Ultrahigh-time-resolution coherent transient spectroscopy with incoherent light

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A new method of picosecond and femtosecond transient spectroscopy yielding information about ultrafast relaxation processes without requiring ultrashort light pulses is presented. It is based on the present theoretical analysis of the resonant degenerate four-wave-mixing process excited by two temporally incoherent light beams with wave vectors $\vec{k}_1$ and $\vec{k}_2$ which are originated from a single beam at frequency ω but have mutual time delay τ. Under the assumption that the incoherent light field has Gaussian random complex amplitude and that the resonant material consists of the usual two-level atoms, the statistically averaged intensity of the output light field with $\vec{k}_3=2\vec{k}_2-\vec{k}_1$ at ω is calculated as a function of τ. Even with the light having a much longer duration than both T_1 (the longitudinal relaxation time) and T_2 (the transverse relaxation time), the correlation trace, i.e., output intensity versus τ, represents a decay profile determined mainly by T_2 for both homogeneously and inhomogeneously broadened transitions, as long as the light correlation time τ_c is much shorter than the relaxation times. The correlation trace does not always represent a single-exponential decay but is sometimes slightly deformed by the T_1 effect. However, it does not cause a significant error in the determination of T_2. Moreover, as $T_2/T_1\rightarrow 0$, the trace becomes a single-exponential decay curve determined only by T_2. The feature of the results obtained by the present method is similar to that obtained by the conventional coherent transient spectroscopy with short pulses, such as the photon echo. The time resolution in the present method, however, is limited only by τ_c much shorter than the light duration. By regarding the incoherent light as a series of random ultrashort pulses, the present four-wave-mixing process is also interpreted as the ensemble of numerous transient four-wave-mixing processes caused by various combinations of these pulses.

I. INTRODUCTION

Studies on the relaxation process associated with excited states of materials are very important in understanding the dynamical behavior of the light-matter interaction. In condensed matter, relaxation times are generally very short, and often fall far below 1 psec. For studies on such ultrafast relaxation processes, various nonlinear spectroscopic methods in the frequency domain[1-5] have been developed and served to determine ultrashort relaxation times in the range down to below 0.1 psec. The frequency domain methods are, however, still indirect and require careful interpretations. As complementary means, there are various techniques of the time-resolved nonlinear spectroscopy, such as a conventional photon echo,[6-10] a stimulated echo,[11-13] an accumulated echo,[14,15] a transient degenerate four-wave mixing,[16,17] an induced transient grating,[18] an optical Kerr shutter,[19,20] and so on. These methods can more directly show relaxation behaviors in the time domain and give more exact relaxation times. Generally speaking, however, the time resolution of such methods is essentially limited by the temporal width of the optical pulses used there. Although recent progress of ultrashort pulse lasers[21,22] and pulse-shortening techniques[23-25] has allowed us to obtain even less than 100-fsec optical pulses, such an extremely short pulse can be generated, for the time being, only in a very limited wavelength region and only with a sophisticated apparatus. Therefore, it is still difficult even now to observe the ultrafast relaxation process occurring in less than 1 psec, in particular, over a wide spectral range. In view of this situation, we propose in the present paper a new method of transient spectroscopy that makes it possible to observe such ultrafast relaxation processes without the limitation of the time resolution by the pulse duration.

A temporally incoherent light with a wide spectral width has a very short correlation time τ_c which corresponds to the reciprocal spectral width and is much less than the temporal duration of the light. This kind of light appears like a single pulse[26,27] with a duration τ_c in the autocorrelation measurement, and is, therefore, expected to play essentially the same role as a short pulse in nonlinear spectroscopy utilizing the correlation technique. As a relevant nonlinear optical process we consider here a kind of resonant degenerate four-wave mixing,[14-17] in which as shown in Fig. 1 a light beam at a frequency ω is spatially divided into two beams with different wave-vectors $\vec{k}_1$ and $\vec{k}_2$ and then they are mixed in a resonant material both in frequency and in wave vector to generate output light beams at the same frequency ω in new directions along $\vec{k}_3=2\vec{k}_2-\vec{k}_1$ and $\vec{k}_4=2\vec{k}_1-\vec{k}_2$ due to the third-order nonlinearity of the material. We suppose that one of the beams with the wave vector $\vec{k}_2$ is temporally delayed by τ relative to the other beam with $\vec{k}_1$. When the energy of the output beam is measured as a function of the time delay τ, we can obtain a kind of correlation profile associated with both the incident light and the resonant material as a function of τ. The transient

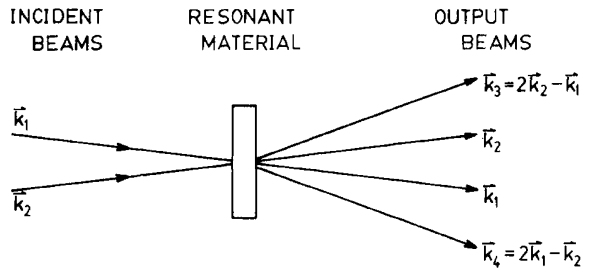

FIG. 1. Relation among the directions of the incident beams and the output beams of the resonant degenerate four-wave mixing. The two incident beams are produced by splitting a single original beam and the $\vec{k}_2$ beam is delayed in time with respect to the $\vec{k}_1$ beam.

resonant four-wave mixing (TRFM) mentioned in the previous phrase corresponds to the case that ultrashort pulses much shorter than the relaxation times are used as the incident light. The TRFM is known[16,17] to include the lowest-order part of the photon echo phenomena, and the correlation trace of the TRFM draws a decay curve determined by the relaxation time. In contrast with the TRFM case, if we use the temporally incoherent light with a much longer duration but a much shorter correlation time than the relaxation times of the resonant material, what shape can we expect for the correlation profile? We show theoretically in the present paper that the correlation profile even in this situation clearly reflects the relaxation times and has a long tail determined by them. This fact means that the use of the temporally incoherent light enables us to measure a much shorter relaxation time than the duration of the light if only the relaxation time is longer than the correlation time τ_c. In other words, the time resolution of the measurement is determined by the correlation time τ_c instead of the duration of the light, unlike the conventional methods of time-resolved spectroscopy. This fact has a possibility of benefiting us greatly because it is far easier to prepare an incoherent light source with a short correlation time τ_c than to produce an ultrashort pulse with the same duration as τ_c in various spectral ranges, especially in the case of less than 1 psec. In an extreme case, even a cw light, which has an infinite duration, enables us to observe the subpicosecond or femtosecond relaxation process in the time domain if only it has an adequate spectral width.

A similar idea has been applied to some research fields, for example, slow neutron-scattering experiments,[28] molecular beam scattering experiments,[29] random modulation cw "lidars"[30] (light detection and ranging), and so on. In these cases, using the pseudorandomly modulated beam[31] of particles or light, they measure the cross correlation between the incident beam and the output beam scattered by an object and obtain the scattering response in the time domain. Their purpose in using such a method is to get a high sensitivity due to the integrated intensity by the quasicontinuous beam.

Such types of ideas, however, have never been applied to the nonlinear spectroscopy on the light-matter system. Besides, we must recognize that this system has some significant differences from other systems mentioned above. In our case the response of the material to be investigated is nonlinear instead of linear, and furthermore the material response and the cross-correlation procedure occur simultaneously in the same material. Therefore, we can never treat these two processes separately. Moreover, in the light-matter interaction, we must consider two kinds of responses, namely, those of population and of coherence. It is also a difference that our purpose is not to get a high sensitivity but a high temporal resolution. In view of these points, it is very significant to examine the effects caused by the characteristics peculiar to the nonlinear interaction between the light and the resonant material. The present result is a new aspect of the four-wave-mixing process and gives us a new method of time-resolved nonlinear spectroscopy with an ultrahigh temporal resolution.

