Two-photon absorption and amplitude mode in conventional superconductors with paramagnetic impurities

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Abstract. The two-photon absorption (TPA) spectrum of BCS superconductors is theoretically investigated. We discuss a possibility of observing the amplitude mode (one of collective modes in superconductors) in this spectrum. An absorption edge by this mode appears at the same frequency as that by quasiparticle excitations in the TPA spectrum of superconductors with time-reversal symmetry, and it will be difficult to distinguish the amplitude mode from quasiparticle excitations. This property originates from the fact that the magnitude of superconducting order parameter is one half of the energy gap in the linear absorption spectrum. In contrast to this one half of the energy gap is smaller than the magnitude of order parameter in superconductors with paramagnetic impurities. Though the amplitude mode is not well-defined owing to the broken time-reversal symmetry, a numerical calculation shows that the peak by this mode persists in the TPA spectrum. This peak appears at the frequency different from that of the absorption edge by quasiparticle excitations, and it is possible to specify the amplitude mode in the TPA spectrum.

1. Introduction
Microwave absorption spectrum reflects various properties in the superconducting state because the value of its frequency (ω) is close to that of the superconducting gap (Δ₀). The linear absorption spectrum shows that there is a finite absorption for ω > 2Δ₀, which is caused by quasiparticle excitations [1]. In the superconducting state collective modes (phase and amplitude modes of the order parameter) exist owing to the spontaneous symmetry breaking. The phase mode does not appear in the microwave absorption spectrum because it couples to the fluctuation of electron density and becomes a plasmon [2]. The amplitude mode is not effective in the linear absorption spectrum when superconductors have inversion symmetry. Then the amplitude mode [3, 4] has been studied in the superconductivity (SC) which coexists with the charge density wave (CDW) state, and was observed in the phonon spectrum through the coupling between SC and CDW [5, 6].

Recently the amplitude mode has been observed in a superconductor without the CDW state with use of the pump-probe spectroscopy [7]. In this experiment the amplitude mode is observed as an oscillation of the optical conductivity in the transient state. In contrast to this, a calculation of two-photon absorption (TPA) shows that there is a method to observe the amplitude mode in the stationary state [8]. By this calculation it is found that there is a
peak around $\omega \simeq \Delta_0$ in the TPA spectrum owing to the amplitude mode. It is also shown that the quasiparticle excitation by TPA gives a finite absorption edge at $\omega = \Delta_0$ in contrast to the vanishing absorption edge in the case of the linear absorption. Then there will be a possibility that an obscurity arises as for the origin of the absorption edge at $\omega = \Delta_0$ in the TPA spectrum (the amplitude mode or the quasiparticle excitation).

In this paper we show that a peak by the amplitude mode appears at the frequency different from an absorption edge by quasiparticle excitations in superconductors with paramagnetic impurities. Though there is no well-defined amplitude mode [9] due to the broken time-reversal symmetry, it is shown that the peak by this mode remains in the TPA spectrum.

2. Formulation

A nonlinear current is written as follows [10],

$$J^{(3)\mu}_\omega = e \int_{FS} \frac{mk_F}{2\pi} \int \frac{d\epsilon}{4\pi i} \sum_{\nu=x,y,z} \text{Tr} [v^\mu_k \hat{g}^{K(3)}_{\nu} (\epsilon + \omega, \epsilon)].$$

(1)

Here, $^{(3)}$ in the superscript indicates the third order of external field. We assume that the system is isotropic, and omit indices indicating directions of velocity ($v_k$) and external field hereafter. $\hat{g}^{K(3)}_\omega (\epsilon + \omega, \epsilon)$ is the quasiparticle Green’s function. $\hat{g}$ and $\text{Tr}$ indicate the Nambu representation and the summation of its diagonal elements, respectively.

The summation over wave number is replaced by the integration as $\sum_k = \frac{mk_F}{2\pi} \int_{FS} d\xi$ with $m$ the mass of electrons, $k_F$ the Fermi wave number, $N^3$ the number of sites and $\int_{FS}$ the integration over the Fermi surface. We put $\hbar = c = 1$ in this paper with $c$ the velocity of light.

