Relaxation Dynamics of Photoinduced Changes in the Superfluid Weight of High-T_c Superconductors

Takanobu JUJO *

Graduate School of Materials Science, Nara Institute of Science and Technology, Ikoma, Nara 630-0101, Japan

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In the transient state of d-wave superconductors, we investigate the temporal variation of photoinduced changes in the superfluid weight. We derive the formula that relates the nonlinear response function to the nonequilibrium distribution function. The latter quantity is obtained by solving the kinetic equation with the electron-electron and the electron-phonon interaction included. By numerical calculations, a nonexponential decay is found at low temperatures in contrast to the usual exponential decay at high temperatures. The nonexponential decay originates from the nonmonotonous temporal variation of the nonequilibrium distribution function at low energies. The main physical process that causes this behavior is not the recombination of quasiparticles as previous phenomenological studies suggested, but the absorption of phonons.

KEYWORDS: nonequilibrium superconductivity, transient state, pump-probe, superfluid weight, vertex correction, unconventional superconductors

1. Introduction

There have been many optical spectroscopic experiments on nonequilibrium superconductors recently, and they have usually been performed by pump-probe techniques. [1–8] As for the interpretation of experimental results, the photoinduced change in reflectivity is often assumed to be directly proportional to the excited nonequilibrium electron density. [1,7,9] Then the temporal evolution of the photoinduced quasiparticle density is analyzed with the use of the Rothwarf-Taylor (RT) equation,[4,5,8] which is first presented in ref. 10. This type of phenomenological equation is claimed to be theoretically derived in refs. 11 and 12 with the use of the kinetic equation for the electron distribution function.

The temporal evolution of the superfluid weight is observed in the experiment with the use of an optical pump and a THz probe,[5] and it is found that the photoinduced change in the superfluid weight decays nonexponentially. The RT equation has been considered to be suitable for describing nonexponential relaxation as the bimolecular relaxation, because this equation includes the quadratic term of the nonequilibrium quasiparticle density as the recombination of the quasiparticles. However, the quadratic term is not allowed in the perturbation expansion

*E-mail address: jujo@ms.aist-nara.ac.jp

of the external field for the reason that it does not satisfy the consistency of the equation with respect to the order of the pump intensity. Therefore, a question arises about the origin of the nonexponential relaxation in the case that the intensity of the pump beam is low.

In this study with regard to this problem, we reexamine two points that have been usually assumed in previous works. One is about the relation between the photoinduced change in optical conductivity and the nonequilibrium quasiparticle density. This relation is not self-evident, but it has not been investigated so far, in addition to the expression of the former quantity itself. The other is about the temporal evolution of the nonequilibrium quasiparticle density. The phenomenological description given by the RT equation[10,13] is based on averaged quasiparticle density. This is also open to doubt for reason that the nonequilibrium quasiparticle density should have a dependence on energy. The kinetic equation is not simply averaged over energy because of the existence of the interaction effect as the kinetic equation in the normal state[14–16] indicates.

To investigate these problems, we microscopically calculate the response function of the transient state. Our calculation shows that the vertex correction term is predominant in photoinduced changes in the superfluid weight, which is same as that in the case of the steady state in a previous study.[17] The nonequilibrium distribution function is related to the response function only through the interaction effect (the vertex correction term). The kinetic equation for this function is solved with the use of the electron-electron and electron-phonon interactions as the collision integral. The numerical calculation shows that, at low temperatures, the photoinduced change in the transient reflectivity is not directly proportional to the number of photoexcited quasiparticles, and the nonexponential decay does not originate from the recombination term, but from the absorption of phonons that enhances the nonequilibrium electron distribution. The effect of nonequilibrium phonons is also considered, and this makes the relaxation dynamics slow as a result of the interaction effect.

2. Response Function in the Transient States

The current under the pump ($A^{\rm pu}$) and probe ($A^{\rm pr}$) beam is written as

$$J^{(3)}(t) = -\int \frac{{\rm d}\omega}{2\pi}\int \frac{{\rm d}\Omega}{2\pi} K^{(3)}(-\omega, -\Omega+\omega/2, \Omega+\omega/2) A^{\rm pu}_{-\Omega+\omega/2} A^{\rm pu}_{\Omega+\omega/2} A^{\rm pr}_{-\omega}.$$

Here, $A^{\rm pu}_{\Omega} = A'_{\rm pu}[{\rm e}^{-\Delta T^2(\Omega-\Omega_0)^2/4} + {\rm e}^{-\Delta T^2(\Omega+\Omega_0)^2/4}]$ and $A^{\rm pr}_{\omega} = {\rm e}^{{\rm i}\omega t} A'_{\rm pr} {\rm e}^{-\Delta t^2\omega^2/4}$ in the case of the Gaussian pulse; $A^{\rm pu}_{\tau} = A_{\rm pu}\cos(\Omega_0\tau){\rm e}^{-(\tau/\Delta T)^2}$ and $A^{\rm pr}_{\tau} = A_{\rm pr}\cos(\omega_0\tau){\rm e}^{-(\tau/\Delta t)^2}$ ($A'_{\rm pu} = A_{\rm pu}\sqrt{\pi}\Delta T/2$ and $A'_{\rm pr} = A_{\rm pr}\sqrt{\pi}\Delta t$). Then $A^{\rm pu}_{\Omega} = \int {\rm d}\tau {\rm e}^{{\rm i}\Omega\tau} A^{\rm pu}_{\tau}$ and $A^{\rm pu}_{\omega} = \int {\rm d}\tau {\rm e}^{{\rm i}\omega\tau} A^{\rm pu}_{\tau-t}$ in which t is the time delay of the probe pulse after the pump excitation. We put $\omega_0 = 0$ to probe the the superfluid weight. The expression of $K^{(3)}$ is given in ref. 18, but the rewritten form is presented below, which clarifies its relation to the nonequilibrium distribution function and its temporal evolution. The integration of $K^{(3)}(-\omega, -\Omega+\omega/2, \Omega+\omega/2)$ by ω and Ω will lead to the temporal variation of the superfluid weight, if possible. However it is difficult to

perform this integration numerically for the reason that the calculation at a very small energy scale is required to determine the variation for a long time. Then we transform the expression of $K^{(3)}$ into a feasible one, as shown below.

We make the following assumptions to calculate the nonlinear response function. The order parameter takes a constant value and is not affected by the external field. The origin of the pairing interaction is not specified (the existence of the superconducting gap is assumed at the outset). The momentum dependence of the vertex correction is weak, which leads to the omission of $\Sigma^{(1)}$ and $\Sigma^{(3)}$ ($\Sigma^{(n)}$ is the self-energy that includes the external fields of the n-th order). We take the local approximation as discussed in ref. 17.

Then the nonlinear response function is written as $K^{(3)} = N_0 \int_{\rm FS} \int {\rm d}\epsilon v_k^4 g^{(3)}_{\epsilon,\epsilon}$. Here, v_k is the quasiparticle velocity, the summation $\sum_k$ is transformed into the integration $N_0 \int_{\rm FS} \int {\rm d}\xi$ (N_0 is the density of states at the Fermi surface), and $g^{(3)}_{\epsilon,\epsilon} = \tanh \frac{\epsilon}{2T}(g^{R(3)}_{\epsilon,\epsilon} - g^{A(3)}_{\epsilon,\epsilon}) + g^{(a)(3)}_{\epsilon,\epsilon}$.[19] $g = \int \frac{{\rm d}\xi}{\pi {\rm i}} G$ (the integration of Green's function G by the electron dispersion ξ), and R, A, and (a) indicate the retarded, advanced, and anomalous parts of Green's function, respectively, as introduced in ref. 20, in which the microscopic formulation for the nonequilibrium superconductors using Green's function is given. (3) indicates the third-order of the external fields, and we represent $\tilde{g}$ (g) as Green's function, which includes (does not include) the external fields H_ω. The predominant term $g^{(a)(3)}_{\epsilon,\epsilon}$ is determined by $\tilde{g}^{(a)(3)}_{\epsilon,\epsilon} = \int \frac{{\rm d}\omega}{2\pi} \int \frac{{\rm d}\Omega}{2\pi} g^{(a)(3)}_{\epsilon,\epsilon} H_{-\Omega+\omega/2} H_{\Omega+\omega/2} H_{-\omega}$, which satisfies the following equation with reference to Éliashberg's formulation.[20]

$$
\begin{aligned}
(\epsilon^R - \epsilon^A)\tilde{g}^{(a)(3)}_{\epsilon,\epsilon} = & \int \frac{{\rm d}\omega}{2\pi}[H_{-\omega}\tilde{g}^{(a)(2)}_{\epsilon,\epsilon-\omega} - H_\omega \tilde{g}^{(a)(2)}_{\epsilon-\omega,\epsilon}] + \int \frac{{\rm d}\omega}{2\pi}[H_{-\omega}(T_{\epsilon-\omega} - T_\epsilon)\tilde{g}^{R(2)}_{\epsilon,\epsilon-\omega} - H_\omega(T_\epsilon - T_{\epsilon-\omega})\tilde{g}^{A(2)}_{\epsilon-\omega,\epsilon}] \\
& + \int \frac{{\rm d}\omega}{2\pi}[\Sigma^{R(2)}_{\epsilon,\epsilon-\omega}\tilde{g}^{(a)(1)}_{\epsilon-\omega,\epsilon} - \tilde{g}^{(a)(1)}_{\epsilon,\epsilon-\omega}\Sigma^{A(2)}_{\epsilon-\omega,\epsilon} + \Sigma^{(a)(2)}_{\epsilon,\epsilon-\omega}\tilde{g}^{A(1)}_{\epsilon-\omega,\epsilon} - \tilde{g}^{R(1)}_{\epsilon,\epsilon-\omega}\Sigma^{(a)(2)}_{\epsilon-\omega,\epsilon}].
\end{aligned}
$$

