Effect of Impurity Scattering on the Nonlinear Microwave Response in High-$T_c$ Superconductors

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We theoretically investigate intermodulation distortion in high-$T_c$ superconductors. We study the effect of nonmagnetic impurities on the real and imaginary parts of nonlinear conductivity. The nonlinear conductivity is proportional to the inverse of temperature owing to the dependence of the damping effect on energy, which arises from the phase shift deviating from the unitary limit. It is shown that the final-states interaction makes the real part predominant over the imaginary part. These effects have not been included in previous theories based on the two-fluid model, enabling a consistent explanation for the experiments with the rf and dc fields.

KEYWORDS: intermodulation distortion, impurity scattering, unconventional superconductor, nonlinear Meissner effect, microwave conductivity, vertex correction

The high-temperature superconductors are attractive for use in microwave circuits because of their low surface resistance as compared to those of normal metals.\textsuperscript{1} This low-loss property is disturbed by the nonlinearity of its response to external fields. This nonlinearity means that the system is unstable with respect to changes in the input power. This effect makes superconductors unsuitable for practical applications. On the other hand, the nonlinear response is useful to investigate the intrinsic properties of superconductivity. It has been predicted that the nonlinear Meissner effect (NLME) shows a peculiar behavior in unconventional superconductors.\textsuperscript{2} This prediction is summarized in the following two points. One is that the nonlinear correction to the magnetic field penetration depth ($\lambda$) is proportional to the inverse of temperature ($T$). Then, the divergence at low temperatures yields a nonanalytic response. The other is that the nonlinear correction takes different values depending on the direction of the external field. These can be evidence of the existence of nodes in superconductors.

Several experiments have been conducted on this effect; these experiments yield different results depending on the measurement methods. An experiment that measures the dependence of $\lambda$ on the magnetic field yielded a result that is inconsistent with the theoretical prediction.\textsuperscript{3} (Neither the low-temperature upturn nor the angle dependence is observed.) Intermodulation distortion (IMD) is theoretically supposed to reflect the presence of the NLME.\textsuperscript{4} An experi-

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ment on the IMD seemingly shows a result consistent with the theoretical prediction.\(^5\) (Only the low-temperature upturn is observed. The angle dependence has not been investigated. In this sense, this experiment is inadequate to be considered as evidence for the NLME.)

In this paper, we microscopically develop the theory of nonlinear microwave response and consider how this contradiction arises. In the IMD experiment, the power is measured, which is expressed as
\[ P_{IMD} \propto |\Delta R_s + i\Delta X_s|^2.\]
Here, \( \Delta R_s = \Delta \sigma_1/(2\sigma_2)\sqrt{\omega \mu/\sigma_2} \) and \( \Delta X_s = -\Delta \sigma_2/(2\sigma_2)\sqrt{\omega \mu/\sigma_2} \) for \( \sigma_2 \gg \sigma_1 \). \((R_s, X_s, \text{and} \sigma = \sigma_1 - i\sigma_2 \text{are the surface resistance, surface reactance, and conductivity, respectively, and} \Delta \text{implies the nonlinear correction.})\)

Previous theories on IMD assume the validity of the two-fluid model in addition to that of the theory of Yip and Sauls; in these theories, only \( \Delta \sigma_2 \) is considered.\(^7\) (We show that this assumption does not necessarily hold.) This is sufficient when the response to the nonlinear dc field\(^3\) is considered. In the case of the IMD, however, there is a contribution from \( \Delta \sigma_1 \) in general.

In the linear response, it is known that \( \sigma_2 \gg \sigma_1 \) holds. On the other hand, the relationship between \( \Delta \sigma_1 \) and \( \Delta \sigma_2 \) is not known. Therefore, we calculate both real and imaginary parts of the nonlinear conductivity to determine which quantity is predominant. We have to specify a dissipation mechanism in order to estimate \( \Delta \sigma_1 \), though this is not the case for \( \Delta \sigma_2 \). The NLME comes into question at the low-temperature region, where the \( 1/T \)-upturn is supposed to occur. Therefore, we mainly consider the effect of nonmagnetic impurities on the nonlinear microwave response. This is because with regard to dissipation, the impurity scattering effect is dominant at low temperatures and the electron-electron correlation is dominant near \( T_c \).\(^8\) In this sense, we do not consider the type of correlation effect that functions as the enhancement factor and can be effective in the response to a static external field.\(^9\) The absence of the NLME under a nonlinear dc field can be explained by taking this effect into account. An explanation to the above contradictory behavior can be provided by combining this effect with the invalidity of the two-fluid model discussed here.

