Coherent population oscillation from a Fermi atom-molecule dark state

Andrew Robertson, Lei Jiang, Han Pu, Weiping Zhang, and Hong Y. Ling

Department of Physics and Astronomy, Rowan University, Glassboro, New Jersey, 08028-1700, USA

Department of Physics and Astronomy, and Rice Quantum Institute, Rice University, Houston, TX 77251-1892, USA

Key Laboratory of Optical and Magnetic Resonance Spectroscopy (Ministry of Education), Department of Physics, East China Normal University, Shanghai 200062, P. R. China

(Dated: February 2, 2008)

We show that a robust macroscopic atom-molecule dark state can exist in fermionic systems, which represents a coherent superposition between the ground molecular BEC and the atomic BCS paired state. We take advantage of the tunability offered by external laser fields, and explore this superposition for demonstrating coherent oscillations between ground molecules and atom pairs. We interpret the oscillation frequencies in terms of the collective excitations of the dark state.

PACS numbers: 03.75.Mn, 05.30.Jp, 32.80.Qk

Association of ultracold atom pairs into diatomic molecules via Feshbach resonance or photoassociation, has made it possible to create coherent superpositions between atomic and molecular species at macroscopic level. This ability is the key to applications that employ the principle of the double pulse Ramsey interferometer for observing coherent population oscillations between atoms and molecules. A particular kind of state, the atom-molecule dark state, has been theoretically proposed and experimentally observed, where population is trapped in a superposition between atom pairs and deeply bound molecules in the electronic ground state. Destructive interference leads to the vanishing population in the excited molecular level. Such a state is the generalization of the usual atomic dark state that lies at the heart of many exciting applications, including electromagnetically induced transparency, slow light propagation and precision spectroscopy. So far, the macroscopic atom-molecule dark state has only been studied in bosonic systems. The purpose of this paper is to show that, under proper conditions, an atom-molecule dark state also exists in fermionic systems, but with quite distinct properties compared with its bosonic counterpart.

To be specific, we consider a homogeneous atom-molecule system where an excited molecular level $m$ is coupled both to a ground molecular level $g$ (bound-bound coupling) by a coherent laser field, and to two free atomic states of equal population labeled as $|\uparrow\rangle$ and $|\downarrow\rangle$ (bound-free coupling) via, for example, a photoassociation laser field. At zero temperature, bosonic molecules all condense to the zero-momentum state, whereas fermionic atoms are of multi-momentum modes in nature due to the Pauli principle, and are thus described by momentum continua of different internal states. This difference has two important ramifications.

The first one is related to the formation of the dark state. As is known, two necessary ingredients for creating a macroscopic atom-molecule dark state are the coherence between its components and the generalized two-photon resonance which, unlike in the linear atomic model, becomes explicitly dependent on the atomic momentum. For bosons at zero temperature, since they all occupy the same zero-momentum mode, properly tuning the laser frequencies can make all the bosons satisfy the two-photon resonance simultaneously. However, for fermions, because of the existence of the fermion momentum sea, the same technique can only render a limited number of atoms with the “right” momentum to satisfy the two-photon resonance. Hence a macroscopic dark state involving all the particles in the system does not seem to be possible for fermions. This difficulty can be circumvented when the attractive interaction between atoms of opposite spins results in a fermionic superfluid state that can be regarded as a condensate of atomic Cooper pairs. As we shall show below, such a fermionic superfluid, together with the ground molecule condensate, can now form a macroscopic dark state under the two-photon resonance condition.

The second ramification of the momentum continuum is related to the collective excitation of the dark state. The excitation spectrum of the fermionic system is far more difficult to analyze than its bosonic counterpart. The zero-temperature spectrum of the bosonic system is discrete. In contrast, the spectrum of the fermionic system is made up of both a discrete and a continuous part, and hence can be regarded as the nonlinear analog of the Fano-Anderson type of models in linear atomic and condensed matter systems. As we demonstrate later, this analogy significantly simplifies our understanding of the excitation spectrum while at the same time enables us to gain profound insights into the dynamical properties of the fermionic dark state.

