Fourth-order coherence-function theory of laser-induced molecular reorientational grating and population grating

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Fourth-order coherence-function theory of laser-induced molecular reorientational grating and population grating

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Résumé. — Nous avons utilisé la théorie des fonctions de corrélation au quatrième ordre pour étudier l'influence des propriétés de cohérence partielle des faisceaux pompe sur les réseaux induits par laser. Tout d'abord, nous examinons la formation du réseau de réorientation moléculaire. Les rôles respectifs des fluctuations de phase et des fluctuations d'amplitude sont dégagés. On propose une méthode de retard temporel pour distinguer le réseau de réorientation moléculaire du réseau de population. Nous appliquons ensuite la théorie au quatrième ordre pour étudier la réflexion de Bragg sur le réseau de population. Nous obtenons une solution analytique qui nous permet d'étudier en détail le comportement temporel du signal de réflexion de Bragg. Cette étude est tout particulièrement utile pour éclaircir le mécanisme de formation du réseau de population.

Abstract. — We have employed fourth-order coherence-function theory to study the influence of the partial-coherence properties of pump beams on the laser-induced gratings. First, we examine the formation of molecular reorientational grating. The different roles of phase fluctuation and amplitude fluctuation have been pointed out. A time-delayed method has been proposed to distinguish molecular reorientational grating from thermal grating. We then apply the fourth-order theory to study the Bragg reflection from a population grating. We obtain an analytic solution which enables us to make an extensive investigation on the temporal behaviour of the Bragg reflection signal. This study is especially helpful for elucidating the generation mechanism of population grating.

1. Introduction.

Recently, a great deal of research works have been devoted to the study of degenerate four-wave mixing (DFWM) because of its ability to generate a phase conjugate wave [1]. When the frequency of the incident beams is near an atomic transition, the resonant nature of the interaction also leads to a strong connection between DFWM and Doppler-free high-resolution nonlinear spectroscopy [2].

DFWM signal comes from the diffraction of light from laser-induced gratings. Since the gratings are originated from the interference of two pump beams in a material, the generation mechanism is influenced by the partial-coherence properties of pump beams. This problem has been studied by Eichler *et al.* [3]. They gave a second-order coherence-function theory, where, the Bragg reflection signal intensity is represented as an absolute square of a nonlinear polarization which is averaged over the stochastic realizations of the electromagnetic field. A similar theory has also been given by Grossman *et al.* [4]. According to these theories, gratings cannot be induced by partially coherent lights from two independent sources. Also, no Bragg reflection signal can be observed when the relative time delay between two pump beams, which are generated from a single laser, is much longer than the coherence time. Since it is the signal intensity which is measured, the correct procedure to treat this problem is to average the absolute square of the polarization over the random variable of the stochastic process. Using this method Trebino *et al.* [5] have studied the effect of pulse-width and grating decay on the formation of thermal grating.

In this paper, we will develop an unified theory which involves fourth-order coherence-function to study the influence of the partial-coherence properties of pump beams on the molecular reorientational grating and population grating. The Bragg reflection signal intensity *versus* relative time delay between two pump beams will be calculated. First, we examine theoretically the formation of molecular reorientational grating. We derive exact expressions for the Bragg reflection signal for two types of radiation model, i.e., amplitude-stabilized radiation and thermal radiation. The different roles of phase fluctuation and amplitude fluctuation have been pointed out. Based on the prediction of fourth-order theory, we propose a time-delayed method to distinguish molecular reorientational grating from thermal grating.

In the second part of this paper, we apply the fourth-order coherence-function theory to study the Bragg reflection from a population grating. The relevant experimental situations to our theoretical consideration involve two correlated pump beams to produce a population grating and a broadband uncorrelated probe beam. The essential difference between population grating and molecular reorientational grating is that the generation of population grating is dependent on transverse relaxation time T_2, while its decay is determined by longitudinal relaxation time T_1. Assuming thermal radiation source with Lorentzian lineshape as pump beams, we obtain an analytic solution for the Bragg reflection signal. One of the relevant problems is the stationary four-wave mixing with incoherent light sources, which was proposed by Morita *et al.* [6] to achieve an ultrafast temporal resolution of relaxation processes. Since they assumed that T_2 is much longer than the light correlation time τ_c, their theory can not be used to study the effect of light bandwidth on the Bragg reflection signal. Asaka *et al.* [7, 8] considered the finite linewidth effect, however the constant background contribution has been ignored in their analysis. Our fourth-order theory includes both the finite light bandwidth effect and constant background contribution. Based on the analytic solution of this model, the influence of various quantities, such as laser coherence time, transverse and longitudinal relaxation time, on the Bragg reflection signal can be investigated in more detail. This study is especially helpful for elucidating the generation mechanism of population grating. In section 2 we study the formation of molecular reorientational grating. Section 3 is devoted to the population grating. Finally, section 4 gives our discussion.

2. Generation of molecular reorientation grating by partially coherent light.

Consider an optical Kerr medium which is illuminated by two partially coherent pump beams with the same mean frequency ω. The complex pump waves E_1 and E_2 can be written as:

$$E_i(\mathbf{r}, t) = \varepsilon_i\, u_i(t) \exp[i(\mathbf{k}_i \cdot \mathbf{r} - \omega t)]\,, \qquad i = 1, 2 \quad (1)$$

where ε_i and $\mathbf{k}_i$ are the constant field magnitude and the wave vector of two pump beams, respectively. $u_i(t)$ is dimensionless field quantity which contains the time dependence of the pump beam fields, i.e., phase and amplitude fluctuations. The expectation value of the modulus of $u_i(t)$ is assumed to be unity.