In the dirty limit $\hat{g}^{K(3)}(\epsilon + \omega, \epsilon)$ is derived from kinetic equations [11], and the result is written with use of quasiclassical Green functions including the second order of external field as follows.

$$J^{(3)}_\omega = -e^2 D_\alpha \frac{mk_F}{2\pi} \int \frac{d\epsilon}{4\pi i} \left\{ A_\omega \text{Tr} \left[ \hat{g}^{+K}_{\epsilon+\omega,\epsilon} \hat{g}^K_{\epsilon+\omega,\epsilon} - \hat{g}^{+K}_{\epsilon,\epsilon+\omega} \hat{g}^K_{\epsilon+\omega,\epsilon} + \hat{g}^{+K}_{\epsilon,\epsilon} \hat{g}^K_{\epsilon-\omega,\epsilon} \right] + A_{\omega+\epsilon} \text{Tr} \left[ \hat{g}^{+K}_{\epsilon,\epsilon+\omega} \hat{g}^K_{\epsilon+\omega,\epsilon} + \hat{g}^{+K}_{\epsilon,\epsilon+\omega} \hat{g}^K_{\epsilon+\omega,\epsilon} + \hat{g}^{+K}_{\epsilon,\epsilon-\omega} \hat{g}^K_{\epsilon-\omega,\epsilon} + \hat{g}^{+K}_{\epsilon,\epsilon-\omega} \hat{g}^K_{\epsilon-\omega,\epsilon} \right] \right\}.$$  

(2)

Here, we omitted terms which are ineffective in the absorption spectrum. $D_\alpha$ is the diffusion constant, $A_{\pm\omega}$ is the external vector potential and $\hat{\tau}_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$. $^+$ and $^-$ in the superscript indicate the retarded and advanced Green function, respectively. $\hat{g}$ with one variable is a quasiclassical Green function in the equilibrium state, and $\hat{g}$ with two variables is a quadratic function of external fields. In the dirty limit kinetic equations for the latter functions are written as

$$B^{+\epsilon}_{\omega+\epsilon} = e^2 D_\alpha A_{\omega} A_{\omega+\epsilon} \sum_{\omega'=\pm\omega} \left( \hat{\tau}_3 \hat{g}^{+\epsilon}_{\omega',\omega} \hat{g}^{+\epsilon}_{\omega,\omega+\epsilon} \hat{g}^{+\epsilon}_{\omega,\omega+\epsilon} - \hat{g}^{+\epsilon}_{\omega',\omega+\epsilon} \hat{g}^{+\epsilon}_{\omega,\omega+\epsilon} \hat{\tau}_3 \right),$$

(3)

$$B^{+\epsilon}_{\omega+\epsilon,-\omega} = e^2 D_\alpha A_{\omega} A_{\omega+\epsilon} \left( \hat{\tau}_3 \hat{g}^{+\epsilon}_{\omega,\omega} \hat{g}^{+\epsilon}_{\omega,\omega+\epsilon} - \hat{g}^{+\epsilon}_{\omega,\omega} \hat{g}^{+\epsilon}_{\omega,\omega+\epsilon} \hat{\tau}_3 \right),$$

(4)

$$B^{-\epsilon(a)}_{\omega+\epsilon} = 2e^2 D_\alpha A_{\omega} A_{\omega+\epsilon} \sum_{\omega'=\pm\omega} (t^h_{\epsilon+\omega} - t^h_{\epsilon}) \left( \hat{\tau}_3 \text{Im} \left( \hat{g}^{+\epsilon}_{\omega+\epsilon,\omega} \hat{g}^{-\epsilon}_{\omega+\epsilon,\omega} \right) \hat{g}^{+\epsilon}_{\omega+\epsilon,\omega} \hat{\tau}_3 \right),$$

(5)

$$B^{-\epsilon}_{\omega+\epsilon,-\omega} = e^2 D_\alpha A_{\omega} A_{\omega+\epsilon} \sum_{s=\pm} (-s) (t^h_{\epsilon+\omega} - t^h_{-\epsilon}) \left( \hat{\tau}_3 \hat{g}^{+\epsilon}_{\omega+\epsilon,\omega} \hat{g}^{-\epsilon}_{\omega+\epsilon,\omega} - \hat{g}^{+\epsilon}_{\omega+\epsilon,\omega} \hat{g}^{+\epsilon}_{\omega+\epsilon,\omega} \hat{\tau}_3 \right).$$

