Cross-polarized wave generation by effective cubic nonlinear optical interaction

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A new cubic nonlinear optical effect in which a linearly polarized wave propagating in a single quadratic medium is converted into a wave that is cross polarized to the input wave is observed in BBO crystal. The effect is explained by cascading of two different second-order processes: second-harmonic generation and difference frequency mixing. © 2001 Optical Society of America

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Third-harmonic generation was reported by Terhune *et al.*[1] in 1962 as the first cubic nonlinear optical effect. Since then several other cubic effects have been discovered, investigated as tools for the study of the properties of matter, and employed for new frequency generation, optical processing, and phase correction. A list of nonresonant cubic nonlinear optical effects can be found in many textbooks (see, e.g., Ref. 2). Third-order effects can be obtained not only on the basis of cubic susceptibility of a nonlinear material (by direct third-order process) but also as a result of cascaded second-order processes.[3] It is now recognized that, owing to second-order cascading, many cubic effects, such as nonlinear phase shift, pulse compression, and soliton propagation, can be observed at lower pump levels in quadratic media than in centrosymmetric media. Nevertheless, in all cases the cubic effects observed in quadratic media are the same as those observed in centrosymmetric media.

We report here what is believed to be the first observation of a new cubic nonlinear optical effect as predicted in Ref. 4, in which three degenerate linearly polarized fields (carried by the same beam) generate through an effective cubic nonlinearity a new wave at the same frequency but polarized in the plane perpendicular to the input one. The nonlinear interaction that we investigate is a direct way of generating a cross-polarized wave (XPW), as opposed to methods of polarization rotation that necessitate splitting the input beam in two beams, manipulation of the phase of one of the beams, and interferometric recombination of the split beams.[5–10]

We demonstrate that the phase-matching dependent nonlinear optical conversion from an ordinary to an extraordinary wave that we observed is a result of two-step cascading of two different second-order processes (TDSOP). This type of cascading is a fast-developing area in nonlinear optics. Large nonlinear phase shift,[11,12] multicomponent solitons,[13–15] and third-harmonic,[16–20] and fourth-harmonic[21] generation are some of the applications that result from the use of TDSOP. Cascading of TDSOP has a different meaning than conventional second-order cascading,[3] in which the effect results from the simultaneous action of two subprocesses that belong to a single second-order interaction, e.g., type I second-harmonic generation (SHG).

The main idea of the experiment with generation of XPW is shown in the inset of Fig. 1. The process used to generate, in a quadratic medium, a wave with a polarization vector perpendicular to the polarization vector of the input wave consists of two steps: (i) SHG with two identical fundamental waves and (ii) difference frequency mixing, in which the second harmonic (SH) is downconverted back to the XPW at the fundamental frequency. When the input wave is ordinary (o) there are two possible combinations of

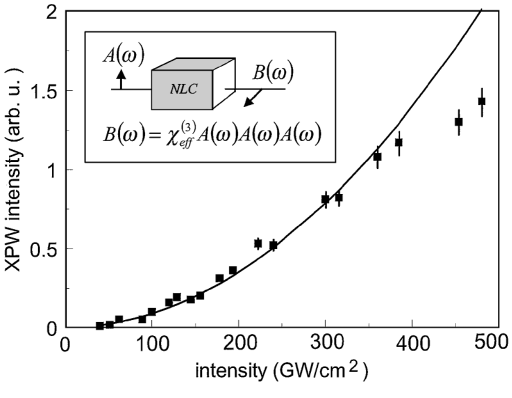

Fig. 1. Dependence of the XPW signal ($I_{e,\mathrm{out}} - I_{\mathrm{bg}}$) on input pump intensity. Solid curve, quadratic fit to the experimental points that were recorded for $I_{o,\mathrm{in}} < 350\ \mathrm{GW/cm^2}$. Inset, diagram of generation of a XPW. NLC, nonlinear $\chi^{(2)}$ crystal.

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second-order processes that will generate an extraordinary (e) wave: $\{o_1o_1 \rightarrow e_2, e_2o_1 \rightarrow e_1\}$ and $\{o_1o_1 \rightarrow o_2, o_2o_1 \rightarrow e_1\}$. The equivalent cubic interaction is $o_1o_1o_1 \rightarrow e_1$. When the input wave is extraordinary the two possible combinations that will generate an ordinary wave are $\{e_1e_1 \rightarrow o_2, o_2e_1 \rightarrow o_1\}$ and $\{e_1e_1 \rightarrow e_2, e_2e_1 \rightarrow o_1\}$. The equivalent cubic interaction is $e_1e_1e_1 \rightarrow o_1$. The general rule is to find the crystal and its associated orientation that will support the two steps simultaneously. For each combination of TDSOP, four possibilities exist: (a) step (i) is phase matched (PM), step (ii) is non-PM; (b) step (ii) is PM, step (i) is non-PM; (c) only the equivalent cubic interaction is PM; (d) steps (i) and (ii) are simultaneously PM.

