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Induced spectral broadening about a second harmonic generated by an intense primary ultrashort laser pulse in ZnSe crystals

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The induced spectral broadening about a weak non-phase-matched 530-nm second-harmonic pulse is observed by propagating an intense primary 1060-nm-ps laser pulse through ZnSe crystals. This broadening is attributed to the induced phase modulation of excitation states.

It is important to obtain information on the spectral distribution of an ultrafast laser pulse propagating through matter for communications,[1] laser-pulse-duration control,[2] and laser spectroscopy.[3] The spectral broadening of an ultrafast laser pulse as it propagates through condensed matter has been attributed to the self-phase-modulation process[3-6] and four-wave mixing.[4] This was first observed by Alfano and Shapiro[4] sixteen years ago. This phenomenon has become extremely important in the reduction of the pulse duration of 8 fs.[7] In this Rapid Communication, we report on the first observation of induced spectral broadening (ISB) about a weak non-phase-matching second-harmonic pulse produced from the propagation of an intense primary picosecond laser pulse through a ZnSe crystal. This observation can lead to the future development of information coding and modulation in different spectral regimes.

The experimental arrangement used to generate and detect the induced spectral broadening is shown in Fig. 1. The laser system consists of a mode-locked Nd:glass laser with single-pulse selector and amplifier. The output laser pulse has about 2 mJ pulse energy and 8 ps duration at 1060 nm wavelength. The 1060-nm laser pulse was weakly focused into the sample. The spot size at the sample was about 1.5 mm in diameter. The second harmonic produced in these samples was about 10 nJ. The incident laser energy was controlled by changing the neutral-density filter F_1. The output signal light was sent through a $\frac{1}{2}$-m Jarrell-Ash spectrograph to measure the spectral distribution of the signal light. The 1060-nm incident laser was filtered out by using color filters F_2. A single crystal of ZnSe of about 1.6 cm in length was grown by Philips Laboratories[8] and a polycrystalline ZnSe sample of about 2.2 cm was purchased from Janos Inc. A 2-ps-time-resolution Hamamatsu streak camera system[9] was used to measure the signal pulse characteristics.

Typical spectra for non-phase-matching second-harmonic generation (SHG) and induced spectral broadening generated in a ZnSe crystal by 1060-nm laser pulses of various pulse energies are displayed in Fig. 2. The spectrum from a quartz sample was included in Fig. 2 for reference. The salient features of the ZnSe spectra indicate that the extent of the spectral broadening about the second-harmonic line at 530 nm depends on the intensity of the incident 1060-nm laser pulse. When the incident laser pulse energy was 2 mJ, there was significant spectral

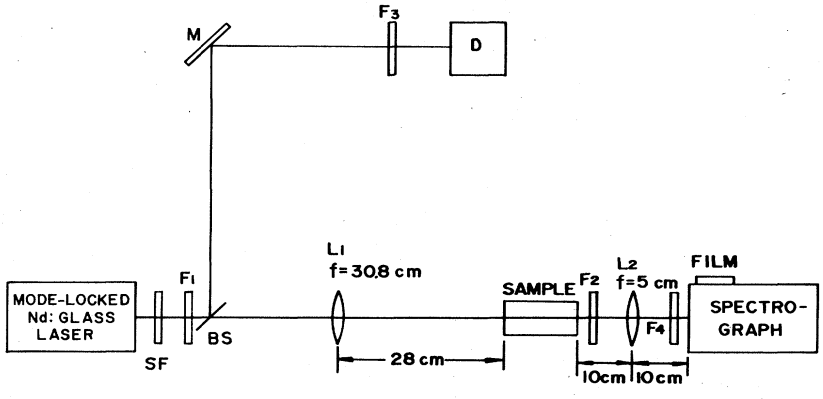

FIG. 1. Experimental setup. L_1 and L_2 are lenses, SF is a spectral filter to select either 1060 nm (1054 nm actual) or 530 nm (527 nm actual) or both wavelength lasers. F_1, F_3, and F_4 are sets of color filters and neutral density filters. F_1 consists of Hoya R720, Corning 2-3-67, and variable neutral density filters. F_2 is a set of Corning color filter.