Here, $T_\epsilon = \tanh \frac{\epsilon}{2T}$, $\epsilon^{R(A)} = \epsilon - \Sigma^{R(A)}_{k,\epsilon}$, and $H_\omega = v_k A_\omega$ with the external field A_ω. The functions $\tilde{g}$ in this equation satisfy similar equations. By taking only the predominant terms into account, $\tilde{g}^{(a)(3)}_{\epsilon,\epsilon}$ is written as

$$
\begin{aligned}
\tilde{g}^{(a)(3)}_{\epsilon,\epsilon} \simeq & \frac{1}{\epsilon^R - \epsilon^A} \int \frac{{\rm d}\omega}{2\pi} \int \frac{{\rm d}\Omega}{2\pi}[R_\omega(\Omega) g^{R(1)}_{\epsilon,\epsilon+\omega} + R^*_{-\omega}(\Omega) g^{A(1)}_{\epsilon-\omega,\epsilon}] H_{-\Omega+\omega/2} H_{\Omega+\omega/2} H_{-\omega} \\
& + \frac{1}{\epsilon^R - \epsilon^A} \int \frac{{\rm d}\omega}{2\pi}[\Sigma^{(a)(2)}_{\epsilon,\epsilon+\omega} g^{A(1)}_{\epsilon+\omega,\epsilon} - g^{R(1)}_{\epsilon,\epsilon+\omega}\Sigma^{(a)(2)}_{\epsilon+\omega,\epsilon}] H_{-\omega}.
\end{aligned}
\tag{1}
$$

Here,

$$
R_\omega(\Omega) = \frac{T_{\epsilon+\omega} - T_{\epsilon-\Omega+\omega/2}}{\epsilon^R - (\epsilon - \Omega + \omega/2)^A} + \frac{T_{\epsilon-\Omega+\omega/2} - T_\epsilon}{\epsilon^R - (\epsilon - \Omega + \omega/2)^R} + \frac{T_{\epsilon+\omega} - T_{\epsilon+\Omega+\omega/2}}{\epsilon^R - (\epsilon + \Omega + \omega/2)^A} + \frac{T_{\epsilon+\Omega+\omega/2} - T_\epsilon}{\epsilon^R - (\epsilon + \Omega + \omega/2)^R}.
$$

To derive the temporal variation of $K^{(3)}$, we consider the following integration with use of some function $f_\omega(\Omega)$;

$$
\int \frac{{\rm d}\omega}{2\pi} \int \frac{{\rm d}\Omega}{2\pi} A^{\rm pu}_{-\Omega+\omega/2} A^{\rm pu}_{\Omega+\omega/2} A^{\rm pr}_{-\omega} f_\omega(\Omega) \simeq \frac{A'^2_{\rm pu} A'_{\rm pr} \exp[-t^2/(\Delta t^2 + \Delta T^2/2)]}{\sqrt{2}\pi \Delta T \sqrt{\Delta t^2 + \Delta T^2/2}} [f_0(\Omega_0) + f_0(-\Omega_0)].
\tag{2}
$$

If the pulse widths Δt and ΔT are several femtoseconds, the effective range of frequency is of the order of the superconducting gap (Δ_0). Here, we consider the case that $\Omega_0 \Delta T \gg 1$, $\Omega_0 \gg \Delta_0$, and the dependences of $f_\omega(\Omega)$ on ω and Ω are weak within $|\omega| < 1/\Delta t$ and $|\Omega \mp \Omega_0| < 1/\Delta T$, respectively. Firstly, we examine the temporal variation of the first term of eq. (1). If we take account of the above consideration and the weak dependence of this term on ω, we can apply the integration eq. (2) to this term. This term is negligible in comparison with the vertex correction term discussed below because of the smallness of $R_0(\Omega_0)$, which is the same as that in the case of ref. 17, in addition to the exponential factor. Next, we consider the second term of eq. (1) (the vertex correction term). By using the relations $\tilde{g}^{R(1)}_{\epsilon,\epsilon+\omega} \simeq H_{-\omega} \partial g^R_{k,\epsilon}/\partial\epsilon$ and $\partial g^A_{k,\epsilon}/\partial\epsilon = -(\partial g^R_{k,\epsilon}/\partial\epsilon)^*$, the vertex correction term is written as

$$\frac{1}{\epsilon^R - \epsilon^A} \int \frac{\mathrm{d}\omega}{2\pi} [\Sigma^{(a)(2)}_{\epsilon,\epsilon+\omega} g^{A(1)}_{\epsilon+\omega,\epsilon} - g^{R(1)}_{\epsilon,\epsilon+\omega} \Sigma^{(a)(2)}_{\epsilon+\omega,\epsilon}] H_{-\omega} \simeq \frac{-2}{\epsilon^R - \epsilon^A} \int \frac{\mathrm{d}\omega}{2\pi} \mathrm{Re} \left(\frac{\partial g^R_{k,\epsilon}}{\partial\epsilon} \right) \Sigma^{(a)(2)}_{k,\epsilon}(\omega) H_{-\omega}.$$

Here, we put $\Sigma^{(a)(2)}_{\epsilon+\omega/2,\epsilon-\omega/2} = \Sigma^{(a)(2)}_{k,\epsilon}(\omega)$, which is a functional of $\tilde{g}^{(a)(2)}_{k,\epsilon}(\omega)$, and its functional form is determined by specifying the interaction and the self-energy. $\tilde{g}^{(a)(2)}_{k,\epsilon}(\omega)$ satisfies the following equation:

$$\begin{aligned}[(\epsilon+\omega/2)^R - (\epsilon-\omega/2)^A] \tilde{g}^{(a)(2)}_{k,\epsilon}(\omega) = \int \frac{\mathrm{d}\Omega}{2\pi} v_k^2 \tilde{R}_{k,\epsilon}(\omega,\Omega) A^{\mathrm{pu}}_{-\Omega+\omega/2} A^{\mathrm{pu}}_{\Omega+\omega/2} - (g^R_{k,\epsilon+\omega/2} - g^A_{k,\epsilon-\omega/2}) \Sigma^{(a)(2)}_{k,\epsilon}(\omega) \\ - (f^R_{k,\epsilon+\omega/2} - f^A_{k,\epsilon-\omega/2}) \Upsilon^{(a)(2)}_{k,\epsilon}(\omega) + (\Upsilon^R_{k,\epsilon+\omega/2} - \Upsilon^A_{k,\epsilon-\omega/2}) \tilde{f}^{(a)(2)}_{k,\epsilon}(\omega). \end{aligned} \tag{3}$$

Here, Υ represents the anomalous (off-diagonal) part of the self-energy, $g^R_{k,\epsilon} = -2\epsilon^R/Z^R_{k,\epsilon}$ and $f^R_{k,\epsilon} = -2\Delta_k/Z^R_{k,\epsilon}$ $\left(Z^R_{k,\epsilon} = \mathrm{sgn}(\epsilon)\sqrt{(\epsilon^R)^2 - \Delta_k^2} \right)$, and $\tilde{R}_{k,\epsilon}(\omega,\Omega) = R_{k,\epsilon}(\omega,\Omega) + R_{k,\epsilon}(\omega,-\Omega)$.

$$\begin{aligned} R_{k,\epsilon}(\omega,\Omega) =& \frac{(T_{\epsilon+\omega/2} - T_{\epsilon+\Omega})(g^R_{k,\epsilon+\omega/2} - g^A_{k,\epsilon+\Omega})}{(\epsilon+\omega/2)^R - (\epsilon+\Omega)^A} + \frac{(T_{\epsilon+\Omega} - T_{\epsilon-\omega/2})(g^R_{k,\epsilon+\omega/2} - g^R_{k,\epsilon+\Omega})}{(\epsilon-\omega/2)^R - (\epsilon+\Omega)^R} \\ & - \frac{(T_{\epsilon+\omega/2} - T_{\epsilon+\Omega})(g^A_{k,\epsilon+\Omega} - g^A_{k,\epsilon-\omega/2})}{(\epsilon+\Omega)^A - (\epsilon-\omega/2)^A} - \frac{(T_{\epsilon+\Omega} - T_{\epsilon-\omega/2})(g^R_{k,\epsilon+\Omega} - g^A_{k,\epsilon-\omega/2})}{(\epsilon+\Omega)^R - (\epsilon-\omega/2)^A}. \end{aligned}$$

This latter quantity is related to the pump-induced term and the initial nonequilibrium distribution function, which directly reflects the values of the self-energy. This induced term vanishes in the case that the self-energy is absent, as indicated in ref. 17.

