Ultrashort pulse characterization by ultra-thin ZnO, GaN, and AlN crystals

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Abstract: Ultra-thin semiconductor crystals were investigated as nonlinear materials for second-harmonic generation. Nonlinear susceptibilities of sub-micrometer-thick ZnO, GaN, and AlN crystals were measured, and these crystals were used for sub-10-fs pulse measurement by a fringe-resolved autocorrelation method. We found that a one-cycle pulse could be characterized by using these ultra-thin-film crystals.

OCIS codes: (320.7100) Ultrafast measurements; (320.7110) Ultrafast nonlinear optics; (310.6860) Thin films, optical properties

References and links

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1. Introduction

Recent progress in ultra-short pulse generation has enabled attosecond pulse generation using high-order harmonics of Ti:sapphire laser pulses [1, 2]. The main difficulty of attosecond pulse generation was how to measure it. The nonlinear effects of multi-photon absorption or optical-electric field acceleration were used for XUV-attosecond pulse measurement. In the

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visible or infrared region, ultra-short pulse generation is approaching the one-optical-cycle pulse regime through the use of a spectrum broadening and chirp compensation technique [3]. Another approach to sub-femtosecond pulse generation in visible and infrared regions is Fourier synthesis of sub-harmonic pulses [4, 5]. The coherent synthesis of independent laser pulses or sub-harmonic pulses from an optical parametric oscillator was investigated in order to generate sub-femtosecond pulses [6-8]. Phase-coherent, multicolor femtosecond pulses were generated in these experiments. The next step is pulse characterization of the synthesized light. There are some difficulties in measuring one-cycle or sub-femtosecond pulses since the spectrum is too broad to adopt conventional autocorrelation methods. First, the pulse duration changes with propagation in the nonlinear material to generate second harmonics (SH). Second, extremely broadband second-harmonic generation (SHG) is necessary. Ultra-thin nonlinear crystals have to be used in order to overcome these problems. The thickness of the commercially available nonlinear crystal is limited to about 5 micrometers. Thinner crystals for SHG in the autocorrelation technique have to be used for one-cycle optical pulse measurement. Some wide-band-gap, thin-film semiconductor crystals are available for this. ZnO, GaN, and AlN thin-film crystals were investigated for SHG [9-12]. However, it was not clear if they could be used for ultra-short pulse characterization in terms of the pulse propagation and the SH conversion bandwidth.

In this report, we characterize the nonlinear properties and pulse propagation of molecular-beam epitaxially grown semiconductor films, and demonstrate ultrashort pulse measurement by using them. Second-order nonlinear coefficients of ZnO, GaN, and AlN were compared with that of BBO crystal, and were found to have large enough nonlinear coefficients and wide enough conversion bandwidth for SHG to measure sub-10-fs Ti:sapphire laser pulses by conventional auto-correlation techniques.

2. Pulse propagation in semiconductor ultra-thin films

A femtosecond pulse is broadened by propagation in a material. The pulse broadening can be estimated by the material dispersions. We adopted Sellmeier's equations of bulk semiconductors for the propagation calculations since the refractive indexes of ZnO, GaN, and AlN thin-film crystals are not clearly established. The adopted Sellmeier's equations of ZnO, GaN, AlN are $n_{ZnO} = 1.9148 + 0.0569/\lambda^2 - 0.0136/\lambda^4 + 0.002168/\lambda^6$, $n_{GaN} = (1 + 4.295 \times \lambda^2/(\lambda^2 - 0.1899^2))^{0.5}$, $n_{AlN} = (1 + 3.335 \times \lambda^2/(\lambda^2 - 0.1392^2))^{0.5}$, where λ denotes the wavelength in micrometers [13-15]. We used the refractive index of the extra-ordinary polarized light for the propagation calculation since the polarization of the incident beam is perpendicular to both a- and b- axes, although the incident angle is not exactly parallel to the c axis.

