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Investigation of degenerate four-wave mixing and phase conjugation in organic dye solutions*

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Abstract

Phase conjugation of backward-and forward-wave produced by degenerate four-wave mixing using a nearly resonantly enhanced nonlinearity in four-level organic dye solutions was investigated. The conversion efficiency of 22% has been obtained in a 2.5×10^{-5}-molar solution of chlorophyll A in methylalcohol with a 5mm interaction length and a pumping energy of ~8 mj from SH of a *Q*-switched Nd:YAG. Degenerate four-wave mixing by two-photon resonant-enhancement in the solution of rhodamine 6G in alcohol was investigated. Measurements were performed which described the dependance of nonlinear reflectivities on the molar concentrations of the media and the intensities of the pumping and object waves. Phase conjugation characteristics of forward-wave in four-wave mixing process were theoretically analysed. Experiments have confirmed the theoretical prediction. The method of quartet degenerate four-wave mixing has been extended to the doublet degenerate case. Using this method, image conversion from 1.06 μm to 5320 Å has been realized with a conversion efficiency of 25% in a 5 mm long saturable absorber made of organic dye 9740 solution in dichloroethane. Theoretical analysis shows that using a medium, which is a near resonant absorber for the infrared object wave but transparent for the visible reconstruction wave, the conversion efficiency may be well in excess of unity.

Introduction

D. M. Bloom et al[1]. proposed that the nonlinear reflectivities can be enhanced in degenerate four-wave mixing (DFWM) using a resonantly enhanced Kerr nonlinearity. Liao, Bloom and Economou first observed resonantly enhanced DFWM in an absorptive two-level medium of atomic sodium vapor[2] and then, in a three-level system of ruby[3]. For the two-level system, the absorption band is very narrow and thus the pumping beam must be accurately tuned to the resonance lines of sodium. Moreover, the backward reflection wave may be interfered by fluorescence of the medium at same wavelengths. The three-level scheme has a number of distinct advantages over the two-level one, but the lifetime of its upper level is long, and so is the response time. We have demonstrated DFWM in four-level organic dye solutions[4]. Their very broad and nearly continuous absorption bands allow operation over a wide range of wavelengths without stringent

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requirements on the pumping laser's wavelength. Secondly, the relaxation times between their levels are very short, and hence they have fast response. Thirdly, peaks of absorption bands of various dye molecules are different and thus an appropriate organic dye can be selected according to one's need. Finally the organic dye solution can be prepared and its concentration can be changed easily.

Using SH of a Q-switched Nd:YAG as the pumping source, we have observed DFWM in various organic dye solutions. For a 2.5×10^{-5}-molar solution of chlorophyll A in methylalcohol with an interaction length of 5mm and a pumping energy of$\sim$8 mj, nonlinear reflection coefficients of 22% have been obtained. Under the same condition, nonlinear reflectivities of 10% for rhodamine 6G solution in alcohol have been achieved. The effect of the backward-wave phase conjugation has been investigated with an aberration plate.

DFWM by two-photon resonant-enhancement was investigated. Using $\sim$70 mj pumping pulse from a Q-switched Nd:YAG laser, for a medium of 10^{-4}-molar rhodamine 6G solution in alcohol, nonlinear reflectivities of 14% were achieved with an interaction length of 5mm.

We have measured the dependence of the nonlinear reflectivities on the molar concentrations of the media and on the intensities of the pumping and object waves.

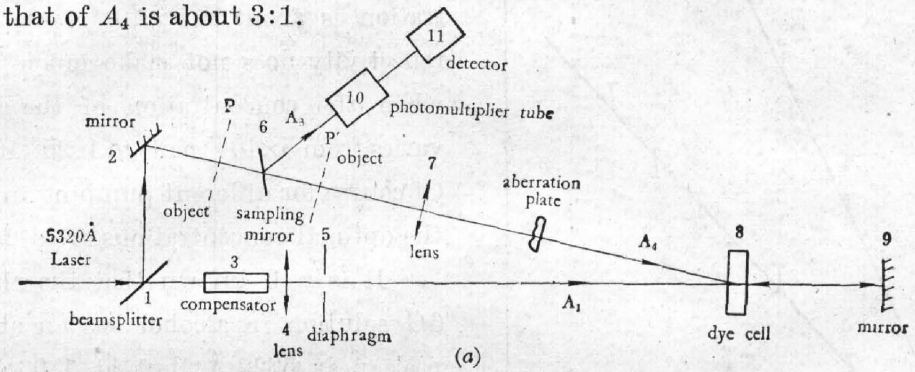
Phase-conjugate forward-wave in four-wave mixing was theoretically analysed. Assuming that two waves, one of them is intense and another is weak, exert on a nonlinear medium and travel at θ, it was proved that when a certain phase-matching condition is satisfied, a phase-conjugate wave in the forward direction at $-\theta$ can be generated. Using SH of a Q-switched Nd:YAG laser, the phase-conjugate forward-wave in rhodamine 6G solution was experimentally demonstrated. It proved the theoretical results.