The order parameter Q of the molecular reorientational grating induced through the interaction of two pump beams with molecules is governed by the following equation of motion [9]:

$$\frac{\partial Q}{\partial t} = -\frac{Q}{T_\mathrm{D}} + \frac{4}{3\,\nu}\,\Delta\alpha\, E_1\, E_2^*\,, \quad (2)$$

where $T_\mathrm{D} = \nu/5\, k_\mathrm{B}\, T$ is the Debye relaxation time, ν is a viscosity coefficient for an individual molecule and $\Delta\alpha = \alpha_\parallel - \alpha_\perp$ is optical polarizability anisotropy. Equation (2) can be solved formally, we have:

$$Q(t) = \frac{4\,\Delta\alpha}{3\,\nu} \int_0^\infty \mathrm{d}t'\, E_1(t - t')\, E_2^*(t - t') \times \times \exp(-\gamma_\mathrm{D}\, t')\,, \quad (3)$$

where $\gamma_\mathrm{D} = T_\mathrm{D}^{-1}$.

The molecular reorientational grating is probed by a light source with complex field amplitude

$$E_3(\mathbf{r}, t) = \varepsilon_3\, u_3(t) \exp[i(\mathbf{k}_3 \cdot \mathbf{r} - \omega_3\, t)]\,. \quad (4)$$

Here, the mean frequency ω_3 is not necessary to be equal to the pump beams frequency ω. The induced polarization which is responsible for Bragg reflection is:

$$\begin{aligned} P(\mathbf{r}, t) &= \frac{2}{3} N\, \Delta\alpha\, Q E_3(\mathbf{r}, t) \\ &= \frac{8\, N(\Delta\alpha)^2}{9\,\nu}\, \varepsilon_1\, \varepsilon_2^*\, \varepsilon_3 \\ &\quad \times \exp\{i[(\mathbf{k}_1 - \mathbf{k}_2 + \mathbf{k}_3) \cdot \mathbf{r} - \omega_3\, t]\} \\ &\quad \times \int_0^\infty \mathrm{d}t'\, u_1(t - t')\, u_2^*(t - t')\, u_3(t) \\ &\quad \times \exp(-\gamma_\mathrm{D}\, t')\,. \end{aligned} \quad (5)$$

The Bragg reflection signal intensity is proportional to the average of the absolute square of the polarization over the random variable of the stochastic process. If pump beams are from a single source with relative time delay τ, i.e., $u_1(t) = u(t)$, $u_2(t) = u(t - \tau)$, while the probe beam is from an independent source, we have the signal intensity $I(\tau) = \beta K(\tau)$ with:

$$K(\tau) = \int_0^\infty \mathrm{d}t' \int_0^\infty \mathrm{d}s' \langle u(t - t')\, u(t - s' - \tau) \times \times u^*(t - t' - \tau)\, u^*(t - s') \rangle \exp[-\gamma_\mathrm{D}(t' + s')]\,. \quad (6)$$

Equation (6) indicates that Bragg reflection signal is dependent on the fourth-order coherence-function of the pump beam. For stationary process $K(\tau)$ is independent of t.

The fourth-order coherence function can be expressed in terms of the second-order coherence function. The recurrence relation depends on the statistical model of the laser field. First, consider the amplitude-stabilized radiation with Lorentzian lineshapes, which has been studied by Picinbono *et al.* [10]. According to equation (59) in reference [10] we can show that :

$$\langle u(t-t')\,u(t-s'-\tau)\,u^*(t-t'-\tau)\,u^*(t-s')\rangle = \exp[-\alpha(|\tau| + |t'-s'| - ||t'-s'| - \tau|)]\,, \tag{7}$$

where $\alpha = \delta\omega/2$ with $\delta\omega$ the linewidth (FWHM) of laser source. Evaluating the integral in (6) yields :

$$K(\tau) = \frac{2\alpha}{\gamma_D^2(\gamma_D + 2\alpha)} \times \times \left\{\frac{\gamma_D}{2\alpha} + \exp[-(\gamma_D + 2\alpha)|\tau|]\right\}. \tag{8}$$

We have the normalized signal intensity

$$\eta(\tau) = I(\tau)/I(0) = \frac{1}{\gamma_D + 2\alpha} \times \{\gamma_D + 2\alpha \exp[-(\gamma_D + 2\alpha)|\tau|]\}. \tag{9}$$

Figure 1 is the theoretical curve of (9). When the molecular reorientational relaxation time is much longer than the laser coherence time, i.e., $\gamma_D \ll \alpha$, we have :

$$\eta(\tau) \simeq \frac{\gamma_D}{2\alpha} + \exp(-2\alpha|\tau|) \simeq \exp(-2\alpha|\tau|). \tag{10}$$

