Spectroscopic Signatures of Nonequilibrium Pairing in Atomic Fermi Gases

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We determine the radio-frequency (rf) spectra for nonstationary states of a fermionic condensate produced by a rapid switch of the scattering length. The rf spectrum of the nonequilibrium state with constant BCS order parameter has two features in contrast with equilibrium where there is a single peak. The additional feature reflects the presence of excited pairs in the steady state. In the state characterized by periodically oscillating order parameter, the rf-absorption spectrum contains two sequences of peaks spaced by the frequency of oscillations. Satellite peaks appear due to a process where a rf photon in addition to breaking a pair emits or absorbs oscillation quanta.

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Cooper pairing in ultracold Fermi gases has been a major focus of research in the past few years. Remarkable experimental techniques such as sweeps across the Feshbach resonance [1,2], generation of collective modes [3] and vortex lattices [4], and radio-frequency (rf) spectroscopy [5,6] have been developed to probe the paired state. While it was crucial to establish for cold gases well-known signatures of fermionic pairing, of a key interest are regimes not easily accessible in superconductors, e.g., strong interactions in the vicinity of the Feshbach resonance and highly imbalanced mixtures.

One of the most interesting possibilities is to access the nonadiabatic coherent dynamics of fermionic condensates [7–15]. Driven out of equilibrium by a sudden change of the pairing strength on the BCS side of the Feshbach resonance, these systems acquire steady states with properties strikingly different from equilibrium ones. Three distinct nonstationary states have been predicted—a state where amplitude of the BCS order parameter \( \Delta(t) \) oscillates periodically [9,14], a state with a constant but reduced gap, and a gapless superfluid state [13–15]. Realization of a particular steady state is determined by the magnitude of change of the pairing strength. Most previous studies concentrated on the time evolution of the order parameter, while direct experimental manifestations of the nonadiabatic dynamics have not been sufficiently explored. The purpose of the present Letter is to address this issue.

Amongst existing experimental techniques the rf spectroscopy appears to have the greatest potential for distinguishing different dynamical states from equilibrium phases. This motivates us to study spectroscopic signatures of the dynamics of fermionic condensates. Our main findings are as follows. In contrast to the BCS ground-state spectrum which has a single peak at a frequency determined by the equilibrium gap, the rf spectrum of a non-equilibrium state with constant but finite \( \Delta_s \) displays two distinct peaks. The second peak reflects the fact that this nonequilibrium state is a superposition of an infinite number of excited stationary states of the condensate. Excited states contain a mixture of ground-state pairs and excited pairs—two-particle excitations of the condensate that conserve the total number of particles and Cooper pairs (see [16,17] and the discussion below).

The “ordinary” peak present already in the ground state is due to a process whereby a photon breaks a ground-state pair, while in the process responsible for the second peak it breaks an excited pair, [Fig. 1(a)]. It is interesting to note that in electronic superconductors excited pairs carry no charge or spin and are therefore difficult to detect.

For a steady state with periodically oscillating order parameter, we show that each of the peaks described above acquires equidistant satellite peaks; i.e., there are two series of equally spaced peaks in this state. The spacing between peaks in each series is equal to the frequency of oscillation \( \Omega \). Satellite peaks appear because a photon can gain optimal energy for breaking a ground-excited pair by emitting or absorbing several “deltons”—oscillation quanta of energy \( \Omega \), [Fig. 1(b)].

FIG. 1 (color online). (a) Nonstationary state of a fermionic condensate with time-independent order parameter \( \Delta_s \). (b) In the steady state where the order parameter \( \Delta(t) \) oscillates with frequency \( \Omega \), the photon can break a pair and emit or absorb several oscillation quanta.
In an atomic Fermi gas the pairing occurs between atoms in two hyperfine states \([1] \text{ and } [2]\). The frequency of external rf radiation can be tuned to induce transitions between one of these states, say \([2]\), to the third atomic state \([3]\). The rf spectrum corresponds to the rate of loss of atoms from \([2]\), i.e., \(I(\omega_{at}) = -dN_2/dt\), measured as a function of the radiation frequency \(\omega_{rf}\). In the normal state of atoms \([1]\) and \([2]\) the quantity \(I(\omega_{at})\) has a sharp peak at \(\omega_{at} = \omega_{a}\), the frequency of atomic transition between \([2]\) and \([3]\). In the paired ground state the peak shifts to a larger frequency since now an additional energy is required to break pairs \([5, 18]\).

