Radiation induced oscillating gap states of nonequilibrium two-dimensional superconductors

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We study effects of infrared radiations on a two-dimensional BCS superconductor coupled with a normal metal substrate through a tunneling barrier. The phase transition conditions are analyzed by inspecting stability of the system against perturbations of pairing potentials. We find an oscillating gap phase with a frequency not directly related to the radiation frequency but resulting from the asymmetry of electron density of states of the system as well as the tunneling amplitude. When such a superconductor is in contact with another superconductor, it will give rise to an unusual alternating Josephson current.

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The rapid development of time-resolved spectroscopy technology have drawn growing interests in the study of nonequilibrium phenomena. The conductivity properties of solid materials can be greatly changed in nonequilibrium states induced by radiation. In two-dimensional electron gas, radiation induces zero resistance states at high Landau filling factors [1–4]. Recent discoveries on high temperature cuprate superconductors reveal that infrared radiations transform nonsuperconducting compounds into transient superconductors or enhance coherent superconducting transport even at temperature above the superconducting transition temperature [5, 6]. For irradiated BCS superconductors, the enhancement of superconducting gap [7–10] or the oscillating amplitude modes in nonadiabatic regime [11–16] induced at near gap frequencies have been discussed in many previous works. Recently, experimental studies on two-dimensional superconductors have shown superconducting order remains robust in ultra thin crystalline films which are a few atomic layers thick [17–21]. It brings new opportunities to the study of non-equilibrium superconducting orders in reduced dimensions.

In this Letter, we investigate effects of radiations on a two-dimensional BCS superconductor coupled with a normal metal through a tunneling barrier and find a new oscillating state induced by radiations which has not been discussed before. Different from the usual (fractional) ac Josephson effect [22, 23], in which oscillation is induced by dc voltages, the frequency of this alternating phase is not directly affected by the radiation frequency \(f_0\) but the radiation intensity, the amplitude of the tunneling interaction and most importantly, the asymmetry of density of states around Fermi-level. Such a state can be a probe of the internal properties of 2D superconductors.

The physical system we concern is a superconducting film coupled to a normal metal substrate by a tunneling barrier. The amplitude of the tunneling matrix element is determined by the thickness of the insulating film. We excite the superconductor with infrared radiations, the wavelength of which is assumed to be resonant with a phonon mode in the superconductor (shown in Fig.1).

As a result, the electrons feel an oscillating crystal field resulting from the excited phonons. The Hamiltonian of this tunneling system can be written as the sum of the Hamiltonians of N, S and the tunneling barrier,

\[
H = H_N + H_S + H_T
\]

\[
H_S = \sum_k \Psi_k^\dagger \begin{pmatrix} \tilde{\epsilon}_k + V_k \cos(\omega_0 t) & 0 \\ 0 & -\tilde{\epsilon}_k - V_k \cos(\omega_0 t) \end{pmatrix} \Psi_k + V_{int}
\]

\[
H_T = \sum_{k, k'} T_{k, k'} \left( c_{k}^\dagger d_{k'}^\dagger + c_{-k}^\dagger d_{-k'}^\dagger \right) + \text{h.c.} \tag{1}
\]

where \(c_k\) and \(d_{k'}\) are annihilation operators of the single particle state in the superconductor and the normal metal substrate respectively. The Hamiltonian of superconductor \(H_S\) with a single frequency oscillating field \(V_k \cos(\omega_0 t)\) is written in the Nambu matrix form [24], where \(\tilde{\epsilon} = \epsilon - \mu\) and \(\Psi_k = \begin{bmatrix} c_k^\dagger \\ c_{-k}^\dagger \end{bmatrix}\) is the basis of Nambu representation, and \(V_{int}\) is the electron-electron interaction. \(T_{k, k'}\) is the tunneling amplitude between S and N. For simplicity, \(\hbar\) and \(e\) are omitted in the discussion.

To determine superconducting phase transition of the irradiated system, we apply the theory of linear response and investigate the system’s stability against a weak ex-
ternal paring potential $\Delta_{\text{ext}}(t)$ which perturbs the system with $V_{\text{ext}} = -\Delta_{\text{ext}}c_{k}^\dagger c_{k} + \Delta_{\text{ext}}^* c_{-k}^\dagger c_{-k}$. The response function $\chi_{k,k'}(t, t')$ of paring amplitude $\psi_k = (c_{-k}^\dagger(t) c_{k}^\uparrow(t))$ to the perturbation is defined by

$$\psi_k(t) = \sum_{k'} \int dt' \chi_{k,k'}(t, t') \Delta_{\text{ext}}(k', t')$$  \hspace{1cm} (2)

In the presence of external paring potential, the disturbance will reach a steady state in the normal state, while in the superconducting state, however small perturbation can lead to a disturbance increasing with the time. Superconductivity is a result of the instability of the system to the perturbing paring potential.