In Sec. II we describe the theoretical model and calculate the correlation traces to show that their profiles reflect the relaxation times of the resonant material. In Sec. III we compare the present results with the case of the TRFM using short pulses and clarify the similarities and the differences between them. Simultaneously, we give an appropriate interpretation to the present results. Finally in Sec. IV, from a viewpoint of applications, we examine the conditions for the present method to give the relaxation times definitely. We also make a slight comment on the experimental results of the present method which have been obtained very recently, as well as a few concluding remarks.

II. BASIC MODEL AND STATISTICAL AVERAGE OF THE SIGNAL INTENSITY

In this section we examine theoretically the behavior of the output light in the degenerate four-wave-mixing process in a resonant material with the temporally incoherent light. In the four-wave-mixing experiment, the configuration consisting of three incident beams[12,13,32,33] are often adopted to satisfy the phase-matching condition. We treat in the present paper, however, only the case of two incident beams as shown in Fig. 1. This is because our purpose is to clarify the essential points of the present idea and the theoretical treatment in this section can be easily extended to the case of three incident beams.

As the temporally incoherent light, we consider a model for the electric field with the following form:[34]

$$E(\vec{r},t)=\mathscr{E}(t-(\vec{n}\cdot\vec{r})/v)\exp(-i\omega t+i\vec{k}\cdot\vec{r})+\text{c.c.}\ , \tag{2.1}$$

$$\mathscr{E}(t)=\epsilon(t)R(t)\ , \tag{2.2}$$

where $\vec{n}$ is the unit vector of $\vec{k}$, v is the light velocity in the material, ignoring the dispersion of the refractive index. While $\epsilon(t)$ is a normal function, $R(t)$ is a complex random function representing a stochastic stationary Gaussian process for which

$$\langle R^*(t)R(t+\tau)\rangle=f(\tau)\ , \tag{2.3}$$

$$\langle R(t)R(t+\tau)\rangle=\langle R^*(t)R^*(t+\tau)\rangle=0\ , \tag{2.4}$$

$$\langle R(t)\rangle=\langle R^*(t)\rangle=0\ , \tag{2.5}$$

where the symbol $\langle\ \rangle$ denotes the statistical average over the random variable of the stochastic process. We assume

that $|f(\tau)|>0$ only at $|\tau| \lesssim \tau_c$, which is the definition of the correlation time τ_c. It should be noted that this mathematical model manifests the fluctuations of both the amplitude and phase of the light field.

As shown in Fig. 1, the total electric field incident on the material is a superposition of the two fields with different wave vectors $\vec{k}_1$ and $\vec{k}_2$ which are produced by splitting a single incoherent light beam at a frequency ω, where the field with the wave vector $\vec{k}_2$ is temporally delayed by τ relative to the other with $\vec{k}_1$. We write the total field in the form

$$E(\vec{r},t)=\{\mathscr{E}(t+\tau-(\vec{n}_1\cdot\vec{r})/v)\exp[-i\omega(t+\tau)+i\vec{k}_1\cdot\vec{r}] + \mathscr{E}(t-(\vec{n}_2\cdot\vec{r})/v)\exp[-i\omega t+i\vec{k}_2\cdot\vec{r}]\}+\text{c.c.}\ , \tag{2.6}$$

where $\vec{n}_1$ and $\vec{n}_2$ are the unit vectors of $\vec{k}_1$ and $\vec{k}_2$, respectively.

As a resonant material we consider a simple model, that is, an ensemble of usual two-level atoms with no degeneracy, in order to clarify the essential points of the theory. The relaxation processes in this system are usually described by two kinds of phenomenological parameters, the longitudinal relaxation time T_1 and the transverse relaxation time T_2, representing the decay of the population difference and the coherence between the two levels, respectively. Furthermore, we also consider the inhomogeneous broadening, i.e., the distribution of the transition frequency between the two levels. The motion of this system is usually described by means of the density matrix formalism, which is given[16,17] with regard to the transient resonant four-wave-mixing process in Appendix A.

The output light field at the frequency ω in the four-wave-mixing process is proportional to the induced polarization of the third order, $P^{(3)}(\vec{r},t)$, which is given in the form

$$P^{(3)}(\vec{r},t)=\hat{P}^{(3)}(\vec{r},t)\exp(-i\omega t)+\text{c.c.}\ , \tag{2.7}$$

$$\hat{P}^{(3)}(\vec{r},t)=N\int_0^\infty d\omega_0\mu_{ab}\hat{\rho}_{ba}^{(3)}(\vec{r},t,\omega_0)g(\omega_0)\ , \tag{2.8}$$

where N is the atomic number density, the subscripts a and b denote lower and upper levels, respectively, μ_{ab} is the electric dipole matrix element of the transition between the two levels, and $g(\omega_0)$ is the distribution function of the transition frequency ω_0, characterizing the inhomogeneous broadening. The third-order off-diagonal density matrix element $\hat{\rho}_{ba}^{(3)}(\vec{r},t,\omega_0)$, represented in the rotating frame at the frequency ω, contains generally four components of different wave vectors $\vec{k}_1$, $\vec{k}_2$, $\vec{k}_3=2\vec{k}_2-\vec{k}_1$, and $\vec{k}_4=2\vec{k}_1-\vec{k}_2$. In the present theory, we consider only the $\vec{k}_3$ component because the analyzing procedure and the results are easily extended to the cases in the other components. Taking out only the $\vec{k}_3$ component, we have (see Appendix A)

$$\hat{\rho}_{ba}^{(3)}(\vec{k}_3)=-2i\rho^{(0)}\left[\frac{\mu}{\hbar}\right]^3\exp(i\vec{k}_3\cdot\vec{r}+i\omega\tau) \times\int_{-\infty}^{t_r}dt_1\int_{-\infty}^{t_1}dt_2\int_{-\infty}^{t_2}dt_3\{\mathscr{E}(t_1)\mathscr{E}(t_2)\mathscr{E}^*(t_3+\tau)\exp[-i(\omega_0-\omega)(t_r-t_1-t_2+t_3)] +\mathscr{E}(t_1)\mathscr{E}^*(t_2+\tau)\mathscr{E}(t_3)\exp[-i(\omega_0-\omega)(t_r-t_1+t_2-t_3)]\} \times\exp[-\gamma_1(t_1-t_2)-\gamma_2(t_r-t_1+t_2-t_3)]\ , \tag{2.9}$$

where $\gamma_1=T_1^{-1}$, $\gamma_2=T_2^{-1}$, $\rho^{(0)}$ is the thermal equilibrium value of the population difference, and we assumed $\mu_{ba}=\mu_{ab}=\mu$. We also introduced the reduced time $t_r=t-(\vec{n}\cdot\vec{r})/v$, assuming $\vec{n}_1\simeq\vec{n}_2=\vec{n}$.