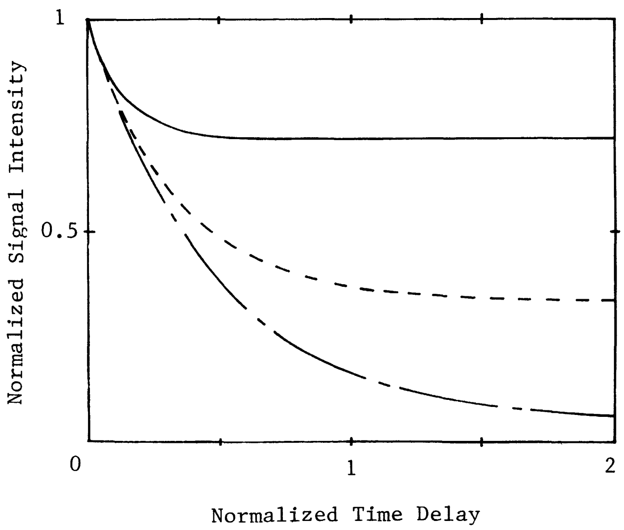

Fig. 1. — Normalized Bragg reflection signal intensity of molecular reorientational grating η *versus* normalized relative time delay $\alpha\tau$ calculated for amplitude-stabilized radiation model. Parameters used in the calculations are $\gamma_D/\alpha = 5$ (——) ; 1 (- - - -) ; 0.1 (– · –).

In this limit signal intensity decreases exponentially as relative time delay between two pump beams increases and our fourth-order theory converts to the second-order theory of Eichler *et al.* [3] and Grossman *et al.* [4]. However, if $\gamma_D \gg \alpha$, we have

$$\eta(\tau) \simeq 1 \tag{11}$$

from (9). In other words, signal intensity is independent of relative time delay when the relaxation time T_D is much shorter than the laser coherence time τ_c. Physically, this novel feature can be understood from the following viewpoint. The Bragg reflection signal intensity is sensitive to the contrast factor of the fringes of molecular reorientational grating, whereas it is insensitive to the induced-grating phase. When $|\tau| \gg \tau_c$, the interference pattern of two pump beams is in constant motion. Now, if $T_D \gg \tau_c$ the integrated effect will cause the induced molecular reorientational grating completely washed out. In this case, no Bragg reflection signal can be observed and the second-order theory is applicable. However, when $\tau_c \gg T_D$ the molecular reorientational grating will follow the field interference pattern whose phase is changing randomly on the scale of coherence time. Since no integrated effect reduces the contrast factor of the induced grating, the Bragg reflection signal intensity should not decrease. In other words, the signal intensity is independent of the relative time delay between pump beams. In this limit the second-order theory fails and our fourth-order theory is of vital importance to give correct results.

In order to clarify the different role of phase fluctuation and amplitude fluctuation, we compare the molecular reorientational grating induced by amplitude-stabilized source with that induced by thermal source. The fourth-order coherence function in (6) for thermal radiation is [11] :

$$\langle u(t-t')\,u(t-s'-\tau)\,u^*(t-t'-\tau) \times \times u^*(t-s')\rangle = \exp(-2\alpha|\tau|) + + \exp(-2\alpha|t'-s'|)\,. \tag{12}$$

Here, Lorentzian lineshape is assumed. After evaluating the integral in (6) we obtain the normalized signal intensity :

$$\eta(\tau) = \frac{1}{2(\gamma_D + \alpha)} \times \times [\gamma_D + (\gamma_D + 2\alpha)\exp(-2\alpha|\tau|)]\,. \tag{13}$$

Figure 2 shows the theoretical curve. When $\gamma_D \ll \alpha$, we have :

$$\eta(\tau) \simeq \frac{\gamma_D}{2\alpha} + \exp(-2\alpha|\tau|)\,. \tag{14}$$

This has exactly the same form as the normalized

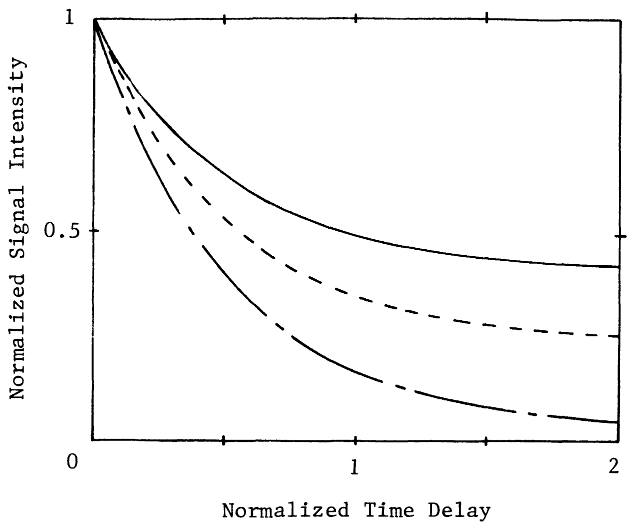

Fig. 2. — Normalized Bragg reflection signal intensity of molecular reorientational grating η *versus* normalized relative time delay $\alpha\tau$ calculated for thermal radiation model. Parameters used in the calculations are γ_D/α = 5 (——) ; 1 (- - - -) ; 0.1 (– · –).

intensity using amplitude-stabilized source as pump beams shown in (10). However, if $\gamma_D \gg \alpha$ we have :

$$\eta(\tau) \simeq \frac{1}{2}\left[1+\exp(-2\,\alpha\,|\tau|)\right], \tag{15}$$

which is quite different from (11) when the pump beam is amplitude-stabilized radiation source. This difference is originated from the amplitude fluctuation of the thermal radiation source. From (2) the order parameter of the molecular reorientational grating is proportional to the product of the amplitude of two pump beams. When the relative time delay between pump beams is shorter than the coherence time, the coincidence of the intensity spikes of two pump beams will enhance the Bragg reflection signal, that explains the τ dependence of the signal intensity in (15) for $\alpha \ll \gamma_D$.