To determine the response function of the system, we employ the RPA-like approach. First, we define $\chi^{\text{int}}_{0, k,k'}(t, t')$ as the pairing response function of a noninteracting system, i.e. $V_{\text{int}} = 0$ in Eq. (1). When the interaction is included, an electron in the system will feel not only the external paring potential, but also an induced pairing potential $\Delta_{\text{ind}}(t)$ exerted by all other electrons in the system. As a result:

$$\psi_k(t) = \sum_{k'} \int dt' \chi_{0,k,k'}(t, t') [\Delta_{\text{ext}}(k', t') + \Delta_{\text{ind}}(k', t')]$$  \hspace{1cm} (3)

The BCS mean field theory [25] corresponds to assuming:

$$\Delta_{\text{ind}}(k, t) = \sum_{k'} U_{k,k'} \langle c_{-k}^\dagger(t) c_{k}^\uparrow(t) \rangle = \sum_{k'} \Gamma_k \chi_{k,k'}(t)$$  \hspace{1cm} (4)

where $U_{k,k'}$ is the effective electron-electron interaction potential. Combine Eq. (2 - 4), we can express $\chi_{k,k'}(t, t')$ of the whole system in the form of operators:

$$\chi = [1 - \chi^{\text{int}}_0 U]^{-1} \chi^0$$  \hspace{1cm} (5)

where $\chi$ is the operator form of the response function and $[1 - \chi^0 U]^{-1}$ is the inverse of the operator $1 - \chi^0 U$.

Thus we can determine the irradiated superconducting system’s response to the perturbation as long as the noninteracting system’s response function $\chi^0$ is obtained.

We derive the expression of $\chi_0(t, t')$ with the Keldysh Green function technique [26, 27]. $\psi_k(t)$ is proportional to the off-diagonal element of the less Green function defined as $G^\less_{\alpha\beta}(k; t, t') = i \langle \bar{\Psi}_a^\dagger(t) \Psi_\alpha(t') \rangle$, where $\Psi_1 = c_k$ and $\Psi_2 = c_{-k}^\dagger$. With the perturbation Hamiltonian $V_{\text{ext}}$, the Dyson equation of $G^\less(t, t')$ can be obtained with the Langreth Theorem. For noninteracting system, $G^\less(t, t')$ can be expanded to the first order of $\Delta_{\text{ext}}$ as

$$G^\less_k(t, t') = G^\less_{0,k} + \int dt_1 G^\less_{0,k}(t, t_1) \Sigma'_k(t_1) G^\less_{0,k}(t_1, t')$$

$$+ \int dt_1 G^\less_{0,k}(t, t_1) \Sigma'_k(t_1) G^\less_{0,k}(t_1, t')$$  \hspace{1cm} (6)

where

$$\Sigma'_k(t) = \begin{pmatrix} 0 & \Delta_{\text{ext}}(k, t) \\ \Delta_{\text{ext}}^*(k, t) & 0 \end{pmatrix}$$  \hspace{1cm} (7)

and $G^\less_{0,k}(t, t')$ is the retarded (lesser) Green function of the noninteracting irradiated system without the external perturbation. We obtain $G^\less_0$ in the presence of radiation:

$$G^\less_0(t, t') = -i\theta(t - t') e^{-i \int_{t'}^t dt' \{ \pm \pi \pm V_k \cos(\omega t_1) \pm i \Gamma \}}$$  \hspace{1cm} (8)

where $\frac{i}{2} \Gamma$ is the energy level broadening resulting from the tunneling into the metal substrate. We assume that the self-energy resulting from the tunneling is nearly a constant around the Fermi level at energy scale of $\Delta \varepsilon \sim \Delta$. While the real part of the self-energy can be absorbed in the electron dispersion, the imaginary part of the self-energy is approximated to be a constant $-\frac{i}{2} \Gamma$. $G^\less_0$ can be obtained similarly. By substituting $G^\less_0$ and $G^\less_0$ into Eq. (6), $\chi_0(t, t')$ is given as

$$\chi_0(t, t') = -i \theta(t - t') \int \frac{d\varepsilon}{2\pi} \Gamma \tanh \left( \frac{\beta \varepsilon}{2} \right)$$

$$\cdot e^{-\frac{i}{2} \Gamma(t-t')} u_k(\varepsilon, t) u_k^*(\varepsilon, t') \delta_{k,k'}$$  \hspace{1cm} (9)

with

$$u_k(\varepsilon, t) = \sum_{n} J_n \left( \frac{V_k}{\omega_0} \right) e^{i(\varepsilon + \omega_0 - \varepsilon_0) t - 2i \frac{\omega_k}{\omega_0} \sin(\omega_0 t)}$$

$$\frac{\omega_0}{2 \omega_0} \sin(\omega_0 t)}$$  \hspace{1cm} (10)

where $J_n(x)$ is the Bessel function of the first kind and $\delta_{k,k'}$ comes from our assumption that the conservation of momentum is kept during the tunneling process.

