Theory of Photoinduced Changes of the Superfluid Weight in Superconductors

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We investigate changes in superfluid weight in photo-excited states. This quantity is obtained by a microscopic calculation of the third-order nonlinear response function on the assumption that the pumping intensity is low. The resultant expression includes the energy dependence of self-energy explicitly. The importance of this dependence is shown by performing numerical calculations with the electron-boson interaction included. Moreover, it is found that the vertex correction makes a predominant contribution to the photoinduced change in superfluid weight. The similarity between the linear and photoinduced responses in its temperature dependence is discussed, and a quantitative estimation of the calculated results is made in comparison with that of the experiments.

KEYWORDS: nonlinear response, superfluid weight, pump-probe, photo-excited state, unconventional superconductor, vertex correction

1. Introduction

Nonlinear optical spectroscopic studies on superconductors, especially high-$T_c$ cuprates, have piled up in recent years. Almost all of these experiments have been performed by pump-probe spectroscopy, and have focused on transient dynamics in the superconducting state in most cases. One of the reasons for this is that nonequilibrium dynamics possibly reflect interaction effects in superconductors.

There are only a few theoretical studies of the nonlinear optical response in superconductors. The kinetic equation approach, in which the excess quasiparticle distribution function is substituted in the linear response function (the nonlocal Mattis-Bardeen formula), is one of these studies, although the validity of such a quasiclassical approach has not yet been confirmed. The other approach is a phenomenological one, in which the nonequilibrium state is represented by introducing effective chemical potential or temperature. (These approaches are called the $\mu^*$-model or $T^*_c$-model.) We attempt a more microscopic approach here.

In this research, we study photoinduced changes (reductions) in superfluid weight. A corresponding experiment using a near-infrared pump and a terahertz probe has been performed recently. (This type of experiment is interpreted as a linear response in terms of the probe

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field in the nonequilibrium electron system, which is photo-excited by pump fields.) Although the transient state is a major concern in these studies, we focus on the static response (supposing that there is no time delay between the irradiation of the pump and probe fields). This is because, even in this simple situation, there has been no theory of the nonlinear response in superconductors, which is sufficient for explaining experimental results. (In this paper, we use the term 'nonlinear response' to describe the linear response in the photo-excited states.)

An example that illustrates such a situation is the expression of photoinduced changes of transmissivity. In the analysis of experiments performed so far, it was assumed that transmissivity is proportional to excited quasiparticle density, and that this quantity is proportional to the energy of a pump pulse and inversely proportional to superconducting gap. These assumptions have no substantial foundations and pose difficulty in interpreting experimental results regarding the temperature dependences of the reduction in superfluid weight. It is shown here that a microscopic calculation of the nonlinear response function enables a straightforward derivation of the reduction in superfluid weight.

In §2, we present a derivation of the nonlinear (the third order in the external field, which corresponds to a low pumping intensity) response function using Green’s function. It is shown that self-energy should be specified to analyze the resulting expression. The numerical calculations using the self-energy arising from the interaction with a bosonic propagator are presented in §3. It is shown numerically that the predominant term is the vertex correction by this interaction; its expression is given in the Appendix.

2. Formulation

The expression of current in the case of two-photon absorption is written as

$$J_\omega = -K^{(1)}(\omega)A_\omega - K^{(3)}(\omega, -\Omega, \Omega)A_\omega A_{-\Omega}A_\Omega.$$  (1)

Here, $K^{(1)}$ is the linear response function, and $A_\omega$ and $A_{\pm\Omega}$ are the vector potentials of the probe and pump fields (their frequencies are $\omega$ and $\pm\Omega$), respectively. We derive the expression of the nonlinear response function $K^{(3)}$ in this section.

Firstly, the linear response function is written as

$$K^{(1)}(\omega) = T \sum_{n,k} \text{Tr}[v_k \hat{G}_k(\epsilon_n + \omega_i)v_k \hat{G}_k(\epsilon_n)] + T \sum_{n,k} \text{Tr}[\frac{\partial v_k}{\partial \hat{\tau}_3}\hat{G}_k(\epsilon_n)]e^{i\epsilon_n 0^+}.  \tag{2}$$