We consider isotropic impurity scattering. The self-energy with the self-consistent t-matrix approximation is
\[
\Sigma_0^R(\epsilon) = \frac{\Gamma_i G_0^R(\epsilon)}{\cot^2 \delta - G_0^R(\epsilon)^2}.
\]
Here, \( \Gamma_i = n_i/\pi N(0) \) \((n_i \text{and} N(0) \text{are the impurity density and the density of states at the Fermi level in the normal state, respectively}) \) and \( G_0^R(\epsilon) = \text{Tr} \sum_k \tilde{G}^R_{e,k}/(2\pi N(0)) \) with the Green function
\[
\tilde{G}^R_{e,k} = \frac{1}{\bar{\epsilon}^2 - \xi_k^2 - \Delta_k^2} \begin{pmatrix}
\bar{\epsilon} + \xi_k & \Delta_k \\
\Delta_k & \bar{\epsilon} - \xi_k
\end{pmatrix}.
\]
(\( \bar{\epsilon} = \epsilon - \Sigma_0^R(\epsilon). \)) The nonlinear response function (third order) can be expressed as follows. (The vertex correction is given by the functional derivative of the self-energy by the one-particle
Green function as that in the conserving approximation,\(^\text{10}\) which is also derived from Keldysh’s method on the nonequilibrium state.\(^\text{11}\) \(K^{(3)}(\omega_1, \omega_2, \omega_3) = \frac{1}{3!} \sum_{i,j,k} \int \mathrm{d}\hat{E}_{i}^{(e)}(\omega_i, \omega_j, \omega_k)\). (The conductivity is expressed as \(\Delta \sigma \propto K^{(3)} / \omega\).) \(\sum_{i,j,k} \) means the sum of all permutations \(\{i, j, k\} = \{1, 2, 3\}\) and \(\omega = \omega_1 + \omega_2 + \omega_3\).

\(\tilde{\Delta}_i^{(3)}(\omega_1, \omega_2, \omega_3) = \text{Tr}[-f_{c}g_{e_1e_2e_3e_4} - (f_{c} - f_{c})g_{e_1e_2e_3e_4} - (f_{e} - f_{e})g_{e_1e_2e_3e_4}]
- (f_{c} - f_{c})g_{e_1e_2e_3e_4} + f_{c}g_{e_1e_2e_3e_4}
+ \text{Tr}[-f_{c}h_{e_1e_2e_3e_4} + f_{c}h_{e_1e_2e_3e_4}] - (f_{c} - f_{c})h_{e_1e_2e_3e_4} + f_{c}h_{e_1e_2e_3e_4}
+ \text{Tr}[-(f_{c} - f_{c})h_{e_1e_2e_3e_4} + f_{c}h_{e_1e_2e_3e_4}]
- (f_{c} - f_{c})h_{e_1e_2e_3e_4} + f_{c}h_{e_1e_2e_3e_4}]
+ \text{Tr}[-(f_{c} - f_{c})h_{e_1e_2e_3e_4} + f_{c}h_{e_1e_2e_3e_4}]
- (f_{c} - f_{c})h_{e_1e_2e_3e_4} + f_{c}h_{e_1e_2e_3e_4}]

(3)

