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Determination of the third- and fifth-order nonlinear refractive indices in InN thin films

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We have combined the reflection and transmission Z-scan (RZ- and TZ-scans) techniques under femtosecond laser at 800 nm to extract both the third- and fifth-order nonlinear refractive indices (n_2 and n_4) in InN thin films. The observation of the nonlinear refractive index saturation in the intensity-dependent RZ-scan measurement indicates the existence of the fifth-order effect. By the aid of the TZ-scan, the fifth-order nonlinear effect has been enhanced by enlarging the cascaded contribution from the increased laser interaction length, where large n_2 of -2.5×10^{-11} cm^2/W and n_4 of 2.1×10^{-19} cm^4/W^2 have been determined. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2813637]

As a promising semiconductor among group III-nitride compounds, indium nitride (InN) has recently received intensive research interest. Even though more and more groups are involved in the study of the structural and optical properties of InN, its nonlinear optical parameters are still seldom investigated, while the nonlinear refractive index of other group III-nitride counterparts such as GaN (Refs. 1 and 2) and AlN (Ref. 3) has been studied intensively. The recent observations of the exceptional large optical bleaching effect and fast bleaching recovery time in InN epitaxial layers[4] indicate InN as a potential material for all-optical switching and related applications, which requires large optical nonlinearities and fast response speed. A deep understanding of the nonlinear optical parameters, especially the nonlinear refractive index, is therefore quite important for the optical communication applications of InN.

Among all techniques exploring the nonlinear optical parameters, the Z-scan measurement including the transmission Z-scan (TZ-scan)[5,6] and the reflection Z-scan (RZ-scan)[7,8] is the most popular method for determining the nonlinear refractive index of a large variety of materials due to its high sensitivity, simple experimental setup, easy alignment, and high accuracy of the obtained data. Both the sign and the magnitude of the nonlinear refractive index can be deduced by this powerful technique. There have also appeared many modifications and improvements of this technique[9–12] to increase the capabilities. Nevertheless, due to the lack of related theories, most of the investigations on nonlinear optical properties were restricted to the third-order nonlinearity, although both the third- and higher-order nonlinear contributions have already been involved in the Z-scan measurement.[13] In fact, the information of the higher-order nonlinear optical properties is of great fundamental and practical importance due to today's fast advance into the region of ultrashort femtosecond pulse duration. With the development in both theories and experiments, much attention has already been paid to the higher-order nonlinear properties of the materials such as semiconductor-doped glasses,[14] thin C_{60} films,[15] glasses,[16] and so on. Recently, based on a Gaussian decomposition method, theories which allow one to quantitatively study the fifth-order nonlinearity have been well developed.[17,18] In this letter, by performing the RZ- and TZ-scan measurements, we are able to observe and separate both the third- and fifth-order nonlinear effects in InN thin films, where the values of the third-order (n_2) and the fifth-order (n_4) nonlinear refractive indices have been quantitatively determined.

The studied 1.905-μm-thick InN thin film was grown on (0001) α-Al_2O_3 substrate by reactive radio frequency magnetron sputtering at the temperature of 100 °C.[19] During the deposition, the sputtering power of 100 W, sputtering pressure of 10 mTorr, gas flow of 3 SCCM (SCCM denotes cubic centimeter per minute at STP) and deposition time of 60 min were maintained. Both the x-ray diffraction and Raman scattering measurements demonstrate that the *c* axis of InN with a wurtzite structure is perpendicular to the substrate surface of (0001) α-Al_2O_3.[19] By the room-temperature optical transmission spectrum and the detailed calculation of the transmission profile,[20] we have yielded the linear refractive index ($n_0=2.548$) and linear absorption coefficient ($\alpha_0=557.8$ cm^{-1}) at the laser wavelength of 800 nm.