Let us begin with the mean-field Hamiltonian...
ten in the frame rotating at the laser frequency:

$$\hat{H} = \sum_{k, \sigma} \epsilon_k \hat{a}_{k, \sigma}^\dagger \hat{a}_{k, \sigma} + \nu_0 \hat{b}_{m}^\dagger \hat{b}_{m} + (\delta_0 + \nu_0) \hat{b}_{g}^\dagger \hat{b}_{g}$$

$$- \sum_k \varphi_k \left( \Delta \hat{a}_{k, \uparrow}^\dagger \hat{a}_{-k, \downarrow}^\dagger + h.c. \right) + \frac{\Omega_0}{2} \left( \hat{b}_{g}^\dagger \hat{b}_{g} + h.c. \right) + \frac{1}{\sqrt{V}} \sum_k g \varphi_k \left( \hat{b}_{m} \hat{a}_{+k, \uparrow}^\dagger \hat{a}_{-k, \downarrow}^\dagger + h.c. \right),$$

(1)

where $\hat{a}_{k, \sigma}$ is the annihilation operator for an atom of spin $\sigma (= \uparrow$ or $\downarrow$), having momentum $\hbar \mathbf{k}$ and kinetic energy $\epsilon_k = \hbar^2 \mathbf{k}^2 / 2m$. $\hat{b}_{m,g}$ are the annihilation operators for a bosonic molecule in state $|m\rangle$ or $|g\rangle$. We have neglected the Hartree mean-field potential as it is usually weak for typical parameters. Here, $V$ is the system volume, $\delta_0$ and $\Omega_0$ ($\nu_0$ and $g$) are respectively the detuning and coupling strength of the bound-bound (bound-free) transition, $\varphi_k = \exp \left[ -k^2 / (2K^2) \right]$ is the regularization function providing momentum cutoff, and $\Delta = -U \sum_k \varphi_k \left( \hat{a}_{-k, \downarrow} \hat{a}_{k, \uparrow} \right) / V$ is the gap parameter. The collisional interaction potential between atoms of opposite spins and the atom-molecule coupling are given by $U \mathbf{k} = U \varphi_k \varphi_k \mathbf{e}^\dagger_k$ and $g \mathbf{k} = g \varphi_k$, respectively, where $U$ and $g$ are momentum independent. Evidently, Eq. (1) preserves the total atom number $N = 2 \langle \hat{b}_{m}^\dagger \hat{b}_{m} \rangle + \langle \hat{b}_{g}^\dagger \hat{b}_{g} \rangle + 2 \sum_k \langle \hat{a}_{k, \uparrow}^\dagger \hat{a}_{k, \downarrow} \rangle$.

The dynamics of the system is governed by the Heisenberg equations of motion for operators. By replacing boson operator $\hat{b}_{m,g}$ with the related c-number $\hat{c}_{m,g} = \langle \hat{b}_{m,g} \rangle / \sqrt{V}$ and fermi operator $\hat{a}_{k,\sigma}(t)$ with $u_k(t)$ and $v_k(t)$ through the Bogoliubov transformation

$$\begin{bmatrix} \hat{a}_{k, \uparrow}(t) \\ \hat{a}_{-k, \downarrow}(t) \end{bmatrix} = \begin{bmatrix} u_k(t) & v_k(t) \\ -v_k^*(t) & u_k(t) \end{bmatrix} \begin{bmatrix} \hat{a}_{k, \uparrow}^\dagger \\ \hat{a}_{-k, \downarrow}^\dagger \end{bmatrix},$$

(2)

with $|u_k(t)|^2 + |v_k(t)|^2 = 1$ and $\hat{a}_{k,\sigma}$ being fermi quasiparticle operators, we obtain the following equations

$$i \hbar \frac{d \hat{c}_{m}}{dt} = \nu_0 \hat{c}_m + \frac{\Omega_0}{2} \hat{c}_g - \frac{g}{\sqrt{V}} \Delta,$$

(3a)

$$i \hbar \frac{d \hat{c}_{g}}{dt} = (\delta_0 + \nu_0) \hat{c}_g + \frac{\Omega_0}{2} \hat{c}_m,$$

(3b)

$$i \hbar \frac{d u_k}{dt} = -\epsilon_k u_k + \varphi_k (g \hat{c}_m - \Delta^* \hat{c}_g),$$

(3c)

$$i \hbar \frac{d v_k}{dt} = \epsilon_k v_k + \varphi_k (g \hat{c}_m - \Delta) \hat{c}_g,$$

(3d)

$$\Delta(t) = -\frac{U}{V} \sum_k \varphi_k u_k(t) v_k(t),$$

(3e)

where we have assumed that the state of the system is the quasiparticle vacuum annihilated by $\hat{a}_{k,\sigma}$.

