研究

Detailed Expression of Phase Conjugate Wave Generated by Degenerate Four-Wave Mixing in Absorbing Media

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An analytic solution of the phase conjugate wave generated by degenerate four-wave mixing (considering both transmission and reflection gratings) in absorbing media has been obtained. The treatment is based on the volume grating theory, and takes into account spatial distributions of waves in a nonlinear medium. The obtained solution is compatible with all previously reported theoretical solutions. In order to prove several specific aspects of the theoretical results, we have carried out experiments using cryptocyanine dissolved in methanol as an absorbing nonlinear medium; we obtained results which agree with the theoretical predictions.

1. Introduction

The generation of a phase conjugate wave by degenerate four-wave mixing (DFWM) in transparent media could be explained by Yariv and Pepper[1] and by Hellwarth.[2] A DFWM theory for a two-level absorbing medium was presented by Abrams and Lind while neglecting the attenuation of pump beams.[3] If the pump-attenuation effect is considered, the nonlinear coupled wave equations become more complicated; thus, most of the recent studies concerning DFWM using absorbing media involve numerical calculations.[4,5]

In DFWM using absorbing media, it is known that the thermal effect becomes the dominant source of nonlinearity; thus, interference between the forward pump beam and the probe beam generates a transmission thermal grating (longer period) and interference between the backward pump beam and probe beam generates a reflection thermal grating (shorter period). A phase conjugate (PC) signal results from the diffraction of a backward and a forward pump beam by the transmission grating and reflection grating, respectively. Kwon and Lee obtained a nonlinear coupled wave equation for the condition that only a transmission grating is generated, owing to the optical-path delay of the backward pump beam (longer than the coherence length of the laser beam).[6] According to the results of Steel *et al.*,[7] the reflection grating effect should not be neglected, even for a small angle between the forward pump and probe beams.

In this study, we derived nonlinear coupled wave equations in which both the transmission and reflection gratings and the pump-attenuation effect caused by linear absorption is considered, and obtained an analytical expression for the phase conjugate reflectivity. Our theoretical results are in agreement with other existing theories (as the limiting cases) and give a reasonable interpretation of our DFWM experimental results obtained from cryptocyanine dissolved in methanol as a nonlinear medium below the saturation intensity.

2. Theoretical Consideration

The basic geometry of a typical DFWM is shown in **Fig. 1**. Four waves with the same frequency (ω) and polarization are propagating through a nonlinear medium. The forward pump beam ($\vec{E}_f$) and the backward pump beam ($\vec{E}_b$) interact with the probe beam ($\vec{E}_p$) in the nonlinear medium

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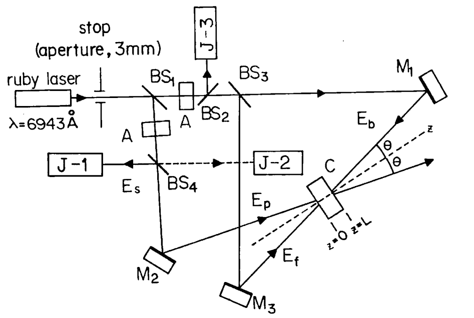

Fig. 1 Geometry of four waves in a nonlinear medium and the experimental arrangement for DFWM. BS_1-BS_4 are beam splitters, M_1-M_3 are full mirrors, A are $CuCl_2$ attenuators, C is a nonlinear cell and J-1, 2, 3 are pulse joule meters.

and a phase conjugate signal beam ($\vec{E}_s$) is generated. The spatial part of the total electric field in the nonlinear medium can be represented as

$$\vec{E}(\vec{r})=\sum_j E_j=\sum_j A_j(\vec{r})\exp(-i\vec{k}_j\cdot\vec{r}), \quad (i=\sqrt{-1}), \tag{1}$$

where j=f, b, p and s. The wave equation describing the field in the nonlinear medium, which has both absorption and dispersion, is given by

$$\nabla^2\vec{E}(r)+k^2\vec{E}(\vec{r})=0, \tag{2}$$

where the propagation constant $k^2=\omega^2\epsilon_r(r)/c^2-i\omega\mu\sigma_0$, In this relation $\epsilon_r(\vec{r})$ is the relative dielectric constant, μ the permeability, σ_0 the average conductivity, and c the velocity f light in free space.[8] The volume grating generated by interference between the incident beams can be expressed by spatial modulations of the relative dielectric constant, represented as:

