

Non-Markovian theory of nonlinear-optical phenomena associated with the extremely fast relaxation in condensed matter

Masaki Aihara

Department of Physics, Faculty of Liberal Arts, Yamaguchi University, Yamaguchi 753, Japan

(Received 28 July 1981)

The transient nonlinear-optical phenomena associated with the extremely fast relaxation processes in condensed matter are investigated from a microscopic viewpoint, taking into account the non-Markovian nature of the system. It is found that, in the ultrashort time region comparable to the thermal-reservoir correlation time, the transient nonlinear-optical response exhibits the characteristic time variation inherent in the non-Markovian nature of the matter system. This result cannot be explained by the conventional phenomenological theory based on the relaxation time description. The general formula obtained is applied, as an example, to the transient four-photon parametric coupling generated by the two-pulse excitation of the localized-electron-phonon system. It is found that not only the time variation of the signal intensity, but also the dependence of the time-integrated intensity of the pulse interval exhibits the characteristic feature which reflects the dynamics of the relaxation of the phonon system. It is emphasized that this result provides us the most practical tool to observe the extremely fast relaxation processes in condensed matter.

I. INTRODUCTION

Recently, the ultrashort relaxation processes in condensed matter have been extensively studied by the resonant nonlinear optical method, such as the resonant Rayleigh-type optical mixing,^{1,2} the polarization spectroscopy,³ the resonant transient parametric coupling,⁴ and so on. In these studies the longitudinal and transverse relaxation times T_1 and T_2 for condensed matter have been estimated to be of the order of the picosecond or less, by comparing the experimental data to the simple theory in which T_1 and T_2 are phenomenologically introduced into the optical Bloch equation.

However, special attention should be paid to the fact that these extremely fast relaxation phenomena, whose time scale is of the order of the thermal-reservoir correlation time t_c , can no longer be analyzed by the conventional phenomenological theory based on the relaxation time description. This is because in such an extremely short time region the effect of the reservoir on the relevant electronic states cannot be regarded as the Markovian process (or equivalently the rapid modulation or the motional narrowing situation); therefore the dynamical motion of the reservoir caused by the strong system-reservoir interaction should properly be taken into consideration.

The purpose of this paper is to present a theoretical study of the resonant nonlinear optical phenomenon associated with the extremely fast relaxation processes in condensed matter, taking into account the non-Markovian character of the interaction between the relevant system and the reservoir. It is

shown by this first-principle theory which is applicable to the transient phenomena in an arbitrary time scale, that the signal radiation exhibits the characteristic transient response reflecting the non-Markovian nature of the condensed matter; this result cannot be explained by the conventional phenomenological theory which leads only to the exponential type decay.

In particular, the two-pulse excitation method is shown to provide us a powerful tool to study the extremely fast relaxation phenomena (comparable or less than picosecond), since the time-integrated signal intensity also shows as the function of the pulse separation the characteristic feature reflecting the dynamical motion of reservoir. This is the decided advantage of the present nonlinear optical method over those associated with the linear optical phenomena, such as the time-resolved spectrum⁶ or the time-dependent degree of polarization⁷ of the resonantly scattered radiation.

In Sec. II we formulate the problem without specifying the details of the matter Hamiltonian, in order to preserve the wide applicability of the present theory to various kinds of materials. The general formula for the electric-dipole moment induced by the third-order nonlinear optical process is derived.

In Sec. III the general formula derived in Sec. II is applied to the case of the two-pulse excitation, in order to investigate the dynamics of the relaxation directly in the time domain. The expression for the intensity of the radiation generated by the transient parametric coupling is obtained by the cumulant expansion method.

Before performing the specific calculation, the two limiting situations of the long- and short-time cases are considered in Secs. IV. and V, respectively. The former leads to the new photon-echo phenomenon⁵ which is caused by the memory effect of the reservoir in the extremely short-time region. The latter case is associated with the opposite situation where the usual relaxation-time description is allowed.

In order to give more specific considerations, the localized-electron phonon system is taken as a typical example of matter. The detailed discussions on the results obtained by the numerical calculations are made in Sec. VI.