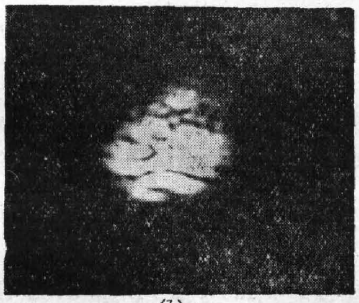
In this paper, the quartet DFWM has been extended to doublet case. By using this method image conversions from 5320 Å to 6300 Å and from 1.06 μm to 5320 Å have been achieved. Using $\sim$5 mj pumping pulse from a Q-switched Nd:YAG laser conversion efficiency of 25% from 1.06 μm to 5320 Å has been obtained by saturable absorber of organic dye 9740 solution in dichloroethane with an interaction length of 5 mm. The theoretical analysis shows that using a medium which is resonantly absorptive at the infrared object wave but transparent at the visible reconstruction wave, the conversion efficiency may be much larger than unity. This scheme of infrared-to-visible image conversion has a number of advantages: real-time, fast response and high efficiency.

DFWM in organic dye solutions

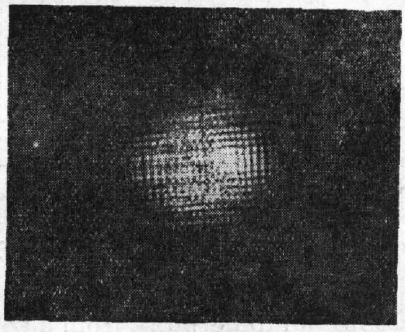
Our experimental apparatus is shown in Fig. 1. A 5320 Å laser pulse from SH of a Q-switched Nd:YAG laser is used as the pumping sources. The FWHM of the pulse is

about 10 nsec. The beam is splitted by beamsplitter 1. The transmitted beam passes through compensator 3, lens 4 with long focal length ($f=1.3$ m), diaphragm 5, and then is impinges on dye cell 8, this beam is denoted as A_1. The reflected beam from the beamsplitter is reflected by mirror 2 and then passes through half-reflective sampling mirror 6 and lens 7 with a focal length of 1m. This beam is also incident on the dye cell, it is denoted as A_4. The angle between A_1 and A_4 is small. Mirror 2 is adjusted to make A_1 and A_4 meet in the dye cell. In our experiments the angle is about 6.2×10^{-2} rad. Beam A_1 passes through the cell and is reflected by mirror 9. Adjusting 9 would make the reflected beam (denoted as A_2) travel in the opposite direction to A_1. The backward-wave is sampled by mirror 6 with reflection/transmission$\approx1:1$ and is detected by photomultiplier 10 (IP28) and photoelectric galvanometer 11. In most of the experiments, the ratio of the intensity of A_1 to that of A_4 is about 3:1.





(b)



(c)

Fig. 1 Experimental arrangement for DFWM (a). Backward-wave field (b). Reconstructed field of the object wave (c).

For a 2.5×10^{-5} molar solution of chlorophyll A in methylalcohol with an interaction length of 5mm, keeping beam A_1 unchanged, I_3 is measured for different intensities of I_4. The 22% conversion efficiency from the incident object beam I_4 to the backward-wave I_3 is obtained. The experimental results are depicted in Fig. 2 as curve a.

If a grid object is placed at P in Fig. 1a, we can obtain the backward-wave reconstruction image of the object P, as shown in Fig. 1b and 1c. When an aberration plate is placed in beam A_4 between the sampling mirror and the dye cell, the reconstructed images are not degraded since the object beam and the backward-wave all pass through

the plate and the phase distortion is thus compensated. If a pure phase object is placed in the position P', no image is observed due to backward-wave phase conjugation character.

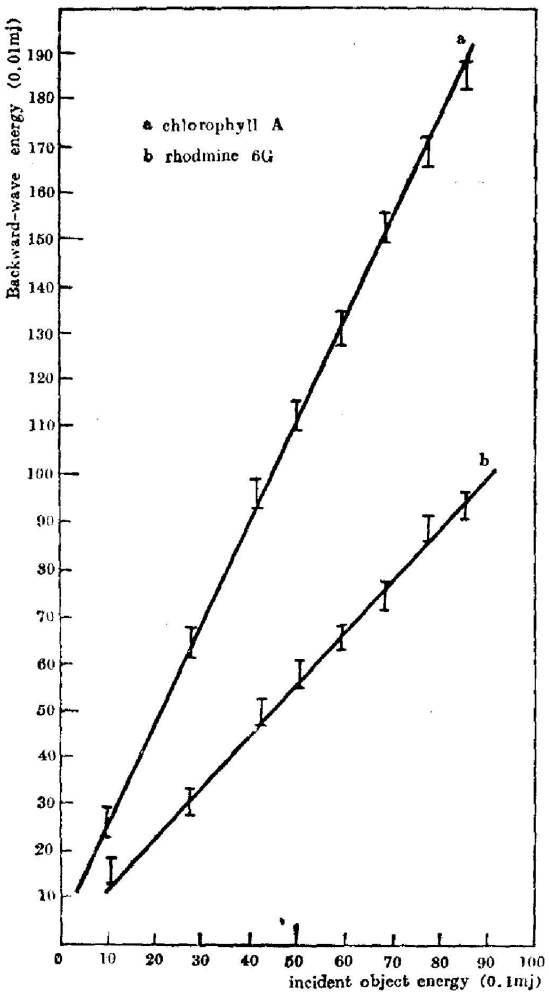


Fig. 2 Results of the experimental measurements of backward-wave energy as a function of incident object wave energy. (*a*) chlorophyll A and (*b*) rhodamine 6G

We have investigated DFWM in rhodamine 6G solution in alcohol. Under the above conditions, nonlinear reflectivities of 10% were obtained, as shown in the curve b of Fig. 2.