We start with the Hamiltonian \(\hat{H} = \hat{H}_{12}^{BCS} + \hat{H}_3 + \hat{H}_T^{EM}\), where

\[
\hat{H}_{12}^{BCS} = \sum_{j, \alpha = 1, 2} e_j \hat{c}_j^\dagger \hat{c}_j \alpha - \frac{\lambda(t)}{\nu_F} \sum_{i,j} \hat{c}_{i j}^\dagger \hat{c}_{j i} \hat{c}_j \hat{c}_j \tag{1}
\]

is the BCS Hamiltonian describing pairing between states \([1]\) and \([2]\), \(\hat{c}_{j a}(\alpha = 1, 2)\) annihilate atoms in states \([1]\) and \([2]\), \(e_j\) are single-particle energy levels relative to the Fermi level of atoms \([1]\) and \([2]\), \(\lambda(t)\) and \(\nu_F\) are the dimensionless coupling and the density of states at the Fermi level, \(\hat{H}_3 = \sum_j e_j \hat{d}_j^\dagger \hat{d}_j\), where \(\hat{d}_j\) annihilate atoms in states \([3]\), represents noninteracting atoms in states \([3]\), and

\[
\hat{H}_T^{EM} = \frac{\omega}{2} \sum_j \left( \hat{c}_{j}^\dagger \hat{c}_j \hat{d}_j - \hat{d}_j^\dagger \hat{d}_j \right) + \hat{H}_T, \tag{2}
\]

accounts for the interaction of atoms \([2]\) and \([3]\) with the rf radiation field \([18]\) in the rotating wave approximation \([19]\). Here \(\omega = \omega_{at} - \omega_a\) is the detuning frequency. Since the size of the trap is much smaller than the photon wavelength, one can take the tunneling matrix to be diagonal, \(T_{ji} = T \delta_{ji}\).

We assume that the pairing strength has been switched from \(\lambda_i\) to \(\lambda_j\) and the rf radiation is turned on after the condensate has reached one of the steady states described above. The magnitude of the change in pairing strength is denoted by the parameter \(\beta\):

\[
\beta = \lambda_j^{-1} - \lambda_i^{-1}.
\]

Our task is to evaluate the current \(\langle \dot{N}_g \rangle = -d(N_g)/dt\). The wave function of the condensate in the steady state without the rf field is of the BCS form \(|\Psi(t)\rangle = \prod_j |u_j(t) \rangle |\hat{c}_j^\dagger |0\rangle\) \([9]\). Treating the tunneling Hamiltonian, \(\hat{H}_T\) in Eq. (2), as a perturbation, we obtain the current out of state \([2]\) to the lowest nonvanishing order in \(T_{ji}\)

\[
I = |T|^2 \int_{-\infty}^{\infty} d\omega \sum_{\omega_{e_j} \pm \delta\mu} \text{Re} [u_j(e_j - \omega - \omega_{\pm}) \bar{u}_j(\omega - e_j) e^{i\omega t}], \tag{3}
\]

where \(u_j(\omega)\) are Fourier components of \(u_j(t)\) and \(\delta\mu = \mu_3 - \mu_2\) is a difference between the corresponding chemical potentials for atoms in states \([3]\) and \([1]\), \([2]\). In recent experiments all states \([3]\) were initially unpopulated \([5, 6]\), which suggests that we set \(\delta\mu \approx -\Delta_F\). However, our model based on truncated BCS Hamiltonian (1) becomes invalid for that case since the so-called “off-diagonal” interaction terms between atoms in states \([1]\) and \([2]\) cannot be discarded \([20]\). To circumvent this problem in what follows we assume \(\delta\mu \ll E_F\).