(6)

Here, $B^{\pm\epsilon}_{\omega+\epsilon} := \hat{\tau}_3 \hat{g}^{\pm\epsilon}_{\omega+\epsilon,-\omega} - \hat{g}^{\pm\epsilon}_{\omega+\epsilon,-\omega} \hat{\tau}_3 - \hat{g}^{\pm\epsilon}_{\omega+\epsilon,-\omega} \hat{\tau}_3$. (The equations for advanced functions are the same as those for retarded ones with $+$ replaced by $-$). $\text{Im}$ indicates the imaginary part. $\hat{g}^{(a)}_{\epsilon+\epsilon} := \hat{g}^{K}_{\epsilon+\epsilon} - t^h_{\epsilon+\epsilon} \hat{g}^{+\epsilon}_{\epsilon+\epsilon} - t^h_{\epsilon+\epsilon} \hat{g}^{-\epsilon}_{\epsilon+\epsilon}$ is the anomalous Green function, which vanishes in the equilibrium state. $t^h_{\epsilon} := \tanh (\beta_T)$ ($T$ is the temperature).

The self-energy is written as $\hat{\Sigma}_{\epsilon,\epsilon'} = \hat{\Sigma}^{(ep)}_{\epsilon,\epsilon'} + \hat{\Sigma}^{(ai)}_{\epsilon,\epsilon'} + \hat{\Sigma}^{(pi)}_{\epsilon,\epsilon'}$. We use Born approximation for
the impurity scattering [12, 13]. \( \Sigma^{(ni)}_{e,e'} = \alpha \tilde{\tau}_3 \tilde{g}_{e,e'} \) and \( \Sigma^{(pa)}_{e,e'} = \alpha P \tilde{g}_{e,e'} \) (\( \alpha = n_i u^2 \text{mK}/2\pi \) and \( \alpha_p = n_i' u_{0}^2 \text{mK}/2\pi \)). \( n_i \) (\( n_i' \)) and \( u \) (\( u' \)) are the concentration and the magnitude of the potential of nonmagnetic (paramagnetic) impurities, respectively. \( D_{\alpha} = v_{F \alpha}^{2}/\alpha = v_{F \alpha}^{2} \tau /3 \) with \( \tau \) the relaxation time. We use the weak coupling approximation for the interaction between electrons and phonons. \( \Sigma^{(ep)}_{e,e',\tau} = g_0 \int \frac{d^{3}q}{(2\pi)^3} \hat{\tau}_{\tau} K_{\tau} \rho \hat{\tau} \hat{\tau}_{\tau} \omega_{\tau} /2 \) with \( g_0 = (g_{ph}^{2}/\omega_D)(\text{mK}/2\pi) \). \( g_{ph} \) and \( \omega_D \) are the coupling constant and Debye frequency, respectively. In this approximation \( \Sigma^{(a)(ep)}_{\tau} = (\tau_{h} - \tau_{h}^{*}) \Sigma^{(ep)}_{\tau} \).

We introduce \( \tilde{g} \) and \( f \) as follows. \( \hat{g}_{e,e} = e^2 D_{\alpha} A_{\omega} A_{-\omega} \left( \tilde{g}_{e(0)} \tilde{\tau}_0 + \tilde{f}_{e(0)} \tilde{\tau}_1 \right) \) and \( \hat{g}_{e+,e- \omega} = e^2 D_{\alpha} A_{\omega} A_{-\omega} \left( \tilde{g}_{e(2\omega)} \tilde{\tau}_0 + \tilde{f}_{e(2\omega)} \tilde{\tau}_1 \right) \) with \( \tilde{\tau}_0 \) the unit matrix and \( \tilde{\tau}_1 = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \). By solving the kinetic equations with use of \( \Sigma^{(ep)}_{\tau} = \Delta \tilde{\tau}_1 (\Delta \text{ is the superconducting gap with paramagnetic impurities included}), \) we obtain the following results.