The effects of the concentrations of the solutions were investigated. Under our experimental conditions, the optimal concentration is about 2.5×10^{-5} mole and the reflectivity does not make much variation when the concentration of the solutions varies from 5×10^{-5} mole to 1.25×10^{-5} mole. Of course for different pumping intensities, the optimal concentrations are different.

It is well known that the rhodamine 6G solution in alcohol has an absorption peak near 5320 Å, but at 1.06 μm it is transparent. In DFWM, the dye moleculer may absorb one photon of 1.06 μm respectively from both beam A_1 and counter-propagating beam A_2, just like the two-photon absorption in the Dopple-free two-photon spectroscopy. We investigated the two-photon resonantly enhanced DFWM. Using ~70 mj pumping pulse from a Q-switched Nd:YAG laser we measured the dependence of intensities of backward-wave on one of object wave for a 10^{-4}-molar solution of rhodamine 6G in alcohol with an interaction length of 5mm. From the experimental curve shown in Fig. 3 we found the energy reflection coefficients to be 14%. When a material has large third-order nonlinear susceptibility but small fluorescent quantum efficiency, this method has some advantages over fluorescent measurements, and thus may be used in the research of high excited states of the materials[5].

Phase conjugation of forward-wave in four-wave mixing

Heer and Griffen[6] have observed phase-conjugate forward-wave in Na vapor using

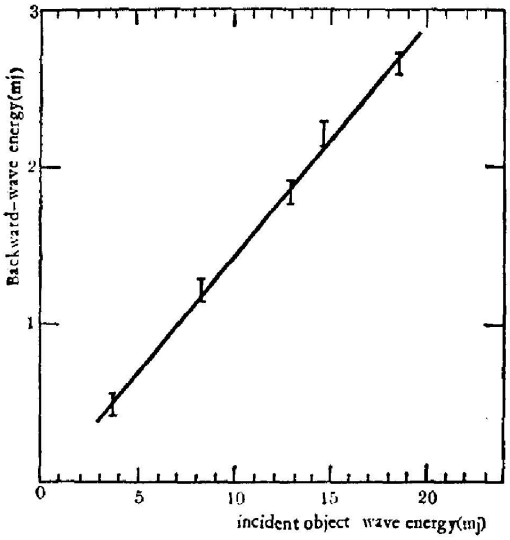


Fig. 3 Results of the experimental measurements of backward-wave energy as a function of input wave energy in two-photon resonant-enhancement four-wave mixing for ~70 mj pumping energy.

a tunable dye laser. We have theoretically predicted the presence of phase-conjugate forward-wave in four-wave mixing and have given the phase-matching conditions. Using SH of a Q-switched Nd:YAG laser as the pumping source the theory has been experimentally proved for rhodamine 6G solution in alcohol.

Let us consider an intense field E_1 and a weak field E_4 simultaneously incident on a nonlinear medium, the fields can be written as

$$\boldsymbol{E}_1(\boldsymbol{r},\ t)=\frac{1}{2}A_1(\boldsymbol{r})\exp(i\omega t-i\boldsymbol{k}\cdot\boldsymbol{r})+C.\,C.$$

$$\boldsymbol{E}_4(\boldsymbol{r},\ t)=\frac{1}{2}A_4(\boldsymbol{r})\exp(i\omega t-i\boldsymbol{k}_4\cdot\boldsymbol{r})+C.\,C.$$

where $\boldsymbol{k}$ and $\boldsymbol{k}_4$ are the wavevectors, not parallel to each other. Suppose E_1 propagates along Z axis and E_4 makes an angle of with it, the wave generated by nonlinear coupling is

$$\boldsymbol{E}_3(\boldsymbol{r},\ t)=\frac{1}{2}A_3(\boldsymbol{r})\exp(i\omega t-i\boldsymbol{k}_3\cdot\boldsymbol{r})+C.\,C.$$

as depicted in Fig. 4. With $\boldsymbol{i}$ and $\boldsymbol{j}$ denoting the unit vectors along X and Z axes respectively, then