The output light intensity, which is the only physical quantity we can detect in usual experiments, is proportional to

$$|\hat{P}^{(3)}(\vec{r},t)|^2\ .$$

In the present case, however, because of the stochastic nature of $\hat{P}^{(3)}(\vec{r},t)$, we should calculate the statistically averaged value $J(\vec{k}_3)$ of this quantity, written down as

$$J(\vec{k}_3)=\langle|\hat{P}^{(3)}(\vec{r},t)|^2\rangle\propto|\epsilon(t_r)|^6F(\tau)\ , \tag{2.10}$$

$$F(\tau)=\sum_{j=1}^{4}\int_{-\infty}^{t_r}dt_1\int_{-\infty}^{t_1}dt_2\int_{-\infty}^{t_2}dt_3\int_{-\infty}^{t_r}ds_1\int_{-\infty}^{s_1}ds_2\int_{-\infty}^{s_2}ds_3M_j^{(6)}(t_1,t_2,t_3,s_1,s_2,s_3,\tau) \times\exp[-\gamma_1(c_1+d_1)-\gamma_2(c_2+d_2)] \times G_j(t_r,t_1,t_2,t_3)G_j'(t_r,s_1,s_2,s_3)\ , \tag{2.11}$$

$$M_1^{(6)}=\langle R(t_1)R(t_2)R^*(t_3+\tau)R^*(s_1)R^*(s_2)R(s_3+\tau)\rangle\ , \tag{2.12}$$

$$M_2^{(6)}=\langle R(t_1)R(t_2)R^*(t_3+\tau)R^*(s_1)R(s_2+\tau)R^*(s_3)\rangle\ , \tag{2.13}$$

$$M_3^{(6)}=\langle R(t_1)R^*(t_2+\tau)R(t_3)R^*(s_1)R^*(s_2)R(s_3+\tau)\rangle\ , \tag{2.14}$$

$$M_4^{(6)}=\langle R(t_1)R^*(t_2+\tau)R(t_3)R^*(s_1)R(s_2+\tau)R^*(s_3)\rangle\ , \tag{2.15}$$

$$c_1=t_1-t_2,\ \ d_1=s_1-s_2,\ \ c_2=t_r-t_1+t_2-t_3,\ \ d_2=t_r-s_1+s_2-s_3\ ,$$

$$G_j(t_r,t_1,t_2,t_3)=\int_0^\infty d\omega_0 g(\omega_0)\exp[-i(\omega_0-\omega)a_j]\ , \tag{2.16}$$

$$G_j'(t_r,s_1,s_2,s_3)=\int_0^\infty d\omega_0 g(\omega_0)\exp[+i(\omega_0-\omega)b_j]\ , \tag{2.17}$$

$$a_1=a_2=t_r-t_1-t_2+t_3,\ \ a_3=a_4=t_r-t_1+t_2-t_3=c_2\ ,$$

$$b_1=b_3=t_r-s_1-s_2+s_3,\ \ b_2=b_4=t_r-s_1+s_2-s_3=d_2\ .$$

The statistically averaged quantity $M_j^{(6)}(t_1,t_2,t_3,s_1,s_2,s_3,\tau)$ represents the sixth-order moment of the stochastic process $R(t)$. In deriving Eq. (2.10), we assumed also that $\epsilon(t)$ varies very slowly in comparison with T_1, T_2, and the correlation time τ_c of the light. Our interests now exist in the behavior of $F(\tau)$, which is independent of t_r because $R(t)$ is the stationary stochastic process and, therefore, the sixth-order moment $M_j^{(6)}$ does not depend on t_r. Although we need an explicit form of this moment as a function of five independent time variables in order to calculate the value of $F(\tau)$, it is impossible to determine such a function with only the properties for $R(t)$ shown in Eqs. (2.3), (2.4), and (2.5). For the stochastic stationary Gaussian process, however, we can make use of the well-known factorization property[35] of the moment, written as

$$\langle R^*(t_1)R^*(t_2)\cdots R^*(t_m)R(t_{m+1})R(t_{m+2})\cdots R(t_{m+n})\rangle$$
$$=\delta_{mn}\sum_p\langle R^*(t_1)R(t_i)\rangle\langle R^*(t_2)R(t_j)\rangle\cdots\langle R^*(t_m)R(t_k)\rangle\ , \tag{2.18}$$

where the subscripts $i,j,\ldots,k$ are given by the permutation

$$\begin{bmatrix} m+1 & m+2 & \cdots & 2m \\ i & j & \cdots & k \end{bmatrix},$$

and the summation $\sum_p$ is made over all possible permutations.

By the factorization property, each $M_j^{(6)}(t_1,t_2,t_3,s_1,s_2,s_3,\tau)$ can be written as the sum of six terms each of which is the product of three second-order moments, for example,

$$\begin{aligned}M_1^{(6)}&=\langle R^*(s_1)R(t_1)\rangle\langle R^*(s_2)R(t_2)\rangle\langle R^*(t_3+\tau)R(s_3+\tau)\rangle+\langle R^*(s_1)R(t_1)\rangle\langle R^*(s_2)R(s_3+\tau)\rangle\langle R^*(t_3+\tau)R(t_2)\rangle\\&\quad+\langle R^*(s_1)R(t_2)\rangle\langle R^*(s_2)R(t_1)\rangle\langle R^*(t_3+\tau)R(s_3+\tau)\rangle+\langle R^*(s_1)R(t_2)\rangle\langle R^*(s_2)R(s_3+\tau)\rangle\langle R^*(t_3+\tau)R(t_1)\rangle\\&\quad+\langle R^*(s_1)R(s_3+\tau)\rangle\langle R^*(s_2)R(t_1)\rangle\langle R^*(t_3+\tau)R(t_2)\rangle+\langle R^*(s_1)R(s_3+\tau)\rangle\langle R^*(s_2)R(t_2)\rangle\langle R^*(t_3+\tau)R(t_1)\rangle\\&=f(s_1-t_1)f(s_2-t_2)f(t_3-s_3)+f(s_1-t_1)f(s_2-s_3-\tau)f(t_3+\tau-t_2)\\&\quad+f(s_1-t_2)f(s_2-t_1)f(t_3-s_3)+f(s_1-t_2)f(s_2-s_3-\tau)f(t_3+\tau-t_1)\\&\quad+f(s_1-s_3-\tau)f(s_2-t_1)f(t_3+\tau-t_2)+f(s_1-s_3-\tau)f(s_2-t_2)f(t_3+\tau-t_1)\ .\end{aligned} \tag{2.19}$$

Therefore, $F(\tau)$ has totally 24 terms each of which includes the product of three correlation functions.

Now we can calculate $F(\tau)$ when a reasonable form is given to the correlation function $f(\tau)$. The calculation is, however, very troublesome to execute if we choose a function with a finite width. For this reason, we assume here

$$f(\tau)=D\delta(\tau)\ , \tag{2.20}$$

where D is a positive constant proportional to the spectral density of the light. Our interests are never diminished by this extreme assumption because our purpose is to examine the effects of the relaxation times of the material that are much longer than the correlation time τ_c.

We show in Appendix B the explicit form of $F(\tau)$ when $g(\omega_0)$ is a Gaussian distribution centered at ω with the width of $\delta\omega$. Because the general form of $F(\tau)$ is rather complicated as seen in Appendix B, we examine here two extreme cases concerning the width $\delta\omega$ of $g(\omega_0)$, as in the following.

