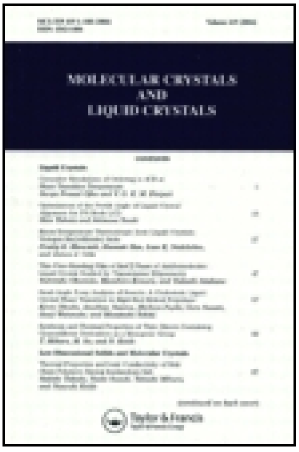

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3-Beam Degenerate 4-Wave Mixing Using Incoherent Light

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3-Beam Degenerate 4-Wave Mixing Using Incoherent Light

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Signal intensity as a function of mutual delay time of three incoherent pump beams in the degenerate four-wave mixing (DFWM) process has been calculated with the second-order correlation time of the fields being much shorter than both the dephasing time T_2 and the inverse of the inhomogeneous broadening width $\delta\omega$. The results show that when broadening is purely homogeneous and extremely inhomogeneous, the curve of the DFWM signal intensity as a function of the delay time τ shows no peak shift, while when the inhomogeneous broadening is finite, the peak shift is related to the dephasing time T_2 and the inhomogeneous width $\delta\omega$, and can thus be used to estimate T_2.

Keywords: dephasing time, inhomogeneous broadening, incoherent beam, four-wave mixing

I. INTRODUCTION

Recently techniques for the measurement of ultrafast dephasing in condensed materials have developed rapidly. Pulses with durations of the order of picoseconds and tens of femtoseconds have become available as powerful tools for ultrafast measurement.[1] However, femtosecond laser devices are still complicated and sophisticated. Even though tunable pulses of durations shorter than 100 fs over the broad visible region have been reported by hybrid mode-locking,[2] it is still inconvenient to perform relaxation experiments using these light sources since the apparatus required is available only in very few laboratories, moreover it is inconvenient to change from one dye laser to another. Instead of using ultrashort pulses, incoherent light with a short correlation time can also achieve a high time resolution, as has been suggested[3] and verified experimentally by several groups.[4–14] In addition, it is easier to produce a wide band incoherent beam with an ultrashort correlation time τ_c than a pulse width; thus the temporally incoherent laser has played an important role in the research of ultrafast phenomena. Degenerate four-wave mixing (DFWM) with a temporally incoherent laser has been widely used to study several ultrafast relaxation processes.[13] In dephasing measurement using DFWM with incoherent beams, the time resolution is limited by the correlation time rather than the pulse width using ultrashort coherent pulses. The relaxation time T_2 determined by the decay tail of the DFWM signal trace becomes ambiguous when T_2 is the same as or shorter than the correlation time τ_c of the pump beams. In these cases, the peak shift of the signal intensity curve as a function of delay time τ between two incoherent beams can be used to estimate the ultrashort relaxation time under some assumptions.[13] By using peak shift for the estimation, a time resolution of several tens of femtoseconds has been reached in recent dephasing measurement experiments using DFWM with two incoherent beams.[13]

Three-beam DFWM has the advantage of a flexible optical arrangement for detecting signals in specific directions of phase matching; thus the general situation of three-beam DFWM is worthy of study. There are many papers on the measurement of ultrafast relaxation processes by DFWM with two incoherent beams.[3,13] In the case of three-incoherent-beam DFWM, however, only the measurement of the relaxation time of a population has been reported.[12] Many problems in dephasing measurements by DFWM with three incoherent beams remain unresolved. Furthermore, many experimental and theoretical studies are limited to two kinds of extreme case: extremely inhomogeneous broadening; and purely homogeneous broadening. The effects of inhomogeneous broadening on dephasing measurement (which may be important in samples such as organic and semiconductor materials) have not yet been well studied. In addition, the peak shift has only been identified in two-incoherent-beam DFWM; here the peak shift is related to inhomogeneous broadening.[13] Therefore, in three-beam-incoherent DFWM, it is necessary to understand whether a peak-shift occurs, and if so, how the shifted value is affected by inhomogeneous broadening of the material system. In this paper, the effect of intermediate inhomogeneous broadening on dephasing measurements by three-incoherent-beam DFWM was examined theoretically. It was found that the peak shift is related to inhomogeneous broadening. Numerical simulation of the peak shift can be used to estimate the dephasing time when the materials measured in experiments have intermediate inhomogeneous broadening. Although this paper only concentrates on dephasing measurements, the results can easily be extended to the study of population relaxation in the case of intermediate inhomogeneous broadening.[13]