$$\boldsymbol{k}=k\boldsymbol{j},\tag{1}$$

$$\boldsymbol{k}_4=k\cos\theta\boldsymbol{j}+k\sin\theta\boldsymbol{i}_{\circ}$$

The total polarization induced in the medium may be expressed as

$$\boldsymbol{P}(t)=\chi(\boldsymbol{E})\cdot\boldsymbol{E},\tag{2}$$

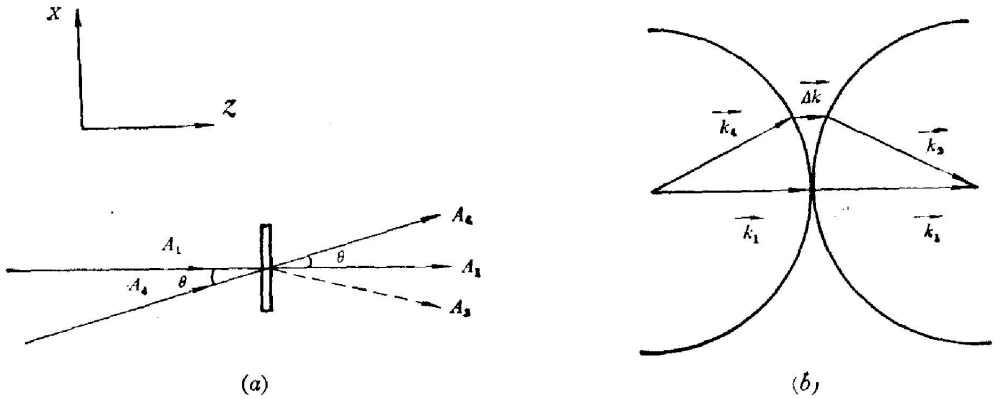
where $\boldsymbol{E}=\boldsymbol{E}_1+\boldsymbol{E}_4+\boldsymbol{E}_3$, $\chi(\boldsymbol{E})$ is the electric susceptibility. $\chi(\boldsymbol{E})$ is expanded about the strong field E_1 to the first order of the weak fields $\boldsymbol{E}_4+\boldsymbol{E}_3$. Then according to the W. K. B. approximation, by taking the time average of $\chi(\boldsymbol{E})$ one obtains[7]

$$\boldsymbol{P}_{NL}(t)=\chi^{(3)}(E_1)\left[\boldsymbol{E}_1^*\cdot(\boldsymbol{E}_4+\boldsymbol{E}_3)+\boldsymbol{E}_1\cdot(\boldsymbol{E}_4+\boldsymbol{E}_3)^*\right](\boldsymbol{E}_1+\boldsymbol{E}_4+\boldsymbol{E}_3), \tag{3}$$

where

$$\chi^{(3)}(\boldsymbol{E}_1)=\frac{2\alpha_0(i+\delta)}{k|E_s|^2}\cdot\frac{1+\delta^2+\dfrac{A_1A_1^*}{2|E_s|^2}}{\left\{(1+\delta^2)\left[(1+\delta^2)+\dfrac{A_1A_1^*}{|E_s|^2}\right]\right\}^{3/2}}, \tag{4}$$

and α_0 is the line-center small-signal-field attenuation coefficient, δ is the normalized detuning from line center, $|E_s|^2$ is the line-center saturation intensity. Expanding eq. (3), the expression of $P_{NL}(t)$ can be obtained



(a) (b)

Fig. 4 Coupling geometry used in phase-conjugate forward-wave (a) and phase-matching geometry (b).

$$\begin{aligned}\boldsymbol{P}_{NL}(t)=&\frac{1}{4}\chi^{(3)}(-3\omega;\ \omega,\ \omega,\ \omega)A_1A_1A_4e^{i(3\omega t-2kz-\boldsymbol{k}_4\cdot\boldsymbol{r})}+C.C.\\&+\frac{1}{4}\chi^{(3)}(-\omega;\ \omega,\ \omega,\ -\omega)A_1A_1A_1^*e^{i(\omega t-2kz+\boldsymbol{k}_4\cdot\boldsymbol{r})}+C.C.\\&+\frac{1}{2}\chi^{(3)}(-\omega;\ \omega,\ -\omega,\ \omega)A_1A_1^*A_4e^{i(\omega t-\boldsymbol{k}_4\cdot\boldsymbol{r})}+C.C.\\&+\frac{1}{4}\chi^{(3)}(-3\omega;\ \omega,\ \omega,\ \omega)A_1A_1A_3e^{i(3\omega t-2kz-\boldsymbol{k}_3\cdot\boldsymbol{r})}+C.C.\\&+\frac{1}{4}\chi^{(3)}(-\omega;\ \omega,\ \omega,\ -\omega)A_1A_1A^*e^{i(\omega t-2kz+\boldsymbol{k}_3\cdot\boldsymbol{r})}+C.C.\\&+\frac{1}{2}\chi^{(3)}(-\omega;\ \omega,\ -\omega,\ \omega)A_1A_1^*A_3e^{i(\omega t-\boldsymbol{k}_3\cdot\boldsymbol{r})}+C.C.+\cdots\cdots\end{aligned} \tag{5}$$

All the third-order nonlinear effects, such as sum frequency and difference frequency generation, first- and second-order, phase conjugate forward-waves etc. are included in this equation. Let us consider the forward-wave which is relevant to phase conjugation of A_4 wave. This involves the second and the fifth terms in Eq. (5). The second term is rewritten as

$$P_2=F_{11}A_4^*e^{i\omega t-2ikz+i\boldsymbol{k}_4\cdot\boldsymbol{r}}=F_{11}A_4^*e^{i\omega t-ik(2z-\cos\theta\cdot z-\sin\theta\cdot x)}.$$

Let

$$\Delta k'=2k(1-\cos\theta). \tag{6}$$

If

$$\Delta k'\ll k\cos\theta, \tag{7}$$

then it follows that

$$P_2 = F_{11}A_4^* e^{i\omega t - ik(\cos\theta z - \sin\theta x)} = F_{11}A_4^* e^{i\omega t - i\boldsymbol{k}_3\cdot\boldsymbol{r}},$$

where

$$\boldsymbol{k}_3 = k(\cos\theta \boldsymbol{j} - \sin\theta \boldsymbol{i})。 \tag{8}$$

It implies that in the forward direction at an angle-θ with Z axis a phase-conjugate wave is generated by the nonlinear coupling.