Here, $\hat{G}_k(\epsilon_n) = (\frac{i\epsilon_n - \Sigma(\epsilon_n) - \xi_k}{-\Delta_k} - \frac{i\epsilon_n - \Sigma(\epsilon_n) + \xi_k}{-\Delta_k})^{-1} (T$ is the temperature, and $\epsilon_n = \pi T(2n - 1)$ and $\omega_i = 2\pi Tl$ are Matsubara frequencies; $\Sigma(\epsilon_n)$ and $\Delta_k$ are the self-energy and superconducting gap, respectively) and $\hat{\tau}_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$. By performing analytic continuation, transforming $\sum_k$ to $N_0 \int_{FS} \int d\xi$, and carrying out integration by $\xi$ firstly (in this case, the note in ref. 19 should be kept in mind), we obtain

$$K^{(1)}(\omega = 0) = N_0 \int_{FS} \int d\epsilon v^2_k \tanh \frac{\epsilon}{2T} \text{Re} \left[ \frac{\partial n^R_k}{\partial \epsilon} \right]. \tag{3}$$

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Here, $\epsilon^R = \epsilon - \Sigma^R$ ($R$ and $A$ indicate the retarded and advanced Green’s functions, respectively), $n^R = \epsilon^R / Z^R$, and $Z^R = \text{sgn}(\epsilon) \sqrt{(\epsilon^R)^2 - \Delta^2}$, $K^{(1)}(\omega = 0)$ is proportional to the inverse square of magnetic field penetration depth and characterizes the Meissner state. (This quantity is called the superfluid weight.)

The general expression of $K^{(3)}$, except for the vertex correction, is given by eqs. (9)-(12) in ref. 20 (the diagrammatic expression is the same as that in Fig. 1 in ref. 21), in which the matrix form of Green’s function is used here in the superconducting state. The nonlinear response function also consists of the paramagnetic term $K_a^{(3)}$ and the diamagnetic term $K_b^{(3)}$. Then $K^{(3)}$ is written as $K^{(3)}(\omega = 0, \Omega \neq 0) = K_a^{(3)}(\omega = 0, \Omega \neq 0) + K_b^{(3)}(\omega = 0, \Omega \neq 0)$.

If we take account of the penetration depth of the pump beam, we can adopt the local limit $q \to 0$ in the calculation of $K^{(3)}$. However, this limit is not trivial and requires some consideration. In the clean limit, the vertex $v_k$ does not induce the transition between the upper and lower bands of Bogoliubov quasiparticle spectra $\pm \sqrt{\xi_k^2 + \Delta_k^2}$ as a result of the odd symmetry of the vertex $v_{-k} = -v_k$, which is in contrast to that in the case of the vertex $\partial v_k / \partial k$, such as in the calculation of the Raman spectrum. Therefore, the paramagnetic term ($K_a^{(3)} \sim T \sum_n v_k^4 \text{Tr} [\hat{G} \hat{G} \hat{G} \hat{G}]$) vanishes for $q \to 0$ in the case of $\Omega \neq 0$ and $\gamma / q \to 0$ ($\gamma$ is the damping rate). On the other hand, this term remains finite in the local limit $q \to 0$ with the finite damping rate $q / \gamma \to 0$ included.

If we take account of the comparison between the mean free path and the penetration depth, we can carry out our calculations with the local limit $q / \gamma \to 0$. By performing the partial integral to derive the paramagnetic and diamagnetic terms separately, the expressions of $K_a^{(3)}$ and $K_b^{(3)}$ are respectively written as

$$K_a^{(3)}(\omega = 0, \Omega \neq 0) = \frac{N_0}{3!} \int \epsilon \int_{FS} \left( v_F \cos \theta \right)^4 \left[ \tanh \frac{\epsilon}{2T} \right] 2 \text{Re} \left( - \frac{\partial^2 n^R}{\partial \epsilon^R} \right) \frac{\alpha}{\epsilon^R - \epsilon}$$

$$+ \left( \tanh \frac{\epsilon}{2T} - \tanh \frac{\epsilon}{2T} \right) 2 \text{Re} \left[ \frac{\partial n^R}{\partial \epsilon^R} \alpha (\epsilon_+ - \epsilon_+) (\epsilon_+ - 2 \epsilon^R + \epsilon^A) \right],$$

(4)

$$K_b^{(3)}(\omega = 0, \Omega \neq 0) = \frac{N_0}{3!} \int \epsilon \int_{FS} \left( \frac{v_F^2 \cos \theta}{2 \epsilon_F} \right)^2 \left[ \tanh \frac{\epsilon}{2T} \right] 2 \text{Re} \left( - \frac{\partial n^R}{\partial \epsilon^R} \right)$$

$$- 4 \Delta_k^2 \left[ \left( \tanh \frac{\epsilon_+}{2T} + \tanh \frac{\epsilon}{2T} \right) 2 \text{Re} \left( \frac{-1/Z^R}{(\epsilon^R)^2 - (\epsilon^R)^2} \right) + \left( \tanh \frac{\epsilon}{2T} - \tanh \frac{\epsilon_+}{2T} \right) 2 \text{Re} \left( \frac{1/Z^R}{(\epsilon^R)^2 - (\epsilon^R)^2} \right) \right].$$

(5)

Here, $\epsilon_+ = \epsilon \pm \Omega$ and $\alpha = 1 - (Z^R / \epsilon_F)^2$. We put $\Delta_k = \Delta_0 \cos(2\theta)$ and $\xi_k = k^2 / 2m - \epsilon_F$. These expressions ensure that $K^{(3)}(\omega = 0, \Omega \neq 0) = 0$ for $\Delta_0 = 0$, which indicates that there is no photo-excited Meissner current in the normal state.