II. FORMULATION

Let us consider a pair of well-separated energy bands in condensed matter which is in resonance with the coherent radiation. The Hamiltonian of this system is expressed by

$$H(t) = H_0 + H_1(\vec{r}, t), \quad (1)$$

$$H_0 = |g\rangle H_g \langle g| + |e\rangle H_e \langle e|, \quad (2)$$

$$H_1(\vec{r}, t) = -|g\rangle d \langle e| E^*(\vec{r}, t) - |e\rangle d^\dagger \langle g| E(\vec{r}, t). \quad (3)$$

In the matter Hamiltonian H_0 given by Eq. (2), H_g (H_e) describes the ground (excited) subspace Hamiltonian which is a function of the so-called reservoir variables. It should be noted that the expression (2) is general in that H_0 represents not only the narrow two-level system in the motional narrowing situation, but also the broad bands in the strongly coupled system such as several color centers in ionic crystals or dye molecules. The matter-radiation interaction is expressed by Eq. (3) in the dipole approximation; d is the matrix element of the dipole-moment operator referring to the ground and excited subspaces, and is in general a function of the reser-

voir variables. Since the exciting radiation is supposed to be so coherent and intense, the positive (negative) frequency part of the electric field is represented by a time-varying complex function $E(r, t)$ [$E^*(r, t)$]. In order to confine our attention to the essential point associated with the coherence effect of many oscillating dipoles, the direct interaction between dipoles at different positions is discarded.

The equation of motion for the density operator in the Schrödinger picture is given by

$$i \frac{d}{dt} \rho(t) = L(t) \rho(t), \quad (4)$$

where the Liouvillian $L(t)$ is defined by the commutator $[H(t), \cdot]$. The corresponding equation of motion in the interaction picture is expressed by

$$i \frac{d}{dt} \rho_I(t) = L_I(t) \rho_I(t), \quad (5)$$

where the density operator in the interaction picture $\rho_I(t)$ and the associated Liouvillian $L_I(t)$ are defined by

$$\rho_I(t) = \exp(iL_0 t) \rho(t), \quad (6)$$

$$L_I(t) = \exp(iL_0 t) L_1(t) \exp(-iL_0 t). \quad (7)$$

In above equations, L_0 and $L_1(t)$ are defined by $[H_0, \cdot]$ and $[H_1(\vec{r}, t), \cdot]$, respectively.

The induced electric-dipole moment is obtained by evaluating the right-hand side of the following equation:

$$\begin{aligned} \langle d(t) \rangle &= \text{Tr}[(|g\rangle d \langle e| + |e\rangle d^\dagger \langle g|) \rho(t)] \\ &= \text{Tr}_R(d \langle e | \rho(t) | g \rangle) + \text{c.c.}, \end{aligned} \quad (8)$$

where Tr_R denotes the trace operation over the reservoir variables.

Let us now consider, as a fundamental example, the third-order nonlinear optical process: the four-photon process. The corresponding term for the density operator is expressed by

$$\rho_I^{(3)}(t) = i \int_{-\infty}^t dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 L_I(t_1) L_I(t_2) L_I(t_3) \rho(-\infty). \quad (9)$$

We assume that the excited band is well separated from the ground one, so that the initial density operator $\rho(0)$ takes the form

$$\rho(-\infty) = |g\rangle \rho_R \langle g|, \quad (10)$$

where

$$\rho_R = \exp(-\beta H_g) / \text{Tr}[\exp(-\beta H_g)]. \quad (11)$$

Thus we find from Eqs. (9)–(11) the third-order off-diagonal density matrix element, which has the following form:

$$\begin{aligned} \langle e | \rho^{(3)}(t) | g \rangle &= \langle e | \exp(-iL_0 t) \rho_I^{(3)}(t) | g \rangle \\ &= i \int_{-\infty}^t dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \langle \langle e g | \exp[-iL_0(t-t_1)] L_I(t_1) \\ &\quad \times \exp[-iL_0(t_1-t_2)] L_I(t_2) \exp[-iL_0(t_2-t_3)] L_I(t_3) | g g \rangle \rangle \rho_R. \end{aligned} \quad (12)$$

Here we have made use of the hyperspace notation $|ab\rangle\rangle$ instead of the usual Dirac notation $|a\rangle\langle b|$, since the former is more convenient when we trace the time evolution of the density matrix element.