II. SIGNAL INTENSITY OF DEGENERATE FOUR-WAVE MIXING

Using the rotating-wave approximation, the three input fields E_i involved in the degenerate four-wave mixing process with vectors $\mathbf{k}_i$ ($i=1$, 2 and 3) are given by

$$E(t,\mathbf{r}) = E_1(t)\exp(i\mathbf{k}_1 \cdot \mathbf{r}) + E_2(t+\tau)\exp(i\mathbf{k}_2 \cdot \mathbf{r}) + E_3(t-T)\exp(i\mathbf{k}_3 \cdot \mathbf{r}). \quad (1)$$

The delay time T and τ are related to the population relaxation time T_1 and the dephasing time T_2, respectively. Here the case when T is much longer than τ is considered. With different combinations of T and τ, various relaxation processes can be treated. For example, by changing T but maintaining $\tau = 0$, the population relaxation time T_1 can be measured by degenerate four-wave mixing with three incoherent beams.[11] This paper concentrates on the measurement of the dephasing time T_2 in a system with finite inhomogeneous broadening by a DFWM process with three incoherent beams. A more general case with a longer delay time T fixed and a relatively small delay time τ meticulously changed to various values was thus chosen.

It was assumed that: the resonant material can be described by a two-level energy system, and that the grating amplitude induced by the three incoherent beams is small, so that perturbation techniques can be applied. In the rotating-wave and electric-dipole approximations, the perturbation expansion of the density matrix is of the third order. Suppose that the signal is detected at a certain direction with a wave vector $\mathbf{k}_s = \mathbf{k}_1 - \mathbf{k}_2 + \mathbf{k}_3$. Then the amplitude of the signal field is proportional to the third-order polarization as follows

$$P_s^{(3)}(t,\tau,T) = C \int_0^{+\infty} d\omega_o g(\omega_o) \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \times \{E_3(t_1 - T)E_2^*(t_2 + \tau)$$
$$\times E_1(t_3)\exp[-i(\omega_0 - \omega)(t - t_1 + t_2 - t_3)] + E_3(t_1 - T)E_1(t_2)E_2^*(t_3 + \tau) \quad (2)$$
$$\times \exp[-i(\omega_o - \omega)(t - t_1 - t_2 + t_3)]\} \times \exp[-(t_1 - t_2)/T_1 -$$
$$(t - t_1 + t_2 - t_3)/T_2]$$

where C is a constant and $g(\omega_o)$ is the distribution function of the resonant frequency ω_o. Here it is assumed that the delay time T is positive and is of the order of T_1, which is much longer than T_2. This means that the nonlinear diffraction of $E_3(t_1 - T)$ through the DFWM process, produced after the transverse relaxation of the transient grating formed by the other two beams (E_1 and E_2) with a variable delay time $\tau(\lesssim 0)$, has disappeared. Therefore, the other two terms proportional to $E_1(t_1)E_2^*(t_2 + \tau) \times E_3(t_3 - T)$ and $E_1(t_1)E_3(t_2 - T)E_2^*(t_3 + \tau)$ are neglected because in these terms the arrival times of the three subsequent electrical fields at the sample are not in the time order of grating-and-diffraction. In addition, when the delay time T is equal to zero, the whole four terms are reduced to two terms proportional to $E_3(t_1)E_2^*(t_2 + \tau)E_1(t_3)$ and $E_3(t_1)E_1(t_2)E_2^*(t_3 + \tau)$ on the right hand side of Equation (2). Therefore equation (2) is suited to cases when $T = 0$ and $T >> T_2$. These two cases will be discussed in detail in this paper, even though it will be seen later that it is not necessary for the delay time T to be much longer than τ, since $T > |\tau|$ is the only limit in the calculation.