The stationary solutions to Eqs. (3) have the form:

$$c_{m,g} = c_{m,g}^0 e^{-2\mu t / \hbar}, \quad \Delta(t) = \Delta^s e^{-i2\mu t / \hbar},$$

$$u_k(t) = u_k^0 e^{iE_k t / \hbar} e^{i\mu t / \hbar}, \quad v_k(t) = v_k^0 e^{iE_k t / \hbar} e^{-i\mu t / \hbar},$$

where quantities with superscript $s$ are time-independent. Inserting this stationary ansatz into Eqs. (3) and searching for solutions with $c_{m,g}^0 = 0$, we find that such a dark-state solution indeed exists as long as the generalized two-photon resonance condition

$$\delta_0 + \nu_0 = 2\mu,$$

(4)

is satisfied. Such a solution is given by $|u_k^0|^2 = 1 - |v_k^0|^2 = (E_k + \epsilon_k - \mu) / 2E_k$, $E_k = \sqrt{(\epsilon_k - \mu)^2 + |\Delta^s|^2 \varphi_k^2}$, where $\mu$, $\Delta^s$, and $\epsilon_k$ are determined from the following equations, representing, respectively, (a) the destructive interference
condition leading to vanishing population in \( |m\rangle \)

\[
\frac{\Omega_0}{2} c_g^s = \frac{g}{U} \Delta^s, \tag{5}
\]

(b) the gap equation

\[
\frac{1}{U} = \frac{1}{2 \pi^2} \int_0^\infty \frac{E_k^2 k^2 dk}{2E_k}, \tag{6}
\]

and (c) the conservation of particle number

\[
\eta = \frac{N}{\sqrt{V}} = 2 |c_g^s|^2 + \frac{1}{2 \pi^2} \int_0^\infty \left( 1 - \frac{\epsilon_k - \mu}{E_k} \right) k^2 dk. \tag{7}
\]

Equation (3) in particular demonstrates the coherent nature of the dark state: for a normal atomic Fermi gas \((\Delta^s = 0)\) which does not possess phase coherence, such a state is impossible as Eq. (3) would imply vanishing population in the molecular level \( |g\rangle \) \((c_g^s = 0)\).

An example of the dark state solution obtained by solving Eqs. (5-7) self-consistently is shown in Fig. 1(a). To remove the ultraviolet divergence in the gap equation (6), we have followed the standard renormalization procedure to replace \( U \) by \( \Gamma U_0 \), where \( U_0 \) is the physical two-body atomic collisional strength. Here \( \Gamma = 1/(1+U_0 U_c^{-1}) \), and \( U_c^{-1} = -m K_c / (4\pi^3/2h^2) \). Further, by replacing \( g \) with \( \Gamma_0 \) while keeping the rest of parameters unchanged, we can easily show that our results become independent of \( K_c \). Figure 1(a) displays the ground molecular population of the dark state \(|c_g^s|^2\), the corresponding chemical potential \( \mu \), and the gap parameter \( \Delta^s \) as a function of the bound-bound coupling strength \( \Omega_0 \). In the limit \( \Omega_0/(g_0 \sqrt{\eta}) \rightarrow \infty \), we have \(|c_g^s|^2 \rightarrow 0 \) and all the population is in a pure atomic BCS state; while in the opposite limit of \( \Omega_0/(g_0 \sqrt{\eta}) \rightarrow 0 \), \(|c_g^s|^2 \rightarrow 0.5 \) and all the population are in the ground molecular state. Thus, in principle, we can adiabatically convert the BCS atom pairs into ground molecular BEC or vice versa by controlling the ratio \( \Omega_0/g_0 \sqrt{\eta} \) in the spirit of STIRAP [14]. Our use of STIRAP here is, however, for preparing a superposition which is a prerequisite for demonstrating coherent oscillations in fermionic systems [13, 16, 17, 18].

Starting from \( t = 0 \) with a pure atomic BCS state at a relatively large \( \Omega_0 \), we adiabatically decrease \( \Omega_0 \) to 4.1 \( E_F \) at \( t = t_s \) [indicated in Fig. 1(b)-(d)] while maintaining the two-photon resonance condition (4) through a proper chirping of the laser frequency [8]. At \( t = t_s \), a dark state, which is indicated by the vertical lines in Fig. 4 is then formed with about 14% of the atoms now converted to ground molecules. Next, immediately after \( t = t_s \), we suddenly change \( \Omega_0 \) from 4.1 to 4.6 \( E_F \) and then keep it fixed for later time, while fixing all other parameters at their respective values at \( t_s \). The dynamical response of the system is illustrated in Fig. 4(b)-(d), which display the ground molecular population as a function of time as obtained by solving Eqs. (3).