Here, \(\dot{g}_{e_1e_2e_3e_4}^{TTTT} = \sum_{k} v_{k} G_{e_1,k}^{T1} G_{e_2,k}^{T2} G_{e_3,k}^{T3} G_{e_4,k}^{T4}\), \(\dot{h}_{e_1e_2e_3e_4}^{TTTT} = \sum_{k} G_{e_1,k}^{T1} v_{k} G_{e_2,k}^{T2} v_{k} G_{e_3,k}^{T3}\), \(v_{k}\) is the quasiparticle velocity, \(f_{c} = \tanh(\epsilon / 2T)\), \(\epsilon_1 = \epsilon, \epsilon_2 = \epsilon = -\omega, \epsilon_3 = \epsilon = -\omega - 2, \epsilon_4 = \epsilon = -\omega\), and \(\omega = \omega_1 + \omega_2 + \omega_3\). \(D_{\epsilon,\epsilon'}^{RR}\) and \(D_{\epsilon,\epsilon'}^{RA}\) are vertex corrections; they are given afterward. The first and second traces represent the variations of the density of states and self-energy under the external field, respectively. The third trace implies the vertex correction, which represents the final-states interaction. The reasons for the invalidity of the application of the two-fluid model to the nonlinear response are as follows. (1) It is based on the assumption that the damping effect is independent of energy. (2) It includes only the nonlinear response of the density of states (the dependence of the damping effect on the external field and the final-states interaction are omitted). Therefore, we investigate these two aspects. The diagrams of the nonlinear response are shown in Fig. 1. Fig. 1(a) shows the diagram of the linear response; the nonlinear corrections are shown in (b) and (c). Fig. 1(b) and (c) show the nonlinear response with the variation of the density of states and vertex correction, respectively. In the linear response, vertex correction does not exist in the case of isotropic impurity scattering.

First, we consider the nonlinear response arising from the variation of the density of states; its response function can be expressed as follows.

\[
\left. \frac{\text{Re}K^{(3)}_{\text{DOS}}}{\omega} \right|_{\omega \to 0} = \int \frac{\partial f_{e}}{\partial \epsilon} \frac{\pi}{3} \text{Re} \left( \frac{\partial^{2} n_{R}^{wR}}{\partial \epsilon^{2}} \right) \frac{1}{\gamma_{e}}.
\]

(4)

\[
\left. \frac{\text{Im}K^{(3)}_{\text{DOS}}}{\omega} \right|_{\omega \to 0} = \int \frac{\partial f_{e}}{\partial \epsilon} \frac{2\pi}{3} \text{Re} \left( \frac{\partial^{2} n_{R}^{wR}}{\partial \epsilon^{2}} \right).
\]

(5)

Here, \(\gamma_{e} = -\text{Im} \Sigma_{0}(\epsilon), n_{R}^{w} = \int_{FS} V_{e} \bar{\epsilon} / \sqrt{\bar{\epsilon}^{2} - \Delta_{k}^{2}}, \) and \(V_{0,v,\nu} = 1, v_{k}^{2}, v_{k}^{4}\). (We substitute \(\Delta_{k} = \Delta_{0} \cos 2\theta\) and take \(\Delta_{0}\) as the unit of energy in the following numerical calculations.) If \(\gamma_{e}\) is independent of energy, we have the same result as that when the two-fluid model is used.

The temperature dependences of \(\text{Re}K^{(3)}_{\text{DOS}} / \omega\) and \(\text{Im}K^{(3)}_{\text{DOS}}\) are shown in Fig. 2. As the
Fig. 1. (b) and (c) Representative diagrams for $K^{(3)}$. The solid and wavy lines denote the one-particle Green function and the external field, respectively. The shaded rectangles denote vertex correction. (a) Linear response diagram. (d) Diagram of the impurity scattering effect with the self-consistent t-matrix approximation.

Fig. 2. Temperature dependences of $\text{Re}K_{DOS}^{(3)}/\omega|_{\omega \to 0}$ and $\text{Im}K_{DOS}^{(3)}$ for various values of the phase shift $\delta$ and $\Gamma_1 = 0.001$.