\[
\begin{align*}
\left( \frac{\tilde{g}_{e}(0)}{\tilde{f}_{e}(0)} \right) & = M_{e,e}^{+} \left( \frac{g_{e+} + g_{e-}}{f_{e+} + f_{e-} + \Sigma^{(ep)}_{e,e}} \right), \\
\left( \frac{\tilde{g}_{e}(2\omega)}{\tilde{f}_{e}(2\omega)} \right) & = M_{e,e}^{+} \left( \frac{g_{e+}^{0} + \Sigma^{(ep)}_{e,e}}{f_{e+}^{0} + \Sigma^{(ep)}_{e,e}} \right), \\
\left( \frac{\tilde{g}_{e}(0)}{\tilde{f}_{e}(0)} \right) & = M_{e,e}^{-} \left( \frac{\Sigma^{(ep)}_{e,e}}{f_{e+}^{0} + \Sigma^{(ep)}_{e,e}} \right), \\
\end{align*}
\]

Here, \( \epsilon_{1,2} = \epsilon \pm \omega, \) \( M_{e,e}^{+} = Z_{e,e}^{++} \tilde{\tau}_3 - X_{e,e}^{++} \tilde{\tau}_0 - Y_{e,e}^{++} \tilde{\tau}_1 \) with \( Z_{e,e}^{ab} = i (\epsilon_{e}^{a} + \epsilon_{e}^{b} + 2i \alpha P X_{e,e})^{-1} \), \( X_{e,e}^{ab} = Z_{e,e}^{ab} (\epsilon_{e}^{a} / \epsilon_{p}^{b} + \Delta_{e}^{a} \Delta_{e}^{b} / \epsilon_{p}^{b}) / (\epsilon_{e}^{a} / \epsilon_{p}^{b} + \Delta_{e}^{a} \Delta_{e}^{b} / \epsilon_{p}^{b}) \). \( \tilde{\tau} = \epsilon_{e} / \epsilon_{e}^{\pm} \) and \( f_{e}^{\pm} = -i \Delta_{e}^{\pm} / z_{e}^{\pm}. \) We obtain \( \Sigma^{(ep)}_{0} \) and \( \Sigma^{(ep)}_{2\omega} \) by solving the equations: \( \overline{\Sigma}^{(ep)}_{0} = -g_0 \int \frac{d\omega}{2\pi} \left( \tilde{f}_{e(0)}^{(a)} + \tilde{f}_{e(0)}^{(b)} \right) \) and \( \overline{\Sigma}^{(ep)}_{2\omega} = -g_0 \int \frac{d\omega}{2\pi} \left( \tilde{f}_{e(2\omega)}^{(a)} + \tilde{f}_{e(2\omega)}^{(b)} \right) \). The results are written as follows.

\[
\begin{align*}
\overline{\Sigma}^{(ep)}_{0} & = \frac{g_0 \overline{D}_{\omega}}{\pi \overline{D}_{\omega}} \int d\omega \sum_{\omega' = \pm \omega} \left( t_{h}^{b} \omega' + t_{h}^{b} \omega' \right) \left( Y_{e,e}^{++} \tilde{g}_{e+}' + \left( Z_{e,e}^{+} + X_{e,e}^{+} \right) \tilde{g}_{e+}' \right) + t_{h}^{b} \omega' \left( Y_{e,e}^{++} \tilde{g}_{e+} + \left( Z_{e,e}^{+} + X_{e,e}^{+} \right) \tilde{g}_{e+} \right), \\
\overline{\Sigma}^{(ep)}_{2\omega} & = \frac{g_0 \overline{D}_{\omega}}{\pi \overline{D}_{\omega}} \int d\omega \sum_{\omega' = \pm \omega} \left( t_{h}^{b} \omega' + t_{h}^{b} \omega' \right) \left( Y_{e,e}^{++} \tilde{g}_{e+} + \left( Z_{e,e}^{+} + X_{e,e}^{+} \right) \tilde{g}_{e+} \right) + t_{h}^{b} \omega' \left( Y_{e,e}^{++} \tilde{g}_{e+} + \left( Z_{e,e}^{+} + X_{e,e}^{+} \right) \tilde{g}_{e+} \right).
\end{align*}
\]