Similarly, the fifth term in Eq. (5) will generate a field which is the phase conjugation of A_3. The wave propagates along the direction at an angle θ with Z axis. It can be concluded that the two waves which propagate along the directions at θ and $-\theta$ with Z axis are in phase conjugation with each other.

If $\boldsymbol{E}_4$ is an intense pump wave, the nonlinear polarization may be expanded in terms of $(\boldsymbol{E}_1 + \boldsymbol{E}_3)$ about $\boldsymbol{E}_4$, and then the phase-conjugate forward-wave of A_1 can be generated. This wave and $\boldsymbol{E}_3$ wave should arrange symmetrically about $\boldsymbol{E}_4$. In order to generate the phase-conjugate forward-waves the phase-matching condition must be satisfied

$$|\Delta k \cdot L| \leqslant \pi,$$

where L is the effective length of the medium and

$$\Delta\boldsymbol{k} = 2\boldsymbol{k} - \boldsymbol{k}_4 - \boldsymbol{k}_3。$$

Substituting Eqs. (1) and (8) in the last equation yields

$$\Delta\boldsymbol{k} = \Delta\boldsymbol{k}' = 4k\sin^2(\theta/2)\boldsymbol{j}。$$

For $\theta \ll 1$, the phase-matching condition requires

$$\theta \leqslant (\lambda/2L)^{1/2}。 \tag{9}$$

From Eq. (7) we get

$$\theta \ll \cos^{-1} 2/3。 \tag{10}$$

For light-frequency radiations and physically thin nonlinear media Eq. (10) is always satisfied when Eq. (9) is satisfied.

Substituting the second and the fifth terms of Eq. (5) in Maxwell equations and taking the adiabatic approximation and the nondepleted pump approximation, the following coupled equations are abtained

$$\frac{dA_1^*}{dz} = \varkappa^* A_3(z) e^{i\Delta\boldsymbol{k}\cdot\boldsymbol{r}},$$

$$\frac{dA_3}{dz} = \varkappa A_4^*(z) e^{-i\Delta\boldsymbol{k}\cdot\boldsymbol{r}},$$

where

$$\varkappa^* = i\pi\frac{\omega}{cn}\chi^{(3)*}A_1^* A_1^*。$$

The energy conversion efficiency may be obtained

$$S = \left|\frac{A_3(L)}{A_4(0)}\right|^2 = \frac{|\varkappa|^2}{|\varkappa|^2 - \left(\frac{4k}{2}\right)^2}\sinh^2\left(\sqrt{|\varkappa|^2 - \left(\frac{\Delta k}{2}\right)^2}\, L\right)。$$

The experimental apparatus used to demonstrate the forward-wave generation is shown in

Fig. 5. Using ~1mj radiation from SH of a Q-switched Nd:YAG laser, for rhodamine 6G solution in alcohol with an effective length of 1mm the phase-conjugate forward-waves were obtained. The angle θ between beams A_1 and A_4 is about 1.7×10^{-2} rad. When the length L of the medium becomes about 2.5 mm, the forward-waves disappear, because in this case $\Delta k\cdot L>\pi$, i.e. the phase-matching condition is not satisfied. Taking L as constant and changing the angle θ, similar phenomena can be observed.

