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

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We theoretically investigate the intermodulation distortion in high-T_c superconductors. We study the effect of nonmagnetic impurities on the real and imaginary part of the nonlinear conductivity. The nonlinear conductivity is proportional to the inverse of the temperature owing to the dependence of the damping effect on the 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 on the basis of the two-fluid model, and then make it possible to give a consistent explanation for the experiments with the rf and dc field.

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 compared to the normal metals.[1] This low-loss property is disturbed by the nonlinearity of its response to the external field. The nonlinearity means that the system is not stable with respect to changes in the input power. This effect makes the superconductor unsuitable to practical use. On the other hand the nonlinear response is useful to investigate intrinsic properties of the superconductivity. It was predicted that the nonlinear Meissner effect (NLME) shows peculiar behavior in unconventional superconductors.[2] This prediction is summarized in the following two points. One is that the nonlinear correction to the magnetic field penetration depth (λ) is proportional to the inverse of the temperature (T). Then the divergence at low temperature leads to 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.

There are several experiments on this effect and these experiments show different results depending on the methods of the measurement. The experiment, which measures the dependence of λ on the magnetic field, show the result inconsistent with the theoretical prediction.[3] (Neither the low-temperature upturn nor the angle dependence is observed.) The intermodulation distortion (IMD) is theoretically supposed to reflect the NLME.[4] The experiment on

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the IMD seemingly shows the 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 as the evidence of the NLME.)

In this paper we develop the theory of the nonlinear microwave response microscopically and consider how this contradiction arises. In the experiment on the IMD the power is measured, which is written as $P_{IMD} \propto |\Delta R_s + \mathrm{i}\Delta X_s|^2$.[6] 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 $\sigma_2 >> \sigma_1$. (R_s, X_s, $\sigma = \sigma_1 - \mathrm{i}\sigma_2$ are the surface resistance, the surface reactance and the conductivity, respectively and Δ means the nonlinear correction.) Previous theories on the IMD assume the validity of the two-fluid model in addition to that of Yip and Sauls theory and only $\Delta\sigma_2$ is considered.[7] (We show that this assumption does not necessarily hold.) This is enough in the case of the response to the nonlinear dc field as in ref.[3] 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 $\sigma_2 >> \sigma_1$ holds. On the other hand the relation between $\Delta\sigma_1$ and $\Delta\sigma_2$ has not been known. Therefore we calculate both of the real and imaginary part of the nonlinear conductivity to see which quantity is predominant. We have to specify a mechanism of the dissipation in order to estimate $\Delta\sigma_1$, though this is not the case of $\Delta\sigma_2$. The NLME comes into question at the low temperature region, where the $1/T$-upturn is supposed to be observed. Therefore we consider mainly the effect of nonmagnetic impurities on the nonlinear microwave response. This is because as for the dissipation the impurity-scattering effect is dominant at low temperature and the electron-electron correlation is dominant near T_c.[8] In this sense we do not take account of the type of correlation effect which works as the enhancement factor and can be effective for the response to the static external field.[9] The absence of the NLME with the nonlinear dc field can be explained by this effect. It is made possible to give an explanation to the above contradictory behavior by combining this effect with the invalidity of the two-fluid model discussed here.

We consider the isotropic impurity scattering. The self-energy with the self-consistent t-matrix approximation is

$$\Sigma_0^R(\epsilon) = \frac{\Gamma_i G_0^R(\epsilon)}{\cot^2\delta - G_0^R(\epsilon)^2}. \tag{1}$$

Here, $\Gamma_i = n_i/\pi N(0)$ (n_i and $N(0)$ are the impurity density and the density of states at the Fermi level in the normal state, respectively) and $G_0^R(\epsilon) = \mathrm{Tr}\sum_k \hat{G}^R_{\epsilon,k}/(2\pi N(0))$ with the Green function,

$$\hat{G}^R_{\epsilon,k} = \frac{1}{\tilde{\epsilon}^2 - \xi_k^2 - \Delta_k^2}\begin{pmatrix} \tilde{\epsilon} + \xi_k & \Delta_k \\ \Delta_k & \tilde{\epsilon} - \xi_k \end{pmatrix}. \tag{2}$$