Finally, \( D_{\omega} = 1 - \frac{g_0 \overline{D}_{\omega}}{\pi \overline{D}_{\omega}} \int \frac{d\omega}{2\pi} \sum_{\omega' = \pm \omega} \left( t_{h}^{b} \omega' + t_{h}^{b} \omega' \right) \left( Z_{e,e}^{+} + X_{e,e}^{+} \right) \tilde{g}_{e+} \). The nonlinear current is written as \( J_{\omega}^{(3)} = -K^{(3)}_{\omega} A_{\omega} \), and then the two-phonon absorption spectrum is given by \( \text{Re} \omega^{(3)}_N = -\text{Im} K^{(3)}_{\omega} / \omega \) with \( -\text{Im} K^{(3)}_{\omega} / \omega = e^4 D_{\overline{A}}^2 (E_{\omega} / 2\pi)^{\overline{a}} \overline{\alpha}_{e} \overline{\alpha}_{e} \). \( \sigma_{e} = \sigma_{e}^{(ap)} + \sigma_{e}^{(am)} \). (\( E_{\omega} \) is the electric field, and \( \text{Re} \) indicates the real part.) Here, \( \sigma_{e}^{(ap)} = \text{Re} \int \frac{d\omega}{\pi} \text{Tr} \left\{ \tilde{f}_{e(0)}^{a} 2 \text{Im} \left( \tilde{g}_{e(0)}^{a} \right) \right\} + \text{Re} \int \frac{d\omega}{\pi} \text{Tr} \left\{ \tilde{f}_{e(0)}^{a} 2 \text{Im} \left( \tilde{g}_{e(0)}^{a} \right) \right\} \). \( \sigma_{e}^{(am)} = 2 \text{Re} \int \frac{d\omega}{\pi} \text{Tr} \left\{ \tilde{f}_{e(0)}^{a} 2 \text{Im} \left( \tilde{g}_{e(0)}^{a} \right) \right\}. \)

3. Numerical calculations

We perform numerical calculations at \( T = 0 \). There is no direct excitation of quasiparticles for \( \omega < 2E_{0} \). (\( 2E_{0} \) is the energy gap for quasiparticle excitations [13].) The quantities introduced...
in the previous section do not depend on $\alpha$ (nonmagnetic impurities) except for $D_\alpha$. We do not specify the value of $\alpha$ in this section.

The nondiagonal part of $\hat{\Sigma}^{(ep)}(\xi_{\epsilon},\epsilon')$ represents the superconducting gap and its correction under external fields. $\Sigma_0^{(ep)}$ gives a static correction to the gap, and $\Sigma_2^{(ep)}$ includes a correction by the amplitude mode represented by $D_{2\omega}$. The calculated results for real and imaginary parts of $D_{2\omega}$ are shown in figures 1 and 2. $\Delta_0$ is the superconducting gap in the absence of paramagnetic impurities, and we take $\Delta_0$ as the unit of energy ($\Delta_0 = 1$). In the presence of finite values of $\alpha_p$ (paramagnetic impurities) $\text{Re}D_{2\omega} \neq 0$ at $\omega = \Delta$, and there is no well-defined amplitude mode. (On the other hand $\text{Re}D_{2\omega} = 0$ at $\omega = \Delta_0$ for $\alpha_p = 0$.) The minimum value of $\text{Re}D_{2\omega}$ increases with increasing $\alpha_p$. $-\text{Im}D_{2\omega}$, which indicates the damping of the amplitude mode and originates from quasiparticle excitations, takes a finite value at smaller $\omega$ with increasing $\alpha_p$ because of decreasing $E_g$.

The nonlinear correction to the absorption spectrum is written as

$$\frac{\text{Re}\sigma_{\omega}^{(3)}}{\sigma_0} = \frac{\Delta_0}{3\alpha} \left( \frac{\pi e \xi_0 |E_g|}{4\Delta_0} \right)^2 \Delta_0^2 \bar{\sigma}_\omega. \quad (9)$$

Here, $\xi_0 = v_F/\pi \Delta_0$ is the coherence length, and $\sigma_0 = e^2 n_c \tau / m$ is the conductivity in the normal state ($n_c = k_f^3 / 3\pi^2$ is electron density). $Z_{\epsilon,\epsilon'}^{+}$, which is a kind of diffusion propagator [14], diverges for $|\epsilon| > E_g$ because paramagnetic impurities also bring about elastic scattering as nonmagnetic impurities do [15]. The diffusion propagator is ineffective in the case that there is no quasiparticle excitation (its condition is $T = 0$ and $\omega < 2E_g$).

