Vector phase conjugation by two-photon-resonant degenerate four-wave mixing

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We have studied the polarization properties of phase conjugation by degenerate four-wave mixing resonantly enhanced by the sodium $3S_{1/2} \rightarrow 6S_{1/2}$ two-photon-allowed transition. As predicted by third-order theories, this interaction leads to simultaneous conjugation of the optical wave front and state of polarization (vector phase conjugation) when the pump intensities are sufficiently weak. However, qualitatively different polarization behavior is observed for pump intensities near or above the two-photon saturation intensity.

Optical phase conjugation is a process that can lead to the correction of wave-front distortions under conditions in which a beam of light passes twice in opposite directions through an aberrating optical medium. Analogously, the state of polarization of an optical beam can become distorted, and these polarization distortions can be removed by the process of polarization conjugation. In this process, the polarization unit vector of the generated wave is equal to the complex conjugate of that of the incident wave. Thus, for example, the handedness of a beam of light is preserved in reflection from a polarization conjugating mirror rather than being inverted, as in the case of reflection from an ideal conventional mirror.

In this Letter, we report our experimental investigations of a new method for producing vector phase conjugation (VPC), that is, the simultaneous conjugation of the optical wave front and state of polarization. Our method involves degenerate four-wave mixing (DFWM) utilizing the special tensor properties of two-photon atomic transitions. It has recently been predicted that, for appropriate choices of the atomic energy levels, this interaction leads to VPC for arbitrary polarizations of the pump waves.[1,2] Phase conjugation by DFWM enhanced by the biexcitonic resonance in copper chloride has been found to lead to good but not perfect VPC.[3] VPC has also been observed in several previous DFWM experiments; however, in these experiments a particular choice of pump-beam polarizations and propagation vector directions was required to obtain VPC.[4] Furthermore, it has been shown theoretically that stimulated Rayleigh-wing scattering can produce VPC under certain conditions.[5] VPC can also be obtained in some cases by conjugating each orthogonal polarization component by using scalar phase-conjugate mirrors[6] or by using a polarization scrambling fiber followed by a scalar phase-conjugate mirror.[7]

Two-photon-resonant DFWM has been analyzed by Grynberg[1] and by Ducloy and Block[2] in the third-order perturbation theory limit for arbitrary atomic angular-momentum states and for arbitrary polarization states of the input fields. These authors show that VPC is achieved, for pump beams of any state of polarization, if the two levels coupled by the two-photon transition have equal angular momenta J, with $J = 0$ or $\frac{1}{2}$. This result can be understood by the following intuitive argument: Since the atom cannot change its angular momentum either by absorbing two pump photons or by emitting a probe and a conjugate photon (because in each case $\Delta J = 0$), the conjugate photon must be emitted with angular momentum equal and opposite to that of the probe photon, which implies that the two beams will be polarization conjugates. The polarization properties of two-photon-resonant DFWM are so different from those of most other DFWM processes because the mechanism responsible for two-photon-resonant DFWM, in the absence of saturation effects [i.e., in the $\chi^{(3)}$ approximation], is the scattering of the probe field from a spatially uniform temporally varying coherence induced by the two pump waves.[8] Conversely, the more usual mechanism for DFWM is scattering from a spatially varying refractive-index distribution induced by the interference between the pump and the probe waves.[9]

It has been shown by Maker and Terhune[10] that the nonlinear polarization of any isotropic medium can be represented in third order as

$$\mathbf{P}^{NL} = A(\mathbf{E}\cdot\mathbf{E}^*)\mathbf{E} + \tfrac{1}{2}B(\mathbf{E}\cdot\mathbf{E})\mathbf{E}^*, \qquad (1)$$

where $\mathbf{E}$ is the complex amplitude of the total electric field. One can readily show[11] that, in the standard four-wave mixing geometry for phase conjugation, the term proportional to B leads directly to VPC for any polarization state of the pump beams, whereas the term proportional to A will lead to VPC only for special choices of pump-beam polarization, as discussed above.[4] To determine the values of A and B relevant to our experimental conditions, we have solved to third order in perturbation theory the density-matrix equations of motion appropriate to the multilevel atomic system shown in Fig. 1(a) in the presence of the three input fields $\mathbf{E}_j = A_j\hat{\epsilon}_j \exp[i(\mathbf{k}_j\cdot\mathbf{r} - \omega t)]$ for $j = 1, 2, 3$. We have found that for a $nS_{1/2} \rightarrow n''S_{1/2}$ transition, A is identically equal to zero and

$$B = \frac{N}{\hbar^3\Delta_1{}^2[\Delta_2 - i(1/T_2)]}\left(\langle n''S|\mu_z|n'P\rangle\langle n'P|\mu_z|nS\rangle\right)^2, \qquad (2)$$

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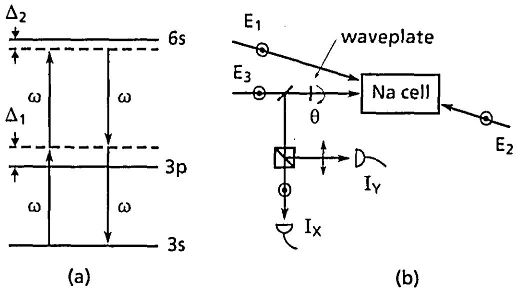

