Spectroscopic Insensitivity to Cold Collisions in a Two-State Mixture of Fermions

Martin W. Zwierlein, Zoran Hadzibabic, Subhadeep Gupta, and Wolfgang Ketterle

Department of Physics, MIT-Harvard Center for Ultracold Atoms, and Research Laboratory of Electronics, MIT, Cambridge, Massachusetts 02139, USA

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We have experimentally demonstrated the absence of spectroscopic resonance shifts in a mixture of two interacting Fermi gases. This result is linked to observations in an ultracold gas of thermal bosons. There, the measured resonance shift due to interstate collisions is independent of the coherence in the system, and twice that expected from the equilibrium energy splitting between the two internal states in a fully decohered cloud. We give a simple theoretical explanation of these observations, which elucidates the effect of coherent radiation on an incoherent mixture of atoms.

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The coherence properties of light and matter are intimately connected with the quantum statistics of the constituent particles. One quantitative measure of the coherence in a system is the two-particle correlation function at zero distance, $g^{(2)}$, which measures the probability that two particles are simultaneously detected. Intensity fluctuations in the incoherent light emitted by a light bulb lead to photon "bunching," making this probability twice higher than in the coherent light of a laser. Identical fermions on the other hand exhibit "antibunching," making such a probability zero.

Interactions in ultracold atomic gases crucially depend on the value of $g^{(2)}$ [1]. The reason is that s-wave scattering relies on particles overlapping in real space. The interaction energy in a many-body system is determined by coherent collisions, for which the outgoing and the incoming two-particle states are identical. Under this constraint, the two colliding particles can at most do two things—either preserve their momenta, or exchange them. We can thus distinguish four cases: (i) Two identical bosons in a thermal gas can collide in both ways, corresponding to $g^{(2)} = 2$. (ii) Two atoms in a Bose-Einstein condensate (BEC) have the same momenta and cannot undergo the exchange interaction. Here $g^{(2)} = 1$. (iii) Two distinguishable particles, fermions or bosons, also cannot exchange their momenta because that would make the outgoing state different from the incoming one. Again, $g^{(2)} = 1$. (iv) Two identical fermions cannot collide at all, so $g^{(2)} = 0$. In all cases, the mean-field energy of a particle with mass m is given by $g^{(2)}(4\pi\hbar^2/m)an$, where a is the s-wave scattering length, and n is the density of atoms it interacts with.

Mean field energies and therefore $g^{(2)}$ can be measured spectroscopically. In experiments on ultracold hydrogen, mean-field shifts of the $1S$-$2S$ two-photon transition were used to prove the existence of a BEC [2]. However, quantitative interpretation of the shifts led to a vivid theoretical discussion about the coherence related "factors of 2" [3–6]. More recently, Harber *et al.* performed Ramsey spectroscopy in a two-component, thermal gas of ^{87}Rb bosons to measure $g^{(2)}$ in the interstate collisional shift [7]. Their measurements yielded $g^{(2)} = 2$, independent of the degree of coherence between the two states. The spectroscopic results thus seemed to correspond to the case of all particles being in an identical coherent superposition of the two internal states, even though the binary mixture was partially decohered and should have had a mean-field energy corresponding to $1 < g^{(2)} < 2$. The authors commented on this mystery [8]: "it is a pleasure to note that a two-level system can still yield surprises, 75 years after the advent of quantum mechanics." The mystery can be formally resolved using a quantum Boltzmann equation [9–13].

Here, we experimentally address the relation between coherence and spectroscopic measurements in a binary mixture of ultracold *fermions*. We demonstrate that shifts of spectroscopic lines are absent even in a fully decohered binary mixture, in which the particles are distinguishable, and the many-body mean-field energy in the system has developed. We theoretically show that this is a direct consequence of the coherent nature of the radiofrequency (rf) *excitation*, which, in general, leads to a final state with $g^{(2)}$ different from the initial state.

Our calculation intuitively explains both our results for fermions, and the results for bosons of Ref. [7].

In a recent paper [14], we demonstrated the absence of mean-field "clock shifts" in a coherent two-state superposition of ^{6}Li fermions. In this case, rf spectroscopy was performed on a gas prepared purely in one internal state. Since an rf pulse acts as a rotation in the two-state Hilbert space, all the atoms stayed in an identical (superposition) state and could not interact. As long as the fermionic atoms were indistinguishable, $g^{(2)} = 0$, and the resonance was thus found to be unperturbed at $\nu_0 = (E_{12}/h)$, where E_{12} is the energy difference between the internal states $|1\rangle$ and $|2\rangle$.