The calculated result of $\bar{\sigma}_\omega$ is shown in figure 3. (The calculation is restricted to $\omega < 2E_g$ as noted above. $\sigma_{\omega}^{(am)}$ is not the case because it does not include $Z_{\epsilon,\epsilon'}^{+}$.) The TPA spectrum shows a peak at $\omega = \Delta_0$ and no absorption for $\omega < \Delta_0$ in the case of $\alpha_p \approx 0$, which is the same result as in [8]. This indicates that the absorption edge by the amplitude mode is the same as that by quasiparticle excitations. The peak in the TPA spectrum ($\omega_p$ is its frequency) shifts to lower frequency with increasing $\alpha_p$. The absorption edge also shifts to lower frequency further. This edge originates from quasiparticle excitations, and its frequency is given by $\omega = E_g$. The difference between $\omega_p$ and

![Figure 1. The dependences of $\text{Re}D_{2\omega}$ (The real part of $D_{2\omega}$, which is in the vertex correction under external field and indicates existence of the amplitude mode of the order parameter) on $\omega$ for various values of $\alpha_p$.](image1)

![Figure 2. The dependences of $\text{Im}D_{2\omega}$ (The imaginary part of $D_{2\omega}$) on $\omega$ for various values of $\alpha_p$.](image2)
Figure 3. The dependences of $\bar{\sigma}_\omega$ (the TPA spectrum without a factor indicating the pump field) on $\omega$ for various values of $\alpha_p$.  

$E_p$ becomes large as $\alpha_p$ increases as shown in figure 4. The peak in the TPA spectrum indicates the existence of the amplitude mode as shown below. This implies that the amplitude mode is separable from quasiparticle excitations by introducing paramagnetic impurities in contrast to the case of $\alpha_p = 0$.

Figure 4 shows that the frequency of the peak is different from the superconducting gap $\Delta$ for $\alpha_p \neq 0$. Though $\omega_p \neq \Delta$, the peak in the TPA spectrum indicates the existence of the amplitude mode as is seen by comparing figures 5 and 6. Figure 5 shows calculated results for $\bar{\sigma}_\omega^{(am)}$ which includes the amplitude mode $D_{2\omega}$ in $\text{Re}\sigma_\omega^{(3)}$. $\bar{\sigma}_\omega^{(na)}$ shown in figure 6 is the term in which $D_{2\omega}$ in $\bar{\sigma}_\omega^{(am)}$ is replaced by 1. These figures show that the existence of $D_{2\omega}$ in $\bar{\sigma}_\omega$ is essential to the appearance of the peak in the TPA spectrum. The difference between $\omega_p$ and $\Delta$ arises from the fact that there is no well-defined amplitude mode for $\alpha_p \neq 0$. Our calculations show that there is a peak in the TPA spectrum even in the case of $\text{Re}D_{2\omega} \neq 0$. The suppression of the peak by increasing $\text{Re}D_{2\omega}$ for large values of $\alpha_p$ is compensated by the decreasing factor $\omega^3$ in the denominator of $\bar{\sigma}_\omega$.  

Figure 5. The dependences of $\bar{\sigma}_\omega^{(am)}$ (there is an amplitude mode in this term) on $\omega$ for various values of $\alpha_p$.  

Figure 6. The dependences of $\bar{\sigma}_\omega^{(na)}$ (there is no amplitude mode in this term) on $\omega$ for various values of $\alpha_p$.  

Figure 4. The dependences of $\omega_p$ (the frequency at which the TPA spectrum shows a peak by the amplitude mode) on $\alpha_p$.  

$E_g$ becomes large as $\alpha_p$ increases as shown in figure 4. The peak in the TPA spectrum indicates the existence of the amplitude mode as shown below. This implies that the amplitude mode is separable from quasiparticle excitations by introducing paramagnetic impurities in contrast to the case of $\alpha_p = 0$.