The Z-scan experimental setup was based on a Spectra-Physics mode-locked Ti:sapphire laser which produces laser pulses of about 100 fs duration with 82 MHz repetition rate at 800 nm wavelength. The measurements were performed using the standard RZ- (Refs. 7 and 8) and TZ-scan[5,6] techniques with a 10 cm focal-length objective lens. A low energy scan for the background signal was performed to eliminate the parasitic effects due to surface roughness or sample nonuniformity in order to accurately determine the nonlinearities in the studied sample.[5] The laser beam incident angle was 30° for the RZ-scan measurement while a small aperture with linear transmittance $S=0.3$ was used for the closed aperture TZ-scan measurement. From open aperture TZ-scan

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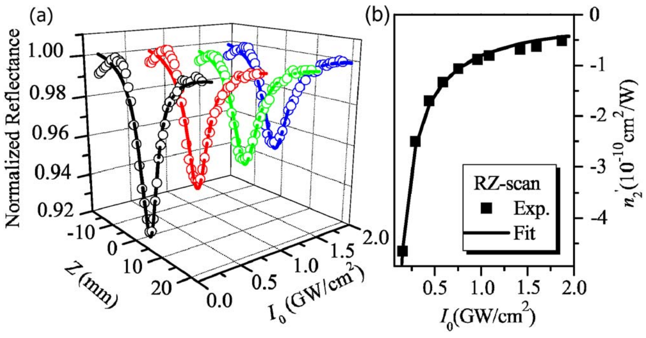

FIG. 1. (Color online) (a) Normalized reflectance and (b) laser intensity-dependent nonlinear refractive indices of InN thin film measured by the open aperture RZ-scan technique. The open circles and solid squares are experimental data while the solid curves are theoretical fitting results.

measurements, the nonlinear absorption coefficient of 2.3 $\times 10^{-9}$ cm/W has been obtained. Following the same procedure described in Ref. 21, we have excluded the possible contribution from the nonlinear absorption through dividing the close aperture results by the open aperture ones in the following calculation. We have also found that no signal can be detected in the sapphire substrate at the highest incident laser intensity, excluding the possible contribution of the sapphire substrate in the TZ-scan measurement. Following the idea in Ref. 22 to examine the thermal effects, we have observed the independence of the signals on different thermal properties of InN samples by different substrates (glass and α-Al_2O_3) in both RZ- and TZ-scans at every incident laser energy level, which indicates that the accumulative heating can be negligible.

The open circles in Fig. 1(a) show the typical results of the open aperture RZ-scan measurement at different incident laser intensities (I_0 from 0.15 to 1.87 GW/cm^2). Around the position of the beam waist ($Z=0$), large decreases of reflectance were observed in all the curves. This indicates that the nonlinear refraction was caused by the self-defocusing of the laser beam at the surface of the InN thin film. The open circles in Fig. 2(a) display the typical result of a closed aperture TZ-scan measurement at the incident laser intensity of 1.87 GW/cm^2. A prefocal transmittance minimum (valley) followed by a postfocal maximum (peak) was observed. TZ-scan curves at other incident laser intensities (I_0 from 0.15 to 1.87 GW/cm^2) show similar valley-peak signature. Compared with the third-order nonlinear effect, the fifth-order nonlinearity is usually weak and often neglected in the Z-scan measurement. However, the fifth-order nonlinear effect may enhance greatly for some materials in the ultrashort femtosecond pulse duration condition. In fact, without considering the contribution from the fifth-order nonlinearity of the InN thin film, the decreases of reflectance of our RZ-scan curves [Fig. 1(a)] indicate a negative sign of n_2,[8] while the valley-peak TZ-scan curve [Fig. 2(a)] represents a positive one.[5] We will demonstrate below that the discrepancy in these two measurements can be resolved into the different contributions of the fifth-order nonlinear effect in the InN thin film, since the laser interaction length with the InN thin film in our TZ-scan measurement is much longer than that in the RZ-scan one, and the fifth-order nonlinear effect is expected to be greatly enhanced in the TZ-scan curves.