However, once decoherence sets in, for example due to inhomogeneous magnetic fields across the cloud, the spatial overlap between atoms in different states grows and mean-field energy density builds up:

$$\mathcal{E}_{\rm int}(\mathbf{r}) = g^{(2)} V_{12} n_1(\mathbf{r})\, n_2(\mathbf{r}), \qquad V_{12} = \frac{4\pi\hbar^2}{m} a_{12}, \quad (1)$$

where n_1 and n_2 are the local densities of particles in states $|1\rangle$ and $|2\rangle$, and a_{12} is the interstate s-wave scattering length. Here decoherence means that off-diagonal matrix elements of the density matrix have vanished locally. As a result, everywhere in the sample, atoms are no longer in one pure state, but occupy two orthogonal states, and s-wave collisions are no longer suppressed by the Pauli principle. In a fully decohered cloud, we have a binary mixture of two distinct species of atoms, with a mean-field energy density $\mathcal{E}_{\rm int} = V_{12} n_1 n_2$. This interaction changes the equilibrium energy level of atoms in state $|1\rangle$ ($|2\rangle$) according to $\delta\mu_{1,2} = V_{12} n_{2,1}$. The difference in equilibrium mean-field energy of the two states is then

$$\Delta E_{\rm int} = \delta\mu_2 - \delta\mu_1 = V_{12}(n_1 - n_2). \quad (2)$$

This suggests [7,8,14] that in a decohering sample, the resonant frequency for population transfer between the two states gradually changes from $\nu_{12} = \nu_0$ to $\nu_{12} = \nu_0 + \frac{1}{h}\Delta E_{\rm int}$. Here, we show both experimentally and theoretically that this conclusion is wrong, and that the spectroscopic resonance frequency ν_{12} is always the unperturbed frequency ν_0.

Our experimental setup was described in [14,15]. About 10^7 fermionic ^{6}Li atoms were confined in an optical dipole trap at a temperature of 35 μK. The two-level system under consideration is formed by the two lowest ground state hyperfine levels, $|1\rangle$ and $|2\rangle$, corresponding to $|F, m_F\rangle = |1/2, 1/2\rangle$ and $|1/2, -1/2\rangle$ in the low field basis, respectively. A dc magnetic field of $B = 320$ G was applied to the sample in order to tune the interstate scattering length a_{12} to a large value of $\sim -300 a_0$, where a_0 is the Bohr radius [14].

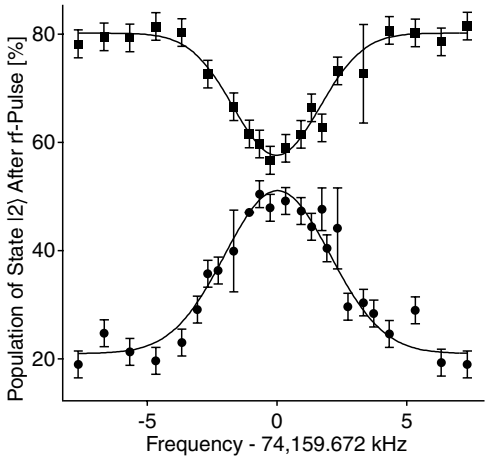

FIG. 1. Absence of mean-field shift of an rf transition in a binary Fermi system. The resonance curves were measured for fully decohered (80/20)% two-state mixtures of fermions. The measured frequency difference between the two lines is (34 ± 146) Hz, even though Eq. (2) would predict a splitting of 20 kHz.

We created a superposition of atoms in states $|1\rangle$ and $|2\rangle$ using a nonadiabatic rf sweep around the energy splitting of 74 MHz. As the sample decohered, efficient evaporative cooling set in, confirming a large elastic scattering length. After 1 sec, we were left with a fully decohered mixture at a mean density $n = 5 \times 10^{13}$ cm^{-3}. The rate of the rf sweep was adjusted so that after decoherence and cooling, 80% of the atoms were in state $|2\rangle$. The mean-field interaction should thus have increased the energy splitting of the two levels by $h\delta\nu = \delta\mu_2 - \delta\mu_1 = V_{12}(n_1 - n_2) \approx h \times 10$ kHz. Our experiments involving a third state [14] have confirmed the presence of such energy shifts, and prove that full decoherence has been reached.