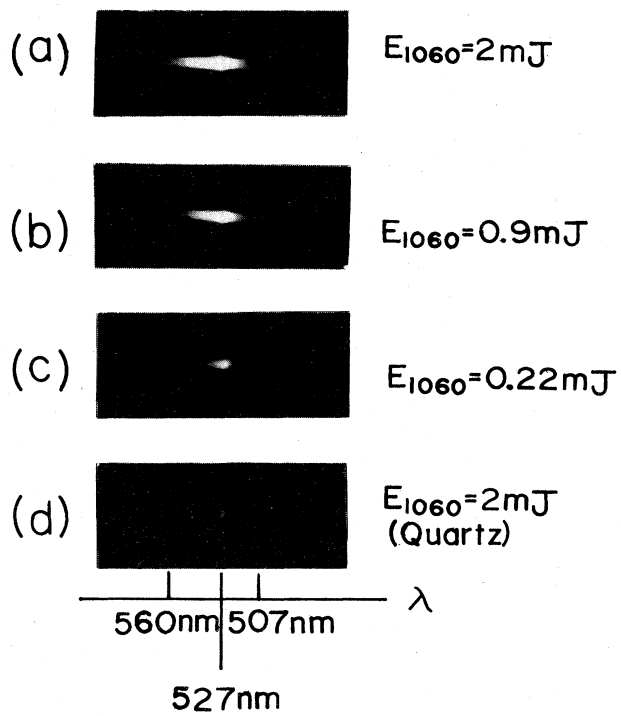

FIG. 2. Induced-spectral-broadening spectra in ZnSe crystal excited by an intense 1060-nm laser pulse. (a) $E_{1060}=2$ mJ, (b) $E_{1060}=0.9$ mJ, (c) $E_{1060}=0.22$ mJ, and (d) $E_{1060}=2$ mJ; ZnSe crystal was replaced by a 3.7-cm-long quartz crystal.

broadening of about 1100 cm^{-1} on the Stokes side and 770 cm^{-1} on the anti-Stokes side. There was no significant difference in the spectral broadening distribution measured in the single and polycrystalline materials. The intensity of induced spectral broadening was greater in polycrystalline ZnSe due to the longer length of the crystal. The ISB width is plotted for the Stokes and anti-Stokes sides as a function of the incident laser pulse energy in Fig. 3. The salient feature of Fig. 3 is that the Stokes side of the ISB is broader than the anti-Stokes side of the ISB. When the incident pulse energy was less than 1 mJ, the spectral broadening was found to be monotonically increasing on the pulse energy of 1060 nm.

The spectral broadening generated by sending an intense 80-μJ 530-nm 8-ps laser pulse alone through these ZnSe crystals was also measured to compare with the $\sim\pm1000$ cm^{-1} induced spectral broadening. The observed spectral broadening was only ~200 cm^{-1} when the energy of the 530-nm pulse was over 0.2 mJ. This measurement suggests that the self-phase modulation process from the 10 nJ SHG pulse in ZnSe is too insignificant to be the observed ~1000 cm^{-1} ISB.

The pulse duration of the ISB was measured by a streak camera at 550 nm using a 10-nm narrow-band filter. The pulse duration of this 10-nm band of ISB was about 9 ps, which was about the same as that of the incident laser pulse.

The observed spectra may arise from second-order and third-order optical nonlinearities. The second-order nonlinearity, Pockels coefficient r_{ijk}, of ZnSe is about 2×10^{-12} m/V (Ref. 10) and the value of the third-order nonlinearity n_2 measured from the optical Kerr effect by us is about half the value of $n_2(CS_2)=2\times10^{-11}$ esu. The Pockels term of ZnSe is responsible for the sharp ~100-cm^{-1} line at the SHG frequency. This weak 530-nm SHG pulse travels through the crystal together with the intense primary 1060-nm laser pulse. Because the large n_2 of ZnSe and the intense incident 1060-nm laser, the spectral width about the weak 530-nm pulse appears to broaden.[6] The intensity ratio of the induced spectral broadening to the non-phase-matched SHG signals displayed in Fig. 2 was ~0.1 for a given 1060-nm pulse energy. This conversion efficiency is much larger than the intensity conversion

