Beam Deflection Measurements of Nondegenerate Nonlinear Refractive Indices in Direct-gap Semiconductors

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Abstract: We use the beam-deflection method to measure nondegenerate nonlinear refractive indices of ZnO and ZnSe and show, in agreement with theory, extremely nondegenerate nonlinear refraction is significantly larger than in the degenerate or near-degenerate case.
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1. Introduction

Bound-electronic nonlinear refraction (NLR), also known as the Kerr effect, is one of the dominant optical nonlinearities in the transparency range of a material, and knowledge of the magnitude, sign and dispersion of the nonlinear refractive index n_2 is critical for applications where cross-phase modulation is the essential mechanism, such as dual-wavelength optical switching [1]. Extensive experimental work has been devoted to the study of degenerate n_2 (D-NLR) in various semiconductors and dielectrics [2, 3], but the nondegenerate $n_2(\omega_1;\omega_2)$ (ND-NLR), namely the NLR at frequency ω_1 caused by the presence of a strong excitation at frequency ω_2, is less often reported particularly for the very nondegenerate case, and also for spectral regions where two-photon absorption (2PA) is present. Sheik-Bahae *et al.* calculated both degenerate and nondegenerate n_2 and the dispersion relations for direct-gap semiconductors by applying a nonlinear Kramers-Kronig (KK) transformation on the nondegenerate nonlinear absorption (ND-NLA) spectrum which is derived from a two-parabolic band model [3, 4]. Through the dispersion integral, $n_2(\omega_1;\omega_2)$ is related with the ND-NLA coefficient $\alpha_{NL}(\omega_1;\omega_2)$ by

$$n_2(\omega_1;\omega_2) = \frac{2}{\pi}\int_0^\infty \frac{\alpha_{NL}(\omega;\omega_2)}{\omega^2-\omega_1^2}d\omega. \qquad (1)$$

The main three mechanisms responsible for $\alpha_{NL}(\omega_1;\omega_2)$ are 2PA, electronic Raman, and optical Stark effect. In general, the Raman effect adds a small, positive and relatively nondispersive overall contribution to $n_2(\omega_1;\omega_2)$; the 2PA contribution to $n_2(\omega_1;\omega_2)$ grows from low frequencies to a maximum near the onset of 2PA and then turns negative as $\hbar\omega_1+\hbar\omega_2$ approaches the bandgap; the Stark effect gives an overall negative contribution which becomes resonant and thus strongly negative at the bandgap. We previously determined both theoretically and experimentally that extremely nondegenerate (END) 2PA gives orders of magnitude enhancement of 2PA compared to its degenerate counterpart [5]. The KK calculations also predict large enhancements for END $n_2(\omega_1;\omega_2)$ although it rapidly changes sign from positive to negative as $\hbar\omega_1+\hbar\omega_2$ nears the bandgap. Using our recently developed optical beam deflection technique with unprecedented sensitivity to nonlinearly induced phase distortion [6], we are able to investigate the ND-NLR theory experimentally. In this work, the ND-NLR indices along with ND-2PA coefficients of ZnO and ZnSe crystals are measured via beam deflection.

2. Experimental Results

The essential principles of the beam deflection measurement technique are shown in Fig. 1(a). As described in Ref [6], a strong excitation pulse (at ω_e) induces an index gradient in the sample due to NLR, which in turn deflects the weak probe pulse (at ω_p) that has been displaced to the wings of the near-Gaussian spatial profile of the excitation beam. A quad-segmented photodetector simultaneously measures the change in total transmission due to NLA and the beam deflection due to NLR from the difference between the signals from the detector segments. In this work, a commercially available Ti:Sapphire amplified laser system (Coherent Legend Elite Duo HE+) producing 12mJ, ~40 fs (FWHM) pulses at a 1 kHz repetition rate pumps an optical parametric amplifier (TOPAS-HE) with 10mJ to produce the 1440nm excitation pulses. The probe pulses are generated using a portion of the excitation to produce a white-light continuum (WLC) in a 5 mm thick sapphire crystal. The WLC is spectrally filtered by narrow bandpass interference filters ($\Delta\lambda\sim 10$ nm) at wavelengths (λ_p) 950nm, 850nm, 750nm, 650nm, 570nm, 532nm and 480nm for each ND combination with the 1440nm excitation. The probe pulse is focused ~3-5× smaller than the excitation spot size w_e and displaced by $w_e/2$ to experience the largest index gradient induced by the excitation. As shown in Fig. 1(a), when the probe undergoes a horizontal deflection, the quad-segmented detector simultaneously measures the total transmitted energy E and the difference between the two detector segments $\Delta E = E_{left} - E_{right}$ as a function of the probe delay τ with respect to the excitation. As a calibration, a 1 mm thick fused silica sample is measured at all wavelength combinations. As shown in Fig.1 (b) for $\lambda_p = 750$ nm, the $\Delta E/E$ trace follows the cross-correlation between excitation and probe pulses which gives a fit of n_2 = 2.4×10^{-7}cm^2/GW, consistent with literature values [3].