Rabi spectroscopy in the interacting binary mixture was performed by applying 200 μs rf pulses of different frequencies, and recording the final populations in the two states by simultaneous absorption imaging (Fig. 1). In order to eliminate the systematic uncertainty in the value of ν_0, we performed a second experiment with the population ratios of states $|1\rangle$ and $|2\rangle$ reversed. According to Eq. (2), one would expect an opposite shift of the resonance.

Within our precision, no interaction shift of the resonance frequency was observed. Comparing the expected difference in mean-field shifts for the two experiments, $2\delta\nu = 20$ kHz, with the measured line separation of (34 ± 146) Hz, we arrive at an apparent value for $g^{(2)} = 0.002(7)$. This demonstrates the universal absence of a resonance shift in a very cold two-level Fermi gas, independent of the coherence in the system.

Evidently, rf spectroscopy does not measure the expected difference in thermodynamic chemical potentials for the two states. Experiments with thermal bosons have posed a similar puzzle [7]. Here we explain that this is a

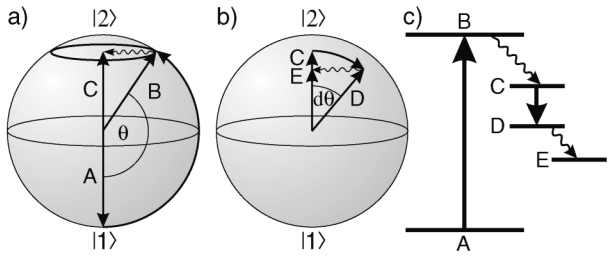

FIG. 2. Bloch sphere representation of rf transitions. (a) An rf pulse rotates a pure state A into B. The superposition state decoheres into a "ring" distribution, represented by its average, C. (b) A second rf pulse transforms the fully decohered state C into a partially coherent state D. The final state E is reached only after further decoherence. (c) Transfers $A \rightarrow B$ and $C \rightarrow D$ are coherent and reversible. $B \rightarrow C$ and $D \rightarrow E$ are irreversible.

direct consequence of the coherent nature of the rf excitation.

In Fig. 2, the average properties of the many-body state at a specific point $\mathbf{r}$ in the trap are described by the three coordinates of the local spin-1/2 Bloch vector $\mathbf{m}(\mathbf{r}) = m_z(\mathbf{r})\hat{\mathbf{e}}_z + \mathbf{m}_\perp(\mathbf{r})$. In the following, we omit the label $\mathbf{r}$. $m_z = [(n_2 - n_1)/2]$ represents the population difference in the two states, whereas the transverse component $\mathbf{m}_\perp$ is a measure of the coherence in the system. The length of the Bloch vector measures the purity of the average state and hence the entropy of the system. Fully decohered statistical mixtures do not have off-diagonal matrix elements of the density matrix and are represented by vectors with $\mathbf{m}_\perp = 0$, with state A being the special case of a pure state. In Fig. 2(a), state B is created by applying an rf pulse on a pure sample A. In this case, there is no interaction energy in the system during the rf pulse, and no frequency shift is expected [14]. State C is formed through subsequent decoherence of state B. States B and C have the same number of particles in $|1\rangle$ and $|2\rangle$, but in C the mean-field has fully developed.

Our experiment is performed on a C-like state [Fig. 2(b)]. Here we explain why Eq. (2) still does not give the correct resonance frequency for an infinitesimal transfer of atoms between $|1\rangle$ and $|2\rangle$. The key point is that even though the sample is fully decohered, the applied rf pulse reintroduces coherence into the system. According to Eq. (6) below, this will change the value of $g^{(2)}$. Let us consider two fully decohered states, C and E. Equation (2) correctly gives the energy of the transformation $C \rightarrow E$. However, these two states have different entropies, as indicated by Bloch vectors of different lengths. An rf pulse is a unitary transformation of the system, and must preserve entropy. The true effect of the rf pulse is thus to change the relative populations of $|1\rangle$ and $|2\rangle$ by tilting the Bloch vector away from the z axis, into state D. It is the energy of *this* transformation, $C \rightarrow D$, that needs to be calculated in order to find the correct resonant rf frequency.