The above expression indicates that the energy dependence of self-energy is possibly influential. Therefore, for the purpose of investigating this effect, we consider the self-energy arising
from the interaction with the bosonic propagator \( D_\omega^R = D_0/(\gamma_b - i\omega) \); such as spin-fluctuation or phonon), which is written as

\[
\Sigma_\epsilon^R = \frac{-N_0}{2} \int_{FS} d\epsilon' \left[ \coth \frac{\epsilon'}{2T} \text{Im}(D_\epsilon^R) n_\epsilon^R + \tanh \frac{\epsilon'}{2T} D_\epsilon^A \text{Re}(n_\epsilon^R) \right].
\]  

(6)

We omit the dependence of \( \Sigma_\epsilon \) on wave number. The expression of the vertex correction by this interaction is given in Appendix.

Here, we comment on the relation of our formulation to experiments. The current eq. (1) is rewritten as \( J_\omega = \sigma_\omega E_\omega \). Conductivity consists of the linear-response part \( \sigma^{(1)}_\omega \) and the photoinduced part \( \Delta \sigma_\omega \). These are related to \( K^{(1)} \) and \( K^{(3)} \) as \( \sigma^{(1)}_\omega = -\frac{K^{(1)}(\omega)}{\text{i}(\omega + \Omega)} \) and \( \Delta \sigma_\omega = -\frac{K^{(3)}(\omega - \Omega, \Omega)}{\text{i}(\omega + \Omega)} \frac{|E|^2}{\Omega^2} \), respectively. We suppose that the photoinduced part \( \Delta \sigma_\omega \) originates from the third-order nonlinear response. This assumption indicates that the excitation fluence is low. (This is presumably satisfied in ref. 10 with taking account of the relation between photoinduced reflectivity and excitation fluence in ref. 12.)

Then we briefly discuss the relation between the nonlinear conductivity at finite \( \omega \) and the photoinduced changes of Meissner response. It is known that the sum rule holds in the nonlinear response: \( \int_0^\infty \omega \text{Im}(\chi^{(3)}(\omega, -\omega', \omega')) d\omega = 0 \). \( \chi^{(3)} \) is the third-order nonlinear optical susceptibility. This relation is equivalent to the relation (Kramers-Kronig transformation)

\[
\text{Re}K^{(3)}(\omega = 0, -\Omega, \Omega) = \frac{1}{\pi} P \int_{-\infty}^{\infty} d\omega' \frac{\text{Im}K^{(3)}(\omega', -\Omega, \Omega)}{\omega'}.
\]  

(7)

The real and imaginary parts of nonlinear conductivity are respectively written as \( \text{Re} \Delta \sigma(\omega, -\Omega, \Omega) = \frac{\text{Im}K^{(3)}(\omega, -\Omega, \Omega)}{\Omega^2} |E|^2 + \pi \delta(\omega) \text{Re}K^{(3)}(\omega, -\Omega, \Omega) |E|^2 \) and \( \text{Im} \Delta \sigma(\omega, -\Omega, \Omega) = \frac{\text{Re}K^{(3)}(\omega, -\Omega, \Omega)}{\Omega^2} |E|^2 \). The relation \( \omega \text{Im}(\chi^{(3)}(\omega, -\Omega, \Omega)) = -\text{Re} \Delta \sigma(\omega, -\Omega, \Omega) \) indicates that the decrease in superfluid weight \( |\text{Re}K^{(3)}(\omega = 0, -\Omega, \Omega)|/|\Omega^2| \) is compensated for by the increase in the real part of the conductivity at a finite frequency. Therefore, we can compare the experimental results of the conductivity at the finite frequencies and the calculated superfluid weight \( \text{Re}K^{(3)}(\omega = 0, -\Omega, \Omega) \).

