1-1-1994

Limitation Due To 3-Photon Absorption On The Useful Spectral Range For Nonlinear Optics In Algaas Below Half Band-Gap

Jin U. Kang
University of Central Florida

Alain Villeneuve
University of Central Florida

Mansoor Sheik-Bahae
University of Central Florida

George I. Stegeman
University of Central Florida

Kadhair Al-hemyari

Limitation due to three-photon absorption on the useful spectral range for nonlinear optics in AlGaAs below half band gap

Cite as: Appl. Phys. Lett. **65**, 147 (1994); https://doi.org/10.1063/1.112654
Submitted: 07 October 1993 . Accepted: 29 April 1994 . Published Online: 04 June 1998

Jin U. Kang, Alain Villeneuve, Mansoor Sheik-Bahae, George I. Stegeman, Kadhair Al-hemyari, J. Stewart Aitchison, and Charles N. Ironside

Limitation due to three-photon absorption on the useful spectral range for nonlinear optics in AlGaAs below half band gap

Jin U. Kang, Alain Villeneuve, Mansoor Sheik-Bahae, and George I. Stegeman
CREOL, University of Central Florida, 12424 Research Parkway, Orlando, Florida 32826

Kadhair Al-hemyari, J. Stewart Aitchison, and Charles N. Ironside
Department of Electronics and Electrical Engineering, University of Glasgow, Glasgow, G12 8QQ, United Kingdom

(Received 7 October 1993; accepted for publication 29 April 1994)

We report measurements of the spectral dispersion and the magnitude of three-photon absorption in $Al_{0.18}Ga_{0.82}As$ for photon energies between one half and one third the band gap and show that a two-parabolic-band model is valid in this material. These results indicate that there is a limited spectral range below half the band gap in AlGaAs (and presumably all semiconductors) in which the bound electronic optical nonlinearity can be used without significant multiphoton absorption.

One of the hardest aspects of nonlinear optics is finding a suitable nonlinear material for various applications. For example, we have been interested in all-optical waveguide switching devices which require solid-state materials that exhibit a large, ultrafast third-order nonlinear response with little or no absorption at the communication wavelengths of 1.3 and 1.55 μm. There are many materials with a large third-order nonlinear response but the major problem has been excessive absorption. The total absorption can have a number of contributions so that $\alpha=\alpha_1+\alpha_2 I=\alpha_3 I^2+...$, where α_2 and α_3 are the two- (2PA) and three- (3PA) photon absorption coefficients, respectively, and I is the intensity. For example, 2PA in semiconductors between the band gap and half the band gap is known to be detrimental to all-optical switching.[1] Below half band gap the 2PA coefficient decreases rapidly with increasing wavelength so that the two-photon figure of merit $T<1$, as required.[1,2] AlGaAs, when used with photons of energy just less than half the band-gap energy, was found to be an ideal material for all-optical switching, leading recently to subpicosecond waveguide switching with 65-pJ energies.[3,4] However, when an AlGaAs device is operated at wavelengths too far below half the band gap, 3PA appears to become a limiting factor for all-optical switching.[5]

There are a number of detailed calculations which have been made on multiphoton absorption in different materials based on models of varying sophistication.[6,7] There is now a wealth of 2PA data available for comparison with theory.[8] In particular, a simple analysis using a two-parabolic-band model by Wherrett has provided a reasonable prediction of 2PA coefficients for semiconductor materials, including AlGaAs.[2,6] For 3PA, experimental data is very limited. No spectral dispersion has been reported and agreement with existing theories is unclear. For AlGaAs in particular, a single value for α_3 has been published near 1.67 μm, which agrees within a factor of two with theory.[3] In this letter, we report measurements of the spectral dispersion and magnitude of α_3 below half the band gap, show that it agrees well with Wherrett's two-parabolic-band model, and evaluate the spectral range over which AlGaAs can be used for efficient nonlinear optics.

The 3PA coefficient can be calculated from Eq. (1) in Ref. 6:

$$\alpha_3=\frac{3^{10}\sqrt{2}}{8}\pi^2\left(\frac{e^2}{\hbar c}\right)^3\frac{\hbar^2P^3}{n^3E_g^7}\left[\frac{(3\hbar\omega/E_g-1)^{1/2}}{(3\hbar\omega/E_g)^9}\right], \quad (1)$$

where P is the Kane parameter, E_g the band-gap energy, $\hbar\omega$ the energy of the photon, n is the refractive index of the material, and $e^2/\hbar c$ the fine-structure constant. For these calculations the wavelength dispersion of the linear refractive index (<3% over the wavelengths investigated) was neglected and a calculated value at $\lambda=1.55$ μm of 3.34 for n was used for 18%, 19%, and 20% aluminum content (band gaps of 1.648, 1.6609, and 1.66436 eV, respectively).[9] For these aluminum concentrations, one half (one third) the band gap occurs around 1.5 μm (2.25 μm).