(a) Homogeneous broadening case ($\delta\omega=0$)

(i) $\tau>0$

$$F(\tau)=C_1\left[u+\frac{2(3-4u)}{1-u}\exp(-2x)-4(1-u)\exp[-(2+u)x]+\frac{u^2(3-u)}{1-u}\exp(-2ux)\right]; \tag{2.21}$$

(ii) $\tau=0$

$$F(0)=C_1(2+2u+\tfrac{1}{4}u^2)\ ; \tag{2.22}$$

(iii) $\tau<0$

$$F(\tau)=C_1[u+2\exp(2x)]\ . \tag{2.23}$$

(b) Extremely inhomogeneous broadening case ($\delta\omega\rightarrow\infty$)

(i) $\tau>0$

$$F(\tau)=C_2\left[u+\frac{32(1-u)^2\exp(-4x)+32u(1-u)\exp[-(2+u)x]+8u^2\exp(-2ux)}{(2-u)^2}\right]\ ; \tag{2.24}$$

(ii) $\tau=0$

$$F(0)=C_2(u+4)\ ; \tag{2.25}$$

(iii) $\tau<0$

$$F(\tau)=C_2u\ , \tag{2.26}$$

where

$$C_1=\frac{D^3}{4\gamma_1^2\gamma_2}\ ,$$

$$C_2=\frac{\sqrt{2\pi}D^3}{8\gamma_1^2\delta\omega}\ ,$$

$u=\gamma_1/\gamma_2$, and we used the normalized time $x=\gamma_2\tau$.

We show the profiles of $F(\tau)$ in the above two cases (a) and (b) in Figs. 2 and 3, respectively. The ratio u of the two relaxation rates obviously takes the value from 0 to 2 in the usual two-level system, because $\gamma_2=\gamma_1/2+\gamma_2'$, where γ_2' represents the rate of the adiabatic relaxation (the pure dephasing rate). Figures 2 and 3 show the profiles for some typical values of u. The discontinuity at $x=0$ in these figures are of course caused by the choice of the δ-function as the correlation function $f(\tau)$. If we choose a correlation function with a nonzero correlation time, each profile should be continuously drawn around $x=0$ with a finite slope associated with the correlation time.

As seen in Figs. 2 and 3, we can find that the correlation traces clearly reflect the relaxation times of the resonant transition, although they are much shorter than the temporal duration of the incident light. This means that we can obtain some information about the relaxation times with the time resolution limited only by the correlation time τ_c. The correlation profiles, however, do not necessarily have single-exponential decay curves. This property is considered to be peculiar to the present method and has a possibility to bring the ambiguity in the determination of relaxation times. We will discuss these points in the following sections.

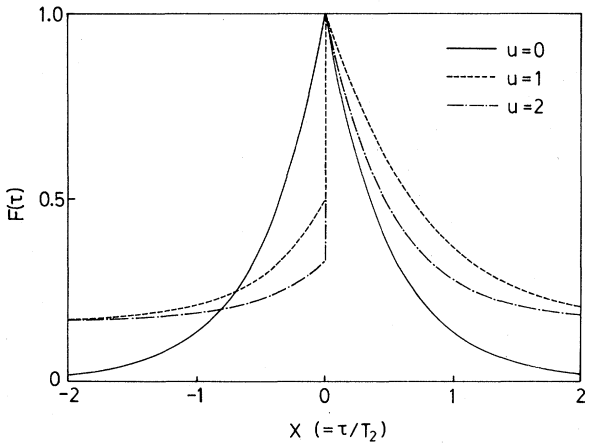

FIG. 2. Correlation profiles of the output intensities with incoherent incident light for $u\ (=T_2/T_1)=0$, 1, and 2 in the homogeneous broadening case; each profile is normalized to unity at its peak.

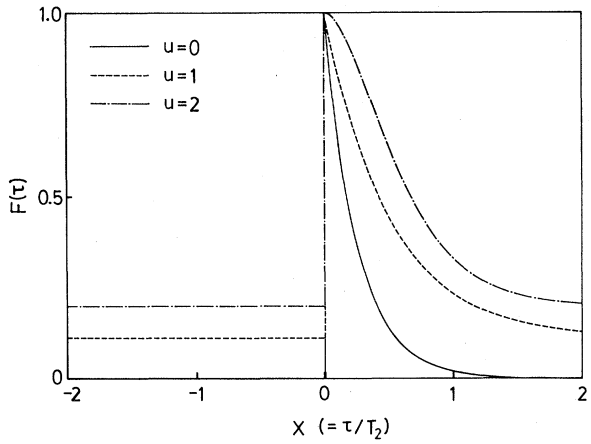

FIG. 3. Correlation profiles of the output intensities with incoherent incident light for $u\ (=T_2/T_1)=0$, 1, and 2 in the extremely inhomogeneous broadening case; each profile is normalized to unity at its peak.

III. COMPARISON WITH THE TRANSIENT FOUR-WAVE MIXING WITH SHORT PULSES

It is very interesting and significant to compare the present results with the signal behaviors of the transient resonant four-wave mixing with two incident short pulses[16,17] (hereafter we call it the two-pulse TRFM). The comparison is made because the situation of the latter case is the same as the present one if both the two incident incoherent light waves are replaced by two coherent light pulses much shorter than T_1 and T_2 with a relative time delay of τ. As discussed in Refs. 16 and 17, the two-pulse TRFM for an inhomogeneously broadened transition becomes the conventional two-pulse photon echo phenomenon in the low-field limit, and therefore has the property peculiar to the echo phenomena that the correlation profile represents the phase relaxation process governed by T_2 not affected by the presence of the dephasing due to the inhomogeneous broadening. The behaviors of the correlation traces in the two-pulse TRFM case are described in detail in Ref. 16 as follows: (a) in the homogeneous broadening case ($\delta\omega=0$), it decreases to zero as proportional to $\exp(-2\gamma_2\tau)$ if $\tau>0$ and is always zero if $\tau<0$, and (b) in the extremely inhomogeneous broadening case ($\delta\omega\rightarrow\infty$), it decreases to zero as proportional to $\exp(-4\gamma_2\tau)$ if $\tau>0$ and is always zero if $\tau<0$. These behaviors are shown in Fig. 4 together with the typical present results for the sake of comparison.

The similarity between the present correlation profiles and those in the two-pulse TRFM is that both profiles reflect the relaxation processes governed dominantly by the phase relaxation time T_2. On the other hand, there are some differences between the present and the two-pulse TRFM cases. The main differences are as follows (see also Figs. 2–4).

(i) The most important difference lies in the fact that the correlation traces of the present four-wave mixing do not necessarily draw the single-exponential decay curves, while those of the two-pulse TRFM always decay single-exponentially by the rate proportional to γ_2.

(ii) The second difference is the presence of the background level, namely, the present correlation profiles have nonzero values at $\tau=\pm\infty$ except in the case $u=0$, while no backgrounds exist in the two-pulse TRFM case.

(iii) The third difference is that the correlation profiles at $\tau<0$ grow up with increasing τ in the homogeneous broadening case, while those in the two-pulse TRFM case are always zero at $\tau<0$.