$$\epsilon_r(\vec{r})=\epsilon_{r0}+(1/2)\{\epsilon_{rt}(\vec{r})\exp(i\vec{K}_t\cdot\vec{r})+\epsilon_{rr}(\vec{r})\exp(i\vec{K}_r\cdot\vec{r})+\text{C. C.}\}, \tag{3}$$

where C. C. is the complex conjugate, ϵ_{rt} and ϵ_{rr} are the modulation depths of the relative dielectric constants in the transmission and reflection gratings, $\vec{K}_t$ and $\vec{K}_r$ are the transmission and reflection volume grating vectors, respectively (subscripts t and r denote the transmission grating and the reflection grating, respectively) and ϵ_{r0} is the average value of $\epsilon_r(\vec{r})$. If we introduce coupling constants κ_t and κ_r as $\kappa_m=\pi\epsilon_{rm}(\vec{r})/2n_0\lambda$ (m=t, r), from the relation between the refractive index and the relative dielectric constant ($\epsilon_{rm}=2n_0n_m$) we obtain κ_m as the following under the assumption that most of the optical materials satisfy $n_0\gg n_m$. That is,

$$\kappa_m=\pi n_m/\lambda, \quad (\text{m}=\text{t, r}), \tag{4}$$

where n_0 and n_m are the average refractive index and changes in the refractive index, respectively.

From Eqs. (1)-(3) we obtain the following coupled wave differential equations from the phase matching conditions of DFWM ($\vec{k}_f=-\vec{k}_b$, $\vec{k}_p=\vec{k}_f+\vec{K}_t=-\vec{k}_f+\vec{K}_r$ and $\vec{k}_s=-\vec{k}_p$) and the slowly varying envelope (SVE) approximation:

$$\frac{dA_f}{dz}\cos\theta+\alpha_0A_f=-i\{\kappa_tA_p+\kappa_r^*A_s\}, \tag{5a}$$

$$\frac{dA_b}{dz}\cos\theta-\alpha_0A_b=i\{\kappa_t^*A_s+\kappa_rA_p\}, \tag{5b}$$

$$\frac{dA_p}{dz}\cos\theta+\alpha_0A_p=-i\{\kappa_t^*A_f+\kappa_r^*A_b\}, \tag{5c}$$

and

$$\frac{dA_s}{dz}\cos\theta-\alpha_0A_s=i\{\kappa_tA_b+\kappa_rA_f\}. \tag{5d}$$

These equations are an extension of Kogelnik's two-wave differential equations to the case of four-waves, where the average absorption coefficient $\alpha_0=\mu c\sigma_0/2n_0$.[8]

Since below saturation the n_m are proportional to the depth of the intensity modulation and the absorption coefficient,[9] from Eq. (4) the coupling constants κ_m are also proportional to them. If we include the contribution of the volume gratings by interference between signal and pump beams,[10] we can express the coupling constants as

$$\kappa_t=C(A_fA_p^*+A_b^*A_s), \quad \kappa_r=C(A_bA_p^*+A_f^*A_s). \tag{6}$$

Here, C is a characteristic constant of the absorbing medium. At $z=0$, $A_fA_p^*\gg A_b^*A_s$; thus, we can express C as $C\simeq(\pi n_t/\lambda A_fA_p^*)=(\pi/\lambda)(dn/dT)(\Phi n_0c\epsilon_0\alpha_0\tau/2\rho C_p)$ for a thermal grating (T is the temperature in the medium, Φ the fraction of the absorbed radiation converted to heat, τ the duration of electric field, ρ the density at constant pressure, C_p the specific heat at constant pressure and ϵ_0 the dielectric constant in free space).[9]

We assume that the intensities of the pump beams are much greater than those of probe and signal beams and approximate the pump beams (with the usual exponential attenuation) as follows:

$$A_f(z)\simeq A_f(0)\exp(-\alpha z) \tag{7}$$

and

$$A_b(z)\simeq A_b(L)\exp[-\alpha(L-z)], \tag{8}$$

where $\alpha=\alpha_0/\cos\theta$ and L is thickness of the nonlinear cell. Then, by substituting Eqs. (7) and (8) into (5c) and (5d) the nonlinear coupled wave equations for A_s and A_p^* are

$$\frac{dA_s}{dz}-[\alpha+iC_1\exp(2\alpha z)+iC_2\exp(-2\alpha z)]A_s=i\gamma_1A_p^* \tag{9}$$

and

$$\frac{dA_p^*}{dz}+[\alpha-iC_1\exp(2\alpha z)-iC_2\exp(-2\alpha z)]A_p^* = i\gamma_2 A_s. \qquad (10)$$