The matrix elements of the free propagator have the forms

$$\langle\langle eg|\exp(-iL_0(t_1-t_2))|eg\rangle\rangle = \exp[-i(L_g + V + E_{eg})(t_1-t_2)] , \quad (13)$$

$$\langle\langle ge|\exp(-iL_0(t_1-t_2))|ge\rangle\rangle = \exp[-i(L_e - V - E_{eg})(t_1-t_2)] , \quad (14)$$

$$\langle\langle gg|\exp(-iL_0(t_1-t_2))|gg\rangle\rangle = \exp[-iL_g(t_1-t_2)] , \quad (15)$$

$$\langle\langle ee|\exp(-iL_0(t_1-t_2))|ee\rangle\rangle = \exp[-iL_e(t_1-t_2)] . \quad (16)$$

In the above equation, $L_{g,e}$ is defined by $[H_{g,e}, \cdot]$, and $E_{eg} = \text{Tr}[\rho_R(H_e - H_g)]$ is the Franck-Condon energy, and $V = H_e - H_g - E_{eg}$ is the system reservoir-interaction. Schematic diagrams associated with Eqs. (13)–(16) are shown in Fig. 1. All off-diagonal elements, such as $\langle\langle ee|\dots|eg\rangle\rangle$, vanish, since H_0 is free from the interband transition caused by the exciting radiation. The matrix elements of the interaction Liouvillian take the forms

$$\langle\langle eg|L_1(t)|gg\rangle\rangle = \langle\langle ee|L_1(t)|ge\rangle\rangle = \langle\langle gg|L_1(t)|ge\rangle\rangle = \langle\langle eg|L_1(t)|ee\rangle\rangle = -E(\vec{r}, t)d^\dagger , \quad (17)$$

$$\langle\langle ge|L_1(t)|gg\rangle\rangle = \langle\langle ee|L_1(t)|eg\rangle\rangle = \langle\langle gg|L_1(t)|eg\rangle\rangle = \langle\langle ge|L_1(t)|ee\rangle\rangle = E^*(\vec{r}, t)d . \quad (18)$$

All diagonal elements vanish, since $H_1(\vec{r}, t)$ causes the transition between two bands.

Substituting Eqs. (13)–(18) into Eq. (12), we obtain the third-order off-diagonal density matrix element which has the following form:

$$\begin{aligned} \langle e|\rho^{(3)}(t)|g\rangle &= i \int_{-\infty}^t dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \\ &\times E(\vec{r}, t_1) \exp[-i(L_g + V + E_{eg})(t-t_1)] d^\dagger [\exp[-iL_g(t_1-t_2)] + \exp[-iL_e(t_1-t_2)]] \\ &\times \{E^*(\vec{r}, t_2)E(\vec{r}, t_3)d \exp[-i(L_g + V + E_{eg})(t_2-t_3)] d^\dagger \\ &+ E(\vec{r}, t_2)E^*(\vec{r}, t_3)d \exp[-i(L_e - V - E_{eg})(t_2-t_3)] d\} \rho_R . \end{aligned} \quad (19)$$

It should be noted that, unlike in the case of the phenomenological theory, $\langle e|\rho^{(3)}(t)|g\rangle$ is still the operator for the reservoir variables. Four third-order processes expressed by the above equation are schematically shown in Fig. 2, which may help our intuitive understanding of the third-order nonlinear optical processes.

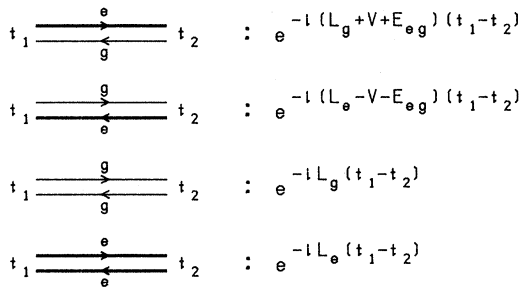


FIG. 1. Schematic representation of the matrix elements of the free propagator in the hyper-space [see Eqs. (13)–(16)]. The thin and thick lines are associated with the ground and excited states, respectively.