With the response function derived, we can determine the conditions of the superconducting phase transitions. The stability of a linear system requires all poles of the response function lie on the lower half plane in the frequency space. The transition condition is determined when the first pole comes across the real axis during the change of the system parameters. Driven by periodic radiation field with frequency $\omega_0$, $\chi_{k,k'}(\omega, \omega')$ and $\Delta_k(\omega)$ can be expressed in matrices defined by

$$\chi_{mn, kk'}(\omega, \omega') = \chi^0_{mn, kk'}(\omega)$$

and

$$\Delta_k(\omega + m \omega_0) = \Delta_m k(\omega_k),$$

where $\omega_k \in [-\frac{i}{2} \omega_0, \frac{i}{2} \omega_0]$ and $m$ and $n$ are integers. Equation (5) in the frequency domain shows that the pole of $\chi(\omega)$ is determined by the zero point of $1 - \chi^0(\omega) U$. We can determine eigenvalues and eigenvectors of $\chi^0(\omega) U$. The stability condition proposed by Bode [28] requires that for $\omega$ on the real axis, each eigenvalue $x_0(\omega)$ of $\chi^0(\omega) U$ satisfies $|x_0(\omega)| \leq 1$ when $\arg x_0(\omega) = 2\pi$. Thus the transitional point for superconductivity is that for the largest $|x_0(\omega)|$ which satisfies $\arg x_0(\omega) = 2\pi$, it should be $|x_0(\omega)| = 1$. By this approach we set up a correspondence between the superconducting transitional temperature $T_c$ and the strength of the effective attractive potential $U_{k,k'}$, as well as the phase frequency $\omega_0$ of the gap phase $\Delta(\omega_0)$. In the approach above, we investigate the most unstable mode of the linear system against perturbation near the phase transition point. To obtain the physical observables at the temperature region far below $T_c$, such as the steady-state gap value, nonlinear gap equations should be applied. Here we make the assumption that the most unstable solution mode of the linearized equation at the transition point will correspond to the steady
The above analysis raises an interesting possibility, i.e., one may find a solution with \( \tilde{\omega}_c \neq 0 \). There is no a priori reason to believe that \( \tilde{\omega}_c \) must be zero or harmonics of the radiation frequency \( \omega_0 \). Such a solution implies an oscillating state whose frequency is neither zero nor integer (or half integer) multiple of the radiation frequency \( \omega_0 \). Thus it will be a new radiation-induced nonequilibrium effect, different from the usual (fractional) ac Josephson effect. Without radiation, it can be proved that \( \tilde{\omega}_c \) must be zero and Eq. (9) is consistent with the BCS gap equation. However in the irradiated state, there is no such conclusion and the occurrence of nonzero \( \tilde{\omega}_c \) is possible. In the following paragraphs, we will discuss the existing conditions of this oscillating gap state numerically.

First we investigate the effects of radiation on the superconducting system with constant density of state (DOS) around the Fermi energy. In our calculation, to involve the possible sub-bands motivated by radiations, the effective interaction potential \( U_{k,k'} \) is taken to be \( U_{k,k'} = -U \theta (\hbar\omega_D - |\xi_k - \xi_{k'}|) \), where \( U \) is the magnitude of the attractive potential and \( \theta(x) \) is the step function. We assume that the radiation frequency is the order of the Debye frequency \( \omega_D \), which is in terahertz regime in most BCS superconductors, and that the radiation energy \( V_0 \) and the imaginary part of self-energy \( \Gamma \) due to tunneling are at the order of magnitude of \( \hbar\omega_D \).

The electron-phonon coupling constant of the superconducting system assumed as \( \lambda = 1/U \rho(0) = 0.9 \), where \( \rho(0) \) is the density of state at the Fermi level.

We show the dependence of the transition temperature \( T_c \) on the magnitude of radiation energy \( V \) at different tunneling amplitudes in Fig. 2(a). Here we make the simplification that the radiation energy \( V_0 \approx V \) close to the Fermi energy. We can see the transition temperature \( T_c \) is suppressed by the radiation. \( V \sim T_c \) curves at different values of \( \Gamma \) show that tunneling to the substrate will also suppress the superconductivity. In this case, the alternating phase frequency \( \tilde{\omega}_c \) we obtain is always zero.