3. Results

In the case that the paramagnetic term \( (K^{(3)}_a) \) predominates over the diamagnetic term \( (K^{(3)}_b) \) and \( \Sigma_\epsilon \) is independent of \( \epsilon \), \( K^{(3)} \) is written as

\[
K^{(3)}(\omega = 0, \Omega \neq 0) = \frac{N_0}{3!} \int_{FS} d\epsilon \int_v \psi_k \frac{\tanh \frac{\epsilon_+}{2T} - \tanh \frac{\epsilon}{2T}}{\Omega^2} \text{Re} \left[ \frac{\partial n_{\epsilon R}}{\partial \epsilon} \right].
\]  

(8)

(We put \( \alpha = 1 \) because of \( \Delta_0 \ll \epsilon_P \).) This expression indicates that the temperature dependences of \( K^{(1)}(\omega = 0) \) and \( K^{(3)}(\omega = 0, \Omega \neq 0) \) are similar to each other. This seemingly explains the experimental results.\(^{10} \) However, it is shown below that this approximation does not give an adequate result quantitatively. The reason for this is that \( \Omega \) is large compared with \( \Delta_0 \), and thus the \( \epsilon \) dependence of \( \Sigma_\epsilon \) cannot be neglected (especially in strongly correlated systems). Furthermore, the predominance of its vertex correction is revealed afterwards.
(In the case of \( \epsilon_F > \Omega \), the paramagnetic term \( K_a^{(3)} \) predominates over the diamagnetic term \( K_b^{(3)} \); moreover, the former term is enhanced compared with the latter term by taking account of the \( \epsilon \) dependence of \( \Sigma_\epsilon \). This is a different point from the two-photon absorption in Mott insulators where the latter terms predominate over the former terms.\(^{20} \))

The above expression of \( K^{(3)} \) also indicates that the temperature dependence of the photoinduced changes of the superfluid weight \( K^{(3)}(\omega = 0, \Omega \neq 0) \) is different from that of the nonlinear Meissner effect \( K^{(3)}(\omega = 0, \Omega = 0) \propto 1/T \).\(^{25} \) The nonlocal effect is predominant in the latter case.\(^{26} \) On the other hand, this is not the case in the photoinduced case owing to the absence of the divergent term.

Here, we attempt a quantitative estimation of the photoinduced changes of superfluid weight, \( K^{(3)}(\omega = 0, -\Omega, \Omega)|A_\Omega|^2 = K^{(3)}(\omega = 0, -\Omega, \Omega)|E_\Omega|^2/\Omega^2 \). The relative change is rewritten as

\[
\frac{K^{(3)} E^2}{K^{(1)} \Omega^2} \rightarrow a^2 \left( \frac{K^{(3)}}{K^{(1)}} \right)_{\text{calc}} \frac{\epsilon^2 E^2}{\Omega^2_{\text{exp}}}.
\]

On the right side, as a preparation for quantitative calculations, we explicitly write the unit of length \( (a) \) and the electric charge \( (e) \); ‘calc’ and ‘exp’ indicate the calculated results and experimental values, respectively. Here, we use the value of the electric field \( E \) as a clue to make a comparison with experimental results. It has been shown that \( \frac{K^{(3)} E^2}{K^{(1)} \Omega^2} \approx 0.16 \) in the experiment,\(^{10} \) and thus \( eE \propto \Omega_{\text{exp}} \sqrt{\frac{0.16}{a \sqrt{\left( \frac{K^{(3)}}{K^{(1)}} \right)_{\text{calc}}}}} \) is required. (The unit of length in calculation \( a \) is given by estimating the coherence length: \( \xi_0^{\text{calc}} \times a = \xi_0^{\text{exp}} \) with \( \xi_0^{\text{calc}} = v_F/\pi \Delta_0 \). For instance, we put \( v_F = 1 \) and \( \Delta_0 = 0.1 \) in the calculation, and suppose that \( \xi_0^{\text{exp}} = 20 \) Å. Then \( a = 6.28 \) Å.) If the self-energy \( \Sigma_\epsilon \) is independent of \( \epsilon \) and the vertex correction is absent as in eq. (8), then \( \left( K^{(3)}/K^{(1)} \right)_{\text{calc}} \approx 1/\Omega^2_{\text{calc}} \). In this case, electric field is estimated as \( E \approx \Omega_{\text{exp}} \sqrt{0.16} \Omega_{\text{calc}}/ea \). If we put \( a = 5 \times 10^{-10} \) m, \( \Omega_{\text{exp}} = 1.5 \) eV, and \( \Omega_{\text{calc}} = 5 \), then \( E = 6 \times 10^9 \) V/m is obtained. Although the experimental value of the electric field is not known exactly, this is considered to be extremely large. (If the refractive index is fixed to 1, this electric field corresponds to an optical intensity of more than 1 TW/cm\(^2 \)). Therefore, we can conclude that eq. (8) and its approximation are inadequate. As noted above, the expression of \( K^{(o)} \) indicates that the \( \epsilon \) dependence of self-energy is relevant; moreover, its vertex correction is found to be predominant by numerical calculations, as shown below.