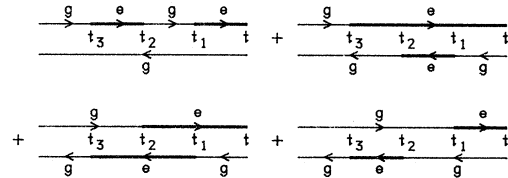


FIG. 2. Schematic representation of the possible four time evolutions of the third-order off-diagonal density-matrix element $\langle e|\rho^{(3)}(t)|g\rangle$ expressed by Eq. (19). The thin and thick lines are associated with the ground and excited states, respectively.

III. TRANSIENT PARAMETRIC COUPLING (TWO-PULSE EXCITATION)

In order to facilitate more explicit discussions, we consider in this section the transient parametric coupling generated by the two-pulse excitation. In this case, the exciting electric field is expressed by

$$E(\vec{r}, t) = E_1(t) \exp[i(\vec{k}_1 \vec{r} - \omega t)] + E_2(t - t_s) \exp[i(\vec{k}_2 \vec{r} - \omega t)] , \quad (20)$$

where $\vec{k}_{1,2}$ are wave vectors for the first and second pulses, $E_{1,2}(x)$ is their pulse envelope function which is peaked at $x=0$, and t_s is a pulse separation. Substituting Eq. (20) into Eq. (19), we have four terms with the wave vectors \vec{k}_1 , \vec{k}_2 , $2\vec{k}_2 - \vec{k}_1$, and $2\vec{k}_1 - \vec{k}_2$. Let us consider the term with the wave vector $2\vec{k}_2 - \vec{k}_1$ which is associated with radiation generated by the optical parametric effect. When two excitation pulses are well separated, Eq. (19) is reduced to

$$\begin{aligned} \langle e | \rho^{(3)}(t) | g \rangle = & i \int_{-\infty}^t dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 E_1(t_3) E_2(t_2 - t_s) E_2(t_1 - t_s) \exp[i(2\vec{k}_2 - \vec{k}_1) \vec{r}] \\ & \times \exp[-i(L_g + V + E_{eg})(t - t_1)] [\exp[-iL_g(t_1 - t_2)] + \exp[-iL_e(t_1 - t_2)]] \\ & \times (d^\dagger)^2 \exp[-i(L_e - V - E_{eg})(t_2 - t_3)] d \rho_R . \end{aligned} \quad (21)$$

Substituting Eq. (21) into Eq. (8), and confining ourselves to the limit of short excitation pulse, we obtain

$$\langle d(t) \rangle = (i/8) \theta_1 \theta_2^2 \exp[-iE_{eg}(t - 2t_s)] \exp[i(2\vec{k}_2 - \vec{k}_1) \vec{r}] \langle \exp[-i(L_g + V)(t - t_s)] \exp[-i(L_e - V)t_s] \rangle , \quad (22)$$

where $\langle \dots \rangle$ denotes $\text{Tr}(\dots \rho_R)$. Figure 3 shows the schematic representation of the time evolution of the system which is expressed by Eq. (22); the off-diagonal element $\langle g | \rho^{(3)}(t) | e \rangle$, which is generated by the first pulse applied at $t=0$, is transferred at $t=t_s$ to its Hermitian adjoint $\langle e | \rho^{(3)}(t) | g \rangle$. In deriving Eq. (22) we have assumed that the dipole moment is independent of the reservoir variables (Franck-Condon approximation), and $\theta_i (i=1, 2)$ are the so-called pulse area defined by $\int 2E_{1,2}(t) dt$.

The averaged value in Eq. (22) is simply evaluated by the cumulant expansion method, and we obtain

$$\begin{aligned} \ln \langle \exp[-i(L_g + V)(t - t_s)] \exp[-i(L_e - V)t_s] \rangle \\ = - \int_0^{t-t_s} dt_1 \int_0^{t_1} dt_2 \langle V(t_1) V(t_2) \rangle + \int_0^{t-t_s} dt_1 \int_0^{t_1} dt_2 \langle V(t_2) V(t_1 + t_s) \rangle - \int_0^{t_s} dt_1 \int_0^{t_1} dt_2 \langle V(t_2) V(t_1) \rangle . \end{aligned} \quad (23)$$

Here, higher-order cumulants have been omitted because of the following two reasons: (i) In the case of the linear system-reservoir coupling, all higher cumulants vanish. (ii) In both limiting situations of weak and strong coupling, they make negligible contribution, irrespective of a specific form of the coupling V .