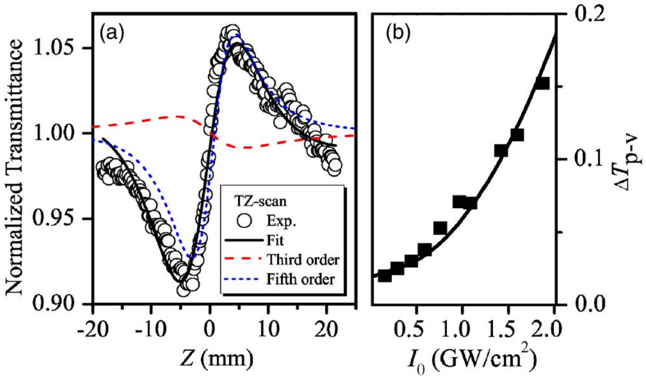

FIG. 2. (Color online) (a) Normalized transmittance and (b) laser intensity-dependent transmittance difference between the normalized peak and valley of the InN thin film obtained by closed aperture TZ-scan. The open circles and solid squares are experimental data. The curves are theoretical fitting results.

Due to the photoinduced modification in the reflection coefficient, a RZ-scan curve includes the information of all the nonlinear contribution. However, it is difficult to separate the third- and fifth-order nonlinear refractive indices from the RZ-scan results because of the lack of related theories. We have followed the same procedure described in Ref. 8 to obtain the nonlinear refractive index n_2' ($n_2'=n_2+n_4I_0$, including both the third- and fifth-order nonlinear contributions) since the influence of nonlinear absorption is negligible in the reflection measurement. The solid curves in Fig. 1(a) represent the theoretical calculation and it is obvious that the large decreases of reflectance become smaller at higher incident laser intensities. Detailed fitting results of the incident intensity dependence of n_2' are illustrated as solid squares in Fig. 1(b). The measured n_2' first increases rapidly at low incident irradiance (<1.0 GW/cm^2), and tends to be a constant as the laser intensity further increases, clearly revealing the saturation of the nonlinear refractive index in the InN thin film.

For saturable materials, the relationship between the nonlinear refractive index and incident intensity is governed by[23] $n_2'=n_{\text{low}}/(1+I_0/I_s)$, where I_s is the saturation intensity and n_{low} is the nonlinear refractive index in the limit of low intensity. A fairly good fitting of the experimental data [solid curve in Fig. 1(b)] yields the parameters of $I_s=3.0 \times 10^7$ W/cm^2 and $n_{\text{low}}=-2.8\times 10^{-9}$ cm^2/W. The saturation intensity I_s is almost one order smaller than that of GaN/AlN quantum wells ($I_s=5.0\times 10^8$ W/cm^2),[24,25] indicating that InN is a strong saturable nonlinear material. This argument is in good agreement with the recent observation of the large and fast recovering absorption bleaching effect in InN epitaxial layers,[4] since a material with saturation nonlinear refraction can also exhibit saturation absorption.[26] The saturable behavior of n_2' can be interpreted by the coexistence in the opposite signs of n_2 and n_4.[26] There are several different nonlinear processes contributing to n_4, which consist of a direct term n_4^{dir} inherent from $\chi^{(5)}$ of the fifth-order nonlinear susceptibility and a cascaded term n_4^{casc} proportional to $[\chi^{(3)}]^2$: $n_4=n_4^{\text{dir}}+n_4^{\text{casc}}$.[27] A possible mechanism resulting to the cascaded nonlinearity is the interference of the incident wave and the wave generated by a slightly phase mismatched process of third-harmonic generation, as described in Ref. 28. Since n_4^{casc} can be enlarged by increasing the laser interaction length of the nonlinear medium,[28] the fifth-order nonlinear effect can be more greatly enhanced in the TZ-scan than in RZ-scan measurement. So the negative n_2' reveals that negative n_2 dominates in the RZ-scan measurement while the saturable behavior of n_2' (i.e., the decrease of the absolute value of n_2') with the increased incident laser intensity comes from the contribution of positive n_4.

