Superconducting pair-breaking under intense sub-gap terahertz radiation

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Superconducting pair-breaking under intense sub-gap terahertz radiation

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Abstract: We study the effect of a strong and low frequency ($\omega < \Delta$, the superconducting gap) electrical field on a superconducting state. It is found that the superconducting gap decreases with the field intensity and wavelength. The physical mechanism for this dependence is the multi-photon absorption by a superconducting electron. By constructing the state of a superconducting electron dressed by photons, we determined the dependence of the superconducting gap on $E/\omega$ and temperature. We show that the critical temperature is determined by the parameter $E/\omega$ which is distinct from that induced by the heating effect. The result is consistent with experimental findings. This result can be applied to study the terahertz nonlinear superconducting metamaterials.

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In the past two decades, a great deal of works has been done in terahertz field, which is mainly because the attractive properties of high-power, long-wavelength, tunable laser sources and other and other potential applications. Several research groups in this area have produced innovative studies of nonlinear effects and ultrafast controlling carriers and spin dynamics. Terahertz lasers have been applied to investigate nonlinear transport and optical properties in an electron gas. Terahertz phenomena like resonant absorption\(^1\), the LO-phonon bottleneck effect\(^2\), terahertz cyclotron resonance\(^3\), terahertz photon-induced impact ionization\(^4\), photon-enhanced hot-electron effects\(^5\) and terahertz photon-assisted tunneling\(^6\) have all been intensely studied.

Terahertz radiation is widely used to study the properties of superconducting materials because the gap energy of many superconductors is of the order of a few terahertz. When a superconductor is under an electromagnetic radiation whose frequency is greater than the Copper-pair gap energy, the electrons can absorb a photon and the pair can be broken. Direct pair breaking does not occur under a field with frequency lower than gap energy. However, recent experimental measurements\(^7\)\(^-\)\(^9\) have revealed that sub-gap terahertz radiation can have a very strong effect on the superconducting properties. By using a high intensity terahertz radiation to study the ultrafast dynamics in superconducting thin film,\(^7\)\(^\text{-}\)\(^9\) it was found that at low temperatures the superconductivity of NbN thin films can be suppressed in the terahertz region by optical pulses.\(^7\)\(^\text{-}\)\(^12\) The experiment revealed that in the region 0.4\(-\)1.2 THz, the corresponding energy is smaller than gap energy of NbN film.\(^9\) Even at the maximum THz pulse energy, the calculated photon number is also 100 times smaller than the carrier density of NbN. However, the measurement of complex conductivity shows clear nonlinear effects at low temperatures and intense THz electric field. These experiments suggest that there may be a strong nonlinear process at the low frequencies whereby electrons can interact with multiple photons.

In this work, we will employ a model which involves superconducting Floquet states and uses the Bogoliubov-de Gennes (BdG) equation to describe the electron-multi-photon interaction. With this model the Cooper pair can be broken even when the incident photon energy is below the superconducting gap due to simultaneous absorption of multiple photons. Energy gap and current density are the most useful quantities in understanding this phenomenon. We first calculate the new form of the energy gap which is dependent on electron-photon coupling strength \(E/\omega\), then the current and other transport information can be derived. This inherently shows that all physical
quantities are strongly dependent on the parameter \( a^2 = \left( \frac{e^2}{\hbar m} \right) \left( \frac{E}{\omega} \right)^2 \) in energy unit. For the notational convenience, we simply denote \( E/\omega \) for \( a \).

Our analysis involves two steps. First, we calculate the single electron states under an intense radiation. The result is an electron dressed with photons. Next we apply the dressed electron states to solve the superconducting gap equation. Here, we choose the laser field to be along the \( x \) direction; \( \mathbf{E} = \text{Ecos}(\omega t)\mathbf{e}_x \), so the vector potential is \( \mathbf{A} = -(E/\omega)\text{sin}(\omega t)\mathbf{e}_x \). The transformation between electron gas under intense laser radiation and single electron is given by

\[
U^\dagger \left[ i \frac{\partial}{\partial t} - \frac{(p - eA)^2}{2m} \right] U = i \frac{\partial}{\partial t} - \frac{p^2}{2m},
\]

where \( U^\dagger = \exp\{i2\gamma_1 \omega t + i\gamma_1 \sin(2\omega t) + i\gamma_0 k_x \chi[1 - \cos(\omega t)]\} \), \( \gamma_0 = eE / (m\omega^2) \) and \( \gamma_1 = (eE)^2 / (8m\omega^3) \). The state of a single electron dressed by photons is given by \( U^\dagger \psi \), and \( \psi \) is the single electron state in the absence of the radiation field.

