.R. acknowledges support from the Engineering and Physical Sciences Research Council of the United Kingdom. R.W.S. acknowledges support from the Royal Society.

APPENDIX: PROOF THAT THE STATE OF THE FERMIONIC REFERENCE FRAME IS UNDISTURBED

The state of the laboratory after the second $\pi/2$ pulse is given by Eq. (85) using Eq. (84) as

$$W_{3L} = \mathcal{S}[(\epsilon|A\rangle\langle A| \otimes |0\rangle\langle 0| + (1 - \epsilon)|\Psi_3\rangle_L\langle\Psi_3|) \otimes \sigma^{\otimes K-1}], \quad (\text{A1})$$

with

⁹An exception is found in some approaches to quantum gravity. The argument for why such superselection rules should be in force for descriptions of the quantum state of the universe is precisely because there are no external reference frames in such a description.

$$|\Psi_3\rangle_L = \cos(\phi/2)|A\rangle|1\rangle - \sin(\phi/2)|M\rangle|0\rangle. \quad (\text{A2})$$

Consider the postselected states of the reference frame conditioned on measuring an atom or a molecule, given by

$$\rho_A^{\text{rf}} = \frac{\langle A|W_{3L}|A\rangle}{p_A}, \quad \rho_M^{\text{rf}} = \frac{\langle M|W_{3L}|M\rangle}{p_M}, \quad (\text{A3})$$

which occur with probabilities given by Eqs. (87) and (86). These postselected states are diagonal in the fermion-number basis and can be expressed as

$$\rho_A^{\text{rf}} = \frac{1}{p_A} \sum_{n=0}^{K-1} c_n^{K-1} (1-\epsilon) [\epsilon \mathcal{S}[|1\rangle^{\otimes n}|0\rangle^{K-n}] + (1-\epsilon) \cos^2(\phi/2) \mathcal{S}[|1\rangle^{\otimes n+1}|0\rangle^{K-n-1}]], \quad (\text{A4})$$

$$\rho_M^{\text{rf}} = \sum_{n=0}^{K-1} c_n^{K-1} (1-\epsilon) \mathcal{S}[|1\rangle^{\otimes n}|0\rangle^{K-n}]. \quad (\text{A5})$$

The postselected state conditioned on an atom, ρ_A^{rf} , can be written as a sum of two terms,

$$\rho_A^{\text{rf}} = \frac{1}{p_A} [\epsilon \rho_M^{\text{rf}} + (1-\epsilon) \cos^2(\phi/2) \tilde{\rho}_A^{\text{rf}}], \quad (\text{A6})$$

where we define

$$\tilde{\rho}_A^{\text{rf}} = \sum_{n=1}^K c_{n-1}^{K-1} (1-\epsilon) \mathcal{S}[|1\rangle^{\otimes n}|0\rangle^{K-n}]. \quad (\text{A7})$$

We now show that the postselected states ρ_A^{rf} and ρ_M^{rf} are indistinguishable from each other, and from the initial state ρ_0^{rf} , in the limit of $K \rightarrow \infty$ for a fixed $\epsilon > 0$. The intuition for expecting this result is that the total number of fermions in the reference frame is indeterminate (a binomial distribution) so that in the limit of large number of fermions, distributed in an even larger number of modes, we will not be able to tell if one of the modes has interacted with the system and possibly lost one fermion. The random coupling of the map \mathcal{E}_U ensures that we do not know which fermion mode to look at.

From (A6) it is clear that the nontrivial step is to show that $\tilde{\rho}_A^{\text{rf}}$ and ρ_M^{rf} are indistinguishable in the limit. We will use the fidelity, defined as [40]

$$F(\rho, \sigma) = \text{Tr}(\sqrt{\rho^{1/2} \sigma \rho^{1/2}}), \quad (\text{A8})$$

as our measure of the indistinguishability of two states ρ and σ , where $F=1$ implies that the states are completely indistinguishable. For states $\rho = \sum_i r_i |i\rangle\langle i|$ and $\sigma = \sum_i s_i |i\rangle\langle i|$ diagonal in the same basis the fidelity reduces to

$$F(\rho, \sigma) = \sum_i \sqrt{r_i s_i}. \quad (\text{A9})$$

Therefore the fidelity of these two states is

$$F(\tilde{\rho}_A^{\text{rf}}, \rho_M^{\text{rf}}) = \sum_{n=1}^{K-1} \sqrt{c_{n-1}^{K-1} c_n^{K-1}} \quad (\text{A10})$$

$$= 1 - \frac{c_0^{K-1}}{2} - \frac{c_{K-1}^{K-1}}{2} - \frac{1}{2} \sum_{n=1}^{K-1} (\sqrt{c_{n-1}^{K-1}} - \sqrt{c_n^{K-1}})^2 \quad (\text{A11})$$