The signal intensity of DFWM using incoherent light is proportional to the statistical average of the square of the polarization

$$I_s^{(3)} = \langle |P_s^{(3)}|^2 \rangle \quad (3)$$

where angular brackets indicate the statistical average. From Equation 2 it can be seen that the statistical average in Equation 3 contains four sixth-order moments of E_i, each of which can be reduced to a sum of six products of three second-order moments if a stationary complex gaussian random process is assumed.[3] In cases of inhomogeneous broadening or homogeneous broadening, Equations 2 and 3 can be analytically or numerically calculated.[12] However, in the case of finite inhomogeneous broadening, the calculation is very complicated. For simplification, in the following calculation, the correlation function of the incoherent fields is assumed to be a δ-function

$$\begin{aligned} <E_i^*(t_i)E_j^*(t_j)> &= <E_i(t_i)E_j(t_j)> = 0, \\ <E_i^*(t_i)E_j(t_j)> &= E_i^*E_j\,\delta(t_i - t_j). \end{aligned} \tag{4}$$

The line-shape function, $g(\omega)$, is assumed to be gaussian centered at ω_o with

$$g(\omega) = \frac{1}{\delta\omega\pi^{1/2}}\exp\left[-\frac{(\omega_o - \omega)^2}{\delta\omega_2}\right]. \tag{5}$$

Using Equation 4, the DFWM signal intensity $I_s^{(3)}$ can be obtained by carrying out the ensemble average

$$I_s^{(3)}(\tau,T) = I(\tau,T) + I'(\tau,T). \tag{6}$$

By introducing the ratio of the transverse relaxation time and the longitudinal relaxation time $u = T_2/T_1$, the ratio of the dephasing linewidth and the inhomogeneous linewidth $a = \sqrt{2}/(T_2\delta\omega)$, and the normalized times $x = \tau/T_2$ and $y = T/T_2$, the analytical expressions of $I(\tau,T)$ and $I'(\tau,T)$ can be given as

$$\begin{aligned} I(\tau,T) = {} & T_1^3 a|C|^2\{au^3/(2-u) <1 - \exp[-(2-u)(x+y)]> \\ & \times \exp<-2u(x+y) + a^2 - [(x+y)/a - a]^2> \\ & + u^3/(2-u)^2(\pi^{1/2}/2) <1 - \exp[-(2-u)(x+y)]>^2 \\ & \times \exp[-2u(x+y) + a^2]\,\phi\,[(x+y)/a - a] \\ & + a^2\exp[-2u(x+y) + a^2](\pi^{1/2}/4)\,\phi\,[(x+y)/a - a] \\ & - (1/2)a^2[(x+y)/a - a]\exp<-2u(x+y) + a^2 \\ & - [(x+y)/a - a]^2>\} + I_1(T) + I_2, \end{aligned} \tag{7}$$

where $I_1(T)$ (independent of τ) and I_2 (independent of τ and T) are the background for dephasing measurement.

If $\tau < 0$, $I'(\tau,T)$ can be expressed as

$$
\begin{aligned}
I'(\tau,T) = {} & T_1^3 a|C|^2 \{u(\pi^{1/2}/2)\exp(a^2)\, \phi\, (x/a - a) + au^2\exp[-u(x + y) \\
& + 2x - x^2/a^2] + u^2\pi^{1/2}/(2 - u)\exp[a^2 - u(x + y)] \\
& <1 - \exp[-(2 - u)\,(x + y)] \quad > \phi\,(x/a - a).
\end{aligned}
\tag{8}
$$

Where the function $\phi(x)$ can be written as

$$
\phi(x) = (2/\pi^{1/2}) \int_{-\infty}^{x} \exp(-t^2)dt.
$$

If $\tau > 0$, $I(\tau,T)$ can be expressed as

$$
\begin{aligned}
I'(\tau,T) = {} & T_1^3 a|C|^2 \{u(\pi^{1/2}/2)\exp(-4x + a^2)\, \phi\, (x/a - a) + au^2\exp \\
& \cdot [-u(x + y) - x^2/a^2] + \pi^{1/2}u^2/(2 - u) \\
& \cdot <1 - \exp[-(2 - u)\,(x + y)]> \\
& \times \exp[-u(x + y) - 2x + a^2]\, \phi\, (x/a - a)\}.
\end{aligned}
\tag{9}
$$

Equations 6–9 are the main analytical results for the dephasing measurement by using three incoherent beams in the case of partial inhomogeneous broadening. Here $T > |\tau|$ has been given for the determination of the integral areas in the calculation of Equation 3 to obtain Equations 6–9.