The BdG equation for superconductors subject to an intense terahertz field can be written in the form

\[
i \frac{\partial}{\partial t} \psi = \begin{pmatrix} \frac{(p - eA)^2}{2m} & \Delta \\ \Delta^* & -\frac{(p - eA)^2}{2m} \end{pmatrix} \psi = H_0 \psi.
\]

where \( \Delta = |\Delta|\exp(i\chi) \). The coupling between an electron and a photon can be eliminated by a unitary transformation \( T^\dagger \left[ i \frac{\partial}{\partial t} - H_0 \right] T = i \frac{\partial}{\partial \epsilon} \hat{I} - H \), where \( T^\dagger = \begin{bmatrix} U^* & 0 \\ 0 & U \end{bmatrix} \), and \( \hat{I} \) is a 2x2 unit matrix and \( H = \begin{bmatrix} \frac{p^2}{2m} & \hat{\Delta} \\ \hat{\Delta}^* & -\frac{p^2}{2m} \end{bmatrix} \), with \( \hat{\Delta} = T^\dagger \Delta T \). The wave function of the system can be written in the form of Floquet states

\[
\Phi = T^\dagger \psi = \begin{pmatrix} \phi_1 \\ \phi_2 \end{pmatrix} = \begin{pmatrix} U^* u_k e^{i\chi/2} \\ U v_k e^{-i\chi/2} \end{pmatrix} \exp(ik \cdot r - i\epsilon t).
\]

Then from (2) we have

\[
i \left( \begin{bmatrix} [-i\epsilon - i\epsilon' t + i2\gamma_1 \omega - i2\gamma_1 \omega \cos(2\omega t) - i\gamma_0 \omega k_x \sin(\omega t)] \phi_1 \\ [-i\epsilon - i\epsilon' t - i2\gamma_1 \omega + i2\gamma_1 \omega \cos(2\omega t) + i\gamma_0 \omega k_x \sin(\omega t)] \phi_2 \end{bmatrix} = \frac{\hbar k^2}{2m} \begin{bmatrix} \hat{\Delta} \\ \hat{\Delta}^* \end{bmatrix} \begin{pmatrix} \phi_1 \\ \phi_2 \end{pmatrix}. \]

By letting \( \alpha(t) = 2\gamma_1 \omega - 2\gamma_1 \omega \cos(2\omega t) - \gamma_0 \omega k_x \sin(\omega t) \) and noting \( \Phi^* \Phi = 1 \), then the new form of BdG equation can be written \( (\epsilon_k + \alpha(t))u_k + |\Delta|v_k = (t\epsilon)'u_k \) and \( -(\epsilon_k + \alpha(t))v_k + |\Delta|u_k = (t\epsilon)'v_k \), where \( \epsilon_k = \hbar^2 k^2 / (2m) \) and \( u_k \) and \( v_k \) are superconducting coherent factors.
defined by $u_k = \frac{1}{\sqrt{2}} \left(1 + \frac{\varepsilon_k + \alpha(t)}{(te')^2}\right)^{1/2}$, $v_k = \frac{1}{\sqrt{2}} \left(1 - \frac{\varepsilon_k + \alpha(t)}{(te')^2}\right)^{1/2}$. The energy spectrum can be easily calculated as following,

$$
\begin{bmatrix}
(t\epsilon') - \varepsilon_k - \alpha(t) & -\hat{\Delta}
\end{bmatrix}
\begin{bmatrix}
(t\epsilon') + \varepsilon_k + \alpha(t)
\end{bmatrix} = 0.
$$

Through some simple operations, the final energy spectrum can be given as

$$
\epsilon(t) = \pm \int_0^t \sqrt{(\varepsilon_k + \alpha(t'))^2 + |\Delta|^2} dt' = \pm \int_0^t \sqrt{(\varepsilon_k + 2\gamma_1 \omega - 2\gamma_1 \omega \cos(2\omega t') - \gamma_0 \omega k_{x} \sin(\omega t'))^2 + |\Delta|^2} dt'.
$$

The modified energy gap can be derived through the self-consistency equation

$$
\hat{\Delta} = V \sum_k (1 - 2f_k) \phi_1 \phi_2^* = V \sum_k (1 - 2f_k) \frac{\Delta}{(te')} \text{ where } f_k \text{ is Fermi-Dirac distribution.}
$$

By replacing the summation with an integration, the final form can be rewritten as

$$
1 = N(0)V \int_0^{E_D} \frac{1}{\sqrt{(\varepsilon_k + \alpha(t'))^2 + |\Delta|^2}} \tan \left(\frac{1}{2} \int_0^t \sqrt{(\varepsilon_k + \alpha(t'))^2 + |\Delta|^2} dt' \right) d\varepsilon_k,
$$

where $E_D$ is Debye energy. The gap function determined from (7) is time dependent because the system is under a time dependent field. Due to the very short period of the field, the parameter $\alpha(t)$ in will be approximated by its time average over one cycle.