We assumed a one-optical-cycle transform-limited pulse at a center wavelength of 800 nm as the incident light. The incident electric field in the time domain is Fourier transformed to the frequency domain and multiplied by an additional phase term by the material. The inverse-Fourier transformation gives the propagated electric field. We calculated only the propagation of the fundamental pulse. The pulse is expressed by using envelope and carrier terms, and this calculation uses no approximation because this calculation deals only the linear dispersion. One should care about the slowly-varying envelope approximation in this time region when the nonlinear processes such as wavelength conversion or nonlinear refractive index are included. Figure 1 plots the result of the pulse propagation calculation. Figure 1(a) depicts the incident and output electric field of 5μm-thick BBO crystal. The output time shifts in accordance with the group velocity delay produced by a crystal, and the output electric field shape changes due to the material dispersion. Although the pulse duration change by the 5μm-thick BBO crystal is not so large, it is too thick to estimate the absolute optical-electric field from this calculation because the optical-electric field shape changes significantly in the material. Thus BBO crystal can not be used for the field characterization of Fourier-synthesized attosecond pulse train by ultra-broad femtosecond pulses [4-6].

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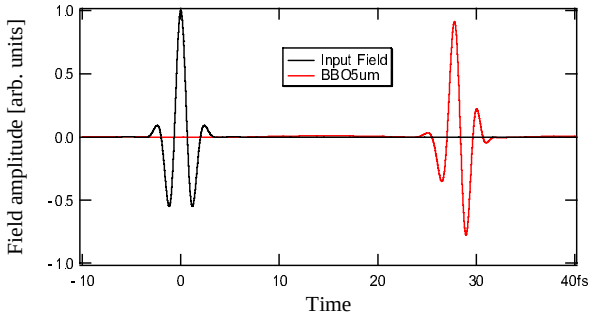
Figure 1(b) illustrates the pulse propagation of ultra-thin-film semiconductor crystals. The crystal thickness is assumed to 0.3μm. The output electric-field shapes do not change so much compared to the BBO calculation. The 0.3μm-thick ZnO, AlN, and GaN crystals can then be used for pulse characterization of a one-cycle optical pulse in terms of the pulse broadening due to the propagation.

(a)



(b)

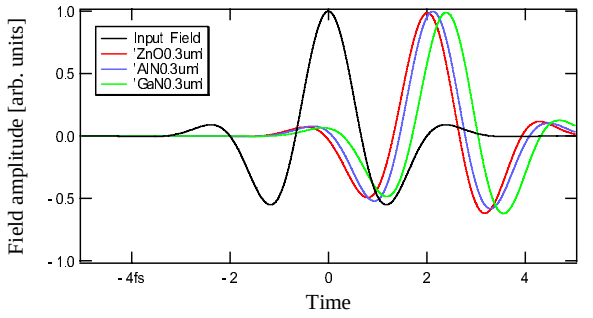


Fig. 1. One-optical-cycle electric field propagation through 5μm-thick BBO crystal (a). Incident (output) electric field is indicated by the black (red) curve. Pulse propagation results of 0.3μm-thick ZnO, GaN, and AlN crystal (b). The output electric fields of ZnO, GaN, and AlN crystals are represented by red, green, and blue curves.

3. Conversion bandwidth for second-harmonic generation of semiconductor crystals

The next problem for extremely wide-band SHG is the conversion bandwidth. Thicker crystals have narrower bandwidths of the SHG due to the phase mismatching. For ultra-wideband, SHG, the phase-matching condition Δk increases at the edge of the spectrum even if the phase-matching angle is defined at a central wavelength. The coherent length and the bandwidth of the frequency conversion have to be numerically calculated. Figure 2 plots the coherent lengths and the wavelength dependence of the relative SH intensity of several nonlinear crystals.