The expression for $F(\tau)$ in Eq. (2.11) does not easily show the relation between the present results and those of the TRFM process with short pulses. We will show below that this relation can well be understood by regarding the incoherent light as a pulse train consisting of numerous random ultrashort pulses, each of which has a duration corresponding to the correlation time and has no correlation with the others. In order to see this feature, it is convenient to use a δ-function-type correlation function as given in Eq. (2.20) and to transform $F(\tau)$ of Eq. (2.11) by using the general relation

$$\delta(a-b)=\int_{-\infty}^{\infty} dx\,\delta(a-x)\delta(b-x)\,.$$

After transformation, we have

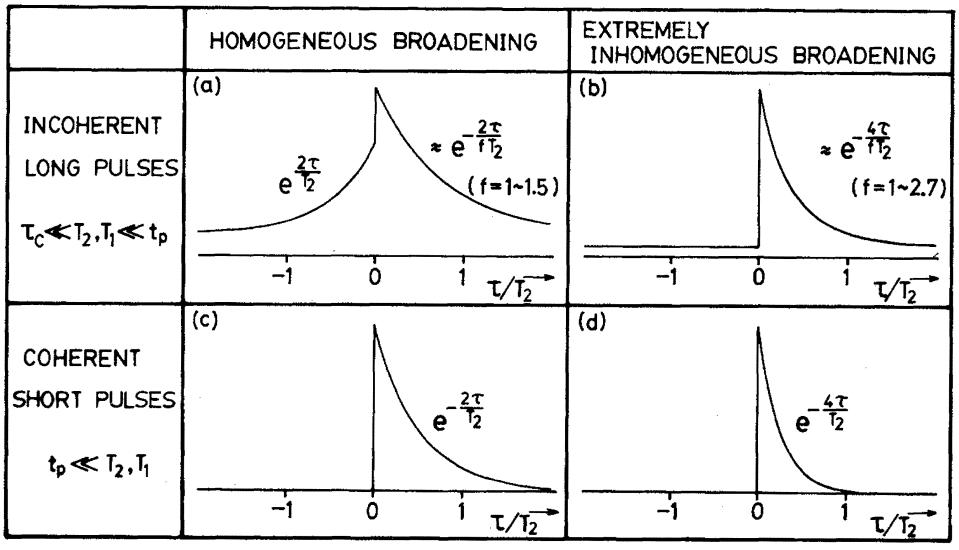

FIG. 4. Typical correlation profiles for the present four-wave mixing with $\tau_c\rightarrow 0$ [(a),(b)] and the two-pulse TRFM with $t_p\rightarrow 0$ [(c),(d)], where t_p and τ_c are the duration and the correlation time of the incident light, respectively; the curves in (a) and (b) are drawn for $u\ (=T_2/T_1)=0.5$. The decay curves in (c) and (d) and the rising curve at $\tau<0$ in (a) are single exponential. Though the decay curves at $\tau>0$ in (a) and (b) are not always expressed by simple functions, they can be approximated to be single-exponential functions of the forms shown nearby the corresponding curves, where f varies with u (see Sec. IV in the text).

$$F(\tau)=D^3A(\tau)+\tfrac{1}{2}D^3B(\tau)\,, \tag{3.1}$$

$$A(\tau)=\int_{-\infty}^{\infty}dt'\left|\int_{-\infty}^{\infty}dt''[H(t',t'',t''-\tau)+H(t'',t',t''-\tau)]\right|^2\,, \tag{3.2}$$

$$B(\tau)=\int_{-\infty}^{\infty}dt'\int_{-\infty}^{\infty}dt''\int_{-\infty}^{\infty}dt'''\,|H(t',t'',t''')+H(t'',t',t''')|^2\,, \tag{3.3}$$

where

$$H(t_a,t_b,t_c)=H_1(t_a,t_b,t_c)+H_2(t_a,t_b,t_c)\,, \tag{3.4}$$

$$H_1(t_a,t_b,t_c)=\int_{-\infty}^{t_r}dt_1\int_{-\infty}^{t_1}dt_2\int_{-\infty}^{t_2}dt_3\delta(t_1-t_a)\delta(t_2-t_b)\delta(t_3-t_c)$$
$$\times\exp[-\gamma_1(t_1-t_2)-\gamma_2(t_r-t_1+t_2-t_3)]G_1(t_r,t_1,t_2,t_3)\,, \tag{3.5}$$

$$H_2(t_a,t_b,t_c)=\int_{-\infty}^{t_r}dt_1\int_{-\infty}^{t_1}dt_2\int_{-\infty}^{t_2}dt_3\delta(t_1-t_a)\delta(t_2-t_c)\delta(t_3-t_b)$$
$$\times\exp[-\gamma_1(t_1-t_2)-\gamma_2(t_r-t_1+t_2-t_3)]G_4(t_r,t_1,t_2,t_3)\,. \tag{3.6}$$

As discussed in Appendix A, $H(t_a,t_b,t_c)$ is proportional to the output field of the transient resonant four-wave-mixing process with three (not two) incident pulses (hereafter we call it the three-pulse TRFM), the field amplitudes of which are proportional to $\delta(t_r-t_a)$, $\delta(t_r-t_b)$, and $\delta(t_r-t_c)$, respectively. These pulses arrive at the material at t_a, t_b, and t_c, respectively, providing the material lies at $\vec{r}=0$.

Equations (3.1)—(3.6) can well be interpreted in terms of the random-pulse-train model of the incoherent light as described before. When the light is divided into two beams and incident on the material, the material senses the two pulse trains, one of which has the wave vector $\vec{k}_2$ and is delayed by τ relative to the other with $\vec{k}_1$. At this time, there occur a number of three-pulse TRFM processes with various combinations of the incident pulses belonging to the two pulse trains. The expression of $F(\tau)$ given in Eqs. (3.1)—(3.6) shows that the present four-wave-mixing process can be interpreted as an assembly of many such three-pulse TRFM processes.

The presence of the two terms proportional to $A(\tau)$ and $B(\tau)$ in the right-hand side of Eq. (3.1) means that the three-pulse TRFM processes occurring in the present situation can be divided into two groups. The first group, corresponding to the first term with $A(\tau)$, consists of the three-pulse TRFM processes with the incident pulses whose fields are proportional to $\delta(t_r-(t''-\tau))$, $\delta(t_r-t'')$, and $\delta(t_r-t')$, designated as p_1, p_2, and p_3, respectively. The pulses p_1 and p_2, separated by τ, have correlation with each other and their phase relation is always constant for any t'' because they come from the same original pulse. Therefore, the output field always has a constant phase for any t'' as long as the pulse p_3 is fixed. This is the reason why the summation over t'' in $A(\tau)$ is not done on the intensities but on the field amplitudes of the outputs, as seen in Eq. (3.2). Because the pulse p_3 has no correlation with p_1 nor p_2, the output fields given for various values of t' have no correlation with each other. For this reason, the output intensities (not the field amplitudes) are summed up over t' in A(τ). On the other hand, the second term with $B(\tau)$ in Eq. (3.1) represents the group consisting of the three-pulse TRFM processes in which the three incident pulses have no correlation among them. Therefore, the output of each three-pulse TRFM process has no correlation with each other. This is the reason why the three integrals in $B(\tau)$ are all performed on the intensities, as seen in Eq. (3.3). This term is, of course, a constant independent of τ and the presence of this term causes the background level to exist in the present correlation traces as mentioned previously in (ii).