From the dynamical simulation, we see that the system follows the dark-state solution up to \( t = t_s \), after which, the sudden change of \( \Omega_0 \) induces oscillations in the population. Note that although the dark-state solution is not explicitly dependent upon the detunings \( \delta_0 \) and \( v_0 \), which must satisfy Eq. (4), the population dynamics for \( t > t_s \) does depend on their specific values. Several conclusions can be drawn from Fig. 1(b)-(d). First, the atom-molecule dark state is robust as, after a sudden “shake” at \( t_s \), the system oscillates around its steady state. Second, the population oscillation occurs between the ground molecular state \(|g\rangle \) and the atomic state, while the excited molecular population (not shown in the figures) remains negligible. Third, the oscillations are dominated by two frequencies whose values depend on the detunings as indicated by the corresponding Fourier spectra shown in the insets.

To better understand these oscillations and gain insight into the dark states, we calculate the collective mode frequencies by linearizing Eqs. (3) around the dark state solution. This procedure leads to a transcendental equation for the collective mode frequency \( \omega \)

\[
f(\omega) \equiv \det \left[ \frac{1}{U_{eff}(\omega)} - \int_0^\infty \frac{dk}{2\pi^2} \frac{E_k^2 \psi_k^2 (\epsilon_k - \mu)^2 + \omega (\epsilon_k - \mu)}{E_k (\omega^2 - 4E_k^2)} \right] = 0, \tag{8}
\]

where \( U_{eff}(\omega) = U + \omega g^2 |\omega (\omega + 2\mu - \nu_0) - |\Omega_0|^2/4|^{-1} \). Here, the integrals in the diagonal elements are automatically renormalized since \( U_{eff}(\omega) \) scales as \( \Gamma U_{eff}^0(\omega) \), where \( U_{eff}^0(\omega) = U_0 + \omega g_{0}^2/|\omega (\omega + 2\mu - \nu_0) - |\Omega_0|^2/4| \)

with \( \nu_0 = v_0 + \Gamma g_{0}^2/U_c \).

Before examining \( f(\omega) \) in detail, we first make a remark. As we have mentioned, our dark state reduces to a pure BCS state in the limit \( \Omega_0/g_0 \sqrt{\eta} \rightarrow \infty \). In this case, \( U_{eff} \rightarrow U_0 \), which is independent of \( \omega \). As is known [19], the collective excitation spectrum of a BCS state contains a continuous part and a discrete mode lying just below the continuum threshold at \( 2\Delta^s \). Due to the coupling between discrete (molecular) states and the continuum (atomic) states, the problem at hand bears much resemblance to the energy diagonalization of the Fano-Anderson type of Hamiltonians in linear atomic and
condensed matter systems [11]. In analogy to these problems, such discrete-continuum coupling may lead to drastic modifications to both parts of the excitation spectrum. Mathematically, this coupling gives rise to \( \omega \)-dependence in \( U_{eff} \) and introduces extra poles in \( f(\omega) \).

We now examine the spectrum by finding the roots of Eq. (5). Since \( f(\omega) \) is an even function of \( \omega \), we only concentrate on the positive-frequency branch. The function of \( f(\omega) \) is plotted in Fig. 2. The left panel [Fig. 2 (a)-(c)] shows the low-frequency part. Here, just as in the pure BCS model, one isolated mode lies not far below the continuum threshold. As the free-bound detuning becomes more negative, this mode decreases and shifts further away from continuum. In the right panel [Fig. 2 (d)-(f)], we show the high-frequency part. Here, the vertical lines are the poles determined by \( \omega = 2E_k \) at discrete momenta. Typically, a single root is trapped between two adjacent poles. These roots will form a continuum. This pattern of root distribution is, however, broken in the region indicated by the arrow, where two roots exist between two adjacent poles. In the continuous \( k \) limit, one of the two roots joins the continuum while the other one becomes part of the discrete spectrum. The two discrete modes (one shown in left and the other in right panel) are the ones that determine the dynamical population oscillation shown in Fig. 2 (b)-(d), while the contribution from the continuous part of the spectrum, due to the destructive interference, may lead to power-law decay of the oscillation at a longer time scale [13, 14].