**Figure 1** shows temperature dependent superconducting gap under various electrical field strength. In general by increasing $E / \omega$ the energy gap narrows and the critical temperature decreases. The
gap approaches zero as the E/ω becomes greater than 0.082. This limiting value is not exact due to the time average used in our calculation.

This can be explained as follows. The photon energy emitted by a terahertz field is about 4.13567meV with ω = 10^{12} Hz. This energy is smaller than the superconducting energy gap 2|Δ| at 0K which is only about 5meV. Hence one photon is unable to provide sufficient energy to break the Cooper pairs directly. However, the coupling energy induced by photon is about 0.8(E/ω)^2ε_k which can effectively modulate the scope of the superconducting energy gap. This indicates that the superconducting energy gap can be overcome with multiple photons whose individual energy is less than the band gap. The coupling coefficient is proportional to the parameter (E/ω)^2. Obviously, this phenomenon is a typical nonlinear effect.

Furthermore, the numerical results of energy gap can be used to calculate the number of superconducting carriers. By taking into account the thermal population of the quasiparticle excitations of the Cooper pairs (Bogoliubov quasiparticles), BCS theory predicts:^{17,18}

\[ n = n_s(T) = n_s(0) \left[ 1 - \frac{2}{k_B T} \int_0^{\varepsilon_F} f(\varepsilon, T) [1 - f(\varepsilon, T)] d\varepsilon \right], \quad (8) \]

where \( f(\varepsilon, T) = \left[ 1 + \exp(\sqrt{\varepsilon_k^2 + |\Delta|^2 / kT} \right]^{-1} \). We obtain,

\[ n = n_s(T) = n_s(0) \left[ 1 - \frac{1}{2 k_B T} \int_0^{\varepsilon_F} \operatorname{sech}^2 \left( \frac{\sqrt{\varepsilon_k^2 + |\Delta|^2} / 2kT \right) d\varepsilon \right], \quad (9) \]

The Fermi sphere is shifted by K when electric field is applied. The energy difference of two electrons in Cooper pairs can be written as

\[ \varepsilon_1 - \varepsilon_2 = \frac{\hbar^2}{2m} (k_F + K)^2 - \frac{\hbar^2}{2m} (k_F - K)^2 = 2 \frac{\hbar^2}{m} k_F K. \quad (10) \]

If \( \varepsilon_1 - \varepsilon_2 \) is equal to the energy gap, the critical momentum of Fermi sphere \( K \) can be calculated by

\[ \hbar K = \Delta \frac{m}{\hbar k_F}. \]

This leads to the critical velocity of superconducting carriers \( v_s = \frac{\Delta}{mv_F}. \)

The supercurrent density can be expressed as

\[ j_s = e n_s v_s = \frac{e n_s \Delta}{mv_F}. \quad (11) \]

In the calculation, the Debye temperature is 330K (equivalent to 0.02844eV).^{19,20} Therefore, the critical temperature \( T_c \) and the thermodynamic parameter \( N(0)V \) are approximately 16.35K and 0.32 respectively.^{21-23} We also choose the electric field used in experiment which is about \( 3 \times 10^6 \text{V/m}. \^9 \)
From Eq.(7) we derive the critical point that the energy gap disappears to be $E / \omega \approx 0.0817$. we compute the variation of energy gap with changing of parameter $E / \omega$ from 0 to 0.0817.

FIG. 2. The relation between ratio of superconducting carriers and energy gap (a), temperature (b), parameters $(E / \omega)^2$ (c).

For superconducting carriers, the formula (9) agrees well with the numerical results of energy gap.
Figure 2(a) reveals that wider energy gaps can sustain more superconducting carriers under different $\frac{E}{\omega}$. The decrease of superconducting carriers becomes more rapid as the energy gap is reduced or the parameter $\frac{E}{\omega}$ is increased. In figure 2(b), the superconducting carriers drop to zero decaying faster as $\frac{E}{\omega}$ approaches its critical value. This is similar to the situation in the top sub-figure. For the critical parameter $E/\omega = 0.082$ the superconducting carriers concentration approaches to zero within 0.1K with the current model. As $\frac{E}{\omega}$ increases, the strength of the electric field rises quickly if the frequency is kept constant. This will dramatically increase the electron energy in Cooper pairs and reduce the superconducting energy gap. Conversely, more Cooper pairs will be destroyed due to excitation and therefore, producing a lower superconducting carrier density. Overall, larger electric field leads to reduction of energy gap and superconducting carrier concentration. The relation between superconducting carrier density and $E/\omega$ under various temperatures is also displayed in figure 2(c).

