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PHOTOINDUCED LASER EFFECTS IN INDIUM NITRIDE FILM

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The optical nonlinear absorption properties in sputtering Indium nitride (InN) film were investigated under the excitations of nanosecond, picosecond and femtosecond pulsed lasers by open-aperture transmission Z-scan technique (TZ-scan). Under the condition of $h\nu > E_g$, the saturable absorption (SA) phenomena induced by one-photon transition were observed in both nanosecond and picosencond pulsed TZ-scan measurements. When $2h\nu > E_g > h\nu$, the film presented SA due to the two-photon transition under the excitation of picosecond laser. However, at femtosecond 800 nm, the film showed the two-photon absorption (TPA) instead of SA, and the TPA coefficient tended to a saturable value as the excitation intensity increased. The results indicate that the InN film is a kind of good saturable absorber.

Keywords: III–V and II–VI semiconductors; nonlinear optics; Z-scan.

1. Introduction

As a promising semiconductor among III-nitride compounds, Indium nitrides (InN) optical nonlinear properties have received intensive research interests. For instance, a large optical bleaching effect[1] in the epitaxial InN layers was observed under the excitation of picosecond pulsed laser. And short carrier lifetimes[1] were also obtained through pump-probe experiment. In addition, the femtosecond pulsed transmission Z-scan (TZ-scan[2–4]) measurements of the saturable absorption (SA) of InN epitaxial films in the range of 720–790 nm were reported.[5] Our group has reported both the third- and fifth-order nonlinear refractive indices in sputtering InN film.[6] These results indicate InN film is a kind of promising material in applications. But different processes responsible for the optical nonlinear absorption (NLA) in different time domains are still seldom investigated. In fact, the information of different nonlinear optical processes is of great fundamental and practical importance in applications. For instance, for optical limiting devices, it may not be merely enough to have high value of nonlinear refractive index or high value of NLA, but the response times of these processes are equally important.[7]

Therefore, in order to deploy the NLA behaviors of InN films into applications in optical communication, it is especially important to study different nonlinear optical processes of InN films.

In this research, we used the TZ-scan technique to investigate the optical NLA in sputtering InN film with ns, ps and fs lasers, respectively. The NLA coefficients under different conditions were obtained. In addition, a great attention was devoted to explain the mechanisms involved in the nonlinear responses. The results obtained in this study underpin the future development of nonlinear optical devices (such as optical switching, optical bistability[8,9] and optical limiters[10,11]) based on InN films.

2. Experimental and Sample

The investigated 1.905 μm-thick InN film was grown on (0001) α-Al_2O_3 substrate by reactive radio frequency magnetron sputtering at 100°. During deposition, the sputtering power of 100 W, sputtering pressure of 10 mTorr, gas flow of 3 SCCM (SCCM denotes cubic centimeter per minute) 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. The film is anisotropic and the density of defects is about in the order of 10^{19} cm^{-3}. By room temperature, the optical absorption edge of 1.97 eV has been obtained.

The optical NLA measurements were performed using a standard open-aperture TZ-scan technique. Nanosecond Q-switched Nd:YLF laser (6 ns, 10 Hz, 532 nm), picosecond mode-locked Nd:YAG laser (38 ps, 10 Hz, 532 nm and 1064 nm), femtosecond mode-locked Ti:sapphire laser (100 fs, 82 MHz, 800 nm) were used as excitation source. And in the fs TZ-scan measurement the beam was chopped at 1000 Hz. Moreover, a low intensity scan for the background signal was performed to eliminate the parasitic effects due to the surface roughness or sample nonuniformity in order to accurately determine the nonlinearities in the studied sample. We have also found that no signal can be detected in the sapphire substrate. And all the measurements were performed at room temperature.

3. Results and Discussion

Figure 1 illustrates the results of open-aperture TZ-scan for InN film measured under excitations of 6 ns and 38 ps pulsed 532 nm lasers. The incident laser intensities I_0 were 48.05 MW/cm^2 and 0.93 GW/cm^2, respectively. Under ns and ps 532 nm excitations, the open-aperture TZ-scan transmittance is symmetric with respect to the focus ($z = 0$), where it has a maximum transmission.