phase shift $\delta$ deviates from unitary scattering ($\delta = \pi/2$), $\text{Re}K_{DOS}^{(3)}/\omega|_{\omega \to 0}$ attains larger values and becomes proportional to the inverse of temperature. On the other hand, $\text{Im}K_{DOS}^{(3)}$ does not show a clear $1/T$-divergence, but it is cut off at low temperatures. (The graph of $\delta = 0.475\pi$ is seemingly divergent, but this is also verified to be cut off by comparing with that of smaller $\delta$ or $1/T$.) This behavior of $K_{DOS}^{(3)}$ can be explained by the dependence of the damping rate on energy. In previous theories on the nonlinear response in the Meissner state, the $1/T$-divergence is supposed to arise from the derivative of the density of states, which is cut off at low temperatures by the impurity scattering.$^{12}$ (In clean systems, $\text{Re} \partial^2 n_{\epsilon}/\partial \epsilon^2 \propto \delta(\epsilon)$ because $\text{Re} n_{\epsilon} \propto |\epsilon|$. If the damping rate takes a constant value, the result shown in Fig. 2 cannot be explained. The energy dependence of the damping rate $\gamma_{\epsilon}$ is shown in Fig. 3. As $\delta$ deviates from $\pi/2$, $\gamma_{\epsilon}$ decreases at around $\epsilon \simeq 0$. Then, $\text{Re} \partial^2 n^R_{\epsilon}/\partial \epsilon^2$ increases, but it is cut off at low
energies because $\gamma_0 \neq 0$. Therefore, the different dependences on temperature arise in Fig. 2. Re$K_{DOS}^{(3)}/\omega$ shows a $1/T$-divergence owing to the energy-dependent damping effect. Im$K_{DOS}^{(3)}$ is cut off at low temperatures reflecting the energy dependence of Re$\partial^2 n_{\epsilon R}^v / \partial \epsilon^2$.

The nonlinear correction to $K^{(3)}$ resulting from the variation of the self-energy is shown as a diagram similar to that in Fig. 1(c). The four-point vertex is expressed as $D_{R R \epsilon \epsilon}^{RA} = n_i T_i R^2 (1 - n_i T_i R^2 i \pi \partial n_{\epsilon R}^0 / \partial \epsilon)$. This term is small as compared to the vertex correction $D_{R A \epsilon \epsilon}^{RA}$, which is verified by a numerical calculation. Therefore, we omit this term.

Next, we consider the contribution of vertex correction to $K^{(3)}$, which is written as

$$K_{VC}^{(3)} = \int d\epsilon \frac{\partial f_\epsilon}{\partial \epsilon} \frac{\pi^2}{3} \sum_{i,j,k} \omega_k (N_1 + i \omega N_2) D_{\omega - \omega_i}^{RA} (N_1 + i \omega N_2). \quad (6)$$

Here, $N_1 = \text{Re}(\partial n_{\epsilon R}^v / \partial \epsilon) / \gamma_\epsilon$, $N_2 = |N_1 / \gamma_\epsilon + \text{Im}(\partial^2 n_{\epsilon R}^v / \partial \epsilon^2)| / (2\gamma_\epsilon)$, and $D_{\omega - \omega_i}^{RA} = (\omega - \omega_i + 2i\gamma_\epsilon) / [\pi(\omega - \omega_i) \text{Re} n_{\epsilon R}^0 / \gamma_\epsilon]$. (The term with $D^{RA}$ does not exist in the case of a nonlinear dc field.) The way in which the vertex correction depends on frequency originates from the identity

$$\hat{\Sigma}_{\epsilon + \omega} - \hat{\Sigma}_{\epsilon} = \Gamma_i \hat{T}_{\epsilon + \omega} R \frac{1}{\pi N(0)} \sum_k (\hat{G}_{k,\epsilon + \omega}^R - \hat{G}_{k,\epsilon}^A) T_{\epsilon}^A, \quad (7)$$

(here, $\hat{T}_{\epsilon}^R = (-\cot \delta \tau_3 - \sum_k \hat{G}_{k,\epsilon}^R / \pi N(0))^{-1}$ and $\tau_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$), which is similar to the identity discussed in the localization problem.\textsuperscript{13}

In the numerical calculation of the two-tone IMD we substitute $\omega_1 = \omega_2 = \omega + \Delta \omega$ and $\omega_3 = -\omega - 2\Delta \omega$ and then maintain $\omega_{1,2,3} / \omega$ as constant for $\omega \to 0$. The contributions from the vertex correction, Re$K_{VC}^{(3)} / |\omega| \to 0$ and Im$K_{VC}^{(3)}$, are shown in Fig. 4. At $\delta = \pi / 2$, both Re$K_{VC}^{(3)} / |\omega| \to 0$ and Im$K_{VC}^{(3)}$ decrease as the temperature decreases. As the phase shift deviates from $\pi / 2$, they show an upturn as $1/T$ increases. These behaviors are explained by the energy dependence of the damping rate and its effect on the density of states. Both Re$K_{VC}^{(3)} / |\omega| \to 0$ and Im$K_{VC}^{(3)}$ decrease as the temperature decreases.