($\tilde{\epsilon} = \epsilon - \Sigma_0^R(\epsilon)$.) The nonlinear response function (the third order) is written as follows. (The vertex correction is given by the functional derivative of the self-energy by the one-particle Green function as in the conserving approximation,[10] which is also derived from Keldysh's

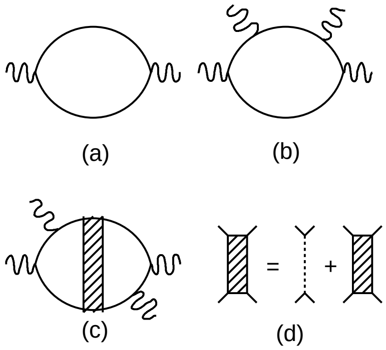

Fig. 1. (b) and (c) The representative diagrams for $K^{(3)}$. The solid and wavy lines express the one-particle Green function and the external field, respectively. The shaded rectangles denote the vertex correction. (a) The diagram of the linear response. (d) The diagram of the impurity-scattering effect with the self-consistent t-matrix approximation.

method on the nonequilibrium state.[11)]) $K^{(3)}(\omega_1,\omega_2,\omega_3) = \frac{1}{3!}\sum_{[i,j,k]}\int \mathrm{d}\epsilon \tilde{K}^{(3)}_\epsilon(\omega_i,\omega_j,\omega_k)$. (The conductivity is written as, $\Delta\sigma \propto K^{(3)}/\omega$.) $\sum_{[i,j,k]}$ means the sum of all permutations $\{i,j,k\} = \{1,2,3\}$ and $\omega = \omega_1+\omega_2+\omega_3$.

$$\begin{aligned}
\tilde{K}^{(3)}_\epsilon(\omega_1,\omega_2,\omega_3) = \mathrm{Tr}[&-f_{\epsilon4}\hat{g}^{RRRR}_{\epsilon1\epsilon2\epsilon3\epsilon4} - (f_{\epsilon3}-f_{\epsilon4})\hat{g}^{RRRA}_{\epsilon1\epsilon2\epsilon3\epsilon4} - (f_{\epsilon2}-f_{\epsilon3})\hat{g}^{RRAA}_{\epsilon1\epsilon2\epsilon3\epsilon4} \\
&- (f_{\epsilon1}-f_{\epsilon2})\hat{g}^{RAAA}_{\epsilon1\epsilon2\epsilon3\epsilon4} + f_{\epsilon1}\hat{g}^{AAAA}_{\epsilon1\epsilon2\epsilon3\epsilon4} \\
+\mathrm{Tr}[&-f_{\epsilon4}\{\hat{h}^{RRR}_{\epsilon4\epsilon1\epsilon2}D^{RR}_{\epsilon2,\epsilon4}\hat{h}^{RRR}_{\epsilon2\epsilon3\epsilon4} + \hat{h}^{RRR}_{\epsilon3\epsilon4\epsilon1}D^{RR}_{\epsilon1,\epsilon3}\hat{h}^{RRR}_{\epsilon1\epsilon2\epsilon3}\} - (f_{\epsilon3}-f_{\epsilon4})\hat{h}^{RAR}_{\epsilon3\epsilon4\epsilon1}D^{RR}_{\epsilon1,\epsilon3}\hat{h}^{RRR}_{\epsilon1\epsilon2\epsilon3} \\
&- (f_{\epsilon1}-f_{\epsilon2})\hat{h}^{ARA}_{\epsilon4\epsilon1\epsilon2}D^{AA}_{\epsilon2,\epsilon4}\hat{h}^{AAA}_{\epsilon2\epsilon3\epsilon4} + f_{\epsilon1}\{\hat{h}^{AAA}_{\epsilon4\epsilon1\epsilon2}D^{AA}_{\epsilon2,\epsilon4}\hat{h}^{AAA}_{\epsilon2\epsilon3\epsilon4} + \hat{h}^{AAA}_{\epsilon3\epsilon4\epsilon1}D^{AA}_{\epsilon1,\epsilon3}\hat{h}^{AAA}_{\epsilon1\epsilon2\epsilon3}\}] \\
+\mathrm{Tr}[&-(f_{\epsilon3}-f_{\epsilon4})\hat{h}^{ARR}_{\epsilon4\epsilon1\epsilon2}D^{RA}_{\epsilon2,\epsilon4}\hat{h}^{RRA}_{\epsilon2\epsilon3\epsilon4} - (f_{\epsilon2}-f_{\epsilon3})\hat{h}^{ARR}_{\epsilon4\epsilon1\epsilon2}D^{RA}_{\epsilon2,\epsilon4}\hat{h}^{RAA}_{\epsilon2\epsilon3\epsilon4} \\
&- (f_{\epsilon2}-f_{\epsilon3})\hat{h}^{AAR}_{\epsilon3\epsilon4\epsilon1}D^{RA}_{\epsilon1,\epsilon3}\hat{h}^{RRA}_{\epsilon1\epsilon2\epsilon3} - (f_{\epsilon1}-f_{\epsilon2})\hat{h}^{AAR}_{\epsilon3\epsilon4\epsilon1}D^{RA}_{\epsilon1,\epsilon3}\hat{h}^{RAA}_{\epsilon1\epsilon2\epsilon3}].
\end{aligned} \tag{3}$$