For absorptive samples, according to Beer's law, the optical intensity I of the optical wave per unit propagation distance is given by

$$\frac{dI}{dz} = -\alpha I\,. \tag{1}$$

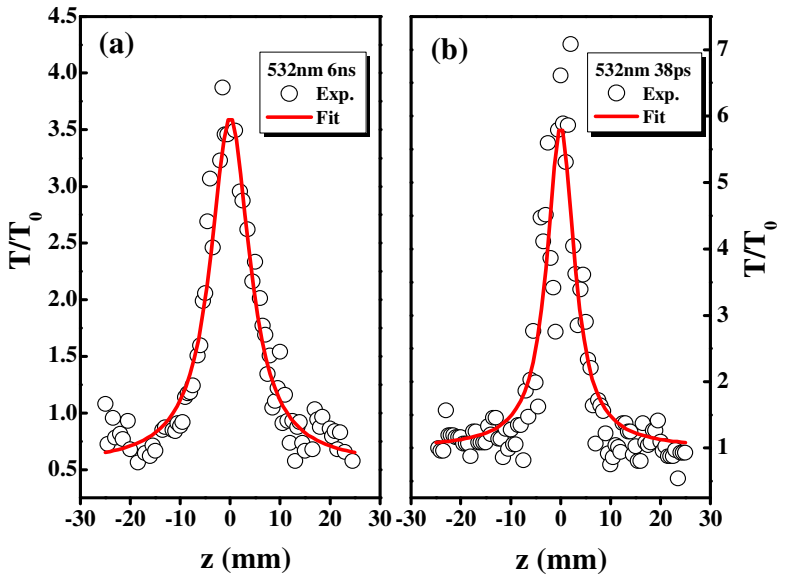

Fig. 1. Results of open-aperture TZ-scan measured under 532 nm by (a) 6 nanosecond pulsed laser and (b) 38 picosecond pulsed laser.

Considering the NLA in semiconductors, we have[12]:

$$\alpha = \alpha_0 + \beta_{\mathrm{TA}} I + \sigma_\alpha \Delta N \,, \tag{2}$$

where α_0 is the linear absorption coefficient, β_{TA} is the TPA coefficient, σ_α is the free carrier absorption (FCA) cross-section and ΔN is the free carrier density which is given by the following equation under the condition of $h\nu > E_g$:

$$\frac{d\Delta N}{dt} = \frac{\alpha_0 I_0}{h\nu} - \frac{\Delta N}{\tau_c} \,, \tag{3}$$

where I_0 is the incident intensity, $h\nu$ is the photon energy and τ_c is the photoexcited free carrier recombination time. In this experiment, one-photon transition occurred as the one photon energy of 532 nm laser (2.33 eV) is larger than the optical absorption edge of the investigated sample. It is known that the free carrier nonlinearities with slow decay time are significant for laser pulses of nanosecond duration and longer.[13,14] But in this ns pulsed measurement, we did not observe FCA which should show a valley in the curve. Maybe it can be attributed to the free carrier density ΔN and FCA cross-section σ_α in InN film are not large enough to generate FCA.

As we know, when $h\nu > E_g$, SA due to band-filling mechanism plays an important role. In our case, the excitation wavelength of 532 nm (2.33 eV) is in the non-resonant absorption region of the investigated sample (1.97 eV). In the open-aperture TZ-scan measurements, the 532 nm laser radiation results in bleaching of acceptor levels, yielding SA. This kind of SA induced by band-filling

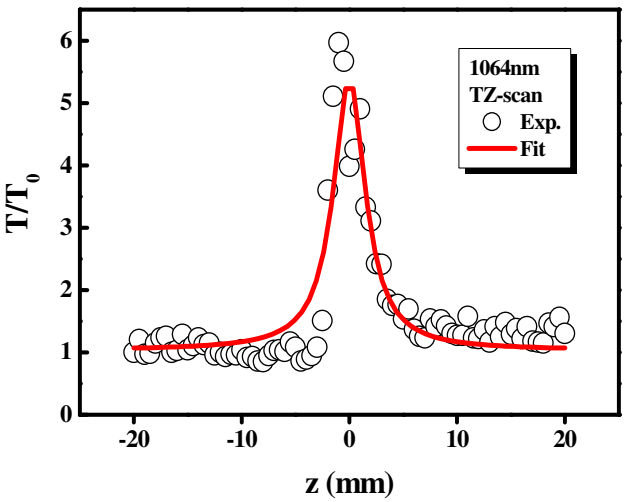

Fig. 2. Result of open-aperture TZ-scan measured by 38 picosecond pulsed laser at 1064 nm.

mechanism in epitaxial InN layers has been observed by Pačebutas *et al.* (2006) and Tsai *et al.* (2009), respectively. Our results confirmed it again.