The results of the numerical calculation are shown below. In the calculations, we omit the \( K_b^{(3)} \) term. This is because, even in the case of \( \Omega \sim \epsilon_F \), the absolute values of \( K_b^{(3)} \) are at most comparable to those of an imprecise \( K_a^{(3)} \) such as eq. (8), which is a lower limit of \( K_a^{(3)} \). In actual numerical calculations, the values of \( K^{(3)} \) with self-energy included are much larger than these values as shown below. We add a small \( \delta = -0.001 \) to the imaginary part of self-energy as effective impurities and a finite mean free path. The range of integration by \( \epsilon \) does not affect the result of \( K^{(3)} \) itself, as long as it exceeds \( \Omega \). On the other hand it affects...
self-energy. Therefore we fix it to 12.5.

The dependences of $K^{(3)}$ and $K^{(3)}/K^{(1)}$ on temperature for several values of $\Delta_0$ are shown in Fig. 1. ($K^{(3)}$ in Fig. 1(a) are calculated results with the constant value of $N_0$ omitted. This does not affect the absolute value of $K^{(3)}/K^{(1)}$. $K^{(3)}/K^{(1)}$ is almost independent of temperature for $\Delta_0 = 0.02$, and its dependence on temperature becomes stronger with increasing $\Delta_0$. The case of $K^{(3)}/K^{(1)}$ except the vertex correction also shows the same tendency. These results reflect the difference in the temperature dependence of self-energy, which originates from the relative magnitudes of $\Delta_0$ and $\gamma_b$. For a small $\gamma_b/\Delta_0$, the variation in self-energy is large for $0 < T/T_c < 1$ compared with that for a large $\gamma_b/\Delta_0$; this makes the temperature dependence of $K^{(3)}/K^{(1)}$ for a smaller $\gamma_b/\Delta_0$ larger. It is also shown that the vertex correction to $K^{(3)}/K^{(1)}$ is larger than the term excluding it by one or two order of magnitudes.

The dependences of $K^{(3)}/K^{(1)}$ on $\Omega$ for several values of $\Delta_0$ are shown in Fig. 2. The inset shows that $K^{(3)}/K^{(1)}$ except the vertex correction varies monotonically with decreasing $\Omega$ as the expression in §2 indicates, although it is not proportional to $1/\Omega^2$ owing to the energy dependence of self-energy. The expression for the vertex correction $K^{(3)}$ indicates that it increases to 0 with $\Omega \to 0$. The nonmonotonic variation in $K^{(3)}$ originates from the relative magnitudes of $\gamma_b$ and $\Omega$, which can be seen in Fig. 4 below. For a smaller $\gamma_b$, the energy dependence of $D_0\omega$ becomes significant, inducing a peak structure in the $\Omega$ dependence of $K^{(3)}$.

The dependences of $K^{(3)}/K^{(1)}$ on temperature for several values of $\gamma_b$ and $N_0D_0$ are shown in Fig. 3. For a smaller $\gamma_b$, the larger variation in $K^{(3)}/K^{(1)}$ with increasing temperature originates from the relative magnitudes of $\gamma_b$ and $\Delta_0$, as discussed above. The increase in $K^{(3)}/K^{(1)}$ with increasing $N_0D_0$ is observed, which originates from the variation in self-
energy and the vertex correction. This increase saturates because of the requirement of self-consistency.

The dependences of $K^{(3)}/K^{(1)}$ on $\Omega$ for several values of $\gamma_b$ and $N_0D_0$ are shown in Fig. 4. The nonmonotonic behavior discussed above is observed. The larger $K^{(3)}/K^{(1)}$ values for smaller $\gamma_b$ values originate from the fact that a strong $\omega$ dependence of $D_\omega$ makes the self-energy large. As for the dependences of $K^{(3)}/K^{(1)}$ on $N_0D_0$, the same explanation as that in the case of Fig. 3 holds.

The results of the numerical calculations indicate that the temperature dependence of
Fig. 4. Dependences of $K^{(3)}/K^{(1)}$ on $\Omega$ for (a) $\gamma_b = 0.2, 0.5, 1.0$, and $2.0$ and $N_0 D_0 = 1.0$, and (b) $N_0 D_0 = 0.2, 0.5, 1.0$, and $1.5$ and $\gamma_b = 0.5$. $\Delta_0 = 0.05$ and $T/T_c = 0.1$. The insets show $K^{(3)}/K^{(1)}$, except the vertex correction. The scales of the horizontal axes of the insets are the same.