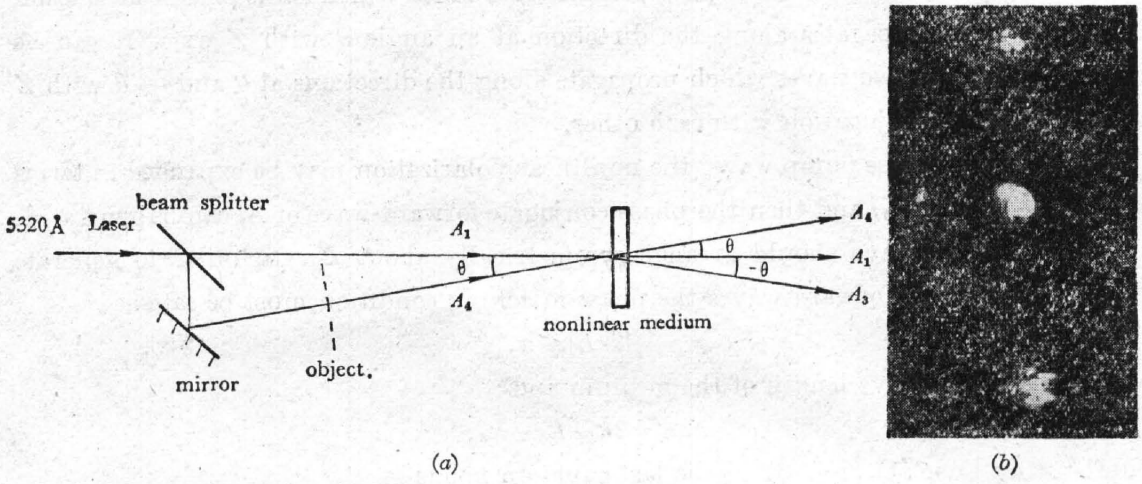
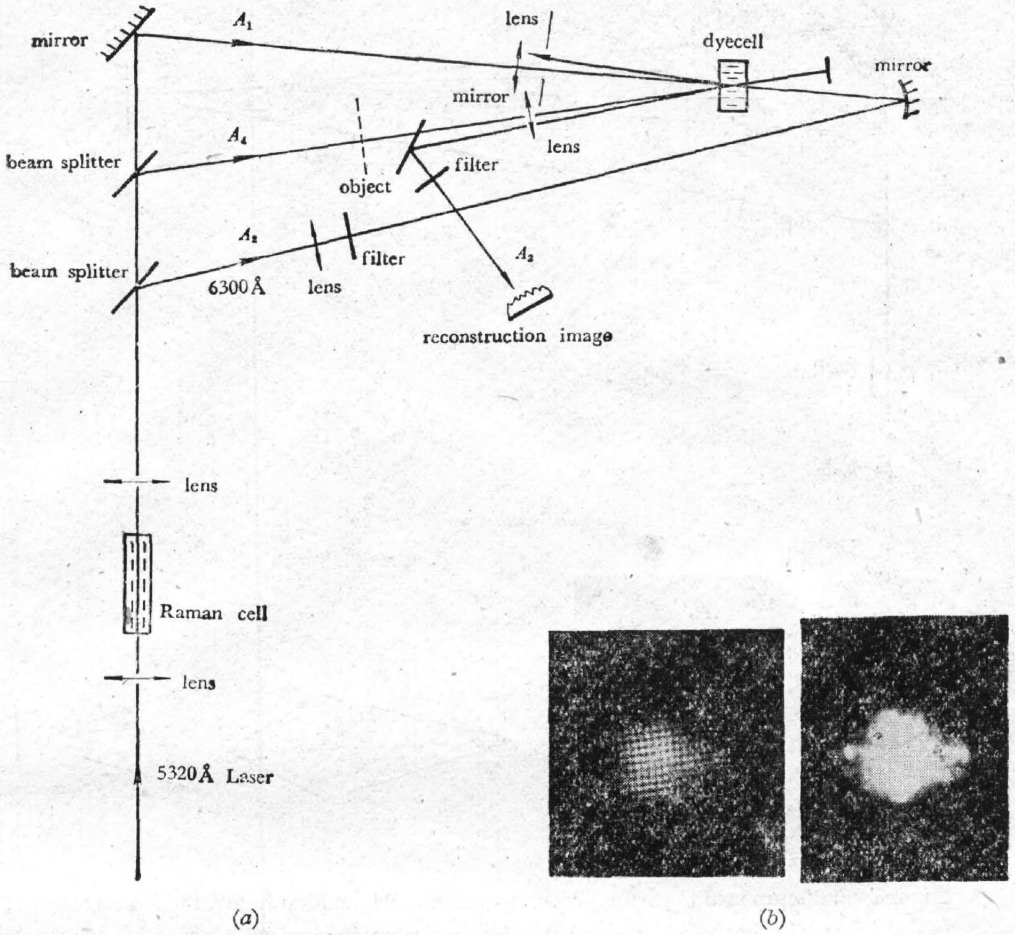


Fig. 5 Experimental setup used to generate phase-conjugate forward-wavd (a) and phase-conjugation fields (b).

Doublet DFWM and image conversion

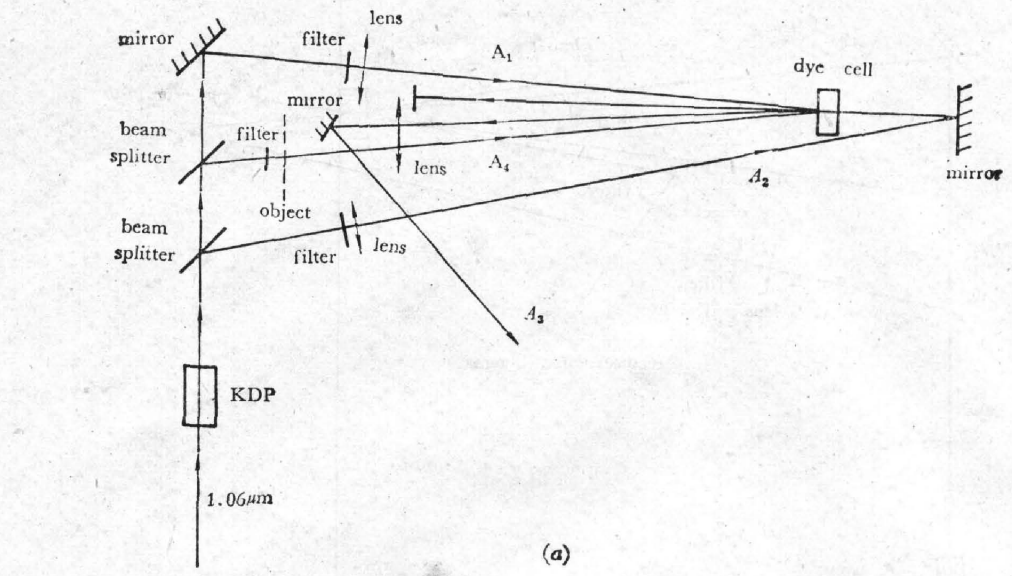
It is well known that there is a formal analogy between the four-wave mixing and the conventional holography[8]. The hologram is formed when the incident object wave and pump wave interfere to produce a spatial phase grating in the nonlinear medium. Therefore, we can reconstruct the image recorded by object wave A_4 using wave A_2 at another frequency. Using the apparatus shown in Fig. 6 we have experimentally demonstrated this conversion. A lens ($f=300$ mm) focuses the 5320 Å laser radiation into a Raman cell with an interaction length of 200 mm. The Raman medium is $(CH_3)_2SO$. The wavelength of the first order Stokes scattering radiation is about 6300 Å. The conversion efficiency of the stimulated Raman scattering is controlled to be about 30%. Using the 5×10^{-4}-molar solution of rhodamine 6G in alcohol with an effective length of 5 mm, the conversion efficiency from the object wave (5320 Å) to the reconstructed wave (6300 Å) is about 12%, with the conditions that the energy of the pump wave A_1 from SH of a Q-switched laser pulse is about 1.2 mj, beam A_4 is ~1 mj and beam A_2 is ~1.2 mj. In order to satisfy the phase-matching condition beam A_2 travels at an angle of 10^{-2} rad approximatly to the direction of beam A_1. The reconstructed image at 6300Å is shown in Fig. 6. In this experiment, the medium used has an absorption band at object wave