Considering the order of the integrals in Eqs. (3.5) and (3.6), we can see that $H_1(t_a,t_b,t_c)$ has a nonzero value only when $t_r\geq t_a\geq t_b\geq t_c$, while $H_2(t_a,t_b,t_c)$ has one only when $t_r\geq t_a\geq t_c\geq t_b$. These conditions represent the orders of the arrival times of the incident pulses at $\vec{r}=0$. Taking these conditions into account, $A(\tau)$ is reduced to the form as

$$A(\tau)=\begin{cases}\displaystyle\int_{-\infty}^{\infty}dt'\left|\int_{-\infty}^{t'}dt''H_1(t',t'',t''-\tau)+\int_{t'}^{t'+\tau}dt''H_1(t'',t',t''-\tau)+\int_{t'}^{\infty}dt''H_2(t''+\tau,t',t'')\right|^2\,, & \tau>0 \qquad (3.7)\\[2ex] \displaystyle\int_{-\infty}^{\infty}dt'\left|\int_{-\infty}^{t'}dt''H_2(t',t''+\tau,t'')\right|^2\,, & \tau<0\,. \qquad (3.8)\end{cases}$$

As discussed in Ref. 17, the three-pulse TRFM in the presence of the inhomogeneous broadening is the lowest-order process of the echo phenomenon with three incident pulses, which is often called the stimulated echo. The output intensity of the three-pulse TRFM behaves in the same way as the stimulated echo intensity; that is, it decreases in the single-exponential manner by the rate proportional to T_2^{-1} with increasing separation between the first and the second pulses, while with increasing separation between the second and the third pulses it decreases

by the rate proportional to T_1^{-1}. The output intensity behaves similarly[16,17] also in the homogeneous broadening case, although the output signal is not a photon echo but a free-induction decay. As seen in the following discussion, the fact that the output behavior of the three-pulse TRFM is more complex than the two-pulse TRFM causes the present correlation traces to be rather complicated, as described in (i).

In $H_1(t',t'',t''-\tau)$ of the first term in Eq. (3.7), τ serves as the separation between the first and the second incident pulses. Therefore, the behavior of $H_1(t',t'',t''-\tau)$ with varying τ represents the decay with the rate proportional to T_2^{-1}. In $H_2(t''+\tau,t',t'')$ of the third term in Eq. (3.7), however, τ corresponds to the separation between the second and the third incident pulses. In this case, it represents the decay with the rate proportional to T_1^{-1}. The behavior of the second term in Eq. (3.7) is more complicated, because τ serves as the separation between the first and the third pulses in $H_1(t'',t',t''-\tau)$ and, moreover, the range of the integral with t'' depends on τ. This term, after all, turns out a multi-exponential function containing both rates T_1^{-1} and T_2^{-1}. Consequently, $A(\tau)$ does not represent such a simple decay as in the case of the two-pulse TRFM.

Finally, the behavior of the correlation trace at $\tau<0$ is given by Eq. (3.8). The presence of this term causes the third property of the present correlation traces as mentioned in (iii). The corresponding term vanishes in the two-pulse TRFM case. Therefore, the behavior at $\tau<0$ is also peculiar to the three-pulse TRFM process. This term, however, vanishes in the extremely inhomogeneous broadening case. This is because the factor $G_4(t_r,t_1,t_2,t_3)$ in this term, representing the dephasing process due to the inhomogeneous broadening, does not have the property of inverting the phase development peculiar to the echo phenomena, while $G_1(t_r,t_1,t_2,t_3)$ has this property.

IV. CONSIDERATION FOR APPLICATION AND CONCLUDING REMARKS

In Sec. II it is shown that we can obtain some information about the relaxation times by the correlation profile in the four-wave-mixing process with the temporally incoherent light whose electric field amplitude is a stochastic stationary Gaussian process. We remarked in Sec. III, however, that the profile does not necessarily give us one of the characteristic relaxation times T_1 and T_2 in such a definite manner as in the usual photon echo phenomena. In this section we further examine the usefulness of the present idea as a practical method to measure the relaxation times.

In order to clarify the decay feature of the correlation profile, we show in Figs. 5 and 6 the semilog representations of $F(\tau)-F(\infty)$, the correlation profiles with the background subtracted, as derived from Figs. 2 and 3, respectively, although the former figures include only the right halves of the latter ones. The right half, where $\tau>0$, of each correlation profile is more significant for practical use, because it remains even in the extremely inhomogeneous broadening case.

When $u=0$, namely, $T_1 \gg T_2$, as we can see from the

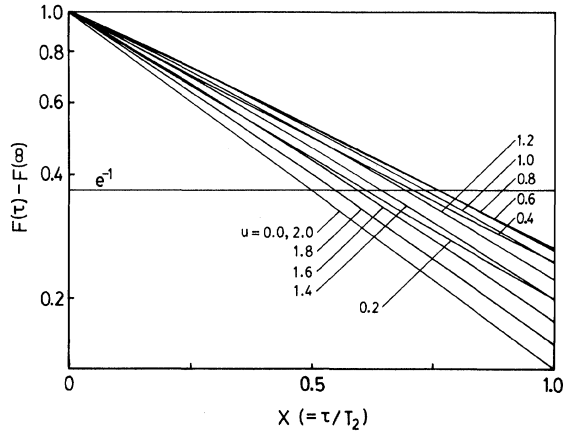

FIG. 5. Semilog representations of the correlation profiles of Fig. 2 minus background levels (homogeneous broadening case); these are displayed only for $\tau>0$. All traces are normalized to unity at $\tau\to 0$. $u=T_2/T_1$.

equations and the figures, the profiles represent the single-exponential decay with the rates $2T_2^{-1}$ and $4T_2^{-1}$ in the homogeneous and extremely inhomogeneous broadening cases, respectively. These decay rates entirely agree with those in the two-pulse TRFM case. In this case we can uniquely determine the relaxation time T_2 by the present method. In condensed matter, such as singlet-singlet transitions of dye molecules, interband transitions of semiconductors, broad absorption bands of impurity ions in insulators and so forth, the phase relaxation time T_2 is generally very short and often falls far below 1 psec. In these transitions, the population relaxation time T_1 is often much longer than T_2. Therefore, the present method is a powerful tool to determine T_2 of these transitions.

In the case $u\neq 0$, the present method cannot definitely determine the relaxation times, except when $u=2$ in the homogeneous broadening case. It can be seen in Figs. 5

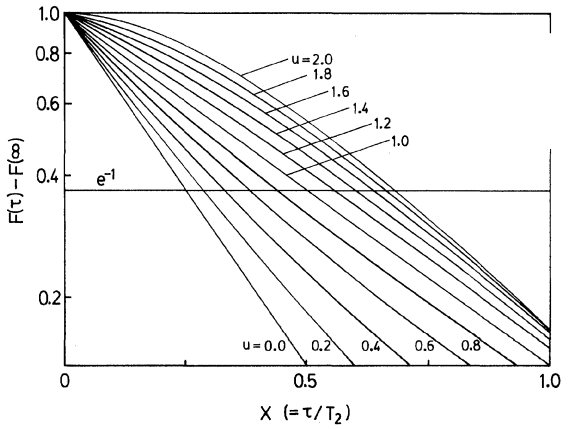

FIG. 6. Semilog representations of the correlation profiles of Fig. 3 minus background levels (extremely inhomogeneous broadening case); these are displayed only for $\tau>0$. All traces are normalized to unity at $\tau\to 0$. $u=T_2/T_1$.

and 6, however, that most profiles are not far from exponential and that for any possible value of u (from 0 to 2) the effective decay time defined by $\tau(e^{-1})$ is not far off the value for $u=0$ where $\tau(e^{-1})$ means the value of τ at which the trace decreases to e^{-1} of its peak. This means that the correlation profile is governed dominantly by T_2 for any value of T_1. Even if we have no information about T_1 at all, it is possible to determine T_2 by $\tau(e^{-1})$ within the error of factors 1.5 and 2.7 in the homogeneous and extremely inhomogeneous broadening cases, respectively. In the extremely ultrafast region of the relaxation process, even such an accuracy is often quite enough.