Figure 1 shows several plots of the 3PA coefficient α_3 as a function of wavelength for 18%, 19%, and 20% aluminum content. From the graphs, it can be clearly seen that the 3PA coefficient peaks at wavelengths just before one third the band gap and then decreases quickly at longer wavelengths.

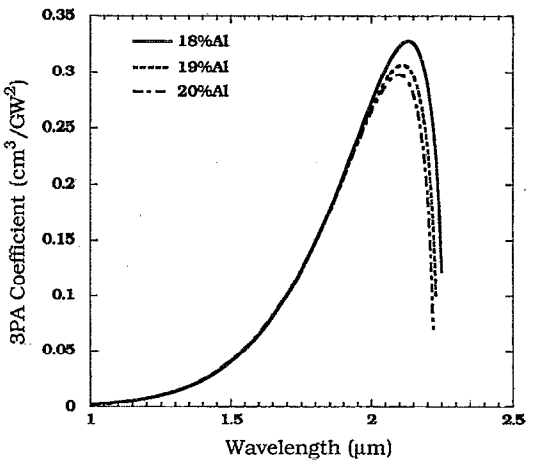

FIG. 1. Wavelength dependence of the three-photon absorption coefficient for three different aluminum contents based on the two-parabolic-band model: the solid, short dashed, and long dashed lines are for 18%, 19%, and 20% aluminum, respectively.

 0003-6951/94/65(2)/147/3/$6.00

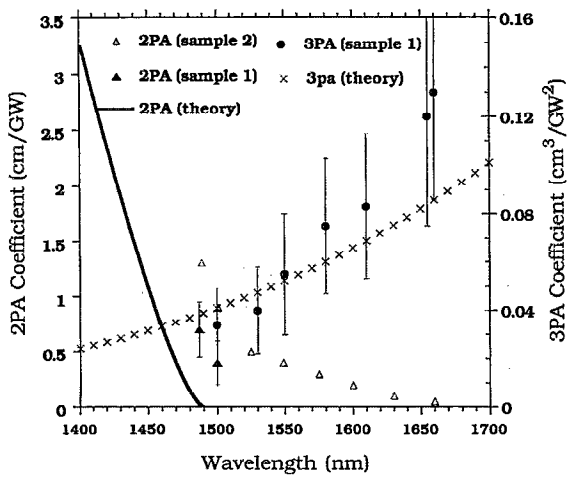

FIG. 2. Measurement and calculated two- (α_2) and three-photon (α_3) absorption coefficients are shown by dots, whereas the theoretical values are shown by cross symbols. The measured values of the two-photon absorption coefficients are shown in two different samples, sample one by filled triangles and sample two by open triangles. The theoretical values of two-photon absorption coefficients are shown by a solid line.

This leads to a contribution to the absorption coefficient of 0.3 cm^{-1} at the peak, and 0.1 cm^{-1} at 1.7 μm for an intensity of 1 GW/cm^2. Since most all-optical waveguide switching devices shorter than 1 cm long require switching intensities of several GW/cm^2, and because 3PA scales as intensity squared, the resulting losses can pose a significant problem for all-optical switching devices below half band gap, especially as one approaches one third the band gap.

Unambiguous 3PA coefficients can be measured by operating below half the band gap and by using high enough intensities so that the 2PA contribution ($\propto I$) is negligible relative to the three-photon component ($\propto I^2$). This was true in our case for the operating wavelength longer than 1.53 μm where the 2PA coefficient is negligible as can be deduced from Fig. 2. The measurement was performed with a NaCl color center laser operating from 1.48 to 1.66 μm with additive pulse mode locking to produce 400–1100 fs pulses with a peak power in excess of 1 kW. In the experiment, in order to minimize the effect of group-velocity dispersion, relatively longer 900–1000 fs pulses were used. A 2.2-cm-long strip-loaded AlGaAs waveguide was used where the guiding region is made of $Al_{0.18}Ga_{0.82}As$ and the cladding region of $Al_{0.24}Ga_{0.76}As$. The effective area of the waveguide was calculated to be 12 μm^2 at 1.5 μm, and 13 μm^2 at 1.66 μm. The linear loss of the waveguide was measured to be 0.1 cm^{-1}. An inverse transmission technique, similar to that used to measure α_2, was used to determine the α_3 coefficient for the operating wavelength above 1.53 μm.[10] For the operating wavelength below 1.53 where the 2PA could not be neglected, a beam propagation method (BPM) was used to fit the data. The BPM code was written based on the equation

