Investigation of Symmetries of Second-Order Nonlinear Susceptibility Tensor Based on THz Generation

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Abstract: We demonstrate that THz generation can be a sensitive technique for investigating symmetries of second-order nonlinear susceptibility tensor.

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1. Introduction

For second-order nonlinear susceptibility, Kleinman symmetry condition must be satisfied when all the frequencies of the waves participating in the parametric interaction are much lower than the lowest resonance frequency of a nonlinear material. In such a case, one can permute the indices of the elements for the second-order nonlinear susceptibility tensor without permuting the frequencies. Furthermore, spatial symmetry of a nonlinear material imposes additional relations among some of the elements of the second-order nonlinear susceptibility tensor.

Here, we demonstrate that THz generation can be a sensitive technique for investigating symmetries of second-order nonlinear susceptibility tensor. Indeed, using GaSe as an example, our result illustrates that both Kleinman and spatial symmetries are violated for THz generation, which can be attributed to the contribution of the slightly-lossy frequency-dependent ionic second-order nonlinear susceptibility of the material.

If Kleinman and spatial symmetry conditions are satisfied, the elements for the second-order nonlinear susceptibility tensor for GaSe satisfy the relations of $d_{16} = d_{21}$ and $d_{22} = - d_{21}$. When the THz output frequency is close to the transverse-optical (TO) phonon frequency of 6.41 THz (46.8 μm) for GaSe [1], GaSe becomes lossy, and therefore, the ionic second-order nonlinear coefficients are frequency dependent. In the past, GaSe was used to generate THz pulses from ultrafast laser pulses with the output power reaching 5.4 μW [2].

2. GaSe crystals and experiment

GaSe crystals in our experiment were grown by a Bridgman method. They were doped by In and Cr in order to improve their hardness. They have a typical dimension of 13×8×1 mm^3 with the two facets perpendicular to the *c* axis being separated by 1 mm. Broadband THz pulses were generated by using a Ti:sapphire regenerative amplifier at the wavelength of 782 nm and a coherent beam at 391 nm achieved through frequency-doubling in a BBO crystal. The pulse duration and repetition rate for the two output beams are 180 fs and 250 kHz, respectively. When the excitation beam was focused onto one of the crystal facets, the THz radiation was collimated and then focused to a power meter for measuring the THz output powers in the transmission and reflection geometries.

Table 1. Output powers measured on GaSe crystals.

Crystal ID	Dopant (ppm)	Output Power	
		μW	nW
GSU7903	undoped	19.41	60
GSI8701	In (500)	22.93	37
GSI7801	In (3000)	16.33	43
GSC8902	Cr (500)	19.45	37

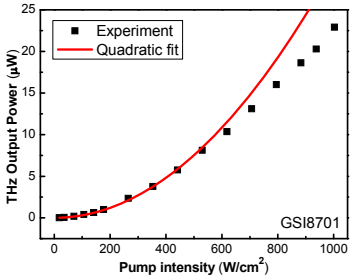

Fig. 1. Typical power dependence of THz generated from a GaSe crystal.

3. Results and discussions

At the average pump power of 1.14 W at 782 nm, the highest THz output powers were measured to be in the range of 16.3-22.9 μW from four GaSe crystals in the transmission geometry, see Table 1. The highest output power

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among different doped crystals was measured to be 22.9 μW from GSI8701, *which is a factor of 4.2 higher than our previous result [2]*. It appears to us that the second-order nonlinear coefficient can be increased by introducing a small amount of In. This is consistent with one of the previous reports [3]. However, based on our result, introducing Cr at the level of 500 ppm did not affect the second-order nonlinear coefficient. According to Fig. 1, the power dependence is close to a quadratic dependence. Above 618 W/cm^2, the deviation of our data from the quadratic dependence is caused by two-photon absorption of GaSe at 782 nm [4]. We have attributed the mechanism of the THz generation in these four GaSe crystals to optical rectification.