The fourth-order theory shows that Bragg reflection signal decays to zero when $\gamma_D \ll \alpha$, while to a significant amount of constant background when $\gamma_D \gg \alpha$. This phenomenon can be employed to distinguish molecular reorientational grating from thermal grating. As is well known, if the absorption coefficient of a sample at pump beam frequency is not zero, the molecular reorientational grating is usually accompanied by an undesired thermal grating [12]. For most liquid the decay time of thermal grating γ_T^{-1} is of order of microsecond while Debye relaxation time is only of few picoseconds. If the laser linewidth is chosen such that $\gamma_T \ll \delta\omega \ll \gamma_D$, thermal grating can be suppressed when relative time delay between pump beams is much longer than laser coherence time. On the other hand, Bragg reflection signal from molecular reorientational grating is still observable. Experiments have been performed in dye-dissolving benzene and results will be published elsewhere. Another way to eliminate thermal grating is to use cross polarization configuration of pump beams. However, it measures the different component of order parameter tensor.

3. Generation of population grating by partially coherent lights.

In this section we will turn our attention to the influence of the partial-coherence properties of pump beams on population grating. For the sake of simplicity, we treat a homogeneous broadened two-level system interacting with resonant light, which can be written as $\tilde{E}(\mathbf{r},t)\exp(-i\omega t)+\tilde{E}^*(\mathbf{r},t)\exp(i\omega t)$. The basic equations of motion are :

$$\frac{\partial\sigma_{21}}{\partial t} = i\,\frac{\mu}{\hbar}\,\tilde{E}\sigma_D - \gamma_2\,\sigma_{21}\,, \tag{16a}$$

$$\frac{\partial\sigma_D}{\partial t} = 2\,i\,\frac{\mu}{\hbar}\,(\tilde{E}^*\,\sigma_{21} - \tilde{E}\sigma_{21}^*) - \gamma_1(\sigma_D - \sigma_D^{(0)})\,, \tag{16b}$$

where $\sigma_{21} = \rho_{21}\exp(i\omega t)$, $\sigma_D = \rho_{11} - \rho_{22}$ with ρ, the density matrix of the system ; μ, the electric dipole matrix element of the transition ; $\sigma_D^{(0)}$, the thermal equilibrium value of σ_D and $\gamma_1(\gamma_2)$, the longitudinal (transverse) relaxation rate. We have already made rotating wave approximation in (16).

Electric field $\tilde{E}(\mathbf{r},t)$ consists of two pump beams, $\tilde{E}_1(\mathbf{r},t)$ and $\tilde{E}_2(\mathbf{r},t)$, and a probe beam $\tilde{E}_3(\mathbf{r},t)$, i.e.,

$$\begin{aligned}\tilde{E}(\mathbf{r},t) &= \tilde{E}_1(\mathbf{r},t)+\tilde{E}_2(\mathbf{r},t)+\tilde{E}_3(\mathbf{r},t)\\ &= \varepsilon_1\,u_1(t)\exp(i\mathbf{k}_1\cdot\mathbf{r})\\ &\quad+\varepsilon_2\,u_2(t)\exp(i\mathbf{k}_2\cdot\mathbf{r})\\ &\quad+\varepsilon_3\,u_3(t)\exp(i\mathbf{k}_3\cdot\mathbf{r})\,.\end{aligned} \tag{17}$$

Again $u_i(t)$ is time dependent field quantity contains phase and amplitude fluctuation.

Neglecting saturation effect, (16) can be solved by perturbation method under the following perturbation chains :

$$\text{(i)}\quad \sigma_D^{(0)} \xrightarrow{\tilde{E}_1} \sigma_{21}^{(1)} \xrightarrow{\tilde{E}_2^*} \sigma_D^{(2)} \xrightarrow{\tilde{E}_3} \sigma_{21}^{(3)}$$

$$\text{(ii)}\quad \sigma_D^{(0)} \xrightarrow{\tilde{E}_2^*} (\sigma_{21}^{(1)})^* \xrightarrow{\tilde{E}_1} \sigma_D^{(2)} \xrightarrow{\tilde{E}_3} \sigma_{21}^{(3)}$$

These chains denote that the establishment of population grating consists of two steps. First, optical coherence $\sigma_{21}^{(1)}$ is induced through the interation of ground state atoms with one of the pump beam $\tilde{E}_1(\tilde{E}_2^*)$. Then, the interaction with another pump beam $\tilde{E}_2^*(\tilde{E}_1)$ gives rise to a population grating

$\sigma_D^{(2)}$. Since optical coherence decays with time constant T_2, the transverse relaxation time plays an important role in the generation of population grating. This feature characterizes the basic difference between population grating and molecular reorientational grating.