Fig. 3. Energy dependence of $\gamma_\epsilon$ with the same parameters as those used in Fig. 2.
Fig. 4. Temperature dependences of $\text{Re}K_{VC}^{(3)}/\omega$ and $\text{Im}K_{VC}^{(3)}$ with various values of the phase shift $\delta$ and $\Gamma_i = 0.001$. We substitute $\Delta\omega/\omega = 0.01$.

and $\text{Im}K_{VC}^{(3)}$ are independent of the phase shifts at high temperatures. This means that the impurity scattering effect is less dependent on the phase shifts in this temperature region, as shown in the high-energy part of Fig. 3. The dependences of $K^{(3)}$ on phase shifts appear in the low-temperature region. The expression of $K^{(3)}$ indicates that $\text{Re}K_{VC}^{(3)}/\omega|_{\omega\to 0}$ and $\text{Im}K_{VC}^{(3)}$ are proportional to $\gamma_0^{-1}$ and $\gamma_0^0$, respectively. This distinguishes the behaviors of the real and imaginary parts of $K^{(3)}$. $\text{Re}K_{VC}^{(3)}/\omega|_{\omega\to 0}$ shows an almost $1/T$-divergence, but $\text{Im}K_{VC}^{(3)}$ is roughly proportional to $1/\sqrt{T}$. The absence of the cut off at low temperatures in $\text{Im}K_{VC}^{(3)}$ (unlike the case of $\text{Im}K_{DOS}^{(3)}$) originates from the energy dependence of the density of states $(n_0^{ER})$ in the vertex correction $D^{RA}$.

In Figs. 2 and 4, we can see that the real part of $K^{(3)}$ shows a $1/T$-divergence at some values of phase shifts. On the other hand, $\text{Im}K^{(3)}$ does not show such a behavior. We should clarify which of $\Delta\sigma_1$ and $\Delta\sigma_2$ is predominant in order to specify the origin of the low-temperature upturn in the IMD power. To see this, we evaluate the following ratio. $\gamma_0(\text{Re}K^{(3)}/\omega)/\text{Im}K^{(3)}$, which is equivalent to $(\gamma_0/\omega)\Delta\sigma_1/\Delta\sigma_2$, is shown in Fig. 5. In the hydrodynamic regime, which is the premise of our calculation, $\gamma_0$ is greater than $\omega$. Therefore, $\Delta\sigma_2$ is always predominant over $\Delta\sigma_1$ if $(\gamma_0/\omega)\Delta\sigma_1/\Delta\sigma_2 < 1$ holds. On the other hand, there is a possibility of $\Delta\sigma_1 > \Delta\sigma_2$ in the case of $(\gamma_0/\omega)\Delta\sigma_1/\Delta\sigma_2 > 1$, depending on the value of $\gamma_0/\omega$. As shown in Fig. 5, if we consider only $K_{DOS}^{(3)}$, $\Delta\sigma_2 > \Delta\sigma_1$ holds in the same way as the two-fluid model. When we take account of $K_{VC}^{(3)}$, $\Delta\sigma_1$ can predominate $\Delta\sigma_2$. As can be seen from Figs. 2 and 4, $\text{Im}K_{VC}^{(3)}$ takes values of the same order as $\text{Im}K_{DOS}^{(3)}$. On the other hand, $\text{Re}K_{VC}^{(3)}/\omega$ takes values that are 100 times greater than $\text{Re}K_{DOS}^{(3)}/\omega$. This difference originates from the following fact. It can be shown that the term $D^{RA} \propto 1/\Delta\omega$ arises in the real part of $K_{VC}^{(3)}$ (this term is cut off by the nonlocal effect mentioned below), but this term is canceled out in the imaginary part. Therefore, $\Delta\sigma_1$ can possibly predominate $\Delta\sigma_2$; as a result, the $1/T$-divergence can be
Fig. 5. Temperature dependences of $\gamma_0 (\text{Re} K_S^{(3)} / \omega) / \text{Im} K_S^{(3)}$ (here, $S = \text{DOS}$ or $\text{DOS} + \text{VC}$) with various values of phase shifts and the same parameters as those used in Fig. 4.

originated from $\Delta \sigma_1$. This yields a solution for the contradiction between the experiments with the nonlinear rf and dc fields, which is not resolved when the two-fluid model is used.