In general, increasing the parameter $E/\omega$ breaks the Cooper pairs in three aspects. First of all, the external electric field can increase the kinetic energy of Cooper pairs. If the field strength is strong enough, it can accelerate the pairs to the critical velocity which leads to pair breaking. This is mainly because electric field augments the kinetic energy of Cooper pairs. Secondly, the frequency also plays an important role in the process of pair breaking. From the wave function of a dressed electron, Eq.(4), one can observe that the superconducting phase is very different from the unperturbed superconductor. The superconducting phase contains a complex factor dependent on both the frequency and strength of the applied field. This complex phase factor destroys the phase stiffness which also greatly contributes to the pair breaking. Finally, increasing the frequency of the electric field leads to higher incident photon energies which can ensure that the electrons have a higher probability to exceed the energy gap. These three reasons together result in the change of carrier density.

The electron-photon coupling dramatically modifies the energy gap and the maximum of critical current, as shown in figure 3(a). The relation between critical current and temperature shown in figure 3(b) is consistent with the temperature-dependent energy gap given in figure. 1.
Figure 3 shows the isothermal curve, showing the relationship between \( E/\omega \) and critical current density. It shows that the maximum current density appears at \( \frac{E}{\omega} \) and zero current emerges as \( \frac{E}{\omega} \to \infty \) in each curve with higher temperatures showing zero current at smaller values of \( \frac{E}{\omega} \). The expression of supercurrent also clearly indicates that the isothermal curve is monotone decreasing.
with increasing the parameter $\frac{E}{\omega}$ and temperature but increases with energy gap.

From Eq. (11), one can see that the critical current only relates to two variables: carrier density and energy gap which in-turn are dependent on temperature. This can be understood by observing the $\Delta - T$ phase diagram in Figure. 1. where the curves with larger parameters $a \frac{E}{\omega}$ clearly show a lower critical temperature. This can be attributed to a larger $\frac{E}{\omega}$ breaking more Cooper pairs. The higher electric field and frequency can break the bonding strength and phase stiffness of Cooper pairs which leads to lower superconducting carrier density. From current density, one can calculate the supercurrent by multiplying the factor $eN(0) \approx 4 \times 10^{10}$ m$^{-3}$. This result is reasonable in comparison with experimental data.$^9$

We also give a simple analysis on the imaginary conductivity from supercurrent. First, the current is proportional to the external electric field strength and inversely proportional to frequency. This implies that the conductivity decreases with increasing electric field strength$^9$ and can be explained phenomenologically by the relationship between energy gap and supercurrent carrier density. When strength of the applied electric field rises, the velocity of superconducting carrier will increase. This implies that the energy gap only leaves a little space (in terms of energy gap) for the supercurrent to reach its critical value. But, the higher speed of the superconducting carriers means it will be more easily to transform into normal carriers and the decline in superconducting carrier leads to a decrease in supercurrent. Therefore, the conductivity drops continually as the electric field frequency increase.$^9$

When the frequency of photon rises, the supercurrent driven by photons will be converted into normal current under constant field strength. As the photon energy becomes higher, the Cooper pairs easily achieve their critical velocity after absorbing part of photon energy. The extra photon energy will further accelerate the Copper pairs. Then, the velocity of superconducting carriers will exceed the critical velocity which leads to the breakup of Cooper pairs. The decrease in superconducting carrier density leads to the decline in supercurrent and conductivity. Therefore, the conductivity drops continually as the frequency grows, which agrees with experimental measurements.$^9$

Our result shows a low frequency field can significantly suppress the superconducting state in the nonlinear regime. In linear regime, gap can be overcome by absorption of a photon with energy greater than the Cooper pair gap. The absorption rate in this process is independent of the field and the can only occur when the photon energy of greater than the gap. Nonlinear absorption occurs when the
field intensity is strong. In the nonlinear regime, Copper pairs can absorb multiple photons with energy lower than the gap energy. Because the absorption of \( n \) photons is proportional to the \( n \)th power of the electrical field, the gap becomes dependent on the field. This indicates that the superconductivity can be suppressed and a strong field at low frequencies. Like the temperature effect, the gap can approach zero under a strong field. In our model, this occurs when \( E/\omega \) exceeds 0.082. This limiting field is only approximate due to the time average used in our calculation. In general, the nonlinear absorption reduces the gap size and the critical temperature, the temperature regime where the superconductivity is retained becomes smaller.

In summary, we have qualitatively and quantitatively determined the effect of an intense field whose frequency is below the superconducting gap on the Copper pair breaking. Frequency and field dependent superconducting energy gap is obtained. The result is in reasonable agreement with experiments. Our result provides a basis for tuning superconductivity with a strong sub-gap electrical field.

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