Here, $\hat{g}^{T1T2T3T4}_{\epsilon1\epsilon2\epsilon3\epsilon4} = \sum_k v_k\hat{G}^{T1}_{\epsilon1,k}v_k\hat{G}^{T2}_{\epsilon2,k}v_k\hat{G}^{T3}_{\epsilon3,k}v_k\hat{G}^{T4}_{\epsilon4,k}$, $\hat{h}^{T1T2T3}_{\epsilon1\epsilon2\epsilon3} = \sum_k \hat{G}^{T1}_{\epsilon1,k}v_k\hat{G}^{T2}_{\epsilon2,k}v_k\hat{G}^{T3}_{\epsilon3,k}$, v_k is the quasiparticle velocity, $f_\epsilon = \tanh(\epsilon/2T)$ and $\epsilon_1 = \epsilon$, $\epsilon_2 = \epsilon-\omega_1$, $\epsilon_3 = \epsilon-\omega_1-\omega_2$, $\epsilon_4 = \epsilon-\omega$, $\omega = \omega_1+\omega_2+\omega_3$. $D^{RR}_{\epsilon,\epsilon'}$ and $D^{RA}_{\epsilon,\epsilon'}$ are vertex corrections and given afterward. The first and second trace represents the variation of the density of states and the self-energy under the external field, respectively. The third trace means the vertex correction which represents the final-states interaction. The reason for the invalidity of an application of the two-fluid model to the nonlinear response is as follows. One of the reasons is that it is based on the assumption that the damping effect is independent of the energy. The other is that it includes only the nonlinear response of the density of states (the omission of the dependence of the damping effect on the external field and the final-states interaction). Therefore we investigate these two aspects. The diagrams for the nonlinear response are shown in Fig. 1. Fig. 1(a) indicates the diagram of the linear response and the nonlinear corrections are shown in (b) and (c). Fig.