As shown in eq. (3) the dependence of $\tilde{g}^{(a)(2)}_{k,\epsilon}(\omega)$ (and $\Sigma^{(a)(2)}_{k,\epsilon}(\omega)$) on ω is strong, and this makes the t-dependence of the vertex correction term different from that of the first term of eq. (1) and larger than that. By using the integration $\int \frac{\mathrm{d}\omega}{2\pi} \tilde{g}^{(a)(2)}_{k,\epsilon}(\omega) \mathrm{e}^{-\mathrm{i}\omega t} \mathrm{e}^{-\Delta t^2 \omega^2/4} = \frac{1}{\sqrt{\pi}\Delta t} \int \mathrm{d}\tau \tilde{g}^{(a)(2)}_{k,\epsilon}(\tau) \mathrm{e}^{-(t-\tau)^2/\Delta t^2} \simeq \tilde{g}^{(a)(2)}_{k,\epsilon}(t)$, which is applicable to the time range $t \gg \Delta t$, we can transform eq. (3) into the following kinetic equation, which describes the long time

behavior of $\tilde{g}^{(a)(2)}_{k,\epsilon}(t)$:

$$\begin{aligned}\mathrm{i}\frac{\partial\tilde{g}^{(a)(2)}_{k,\epsilon}(t)}{\partial t} =&\alpha(t)v_k^2\tilde{R}_{k,\epsilon}(0,\Omega_0) + (\Sigma^R_{k,\epsilon}-\Sigma^A_{k,\epsilon})\tilde{g}^{(a)(2)}_{k,\epsilon}(t). - (g^R_{k,\epsilon}-g^A_{k,\epsilon})\Sigma^{(a)(2)}_{k,\epsilon}(t)\\ &+(\Upsilon^R_{k,\epsilon}-\Upsilon^A_{k,\epsilon})\tilde{f}^{(a)(2)}_{k,\epsilon}(t). - (f^R_{k,\epsilon}-f^A_{k,\epsilon})\Upsilon^{(a)(2)}_{k,\epsilon}(t).\end{aligned} \tag{4}$$

Here, $\alpha(t) = \frac{A'^2_{\mathrm{pu}}\exp[-t^2/(\Delta t^2+\Delta T^2/2)]}{\sqrt{2}\pi\Delta T\sqrt{\Delta t^2+\Delta T^2/2}}$. To obtain $K^{(3)}$, the solution of this kinetic equation is substituted into $\Sigma^{(a)(2)}_{k,\epsilon}(t)$ in the following vertex correction term:

$$J^{(3)}_{\mathrm{vc}}(t) = -N_0\int_{\mathrm{FS}}\int \mathrm{d}\epsilon v_k^2\frac{-2}{\epsilon^R-\epsilon^A}\mathrm{Re}\left(\frac{\partial g^R_{k,\epsilon}}{\partial\epsilon}\right)\Sigma^{(a)(2)}_{k,\epsilon}(t)A'_{\mathrm{pr}}. \tag{5}$$

The kinetic equation for nonequilibrium phonons is similarly written as

$$\mathrm{i}\frac{2\omega}{\omega_\phi^2}\frac{\partial D^{(a)(2)}_\omega(t)}{\partial t} = (\Pi^R_\omega-\Pi^A_\omega)D^{(a)(2)}_\omega(t). - (D^R_\omega-D^A_\omega)\Pi^{(a)(2)}_\omega(t). \tag{6}$$

Here, $D^{R,(A)}_\omega$ and $\Pi^{R,(A)}_\omega$ are the retarded (advanced) phonon Green's function and the self-energy by phonon-electron interaction, respectively. (a) and (2) indicate the anomalous part and the order of the external field, respectively, which is same as that in the case of electrons.

3. Kinetic Equation of the Nonequilibrium Distribution Function

In this section, we derive the kinetic equations for the distribution functions of electrons and phonons. We put the deviation of the distribution function from the equilibrium state by the pump excitation as $\delta n_\epsilon(t)$ and $\delta N_{\omega_\phi}(t)$ for electrons and phonons, respectively. We consider a two-dimensional system and replace $\int_{\mathrm{FS}}$ by $\int\frac{\mathrm{d}\varphi}{2\pi}$. $v_k = v_{\mathrm{F}}\cos\varphi$ (v_{F} is the Fermi velocity) and $\Delta_k = \Delta_0\cos 2\varphi$ for d-wave superconductors.

We adopt the second-order perturbation expansion for the electron-electron interaction and the one-loop approximation for the electron-phonon interaction as the self-energy. We rewrite the nonequilibrium Green function as $\tilde{g}^{(a)(2)}_{\varphi,\epsilon}(t) = -2\delta n_\epsilon(t)(g^R_{\varphi,\epsilon}-g^A_{\varphi,\epsilon})$, $\tilde{f}^{(a)(2)}_{\varphi,\epsilon}(t) = -2\delta n_\epsilon(t)(f^R_{\varphi,\epsilon}-f^A_{\varphi,\epsilon})$, and $D^{(a)(2)}_\omega(t) = 2\delta N_\omega(t)(D^R_\omega-D^A_\omega)$ with the use of $D^R_\omega-D^A_\omega = -\pi\mathrm{i}\omega[\delta(\omega-\omega_\phi)+\delta(\omega+\omega_\phi)]$ (for example, see ref. 21). We consider acoustic phonons and put $\omega_\phi = v_{\mathrm{s}}k_{\mathrm{F}}|\phi|$ (v_{s} is the sound velocity). Then the kinetic equation for electrons, eq. (4), is written as

$$\begin{aligned}\frac{\partial\delta n_\epsilon(t)}{\partial t} =&\frac{1}{\bar{g}^R_\epsilon}\frac{\alpha(t)}{4\mathrm{i}}\int\frac{\mathrm{d}\varphi}{2\pi}v_\varphi^2\tilde{R}_{\varphi,\epsilon}(0,\Omega_0) - \frac{1}{\bar{g}^R_\epsilon}\frac{\pi}{2}N_0g^2\int_{-\phi_D}^{\phi_D}\frac{\mathrm{d}\phi}{2\pi}\omega_\phi I^{\mathrm{el-ph}}_{\phi,\epsilon}[\delta n,\delta N]\\ &-\frac{1}{\bar{g}^R_\epsilon}\frac{\pi}{2}\frac{U^2N_0^2}{v_{\mathrm{F}}k_{\mathrm{F}}}\iiint\frac{\mathrm{d}\varphi\mathrm{d}\varphi_1\mathrm{d}\varphi_2}{(2\pi)^3}\iint\mathrm{d}\epsilon_1\mathrm{d}\epsilon_2\tilde{\delta}_{\varphi,\varphi_1,\varphi_2}I^{\mathrm{el-el}}_{\epsilon,\varphi;\epsilon_1,\varphi_1;\epsilon_2,\varphi_2}[\delta n].\end{aligned} \tag{7}$$

Here, $\bar{g}_\epsilon := -\int\frac{\mathrm{d}\varphi}{2\pi}\mathrm{Re}g^R_\varphi(\epsilon)$ and $\tilde{\delta}_{\varphi,\varphi_1,\varphi_2} := \delta[1-\cos(\varphi-\varphi_1)+\cos(\varphi-\varphi_2)-\cos(\varphi_1-\varphi_2)]$ is a delta function. U is the effective short-range Coulomb repulsion energy, which is called the on-site Coulomb interaction in the Hubbard model. g is the coefficient of the electron-phonon interaction, which is usually written as $g = \sqrt{\hbar/2M_{\mathrm{i}}v_{\mathrm{s}}}V_{\mathrm{i}}$ (M_{i} is the mass of the

ion and $V_{\rm i}$ is the renormalized ion potential). In the next section, we use UN_0 and g^2N_0 as dimensionless parameters that characterize the strengths of the electron-electron and electron-phonon interactions, respectively. The collision terms for the electron-electron and electron-phonon interactions are written as

$$I^{\rm el-el}_{\epsilon,\varphi;\epsilon_1,\varphi_1;\epsilon_2,\varphi_2}[\delta n] = \frac{{\rm Re}g^R_{\varphi,\epsilon}{\rm Re}g^R_{\varphi_1,\epsilon_1}{\rm Re}g^R_{\varphi_2,\epsilon_2}{\rm Re}g^R_{\varphi-\varphi_1+\varphi_2,\epsilon-\epsilon_1+\epsilon_2}}{\cosh\frac{\epsilon}{2T}\cosh\frac{\epsilon_1}{2T}\cosh\frac{\epsilon_2}{2T}\cosh\frac{\epsilon-\epsilon_1+\epsilon_2}{2T}} \times\left[(\cosh\tfrac{\epsilon}{2T})^2\delta n_\epsilon(t)-(\cosh\tfrac{\epsilon_1}{2T})^2\delta n_{\epsilon_1}(t)+(\cosh\tfrac{\epsilon_2}{2T})^2\delta n_{\epsilon_2}(t)-(\cosh\tfrac{\epsilon-\epsilon_1+\epsilon_2}{2T})^2\delta n_{\epsilon-\epsilon_1+\epsilon_2}(t)\right],$$

and

$$\begin{aligned} I^{\rm el-ph}_{\phi,\epsilon}[\delta n,\delta N] =& \left(\coth\tfrac{\omega_\phi}{2T}+\tanh\tfrac{\epsilon-\omega_\phi}{2T}\right)g^-_{\phi,\epsilon}\delta n_\epsilon(t)+\left(\coth\tfrac{\omega_\phi}{2T}-\tanh\tfrac{\epsilon+\omega_\phi}{2T}\right)g^+_{\phi,\epsilon}\delta n_\epsilon(t)\\ &-\left(\coth\tfrac{\omega_\phi}{2T}-\tanh\tfrac{\epsilon}{2T}\right)g^-_{\phi,\epsilon}\delta n_{\epsilon-\omega_\phi}(t)-\left(\coth\tfrac{\omega_\phi}{2T}+\tanh\tfrac{\epsilon}{2T}\right)g^+_{\phi,\epsilon}\delta n_{\epsilon+\omega_\phi}(t)\\ &+\left(\tanh\tfrac{\epsilon-\omega_\phi}{2T}-\tanh\tfrac{\epsilon}{2T}\right)g^-_{\phi,\epsilon}\delta N_\phi(t)+\left(\tanh\tfrac{\epsilon+\omega_\phi}{2T}-\tanh\tfrac{\epsilon}{2T}\right)g^+_{\phi,\epsilon}\delta N_\phi(t). \end{aligned}$$

Here, $g^\mp_{\phi,\epsilon}:=\int\frac{{\rm d}\varphi}{2\pi}\left[{\rm Re}g^R_{\varphi-\phi}(\epsilon\mp\omega_\phi){\rm Re}g^R_\phi(\epsilon)-{\rm Re}f^R_{\varphi-\phi}(\epsilon\mp\omega_\phi){\rm Re}f^R_\phi(\epsilon)\right]$.