By solving (16) yields the population grating

$$\sigma_D^{(2)}(\mathbf{r}, t) = 2\left(i \frac{\mu}{\hbar}\right)^2 \varepsilon_1 \varepsilon_2^* \exp[i(\mathbf{k}_1 - \mathbf{k}_2)\cdot \mathbf{r}] \int_0^\infty \mathrm{d}t_1 \int_0^\infty \mathrm{d}t_2 \exp[-(\gamma_2 t_1 + \gamma_1 t_2)] \times \\ \times [u_1(t - t_1 - t_2) u_2^*(t - t_2) + u_1(t - t_2) u_2^*(t - t_1 - t_2)] . \quad (18)$$

It is probed by a light field $\tilde{E}_3(\mathbf{r}, t)$ and the nonlinear polarization responsible for Bragg reflection is given by :

$$P(\mathbf{r}, t) = N\mu\sigma_{21}^{(3)} \exp(-i\omega t) \\ = i\hbar N \left(\frac{\mu}{\hbar}\right)^2 \varepsilon_3 \exp[i(\mathbf{k}_3 \cdot \mathbf{r} - \omega t)] \int_{-\infty}^t \mathrm{d}t'\, u_3(t')\, \sigma_D^{(2)}(t') \exp[-\gamma_2(t - t')] . \quad (19)$$

Here N is the atomic density. The Bragg reflection signal intensity, which is the stochastic average of the absolute square of the nonlinear polarization, is :

$$I \alpha \int_{-\infty}^t \mathrm{d}t' \int_{-\infty}^t \mathrm{d}t'' \exp\{-\gamma_2[(t - t') + (t - t'')]\} \langle u_3(t')\, u_3^*(t'')\, \sigma_D^{(2)}(t')(\sigma_D^{(2)}(t''))^* \rangle . \quad (20)$$

If pump beams are from a single source with relative time delay τ, i.e., $u_1(t) = u(t)$, $u_2(t) = u(t - \tau)$, while the probe beam is from an independent source with extra-short correlation time, i.e., $\langle u_3(t')\, u_3^*(t'')\rangle = \delta(t' - t'')$, we have the signal intensity

$$I(\tau) = \beta[K_1(\tau) + K_2(\tau) + 2K_3(\tau)] . \quad (21)$$

Here β is a constant and :

$$K_1(\tau) = \int_0^\infty \mathrm{d}t_1 \int_0^\infty \mathrm{d}t_2 \int_0^\infty \mathrm{d}s_1 \int_0^\infty \mathrm{d}s_2\, f(t_1, t_2, s_1, s_2) \times \\ \times \langle u(t - t_1 - t_2)\, u(t - s_2 - \tau)\, u^*(t - t_2 - \tau)\, u^*(t - s_1 - s_2)\rangle , \quad (22a)$$

$$K_2(\tau) = \int_0^\infty \mathrm{d}t_1 \int_0^\infty \mathrm{d}t_2 \int_0^\infty \mathrm{d}s_1 \int_0^\infty \mathrm{d}s_2\, f(t_1, t_2, s_1, s_2) \times \\ \times \langle u(t - t_2)\, u(t - s_1 - s_2 - \tau)\, u^*(t - t_1 - t_2 - \tau)\, u^*(t - s_2)\rangle , \quad (22b)$$

$$K_3(\tau) = \int_0^\infty \mathrm{d}t_1 \int_0^\infty \mathrm{d}t_2 \int_0^\infty \mathrm{d}s_1 \int_0^\infty \mathrm{d}s_2\, f(t_1, t_2, s_1, s_2) \times \\ \times \langle u(t - t_1 - t_2)\, u(t - s_1 - s_2 - \tau)\, u^*(t - t_2 - \tau)\, u^*(t - s_2)\rangle , \quad (22c)$$

with $f(t_1, t_2, s_1, s_2) = \exp\{-[\gamma_2(t_1 + s_1) + \gamma_1(t_2 + s_2)]\}$. $K_1(\tau)$ and $K_2(\tau)$ are the auto-correlation of population gratings originated from perturbation chain (i) and chain (ii), respectively. $K_3(\tau)$ is the cross-correlation of population gratings from two different chains.

Analytic expressions for $K_i(\tau)$ can be obtained if we assume thermal radiation source with Lorentzian lineshape. We get

$$K_1(\tau) = G_1(\tau) + G_4(\tau),\; K_2(\tau) = G_2(\tau) + G_4(\tau),\; K_3(\tau) = G_3(\tau) + G_5(\tau) ,$$

with

$$G_1(\tau) = \int_0^\infty \mathrm{d}t_1 \int_0^\infty \mathrm{d}t_2 \int_0^\infty \mathrm{d}s_1 \int_0^\infty \mathrm{d}s_2\, f(t_1, t_2, s_1, s_2) \exp[-\alpha(|t_1 - \tau| + |s_1 - \tau|)] = \\ = \left[\frac{1}{\gamma_1(\gamma_2 - \alpha)}\right]^2 \left[\exp(-\alpha|\tau|) - \frac{2\alpha}{\gamma_2 + \alpha} \exp(-\gamma_2|\tau|)\right]^2 , \quad (23a)$$