$K^{(3)}/K^{(1)}$ varies with several parameters, but that there are some parameters in which $K^{(3)}/K^{(1)}$ shows a weak temperature dependence similar to that of experimental results. On the other hand, the vertex correction term is predominant irrespective of several parameters. If we perform a quantitative estimation similar to that discussed above, $(K^{(3)}/K^{(1)})_{\text{calc}} \approx 800$ gives a 10% reduction in superfluid weight at a pump intensity less than 1GW/cm$^2$.

It is possible to verify the predominance of the vertex correction in $K^{(3)}$ by experiments. $K_a^{(3)}$ includes the integral $\int_{FS} \cos^4 \theta$ or $\int_{FS} \cos^2 \theta \sin^2 \theta$, depending on the relative configuration of the pump and probe beams, for the copolarized or cross-polarized configuration, respectively. Therefore if $K_a^{(3)}$ predominates in $K^{(3)}$, then the former configuration gives a response three times larger than the latter. On the other hand, $K_{vc}^{(3)}$ shows none of this behavior. This is because the term including $\Sigma^{(a)(2)}_{\epsilon,\epsilon'}$ predominates in $K_{vc}^{(3)}$. ($\Sigma^{(a)(2)}_{\epsilon,\epsilon'}$ includes only the pump fields whose frequencies are $\pm \Omega$, and then two energy variables are both $\epsilon$.) In this term, the above two configurations include the integrals $\int_{FS} \cos^2 \theta \times \int_{FS} \cos^2 \theta$ or $\int_{FS} \cos^2 \theta \times \int_{FS} \sin^2 \theta$, which give the same value.

4. Summary and Discussion

In this research we calculated the photoinduced changes in superfluid weight in $d$-wave superconductors. We microscopically obtain the third-order nonlinear response function, which corresponds to the change in magnetic field penetration depth in photo-excited states. The derived expression of the nonlinear response shows that the explicit energy dependence of self-energy is relevant and necessary to clarify the temperature dependence and absolute value of the photoinduced reduction in superfluid weight. By numerical calculations with the boson-electron interaction included, it is shown that the term of the vertex correction is predominant in the nonlinear response. The temperature dependence of the nonlinear response is found to
be similar to that of the linear response for some parameters, which is consistent with the result of the optical-pump and THz-probe experiments.

Our calculation was performed by integrating $\xi$ analytically, which indicates that the band structure is simplified to be constant. The photoinduced change in superfluid weight results from the excitations around 1.5 eV. Therefore, a realistic band structure, especially at high energies, is needed to perform a precise quantitative estimation. We omitted the momentum dependence of the interaction, although it may also be required to include this effect in more detailed studies. By examining these details, the temperature dependences and the absolute values of the nonlinear response, which is parameter-dependent in this study, will be precisely determined.

In our calculations, the pump and probe fields are set to act on systems simultaneously. If we consider the relation between this setting and the transient response, our case corresponds to operating probe beams immediately after optical excitation by pump beams. (For example, the time delay is 1.3 ps in the experiments discussed in ref. 10.) To discuss the time evolution of the response for longer time delays, it is required to develop our formulation to include time variables. This could be carried out by extending the range of the frequency variables of $K^{(3)}$. This is because variations in the time domain are related to those in the frequency domain. If we take account of this and the variations in the time-dependent external field $E$ in the frequency domain, the integration of $K^{(3)}$ with $E$ by frequency variables would give the time evolution of the photoinduced changes in superfluid weight.

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Appendix: Vertex corrections

Here, we present the expression of the vertex correction to $K^{(3)}$ originating from the interaction with the boson propagator given in §2. The vertex correction to $K^{(3)}$ is written as (the original expression prior to transformations by integration and others is obtained by referring to ref. 27.)

$$K^{(3)}_{vc}(\omega = 0, \Omega \neq 0) = \frac{-4}{3\epsilon}(K^{RA}_{vc} + K^{(a)}_{vc}).$$

$$K^{RA}_{vc} = \frac{1}{2} \int d\epsilon \{ \tanh \frac{\epsilon}{2T} \text{Re}[(\tilde{\Sigma}_{\epsilon,\epsilon+}^{(2)} + \tilde{\Sigma}_{\epsilon+}^{(2)})g_a(\epsilon) + 2\tilde{\Sigma}_{\epsilon,\epsilon}^{(2)}g_b(\epsilon)] + (\tanh \frac{\epsilon_+}{2T} - \tanh \frac{\epsilon}{2T}) \text{Re}[\tilde{\Sigma}_{\epsilon,\epsilon+}^{(2)}g_c(\epsilon)] \}.$$  \hfill (A.1)

$$K^{(a)}_{vc} = \frac{1}{2} \int d\epsilon \{ \text{Re}[\tilde{\Sigma}_{\epsilon,\epsilon+}^{(a)(2)}g_c(\epsilon)] + i\Sigma^{(a)(2)}_{\epsilon,\epsilon+}\text{Im}[g_c(\epsilon)] \}.$$  \hfill (A.2)