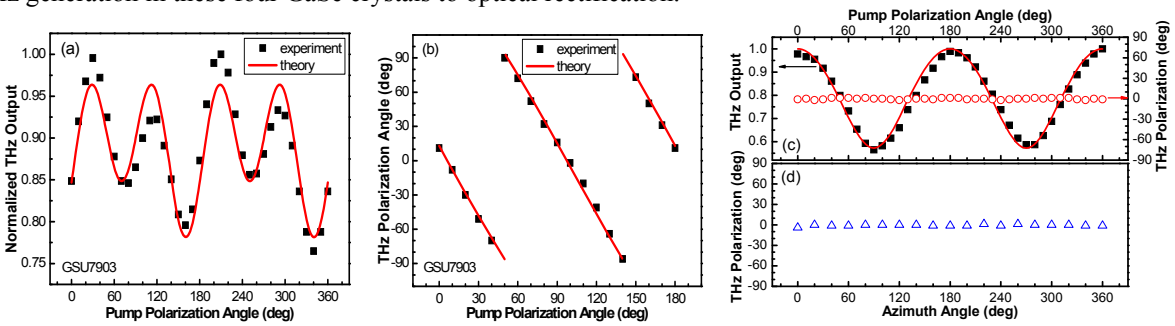

Fig. 2. (a) THz output power vs. pump polarization angle at 782 nm, (b) THz polarization vs. pump polarization angle at 782 nm, (c) THz output power and polarization vs. azimuth angle at 391 nm, and (d) THz polarization vs. azimuth angle at 391 nm. Squares, circles, and triangles correspond to data; solid curves correspond to fitting to the data points.

At normal incidence, THz nonlinear polarizations are given by $P_x = 4d_{16}E_xE_y$ and $P_y = 2d_{21}E_x^2+2d_{22}E_y^2$. Using $E_x = E_0\cos\theta$ and $E_y = E_0\sin\theta$, where θ is the pump polarization angle, the THz intensity and polarization angle are

$$I_T \propto 2\left(\frac{d_{16}}{d_{22}}\right)^2 \sin^2(2\theta)+\left(\frac{d_{21}}{d_{22}}\right)\sin^2(2\theta)+2\left(\frac{d_{21}}{d_{22}}\right)^2 \cos^4\theta+2\sin^4\theta, \theta_T = \tan^{-1}\left[\frac{(d_{21}/d_{22})\cos^2\theta+\sin^2\theta}{(d_{16}/d_{22})\sin(2\theta)}\right] \quad (1)$$

By adjusting d_{16}/d_{22} and d_{21}/d_{22}, we can use Eq. (1) to achieve the nonlinear least square fit to the data presented in Fig. 2(a). As a result, we have obtained $d_{16} \approx 0.979d_{21}$ and $d_{22} \approx -1.04d_{21}$. In comparison, if $d_{16} = d_{21}$ and $d_{22} = -d_{21}$, the THz output power would be independent of the pump polarization angle. *Therefore, even when d_{16} and d_{22} deviate from d_{21} and $-d_{21}$ by as small as 2% and 4%, respectively, the THz output power exhibits a measurable oscillation as a function of the pump polarization angle.* Based on our result, we have demonstrated that such an oscillation characteristic can be used to determine d_{16}/d_{21} and d_{22}/d_{21} for GaSe crystals. We believe that such small deviations originate from the low loss for the THz wave. Indeed, a THz wave can be coupled with TO phonons in GaSe, resulting in the absorption of the THz wave [5]. Since the peak output wavelength of 300 μm is much longer than the corresponding wavelength for the TO phonons, the THz output wave suffers from low loss. Consequently, due to the slightly-lossy ionic second-order nonlinear susceptibility contributing to the THz generation, Kleinman symmetry condition is no longer satisfied. On the other hand, due to asymmetric residual strain following the crystal growth and cleavage, spatial symmetry is violated. Despite of the oscillation observed in Fig. 2(a), the dependence of the THz polarization angle on the pump polarization angle is close to linear, see Fig. 2(b).

When we used the wavelength of 391 nm to pump the GaSe crystals, the THz output powers measured under the reflection geometry are 2-3 orders of magnitude lower, see Table 1. At an incident angle of 71° for the pump beam, the polarization angle for the THz beam is kept as a constant regardless of the pump polarization angle or azimuth angle, see Figs. 2(c) and 2(d). Such a behavior is a clear indication that the mechanism for the THz generation pumped at 391 nm is photocurrent surge. THz output power vs. polarization angle exhibits two-period oscillation within 0-360°. This is completely different from the THz characteristic due to optical rectification. After including the Fresnel loss of the pump beam, our data in Fig. 2(c) can be well fitted by our theory based on photocurrent surge.

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4. References

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