In summary, we have shown that it is possible to construct a macroscopic atom-molecule dark state in a fermionic superfluid. The superfluidity of the fermionic atoms is a necessary ingredient for such a state. Therefore characteristics of the dark state may serve as a diagnostic tool for Fermi superfluids. Via direct dynamical simulation, we have shown that the dark state is quite robust. By perturbing the state, we are able to generate coherent oscillations reminiscent of the oscillating current across Josephson junctions. A remarkable feature here is that the population oscillation occurs between the ground molecules and the BCS atom pairs, while the excited molecular population remains highly suppressed. This has the obvious advantage of preserving the atom-molecule coherence for a time much longer than the excited molecular lifetime. Thus, this technique has the potential to increase the sensibility in interference-based high-precision measurements. In particular, the low frequency mode is directly related to the gap parameter \( \Delta_s \) and measurement of this frequency will allow us to gain insight into the atom-atom and atom-molecule interactions as they will strongly affect \( \Delta_s \).

This work is supported by the NSF (HYL, HP), ARO (HYL), the Welch and the Keck Foundations (HP), and by the National Natural Science Foundation of China under Grant No. 10474055 and No. 10588402, the National Basic Research Program of China (973 Program) under Grant No. 2006CB921104, the Science and Technology Commission of Shanghai Municipality under Grant No. 05PJ14038, No. 06JC14026 and No. 04DZ14009 (WZ).

†To whom correspondence should be addressed E-mail: ling@rowan.edu

[1] E. Tiesinga, B.J. Verhaar, and H.T.C. Stoof, Phys. Rev A 47, 4114 (1993); E. Timmermans, P. Tommasini, M. Hussein, and A. Kerman, Phys. Rep. 315, 199 (1999).
[2] H. R. Thorsheim, J. Weiner, and P. S. Julienne, Phys. Rev. Lett. 58, 2420 (1987).
[3] N. F. Ramsey, Molecular Beams, (Oxford University Press, New York 1985).
[4] E. A. Donley, N. R. Classen, S. T. Thompson, and C. E. Wieman, Nature (London) 417, 529 (2002).
[5] S. J. J. M. F. Kokkelmans and M. J. Holland, Phys. Rev. Lett. 89, 180401 (2002).
[6] M. Mackie, K.-A. Suominen, and J. Javanainen, Phys. Rev. Lett. 89 , 180403 (2002).
[7] M. Mackie, R. Kowalski, and J. Javanainen, Phys. Rev. Lett. 84, 3803 (2000); M. Mackie et al., Phys. Rev. A 70, 013614 (2004).
[8] H. Y. Ling, H. Pu, and B. Seaman, Phys. Rev. Lett. 93, 250403 (2004); H. Y. Ling, P. Maenner, and H. Pu, Phys.
Rev. A 72, 013608 (2005).

[9] K. Winkler et al., Phys. Rev. Lett. 95, 063202 (2005); R. Dumke et al., Phys. Rev. A 72, 041801(R) (2005); S. Moal et al., Phys. Rev. Lett. 96, 023203 (2006).

[10] See, for example, M. O. Scully and M. S. Zubairy, Quantum Optics (Cambridge University Press, Cambridge, 1997).

[11] U. Fano, Phys. Rev. 124, 1866 (1961); P. W. Anderson, Phys. Rev. 124, 41 (1961).

[12] P. G. de Gennes, Superconductivity of Metals and Alloys (Addison-Wesley Publishing Company, New York, 1989).

[13] S. J. J. M. F. Kokkelmans et al., Phys. Rev. A 65, 053617 (2002); J. Stajic et al., Phys. Rev. A 69, 063610 (2004).

[14] K. Bergmann, H. Theuer, and B. W. Shore, Rev. Mod. Phys. 70, 1003 (1998).

[15] E. A. Yuzbashyan, O. Tsypliyatev, and B. L. Altshuler, Phys. Rev. Lett. 96, 097005 (2006).

[16] A. V. Andreev, V. Gurarie, and L. Radzihovsky, Phys. Rev. Lett. 93, 130402 (2004).

[17] R. A. Barankov, L. S. Levitov, and B. Z. Spivak, Phys. Rev. Lett. 93, 160401 (2004); R. A. Barankov and L. S. Levitov, Phys. Rev. A 73, 033614 (2006).

[18] M. H. Szymanska, B. D. Simons, and K. Burnett, Phys. Rev. Lett. 94, 170402 (2005).

[19] A. F. Volkov and Sh. M. Kogan, Sov. Phys. JETP 38, 1018 (1974).