The constants in these equations are defined as

$$C_1=\frac{2C}{cn_0\epsilon_0\cos\theta}I_b(L)\exp(-2\alpha L),$$

$$C_2=\frac{2C}{cn_0\epsilon_0\cos\theta}I_f(0)$$

and

$$\gamma_1=\frac{2C}{\cos\theta}A_f(0)A_b(L)\exp(-\alpha L),$$

$$\gamma_2=\frac{2C}{\cos\theta}A_f^*(0)A_b^*(L)\exp(-\alpha L),$$

where $I_b(L)=(c\epsilon_0 n_0/2)|A_b(L)|^2$ and $I_f(0)=(c\epsilon_0 n_0/2)|A_f(0)|^2$.

In order to solve the above nonlinear coupled wave equations, we define $A_s(z)$ and $A_p(z)$ using $f(z)$ and $g(z)$, the detailed modulation function, as

$$A_s(z)=f(z)\exp\left[\alpha z+i\frac{C_1}{2\alpha}\exp(2\alpha z) - i\frac{C_2}{2\alpha}\exp(-2\alpha z)-i\frac{C_1}{2\alpha}+i\frac{C_2}{2\alpha}\right] \qquad (11)$$

and

$$A_p^*(z)=g(z)\exp\left[-\alpha z+i\frac{C_1}{2\alpha}\exp(2\alpha z) - i\frac{C_2}{2\alpha}\exp(-2\alpha z)-i\frac{C_1}{2\alpha}+i\frac{C_2}{2\alpha}\right]. \qquad (12)$$

By substituting Eqs. (11) and (12) into Eqs. (9) and (10), the ordinary differential equations for $f(z)$ and $g(z)$ can be obtained, respectively, as

$$\frac{d^2}{dz^2}f(z)+2\alpha\frac{d}{dz}f(z)+\gamma_1\gamma_2 f(z)=0, \qquad (13)$$

and

$$\frac{d^2}{dz^2}g(z)-2\alpha\frac{d}{dz}g(z)+\gamma_1\gamma_2 g(z)=0. \qquad (14)$$

Thus, we easily obtain solutions ; the final forms are

$$f(z)=\frac{1}{2}\left[\frac{\gamma_1}{\sqrt{\Delta}}A_p^*(0)+\left(1-\frac{i\alpha}{\sqrt{\Delta}}\right)A_s(0)\right]\times\exp[(-\alpha+i\sqrt{\Delta})z] +\frac{1}{2}\left[-\frac{\gamma_1}{\sqrt{\Delta}}A_p^*(0)+\left(1+\frac{i\alpha}{\sqrt{\Delta}}\right)A_s(0)\right]\times\exp[(-\alpha-i\sqrt{\Delta})z]$$
$$=e^{-\alpha z}\left[\frac{i\gamma_1\sin\{(z-L)\sqrt{\Delta}\}}{\sqrt{\Delta}\cos(L\sqrt{\Delta})+\alpha\sin(L\sqrt{\Delta})}\right]\times A_p^*(0), \qquad (15)$$

and

$$g(z)=e^{\alpha z}\left[\frac{\cos\{(L-z)\sqrt{\Delta}\}+\alpha\sin\{(L-z)\sqrt{\Delta}\}}{\sqrt{\Delta}\cos(L\sqrt{\Delta})+\alpha\sin(L\sqrt{\Delta})}\right]\times A_p^*(0), \qquad (16)$$

where $\Delta=\gamma_1\gamma_2-\alpha^2$. In deriving Eqs. (15) and (16) we used the conditions $A_s(0)=f(0)$, $df(z)/dz|_{z=0}=i\gamma_1 A_p^*(0)$, $A_p^*(0)=g(0)$, $dg(z)/dz|_{z=0}=i\gamma_2 A_s(0)$ and $A_s(L)=f(L)=0$. Therefore, $A_s(z)$ and $A_p^*(z)$ can be obtained by substituting Eqs. (15) and (16) into Eqs. (11) and (12) ; therefore, the reflectivity can be expressed as

$$R=\left|\frac{A_s(0)}{A_p^*(0)}\right|^2 = \left|\frac{(4C/cn_0\epsilon_0\cos\theta)\sqrt{I_f(0)I_b(L)}}{\sqrt{\Delta}\cos(L\sqrt{\Delta})} * \frac{\exp(-\alpha L)\sin(L\sqrt{\Delta})}{+\alpha\sin(L\sqrt{\Delta})}\right|^2. \qquad (17)$$