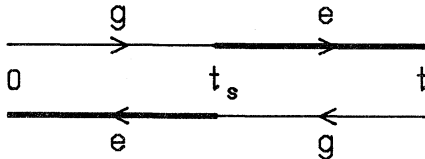


FIG. 3. Schematic representation of the time evolution of the system which is excited by the sequence of two short pulses [see Eq. (22)]. The off-diagonal element $\langle g | \rho^{(3)}(t) | e \rangle$, which is generated by the first pulse at $t=0$, is transferred to its conjugate element $\langle e | \rho^{(3)}(t) | g \rangle$ by the second pulse at $t=t_s$.

From Eqs. (22) and (23) we obtain the intensity of radiation emitted into the direction $2\vec{k}_2 - \vec{k}_1$, which takes, aside from the unimportant multiplicative factor, the form

$$I^{(3)}(t) = \theta_1^2 \theta_2^4 \exp\{-2[2S(t - t_s) + 2S(t_s) - S(t)]\} , \quad (24)$$

where $S(t)$ is the real part of the cumulant, which is defined by

$$\begin{aligned} S(t) &= \int_0^t dt_1 \int_0^{t_1} dt_2 \text{Re}[\langle V(t_1) V(t_2) \rangle] \\ &= \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \omega^{-2} J(\omega) (1 - \cos \omega t) . \end{aligned} \quad (25)$$

In Eq. (25) $J(\omega)$ is the power spectrum of the system-reservoir coupling:

$$J(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle V(t) V(0) \rangle . \quad (26)$$

The set of equations (24) and (25) enables us to

know how the transient response associated with the optical parametric process reflects the dynamical behavior of the system-reservoir coupling. Before performing specific model calculations in Sec. VI, we will consider the long- and short-time limits in the following two sections.

IV. LONG-TIME BEHAVIOR (MARKOVIAN LIMIT)

When observation time scale is much longer than the correlation time t_c [t_c is the measure of the inverse of the width of $J(\omega)$], we obtain the following result independent of the specific form of the system-reservoir interaction. In this case ($t \gg t_c$), it is convenient to rewrite Eq. (25) into the form

$$S(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{1 - \cos\omega t}{t\omega^2} J(\omega) t, \quad (27)$$

which enables us to use the following well-known asymptotic formula:

$$\frac{1 - \cos\omega t}{t\omega^2} \sim \delta(\omega). \quad (28)$$

From this, $S(t)$ defined by Eq. (27) [or (25)] is reduced to

$$S(t) \sim \frac{1}{2} J(0) t. \quad (29)$$

We should here remark that, in the present long-time (i.e., Markovian) limit, the function $S(t)$ is found to be linear in t . This situation causes the fact that, in the argument of the exponential function in Eq. (24), the t_s dependence of the radiation intensity cancels out; we obtain the following simple expression for the radiation intensity:

$$I^{(3)}(t) \sim e^{-J(0)t} \sim e^{-(2/T_2)t}. \quad (30)$$

From this, we find that the radiation intensity decays exponentially with the transverse relaxation time T_2 defined by $2/J(0)$. Thus, if the present theory is applied to the limiting case of the long time ($t \gg t_s$), the result obtained by the phenomenological theory⁴ is reproduced.

V. SHORT-TIME BEHAVIOR

In this section, let us consider the opposite limiting situation in which the observation time scale is much shorter than the correlation time ($t \ll t_s$). In this case, we can approximate the expression (25) to its leading term of the power series in time t , which has the form

$$S(t) \sim D^2 t^2. \quad (31)$$

Here D is defined by

$$D^2 \sim \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} J(\omega) = \langle V^2 \rangle, \quad (32)$$

and is the measure of the system-reservoir interaction energy. From Eq. (31), the radiation intensity expressed by Eq. (24) becomes