Figure 4 shows that the frequency of the peak is different from the superconducting gap $\Delta$ for $\alpha_p \neq 0$. Though $\omega_p \neq \Delta$, the peak in the TPA spectrum indicates the existence of the amplitude mode as is seen by comparing figures 5 and 6. Figure 5 shows calculated results for $\bar{\sigma}_\omega^{(am)}$ which includes the amplitude mode $D_{2\omega}$ in $\text{Re}\sigma_\omega^{(3)}$. $\bar{\sigma}_\omega^{(na)}$ shown in figure 6 is the term in which $D_{2\omega}$ in $\bar{\sigma}_\omega^{(am)}$ is replaced by 1. These figures show that the existence of $D_{2\omega}$ in $\bar{\sigma}_\omega$ is essential to the appearance of the peak in the TPA spectrum. The difference between $\omega_p$ and $\Delta$ arises from the fact that there is no well-defined amplitude mode for $\alpha_p \neq 0$. Our calculations show that there is a peak in the TPA spectrum even in the case of $\text{Re}D_{2\omega} \neq 0$. The suppression of the peak by increasing $\text{Re}D_{2\omega}$ for large values of $\alpha_p$ is compensated by the decreasing factor $\omega^3$ in the denominator of $\bar{\sigma}_\omega$.  

Figure 4. The dependences of $\omega_p$ (the frequency at which the TPA spectrum shows a peak by the amplitude mode) on $\alpha_p$.  

Figure 5. The dependences of $\bar{\sigma}_\omega^{(am)}$ (there is an amplitude mode in this term) on $\omega$ for various values of $\alpha_p$.  

Figure 6. The dependences of $\bar{\sigma}_\omega^{(na)}$ (there is no amplitude mode in this term) on $\omega$ for various values of $\alpha_p$.
\( \tilde{\sigma}^{(qp)}_\omega = \tilde{\sigma}^{(am)}_\omega \) does not include the amplitude mode \( D_{2\omega} \). This quantity is small as seen by making a comparison between figures 3 and 5. \( \tilde{\sigma}^{(qp)}_\omega \) includes a diffusion propagator \( (Z^+_\omega Z^-_{\omega'}) \), and makes a large contribution to the TPA spectrum in the case of \( \omega > 2E_g \) (there is a direct excitation of quasiparticles).

4. Discussion
When we put \( \xi_0 = 5 \text{ nm} \), \( |E_\omega| = x \text{ kV/cm} \), and \( \Delta_0 = 5 \text{ meV} \) to estimate \( \text{Re} \sigma^{(3)}_\omega / \sigma_0 \) quantitatively, the dimensionless factor is calculated as \( (\pi e^2 \xi_0 |E_\omega|/4\Delta_0)^2\Delta_0/(3\alpha) \approx (2x^2/(\alpha/\Delta_0)) \times 10^{-3} \). Numerical calculations in the previous section show that \( \Delta_0^2 \tilde{\sigma}_\omega > 100 \). Then \( \text{Re} \sigma^{(3)}_\omega / \sigma_0 \approx 0.1 \) for \( |E_\omega| > 2 \text{ kV/cm} \) even in the dirty limit \( \alpha = 10\Delta_0 \). This result indicates that a nonlinear correction to the absorption spectrum will be observable in experiments.

In this paper we used a monochromatic external field to discuss the absorption spectrum in the steady state. The direct excitation of quasiparticles should be avoided to obtain meaningful results. This is the reason why we suppose \( T = 0 \) and \( \omega < 2E_g \). In the presence of quasiparticle excitations we have to take account of mechanisms for the dissipation of energy such as inelastic scattering and nonlocality (nonconservation of momentum). In the case of \( \omega > 2E_g \) the direct excitation of quasiparticles above the energy gap narrows the range of \( |E_\omega| \) in which the perturbation expansion by external fields is valid. In contrast to this, the perturbation expansion is considered to be valid at \( T \neq 0 \) for \( \omega < 2E_g \) (there are only thermally excited quasiparticles in this case) when there is a dissipation mechanism. The difference between \( T = 0 \) and \( T \neq 0 \) is that there is a negative nonlinear correction to the absorption spectrum for \( \omega < E_g \) in the latter case [15]. Therefore, our calculation on the peak by the amplitude mode at \( T = 0 \) is considered to be valid at finite but low temperatures.

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