Consider first the steady state with a constant order parameter \(\Delta_s\), that is realized for \(-\pi/2 \leq \beta \leq \pi/2\). The steady state wave function has been determined exactly in Ref. \([15]\]

\[
\begin{bmatrix} u_j \\ v_j \end{bmatrix} = \frac{\theta_j}{2} \begin{bmatrix} u_0^l \\ v_0^l \end{bmatrix} e^{i\xi_j} + \frac{\cos \theta_j}{2} \begin{bmatrix} \bar{v}_0^l \\ -\bar{u}_0^l \end{bmatrix} e^{-i\xi_j} e^{i\phi_j}, \tag{4}
\]

where \(\xi_j = (e_j^2 + \Delta_s^2)^{1/2}, \phi_j\) is the time-independent relative phase, and \(u_0^l = (\xi_j - e_j)/2\xi_j\) and \(v_0^l = (\xi_j + e_j)/2\xi_j\) are the Bogoliubov amplitudes in the BCS ground state with gap \(\Delta_s\). The distribution function \(\cos^2(\theta_j/2)\). (Fig. 3), and \(\Delta_s\) are known exactly in terms of the initial and final equilibrium BCS gaps \(\Delta_i\) and \(\Delta_f\) \([15]\). The first term in Eq. (4) is the wave function of a ground-state pair of energy \(-\xi_j\). The second term is the wave function of an excited pair with energy \(\xi_j\) \([16]\). Excited pairs are excitations of the condensate and should be contrasted to the single-particle excitations, which are created outside of the condensate. When the BCS wave function is projected onto the subspace of fixed particle number \([21]\), excited pairs conserve the total number of paired atoms, while quasi-particle excitations break Cooper pairs. In the Anderson pseudospin representation \([21]\), excited and ground state pairs correspond to a pseudospin, respectively, aligned parallel or antiparallel to its effective magnetic field. In this case \(\theta_j\) is the angle between the pseudospin and the field.

Using Eqs. (3) and (4), we derive the rate of loss in state \([2]\)

\[
\frac{I(\omega)}{2\pi|T|^2} = \frac{\Delta_s^2}{\omega^2} \left[ \sin^2 \left( \frac{\theta(\omega)}{2} \right) \partial(\omega - \omega_R^+) + \cos^2 \left( \frac{\theta(\omega)}{2} \right) \partial(\omega + \omega_R^+) \right], \tag{5}
\]

where \(\omega_R^+ = \sqrt{\delta\mu^2 + \Delta_s^2} \pm \delta\mu\) and \(\theta = (\omega^2 - \Delta_s^2)/2\omega\). The first term represents the contribution of ground-state pairs, \([\text{Fig. } 2(b)]\), corresponding to a process where a photon breaks a ground-state pair and creates an unpaired atom in state \([3]\), \([\text{Fig. } 1(a)]\). Energy balance yields \(\omega = e_j + \xi_j\). The first term is nonzero when \(\omega\) exceeds the threshold energy \(\omega_R^+\). In the ground state \(\theta(\omega) = \pi\) and only this term remains. The second term derives from excited pairs and corresponds to the process where a photon breaks an excited pair, \([\text{Fig. } 1(a)]\). The energy balance now implies \(\omega = e_j - \xi_j\), which is negative for all \(j\). We see that an additional peak appears at \(\omega \approx -\omega_R^+\),
\[
\text{FIG. 2 (color online). rf spectra (5) for a nonstationary state with a constant order parameter } \Delta_j \neq 0 \text{ produced by an abrupt change in the pairing strength, } \lambda_i \to \lambda_j \text{ for } \delta \mu = -0.75 \Delta_j; \text{ (a) The spectral weight for } \omega < 0 \text{ where the peak is due to processes where the photon breaks an excited pair. This peak is absent in the ground state, } \beta = 0; \text{ (b) The peak at } \omega > 0 \text{ is due to processes where an rf photon breaks a ground-state Cooper pair. A similar peak is present in the paired ground state. } \Delta_j \text{ is the equilibrium gap for the final coupling } \lambda_j. \]