Here, $\tilde{\Sigma}_{\epsilon,\epsilon+}^{(2)}$ and $\Sigma^{(2)}_{\epsilon,\epsilon+}$ respectively satisfy the integral equations.

$$\begin{pmatrix}
\tilde{\Sigma}_{\epsilon,\epsilon+}^{(2)} \\
\Sigma_{\epsilon,\epsilon+}^{(2)} \\
\tilde{\Sigma}_{\epsilon,\epsilon+}^{(a)(2)} \\
\Sigma^{(2)}_{\epsilon,\epsilon+}
\end{pmatrix} = -\frac{1}{4} \int d\epsilon' \tilde{I}(\epsilon, \epsilon', \Omega) \begin{pmatrix}
g_{d}(\epsilon') \\
g_{f}(\epsilon') \\
[g_{d}(\epsilon')]^{*} \\
[g_{f}(\epsilon')]^{*}
\end{pmatrix} - \begin{pmatrix}
(\partial \tilde{n}_{\epsilon'}^{R}/\partial \epsilon' + \partial \tilde{n}_{\epsilon'}^{A}/\partial \epsilon')\tilde{\Sigma}_{\epsilon',\epsilon+}^{(2)}/(\epsilon^{R} - \epsilon_{+}^{R}) \\
(\partial \tilde{n}_{\epsilon'}^{R}/\partial \epsilon' + \partial \tilde{n}_{\epsilon'}^{A}/\partial \epsilon')\tilde{\Sigma}_{\epsilon',\epsilon+}^{(a)(2)}/(\epsilon^{R} - \epsilon_{+}^{A}) \\
(\partial \tilde{n}_{\epsilon'}^{A}/\partial \epsilon' - \partial \tilde{n}_{\epsilon'}^{R}/\partial \epsilon')\Sigma_{\epsilon',\epsilon+}^{(2)}/(\epsilon^{A} - \epsilon_{+}^{A}) \\
(\partial \tilde{n}_{\epsilon'}^{A}/\partial \epsilon' - \partial \tilde{n}_{\epsilon'}^{R}/\partial \epsilon')\Sigma_{\epsilon',\epsilon+}^{(a)(2)}/(\epsilon^{A} - \epsilon_{+}^{R})
\end{pmatrix},$$  \hfill (A.3)
\[ \left( \begin{array}{c} \nu^{R(2)}_{\tau, \epsilon} \\ \nu^{A(2)}_{\tau, \epsilon} \\ \nu^{A(2)}_{\tau, \epsilon} \end{array} \right) = -\frac{1}{4} \int d\epsilon' \hat{I}(\epsilon, \epsilon', 0) \left[ \begin{array}{c} \nu_{\epsilon}^{R} \\ \nu_{\epsilon}^{A} \\ -[\nu_{\epsilon}^{R}]^* \end{array} \right] - \left( \begin{array}{c} \frac{\partial \nu_{\epsilon}^{R}}{\partial \epsilon^{R}} - \frac{\partial \nu_{\epsilon}^{A}}{\partial \epsilon^{A}} \nu_{\epsilon}^{A(2)}_{\tau, \epsilon} \\ \frac{\partial \nu_{\epsilon}^{R}}{\partial \epsilon^{R}} - \frac{\partial \nu_{\epsilon}^{A}}{\partial \epsilon^{A}} \nu_{\epsilon}^{A(2)}_{\tau, \epsilon} \\ \frac{\partial \nu_{\epsilon}^{R}}{\partial \epsilon^{R}} - \frac{\partial \nu_{\epsilon}^{A}}{\partial \epsilon^{A}} \nu_{\epsilon}^{A(2)}_{\tau, \epsilon} \end{array} \right) \].