III. DISCUSSION

The signal intensity analytically expressed by equation (6) is too complicated for the physical meaning to be seen. At first, the signal intensity $I_s^{(3)}(\tau,T)$ is plotted against the normalized delay time τ/T_2 with various parameters as shown in Figure 1. To detect T_2 using coherent short pulses, the resolution limit is the pulse width. In the frequency domain, completely incoherent beams with a δ-function correlation correspond to ultrashort pulses in the time domain with a δ-function pulse width. The δ-function correlation approximation means that the correlation width τ_c of the practical pump fields is much shorter than T_1 and T_2 of the atomic system to be detected. In this case the dephasing time T_2 can be approximately estimated by the decay tails of the curves $I_s^{(3)} - \tau$ as shown in Figure 1 even though in Equations 6–9 the signal intensity $I_s^{(3)}$ is not exponential.

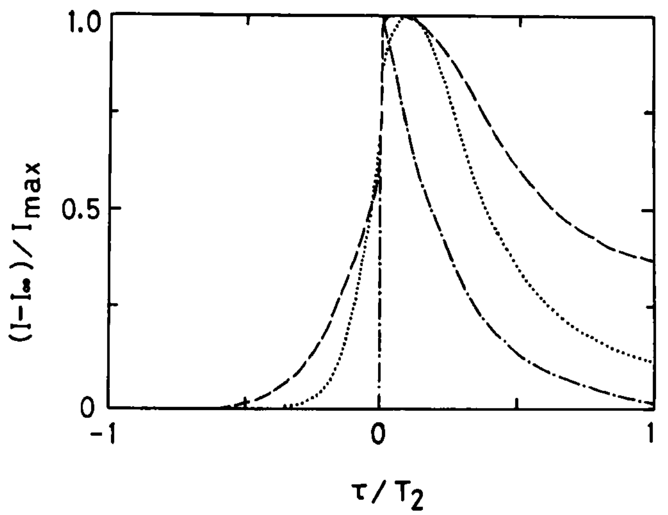

FIGURE 1 Calculated signal intensities of the DFWM with three incoherent beams as a function of normalized delay time τ/T_2 (T_2/T_1 = 0.2; T/T_2 = 10). a) —.—.—.—.$T_2\delta\omega$ = 0; b) ——— $T_2\delta\omega$ = 4; c) . . . . . . $T_2\delta\omega$ = 8.

Equation 7 can be arranged to four groups of signals according to time delay properties

$$I(\tau,T) = I_1(\tau,T) + I_2(\tau) + I_3(T) + I_4 \quad (10)$$

where I_4 is the background, and I_3 is dependent only on T. I_1 and I_2 correspond to photon echo, from which the peak shift is expected.

The determination of T_2 from the tail of the DFWM signal, however, is limited by the width of the pump-source coherence spike. At this time the peak shift must be taken into consideration. In Figure 1 it is clearly seen that the peak of the curve $I_s^{(3)} - \tau$ has shifted from zero delay time ($\tau = 0$). In the case of ultrashort pulses, when T_2 is comparable to the pulse width and thus the tail of the curve $I_s^{(3)} - \tau$ becomes ambiguous for the determination of T_2, the peak shift can be used to estimate T_2.[1] For dephasing measurement using incoherent beams, the width of the coherent spike is usually about several tens to 100 femtoseconds and thus no tail, but a coherent spike, can be detected if the atomic system detected has a T_2 of duration some tens or several femtoseconds. Therefore, again the peak shift as shown in Figure 1 can be used to determine T_2 as has been demonstrated in earlier experiments with two incoherent beams.[10]

In the ideal case, for signal detection of the DFWM with three ultrashort pulses, a finite peak shift exists even though the pulse form in the time domain is assumed to be a δ-function.[3] In the DFWM of two incoherent beams with δ-function correlation, however, the delay time dependence of the signal intensity shows no peak shift in two extreme cases, the purely homogeneous broadening and the extremely inhomogeneous broadening, as has been investigated by Morita and Yajima.[3] In