[Fig. 2(a)]. The maximum in absorption is reached at } \omega = -\Delta_j. \text{ Its height is suppressed, since } \theta(\omega) \text{ can only deviate significantly from } \pi \text{ in a narrow window of width } \Delta_j \text{ around the Fermi energy, where there is a significant density of excited pairs. Finally, we note that when } \Delta_j \to 0 \text{ the two peaks merge at zero frequency; i.e., the rf spectrum of the gapless steady state is reversed to that of a normal state.} \n
Now let us turn to the regime of periodically oscillating order parameter, which occurs when } \beta > \pi/2 \text{ [9,14]. In this state } \Delta(t) \text{ is given by the Jacobi elliptic function dn with an amplitude comparable to } \Delta_j \text{ and a period of order } 2\pi/\Delta_j. \text{ We are to analytically determine the Bogoliubov amplitudes using the exact solution for the BCS dynamics [11,13], yielding}

\[
\begin{bmatrix}
\begin{array}{c}
u_j \\
\end{array}
\end{bmatrix} = \sum_{n=-\infty}^{\infty} \left( \frac{\sin \theta_j}{2} \right) \begin{bmatrix}
\begin{array}{c}
a_{jn} \\
b_{jn}
\end{array}
\end{bmatrix} e^{i(n\omega_j - n\Omega)t} + \cos \frac{\theta_j}{2} \begin{bmatrix}
\begin{array}{c}
\bar{b}_{jn} \\
\bar{a}_{jn}
\end{array}
\end{bmatrix} e^{-i(n\omega_j - n\Omega)t},
\end{align}
\]

(6)

\[
\text{FIG. 3 (color online). The probability } n(e) = \cos^2[\theta(e)/2] \text{ of having an excited pair [see the text below Eq. (5)] at energy } e \text{ in all steady states produced by a switch of the BCS coupling constant } \lambda_i \to \lambda_j \text{ [15]. Plots for three values of } \beta = 1/\lambda_i - 1/\lambda_j \text{ are shown. The presence of excited pairs leads to additional peaks in rf spectra shown in Figs. 2 and 4. In the ground state } n(e) = 0. \]

where } \Omega \text{ is the frequency of oscillations of } \Delta(t), \nu_j = \nu(e_j) \text{ is a function of single-particle energy, and } \theta_j \text{ has been discussed below Eq. (4). For brevity, the analytic expressions for } \Omega, a_{jn}, b_{jn}, \text{ and } \nu_j \text{ are omitted. We note, however, that } \nu_j \text{ plays the analogous role of excitation energy } \xi_j \text{ for the periodic regime. For example, } \nu_j \to \xi_j \text{ as we approach the regime of constant steady state gap, } \beta \to \pi/2. \text{ One can also show that } \nu(e) \text{ is a monotonic function of } |e|, \nu(e) \geq |e|, \nu(0) = \Omega/2, \text{ and } \nu(e) \to |e| \text{ for } \lambda_i \to 0 \text{ and for large } |e|.}

Comparison of the steady state wave functions (4) and (6) suggests the two terms in Eq. (6) may be interpreted as two orthogonally paired states for each level } j. \text{ These are the analog of ground-state and excited pairs and have energies } \pm \nu_j. \text{ In addition, these states contain } n \text{ quanta of the oscillating pairing field } \Delta(t) \text{ each carrying energy } \Omega. \text{ We will refer to these quanta as deltons. These are quanta of the amplitude mode of the pairing field and can be interpreted as Higgs bosons [22,23].}

Equations (3) and (6) determine the rf spectrum in the periodic regime

\[
\frac{I(\omega)}{2\pi|T|^2} = \sum_{n,\epsilon_j,\epsilon_j} \left\{ \sin^2 \frac{\theta_j}{2} |a_{jn}|^2 \delta(\omega - \nu_j - \epsilon_j - n\Omega) + \cos \frac{\theta_j}{2} |b_{jn}|^2 \delta(\omega + \nu_j - \epsilon_j - n\Omega) \right\},
\]