Fig. 1. (a) Energy-level diagram showing the two-photon resonantly enhanced DFWM process. (b) Experimental setup.

where $\Delta_1 = \omega_{n'P \rightarrow nS} - \omega$ is the one-photon detuning (which is large), $\Delta_2 = \omega_{n''S \rightarrow nS} - 2\omega$ is the two-photon detuning, T_2 is the dipole dephasing time of the two-photon coherence, and $\langle \alpha' | \mu_z | \alpha \rangle$ is the z component of the dipole matrix element connecting the states α and α'. Since A vanishes identically, the VPC process is expected to be perfect whenever the predictions of the third-order theory are valid, that is, whenever the input intensities are much smaller than the two-photon saturation intensity. For the $3S_{1/2} \rightarrow 6S_{1/2}$ transition in atomic sodium, the two-photon saturation intensity, $I_{\mathrm{sat}} = (cn\hbar^2\Delta_1)/[4\pi(T_1T_2)^{1/2}\langle n''S|\mu_z|n'P\rangle\langle n'P|\mu_z|nS\rangle]$, is equal to 1.2 MW cm^{-2}, where T_1 is the lifetime of the $6S$ level.

In our experiment to investigate the polarization properties of the two-photon-resonant DFWM process, the output from an excimer-laser-pumped dye laser was set to ~549 nm to excite the sodium $3S_{1/2} \rightarrow 6S_{1/2}$ two-photon transition. A sodium heat-pipe oven with a 20-cm interaction length was operated at an atomic number density of $\sim 10^{17}$ atoms cm^{-3} with a helium buffer gas at a pressure of ~2 Torr. The dye laser was operated with pulse energies up to 10 mJ, a pulse duration of ~15 nsec, and a bandwidth of ~0.04 cm^{-1}. Two beams of equal intensity and independently controllable states of polarization formed the counterpropagating pump beams. The probe beam, with an intensity equal to 2% of that of one of the pump beams, was directed into the sodium heat pipe at an angle of 0.5° with respect to the pump beams to ensure that the beams overlapped over the entire sodium region.

The polarization of the probe beam is controlled by passing an initially linearly polarized beam through a quarter-wave plate, as shown in Fig. 1(b). This procedure can be thought of as impressing a known polarization distortion upon the initially linearly polarized beam. The experiment then entails determining the extent to which the VPC process is capable of removing the effects of the polarization distortion from the conjugate beam after its return pass through the quarter-wave plate. The state of polarization is analyzed by a cube polarizer oriented so that its transmission axis is parallel to the direction of linear polarization of the initial (i.e. undistorted) probe beam. The fidelity of the VPC process is then determined by measuring the intensity of the beam transmitted through (denoted I_x) and rejected by (denoted I_y) the polarizer. If the VPC process is perfect, the conjugate beam will be returned to its original polarization state after passing through the distorter and I_y will be equal to zero.

Figure 2 shows I_x and I_y plotted as functions of the laser detuning from the two-photon resonance for the case of linear, parallel, pump-beam polarizations and a probe beam that is circularly polarized at the entrance of the sodium cell. Figure 2(a) shows that for a pump-beam intensity of $0.16I_{\mathrm{sat}}$, for which the predictions of the third-order theory should be applicable, the polarization distortion introduced by the waveplate is removed by the DFWM process (i.e., $I_y \ll I_x$). This result demonstrates that DFWM resonantly enhanced by a two-photon transition produces high-fidelity VPC, in agreement with the predictions of the third-order theory outlined above. However, the maximum phase-conjugate reflectivity for this pump-beam intensity is only $\sim 10^{-5}$. We have also studied the fidelity of the VPC process for higher pump intensities, for which the phase-conjugate reflectivity is expected to be larger. Figure 2(b) shows the results for a saturating pump beam of intensity equal to $1.7I_{\mathrm{sat}}$ and where the reflectivity is $\sim 10^{-3}$. In this case the fidelity of the VPC process is badly degraded in that I_y is comparable to I_x. Furthermore, the DFWM resonance is broadened considerably, showing that saturation effects are indeed present in this case.[12]

To quantify further the polarization properties of the two-photon-resonant DFWM process, we have measured the values of I_x and I_y as functions of the probe-beam polarization distortion. By rotating the waveplate, the polarization of the distorted probe beam is varied from circular to linear. Figure 3(a) shows that, for a weak pump-beam intensity equal to $0.16I_{\mathrm{sat}}$, the generated beam is a high-fidelity vector phase conjugate, since $I_y \ll I_x$ in all cases and I_x is independent of the polarization distortion. Figure 3(b) shows the results for the case of saturating pump beams with intensities equal to $1.7I_{\mathrm{sat}}$. The phase-conjugate reflectivity is seen to depend on the state of