In the case of fermions with short-range (delta function) interactions, we can prove very generally that the resonance frequency will always be ν_0, by showing that the interaction Hamiltonian is invariant under rotations on the Bloch sphere. The interstate s-wave interaction at point $\mathbf{r}$ is described by the second-quantized Hamiltonian density

$$H_{\rm int}(\mathbf{r}) = V_{12}\psi_1^\dagger(\mathbf{r})\psi_2^\dagger(\mathbf{r})\psi_2(\mathbf{r})\psi_1(\mathbf{r}). \tag{3}$$

Under a general rotation, described by polar angles θ, ϕ, the field operators $\psi_{1,2}^\dagger$ transform according to:

$$\begin{aligned}\psi_{1\theta,\phi}^\dagger &= \cos\frac{\theta}{2}e^{-i\phi/2}\psi_1^\dagger + \sin\frac{\theta}{2}e^{i\phi/2}\psi_2^\dagger,\\ \psi_{2\theta,\phi}^\dagger &= -\sin\frac{\theta}{2}e^{-i\phi/2}\psi_1^\dagger + \cos\frac{\theta}{2}e^{i\phi/2}\psi_2^\dagger.\end{aligned} \tag{4}$$

Using the standard fermionic anticommutation relations ($\psi_1\psi_2 = -\psi_2\psi_1$, $\psi_1\psi_1 = 0$, etc.), it is easy to show that

$$H_{\rm int}^{\theta,\phi} = V_{12}\psi_{1\theta,\phi}^\dagger\psi_{2\theta,\phi}^\dagger\psi_{2\theta,\phi}\psi_{1\theta,\phi} = H_{\rm int}. \tag{5}$$

We therefore see that an rf-induced rotation on the Bloch sphere commutes with the interaction Hamiltonian, and hence does not change the energy of the many-body state. It is then obvious that the resonant frequency will always be ν_0, independent of the coherence of the system.

We now present a more general calculation of the mean-field frequency shifts, which holds for both fermions and bosons. To reduce complexity and concentrate on the only controversial case of interstate interactions, we consider a fictitious boson with no intrastate interactions ($a_{11} = a_{22} = 0$). The (local) mean-field expectation value of the Hamiltonian density in Eq. (3) is [16]

$$\begin{aligned}\mathcal{E}_{\rm int}(\mathbf{r}) &= \langle H_{\rm int}\rangle = V_{12}(n_1 n_2 + \epsilon n_{12}n_{21})\\ &\Rightarrow g^{(2)} = 1 + \epsilon\frac{n_{12}n_{21}}{n_1 n_2},\end{aligned} \tag{6}$$

where $n_1 = \langle\psi_1^\dagger\psi_1\rangle$ and $n_2 = \langle\psi_2^\dagger\psi_2\rangle$ are the local densities in the two states, we have introduced "coherences" $n_{12} = \langle\psi_1^\dagger\psi_2\rangle$ and $n_{21} = \langle\psi_2^\dagger\psi_1\rangle$, and $\epsilon = \pm 1$ for bosons/fermions. In a fully coherent sample $n_{12}n_{21} = n_1 n_2$ and $g^{(2)} = 1 + \epsilon$. As decoherence sets in, $g^{(2)}$ increases (decreases) from 0 (2) to 1 for fermions (bosons). For the most general case of a partially decohered sample, we can rewrite Eq. (6) in terms of the (local) Bloch vector, using $n_{1,2} = \frac{n}{2} \mp m_z$, $n_{12} = m_x + i\,m_y = n_{21}^*$, and $n_{12}n_{21} = m_x^2 + m_y^2 = m_\perp^2$, where n is the total particle density. This gives

$$\mathcal{E}_{\rm int} = V_{12}\frac{n^2}{4} + \epsilon V_{12}|\mathbf{m}|^2 - (1+\epsilon)V_{12}m_z^2. \tag{7}$$

Two samples with the same numbers of atoms in states $|1\rangle$ and $|2\rangle$, but different levels of coherence, have the same m_z, but different $|\mathbf{m}_\perp|$ [e.g., states D and E in Fig. 2(b)]. Again we see that two such samples indeed have different interaction energies.