In the limit of lower reflectivity, if we substitute $A_s\simeq 0$ into Eqs. (9) and (10), we can express $A_p^*(z)$ as $A_p^*(0)\exp(-\alpha z)$ and the reflectivity becomes $R\simeq(|\gamma_1|^2/\alpha)[1-\exp(-\alpha L)]^2$; this is the same as the result obtained by Caro and Gower.[9] In a transparent media ($\alpha\simeq 0$), Eq. (17) becomes $R\simeq\tan^2[(2LC/\cos\theta)\sqrt{I_f(0)I_b(L)}(2/cn_0\epsilon_0)]$, which agrees with the result of Yariv and Pepper.[1] If the absorptions of the pump beams are ignored ($I_f(0)I_b(L)\exp(-2\alpha L)\simeq I_f(0)I_b(L)$), R becomes $|B'\sin(wL)/[w\cos(wL)+\sin(wL)]|^2$, where $B'=[4C/cn_0\epsilon_0\cos\theta]I_f(0)I_b(L)$ and $w=\sqrt{B'^2-\alpha^2}$. This coincides with the result of Abrams and Lind.[3] Therefore, we know that the analytical expression of reflectivity represented in Eq. (17) agrees with several previous theories concerning DFWM in the limiting cases and can be applied to any absorbing media if we know the absorption coefficients.

3. Experimental Procedure

The experimental arrangement is shown schematically in **Fig. 1**. Experiments were performed using a linearly polarized TEM_{00} mode beam from a 50-ns pulse duration and a Q-switched ruby laser with a beam diameter of 3 mm. The nonlinear medium was cryptocyanine dissolved in methanol. The incoming laser beam was split into three separate beams using beam splitters BS_1(R=4%) and BS_3(R=50%). The optical path differences among the three incident beams were less than the coherence length of the ruby laser. The angle between the wave vectors of the forward pump beam and the probe beam was fixed at 2°. The intensities of the incident beams were controlled by using absorbing cells with various concentrations of a $CuCl_2$ aqueous solutions. Experiments were performed below the saturation intensity of the cryptocyanine solution[11] and energies of the PC signal beam, probe beam and pump beams were measured at positions J-1, J-2 and J-3, respectively, using pyroelectric joule meters.

4. Results and Discussions

In this section, we graphically show results of our analytical theory and experimental data for several interesting cases. The open circles show our experimental results. In **Fig. 2.** the reflectivity is presented as a function of the molar concentration (or the value of C) of the nonlinear medium (from 10^{-7} mol/l to 10^{-5} mol/l when the thickness of nonlinear medium is 5.0 mm, $I_f = I_b = 5.6$ MW/cm^2 and $I_p = 0.5$ MW/cm^2). The reflectivity has a maximum value of 14.0% when the molar concentration is 8×10^{-6} mol/l ($C = 3.5 \times 10^{-4}$ F·cm/MWs) and the absorption coefficient $\alpha_0 = 1.23$ cm^{-1}. At this concentration, n_t was 4×10^{-6} when $I_f(0) = 3$ MW/cm^2 and $I_p(0) = 0.3$ MW/cm^2;[6] then, C could be calculated as $C = [2\pi n_t / \lambda n_0 \epsilon_0 c \times (I_f(0) I_p(0))^{-1/2}] = 3.5 \times 10^{-4}$ F·cm/MWs, with $\epsilon_0 = 8.9 \times 10^{-14}$ F/cm and $n_0 = 1.33$. **Figure 3** shows the reflectivity as a function of the thickness of the nonlinear medium when the molar concentration is 8×10^{-6} mol/l and R has a maximum value of 14.0% when the thickness is 4.6 mm. For higher dye concentrations and larger cell thickness, as the reflectivity drops owing to an increasing exponential absorption.