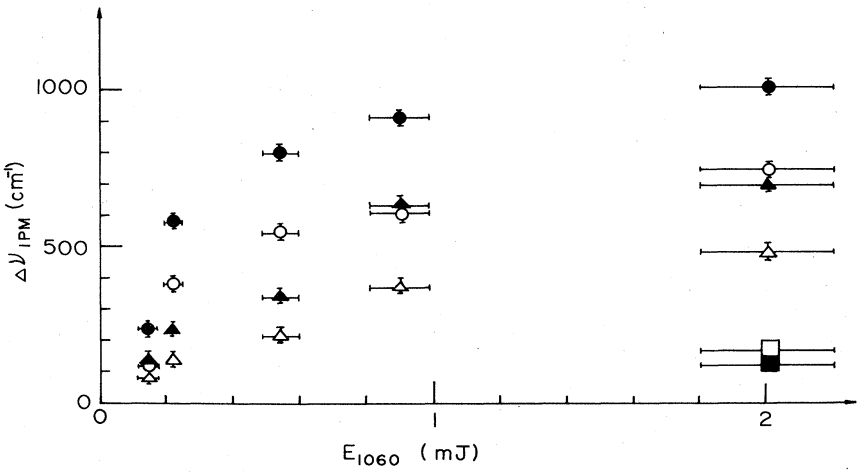

FIG. 3. Intensity dependence of induced spectral width about 530 nm in ZnSe pumped by 1060-nm laser pulse. The horizontal line is the incident laser pulse energy. ○: 2.2-cm-long polycrystalline ZnSe anti-Stokes broadening. ●: 2.2-cm-long polycrystalline ZnSe Stokes broadening. △: 1.6-cm-long single crystal ZnSe anti-Stokes broadening. ▲: 1.6-cm-long single crystal ZnSe Stokes broadening. □: 3.7-cm-long quartz crystal anti-Stokes broadening. ■: 3.7-cm-long quartz crystal Stokes broadening. The measured $\Delta\nu$ is defined to be the frequency spread from the 527 nm to the furthest detectable wavelengths measured either photographically or by an optical multichannel analyzer.

ratio of $\sim 10^{-4}$ observed for the induced supercontinuum about the 530-nm pulse in a BK-7 glass[11] by propagating an intense 2-mJ 1060-nm laser pulse and a weak 80-μJ 530-nm laser pulse simultaneously. By replacing the ZnSe with a quartz crystal, only a narrow line of 530 nm was measured as shown in Fig. 2(d). This observation can be explained by the lower n_2 value of quartz which is about 50 times smaller than n_2 of ZnSe. It follows that $\Delta\nu_{\text{quartz}}$ could be 50 times narrower than $\Delta\nu_{\text{ZnSe}}$.

A simple model can be used to explain the general features observed for SHG and ISB produced by an intense laser pulse in a medium with large second-order and third-order optical nonlinearities. The incident electric field at ω_0 can be assumed to be

$$E(t)=E_0\exp(-t^2/\tau^2)\cos[\omega_0 t+\phi(t)] , \tag{1}$$

where E_0 is the amplitude of the primary wave and $\phi(t)$ is the phase term of the incident field propagating in ZnSe. The phase behaves as

$$\phi(t)=nz\omega_0/c\sim z_0\omega_0/c\,[n_0+n_1E_0\exp(-t^2/\tau^2)\cos(\omega_0 t-n_0\omega_0 z_0/c)+n_2E_0^2\exp(-2t^2/\tau^2)\cos^2(\omega_0 t-n_0\omega_0 z_0/c)+\cdots] , \tag{2}$$