In the case when n_2 and n_4 have the opposite signs, a simple peak-valley TZ-scan trace will be formed by a negative n_2 if the contribution of the fifth-order nonlinear effect is not large enough. Nevertheless, when the contribution of the fifth-order nonlinear effect is enhanced, it is even possible to obtain a valley-peak configuration, as observed in Fig. 2(a), due to the combination effect of negative n_2 and positive n_4. From the TZ-scan theory based on a Gaussian decomposition method,[18] a pure normalized transmittance related to $(2n+1)$th order nonlinearity has the following form:

$$T_n(x,\Phi_{0n}) = \lim_{M\to\infty}\left|\sum_{m=0}^{M}\frac{1}{m!}\left[\frac{\Phi_{0n}}{(1+x^2)^n}\right]^m\frac{i^m(x+i)}{x+i(2mn+1)}\right|^2, \qquad (1)$$

where Φ_{0n} is the $(2n+1)$th order on-axis nonlinear refraction phase shift and $x=Z/Z_0$ is the Z position divided by waist radius. The quantitative determination of n_2 and n_4 from our TZ-scan curves can be realized via the normalized transmittance with simultaneous third- and fifth-order nonlinear refractions

$$T(x,\Phi_{01},\Phi_{02}) = -1 + T_1(x,\Phi_{01}) + T_2(x,\Phi_{02}) + \frac{48\Phi_{01}\Phi_{02}(x^4+14x^2-35)}{(x^2+1)^3(x^2+9)(x^2+25)(x^2+49)}. \qquad (2)$$

Figure 2(a) depicts the detailed theoretical fitting (solid curve) considering both the third- and fifth-order effects, from which we can obtain the parameters of $n_2=-2.5\times10^{-11}$ cm^2/W and $n_4=2.1\times10^{-19}$ cm^4/W^2. Theoretical fittings of the TZ-scan curves at other incident laser intensities show similar results with the deviation of less than 10% for both n_2 and n_4 due to the instability of the incident laser intensity. For comparison, the extracted value of n_2 is of the same negative sign but two order magnitude larger than that of the GaN thin film at the same laser wavelength of 800 nm ($n_2=-7.3\times10^{-14}$ cm^2/W).[29] Separated contribution of pure third- and fifth-order nonlinear effects has also been calculated by Eq. (1) using the above n_2 and n_4 values, as illustrated in Fig. 2(a) with dashed and dotted curves, respectively. It is clear that the third- and fifth-order contributions behave totally reverse valley and peak configuration and the fifth-order contribution is much larger than that of the third-order one as a larger n_4^{casc} is expected in the TZ-scan measurement of InN.

TZ-scan measurement carried out at different incident laser intensities can further give us an insight into the detailed nonlinearities of a material, since I_0 dependence of $\Delta T_{p\text{-}v}$ [see Fig. 2(b)] of the InN thin film allows one to directly judge the presence or absence of the fifth-order nonlinearity contribution. Here, we define $\Delta T_{p\text{-}v}$ as the difference between the normalized peak and valley transmittance: T_p-T_v. With regard to the contribution from the third- and fifth-order nonlinearities, $\Delta T_{p\text{-}v}$ as a function of I_0 for the given order of nonlinearity is proportional to $kL_{\mathrm{eff}}n_2I_0$ and $kL'_{\mathrm{eff}}n_4I_0^2$, respectively,[5] where k is the wave vector of the laser beam, $L_{\mathrm{eff}}=[1-\exp(-\alpha_0L)]/\alpha_0$, $L'_{\mathrm{eff}}=[1-\exp(-2\alpha_0L)]/2\alpha_0$, and L is the sample thickness. In our intensity-dependent TZ-scan measurement, instead of growing linearly with I_0, $\Delta T_{p\text{-}v}$ behaves quadratic with I_0, which clearly reveals the presence of the fifth-order nonlinearity in the InN thin film. From fitting the experimental data with the obtained n_2 and α_0 values [solid curve in Fig. 2(b)], we can get $n_4=1.4\times10^{-19}$ cm^4/W^2, which has the same magnitude as from the above TZ-scan curves. We can therefore confirm the enhancement of the fifth-order nonlinear contribution in InN thin film under the TZ-scan measurement, with the positive n_4 resulting in the valley-peak TZ-scan signature in Fig. 2(a).

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