The kinetic equation for nonequilibrium phonons, eq. (6), is written as

$$\frac{\partial\delta N_{\omega_\phi}}{\partial t}=-\gamma_{\rm esc}\delta N_{\omega_\phi}+\frac{v_{\rm s}}{v_{\rm F}}\frac{1}{2}N_0g^2\int_0^\infty{\rm d}\epsilon I^{\rm ph-el}_{\phi,\epsilon}[\delta n_\epsilon,\delta N_\phi].$$

Here, we add a phenomenological term, $\gamma_{\rm esc}\delta N_{\omega_\phi}$ (damping by phonon escape), which describes the equilibration between the electron-phonon system and the reservoir.

$$\begin{aligned} I^{\rm ph-el}_{\phi,\epsilon}[\delta n_\epsilon,\delta N_\phi] =& \left(\coth\tfrac{\omega_\phi}{2T}+\tanh\tfrac{\epsilon-\omega_\phi}{2T}\right)g^-_{\phi,\epsilon}\delta n_\epsilon(t)-\left(\coth\tfrac{\omega_\phi}{2T}-\tanh\tfrac{\epsilon+\omega_\phi}{2T}\right)g^+_{\phi,\epsilon}\delta n_\epsilon(t)\\ &-\left(\coth\tfrac{\omega_\phi}{2T}-\tanh\tfrac{\epsilon}{2T}\right)g^-_{\phi,\epsilon}\delta n_{\epsilon-\omega_\phi}(t)+\left(\coth\tfrac{\omega_\phi}{2T}+\tanh\tfrac{\epsilon}{2T}\right)g^+_{\phi,\epsilon}\delta n_{\epsilon+\omega_\phi}(t)\\ &+\left(\tanh\tfrac{\epsilon-\omega_\phi}{2T}-\tanh\tfrac{\epsilon}{2T}\right)g^-_{\phi,\epsilon}\delta N_\phi(t)-\left(\tanh\tfrac{\epsilon+\omega_\phi}{2T}-\tanh\tfrac{\epsilon}{2T}\right)g^+_{\phi,\epsilon}\delta N_\phi(t). \end{aligned}$$

The energy conservation is also discussed using the kinetic equations. The additional energy induced by the external field in the system of electrons and phonons is given by

$$2\sum_k E_k\delta n_{E_k}(t)+\sum_q\omega_q\delta N_{\omega_q}(t)=2N_0\int\frac{{\rm d}\varphi}{2\pi}\int{\rm d}\epsilon\frac{|\epsilon|}{\sqrt{\epsilon^2-\Delta^2_\varphi}}\epsilon\delta n_\epsilon(t)+\frac{N_0}{2}\int_{-\phi_D}^{\phi_D}\frac{{\rm d}\phi}{2\pi}2\pi v_{\rm F}k_{\rm F}|\phi|\omega_\phi\delta N_\phi(t).$$

With the use of the kinetic equation, it is shown that this quantity is equal to the energy injected by the external field, which is written as

$$\int_{-\infty}^t{\rm d}t'N_0\int{\rm d}\epsilon\int\frac{{\rm d}\varphi}{2\pi}\frac{1}{4{\rm i}}\alpha(t')v^2_\varphi\tilde{R}_{\varphi,\epsilon}(0,\Omega_0).$$

In the above discussion, we put $\gamma_{\rm esc}=0$ for the energy conservation. In the case of $\gamma_{\rm esc}\neq0$, the energy of the electron-phonon system dissipates into the reservoir system.

The solution of the kinetic equations is related to the nonlinear response function as follows. $\delta n_\epsilon(t)$ and $\delta N_\omega(t)$ obtained by solving the kinetic equations is substituted to $\Sigma^{(a)(2)}_{\varphi,\epsilon}(t)$

of the previous section as $\Sigma^{(a)(2)}_{\varphi,\epsilon}(t) = \Sigma^{(a)\mathrm{el-el}}_{\varphi,\epsilon}(t) + \Sigma^{(a)\mathrm{el-ph}}_{\varphi,\epsilon}(t)$. Here,

$$\Sigma^{(a)\mathrm{el-el}}_{\varphi,\epsilon}(t) = -\pi \mathrm{i}\frac{U^2N_0^2}{v_\mathrm{F}k_\mathrm{F}}\iint\frac{\mathrm{d}\varphi_1\mathrm{d}\varphi_2}{(2\pi)^2}\iint \mathrm{d}\epsilon_1\mathrm{d}\epsilon_2\tilde{\delta}_{\varphi,\varphi_1,\varphi_2}\frac{\mathrm{Re}g^R_{\varphi_1,\epsilon_1}\mathrm{Re}g^R_{\varphi_2,\epsilon_2}\mathrm{Re}g^R_{\varphi-\varphi_1+\varphi_2,\epsilon-\epsilon_1+\epsilon_2}}{\cosh\frac{\epsilon}{2T}\cosh\frac{\epsilon_1}{2T}\cosh\frac{\epsilon_2}{2T}\cosh\frac{\epsilon-\epsilon_1+\epsilon_2}{2T}}$$
$$\times\left[(\cosh\tfrac{\epsilon_1}{2T})^2\delta n_{\epsilon_1}(t) - (\cosh\tfrac{\epsilon_2}{2T})^2\delta n_{\epsilon_2}(t) + (\cosh\tfrac{\epsilon-\epsilon_1+\epsilon_2}{2T})^2\delta n_{\epsilon-\epsilon_1+\epsilon_2}(t)\right],$$

and

$$\Sigma^{(a)\mathrm{el-ph}}_{\varphi,\epsilon}(t) = -\pi\mathrm{i}N_0g^2\int_{-\phi_D}^{\phi_D}\frac{\mathrm{d}\phi}{2\pi}\omega_\phi\{\mathrm{Re}g^R_{\varphi-\phi,\epsilon-\omega_\phi}[\left(\coth\tfrac{\omega_\phi}{2T}-\tanh\tfrac{\epsilon}{2T}\right)\delta n_{\epsilon-\omega_\phi}(t) - \left(\tanh\tfrac{\epsilon-\omega_\phi}{2T}-\tanh\tfrac{\epsilon}{2T}\right)\delta N_\phi(t)]$$
$$+\mathrm{Re}g^R_{\varphi-\phi,\epsilon+\omega_\phi}[\left(\coth\tfrac{\omega_\phi}{2T}+\tanh\tfrac{\epsilon}{2T}\right)\delta n_{\epsilon+\omega_\phi}(t) - \left(\tanh\tfrac{\epsilon+\omega_\phi}{2T}-\tanh\tfrac{\epsilon}{2T}\right)\delta N_\phi(t)]\}.$$

Then this $\Sigma^{(a)(2)}_{\varphi,\epsilon}(t)$ is substituted into eq. (5), which gives the nonlinear response function.

4. Results

The results of the numerical calculation are shown below. The superconducting gap Δ_0 is taken as the unit of energy $\Delta_0 = 1.0$ (this leads to the superconducting transition temperature $T_\mathrm{c} = 0.465$), and we put $\omega_\phi \le v_\mathrm{s}k_\mathrm{F}$. We fix the values of several parameters as follows: $\Omega_0 = 6.0$,[22] $v_\mathrm{s}k_\mathrm{F} = 2.0372$, $v_\mathrm{s}/v_\mathrm{F} = 0.05$ (which leads to the Fermi energy $E_\mathrm{F} = v_\mathrm{F}k_\mathrm{F}/2 \simeq 20$), $UN_0 = 0.2$, and $g^2N_0 = 0.05$ (this value corresponds to $\lambda = 0.1$ as the electron-phonon coupling constant). We add a small $\delta = -0.01$ to the imaginary part of the self-energy as effective impurities and a finite mean free path.

In the calculation of the kinetic equation eq. (7), the following approximation is used to make a multiple integration in the electron-electron collision term a feasible one: $\iiint\frac{\mathrm{d}\varphi\mathrm{d}\varphi_1\mathrm{d}\varphi_2}{(2\pi)^3}\tilde{\delta}_{\varphi,\varphi_1,\varphi_2}\mathrm{Re}g^R_{\varphi,\epsilon}\mathrm{Re}g^R_{\varphi_1,\epsilon_1}\mathrm{Re}g^R_{\varphi_2,\epsilon_2}\mathrm{Re}g^R_{\varphi-\varphi_1+\varphi_2,\epsilon-\epsilon_1+\epsilon_2} \to \mathrm{Re}\bar{g}^R_\epsilon\mathrm{Re}\bar{g}^R_{\epsilon_1}\mathrm{Re}\bar{g}^R_{\epsilon_2}\mathrm{Re}\bar{g}^R_{\epsilon-\epsilon_1+\epsilon_2}$. We perform the same approximation for $\Sigma^{(a)\mathrm{el-el}}_{\varphi,\epsilon}$. This leads to violation of the momentum conservation, although the energy conservation is satisfied. The numerical calculation indicates that, by this replacement, the interaction effect is underestimated at low energies ($|\epsilon| < \Delta_0$) owing to averaging the angular dependence. However, the quantitative difference is small especially at $|\epsilon| > \Delta_0$. Then it is not considered that this approximation causes qualitative changes for numerical results.