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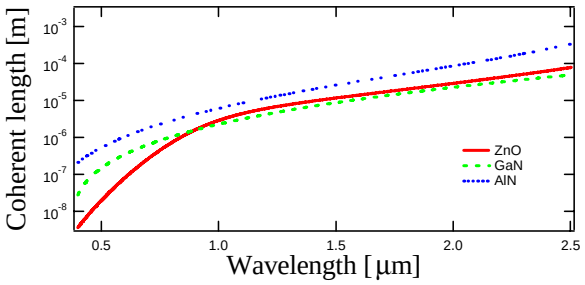
(a)



(b)

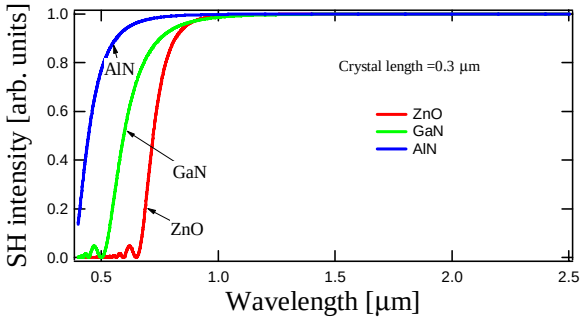


Fig. 2. (a). Coherent lengths of ZnO (red curve), GaN (green curve), and ZnO (blue curve) crystals. (b) Relative intensity of the SH light versus the fundamental wavelength.

The coherent length of AlN is the greatest due to the similarity of the refractive indexes for ordinary and extra-ordinary waves. The conversion bandwidth of AlN is greatest because its band gap is widest. To measure ultra-short Ti:sapphire laser pulses, GaN and AlN should be used because of their conversion bandwidth. The next question is the conversion efficiency of each crystal.

4. Nonlinear coefficient measurements of ultra-thin films

ZnO, GaN, and AlN thin-film crystals were grown by molecular-beam-epitaxial (MBE) methods on sapphire crystals. Their c-axes are perpendicular to the substrate surfaces. The uniformity of a- and b-axes of these samples is not clear, but they should be good since they were grown by MBE. The refractive indexes of polarized light parallel to the c-axes are much different from those of a- and b-axes polarized light. However, the difference is not large to achieve phase matching. Therefore, full-tensor of the second-order nonlinear optical susceptibility should be considered for SHG. ZnO, GaN, and AlN crystals we used are hexagonal materials with 6-mm symmetry, then d_{33}, $d_{31}=d_{32}$, and $d_{15}=d_{24}$ should be considered. The effective nonlinear susceptibility can be derived considering the wave vector of the fundamental beam and the projection of the second-order polarization. For hexagonal materials with 6-mm symmetry, the effective second-order nonlinear susceptibility of the s- and p-polarized SH lights are given by $d_{eff}^{s} = 2d_{15}\sin\theta\sin\rho\cos\rho$, and $d_{eff}^{p} = \left(d_{33}\sin^3\theta + 2d_{15}\sin\theta\cos^2\theta + d_{31}\sin\theta\cos^2\theta\right)\cos^2\rho + d_{31}\sin\theta\sin^2\rho$, respectively, where ρ is the fundamental polarization angle, and θ is the incidence angle in the crystal. In our experiment, both p- and s-polarized SH lights were observed. Then the total effective second-order nonlinear susceptibility can be written by $d_{eff} = \sqrt{d_{eff}^{s\ 2} + d_{eff}^{p\ 2}}$. Since there is a relation of

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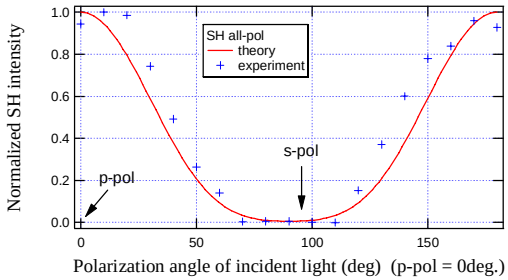
$d_{31}=d_{32}=d_{15}=d_{24}$ in a case of AlN, d_{eff} can be simplified as $d_{eff} = d_{33}\sin^3\theta + 3d_{31}\sin\theta\cos^2\theta$ when the fundamental light is p-polarized.