$$I^{(3)}(t) \sim \exp[-D^2(t - 2t_s)^2]. \quad (33)$$

If the system-reservoir interaction is so strong ($D \gg 1$) that D^{-1} is shorter than the pulse separation t_s , then the right-hand side of the above equation expresses the fact that the echo with the Gaussian profile is formed at $2t_s$. This is the new type of photon-echo phenomenon which was recently predicted by the present author.⁵ We should remark that Eq. (33) expressing the echo profile has been obtained without introducing the inhomogeneous line-shape function which plays the essential role in the conventional echo phenomena. The expressions (33) and (30) have derived by the same equation (24); the difference between them is due simply to the different observation time scale, as is schematically shown in the Fig. 4. This new echo phenomenon is caused by the memory effect of the reservoir in the extremely short-time region; or equivalent statement is that the slow frequency modulation due to the reservoir plays the role similar to the inhomogeneous broadening. Thus, the present theory unifies the two concepts, the homogeneous and inhomogeneous broadenings, from the microscopic viewpoint.

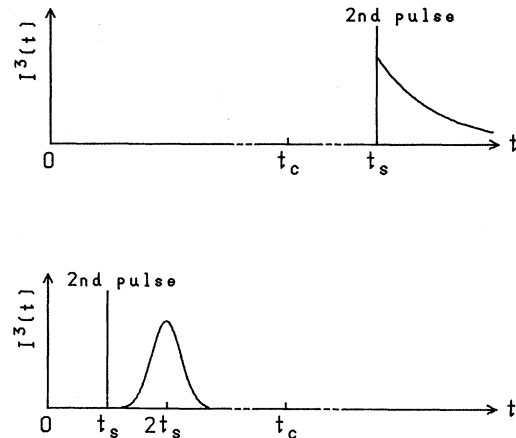


FIG. 4. Schematic display of the time dependence of the intensity of radiation generated by the two short excitation pulses, for the two limiting situations: the long- and short-time regions. The upper (lower) graphs is for the case that the excitation pulse separation t_s is much larger (shorter) than the correlation time t_c .

VI. MODEL CALCULATION (ARBITRARY TIME REGION)

We are now in a position to answer the following question: what kind of phenomena will arise in the intermediate time region in which observation times are of the same order as the correlation time t_c ? To answer this question we need specific information on

the frequency dependence of the power spectrum $J(\omega)$ of the system-reservoir interaction. [We should recall that in the preceding sections only the two quantities $J(0)$ and $D = \langle V^2 \rangle$ are of interest.]

In this section, we consider as a typical example a localized-electron phonon system. The system-reservoir (i.e., electron-phonon) interaction Hamiltonian is expressed, up to the quadratic interaction term, by

$$V = H_e - H_g - \langle (H_e - H_g) \rangle = \sum_k h_k e_k (b_k + b_k^\dagger) + \frac{1}{2} \sum_k \sum_q h_{k,q} (e_k e_q)^{1/2} (b_k + b_k^\dagger) (b_q + b_q^\dagger) , \quad (34)$$

where e_k is the phonon energy for mode k , and b_k and b_k^\dagger are the associated annihilation and creation operators. The first and second terms in Eq. (34) describe the linear and quadratic interactions, and h_k and $h_{k,q}$ are their dimensionless interaction constants. In many localized-electron phonon system, the quadratic interaction is very weak, so that the higher cumulants can be discarded [see Eq. (23) and the discussion below].

The time-correlation function for the electron-phonon interaction V expressed by Eq. (34) can be easily calculated, and is found to be composed of the two terms for the linear and quadratic interactions;

$$\langle V(t) V(0) \rangle = \langle V(t) V(0) \rangle_L + \langle V(t) V(0) \rangle_Q , \quad (35)$$

$$\langle V(t) V(0) \rangle_L = \sum_k h_k^2 e_k^2 \{ [n(e_k) + 1] \exp(-ie_k t) + n(e_k) \exp(ie_k t) \} , \quad (36)$$

$$\begin{aligned} \langle V(t) V(0) \rangle_Q = & \sum_k \sum_q e_k e_q h_{k,q}^2 \{ [n(e_k) + 1][n(e_q) + 1] \exp[-i(e_k + e_q)t] \\ & + 2n(e_k)[n(e_q) + 1] \exp[i(e_k - e_q)t] + n(e_k)n(e_q) \exp[i(e_k + e_q)t] \} , \end{aligned} \quad (37)$$