Below we consider two typical cases for a nonequilibrium phonon system. The numerical calculation of $K^{(3)}$ shows that calculations with large values of the damping γ_esc lead to results similar to those in the case of $\delta N = 0$ (phonons in thermal equilibrium). On the other hand, the results with small values of γ_esc are approximated by those for $\gamma_\mathrm{esc} = 0$. Therefore, we show the calculated results only for $\delta N = 0$ and $\delta N \neq 0$ with $\gamma_\mathrm{esc} = 0.0$ as characteristic cases of the phonon system.

To solve the kinetic equations, we consider the square pulse and put $\alpha(t) = 1$ for $0 < t < 0.25$ and $\alpha(t) = 0$ otherwise. (The long time behavior is not affected if we use a Gaussian pulse.) Then we redefine $K^{(3)}$ as $K^{(3)} = -J^{(3)}_\mathrm{vc}(t)/A'_\mathrm{pr}$ from eq. (5) with the use of this $\alpha(t)$, and this is the photoinduced change in the superfluid weight. (We can put $J^{(3)}(t) \simeq J^{(3)}_\mathrm{vc}(t)$

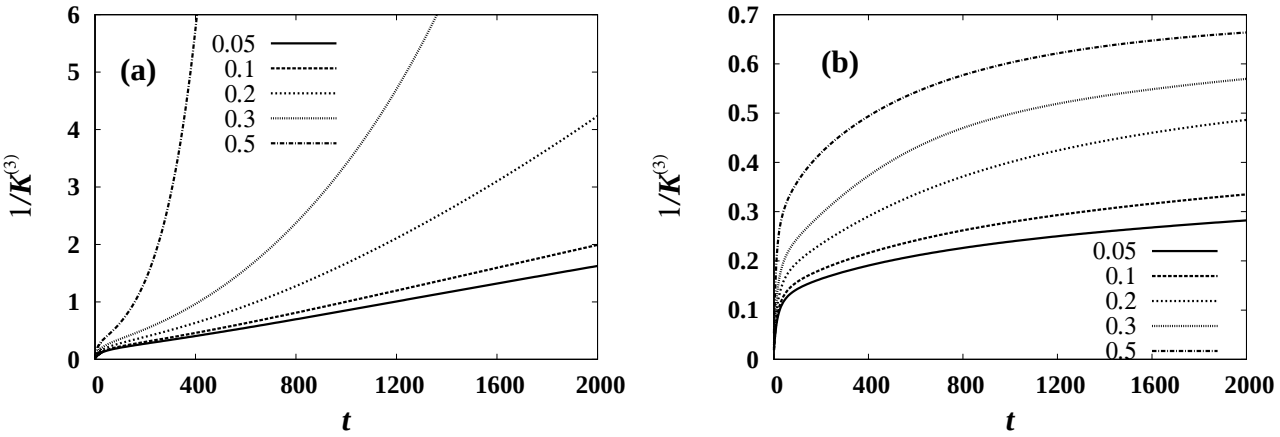

Fig. 1. Dependences of $1/K^{(3)}$ on time for various values of T/T_c (numbers in the figure). The phonons are supposed to be (a) in thermal equilibrium ($\delta N = 0$), and (b) in nonequilibrium state ($\delta N \neq 0$).

as noted above.) If we use only $\Sigma^{(a)\mathrm{el-el}}$ ($\Sigma^{(a)\mathrm{el-ph}}$) as $\Sigma^{(a)(2)}$ in this equation, we write this quantity as $K^{(3)\mathrm{el-el}}$ ($K^{(3)\mathrm{el-ph}}$) below.

The temporal variation of $1/K^{(3)}$ is shown in Fig. 1. If we put $\Delta_0 = 30$ meV, $t = 1000$ corresponds to about 22 ps. This is comparable to the range of time taken in the experiments,[5] and we present the numerical results for this range hereafter. $K^{(3)}$ shows a nonexponential decay for low T/T_c, and it becomes an exponential decay for high T/T_c as in the experimental result.[5] Here, the nonexponential decay and exponential decay indicate $1/K^{(3)} \propto 1 + \gamma t$ and $1/K^{(3)} \propto \mathrm{e}^{\gamma t}$, respectively. At first sight, the former seems to be an approximation of the latter using $\mathrm{e}^{\gamma t} \simeq 1 + \gamma t$ with $\gamma t \ll 1$, but this is not the case. For example, at $T/T_c = 0.05$, $1/K^{(3)}$ is fitted to $0.1(1 + 0.0076t)$ ($0.0076t \gg 1$ for large t). Therefore, some qualitative differences exist between low and high T/T_c. It is shown below that this difference in relaxation dynamics arises from the temporal variation of the nonequilibrium quasiparticles (δn_ϵ), and this traces back to the change of the predominant physical process in the collision integral. In the case of $\delta N \neq 0$, the relaxation becomes slower than that in the case of $\delta N = 0$. As shown below this originates from the existence of δN, which directly affects $K^{(3)}$ through the electron-phonon interaction, in addition to the slow relaxation of nonequilibrium electrons. If we vary $g^2 N_0$, we obtain qualitatively the same results with regard to the t- and T-dependences of $1/K^{(3)}$, but there are several quantitative differences. In the case of $\delta N = 0$ $1/K^{(3)}$ increases, and the exponential decay of $1/K^{(3)}$ is observed at lower T/T_c with increasing $g^2 N_0$. This is because the equilibration between electrons and phonons becomes faster, and δn rapidly decreases. On the other hand, in the case of $\delta N \neq 0$, the absolute value of $1/K^{(3)}$ decreases with increasing $g^2 N_0$. In this case, the nonequilibrium electrons remain finite, and the interaction effect directly enhances $K^{(3)}$.

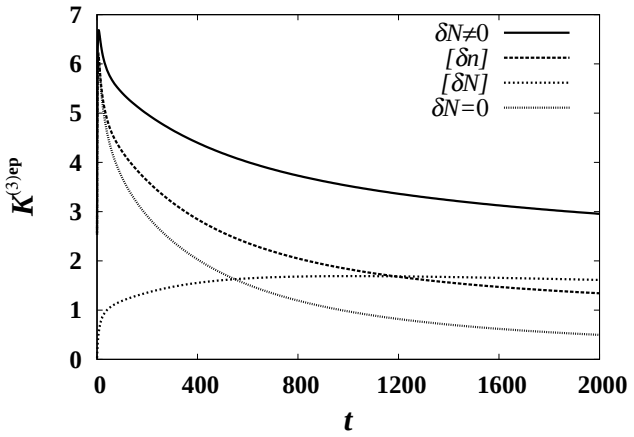

Fig. 2. Dependences of $K^{(3)\mathrm{el-ph}}$ on time for $T/T_\mathrm{c} = 0.1$. $\delta N = 0$ and $\delta N \neq 0$ indicate the results of $K^{(3)\mathrm{el-ph}}$ in the cases of $\delta N = 0$ and $\delta N \neq 0$, respectively. $[\delta n]$ and $[\delta N]$ are components of $K^{(3)\mathrm{el-ph}}$ for $\delta N \neq 0$ and include only δn and δN in $\Sigma^{(a)\mathrm{el-ph}}_{\varphi,\epsilon}$, respectively.

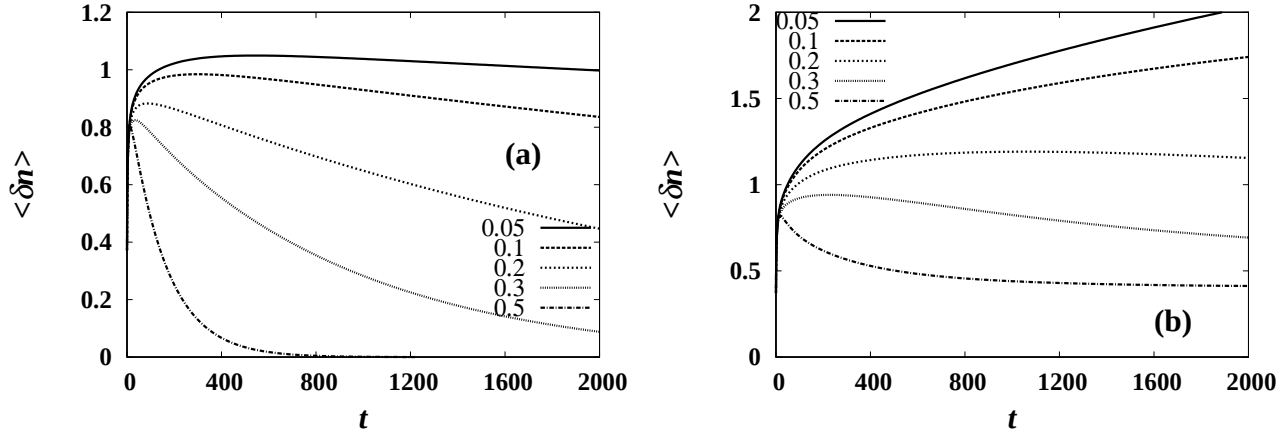

Fig. 3. Dependences of $\langle \delta n \rangle$ on time for various values of T/T_c (numbers in the figure). The phonons are supposed to be (a) in thermal equilibrium ($\delta N = 0$) and (b) in a nonequilibrium state ($\delta N \neq 0$).