$$G_2(\tau) = \int_0^\infty \mathrm{d}t_1 \int_0^\infty \mathrm{d}t_2 \int_0^\infty \mathrm{d}s_1 \int_0^\infty \mathrm{d}s_2\, f(t_1, t_2, s_1, s_2) \exp[-\alpha(|t_1+\tau| + |s_1+\tau|)] = \\ = \left[\frac{1}{\gamma_1(\gamma_2+\alpha)}\right]^2 \exp(-2\alpha|\tau|)\,, \tag{23b}$$

$$G_3(\tau) = \int_0^\infty \mathrm{d}t_1 \int_0^\infty \mathrm{d}t_2 \int_0^\infty \mathrm{d}s_1 \int_0^\infty \mathrm{d}s_2\, f(t_1, t_2, s_1, s_2) \exp[-\alpha(|t_1-\tau| + |s_1+\tau|)] = \\ = \frac{1}{\gamma_1^2(\gamma_2-\alpha)(\gamma_2+\alpha)} \left\{\exp(-2\alpha|\tau|) - \frac{2\alpha}{\gamma_2+\alpha}\exp[-(\gamma_2+\alpha)|\tau|]\right\}\,, \tag{23c}$$

$$G_4(\tau) = \int_0^\infty \mathrm{d}t_1 \int_0^\infty \mathrm{d}t_2 \int_0^\infty \mathrm{d}s_1 \int_0^\infty \mathrm{d}s_2\, f(t_1, t_2, s_1, s_2) \exp[-\alpha(|t_1+t_2-s_1-s_2| + |t_2-s_2|)] = \\ = \frac{\gamma_1+\gamma_2+2\alpha}{\gamma_1\gamma_2(\gamma_1+2\alpha)(\gamma_2+\alpha)(\gamma_1+\gamma_2+\alpha)}\,, \tag{23d}$$

$$G_5(\tau) = \int_0^\infty \mathrm{d}t_1 \int_0^\infty \mathrm{d}t_2 \int_0^\infty \mathrm{d}s_1 \int_0^\infty \mathrm{d}s_2\, f(t_1, t_2, s_1, s_2) \exp[-\alpha(|t_1+t_2-s_2| + |s_1+s_2-t_2|)] = \\ = \frac{\gamma_1+\gamma_2+3\alpha}{\gamma_1(\gamma_1+2\alpha)(\gamma_2+\alpha)^2(\gamma_1+\gamma_2+\alpha)}\,. \tag{23e}$$

The Bragg reflection signal is :

$$I(\tau) = \beta\left\{\left[\frac{2}{\gamma_1(\gamma_2-\alpha)(\gamma_2+\alpha)}\right]^2 [\gamma_2\exp(-\alpha|\tau|) - \alpha\exp(-\gamma_2|\tau|)]^2 + \\ + \frac{2(\gamma_1+\gamma_2+2\alpha)}{\gamma_1\gamma_2(\gamma_1+2\alpha)(\gamma_2+\alpha)(\gamma_1+\gamma_2+\alpha)} + \frac{2(\gamma_1+\gamma_2+3\alpha)}{\gamma_1(\gamma_1+2\alpha)(\gamma_2+\alpha)^2(\gamma_1+\gamma_2+\alpha)}\right\}\,. \tag{24}$$

Equation (24) is the main result of this section. Because of its analytic form, the influence of various quantities, such as laser coherence time, transverse and longitudinal relaxation time, on the Bragg reflection signal can be investigated in more detail.

The first term in (24) is just the absolute square of the statistical average of population grating, i.e., $|\langle\sigma_D^{(2)}\rangle|^2$. Unlike the case of molecular reorientational grating, both coherence time and transverse relaxation time influence the temporal behaviour of the signal intensity. The second and third terms give a constant background.

Figure 3 is the theoretical curves of normalized intensity *versus* $\alpha\tau$ for different γ_2/α and γ_1/α. It shows that the decay time of the Bragg reflection signal increases with the decrease of γ_2/α. The underlying physics can better be understood in time domain. As pointed out by Morita *et al.* [6], a light with coherence time τ_c can be considered as composed of a sequence of constant phase subpulses with duration τ_c. When the relative time delay between pump beams is τ, the subpulse pairs within two pump beams can be classified into coherent pairings, which are separated by τ, and incoherent pairings for the other cases [8]. Population gratings generated by coherent pairings have a common phase and therefore grow up coherently. This coherent grating determines the temporal behaviour of τ. On the other hand, gratings generated by incoherent pairings have random spatial phases. This incoherent grating contributes to the Bragg reflection signal intensity a constant background.

Considering the coherent grating, it can be generated only when the separation between coherent pairings is less than T_2, because the optical coherence $\sigma_{21}^{(1)}$ decays with time constant T_2. When $T_2 \ll \tau_c$, overlap between coherent pairings is required. Since the duration of constant phase subpulse is τ_c, coherent grating decays according to $\exp(-\alpha|\tau|)$ in this limit. When T_2 increases, grating can be generated by coherent pairings with longer separation between them. This leads to the increase of the decay time of Bragg reflection signal. In this region, the temporal behaviour of η depends on both γ_2 and α. When $T_2 \gg \tau_c$, the finite coherent time of light can be neglected. In other words, coherent grating is only dependent on T_2, which leads to a temporal behaviour of $\exp(-\gamma_2|\tau|)$ in this limit [6-8].