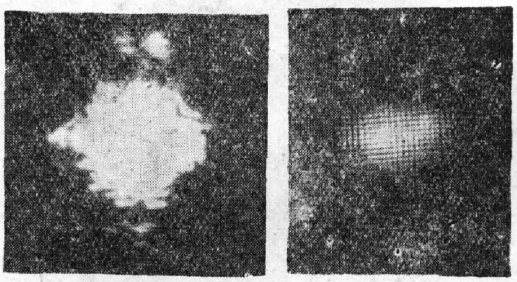
g. 6 Experimental apparatus used in doublet DFWM which performed conversion from 5320 Å to 6300 Å (*a*) and the reconstructed wave fields (*b*).

frequency but is transparent at the reconstruction frequency, hence we are able to enhance resonantly nonlinearity without loss of the reconstruction beam. By this method if we select a medium which is strong absorptive with large oscillator strength for infrared object wave but transparent for visible reconstruction wave, resonantly enhanced image conversion from infrared to visible frequency may be achieved. The experiment was performed by the setup shown in Fig. 7. The nonlinear medium used is organic dye 9740 solution in dichloroethane with an interaction length of 5 mm. The dye molecule has strong absorption at 1.06 μm but transparent at 5320 Å. We adjusted the concentration of the dye solution so that the pump beam A_1 was absorbed totally by the medium. For the energy of beam A_1 is $\sim$5 mj, the concentration of the dye solution is about 10^{-4}mole. The angle φ between the two infrared beams A_1 and A_4 is about 6×10^{-2} rad. For the energies of beams A_1 and A_4 are 4.2 mj, 1.5 mj respectively, the energy conversion efficiency from beam A_4 to A_3 is about 25%.

For the doublet DFWM, the nonlinear polarization may be expressed as



(a)



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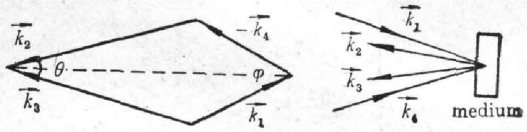

(c)

Fig. 7 Experimental setup used to perform conversion from 1.06 μm 5320 Å (*a*), the reconstructed visible image field (*b*) and the phase-matching diagram (*c*).

$$P_{NL}(\omega_{vis}) = \frac{3}{4}\chi_1^{(3)} A_1 A_2 A_4^* \exp\left[i(\omega_{vis}t - \boldsymbol{k}_3 \cdot \boldsymbol{r})\right],$$

$$P_{NL}(\omega_{If}) = \frac{3}{4}\chi_2^{(3)} A_1 A_2 A_3^* \exp\left[i(\omega_{If}t - \boldsymbol{k}_4 \cdot \boldsymbol{r})\right],$$

where

$$\chi_1^{(3)} = \chi^{(3)}(-\omega_{vis};\ \omega_{If},\ \omega_{If},\ -\omega_{vis}),$$

$$\chi_2^{(3)} = \chi^{(3)}(-\omega_{If};\ \omega_{If},\ \omega_{vis},\ -\omega_{vis}),$$

and

$$\boldsymbol{k}_3 = \boldsymbol{k}_1 + \boldsymbol{k}_2 - \boldsymbol{k}_4,$$

$$\boldsymbol{k}_4 = \boldsymbol{k}_1 + \boldsymbol{k}_2 - \boldsymbol{k}_3,$$

For a medium which is absorptive for the infrared but transparent for the visible, it follows that

$$\chi_1^{(3)} > \chi_2^{(3)}.$$

After applying the standard methods of nonlinear optics we obtain the coupled wave equations in the small angle approximation

$$\frac{dA_3}{dz} = -i\varkappa_1 A_4^*,$$

$$\frac{dA_4^*}{dz} = i\varkappa_2^* A_3,$$

where the coupling constants $\varkappa_i$ are given by

$$\varkappa_1=\frac{2\pi\omega_{vis}}{cn_{vis}}\chi_1^{(3)}A_1A_2,$$

and

$$\varkappa_2=\frac{2\pi\omega_{IR}}{cn_{IR}}\chi_2^{(3)}A_1A_2.$$

If we specify the complex amplitudes $A_3(L)$ and $A_4(0)$ at their respective input planes ($Z=L$, $Z=0$), the following solution is obtained