where $n(x)$ is the Bose-Einstein distribution function. In order to perform further calculations, let us assume that h_k and $h_{k,q}$ are constants ($S_L = h_{k,q}^2$ and $S_Q = h_{k,q}^2$), and that the phonon density of states has the Gaussian profile with the maximum at ω_p and with the width γ_p . Equations (36) and (37) are thus rewritten into the following forms:

$$J_L(\omega) = (2\sqrt{\pi} S_L / \gamma_p) \omega^2 \{ [n(\omega) + 1] \exp[-(\omega - \omega_p)^2 / \gamma_p^2] \theta(\omega) + n(-\omega) \exp[-(\omega + \omega_p)^2 / \gamma_p^2] \theta(-\omega) \} , \quad (38)$$

$$\begin{aligned} J_Q(\omega) = & (4S_Q / \gamma_p^2) \int_{-\infty}^{\infty} de \exp[-(e - \omega_p)^2 / \gamma_p^2] \\ & \times \{ e(\omega - e)[n(e) + 1][n(\omega - e) + 1] \exp[-(\omega - e - \omega_p)^2 / \gamma_p^2] \theta(\omega - e) \\ & - 2e(\omega - e)[n(e) + 1]n(\omega - e) \exp[-(e - \omega - \omega_p)^2 / \gamma_p^2] \theta(e - \omega) \\ & - e(\omega + e)n(e)n(-\omega - e) \exp[-(e + \omega + \omega_p)^2 / \gamma_p^2] \theta(-\omega - e) \} . \end{aligned} \quad (39)$$

We show in Fig. 5 the numerical displays of $J(\omega)$, $J_L(\omega)$ and $J_Q(\omega)$, which are displayed by the thick solid, the thin solid, and the dashed line, respectively. (Here and henceforth, we use the unit of $\omega_p = 1$.) Under the situation where the linear interaction is much stronger than the quadratic one ($S_L \gg S_Q$), the power spectrum $J(\omega)$ is mainly determined by the linear term $J_L(\omega)$. However, for finite tempera-

tures, we should remark that $J_Q(\omega)$ at low frequencies becomes larger than $J_L(\omega)$, as shown in Fig. 5. This is because the simultaneous phonon absorption and emission process is induced by the quadratic interaction, which makes the predominant contribution to the power spectrum at low frequencies. As was discussed in Sec. III, this low-frequency component determines the long-time behavior.

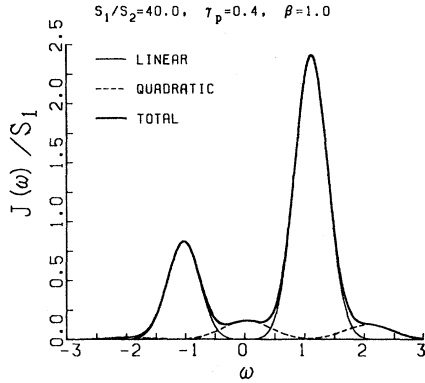


FIG. 5. The power spectrum $J(\omega)$ of the localized-electron phonon system with the linear and quadratic interaction. The linear part $J_L(\omega)$ and the quadratic one $J_Q(\omega)$ are displayed by the thin solid line and the dashed line, respectively. The energy scale of the abscissa is normalized by the average phonon frequency ω_p (i.e., $\omega_p = 1$).

We show in Fig. 6 the logarithmic display of the transient response of the radiation intensity, which is obtained by the numerical integration of Eq. (25) with Eqs. (35)–(37), for several values of the time interval t_s between the two excitation pulses. From this figure, it is clearly observed that, in the extremely short-time region comparable to the damping time of the phonon system (γ_p^{-1}), the emitted radiation exhibits the characteristic transient behavior reflecting the dynamical motion of the phonon system. We should also note that, for shorter time interval t_s , the radiation intensity does grow immediately after the second pulse. This unusual effect is the new photon echo phenomenon⁵ which is caused by the memory