To obtain the SA coefficient β, we calculate it from the decrease of the transmittance (T_i) versus input irradiance according to[15]:

$$T_{OA}(z) = \sum_{m=0}^{\infty} \frac{(-1)^m q_0^m}{(m+1)^{3/2}}\,, \tag{4}$$

where $q_0 = \beta I_0 L_{\text{eff}}/(1+z_2/z_0^2)$, in which L_{eff} ($= [1-\exp(-\alpha_0 L)]/\alpha_0$) is the effective path-length with α_0 being the linear absorption coefficient, L being the thickness of the sample; $z_0 = k\omega_0/2$ is the diffraction length of the beam, ω_0 is the beam waist radius at the focus point and $k = 2\pi/\lambda$ is the wave vector. With the linear absorption coefficient value of 6.7×10^4 cm^{-1} at 532 nm, the best fittings give the coefficient β in ns and ps cases as -1.03×10^{-3} cm/W and -0.83×10^3 cm/GW, respectively. It is found the value of β in ns case is three-order larger than in ps case. This is due to the fact that the free carrier density in conduction band is mainly dependent on laser fluence. More free carriers were generated in the ns pulsed TZ-scan measurement because of the larger ns laser fluence (1.99×10^{-1} J/cm^2) than that of ps (3.53×10^{-2} J/cm^2), which makes the sample more transparent in ns case. So we observed stronger SA effect in ns pulsed TZ-scan measurement. Similar phenomena[16] were also reported in other materials.

Previously, the SA phenomena induced by one-photon transition in InN film were observed under the condition of $h\nu > E_g$. To study InN film more deeply, we performed TZ-scan measurements under the condition of $2h\nu > E_g > h\nu$. The result of 38 ps pulsed measurement is shown in Fig. 2 in which the wavelength and incident intensity were 1064 nm and 1.14 GW/cm^2, respectively. The trace suggests

under the excitation of picosecond 1064 nm laser strong SA effect still occurred in InN film.

Under the condition of $2h\nu > E_g > h\nu$, the free carrier density could be expressed as[17]:

$$\frac{d\Delta N}{dt} = \frac{\beta_{\mathrm{TA}} I_0^2}{2h\nu} - \frac{\Delta N}{\tau_c}. \tag{5}$$

The first and second term of Eq. (5) represent the generation and recombination rate of free carrier, respectively. As we know, in the transparency region ($2h\nu > E_g > h\nu$) of semiconductors the nonlinear responses can be attributed to the contributions of the intrinsic optical nonlinearities arising from TPA and the free carrier nonlinearities[18] due to the free carriers generated by TPA. However, neither TPA nor FCA effect was observed in this measurement. It is due to much larger the generation rate of photoexcited free carrier than the recombination rate. So a mass of photoexcited free carriers induced by TPA occupy the holes in conduction band quickly, leading to band-filling effect directly which resulted in the SA under the excitation of picosecond 1064 nm. At last, the fitting gives the SA coefficient β as -11.6×10^2 cm/GW.

Figure 3(a) shows the typical curve of TZ-scan measurements for InN film by 100 fs pulsed laser at 800 nm with the incident intensity of 7.73 GW/cm^2. We