The constant background is originated from the incoherent grating. Define a parameter C as $C = \eta(\infty)/(1-\eta(\infty))$, which measures the con-

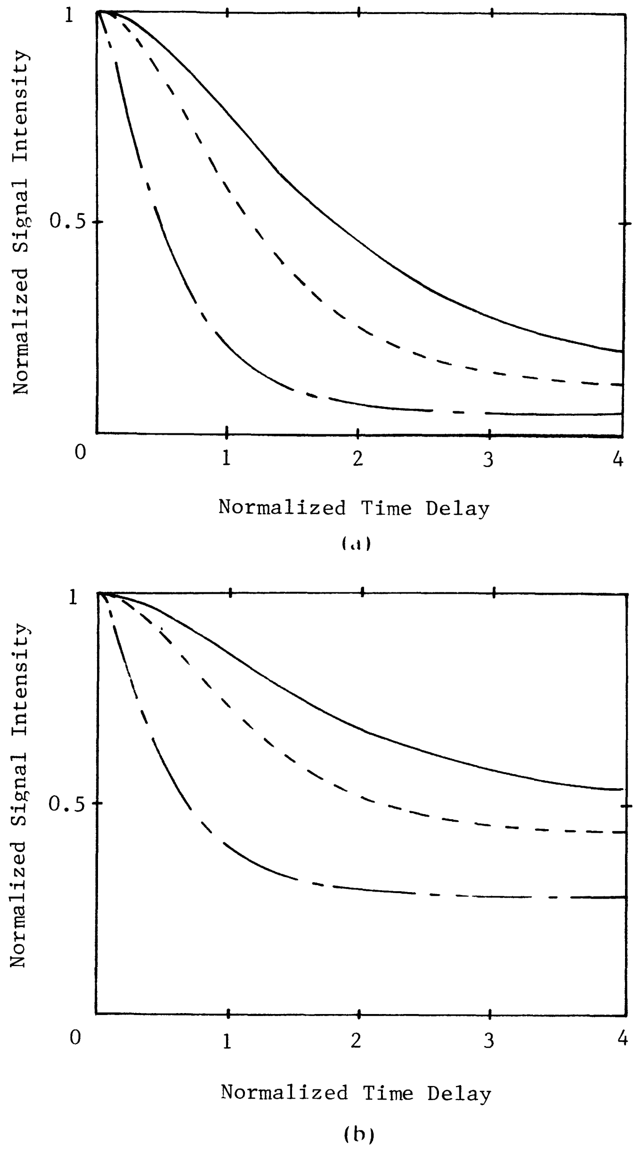

Fig. 3. — Normalized Bragg reflection signal intensity of population grating η *versus* normalized relative time delay $\alpha\tau$. Parameters used in the calculations are (a) $\gamma_1/\alpha = 0.1$ and $\gamma_2/\alpha = 0.5$ (——) ; 1 (- - - -) ; 10 (— · —). (b) $\gamma_1/\alpha = 1$ and $\gamma_2/\alpha = 0.5$ (——) ; 1 (- - - -) ; 10 (— · —).

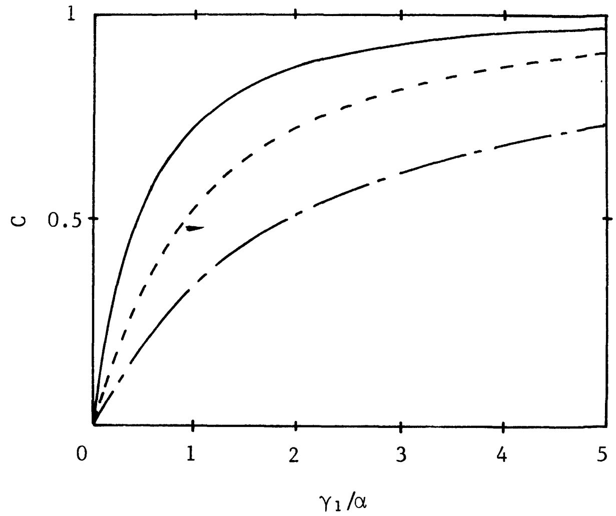

Fig. 4. — Theoretical curves of C *versus* γ_1/α with $\gamma_2'/\alpha = 0.5$ (——) ; 2 (- - - -) ; 100 (— · —).