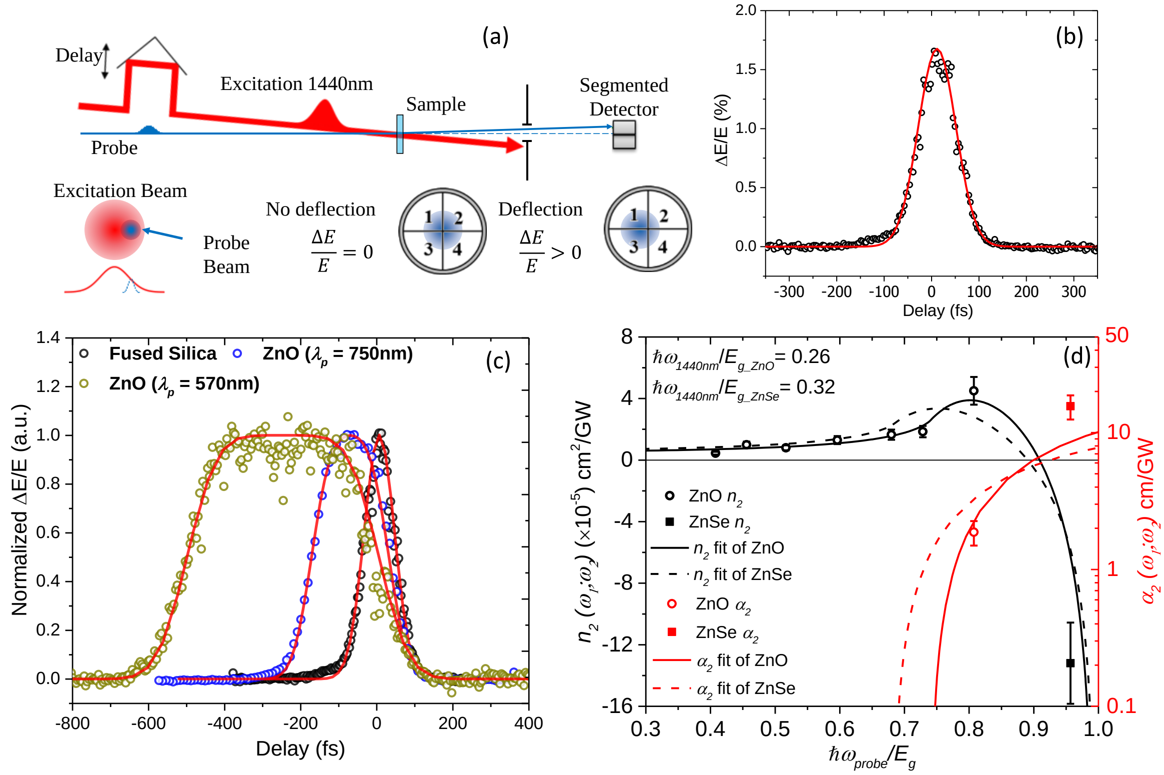

Fig. 1. (a) The beam deflection measurement schematics: excitation and probe overlap geometry and position sensitive quad-segmented photodetector; (b) $\Delta E/E$ of fused silica with λ_p = 750 nm, which is fit with n_2 = 2.4×10^{-7} cm^2/GW; (c) Typical $\Delta E/E$ (normalized) of ZnO with λ_p = 750 nm and 570 nm where GVM is considered in the analysis, compared with fused silica; (d) ND-NLR and ND-2PA of ZnO and ZnSe, along with the calculated curves by two-band theory with the KK transformation method [4].

We perform the beam deflection measurements on a 0.53mm thick ZnO with probes at $\hbar\omega_p/E_{g\_ZnO}$ = 0.40, 0.46, 0.52, 0.60, 0.68, 0.73 and 0.81 of the bandgap energy (E_{g_ZnO}). Fig. 1(c) shows the typical $\Delta E/E$ (normalized) in such ND beam deflection measurements, where at shorter λ_p the group velocity mismatch (GVM) causes excitation (fast) and probe (slow) pulses to walk through each other as they propagate through the sample. Compared with fused silica where GVM is much less pronounced, substantial GVM has to be accounted for in the analysis for ZnO to extract values of n_2 [7]. The moderate normal dispersion of n_2 approaching the 2PA resonance (when λ_p varies from 0.4-0.73E_{g_ZnO}) can be resolved from beam deflection measurements, as shown in Fig.1(d). Particularly at λ_p=480nm (0.81E_{g_ZnO}) where 2PA starts, ND-NLR and ND-2PA are measured simultaneously, and the analysis of n_2 takes into account the small contribution to $\Delta E/E$ from the translational beam shift due to 2PA. The KK transformation agrees on both the dispersion relation and the magnitude of $n_2(\omega_p;\omega_{1440nm})$ and $\alpha_2(\omega_{480nm};\omega_{1440nm})$ when using K=3100cmGW^{-1}eV$^{2.5}$ (the experimental best fit) defined in the two-band model [3]. Due to the near-UV portion of the WLC being weak, the anomalous dispersion feature and negative n_2 could not be measured in ZnO. However, we show in 0.51mm thick ZnSe with λ_p=480nm (0.96E_{g_ZnSe}), $n_2(\omega_{480};\omega_{1440nm})$ is strongly negative due to the vicinity of the 1PA resonance, accompanied by ND-2PA, where the magnitudes are consistent with the theoretical values.

3. Conclusion

We measure the nondegenerate nonlinear refractive indices of ZnO and ZnSe using the beam deflection technique and find good agreement in both magnitude and spectral dependence with the predictions from nonlinear KK transformation. We therefore confirm the theory of Ref [4], allowing predictions of the operation of nonlinear optical devices engineered to use cross-phase modulation in direct-gap semiconductors.

4. References

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