$$\frac{dI}{dz} = -\alpha_1 I - \alpha_2 I^2 - \alpha_3 I^3.$$

In the code, an incident $\mathrm{sech}^2(t)$ temporal pulse shape was assumed. Free-carrier absorption generated by 2PA or 3PA was neglected in the analysis based on previous pump-probe experiments on similar samples, and a simple calculation which shows that the maximum carrier density is less than $10^{16}/cm^3$.[4] The wavelength dependence of α_3 was measured over the range 1.50 μm→1.66 μm and is shown in Fig. 2. The good agreement between experiment and theory indicates that the two-parabolic-band model should be useful for predicting α_3 in the $Al_xGa_{1-x}As$ system for photon energies between one half and one third the band gap. For example, at λ=1.55 μm, after including all of the corrections, we obtained a value of $\alpha_3 = 5 \pm 2 \times 10^{-20}$ cm^3/W^2. The corresponding theoretical value of 5.4×10^{-20} cm^3/W^2 was obtained for α_3. This number is also in good agreement with a value (5.5×10^{-20} cm^3/W^2) deduced at 1.55 μm from fitting the response of a nonlinear directional coupler at this wavelength.[5]

In Fig. 2, the dispersion of α_2 is also shown, theoretically (Ref. 11) by a solid line and experimentally for two different samples by open and filled triangle symbols, respectively. Note that the experimental values of α_2 are different for the two samples with the same aluminum concentration. They are also different from the theoretical values where the theoretical values for α_2 are zero below half the bandgap. This additional two-photon absorption is a consequence of the two-photon equivalent of the Urbach tail, and defect states which are not accounted for in the two-parabolic-band model.[12] It is clear from the figure that sample 1 is superior in quality to sample 2 for which α_2 extends far below half the band gap. Thus, the width of the useful spectral region also depends on the quality of the sample and the theoretical values represent the best useful spectral region one can hope to achieve.

The effects of multiphoton absorption on all-optical switching are now well known.[1,5] In order to quantify the effects of nonlinear absorption, the two figures of merit $T = 2\alpha_2 \lambda_{vac}/n_2$ and $V = \alpha_3 \lambda_{vac} I/n_2$ have been formulated where I is the intensity needed to produce a desired nonlinear effect.[1,13] For example, for a NLDC the relevant intensity is $I_c = \lambda_{vac}/n_2 L_c$ where L_c is the one half beat length of the device. When $T>1$ or/and $V>0.68$, multiphoton absorption degrades seriously the ideal response of a NLDC which requires a nonlinear phase shift of 2π for switching.[5] Although other switching devices or nonlinear interactions are limited by different maximum values for V and T, the required figures of merit all fall within a factor of 4 of unity. That is, the figures of merit V and T are generally applicable in nonlinear optics for choosing appropriate materials.

Just as multiphoton transitions contribute to the total absorption, they also contribute to the intensity-dependent refractive index. The total refractive index can be written as $n = n_0 + n_2 I + n_4 I^2 + \ldots$, where the major contribution of n_4 comes from virtual three-photon transitions. An estimation of the n_4 coefficient associated with the three-photon absorption can be obtained using a nonlinear Kramers–Kronig (KK) transformation of α_3. In principle, this requires a nondegenerate α_3 coefficient. However, similar to the procedure given in Ref. 14 for the two-photon absorption process, we approximated the nondegenerate α_3 with the degenerate one