To clarify the effect of nonequilibrium phonons on $K^{(3)}$, we decompose $K^{(3)\mathrm{el-ph}}$ to contributions from nonequilibrium electrons and phonons by taking only the δn term or δN term in $\Sigma^{(a)\mathrm{el-ph}}_{\varphi,\epsilon}$. The temporal variations of these terms are shown in Fig. 2. This result indicates that the δN term has a sufficient contribution to $K^{(3)}$ at a large t. This makes the relaxation slower than that in the case of $\delta N = 0$.

Here, we examine the question as to whether the nonlinear response is proportional to the nonequilibrium electron density. The temporal variation of the integrated nonequilibrium electron density, which is written as $\langle \delta n_\epsilon(t) \rangle = \int_0^\infty \mathrm{d}\epsilon \int_\mathrm{FS} \frac{2\epsilon}{\sqrt{\epsilon^2 - \Delta_\varphi^2}} \delta n_\epsilon(t)$, is shown in Fig. 3. In the case of $\delta N = 0$, $\langle \delta n \rangle$ shows the exponential decay for high T/T_c, as $K^{(3)}$ does. For low T/T_c, $\langle \delta n \rangle$ does not show the same t-dependence as $K^{(3)}$. Therefore, in the region of

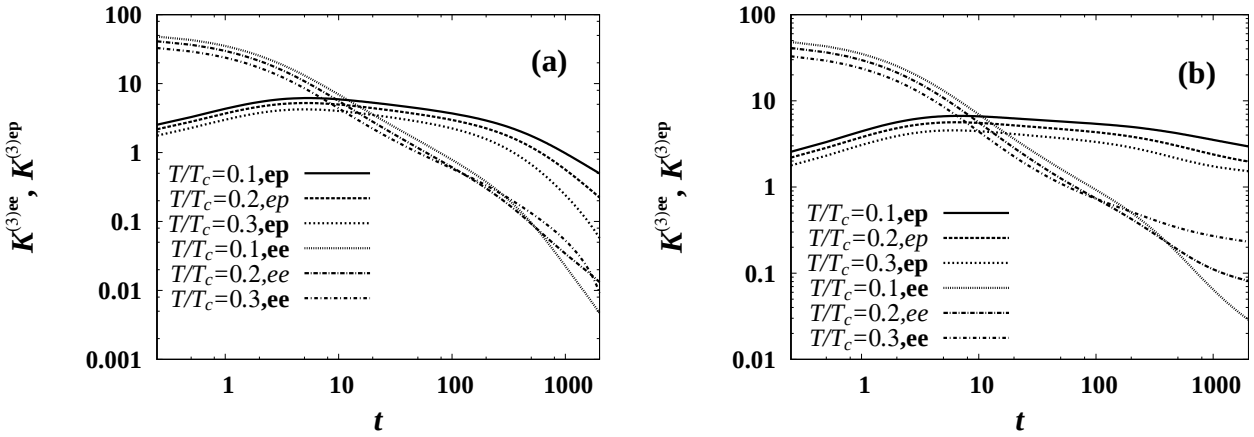

Fig. 4. Dependences of $K^{(3)\mathrm{el-el}}$ (ee) and $K^{(3)\mathrm{el-ph}}$ (ep) on time for various values of T/T_c. The phonons are supposed to be (a) in thermal equilibrium ($\delta N = 0$) and (b) in a nonequilibrium state ($\delta N \neq 0$).

the nonexponential decay, there is no proportionality between the nonlinear response and nonequilibrium electron density, which is different from the assumption adopted in previous studies. The similarity in relaxation dynamics between these two quantities is limited to the region of the exponential decay. In the case of $\delta N \neq 0$, $\langle \delta n \rangle$ does not decrease to 0, but it varies toward the nonequilibrium steady state. This state is rapidly achieved for high T/T_c as for $\langle \delta n \rangle$. The increase of $\langle \delta n \rangle$ with t at low T/T_c is understood by considering the temporal evolution of $\delta n_\epsilon(t)$ (shown in Fig. 7) and the collision integral $\tilde{I}_\epsilon(t)$ (in Figs. 6 and 9). As discussed there, the distribution of nonequilibrium electrons is enhanced at low energies by the absorption of phonons. (The particle conservation is satisfied because $\delta n_{-\epsilon} = -\delta n_\epsilon$ in our calculation.)

Next, we investigate which of the electron-electron and electron-phonon interactions predominate in the nonlinear response. The temporal variations of $K^{(3)\mathrm{el-ph}}$ and $K^{(3)\mathrm{el-el}}$ are shown in Fig. 4. For small (large) t, $K^{(3)\mathrm{el-el}}$ ($K^{(3)\mathrm{el-ph}}$) is predominant in $K^{(3)}$. This change of the predominant term originates from the temporal variation of the functional form of $\delta n_\epsilon(t)$. $\delta n_\epsilon(t)$ has a broad spectrum as a function of ϵ at small t, and its spectrum concentrates at low energies as time passes, as discussed below. The time at which $K^{(3)\mathrm{el-el}}$ and $K^{(3)\mathrm{el-ph}}$ intersect is weakly dependent on whether phonons are in the equilibrium or nonequilibrium state, which indicates that the effect of nonequilibrium phonons is small in this time range. Although $K^{(3)\mathrm{el-ph}}$ decreases with increasing T/T_c, $K^{(3)\mathrm{el-el}}$ shows a different behavior at large t, which also reflects the ϵ-dependence of $\delta n_\epsilon(t)$, but this behavior is not important because $K^{(3)\mathrm{el-el}} \ll K^{(3)\mathrm{el-ph}}$ at this time scale.

Hereafter, we consider the microscopic quantities that cause the above behavior of the nonlinear response. Firstly, we show the temporal variation of the collision integral, which is

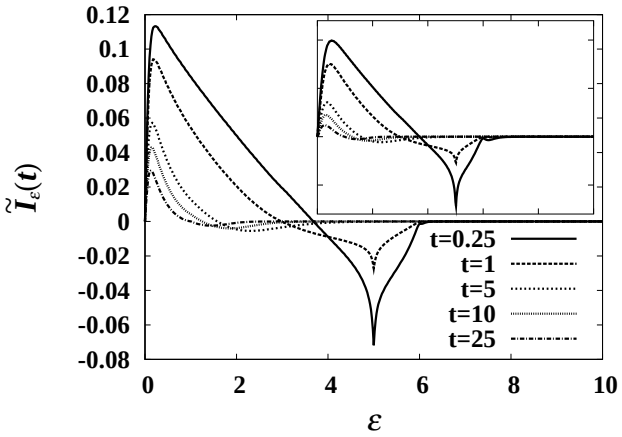

Fig. 5. Collision integrals $\tilde{I}_\epsilon(t)$ for various values of t. $T/T_c = 0.1$. The phonons are supposed to be in thermal equilibrium ($\delta N = 0$). The results at $T/T_c = 0.3$ are shown in the inset.

the second and third terms on the right side of eq. (7) and written as $\tilde{I}_\epsilon(t) = \tilde{I}_\epsilon^{\rm el-ph}(t) + \tilde{I}_\epsilon^{\rm el-el}(t)$ hereafter. As indicated above in the cases of $K^{(3){\rm el-el}}$ and $K^{(3){\rm el-ph}}$, in the collision integral $\tilde{I}_\epsilon(t)$, the electron-electron interaction predominates the relaxation dynamics over the electron-phonon interaction (roughly $\tilde{I}_\epsilon(t) \simeq \tilde{I}_\epsilon^{\rm el-el}(t)$) at small t, and it is the other way around ($\tilde{I}_\epsilon(t) \simeq \tilde{I}_\epsilon^{\rm el-ph}(t)$) at large t. This holds irrespective of the values of T/T_c and the existence of δN. We show the collision integral $\tilde{I}_\epsilon(t)$ (only for $\epsilon \geq 0$ because of $\tilde{I}_{-\epsilon}(t) = -\tilde{I}_\epsilon(t)$) at small t in Fig. 5 and at large t in Fig. 6. For small t, there seems to be no qualitative difference in the ϵ-dependence of $\tilde{I}_\epsilon$ between $T/T_c = 0.1$ and 0.3. The energy range in which $\tilde{I}_\epsilon$ takes finite values is broad, and it takes negative and positive values at high and low energies, respectively. This brings about a shift in the weight of the nonequilibrium distribution function from high energy to low energy. The results in the case of $\delta N \neq 0$ are omitted here, but they are similar to those in the case of $\delta N = 0$. In contrast to that in the case of small t, ϵ-dependences of $\tilde{I}_\epsilon$ are different depending on the values of T/T_c and δN for large t. For $T/T_c = 0.3$, $\tilde{I}_\epsilon$ becomes negative all over the range of $\epsilon > 0$. This property is the same in the case of $\delta N \neq 0$, although its degree is small. For $T/T_c = 0.1$, there remains a positive part in $\tilde{I}_\epsilon$ at low energy. The shift in the weight of the nonequilibrium distribution function from high energy to low energy occurs, as in the case of small t, but its energy scale becomes narrower. The result for $\delta N \neq 0$ shows that the negative part in $\tilde{I}_\epsilon$ is small and that the decreasing rate of the positive part is slower than that of $\delta N = 0$.