(7)

Here we dropped oscillatory terms assuming they average to zero on the time scale of the measurement. Expression (7) describes two series of equidistant peaks, (Fig. 4), corresponding to the processes where an rf photon breaks one of the two paired states on level } j \text{ and emits or absorbs } n \text{ deltons, [Fig. 1(b)]. The energy balance reads } \omega = \epsilon_j \pm \nu_j + n\Omega. \text{ The first series of peaks is described by the first term in Eq. (7) and is analogous to the ground-state pair peak in Eq. (5). In this case, the } n = 0 \text{ peak is located at}
FIG. 4 (color online). rf absorption spectra Eq. (7) for the state with periodic in time order parameter $\Delta(t)$ produced by a sudden switch of the pairing strength $\lambda_j - \lambda_j$ for $\delta \mu \approx -0.75 \Delta_j$. The detuning frequency $\omega$ is in units of time-averaged order parameter $\Delta_j$. Note two sequences of peaks at even and odd multiples of $\Delta_j$ and also jumps at $\omega = (2n + 1)\Delta_j$ that sometimes are on top of the odd peaks. The frequency of oscillations of $\Delta(t)$ is $\Omega = 2\Delta_j$. Multiple peaks are due to processes where an rf photon breaks an excited-state–ground-state Cooper pair and emits or absorbs several oscillation quanta (deltons).

The minimum detuning frequency $\omega^+ = \nu(\delta \mu) + \delta \mu$ cf. Eq. (5). Thus, peaks in the first sequence are at $\omega = \omega^+ + n\Omega$. When $\Delta(t)$ is the Jacobi elliptic function $dn$, $\Omega = 2\Delta_j$ [24], where $\Delta_j$ is the time average of $\Delta(t)$ over the period. We note that the Fourier components of Bogoliubov amplitudes, $a_{j\mu}$ and $b_{j\mu}$ in Eq. (7), are discontinuous at the Fermi level $\varepsilon_j = 0$ similar to $T = 0$ Fermi distribution. This is a consequence of the fact that initial states for the periodic regime are close to the normal state [9,13,14]. The discontinuities lead to jumps in the rf spectra at $\omega = (2n + 1)\Delta_j$, (Fig. 4).

The second series of peaks is the analog of the excited pair peak in Eq. (5). These peaks are at $\omega = -\Delta_j + n\Omega = (2n - 1)\Delta_j$, (Fig. 4). Their heights are suppressed for the same reason as in the excited pair peak in Eq. (5). Their width is determined by the width of the excited pair distribution function and becomes extremely narrow for large $\beta$, (Fig. 3). In this limit, they can be superimposed by jumps in the first sequence of peaks, (Fig. 4).

The sharp features of the rf spectra detailed above will be broadened by variety of effects in practice such as changes in particle number between experiments. More significant deviations will occur as one gets closer to the Feshbach resonance as our treatment is based on BCS theory. Finally, rf probing should be performed on a time scale shorter than the quasiparticle relaxation time $\tau_e \approx E_F/\Delta^2$ [14], which limits the lifetime of the steady states considered here. At times larger than $\tau_e$ an isolated system is expected to rethermalize to a state with a nonzero effective temperature which can be determined by balancing the total internal energy [14,15].

In conclusion, we have obtained rf spectra for the non-equilibrium steady states formed in a fermionic condensate due to a rapid switching of the pairing strength. The rf spectrum of the steady state with constant order parameter $\Delta_j \neq 0$ has two peaks in contrast to the spectrum of the paired ground state where there is a single peak. The peak at negative detuning frequencies reflects the presence of excited pairs—elementary excitations of the condensate and its shape is a direct measure of their distribution function. The other peak is a counterpart of the ground-state spectroscopic response. In the steady state characterized by a periodically oscillating $\Delta(t)$, each of the two peaks splits into a sequence of equidistant peaks with the spacing between peaks given by the frequency of oscillations $\Omega$.

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