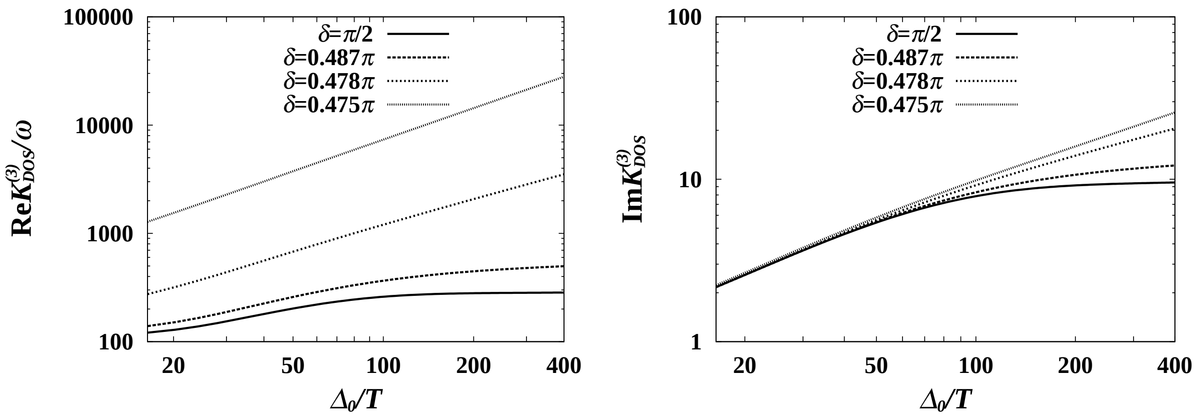

Fig. 2. The temperature dependences of $\mathrm{Re}K^{(3)}_{DOS}/\omega|_{\omega\to 0}$ and $\mathrm{Im}K^{(3)}_{DOS}$ with various values of the phase shift δ and $\Gamma_i = 0.001$.

1(b) and (c) represent the nonlinear response with the variation of the density of states and the vertex correction, respectively. In the linear response the vertex correction does not exist in the case of the isotropic impurity scattering.

First we consider the nonlinear response arising from the variation of the density of states, the response function is written as follows.

$$\left.\frac{\mathrm{Re}K^{(3)}_{DOS}}{\omega}\right|_{\omega\to 0} = \int \mathrm{d}\epsilon \frac{\partial f_\epsilon}{\partial \epsilon}\frac{\pi}{3}\mathrm{Re}\left(\frac{\partial^2 n^{wR}_\epsilon}{\partial \epsilon^2}\right)\frac{1}{\gamma_\epsilon}. \tag{4}$$

$$\mathrm{Im}K^{(3)}_{DOS} = \int \mathrm{d}\epsilon \frac{\partial f_\epsilon}{\partial \epsilon}\frac{2\pi}{3}\mathrm{Re}\left(\frac{\partial^2 n^{wR}_\epsilon}{\partial \epsilon^2}\right). \tag{5}$$

Here $\gamma_\epsilon = -\mathrm{Im}\Sigma_0(\epsilon)$, $n^{xR}_\epsilon = \int_{\mathrm{FS}} V_x\tilde{\epsilon}/\sqrt{\tilde{\epsilon}^2 - \Delta_k^2}$ and $V_{0,v,w} = 1, v_k^2, v_k^4$, respectively. (We put $\Delta_k = \Delta_0\cos 2\theta$ and take Δ_0 as the unit of energy in the following numerical calculations.) If γ_ϵ is independent of the energy, we have the same result as with the two-fluid model.