tribution of constant background to the Bragg reflection signal. Figure 4 is the theoretical curves of C *versus* γ_1/α for different γ_2'/α, where γ_2' is a pure dephasing rate defined as $\gamma_2' = \gamma_2 - \gamma_1/2$. It shows that the constant background increases with the increase of T_2 or the decrease of T_1. This behaviour of background can also be understood in time domain. First, consider the case when $T_1 \gg \tau_c$. Since population grating decays with time constant T_1, both coherent and incoherent grating are results of the accumulation of gratings induced by $N \approx T_1/\tau_c$ pairs when $T_2 \ll \tau_c$. Now, coherent grating grows linearly with N while incoherent grating grows linearly with $N^{1/2}$, we have $C \simeq N/N^2 \sim \gamma_1/\alpha$, which is consistent with (24) in the limit of $\gamma_1 \ll \alpha$ and $\gamma_2 \gg \alpha$. On the other hand, there are about $N(T_2/\tau_c)$ incoherent pulse pairs which can contribute to the incoherent grating when $T_2 \gg \tau_c$, because grating can be induced by two subpulses separated by less than T_2. In this case, we have $C \simeq N(T_2/\tau_c)/N^2 \sim \gamma_1/\gamma_2$, which is consistent with (24) in the limit of $\gamma_1 \ll \alpha$ and $\gamma_2 \ll \alpha$. The situation is different when $T_1 \ll \tau_c$. In this case, no accumulation of population grating can exist, therefore, the incoherent grating is always comparable to the coherent grating. Actually, we have $C \simeq 1$ from (24) in the limit of γ_1, $\gamma_2 \gg \alpha$. It should be noted that T_1 and T_2 influence the constant background in a completely different manner. The accumulation of gratings is determined by the ratio T_1/τ_c. It causes the decrease of C as T_1/τ_c increases. On the other hand, T_2/τ_c determined the number of incoherent pairings which can contribute to the incoherent grating. It leads to an increase of C when T_2/τ_c increases.

4. Discussion.

We have studied the influence of the partially coherent properties of light on molecular reorientational grating and population grating. Although the mechanism for the generation of these two types of gratings is quite different, there exists a connection between them. As mentioned before, transverse relaxation time plays an important role in the generation of population grating. However, when T_2 is extremely short the optical coherence induced by one pump beam will decay quickly. Therefore, no grating can be generated unless the induced optical coherence interacts immediately with the other pump beam. This situation is similar to the generation process of molecular reorientational grating since the order parameter responses directly to the field intensity. Actually, when $\gamma_2 \gg \gamma_1$, α, we have from (24) :

$$\eta(\tau) \simeq \frac{1}{2(\gamma_1 + \alpha)} \times \times [\gamma_1 + (\gamma_1 + 2\alpha)\exp(-2\alpha|\tau|)] . \quad (25)$$

With the decay rate of population grating γ_1 replaced by the decay rate of molecular reorientational grating γ_D, equation (25) converts to (13) exactly.

In section 3 an analytic expression (24) has been derived for the Bragg reflection signal intensity assuming thermal radiation source (which is often used to approximate a multimode laser) with Lorentzian lineshape for the pump beams. This analytic solution enables us to make a detailed investigation on the temporal behaviour of the Bragg reflection signal. Moreover, if the thermal radiation source has a general form of lineshape, a formal expression is still obtainable. The τ dependence of the Bragg reflection signal is :

$$I_{\text{s.o.}}(\tau) = \beta\,|L(\tau)|^2 , \quad (26)$$

with $L(\tau)$ proportional to $\langle \sigma_D^{(2)}(\tau) \rangle$, i.e.,

$$L(\tau) = \int_0^\infty \mathrm{d}t_2 \exp(-\gamma_1 t_2) \int_0^\infty \mathrm{d}t_1 \exp(-\gamma_2 t_1) \times \times [\langle u(t - t_1 - t_2)\, u^*(t - t_2 - \tau)\rangle + \langle u(t - t_2)\, u^*(t - t_1 - t_2 - \tau)\rangle] . \quad (27)$$

It is instructive to note that equation (26) is exactly the result of second-order coherence-function theory. According to the Wiener-Khintchine theorem [13], the relation between field autocorrelation function and the power spectrum $S_0(\Delta)$ is

$$\langle u(t - \tau)\, u^*(t)\rangle \propto \int_{-\infty}^{\infty} \mathrm{d}\Delta\, S_0(\Delta) \exp(-i\,\Delta\tau) . \quad (28)$$

We have from (27)

$$L(\tau) \propto \int_{-\infty}^{\infty} \mathrm{d}\Delta \frac{S_0(\Delta)\exp(i\,\Delta\tau)}{\Delta^2 + \gamma_2^2} . \quad (29)$$

Or the Bragg reflection signal intensity is :

$$I_{\text{s.o.}}(\tau) \propto \left| \int_{-\infty}^{\infty} \mathrm{d}\Delta \frac{S_0(\Delta)\exp(i\,\Delta\tau)}{\Delta^2 + \gamma_2^2} \right|^2 . \quad (30)$$

It is not difficult to show that (30) is equivalent to the corresponding results in references [7, 8]. However, Asaka *et al.* [7] employed rate equation approximation, therefore an assumption of $\gamma_2 \gg \gamma_1$ should be imposed in their derivation at the beginning. Here we note that in the limit of $\gamma_1 \ll \gamma_2$, α, the constant background can be ignored.

In conclusion, we have employed fourth-order coherence-function theory to study the influence of the partial-coherence properties of pump beams on the laser-induced gratings. First, we examine theoretically the formation of molecular reorientational grating. By comparing the Bragg reflection signal for two types of radiation model, i.e., amplitude-stabilized radiation and thermal radiation, the different roles of phase fluctuation and amplitude fluctuation have been pointed out. Based on the prediction of our fourth-order theory, we propose a time-delayed method to distinguish molecular reorientation grating from thermal grating. We then apply the fourth-order theory to study the Bragg reflection from a population grating. Assuming thermal radiation source with Lorentzian lineshape as pump beams, we obtain an analytic solution which enables us to make an extensive investigation on the temporal behaviour of the Bragg reflection signal. This study is especially helpful for elucidating the generation mechanism of population grating.

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