Sub-10-fs Ti:sapphire laser pulses were focused by an f=25.4mm parabolic mirror with an incident angle of 45 degrees. The SH powers of thin crystals were measured by a photomultiplier tube (Hamamatsu R7400 U-3). Figure 3(a) shows the measured dependence of the SH intensity on the fundamental polarization angle with AlN crystal. Measured SH power was well agreed with the theory. The SH powers of semiconductor crystals were compared with that obtained by a 10μm-thick BBO crystal. The ZnO crystal is 220nm thick; the AlN crystal, 340nm thick; and the GaN crystal, 340nm thick. 340-nm GaN is grown with a thin buffer layer on a sapphire crystal. This layer is negligible since the generated SH power from GaN layer is two-orders higher than that from this buffer layer. The SH power with thinner AlN crystal was below the detection limit, then the observed SHG was not the surface SHG. The obtained SH powers and relative nonlinear coefficients of thin-film crystals are depicted in Fig. 3(b). Blue rectangles represent measured SH powers, and red circles, the normalized nonlinear coefficients (d_{eff}) relative to that of BBO. The phase-match angle of BBO crystal is θ=29.2 degrees. The SH power of BBO crystal is highest due to the greater crystal thickness, although its d_{eff} was lowest. The d_{eff} of GaN is seven times larger than that of BBO crystal. This measurement indicates that ultra-thin-film semiconductor crystals have higher nonlinear coefficients than BBO crystals. These crystals can thus be used for ultra-short pulse characterization by the autocorrelation technique.

(a)



(b)

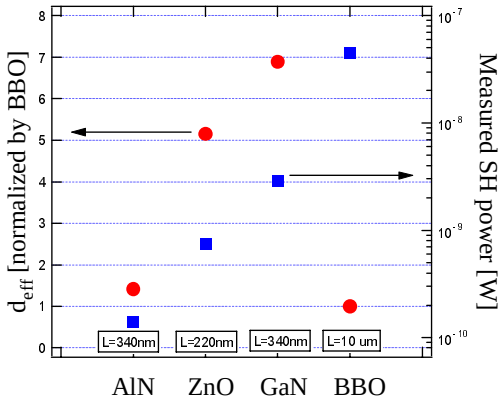


Fig. 3. (a). Measured dependence of the SH intensity on the fundamental polarization angle by AlN crystal. (b) Second-harmonic powers and nonlinear efficiencies of semiconductor thin-film crystals.

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5. Auto-correlation measurement of sub-10fs Ti:sapphire laser pulse

A mode-locked Ti:sapphire laser oscillator produces an ultra-short femtosecond pulse train with a repetition frequency of 80MHz. The output power is about 200mW with a spectrum width exceeding 140nm. The Fourier-transformed-limited pulse width is about 7fs in this spectrum. The pulse train was split by a Cr-coated beam splitter with a broad spectrum bandwidth. The split beams were recombined collinearly after some time delay by a piezoelectric transducer. The superimposed pulse was focused into thin-film crystals by a parabolic mirror, and generated SH was detected by a photomultiplier tube. SH power versus changing time delay was recorded by an oscilloscope. Figure 4 plots the obtained laser spectrum and autocorrelation traces. Figure 4(a) is the mode-locked Ti:sapphire laser spectrum. The spectrum is in the region of 670nm to 950nm, thus the absorption edge of the crystal should be less than 335 nm.