The temperature dependences of $\mathrm{Re}K^{(3)}_{DOS}/\omega$ and $\mathrm{Im}K^{(3)}_{DOS}$ are shown in Fig. 2. As the phase shift δ deviates from the unitary scattering ($\delta = \pi/2$), $\mathrm{Re}K^{(3)}_{DOS}/\omega|_{\omega\to 0}$ has larger values and becomes proportional to the inverse of the temperature. On the other hand $\mathrm{Im}K^{(3)}_{DOS}$ does not show clear $1/T$-divergence but is cut off at low temperature. (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 δ or $1/T$.) This behavior of $K^{(3)}_{DOS}$ can be explained by the dependence of the damping rate on the energy. In previous theories on the nonlinear response in the Meissner state the $1/T$-divergence is supposed to come from the derivative of the density of states, which is cut off at low temperatures by the impurity scattering.[12] (In clean systems $\mathrm{Re}\partial^2 n_\epsilon/\partial\epsilon^2 \propto \delta(\epsilon)$ because of $\mathrm{Re}n_\epsilon \propto |\epsilon|$.) If the damping rate takes a constant value, the result of Fig. 2 cannot be explained. The energy dependence of the damping rate γ_ϵ is shown in Fig. 3. As δ deviates from $\pi/2$, γ_ϵ decreases around $\epsilon \simeq 0$. Then $\mathrm{Re}\partial^2 n^{wR}_\epsilon/\partial\epsilon^2$ increases, but is cut off at low

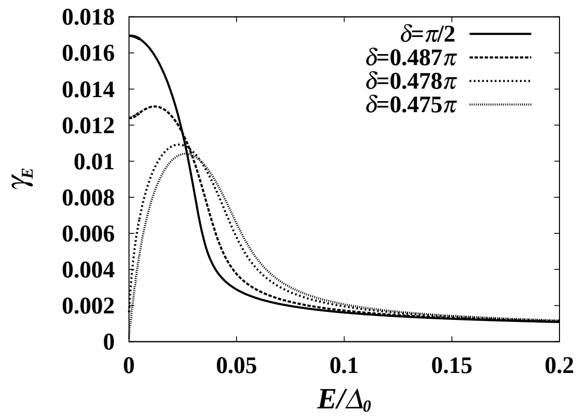

Fig. 3. The energy dependence of γ_ϵ with the same parameters as in Fig. 2.

energy because of $\gamma_0 \neq 0$. Therefore the different dependences on the temperature arise in Fig. 2; $\mathrm{Re}K^{(3)}_{DOS}/\omega$ shows $1/T$-divergence owing to the energy-dependent damping effect and $\mathrm{Im}K^{(3)}_{DOS}$ is cut off at low temperature reflecting the energy dependence of $\mathrm{Re}\partial^2 n^{wR}_\epsilon/\partial\epsilon^2$.

The nonlinear correction to $K^{(3)}$ resulting from the variation of the self-energy is represented with the diagram similar to Fig. 1(c). The four-point vertex is expressed as $D^{RR}_{\epsilon,\epsilon} = n_i T^{R2}_\epsilon/(1 - n_i T^{R2}_\epsilon \mathrm{i}\pi\partial n^{0R}_\epsilon/\partial\epsilon)$. This term is small compared with the vertex correction $D^{RA}_{\epsilon,\epsilon}$, which is verified with numerical calculation. Therefore we omit this term.

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

$$K^{(3)}_{VC} = \int \mathrm{d}\epsilon \frac{\partial f_\epsilon}{\partial\epsilon}\frac{\pi^2}{3}\sum_{[i,j,k]}\omega_k(N_1 + \mathrm{i}\omega N_2)D^{RA}_{\omega-\omega_i}(N_1 + \mathrm{i}\omega_k N_2). \tag{6}$$

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

$$\hat{\Sigma}^R_{\epsilon+\omega} - \hat{\Sigma}^A_\epsilon = \Gamma_i \hat{T}^R_{\epsilon+\omega}\frac{1}{\pi N(0)}\sum_k(\hat{G}^R_{k,\epsilon+\omega} - \hat{G}^A_{k,\epsilon})\hat{T}^A_\epsilon \tag{7}$$

(here $\hat{T}^R_\epsilon = (-\cot\delta\hat{\tau}_3 - \sum_k \hat{G}^R_{k,\epsilon}/\pi N(0))^{-1}$ and $\hat{\tau}_3 = \begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}$), which is similar to the identity discussed in the localization problem.[13)]