Here, we mention some issues that are not discussed above. Strictly speaking, in the case of $\delta \neq \pi/2$, the self-energy should be written as the matrix $\hat{\Sigma}^R(\epsilon) = \Sigma^R_0(\epsilon) \hat{\tau}_0 + \Sigma^R_3(\epsilon) \hat{\tau}_3$. $(\Sigma^R_3(\epsilon) = -\Gamma_i \cot \delta / [\cot^2 \delta - G^R_0(\epsilon)^2]$ and $\hat{\tau}_0$ is a unit matrix.) In this paper, we present the formula with $\Sigma^R_3(\epsilon) \to 0$ because it yields an intricate expression of $K^{(3)}$, and this gives almost the same numerical results as those for $\Sigma^R_3(\epsilon) \neq 0$. We present the numerical results of $K^{(3)}$ calculated by using the original expressions $(\Sigma^R_3(\epsilon) \neq 0)$. The diamagnetic terms that include the factor $\partial v_k / \partial k \hat{\tau}_3$ are omitted. This is because out of the two branches, only the gap-full branch remains in the vertex correction $\hat{D}^{RA}$; these branches arise from the matrix structure in the superconducting state. The nonlocal effect is not considered here. This effect also broadens the singular behavior of the derivative of the density of states in the same way as that by the impurity scattering effect.\(^9, 14\) However, the thickness of the film used in the IMD experiments\(^5\) is nearly 4000 Å, which is almost 100 times thinner than that of the experiment with the nonlinear dc field.\(^3\) This is almost the same order of magnitude as $\lambda$. Therefore, we omitted this effect here. (The numerical calculation of the current distribution with various values of $\lambda$ is given in ref. 15.) We show only the numerical results in which the impurity concentration $\Gamma_i$ was fixed. This is because our argument on $K^{(3)}$ can be similarly discussed when $\Gamma_i$ is varied. The different points are that the phase shift at which the $1/T$-divergence appears depends on $\Gamma_i$ and the absolute value of $K^{(3)}$ varies with $\Gamma_i$.

In our theory, whether $P_{\text{IMD}} \propto 1/T^2$ or not depends on the value of the phase shift, which is not known so far. As for the phase shift deviating from the unitary limit, however, there are several discussions related to the low-temperature thermal conductivity that suggests neither unitary nor Born limits.\(^16, 17\) With regard to the comparison between the real and
imaginary parts of the nonlinear conductivity, there is an experimental suggestion that $\Delta R_s$ is predominant over $\Delta X_s$,\cite{18} though $1/T$-divergence is not expected to exist in their temperature range. One of the possible experiments that can verify our theory is the third harmonic generation. When $\omega_1 = \omega_2 = \omega_3$, the contribution from vertex correction to $\text{Re}K^{(3)}$ is reduced to the same order as $\text{Re}K^{(3)}_{\text{DOS}}$. Therefore, it is expected that $\sigma_2 > \sigma_1$ holds and the $1/T$-divergence is cut off at low temperatures.

In this paper, we derived the general formalism of the nonlinear microwave conductivity under the influence of nonmagnetic impurities. We evaluated this formula by varying the value of the impurity scattering phase shift. As the phase shift deviates from the unitary limit, the nonlinear response shows a $1/T$-divergence owing to the dependence of the damping rate on energy. This is one of differences from previous theories where the $1/T$-divergence originates from the second derivative of the density of states. The predominance of the resistive part over the reactive part arises when the vertex correction is included. This term is not included in the two-fluid model. Therefore, the upturn of the IMD power at low temperatures can originate in the resistive part. This upturn does not need to be accompanied with $1/T$-divergence in the reactive part; this is a possible explanation to the seemingly contradictory results between the static and microwave experiments.

The numerical computation in this study was carried out at the Yukawa Institute Computer Facility.
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