The above \( g \)'s are written as

\[ g_{a}(\epsilon) = \frac{\nu_{\epsilon}^{R} - \nu_{\epsilon}^{R}}{\epsilon^{R} - \epsilon^{R}} - \frac{\nu_{\epsilon}^{R}}{\epsilon^{R} - \epsilon^{R}}, \]

\[ g_{b}(\epsilon) = \frac{1}{2} \frac{\partial^{2} \nu_{\epsilon}^{R}}{\partial \epsilon^{R}}, \]

\[ g_{c}(\epsilon) = \frac{\nu_{\epsilon}^{R} - \nu_{\epsilon}^{R}}{\epsilon^{R} - \epsilon^{R} - \epsilon^{A}} + \frac{\nu_{\epsilon}^{A}}{\epsilon^{R} - \epsilon^{A}} \]

\[ g_{d}(\epsilon) = \frac{\partial \nu_{\epsilon}^{R}}{\partial \epsilon^{R}} - \frac{\partial \nu_{\epsilon}^{R}}{\partial \epsilon^{R}}, \]

\[ g_{e}(\epsilon) = \frac{-\nu_{\epsilon}^{R} + \nu_{\epsilon}^{A}}{\epsilon^{R} - \epsilon^{A}} + \frac{\partial \nu_{\epsilon}^{R}}{\partial \epsilon^{R}} \]

\[ g_{f}(\epsilon) = \left( \frac{\tanh^{\epsilon_{+}}_{2T}}{2T} - \tanh^{\epsilon_{-}}_{2T} \right) \frac{\partial \nu_{\epsilon}^{R}}{\partial \epsilon^{R}} - \partial \nu_{\epsilon}^{A} / \partial \epsilon^{A} \]

and

\[ \tilde{g}_{c}(\epsilon) = \left( \frac{\tanh^{\epsilon_{+}}_{2T}}{2T} - \tanh^{\epsilon_{-}}_{2T} \right) 2\ii \text{Reg}_{c}(\epsilon). \]

The definitions of \( \nu_{\epsilon}^{R} \) and \( \nu_{\epsilon}^{R} \) are \( \nu_{\epsilon}^{R} = N_{0} \int_{FS} v_{\epsilon}^{2} n_{\epsilon}^{R} \) and \( \nu_{\epsilon}^{R} = N_{0} \int_{FS} n_{\epsilon}^{R} \), respectively.

With use of the boson propagator \( D_{\omega} \) given in §2, the matrix elements of the interaction \( \hat{I}(\epsilon, \epsilon', \Omega) \) are written as \( \hat{I}_{11} = \tanh^{\epsilon_{-}}_{2T}(D_{\epsilon}^{R} - D_{\epsilon}^{A}) + \tanh^{\epsilon_{+}}_{2T}(D_{\epsilon}^{A} - D_{\epsilon}^{R}), \hat{I}_{12} = D_{\epsilon}^{A}, \hat{I}_{13} = 0, \hat{I}_{21} = \tanh^{\epsilon_{-}}_{2T}(D_{\epsilon}^{R} - D_{\epsilon}^{A}) + \tanh^{\epsilon_{+}}_{2T}(D_{\epsilon}^{A} - D_{\epsilon}^{R}), \hat{I}_{22} = D_{\epsilon}^{A}, \hat{I}_{23} = -\tanh^{\epsilon_{-}}_{2T}(D_{\epsilon}^{R} - D_{\epsilon}^{A}) + \tanh^{\epsilon_{+}}_{2T}(D_{\epsilon}^{A} - D_{\epsilon}^{R}), \hat{I}_{31} = \tanh^{\epsilon_{-}}_{2T}(D_{\epsilon}^{R} - D_{\epsilon}^{A}) + \tanh^{\epsilon_{+}}_{2T}(D_{\epsilon}^{A} - D_{\epsilon}^{R}), \hat{I}_{32} = D_{\epsilon}^{A}, \) and \( \hat{I}_{33} = 0 \).

The above expression also indicates that \( K_{b}^{(3)} = 0 \) for \( \Delta_{0} = 0 \), which is same as the term excluding vertex corrections. It is shown that the vertex correction to \( K_{b}^{(3)} \) is absent by considering the symmetry of the vertex \( \hat{v}_{\tau}^{2} \partial v_{\tau}^{2} / \partial k \).
References

22) In the clean limit, there is no gap structure, which is shown by mid-infrared absorption spectroscopy in cuprates. In this paper, we indicate the clean limit as $1/\tau \ll 2\Delta$ and $\xi_0 \ll l \ll \lambda (\tau, \Delta, \xi_0, l, and \lambda$ are the relaxation time, superconducting gap, coherence length, mean free path, and penetration depth, respectively), which holds in cuprates. In conventional superconductors, the definition of the clean limit is different. The clean (dirty) limit indicates $\lambda \ll \xi_0, l (\lambda \gg \xi_0, l)$ and its response is nonlocal (local).