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

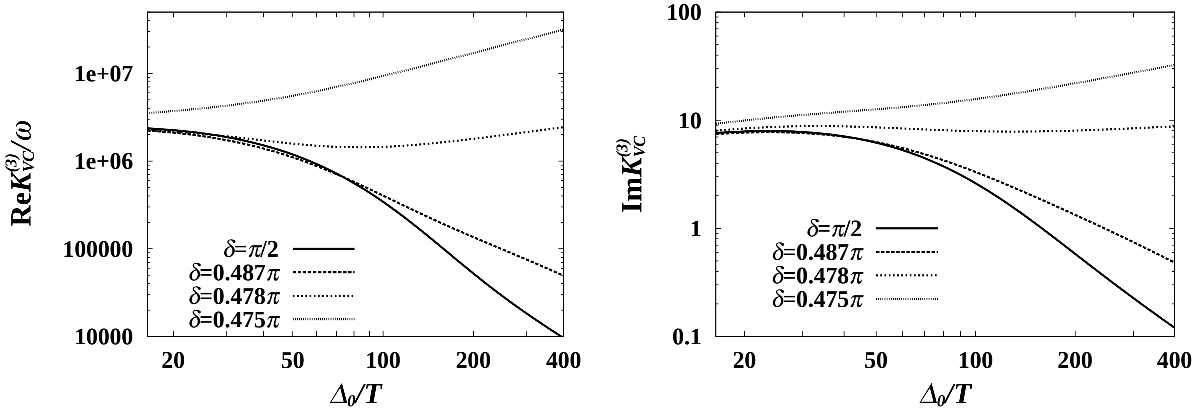

Fig. 4. The temperature dependences of $\mathrm{Re}K^{(3)}_{VC}/\omega$ and $\mathrm{Im}K^{(3)}_{VC}$ with various values of the phase shift δ and $\Gamma_i = 0.001$. We put $\Delta\omega/\omega = 0.01$.

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

We see in Figs. 2 and 4 that the real part of $K^{(3)}$ shows $1/T$-divergence at some values of phase shifts. On the other hand $\mathrm{Im}K^{(3)}$ does not show such 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. $\gamma_0(\mathrm{Re}K^{(3)}/\omega)/\mathrm{Im}K^{(3)}$, 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, γ_0 is larger than ω. 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 γ_0/ω. As is shown in Fig. 5, if we consider only $K^{(3)}_{DOS}$, $\Delta\sigma_2 > \Delta\sigma_1$ holds in the same way as the two-fluid model. When we take account of $K^{(3)}_{VC}$, $\Delta\sigma_1$ can be predominant over $\Delta\sigma_2$. As can be seen from Figs. 2 and 4 $\mathrm{Im}K^{(3)}_{VC}$ takes values of the same order as $\mathrm{Im}K^{(3)}_{DOS}$. On the other hand $\mathrm{Re}K^{(3)}_{VC}/\omega$ takes 100 times larger values than $\mathrm{Re}K^{(3)}_{DOS}/\omega$. This difference originates from the following fact. It can be shown that the term $D^{RA} \propto 1/\Delta\omega$ arises in real part of $K^{(3)}_{VC}$ (this term is cut off by the nonlocal effect which is mentioned below), but this term is canceled out in the imaginary part. Therefore $\Delta\sigma_1$ is possible to be predominant over $\Delta\sigma_2$ and then $1/T$-divergence can be