The theoretical model in the present paper requires a considerably restricted condition for the property of the temporally incoherent light; the complex electric field amplitude of the light is a stochastic stationary Gaussian. It is expected, however, that the basic features of the results may not largely be changed by the statistical property of the incoherent light, as far as the correlation time can be defined. As a practical light source, natural incoherent light such as thermal radiation can hardly be used in the present method because the light source must satisfy two more conditions as follows: the light should have enough high intensity to cause the third-order nonlinear process in the material and should have fairly good directivity, i.e., the transverse spatial coherence, because the output can be taken out only by separating it spatially from the incident light. One of the candidates for useful source is the amplified spontaneous emission from a dye solution. This source will also have a statistical property close to the Gaussian. From a practical point of view, it may be important to try to use a broad-band dye laser light with poor temporal coherence but with good directivity, tunability and intensity, even though probably it cannot be regarded as a Gaussian noise.

Here we note that the present authors have made a preliminary experiment[36] in Na vapor by using a broad-band imperfectly mode-locked cw dye laser and successfully demonstrated the fact that the transient phenomena can be observed with a temporal resolution determined by the correlation time of the light which is much shorter than the pulse duration. The detailed results will be described elsewhere. Very recently, Asaka *et al.*[37] also demonstrated this fact in Nd^{3+}:glass by using a broad-band cw dye laser and an imperfectly mode-locked cw dye laser. They obtained some correlation traces with much longer tails than the correlation time of the light. The decay curves of these profiles were in good agreement with that obtained by the usual photon echo technique, and the decay time of both curves is determined by the phase relaxation time T_2. In their experimental condition, T_2 is much shorter than the population relaxation time T_1. Therefore, their result is consistent with our present theoretical result, as discussed in the beginning of this section. Asaka *et al.*[37] provided also the theoretical analysis. Their analysis, however, is valid only when $T_2 \ll T_1$, while the present analysis can be applied to any case concerning the relaxation rates. A theoretical problem which needs further refinement is to clarify the effect of the statistical property of the incoherent light on the correlation profiles under any condition of relaxation.

The significant point of the present idea is to utilize the correlation technique. In this situation, the information of the coherence in the atomic levels caused by a certain instantaneous field of the incoherent light can be retrieved later by the same field, although the statistical average of the coherence itself falls to zero. In the field of picosecond and femtosecond spectroscopy, a great number of attempts have hitherto been made to produce coherent pulses as short as possible in order to observe the extremely fast phenomena associated with the light-matter interaction. In view of this situation, it is paradoxical and interesting that there exists a great possibility to observe such ultrafast phenomena in the time domain without any difficulty to generate ultrashort pulses.

ACKNOWLEDGMENTS

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APPENDIX A

A two-level system interacting with light field is described by the density matrix formalism. We consider here two nearly resonant light beams with the same frequency ω. The basic equations describing the motion of this system are given as

$$\frac{\partial}{\partial t}\rho_D = -2\frac{i}{\hbar}(H_{ab}\rho_{ba} - \rho_{ab}H_{ba}) - \frac{1}{T_1}(\rho_D - \rho_D^{(0)}) , \quad \text{(A1)}$$

$$\frac{\partial}{\partial t}\rho_{ba} = \frac{\partial}{\partial t}\rho_{ab}^{*} = -\frac{i}{\hbar}H_{ba}\rho_D - \left[\frac{1}{T_2} + i\omega_0\right]\rho_{ba} , \quad \text{(A2)}$$

where $\rho_D = \rho_{aa} - \rho_{bb}$, the subscripts a and b denote the lower and upper levels, respectively, ω_0 ($\simeq\omega$) is the transition frequency, $\rho_D^{(0)}$ is the thermal equilibrium value of ρ_D, and T_1 and T_2 are the longitudinal and transverse relaxation times, respectively. The matrix elements of the interaction Hamiltonian are given by

$$H_{ba} = H_{ab}^{*} = -\mu_{ba}\hat{E}(\vec{r},t)\exp(-i\omega t) + \text{c.c.} , \quad \text{(A3)}$$

$$\hat{E}(\vec{r},t) = \tilde{E}_1(\vec{r},t)\exp(i\vec{k}_1\cdot\vec{r}) + \tilde{E}_2(\vec{r},t)\exp(i\vec{k}_2\cdot\vec{r}) , \quad \text{(A4)}$$

where μ_{ba} is the electric dipole matrix element of the transition, and $\tilde{E}_1, \tilde{E}_2$ and $\vec{k}_1, \vec{k}_2$ are the electric field amplitudes and wave vectors of the two incident beams, respectively.

In the perturbation and rotating-wave approximations, the nth-order density matrix elements proportional to the nth power of the electric field can be calculated as

$$\hat{\rho}_{ba}^{(n)}(\vec{r},t) = \frac{i\mu}{\hbar}\int_{-\infty}^{t} dt_1 \hat{E}(\vec{r},t)\rho_D^{(n-1)}\exp\left[-\left[\frac{1}{T_2} + i\Delta\omega\right](t-t_1)\right] , \quad \text{(A5)}$$

$$\rho_D^{(n)}(\vec{r},t)=\frac{2i\mu}{\hbar}\int_{-\infty}^{t}dt_1[-\hat{E}(\vec{r},t)\hat{\rho}_{ab}^{(n-1)}+\hat{E}^*(\vec{r},t)\hat{\rho}_{ba}^{(n-1)}]\exp\left[-\frac{1}{T_1}(t-t_1)\right], \tag{A6}$$

where $\Delta\omega=\omega_0-\omega$, $\hat{\rho}_{ba}^{(n)}=\rho_{ba}^{(n)}\exp(i\omega t)$, and we assumed $\mu_{bq}=\mu_{ab}=\mu$.

The third-order off-diagonal density matrix element $\hat{\rho}_{ba}^{(3)}$ at ω contains generally four components of different wave vectors $\vec{k}_1$, $\vec{k}_2$, $\vec{k}_3=2\vec{k}_2-\vec{k}_1$, and $\vec{k}_4=2\vec{k}_1-\vec{k}_2$. Taking out only the $\vec{k}_3$ component, we have

$$\begin{aligned}\hat{\rho}_{ba}^{(3)}(\vec{k}_3)=&-2i\rho^{(0)}\left[\frac{\mu}{\hbar}\right]^3\exp(i\vec{k}_3\cdot\vec{r})\\&\times\int_{-\infty}^{t}dt_1\int_{-\infty}^{t_1}dt_2\int_{-\infty}^{t_2}dt_3\{\tilde{E}_2(\vec{r},t_1)\tilde{E}_2(\vec{r},t_2)\tilde{E}_1^*(\vec{r},t_3)\exp[-i\Delta\omega(t-t_1-t_2+t_3)]\\&\qquad+\tilde{E}_2(\vec{r},t_1)\tilde{E}_1^*(\vec{r},t_2)\tilde{E}_2(\vec{r},t_3)\exp[-i\Delta\omega(t-t_1+t_2-t_3)]\}\\&\times\exp[-\gamma_1(t_1-t_2)-\gamma_2(t-t_1+t_2-t_3)],\end{aligned} \tag{A7}$$

where $\gamma_1=T_1^{-1}$, $\gamma_2=T_2^{-1}$, and we assumed $\rho_D^{(0)}=\rho^{(0)}$, and $\rho_{ba}^{(0)}=0$.