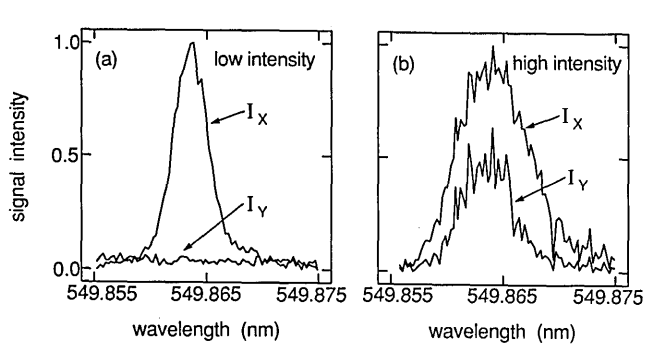

Fig. 2. Intensities of the polarization components parallel (I_x) and perpendicular (I_y) to the initial probe-beam polarization direction as functions of the excitation wavelength. (a) For low pump intensities (200 kW cm^{-2}, where I_{sat} = 1.2 MW cm^{-2}), the VPC process is of high quality in that $I_y \ll I_x$. (b) At high pump intensities ($1.7I_{\mathrm{sat}}$), the VPC process is severely degraded in that I_y is comparable with I_x. In each case, the waveplate in Fig. 1(b) was oriented to render the probe beam circularly polarized at the entrance to the sodium cell.

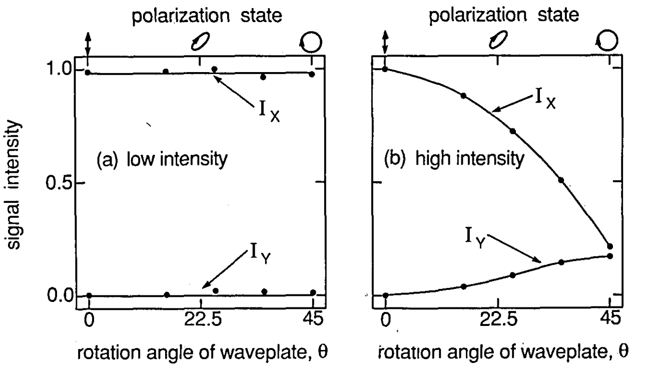

Fig. 3. Intensity of each polarization component plotted as a function of the degree of polarization distortion introduced into the probe wave. (a) For low pump intensities ($0.16 I_{sat}$), the effects of the polarization distortion are removed essentially completely (I_x is independent of θ and $I_y \ll I_x$. (b) For high pump intensities ($1.7 I_{sat}$), severe degradation of the VPC is observed (I_x and I_y are comparable and depend on θ).

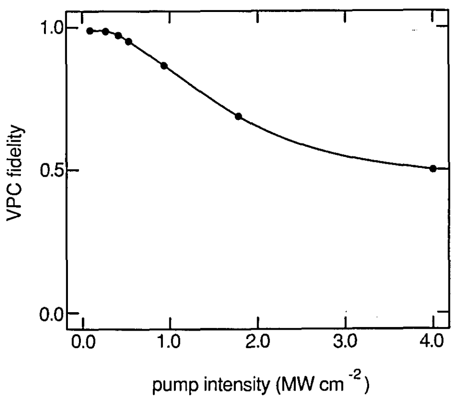

Fig. 4. Fidelity of the VPC process, quantified by the ratio $I_x/(I_x + I_y)$ (i.e., the ratio of the intensity of the proper polarization component to the total output intensity), plotted as a function of the intensity of one of the pump waves. The waveplate in Fig. 1(b) has been oriented to produce circular polarization.

polarization of the input probe beam, demonstrating that saturation of the two-photon resonance significantly degrades the fidelity of the VPC process.

We have also determined how quickly the fidelity of the VPC process is degraded as the pump-beam intensities are increased. We take the quantity $I_x/(I_x + I_y)$ to be a measure of the fidelity of the VPC process. Figure 4 shows the fidelity as a function of the intensity of either pump beam for the case of a probe beam that is circularly polarized at the entrance of the sodium cell. For pump intensities well below the saturation intensity, this ratio is equal to 1, indicating that the VPC is perfect. At intensities well above I_{sat}, the polarization of the phase-conjugate beam at the exit of the sodium cell is linearly polarized parallel to the pump-beam polarization. After propagating back through the waveplate, the polarization of this beam is converted to circular, thus leading to the measured ratio of 0.5.

We have also verified that, for input intensities well below the two-photon saturation intensity, high-quality VPC occurs for pump beams of any state of polarization. However, the overall phase-conjugate reflectivity is found to depend on the scalar product of the pump-beam polarizations, as expected from Eq. (1). For intensities much greater than the saturation intensity, we find that the polarization of the phase-conjugate beam is the same as that of the pump beam, which is nearly copropagating with the conjugate beam. We attribute this result to the scattering of the pump beam from a refractive-index grating induced by the transfer of population to the $6S_{1/2}$ excited state. Such transfer of population is predicted by nonlinearities of higher order than $\chi^{(3)}$.[13]

In conclusion, we have shown that DFWM resonantly enhanced by a two-photon transition produces high-quality VPC when the pump-beam intensity is well below the two-photon saturation intensity, as predicted by third-order theories. At pump-beam intensities comparable with or above the saturation intensity, the quality of the VPC process is severely degraded.

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