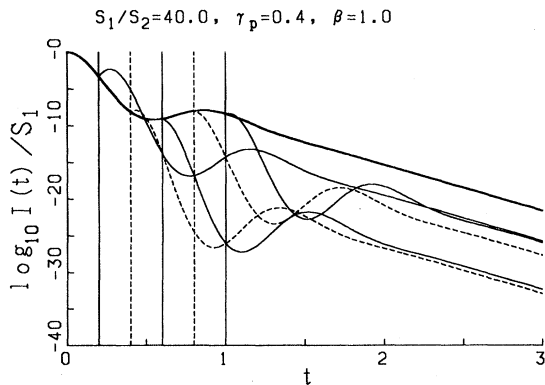


FIG. 6. The logarithmic display of the time dependence of the radiation intensity for $t_s = 0.2, 0.4, 0.6$, and 1.0 ; the time axis is normalized by the average phonon oscillation period $2\omega_p^{-1}$. The first excitation pulse is applied at $t = 0$, and the position of the second one is indicated by the vertical straight line.

effect of the reservoir, as was discussed in Sec. V.

In the longer-time region, we obtain the expected result that the radiation intensity decays exponentially with the transverse relaxation time T_2 . However, as is observed from Fig. 6, the magnitude of the intensity takes different values depending on the position of the second excitation pulse t_s ; we should recall that according to the phenomenological theory the radiation intensity is independent of t_s .⁴ This effect obtained by the present theory indicates that the time-integrated intensity (i.e., the total energy) of the emitted radiation also exhibits, as a function of t_s , the damped oscillation which reflects the lattice relaxation, as is shown in Fig. 7. We should remark that this fact is caused by the nonlinearity of the radiation-matter interaction. That is to say, the behavior of the radiation generated by the two-pulse excitation cannot be simply described by the superposition of the responses due to the two single-pulse excitations, but depends essentially on the correlation between two excitations. This is the reason why the dynamics of the system-reservoir interaction is reflected to the t_s dependence of the emitted radiation energy.

It should be emphasized here that this result sheds light on the experimental investigation of the dynamics of the extremely fast relaxation phenomena in condensed matter, since the measurement of the total radiation energy as a function of t_s (Fig. 7) is much easier than the measurement of the time dependence of radiation intensity in the picosecond or subpicosecond region (Fig. 6). Furthermore, we should also remark that the transient parametric method has the advantage in that it is free from the restriction due to the energy-time uncertainty principle which limits the effectiveness of the method based on the time-resolved spectrum,⁶ and is also free from the details of the level degeneracy which is essential in the method using the radiation polarization.⁷

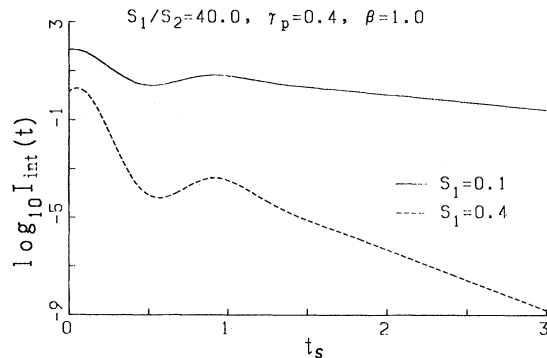


FIG. 7. The logarithmic display of the time-integrated intensity as a function of the pulse separation t_s . The abscissa is normalized by the average phonon oscillation period $2\omega_p^{-1}$.

ACKNOWLEDGMENT

The author would like to thank Professor T. Yajima for stimulating discussions.

¹T. Yajima and H. Soma, Phys. Rev. A 17, 309 (1978); 17, 324 (1978).

²H. Soma, T. Yajima, and Y. Taira, J. Phys. Soc. Jpn. 48, 2040 (1980).

³J. J. Song, J. H. Lee, and M. D. Levenson, Phys. Rev. A 17, 1439 (1978).

⁴T. Yajima, and Y. Taira, J. Phys. Soc. Jpn. 47, 1620 (1979).

⁵M. Aihara, Phys. Rev. B 21, 2051 (1980); in *Relaxation of Elementary Excitations* (Springer Series in Solid-State Sciences 18), edited by R. Kubo and E. Hanamura (Springer-Verlag, New York, 1980).

⁶Y. Toyozawa, A. Kotani, and A. Sumi, J. Phys. Soc. Jpn. 42, 1495 (1977).

⁷M. Aihara, Phys. Rev. A 18, 606 (1978); J. Phys. Soc. Jpn. 48, 773 (1980).