$$\geq 1 - \frac{c_0^{K-1}}{2} - \frac{c_{K-1}^{K-1}}{2} - \frac{1}{2} \sum_{n=1}^{K-1} |\sqrt{c_{n-1}^{K-1}} - \sqrt{c_n^{K-1}}| \quad (\text{A12})$$

$$= 1 - c_0^{K-1} - c_{K-1}^{K-1} - c_{\text{max}}^{K-1} \quad (\text{A13})$$

$$\rightarrow 1, \quad (\text{A14})$$

where $c_{\text{max}}^{K-1} = \max_n c_n^{K-1}$ is the probability of the most likely number of fermions (the floor or ceiling of the mean) and we have not written explicitly the dependence on $(1-\epsilon)$ of the binomial probabilities.

Therefore from (A6) it is clear that the fidelity $F(\tilde{\rho}_A^{\text{rf}}, \rho_M^{\text{rf}})$ between the two postselected states also approaches 1 in the limit $K \rightarrow \infty$. Similar calculations show the same result for the postselected states of the reference frame at either of the intermediate times in the experiment, Eq. (82) or (83) substituted into Eq. (85). Furthermore either of the postselected states of the reference frame at any time during the experiment can be shown to have fidelity approaching 1 with the initial state ρ_0^{rf} in the same limit. Therefore we have the result we claimed—the reference frame is undisturbed by the interaction in the limit of a large number of modes.

-
- [1] G. C. Wick, A. S. Wightman, and E. P. Wigner, *Phys. Rev.* **88**, 101 (1952).
[2] E. Joos, H. D. Zeh, C. Kiefer, D. Giulini, K-Kupsch, I.-O. Stamatescu, *Decoherence and the Appearance of a Classical World in Quantum Theory* (Springer-Verlag, Berlin, 1996).
[3] F. Strocchi and A. S. Wightman, *J. Math. Phys.* **15**, 2198 (1974).
[4] Y. Aharonov and L. Susskind, *Phys. Rev.* **155**, 1428 (1967).
[5] R. Mirman, *Phys. Rev.* **186**, 1380 (1969); *Phys. Rev. D* **1**, 3349 (1970).
[6] A. Shimizu and T. Miyadera, e-print cond-mat/0102429.
[7] A. Kitaev, D. Mayers, and J. Preskill, *Phys. Rev. A* **69**, 052326 (2004).
[8] S. D. Bartlett, A. C. Doherty, R. W. Spekkens, and H. M. Wiseman, *Phys. Rev. A* **73**, 022311 (2006).
[9] M. O. Terra Cunha, J. Dunningham, and V. Vedral, e-print quant-ph/0606149.
[10] J. I. Cirac, C. W. Gardiner, M. Naraschewski, and P. Zoller, *Phys. Rev. A* **54**, R3714 (1996).
[11] M. Naraschewski, H. Wallis, A. Schenzle, J. I. Cirac, and P. Zoller, *Phys. Rev. A* **54**, 2185 (1996).
[12] C. W. Gardiner, *Phys. Rev. A* **56**, 1414 (1997).
[13] A. J. Leggett, *Rev. Mod. Phys.* **73**, 307 (2001).
[14] F. Verstraete and J. I. Cirac, *Phys. Rev. Lett.* **91**, 010404