where n_1 in ZnSe is related to three nonzero γ_{ijk} coefficients: $\gamma_{231}=\gamma_{312}=\gamma_{123}$. If the incident laser field is polarized along the x direction, $n_1\sim(\frac{1}{2})n_0^3\gamma_{231}$. For a thin medium and neglecting dispersion, Eq. (2) can be substituted into Eq. (1) to yield the total field as

$$\begin{aligned}E(t)=E_0\exp(-t^2/\tau^2)[&A\cos(\omega_0 t-\Omega-\alpha_2)+B\cos(\omega_0 t-\Omega+\alpha_2)+C\sin(\omega_0 t-\Omega-\alpha_2)+D\sin(\omega_0 t-\Omega+\alpha_2)\\&+E\cos(2\omega_0 t-2\Omega-\alpha_2)+F\cos(2\omega_0 t-2\Omega+\alpha_2)\\&+G\sin(2\omega_0 t-2\Omega-\alpha_2)+H\sin(2\omega_0 t-2\Omega+\alpha_2)] ,\end{aligned} \tag{3}$$

where

$$\Omega=n_0\omega_0 z_0/c , \tag{4}$$

$$\alpha_2=\left(\frac{1}{2c}\right)n_2\omega_0 z_0 E_0^2\exp(-2t^2/\tau^2) , \tag{5}$$

and

$$\alpha_1=(1/c)n_1\omega_0 z_0 E_0\exp(-t^2/\tau^2) \tag{6}$$

are induced parameters and $A,B,C,\ldots$ are Taylor's-series-expanded Bessels functions with arguments α_1 and α_2. In Eq. (3), one can clearly identify the induced spectral broadening (ISB) terms $2\omega_0\pm\partial\alpha_2/\partial t$ around the second-harmonic frequency $2\omega_0$ due to E_0^2, while regular self-phase-modulation (SPM) arises from $\omega_0\pm\partial\alpha_2/\partial t$ terms. The maximum extent of the spectral broadening by induced phase modulation (IPM) about $2\omega_0$ is given by

$$\Delta\nu_{\text{IPM}}\propto n_2E_0^2L/\tau . \tag{7}$$

Qualitatively, Eqs. (3) and (7) explain the observed spectral extent of the induced broadening as a function of incident pulse energy, crystal length, and samples of different n_2.

The induced spectral broadening generated around $2\omega_0$ by an intense picosecond laser pulse at ω_0 in ZnSe crystals has been observed and attributed to the induced phase modulation of the excitation in the crystal at $2\omega_0$ caused by the intense ω_0 laser pulse. The intensity ratio of the induced-spectral-broadening spectra to the non-phase-matched SHG is about 0.1. In all of this ISB observation, the spectral broadening of the Stokes side was broader than that of the anti-Stokes side. These results are displayed in Fig. 3. This measurement is opposite to our previous measurements[4,11] and theories[5,6] of SPM and IPM processes. The spectral broadening of SPM and IPM in dielectrics like glasses or water has shown wider anti-Stokes broadening. There are at least two reasons which may explain the difference. First, the anti-Stokes side of the 530-nm broadening has a larger absorption coefficient. Below 500 nm, there is little light which can be transmitted through these long ZnSe crystals. Second, in our previous theoretical derivation, the group velocity was assumed to be constant, independent of frequency. This assumption is generally true for materials we chose where group velocity differences of the generated spectrum are small. In the induced spectral experiment in ZnSe, the difference of the index of refraction between the incident pump 1060-nm pulse and the observed 530-nm spectral regime is about 0.8. Furthermore, the group velocity dispersion between the anti-Stokes side of 530 nm and Stokes side of 530 nm is not small. Therefore, our previous observations and theories in the transparency wavelength regimes in materials may require some modifications. The conversion efficiency of anti-Stokes continuum generation decreased more than that of the Stokes continuum around 530 nm. There is more absorption at the trailing half of the pulse than that from the leading half.[12] These two factors may also explain the saturation of the induced spectral width as a function of the pump pulse energy displayed in Fig. 3. Further theoretical analysis and time-dependent measurements are being pursued to clarify these points.

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