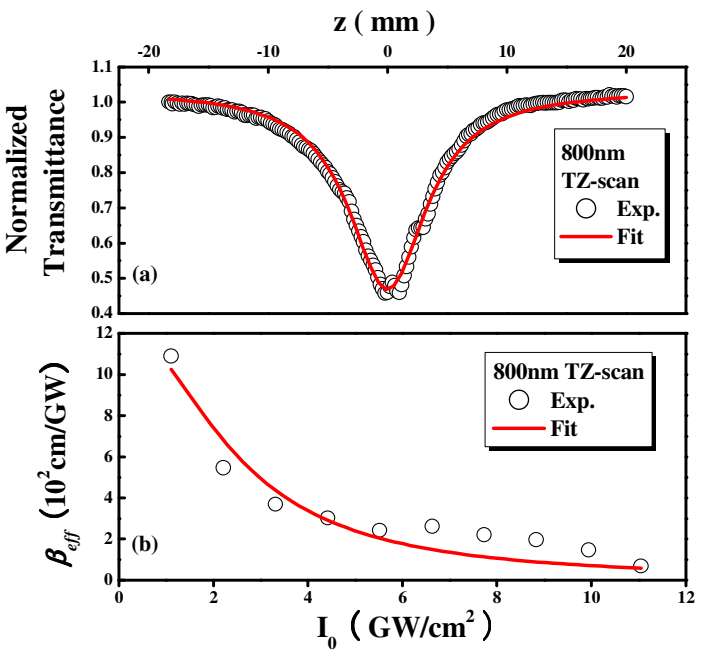

Fig. 3. (a) Open-aperture TZ-scan for InN film measured by femtosecond pulsed laser at 800 nm. (b) Laser intensity dependent nonlinear absorption coefficient of InN thin film measured by the femtosecond open-aperture TZ-scan technique.

can see that the result is absolutely different from that of ns and ps. In the curve around the position of the beam waist ($z = 0$) a large decrease of transmittance was observed, which indicates a positive NLA effect.

As mentioned above, when $2h\nu > E_g > h\nu$, both TPA and FCA should be taken into consideration. However, the fluence of fs laser was much smaller than that of ns and ps lasers. In both ns and ps pulsed TZ-scan measurements FCA did not occur, not to mention the fs case. So FCA can be ignored in fs case. Moreover, it is well known that the intrinsic nonlinearities which have fast response are important in the femtosecond time domain.[14] So in the fs pulsed TZ-scan measurements at 800 nm, it is considered that the positive NLA is mainly contributed by TPA. The NLA coefficient at 7.73 GW/cm^2 was obtained as 2.204×10^2 cm/GW which indicates there is a strong TPA effect in InN film.

In order to find out if SA would occur in fs pulsed TZ-scan measurements, we carried out similar TZ-scans and fittings at the incident intensities ranging from 1.11 GW/cm^2 to 11.05 GW/cm^2. All the curves show similar signatures as Fig. 3(a). A plot of β versus I_0 is shown in Fig. 3(b). All the positive values of β indicate that SA was not observed directly in these measurements. But it is found that β decreases with the I_0 increasing. If the NLA was dominated by a pure TPA process,[15] the values of would be independent on I_0. Therefore, the decrease of β indicates that another negative NLA effect besides the TPA occurred as the intensity increased. We consider it is attributed to that the duration of laser pulse (100 fs) is much shorter than electron-hole (e-h) recombination time,[19] based on which the second term of Eq. (5) can be ignored. So the generation rate of photoexcited free carrier will be directly proportional to the incident laser intensity. With the incident intensity I_0 increasing, more and more free carriers aggregate in the conduction band, giving rise to saturation trend in the absorption. Similar conclusions were reached in other materials, such as bulk silicon[12] and silicon nanocrystals.[20]

For saturable materials, the NLA is expressed in terms of the saturation intensity I_s as:

$$\beta = \frac{\beta_0}{1 + \left(\dfrac{I_0}{I_s}\right)}^2. \tag{6}$$

A fit of data reported in Fig. 3(b) by Eq. (6) allows extracting $\beta_0 = 12.3 \times 10^2$ cm/GW and $I_s = 2.46$ GW/cm^2.

On the whole, in our study, the condition $h\nu > E_g$ and $2h\nu > E_g > h\nu$ indicated that there would mainly exist one-photon and two-photon excitations. Compared to the former two excitations, the probability that multi-photon excitations occurred would be much less. As we know, electron-phonon interaction also contributes to the bleaching effect. Usually, the weaker the electron-phonon interaction is, the stronger the bleaching effect would be. So in our results, the electron-phonon interaction is also one factor of the strong bleaching effects in ns and ps measurements which

are contrary to the fs. In addition, the defect states[21] may also contribute to the third-order susceptibility in InN film.

4. Conclusion

In conclusion, we have studied the NLA properties in InN film by ns, ps and fs pulsed TZ-scan measurements. Different nonlinear absorption mechanisms occurred under different laser conditions. In 6 ns and 38 ps pulsed TZ-scan measurements at 532 nm, SA effects induced by one-photon transition were observed directly. And the film showed SA effect induced by two-photon transition in 38 ps pulsed TZ-scan measurement at 1064 nm. In 100 fs pulsed TZ-scan measurements at 800 nm, TPA was the dominated process at low optical intensity. However, as the incident laser intensity increased, the TPA coefficient trended to saturable value. The strong SA properties measured in ps and ns lasers suggest that it may be a material with potential application in laser pulse compression, while the TPA presented in fs laser underpins the future development in the optical limiting.

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