The change in the phase conjugate reflectivity for varying intensities of the probe beam and pump beams is plotted in **Fig. 4** (a) and (b), respectively, for a molar concentration of 8×10^{-6} mol/l and a nonlinear cell thickness of 4.0 mm. The solid lines are plotted according to Eq. (17) and the dashed lines refer to the previous theory which ignores effects of the reflection grating.[6] As shown in **Fig. 4**(a), when the intensity of the probe beam increases from 0.2 to 2 MW/cm^2 at a constant pump intensity of 5.6 MW/cm^2, the experimental reflectivities are nearly constant with a value of 13.9%. This result agrees with the analytic solution derived in the previous section but is higher than that predicted in the absence of a reflection grating (3.0%). When the intensities of the backward and forward beams are simultaneously

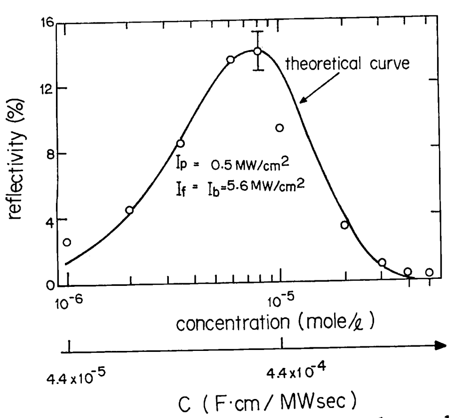

Fig. 2 Reflectivity as a function of the molar concentration of cryptocyanine and the value of C. The solid line is the theoretical curve.

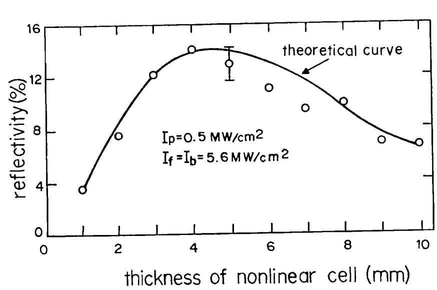

Fig. 3 Reflectivity versus the thickness of the nonlinear cell. The solid line is the theoretical curve.

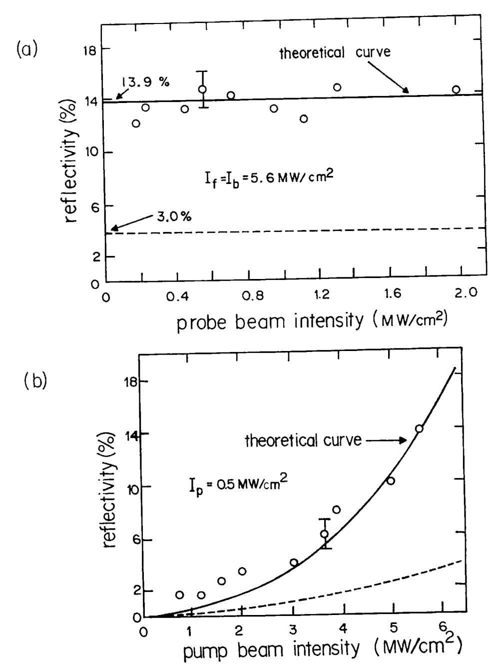

Fig. 4 (a) Reflectivity versus probe beam intensity. The solid line is due to the present theory and the dashed line is from a theory which considers only a transmission grating. (b) Reflectivity versus the pump beam intensities. The solid line is the theoretical curve and the dashed line is for a theory considering only the transmission grating.

changed from 0.7 to 5.6 MW/cm^2 for a fixed probe beam intensity of 0.5 MW/cm^2, the reflectivity increases and the profile is nearly proportional to the square of the pump beam intensity. Our theory (solid line), including both the transmission and reflection gratings, shows satisfactory agreement with the experimental results (in contrast to a previous theory (dashed line)[6] which neglects the reflection grating). This large discrepancy is due to constructive interference between two phase conjugate signals from the transmission and reflection gratings which contain the interference between the signal and pump beams (Eq. (6)). Martin's[12] measurements of the relaxation times of the thermal transmission and reflection volume gratings in methanol when the angle between the forward and probe beams is 2°, show relaxation times of about 30 and 1 μs, respectively. These are very long compared to the pulse width of a ruby laser beam (50 ns); thus, the contribution from the reflection grating must be considered in a correct analysis of the phase conjugate wave and to obtain large signals.

5. Conclusion

We derived an analytical solution for degenerate four-wave mixing in an absorbing media while considering both the transmission and reflection thermal gratings simultaneously. When there is coherence among the forward, backward, probe and signal beams we could obtain a much stronger signal; the present theory gives a correct theoretical interpretation of the experimental results. This theory, which considers only the transmission grating, should show large deviation from the experimental results. If we align the DFWM system carefully while keeping coherence among the four waves, we can obtain a maximum efficiency in the generation of a PC signal.

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