Now, let us evaluate the effect of coherence on the resonant rf frequency. A coherent rf excitation preserves entropy ($|\mathbf{m}| = \text{const}$), and the total density n. In an infinitesimal tilt of the Bloch vector, the density of atoms transferred from $|1\rangle$ to $|2\rangle$ is $dn_2 = -dn_1 = dm_z$. Therefore, the change of interaction energy per transferred particle, and thus the shift in the resonant frequency $\Delta\nu$, comes out to be

$$\Delta\nu = \frac{1}{h}\frac{\partial\mathcal{E}_{\rm int}}{\partial m_z}\bigg|_{n,|m|} = \frac{1}{h}(1+\epsilon)V_{12}\,(n_1 - n_2). \tag{8}$$

In analogy with a spinning top which precesses in the gravitational field, the resonant frequency for an infinitesimal tilt of the Bloch vector is also equal to the frequency of its free precession. In the traditional language of atomic physics, this analogy just reiterates

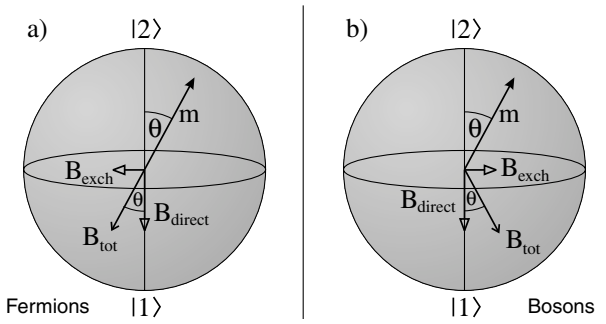

FIG. 3. Mean-field represented as effective magnetic field. (a) Fermions: The exchange and direct interaction add up to form a magnetic field aligned with the average spin ($V_{12} < 0$ in the drawing). The net torque vanishes and the Bloch vector $\mathbf{m}$ precesses at the unperturbed frequency ν_0. (b) Bosons: The exchange interaction has opposite sign to that in fermions. It exerts a torque on the average spin equal to the torque induced by the direct interaction, as can be seen by comparing the two cross products with $\mathbf{m}$. The Bloch vector thus precesses at ν_0 plus twice the frequency shift due to direct interaction.

that Rabi [14] and Ramsey [7] spectroscopy measure the same characteristic frequency of the system. The striking result is that in contrast to the interaction energy [Eqs. (6) and (7)], the precession of the Bloch vector, or equivalently the rf frequency shift [Eq. (8)], *does not* depend on the level of coherence in the sample. Remarkably, the final state may have a value of $g^{(2)}$ different from the initial state, such that the energy difference per transferred particle is *independent* of the initial $g^{(2)}$. Equation (8) explains both our measurements with fermions, and the experiment with thermal bosons of Ref. [7].

In order to further elucidate the role of coherences in the precession of the Bloch vector, we employ the interpretation of the mean-field energy as the interaction of the average spin with an effective magnetic field [12,13]. Using Eq. (7), we obtain $\mathcal{E}_{\rm int} = \text{const} - \frac{1}{2}\mathbf{B}_{\rm eff} \cdot \mathbf{m}$ [17] with

$$\mathbf{B}_{\rm eff} = 2V_{12}(m_z\hat{\mathbf{e}}_z - \epsilon\mathbf{m}_\perp). \tag{9}$$

In this picture, the precession of the spin due to interactions is driven by the torque $\mathbf{B}_{\rm eff} \times \mathbf{m}$. The magnetic field along the z axis is induced by the direct interaction, and has the same sign for fermions and bosons (Fig. 3). The transverse magnetic field comes from the exchange interaction, and has opposite signs for fermions and bosons. For fermions, $\mathbf{B}_{\rm eff}$ is parallel to $\mathbf{m}$ [Eq. (9)] and hence does not cause any precession. Equivalently, the direct and exchange interaction exert torques equal and opposite to each other. For bosons, the two contributions add up to yield exactly twice the precession frequency given by the direct interaction alone. During decoherence, the exerted torque shrinks in proportion to the decaying transverse spin. Therefore, the precession frequency remains constant, no matter how small the coherences are.

In conclusion, we have demonstrated the absence of the mean-field shift of rf transitions in a fully decohered, interacting binary mixture of fermions. This was explained by proving the invariance of the interaction energy under coherent Hilbert space rotations. This result is relevant for the potential use of a fermionic atom supplying the frequency standard in an atomic or optical clock, since it implies a robust elimination of the systematic errors due to density dependent frequency shifts. Previously, the absence of such clock shifts was explained by the absence of mean-field energy in a purely coherent superposition state [14]. Now we have shown that there is no spectroscopic shift even after decoherence has led to measurable mean-field energies. Further, we have presented a simple theoretical framework for calculating the precession frequency of the Bloch vector which describes an arbitrary spin state of either fermions or bosons. This resolves "the mystery of the Ramsey fringe that did not chirp" [8] with a simple and intuitive picture.

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