The nonequilibrium distribution functions for electrons, $\delta n_\epsilon(t)$, at various times t for $T/T_c = 0.1$ and 0.3 in the case of $\delta N = 0$ are shown in Fig. 7. (We consider the particle-hole symmetric case: $\delta n_{-\epsilon} = -\delta n_\epsilon$.) At small t, immediately after the pump excitation, $\delta n_\epsilon(t)$ has a broad spectrum. The spectrum at high energy rapidly decreases with time. This is because the damping effect is large at high energy owing to the electron-electron interaction. Then

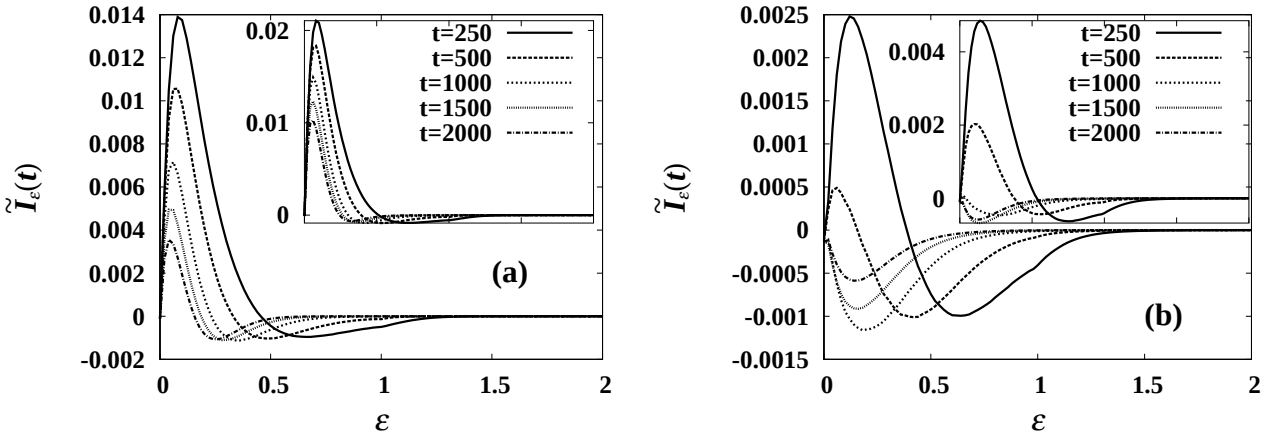

Fig. 6. Collision integrals $\tilde{I}_\epsilon(t)$ for various values of t. (a) $T/T_c = 0.1$ and (b) $T/T_c = 0.3$. The phonons are supposed to be in thermal equilibrium ($\delta N = 0$). The results in the case of $\delta N \neq 0$ are shown in the inset.

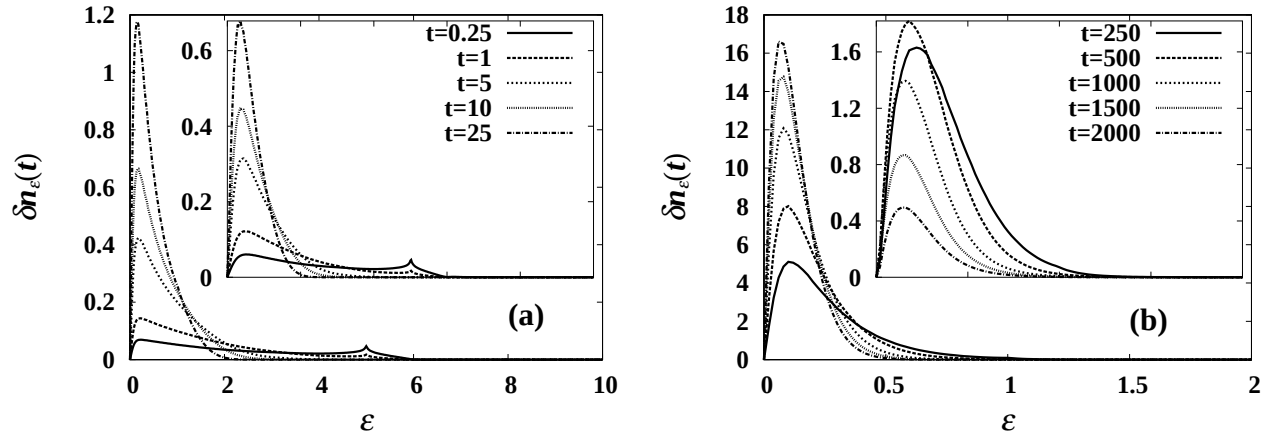

Fig. 7. Nonequilibrium distribution function $\delta n_\epsilon(t)$ (a) at small values of t and (b) at large values of t. $T/T_c = 0.1$. The results for $T/T_c = 0.3$ are shown in the inset. The phonons are supposed to be in thermal equilibrium ($\delta N = 0$).

$\delta n_\epsilon(t)$ takes a functional form, which is similar to that of the T^*-model:[23] $\frac{1}{e^{\epsilon/T^*}+1} - \frac{1}{e^{\epsilon/T}+1} \simeq \frac{\epsilon(T^*-T)}{[2T\cosh(\epsilon/2T)]^2}$, where T^* is the temperature that characterizes nonequilibrium electrons and $T^* > T$. For $T/T_c = 0.1$, the shift of the spectrum from high energy to low energy occurs. (This indicates a nonthermal state, as discussed in ref. 14, with taking account of a change in the sign of $\partial\delta n_\epsilon/\partial t \simeq \tilde{I}_\epsilon^{\mathrm{el-ph}}$ for large t.) On the other hand, $\delta n_\epsilon(t)$ starts to decrease all over the range of $\epsilon > 0$ after a certain t for $T/T_c = 0.3$. This behavior is understood by examining the ϵ-dependences of the collision integral, as shown above. For $\delta N \neq 0$, these tendencies are qualitatively the same, which is also indicated in the results of the collision integral.

The energy injected by a pump beam is transferred to the phonon system via the electron

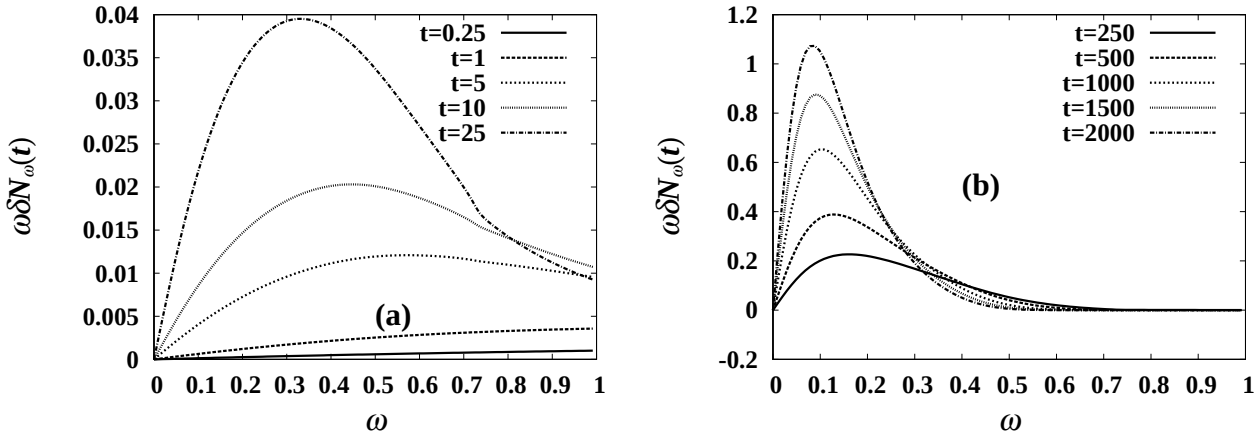

Fig. 8. $\omega\delta N_\omega(t)$ (a) at small values of t and (b) at large values of t. $T/T_c = 0.1$.

system. In the case of $\delta N = 0$, we presume that this energy transferred to the phonon system dissipates by the damping effect $\gamma_{\rm esc}$ and that there is no influence on the electron system by δN. On the other hand, δN is finite if the effect of $\gamma_{\rm esc}$ is small. The distributions of nonequilibrium phonons $\omega\delta N_\omega(t)$ at various values of t for $T/T_c = 0.1$ are shown in Fig. 8. The tendency in the changes of the spectrum is similar to that observed in nonequilibrium electrons. However, the decrease in the spectrum does not occur in a phonon system, and nonequilibrium phonons accumulate at small ω with increasing t. For high T/T_c, δN shows a similar but broader spectrum than that for low T/T_c.