For practical realization of the experiment we used a BBO crystal cut for PM type II SHG ($e_1o_1 \rightarrow e_2$) at the fundamental beam, with $\lambda_1 = 620$ nm, i.e., $\theta_{\mathrm{PM}} = 58°33$. The azimuthal angle, φ, was chosen to be optimal for the simultaneous action of steps (i) and (ii), i.e., $\varphi = 15°$ for BBO crystal. This nonlinear crystal supports cascading with step (i) type I SHG ($o_1o_1 \rightarrow e_2$) and step (ii) difference-frequency mixing process ($e_2o_1 \rightarrow e_1$). This cascading corresponds to the $o_1o_1o_1 \rightarrow e_1$ cubic process.

An input beam at $\lambda_1 = 620$ nm was produced by a colliding-pulse mode-locked dye system. The laser pulses that were used had the following parameters: duration, ~100 fs; maximum energy, 5 mJ; and a repetition rate of 10 Hz. The laser beam was focused with a lens ($f = 1.5$ m) that produced a spot with radius $r = 0.2$ mm in the plane of the crystal. The BBO crystal was situated between a crossed polarizer and an analyzer. To avoid depolarization of the ordinary wave in the crystal we tuned the input polarization perpendicularly to the plane formed by the fundamental wave vector and the optical axis of the BBO crystal. In this way, in the linear regime, only one of the two allowed eigenwaves propagated in the medium: The extinction ratio of the polarizer–BBO crystal–analyzer system, measured at relatively low input power, was reduced to $R_X{}^{(0)} = I_{\mathrm{bg}}/I_{o,\mathrm{in}} = 6 \times 10^{-6}$. We checked that the observed SH signal at the output of the crystal resulted from the non-PM process, $o_1o_1 \rightarrow e_2$. When the angle θ was tuned to θ_{PM}, no additional SH signal (which would result from the PM interaction $o_1e_1 \rightarrow e_2$) could be detected. This was an additional check that only one ordinary polarized wave entered the crystal. From these initial conditions, an increase of the input power led to a worsening of the extinction ratio at the fundamental wavelength, $R_X(I_{o,\mathrm{in}}) = R_X{}^{(0)} + R_{\mathrm{NL}}(I_{o,\mathrm{in}})$, an indication that a new signal polarized perpendicularly to the input wave was generated by a nonlinear optical process in the crystal. In Fig. 1 the increase of the extraordinary component of the signal ($I_{e,\mathrm{out}} - I_{\mathrm{bg}}$) at the output of the polarizer–BBO crystal–analyzer system is shown as a function of the input intensity for the case in which the input wave is propagating in the BBO crystal as an ordinary wave.

As we expected, the magnitude of the XPW was sensitive to the deviation from the exact PM angle, $\Delta\theta_{\mathrm{PM}} = \theta - \theta_{\mathrm{PM}}$. In this set of experiments we measured the XPW at the fundamental frequency and the SH signal generated in the crystal simultaneously while the crystal was tuned in a range of θ from 56° to 60°. The results of these recordings (again for a pure ordinary input wave) are shown in Fig. 2. The maximum of the XPW coincides with angle θ_{PM}, at which the type II SHG process is expected to be maximal (type II SH signal was measured separately when the two eigenpolarizations were present in the BBO crystal). At the same time, it can clearly be seen that the SH signal goes through a minimum at this angle; this result is related to the depletion of the SH wave owing to the generation of the XPW. The general decrease of the SH signal with increasing angle θ is related to the decrease of the coherence length for non-PM type I SHG in the first step of this cascading interaction. The maximum efficiency of the generated XPW, achievable for input intensities of ~500 GW/cm^2, was measured to be $R_{\mathrm{NL}} = 1.8 \times 10^{-5}$.

The effects of the input power and the length of the BBO crystal on the width of the PM curve, $\Delta\theta_{\mathrm{PM}}$, are illustrated in Fig. 3. A decrease of the BBO crystal length at fixed input power leads to an increase of $\Delta\theta_{\mathrm{PM}}$; at the same time, a decrease of the input power results in a decrease of $\Delta\theta_{\mathrm{PM}}$.