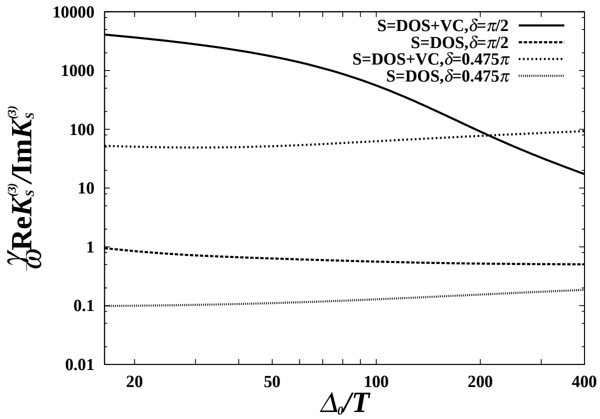

Fig. 5. The temperature dependences of $\gamma_0(\mathrm{Re}K_S^{(3)}/\omega)/\mathrm{Im}K_S^{(3)}$ (here $S = DOS$ or $DOS+VC$) with various values of the phase shift and the same parameters as in Fig. 4.

originated from $\Delta\sigma_1$. This gives a solution to the contradiction between the experiments with the nonlinear rf and dc field, which is not solved with the two-fluid model.

Here we mention some issues which are not discussed above. Strictly speaking in the case of $\delta \neq \pi/2$ the self-energy should be written as a matrix, $\hat{\Sigma}^R(\epsilon) = \Sigma_0^R(\epsilon)\hat{\tau}_0 + \Sigma_3^R(\epsilon)\hat{\tau}_3$. ($\Sigma_3^R(\epsilon) = -\Gamma_i \cot\delta/[\cot^2\delta - G_0^R(\epsilon)^2]$ and $\hat{\tau}_0$ is a unit matrix.) In this paper we present the formula with $\Sigma_3^R(\epsilon) \to 0$ because an intricate expression of $K^{(3)}$ appears and this gives almost the same numerical results as those of $\Sigma_3^R(\epsilon) \neq 0$. We presented numerical results of $K^{(3)}$ calculated with original expressions ($\Sigma_3^R(\epsilon) \neq 0$). The diamagnetic terms which include the factor $\partial v_k/\partial k\hat{\tau}_3$ are omitted. This is because only the gap-full branch remains out of two branches in the vertex correction $\hat{D}^{RA}$, which 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 the impurity-scattering effect does.[9,14] However the film used in the IMD experiments[5] is nearly 4000Å, which is almost 100 times thinner than that of the experiment with the nonlinear dc field.[3] This is almost same order of magnitude with λ. Therefore we omitted this effect here. (The numerical calculation of the current distribution with various values of λ is given in ref.[15]) We show only numerical results with the impurity concentration Γ_i fixed. This is because our argument on $K^{(3)}$ can be discussed similarly when Γ_i is varied. The different points are that the phase shift at which $1/T$-divergence appears depends on Γ_i, and the absolute value of $K^{(3)}$ varies with Γ_i.

In our theory whether $P_{IMD} \propto 1/T^2$ 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 which suggests neither unitary nor Born limit[16,17] In connection with the comparison between the real and imaginary part of the nonlinear conductivity there is an experimental suggestion that ΔR_s

is predominant over ΔX_s[18) though the temperature range is not where $1/T$-divergence is expected. One of the possible experiments which verify our theory is the third harmonic generation. When $\omega_1 = \omega_2 = \omega_3$ the contribution from the vertex correction to $\mathrm{Re}K^{(3)}$ is reduced to the same order as $\mathrm{Re}K^{(3)}_{DOS}$. Therefore it is expected that $\sigma_2 > \sigma_1$ holds and $1/T$-divergence is cut off at low temperature.

In this paper we derived the general formalism of the nonlinear microwave conductivity under the influence of nonmagnetic impurities. We evaluated this formula with the value of the impurity-scattering phase shift varied. As the phase shift deviates from the unitary limit, the nonlinear response shows $1/T$-divergence owing to the dependence of the damping rate on the energy. This is one of different points from previous theories where $1/T$-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 temperature can originate in the resistive part. This upturn does not need to be accompanied with $1/T$-divergence in the reactive part, and then this is a possible explanation to seemingly contradictory results between the static and microwave experiments.

Numerical computation in this work was carried out at the Yukawa Institute Computer Facility.

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