- (2003).
- [15] H. M. Wiseman and J. A. Vaccaro, *Phys. Rev. Lett.* **91**, 097902 (2003).
- [16] E. P. Wigner, in *Quantum Theory and Measurement*, edited by J. Wheeler and W. Zurek (Princeton University Press, Princeton, NJ, 1983), pp. 260–314.
- [17] P. Bertet, S. Osnaghi, A. Rauschenbeutel, G. Nogues, A. Auffeves, M. Brune, J. M. Raimond, and S. Haroche, *Nature (London)* **411**, 166 (2001).
- [18] N. H. Bonadeo, J. Erland, D. Gammon, D. Park, D. S. Katzer, and D. G. Steel, *Science* **282**, 1473 (1998).
- [19] Y. Nakamura, Yu. A. Pashkin, and J. S. Tsai, *Nature (London)* **398**, 786 (1999).
- [20] H. Lee, P. Kok, and J. P. Dowling, *J. Mod. Opt.* **49**, 2325 (2002).
- [21] D. J. Heinzen, R. Wynar, P. D. Drummond, and K. V. Kheruntsyan, *Phys. Rev. Lett.* **84**, 5029 (2000).
- [22] J. Calsamiglia, M. Mackie, and K.-A. Souminen, *Phys. Rev. Lett.* **87**, 160403 (2001).
- [23] E. A. Donley, N. R. Claussen, S. T. Thompson, and C. E. Weiman, *Nature (London)* **417**, 529 (2002).
- [24] J. A. Dunningham and K. Burnett, *Phys. Rev. Lett.* **82**, 3729 (1999).
- [25] S. D. Bartlett, T. Rudolph, and R. W. Spekkens, *Int. J. Quantum Inf.* **4**, 17 (2006).
- [26] F. Dalfovo, S. Giorgini, L. P. Pitaevskii, and S. Stringari, *Rev. Mod. Phys.* **71**, 463 (1999).
- [27] M. R. Andrews, C. G. Townsend, H.-J. Miesner, D. S. Durfee, D. M. Kurn, and W. Ketterle, *Science* **275**, 637 (1997).
- [28] J. Javanainen and S. M. Yoo, *Phys. Rev. Lett.* **76**, 161 (1996).
- [29] K. Mølmer, *Phys. Rev. A* **55**, 3195 (1997); *J. Mod. Opt.* **44**, 1937 (1997).
- [30] B. C. Sanders, S. D. Bartlett, T. Rudolph, and P. L. Knight, *Phys. Rev. A* **68**, 042329 (2003).
- [31] H. Cable, P. L. Knight, and T. Rudolph, *Phys. Rev. A* **71**, 042107 (2005).
- [32] J. F. Corney and P. D. Drummond, *Phys. Rev. Lett.* **93**, 260401 (2004).
- [33] K. Nemoto and S. L. Braunstein, *Phys. Rev. A* **68**, 042326 (2003).
- [34] C. A. Regal, C. Ticknor, J. L. Bohn, and D. S. Jin, *Nature (London)* **424**, 47 (2003).
- [35] J. Javanainen, M. Kostrun, Y. Zheng, A. Carmichael, U. Shrestha, P. Meinel, M. Mackie, O. Dannenberg, and K.-A. Souminen, *Phys. Rev. Lett.* **92**, 200402 (2004).
- [36] S. B. Bravyi and A. Yu. Kitaev, *Ann. Phys. (N.Y.)* **298**, 210 (2002).
- [37] K. E. Cahill and R. J. Glauber, *Phys. Rev. A* **59**, 1538 (1999).
- [38] M. Grigorescu and W. E. Baylis, *Phys. Rev. B* **66**, 014530 (2002).
- [39] T. Tyc, B. Hamilton, B. C. Sanders, and W. D. Oliver, e-print quant-ph/0507167.
- [40] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, Cambridge, England, 2000).
- [41] P. Kok and S. L. Braunstein, *Phys. Rev. A* **61**, 042304 (2000).
- [42] G. Modugno, G. Ferrari, G. Roati, R. J. Brecha, A. Simoni, and M. Inguscio, *Science* **294**, 1320 (2001).
- [43] G. Modugno, M. Modugno, F. Riboli, G. Roati, and M. Inguscio, *Phys. Rev. Lett.* **89**, 190404 (2002).
- [44] G. Roati, F. Riboli, G. Modugno, and M. Inguscio, *Phys. Rev. Lett.* **89**, 150403 (2002).
- [45] Z. Hadzibabic, C. A. Stan, K. Dieckmann, S. Gupta, M. W. Zwierlein, A. Görlitz, and W. Ketterle, *Phys. Rev. Lett.* **88**, 160401 (2002).
- [46] C. Silber, S. Günther, C. Marzok, B. Deh, Ph. W. Courteille, and C. Zimmermann, *Phys. Rev. Lett.* **95**, 170408 (2005).
- [47] A. Simoni, F. Ferlaino, G. Roati, G. Modugno, and M. Inguscio, *Phys. Rev. Lett.* **90**, 163202 (2003).
- [48] M. Köhl, H. Moritz, T. Stöferle, K. Günter, and T. Esslinger, *Phys. Rev. Lett.* **94**, 080403 (2005).
- [49] P. Treutlein, T. Steinmetz, Y. Colombe, B. Lev, P. Hommelhoff, J. Reichel, M. Greiner, O. Mandel, A. Widera, T. Rom, I. Bloch, and T. W. Hänsch, e-print quant-ph/0605163.
- [50] C. W. J. Beenakker, *Quantum Computer, Algorithms and Chaos*, in Proceedings of the International School of Physics “Enrico Fermi”, Vol. 162, edited by G. Casati, D. L. Shepelyansky, P. Zoller, and Benenti (IUS Press, Amsterdam, 2006); e-print cond-mat/0508488.
- [51] D. Kershaw and C. H. Woo, *Phys. Rev. Lett.* **33**, 918 (1974).