$$A_4^*(z)=-\left|\frac{\varkappa_2^*}{\varkappa_1}\right|^{1/2}\cdot\frac{\sin(\sqrt{|\varkappa_1\varkappa_2^*|}\,z)}{\cos(\sqrt{|\varkappa_1\varkappa_2^*|}\,L)}A_3(L)-\frac{\cos[\sqrt{|\varkappa_1\varkappa_2^*|}\,(z-L)]}{\cos[\sqrt{|\varkappa_1\varkappa_2^*|}\,L)}A_4^*(0),$$

$$A_3(z)=\frac{\cos\sqrt{|\varkappa_1\varkappa_2^*|}\,z}{\cos\sqrt{|\varkappa_1\varkappa_2^*|}\,L}A_3(L)+i\left|\frac{\varkappa_1}{\varkappa_2^*}\right|^{1/2}\cdot\frac{\sin[\sqrt{|\varkappa_1\varkappa_2^*|}\,(z-L)]}{\cos(\sqrt{|\varkappa_1\varkappa_2^*|}\,L)}A_4^*(0).$$

From this solution we obtain

$$A_3(0)=\frac{1}{\cos(\sqrt{|\varkappa_1\varkappa_2^*|}\,L)}A_3(L)+i\left|\frac{\varkappa_1}{\varkappa_2^*}\right|^{1/2}\tan(\sqrt{|\varkappa_1\varkappa_2^*|}\,L)A_4^*(0),$$

$$A_4^*(L)=-i\left|\frac{\varkappa_2^*}{\varkappa_1}\right|^{1/2}\tan(\sqrt{|\varkappa_1\varkappa_2^*|}\,L)A_3(L)-\frac{1}{\cos(\sqrt{|\varkappa_1\varkappa_2^*|}\,L)}A_4^*(0).$$

In our case, the object wave input at $Z=0$ is $A_4(0)$ and $A_3(L)=0$, thus the nonlinear conversion efficiency is

$$R=\left|\frac{A_3(0)}{A_4(0)}\right|^2=\left|\frac{\varkappa_1}{\varkappa_2^*}\right|\tan^2(\sqrt{|\varkappa_1\varkappa_2^*|}\,L),$$

where $\left|\frac{\varkappa_1}{\varkappa_2^*}\right|>1$. When

$$\sqrt{|\varkappa_1\varkappa_2^*|}\cdot L=\pi/2,$$

we have

$$\left|\frac{A_3(0)}{A_4(0)}\right|=\infty,$$

which corresponds to parametric oscillation. This implies that by using this scheme the conversion efficiency from infrared to visible image may be much larger than unity.

This image conversion has a number of distinct advantages: 1. this is a four-wave parametric process, the conversion is achieved by the nonlinear coupling of the fields in nonlinear medium. In principle,the scheme may be used at any wavelengths. 2. if the medium has an absorption band with large oscillator strength but is transparent at visible reconstruction wave, the third-order nonlinear susceptibility may be resonantly enhanced. Since the nonlinear conversion efficiency is proportional to $|\chi^{(3)}|^2$, the efficiency may thus be greatly increased. If strong pumping and reconstruction waves are used, the efficiency may be much larger than unity. 3. since the backward-wave generated in the four-wave mixing is relevant to the phase conjugation of the object wave, a phase distortion produced in the propagation of the image may be compensated. 4. it can be performed in real-time. Finally, this scheme has fast response time. For many organic dye molecules, the typical lifetimes of metastable singlets are shorter than or nearly equal to 1 nsec.

Obviously, an infrared image conversion system with a response time in the order of 1 nsec is rather attractive.

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有机染料溶液中简并的四波混频和位相复共轭研究*

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提 要

研究了近共振增强的四能级系统有机染料溶液中简并的四波混频后向反射波及前向波的位相复共轭特性。用有效长度为5mm，浓度为2.5×10^{-5}mol.的叶录素A甲醇溶液作介质，当调Q Nd:YAG倍频泵浦光的能量为8mj时，非线性反射率为22%。对于若丹明6G酒精溶液，在相同条件下的反射率为10%。用象差板检证了位相复共轭特性。用调Q Nd:YAG激光辐射作泵浦源，研究了若丹明6G酒精溶液双光子共振增强的简并的四波混频作用。当介质浓度为10^{-4}克分子，有效长度为5mm，泵浦光能量为70mj时，非线性反射率达14%。测量了非线性反射率与介质浓度及入射物波与后向反射波的强度的关系曲线。理论分析了四波混频前向波的位相复共轭特性，用调Q Nd:YAG倍频激光作泵浦源，若丹明6G酒精溶液作介质，实验上证实了理论的预言。另外，把四重简并的四波混频推广到二重简并的情况。用若丹明6G酒精溶液作介质，实现了由6300Å到5320Å的象转换。用9740有机染料二氯乙烷饱和吸收溶液作介质，实现了1.06μm到5320Å的象转换。当介质长度为5mm，泵浦光束的能量为5mj时，后者的转换效率为25%。理论分析表明，用对红外近共振吸收，而对可见辐射是透明的介质可以实现高效率的红外象转换。这种红外象转换的方法还具有实时、快响应等优点。

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