To search for cases of nonzero \( \tilde{\omega}_c \), we investigate the superconducting system with particle-hole asymmetry near the Fermi surface. We assume the superconductor in our system has a DOS with a linear inclination at the Fermi surface, which is expressed as \( \rho(\xi_k) = \rho(0) (1 + a|\xi_k|/\hbar\omega_D) \), where \( a \) is the slope and the total range of \( \rho \) is limited to \([0.95, 1.05]\) to avoid unphysical results. In such a system, we observe the occurrence of the radiation-induced oscillating state. In Fig. 3(a), the dependence of the oscillating frequency \( \tilde{\omega}_c \) on the radiation intensity \( V \) at different values of slope is shown. We see that \( \tilde{\omega}_c \) increase with the DOS slope \( a \) as well as the radiation intensity. \( \tilde{\omega}_c \) shows a nearly linear relationship with the asymmetric degree of the DOS [Fig. 3(a) inset]. \( \tilde{\omega}_c \) we obtain is in the regime of GHz, three or more orders of magnitude smaller than the radiation frequency. The significant difference of magnitude between this al-

FIG. 2. (a) The transition temperature \( T_c \) versus the radiation intensity \( V \) for superconductor with constant DOS at different tunneling amplitudes and \( \omega_0 = 0.8\omega_D \). The evolution of the relative gap modulus at the Fermi surface with time over one period of the radiation for superconductor (b) at constant DOS or (c) at \( \rho(\xi_k) = \rho(0) (1 + 0.08|\xi_k|/\hbar\omega_D) \) with \( \Gamma = 0.5 \).

FIG. 3. (a) The alternating phase frequency \( \tilde{\omega}_c \) versus the radiation intensity \( V \) at different values of the slope \( a \) of the DOS of \( \rho(\xi_k) = \rho(0) (1 + a|\xi_k|/\hbar\omega_D) \) at \( \Gamma = 0.5\omega_D \) and \( \omega_0 = 0.8\omega_D \). Here \( \tilde{\omega}_c \) is normalized by \( 10^{-3}\omega_D \). Insets, dependence of \( \tilde{\omega}_c \) on \( a \) at \( V = 0.4\hbar\omega_D \), (b) \( \tilde{\omega}_c \) versus the radiation intensity \( V \) at different values of the tunneling amplitude \( \Gamma \) at \( a = 0.08 \) and \( \omega_0 = 0.8\omega_D \). Insets, dependence of \( \tilde{\omega}_c \) on \( \Gamma \) at \( V = 0.6\hbar\omega_D \).
ternating phase and the radiation frequency could make the experimental observation of the effect easier.

Besides the asymmetry of the DOS, the occurrence of nonzero $\hat{\omega}_c$ also depends on the tunneling amplitudes between the superconductor and the substrate. Fig. 3(b) shows $\hat{\omega}_c$ with respect to the radiation intensity at different values of $\Gamma$. $\hat{\omega}_c$ increase rapidly with the tunneling amplitudes. $\Gamma = 0$ corresponds to the bulk superconducting system, where the interactions with the substrate are screened over the length of penetration. At this case, $\hat{\omega}_c$ is always zero, which indicates this radiation induced effect is special to the two-dimensional superconductor.

We calculate the relative modulus of the gap by solving the eigenvectors of the operator $(\mathbf{U} \chi_0)^{-1} - 1$. The evolution of the gap modulus over one period of radiation at the Fermi surface for systems with constant DOS [Fig. 2(b)] and with nonzero $\hat{\omega}_c$ motivated at $\alpha = 0.06$ and $\Gamma = 0.5$ [Fig. 2(c)] are plotted respectively. We can see that the radiation induce oscillation of the gap amplitude with the same frequency as the radiation. For system with nonzero $\hat{\omega}_c$, large proportion of the secondary and higher harmonic wave is also excited, as shown in Fig. 2(e).

When the system in Fig. 1 with nonzero $\hat{\omega}_c$ is connected to a bulk superconductor or a 2D superconductor with particle-hole symmetry, an unusual alternating Josephson current can be observed experimentally. When irradiated with infrared radiation, the phase difference of the two superconductor is $\phi = \hat{\omega}_c t$. The fast oscillating term due to the infrared radiation is ignored. Thus the tunneling current is $I = I_c \sin (\hat{\omega}_c t)$, where $I_c$ is affected by the radiation intensity and frequency. Unconventionally, the frequency of this alternating current depends on the radiation intensity and the degree of the electron-hole asymmetry of the 2D superconductor.

In conclusion, we find a radiation-induced oscillating state of the gap phase in a two-dimensional BCS superconductor with particle-hole asymmetry coupled to a normal metal substrate. Its oscillating frequency is determined by the radiation intensity, the tunneling amplitude with the substrates and the electron-hole asymmetry of the superconducting system, which is different from the (fractional) ac Josephson effect. When this system is connected to another superconductor in a Josephson junction, alternating current corresponding to this phase occurs.

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