When the total electric field of the incident light beams is given as Eq. (2.6), we have

$$\tilde{E}_1(\vec{r},t)=\mathscr{E}(t+\tau-(\vec{n}_1\cdot\vec{r})/v)\exp(-i\omega\tau), \tag{A8}$$

$$\tilde{E}_2(\vec{r},t)=\mathscr{E}(t-(\vec{n}_2\cdot\vec{r})/v). \tag{A9}$$

Substituting Eqs. (A8) and (A9) in Eq. (A7), assuming $\vec{n}_1\simeq\vec{n}_2=\vec{n}$, and using the reduced time $t_r=t-(\vec{n}\cdot\vec{r})/v$, we get the form of Eq. (2.9) in the text.

Next, we consider the case of the three-pulse TRFM with the incident pulses whose electric fields are given as

$$E_a(\vec{r},t)=\hbar\mu^{-1}\theta_a\delta(t-t_a-(\vec{n}_2\cdot\vec{r})/v)\exp[-i\omega(t-t_a)+i\vec{k}_2\cdot\vec{r}]+\text{c.c.}, \tag{A10}$$

$$E_b(\vec{r},t)=\hbar\mu^{-1}\theta_b\delta(t-t_b-(\vec{n}_2\cdot\vec{r})/v)\exp[-i\omega(t-t_b)+i\vec{k}_2\cdot\vec{r}]+\text{c.c.}, \tag{A11}$$

$$E_c(\vec{r},t)=\hbar\mu^{-1}\theta_c\delta(t-t_c-(\vec{n}_1\cdot\vec{r})/v)\exp[-i\omega(t-t_c)+i\vec{k}_1\cdot\vec{r}]+\text{c.c.}, \tag{A12}$$

where θ_j ($j=a$, b, and c) is the pulse area and we assume $t_a>t_b$. In this case, $\hat{\rho}_{ba}^{(3)}(\vec{k}_3)$ consists of two parts; one of them represents the pure three-pulse TRFM process, which means that this part is generated only by mixing all the three incident fields. The other part is quite equivalent to the two-pulse TRFM process, which requires only two incident fields E_c and E_a (or E_b). (The output fields of these two processes cannot be separated in the present situation with only two different directions of incident beams. They, however, can be spatially separated if the three incident pulses have different wave vectors from each other.) The former part is the lowest-order process of the stimulated photon-echo phenomena. The term "three-pulse TRFM" which we used in the text refers to the former part. Taking out only this part, we have

$$\begin{aligned}\hat{\rho}_{ba}^{(3)}(\vec{k}_3)=&-2i\rho^{(0)}\theta_a\theta_b\theta_c\exp[i\vec{k}_3\cdot\vec{r}+i\omega(t_a+t_b-t_c)]\\&\times\int_{-\infty}^{t_r}dt_1\int_{-\infty}^{t_1}dt_2\int_{-\infty}^{t_2}dt_3\{\delta(t_1-t_a)\delta(t_2-t_b)\delta(t_3-t_c)\exp[-i(\omega_0-\omega)(t_r-t_1-t_2+t_3)]\\&\qquad+\delta(t_1-t_a)\delta(t_2-t_c)\delta(t_3-t_b)\exp[-i(\omega_0-\omega)(t_r-t_1+t_2-t_3)]\}\\&\times\exp[-\gamma_1(t_1-t_2)-\gamma_2(t_r-t_1+t_2-t_3)],\end{aligned} \tag{A13}$$

where we assumed $\vec{n}_1\simeq\vec{n}_2=\vec{n}$. Substituting Eq. (A13) in Eq. (2.8), we have

$$\hat{P}^{(3)}(\vec{k}_3)=-2i\rho^{(0)}N\mu\theta_a\theta_b\theta_c\exp[i\vec{k}_3\cdot\vec{r}+i\omega(t_a+t_b-t_c)][H_1(t_a,t_b,t_c)+H_2(t_a,t_b,t_c)], \tag{A14}$$

where $H_1(t_a,t_b,t_c)$ and $H_2(t_a,t_b,t_c)$ are given in Eqs. (3.5) and (3.6), respectively.

The two-pulse TRFM case is realized by letting $E_b=0$ (or $E_a=0$). The expressions for $\hat{\rho}_{ba}^{(3)}(\vec{k}_3)$ and $\hat{P}^{(3)}(\vec{k}_3)$ in this case are consequently given by replacing the subscript b with a (or a with b) in Eqs. (A13) and (A14), respectively.

APPENDIX B

When the distribution function of the transition frequency is given as

$$g(\omega_0)=\frac{1}{\sqrt{\pi}\delta\omega}\exp\left[-\frac{(\omega_0-\omega)^2}{\delta\omega^2}\right], \tag{B1}$$

the explicit form of $F(\tau)$ under the assumption $\delta\omega \ll \omega$ is calculated to be

(i) $\tau>0$

$$F(\tau)=D^3(2\sqrt{2}\gamma_1^2\delta\omega)^{-1}(F_1+F_2+F_3+F_4), \tag{B2}$$

$$F_1=u[(1-a^2)\Phi(a)\exp(a^2)+a], \tag{B3}$$

$$F_2=[4/(2-u)^2]\{4(1-u)^2\Phi(a-x/a)\exp(a^2-4x)+4u(1-u)\Phi(b-x/a)\exp[b^2-(2+u)x] + u^2\Phi(c-x/a)\exp(c^2-2ux)\}, \tag{B4}$$

$$F_3=[8u(1-u)/(2-u)^2]\{2\Phi(a)\exp(a^2)-2\Phi(b)\exp(b^2)+(2-u)a[1-2a\Phi(a)\exp(a^2)]\}\exp[-(2+u)x], \tag{B5}$$

$$F_4=[2u^2/(2-u)^2]\{2\Phi(a)\exp(a^2)-2\Phi(c)\exp(c^2)+2(2-u)a[1-2a\Phi(a)\exp(a^2)] + (2-u)^2a^2[(2a^2+1)\Phi(a)\exp(a^2)-a]\}\exp(-2ux); \tag{B6}$$

(ii) $\tau=0$

$$F(0)=D^3(2\sqrt{2}\gamma_1^2\delta\omega)^{-1}\{(u+4)\Phi(a)\exp(a^2)+3ua[1-2a\Phi(a)\exp(a^2)]+(u^2a^2/2)[(2a^2+1)\Phi(a)\exp(a^2)-a]\}, \tag{B7}$$

(iii) $\tau<0$

$$F(\tau)=D^3(2\sqrt{2}\gamma_1^2\delta\omega)^{-1}\{u[(1-2a^2)\Phi(a)\exp(a^2)+a]+4\Phi(a-x/a)\exp(a^2)\}, \tag{B8}$$

where $x=\tau/T_2$, $u=T_2/T_1$, $a=\sqrt{2}(T_2\delta\omega)^{-1}$, $b=au/2$, $c=a(u-1)$, and $\Phi(x)$ is the error function defined as

$$\Phi(x)=\int_x^\infty dy\exp(-y^2). \tag{B9}$$

From this general form, the expressions in two extreme cases ($\delta\omega\to 0$ or ∞) are derived as in Eqs. (2.21)—(2.26) in the text.

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