Finally, we investigate which of the physical processes causes the nonexponential decay of $K^{(3)}$ by increasing δn_ϵ. We rewrite the collision integral by the electron-phonon interaction $\tilde{I}_\epsilon^{\rm el-ph} := -\frac{1}{\bar{g}_\epsilon^R}\frac{\pi}{2}N_0 g^2 \int_{-\phi_D}^{\phi_D}\frac{{\rm d}\phi}{2\pi}\omega_\phi I_{\phi,\epsilon}^{\rm el-ph}[\delta n_\epsilon, \delta N_\phi]$ to specify the physical processes in this term. In the case of $\epsilon > 0$, $I_{\phi,\epsilon}^{\rm el-ph}$ is decomposed into three terms, $I_{\phi,\epsilon}^{\rm el-ph} = I_{\phi,\epsilon}^a + I_{\phi,\epsilon}^b + I_{\phi,\epsilon}^c$, in which each term is written as follows.

$$I_{\phi,\epsilon}^a = 2g_{\phi,\epsilon}^- \delta^{(2)}[n_\epsilon(1-n_{\epsilon-\omega_\phi})(1+N_{\omega_\phi}) - n_{\epsilon-\omega_\phi}(1-n_\epsilon)N_{\omega_\phi}]\theta(\epsilon-\omega_\phi).$$

$$I_{\phi,\epsilon}^b = -2g_{\phi,\epsilon}^+ \delta^{(2)}[n_{\epsilon+\omega_\phi}(1-n_\epsilon)(1+N_{\omega_\phi}) - n_\epsilon(1-n_{\epsilon+\omega_\phi})N_{\omega_\phi}].$$

$$I_{\phi,\epsilon}^c = 2g_{\phi,\epsilon}^- \delta^{(2)}[n_\epsilon n_{\omega_\phi-\epsilon}(1+N_{\omega_\phi}) - (1-n_{\omega_\phi-\epsilon})(1-n_\epsilon)N_{\omega_\phi}]\theta(\omega_\phi-\epsilon).$$

Here, $\delta^{(2)}$ operates n or N ((2) indicates the order of the external fields), and then $\delta^{(2)}n_\epsilon = \delta n_\epsilon(t)$ and $\delta^{(2)}N_{\omega_\phi} = \delta N_{\omega_\phi}(t)$ (other n and N are replaced by $n_\epsilon^0 = 1/({\rm e}^{\epsilon/T}+1)$ and $N_{\omega_\phi}^0 = 1/({\rm e}^{\omega_\phi/T}-1)$, respectively). Each term describes the emission of phonons (I^a), the absorption of phonons (I^b), and the recombination term (I^c), respectively. (The recombination does not mean that quasiparticles recombine into Cooper pairs, as the misleading picture in ref. 11 suggests. This term exists in the case of the electron-phonon interaction for d-wave superconductors.) We substitute these terms into the above collision integral $\tilde{I}_\epsilon^{\rm el-ph}$,

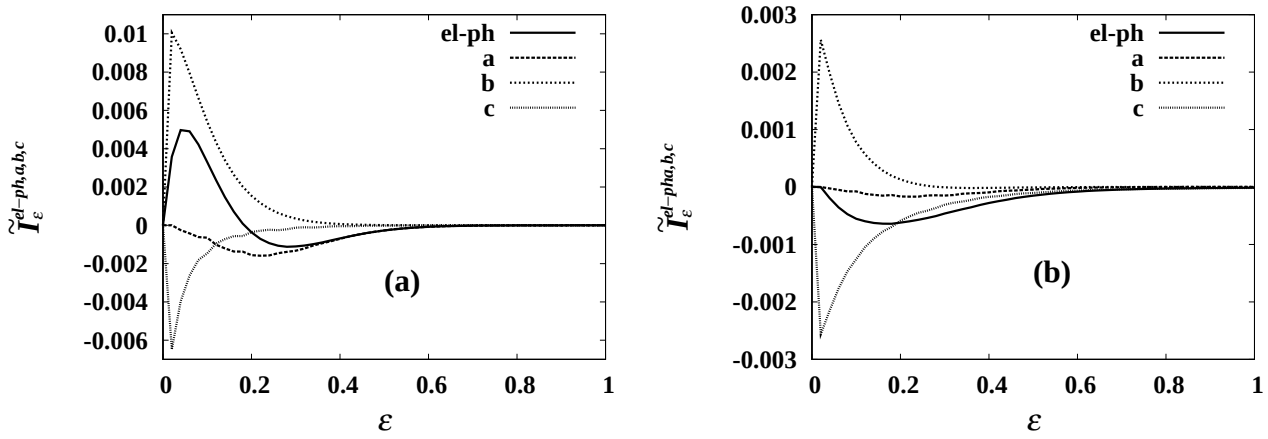

Fig. 9. Energy dependences of $\tilde{I}_\epsilon^{\mathrm{el-ph}}(t)$ and $\tilde{I}_\epsilon^{\mathrm{a,b,c}}(t)$ at $t = 1500$ for (a) $T/T_\mathrm{c} = 0.1$ and (b) $T/T_\mathrm{c} = 0.3$. The phonons are supposed to be in thermal equilibrium ($\delta N = 0$).

and write each term as $\tilde{I}_\epsilon^{a,b,c}$.

The energy dependences of $\tilde{I}_\epsilon^{\mathrm{el-ph}}(t)$ and $\tilde{I}_\epsilon^{a,b,c}(t)$ in the case of $\delta N = 0$ are shown in Fig. 9. A scattering term (emission of phonons) $\tilde{I}_\epsilon^a$ and the recombination term $\tilde{I}_\epsilon^c$ are negative, and another scattering term (absorption of phonons) $\tilde{I}_\epsilon^b$ is positive. At low T/T_c, $\tilde{I}_\epsilon^b$ is predominant over $\tilde{I}_\epsilon^a + \tilde{I}_\epsilon^c$, and then the collision term $\tilde{I}_\epsilon^{\mathrm{el-ph}}$ takes a positive value at low energy. Although the absolute values of each term decrease with increasing T/T_c, $\tilde{I}_\epsilon^{\mathrm{el-ph}}$ takes a negative value at $T/T_\mathrm{c} = 0.3$ because of a rapid decrease in the degree of absorption of phonons. The negative $\tilde{I}_\epsilon^{\mathrm{el-ph}}$ for $\epsilon > 0$ brings about a decrease in $\delta n_\epsilon(t)$ all over $\epsilon > 0$, as shown above. This causes the exponential decay of $K^{(3)}$. On the other hand, the positive $\tilde{I}_\epsilon^{\mathrm{el-ph}}$ at low energy increases $\delta n_\epsilon(t)$, and then reduces the decreasing rate of $K^{(3)}$ as compared to that of the exponential decay. Therefore, we regard the absorption of phonons as the main process for the nonexponential decay of $K^{(3)}$. This is in contrast to the phenomenological interpretation based on the RT equation,[5] in which the recombination term causes the nonexponential relaxation as the bimolecular decay. In the case of $\delta N \neq 0$, the energy dependences of $\tilde{I}_\epsilon^{\mathrm{el-ph}}(t)$ and $\tilde{I}_\epsilon^{a,b,c}(t)$ are qualitatively similar to those of $\delta N = 0$. However, in this case, the relaxation dynamics is slower than that of $\delta N = 0$ because of the presence of δN in $K^{(3)\mathrm{el-ph}}$, as shown above.

This predominance of the phonon-absorption term at low temperatures is restricted within some range of time. By performing the calculation further at large t, it is found that the recombination term becomes predominant over the absorption of phonons. At $T/T_\mathrm{c} = 0.1$, for instance, $\tilde{I}_\epsilon^{\mathrm{el-ph}}(t)$ takes negative values in the entire range of $\epsilon > 0$ at approximately $t \simeq 5000$. Then, at about this time, $1/K^{(3)}$ deviates from the t-linear behavior, and ceases to show the nonexponential decay.

5. Summary and Discussion

In this study, we investigated the photoinduced change in the superfluid weight in the transient state of d-wave superconductors. We clarified how the nonexponential decay occurs. At first, we obtained the formula that relates the nonequilibrium distribution function to the physical response function. Although the former quantity is usually discussed in theoretical works, it is the latter quantity that is necessary for a comparison with the experiments. In this derivation, we found that the vertex correction is predominant and that the nonequilibrium distribution function is effective only through this interaction term. We numerically solved the kinetic equation for the nonequilibrium distribution function with taking account of the electron-electron and electron-phonon interactions and substituted its solution into the expression of the nonlinear response function.

The numerical calculation shows that the electron-phonon interaction predominates over the electron-electron interaction in the long-time behavior. In contrast to the previous studies based on the phenomenological RT equation, the nonexponential decay does not originate from the bimolecular recombination. Rather, it results from the enhancement of the nonequilibrium distribution at low energies, which is caused by the absorption of phonons. This fact is revealed using the nonequilibrium distribution functions that couple with each other at different energies through the interaction effect, and is not known from the RT equation in which the quasiparticle density averaged over energy is used. This leads to an explanation of the nonexponential decay which is consistent in terms of the order of the external field under the condition that the pumping intensity is low. (As noted in ref. 17, this condition is presumably satisfied in the experiment[5] with reference to the excitation fluence in ref. 7.)

Finally, we comment on several problems related to but beyond the scope of this paper. Strictly speaking, it requires numerical integration with a very small energy scale to discuss the long-time behavior of a response function. In this paper, however, we avoided this by performing Fourier transformation at the outset. This is achieved at the cost of accuracy in the short-time behavior of a response function. Therefore, our formulation is not suitable for discussing physical quantities such as the exact form of the spectrum at this time scale; however, the results presented in this paper are not considered to be affected by these fine structures because of the rapid smoothing by the electron-electron interaction.

There are very few experiments that probed low-energy phenomena such as the superfluid weight reported in ref. 5. Most pump-probe experiments have been performed with the use of an optical probe beam. The formulation in this paper is restricted to the case in which the probe frequency is zero. The extension to a finite probe frequency is required to discuss optical probe cases, but it will not alter the importance of the interaction effect if we consider the case of the local limit.

The formulation based on the local limit is appropriate for cuprate superconductors as

discussed in ref. 17. Other superconductors are also investigated by nonlinear optical spectroscopy, and the pump-probe experiments have been performed in iron-based superconductors (for example, ref. 24.) Infrared spectroscopy indicates that the nonlocal limit is realized in superconductors of this kind,[25] and in this case the Mattis-Bardeen formula[26] is valid. It is possible that the nonlocal limit gives a different result regarding the nonlinear response from the local limit in which the interaction effect is essential for the optical response.

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