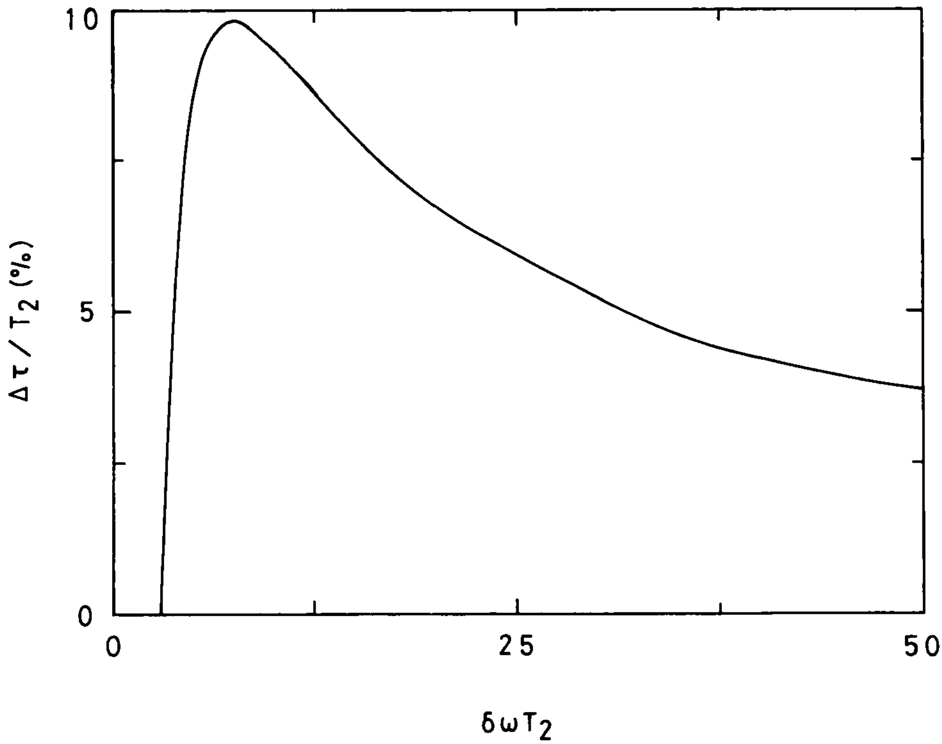

FIGURE 2 Peak shifted values with $T_2/T_1 = 0.2$ and $T/T_2 = 5$, and various inhomogeneous broadening widths $\delta\omega$.

the present case of three incoherent beams with δ-function correlation, the peak shift value $\Delta\tau$ of the curve $I - \tau/T_2$ as a function of τ/T_2 was calculated with fixed $T_2/T_1 = 0.2$ and $T/T_2 = 5$ as shown in Figure 2. The results also show that the peak shift appears as the width $\delta\omega$ of the inhomogeneous broadening takes some intermediate values while in the homogeneous broadening or extremely inhomogeneous broadening cases it disappears as shown in Figure 1 (curve 1). This is one of the main differences between the two extreme cases and the partial inhomogeneous broadening case.

As for the two-beam experiment, a peak shift can still be expected in the three-beam case when the coherent time τ_c of the pump beams is comparable to the dephasing time T_2. Fields with finite width τ_c correspond to the statistical property of the fields with gaussian correlation function. In this case, however, an analytical solution like that of Equations 6–9 is too complicated to obtain for moderately inhomogeneous broadening, and thus only a numerical solution may be available. The present ideal case of δ-function correlation indicates the possibility of the estimation of the dephasing time T_2 by the peak shift if the peak shift value is assumed to be insensitive to pulse correlation width τ_c. As shown in Figure 2, peak shift is related to both T_2 and inhomogeneous width $\delta\omega$. In order to determine T_2

from Figure 1, the inhomogeneous width $\delta\omega$ must be estimated from the absorption-spectrum of the sample.[10]

In principle, two-beam DFWM is one of the special cases of three-beam DFWM. To treat the two-beam case, the parameters of the fields in Equation 3 simply need to be changed to $T = 0$ and $\mathbf{k}_3 = \mathbf{k}_1$ and the calculated results (Equations 6–9) still hold for $T = 0$.

In conclusion, the signal intensity of DFWM of three incoherent beams with a δ-function correlation has been calculated as a function of the delay time τ for a finite inhomogeneous broadening case. The peak shift of three-incoherent-beam DFWM has been identified and found to be related to the product $\delta\omega \cdot T_2$ of the inhomogeneous broadening width $\delta\omega$ and the dephasing time T_2. In addition, this calculation can be extended to the determination of T_1 for intermediate inhomogeneous broadening with a variable delay time T and a fixed $\tau = 0$.

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