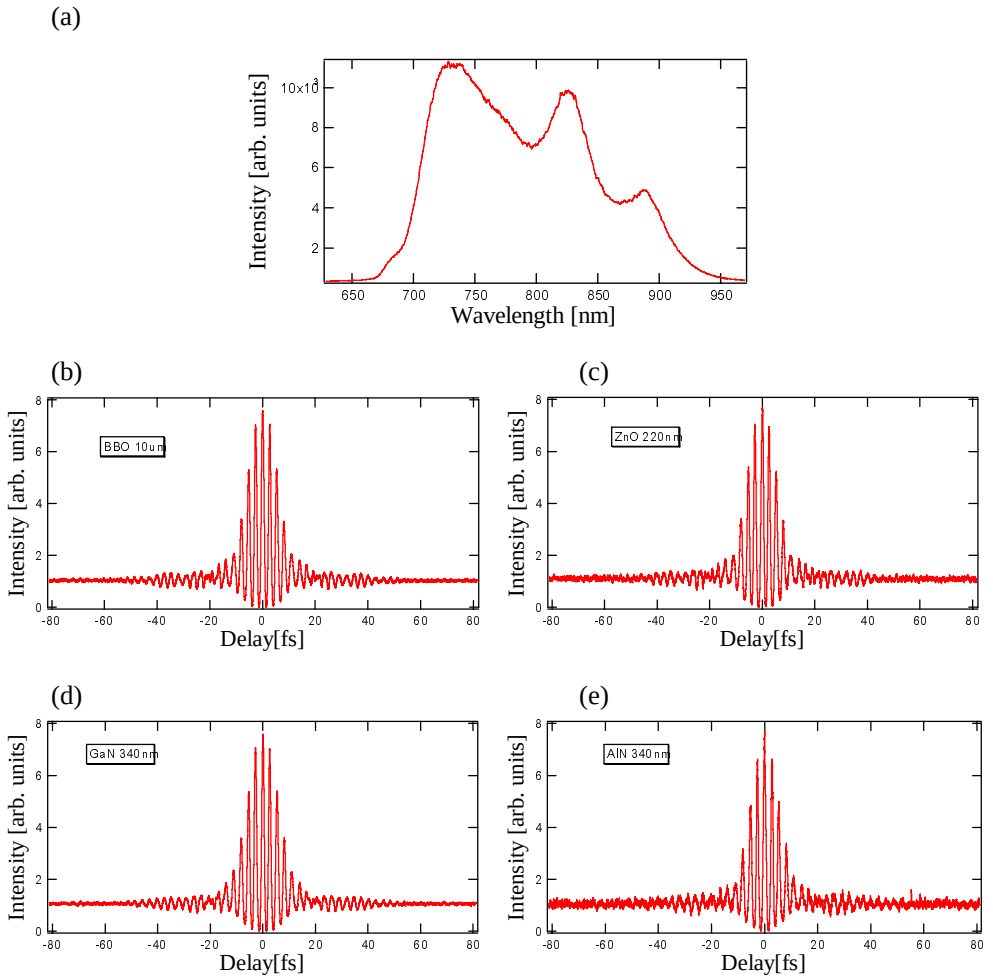

Fig. 4. Ti:sapphire laser spectrum (a), and autocorrelation traces obtained by 10µm BBO (b), 220nm ZnO (c), 340nm GaN (d), and 340nm thick AlN (e) crystals.

Figure 4(b) graphs the autocorrelation trace of the Ti:sapphire laser pulse obtained by using a 10µm-thick BBO crystal, which is shown as a reference. Figures 4(c), 4(d), and 4(e) are the

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autocorrelation traces obtained by using 220nm-thick ZnO, 340nm-thick GaN, and 340nm-thick AlN crystals. A contrast ratio of 1:8 was obtained for all crystals, thus all ultra-thin crystals worked well for ultra-short pulse measurement. All crystals can be used for 7fs pulse characterization. However, 10μm-thick BBO crystals cannot be used for shorter pulse measurements due to their narrow conversion bandwidth. AlN crystals have the greatest bandwidth and least propagation effects. Thus, thin AlN crystals are promising candidates for one-cycle optical pulse characterization, although they have poor conversion efficiency.

6. Summary

The pulse propagation and the second-harmonic conversion of ultra-thin semiconductor crystals were investigated by numerical calculation. The results reveal that sub-micrometer-thick ZnO, GaN, and AlN crystals are appropriate for ultra-wide-spectrum pulse characterization. The nonlinear coefficients of these crystals for SHG were measured and were found to exceed that of BBO crystal. These crystals were thus found to be appropriate for ultrashort pulse measurement by autocorrelation techniques and were used to characterize 7-fs Ti:sapphire laser pulses.

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