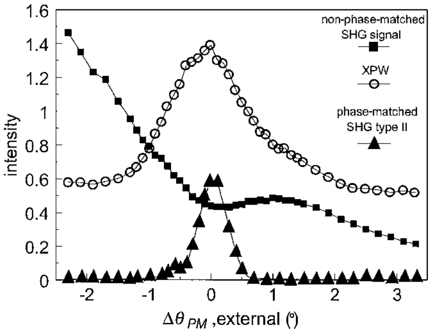

Fig. 2. Experimentally measured XPW and non-PM SH signals as a function of the deviation $\Delta\theta$ from the PM angle for type II SHG. The input power for these two curves is $I_{o,\mathrm{in}} \approx 300$ GW/cm^2. The bottom curve, taken with $I_{o,\mathrm{in}} \approx 30$ GW/cm^2, represents the PM type II SHG signal measured in a separate experiment when both ordinary and extraordinary waves entered the BBO crystal (the purpose of this recording is to demonstrate the position of the phase-matching angle for the second step).

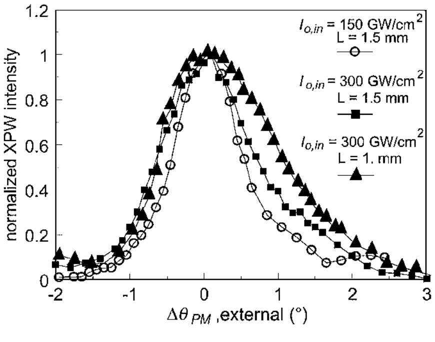

Fig. 3. Normalized XPW signal for three different sets of input and crystal length.

The XPW generation effect in the plane-wave approximation and in lossless quadratic media is described by the equations discussed in Refs. 4 and 12. We solved these equations in the approximation of nondepletion of the fundamental wave. Here we present only the final result for the intensity of the XPW at exact PM conditions for the second step, $e_2 o_1 \rightarrow e_1$:

$$I_e = \frac{\epsilon_o c n}{2} \frac{{\sigma_1}^2 |A|^4}{2 \Delta {k_1}^2} \sin^2(\sqrt{2}\, \sigma_2 |A| L) , \tag{1}$$

where A denotes the complex amplitude of the input fundamental wave; $\Delta k_1 = k_2 - 2k_1$ is the phase mismatch of step (i), $o_1 o_1 \rightarrow e_2$; $\sigma_1 = 2\pi d_{\text{eff},ooe}/(\lambda_1 n_1)$; and $\sigma_2 = 2\pi d_{\text{eff},oee}/(\lambda_1 n_1)$; since $d_{15} \ll d_{22}$ in BBO crystal, we can write $d_{\text{eff},ooe} \approx -d_{22} \sin(3\varphi) \cos(\theta)$ and $d_{\text{eff},oee} \approx d_{22} \cos(3\varphi) \cos^2\theta$.

Equation (1) reveals that at low input intensity, when $|\sigma_2 A| L << 1$, the intensity of the XPW has a cubic dependence with respect to the fundamental intensity. At higher input intensities, when $|\sigma_2 A| L > 1$, the dependence is quadratic. For BBO crystal, $|\sigma_2 A| L = 1$ corresponds to an input intensity of 12.6 GW/cm^2 if a value of $d_{22} = 2.2$ pm/V is taken for the relevant second-order component and a crystal length of 0.15 cm is used. Our experimental investigation of the XPW power dependence (see Fig. 1) shows a quadratic dependence that is in good accordance with the prediction of Eq. (1). A similar experimental observation, i.e., quadratic dependence of a cubic process, was reported in Refs. 19 and 20, in which the authors investigated third-harmonic generation owing to cascading of TDSOP in a single quadratic crystal.

For $I_{o,\text{in}} \approx 500$ GW/cm^2, i.e., $|\sigma_1 A| L = 12$ and $L = 0.15$ cm, Eq. (1) predicts an efficiency of conversion of a linearly polarized wave into a XPW that is equal to 4×10^{-5}, i.e., twice as large as the measured efficiency. This prediction can also be considered to be in good agreement with the theory according to the approximation levels in the theoretical analysis presented above. As can be seen from Eq. (1), the efficiency of conversion of an ordinary wave into a XPW depends strongly on the magnitude of parameter Δk_1. For the BBO experiment reported here, the magnitude of Δk_1 was dramatically high, $\sim$8900 cm^{-1}, and this was the preponderant cause of the low conversion of the input ordinary wave into a XPW.

In conclusion, we report what is believed to be the first experimental demonstration of a direct nonlinear optical transformation of an ordinary wave into an extraordinary wave. Generation of a cross-polarized wave is not only important from the fundamental point of view but also has definite practical interest. We have calculated that a conversion efficiency that is 2 orders of magnitude higher than that reported here can be reached in an experiment performed in the infrared spectral region. However, the best approach for achieving high conversion of a linearly polarized wave into a XPW is to use some of the previously reported methods for simultaneous phase matching of two second-order processes.[18,22,23] In this case XPW generation will be efficient enough[4] for realization of intensity-dependent polarization switching or optical limiting.

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