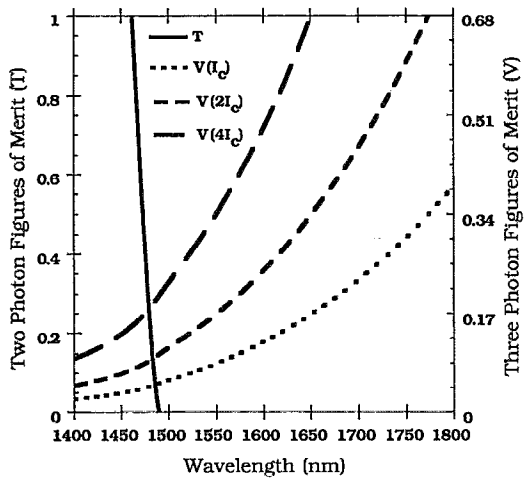

FIG. 3. Wavelength dependence of the figures of merit, T and V. The solid line represents the two-photon figure of merit T based on the two-parabolic-band model in Ref. 11. The dotted, short dashed, and long dashed lines represent theoretical values of V for 19% aluminum content and 2, 4, and 8π nonlinear phase shifts, respectively.

[Eq. (1)] and performed the KK transformation to obtain a crude estimate of n_4. For $Al_xGa_{1-x}As$ with $x=0.18\rightarrow0.20$, the values of n_4 between 1.5 and 1.8 μm were estimated to be of order 6×10^{-25} cm^4/W^2. The third-order nonlinear refractive index $n_2>0.5\times10^{-13}$ cm^2/W right up to 1.8 μm. Therefore, the relative contribution of the three-photon transition to the nonlinear refractive index change, n_4I^2/n_2I, varies linearly with the intensity, and is always less than a few percent at 1 GW/cm^2. For the typical switching intensities encountered in a NLDC, the induced fifth-order refractive index change was less than one order of magnitude smaller than the third-order nonlinear refractive index change. This remains true for wavelengths up to 1.8 μm, the maximum considered here. Therefore the dominant source of $\Delta n(I)$ comes from virtual two-photon transitions and we include only n_2 effects in our definition and calculations of V.[2,5] Note, however, that for wavelengths near the peak of the three-photon absorption coefficient, index changes due to n_4I^2 cannot be neglected at >10 GW/cm^2 intensities.

We now use calculations of n_2, α_2, and α_3 to calculate T and V, and hence identify the largest spectral window possible within which $T<1$ and $V<0.68$ for $Al_{0.18}Ga_{0.82}As$.[2,11] Figure 3 shows the calculated values of T and V as a function of wavelength for photon energies around one half the band gap. The solid line represents the values of T as a function of wavelength. Dotted, short dashed, and long dashed lines represent the values of V for nonlinear phase shifts of 2, 4, and 8π, respectively, accumulated in the absence of all losses over a distance equal to one absorption length. For example, if a phase shift of 4π is needed for a nonlinear optical interaction, then the available spectral range for which $T<1$ and $V<0.68$ is limited to 1.46 μm$<\lambda<$1.77 μm. Therefore, the useful spectral range is determined by the individual requirements of each nonlinear interaction, as quantified by the required nonlinear phase shift.

We now specialize the discussion to all-optical waveguide switching devices such as the nonlinear directional coupler. The reported critical intensity (which requires a 2π phase shift) of a 2.2-cm-long device in $Al_{0.18}Ga_{0.18}As$ at 1.55 μm is approximately 0.7 GW/cm^4. The corresponding experimental value of V if 0.07 (at $I=I_c$), which indicates that this material is suitable for all-optical switching. However, switching with good contrast requires $4I_c$, i.e., nonlinear phase shifts of 8π. This limits the spectral range to only 1.46 μm$<\lambda<$1.65 μm for efficient switching of NLDCs. Clearly this material may not be suitable for all-optical switching if one deviates too much from just below half the band gap.

In conclusion, using the two-parabolic-band model, we have calculated the wavelength dispersion of three-photon absorption and have shown that three-photon absorption limits the spectral range over which AlGaAs is useful for nonlinear optics below one half of its band gap. Our measurements of the three-photon absorption coefficients of an $Al_{0.18}Ga_{0.82}As$ channel waveguide agreed with the theoretical values, showing that the two-parabolic-band model works well for this material.

This work was supported in the U.K. by SERC through the nonlinear optics initiative and the work at CREOL was sponsored by NSF (ECS-9203877).

[1] K. DeLong, G. I. Stegeman, Appl. Phys. Lett. **57**, 2063 (1990); J. S. Aitchison, M. K. Oliver, E. Kapon, E. Colas, and P. W. E. Smith, Appl. Phys. Lett. **56**, 1305 (1990).
[2] A. Villeneuve, C. C. Yang, G. I. Stegeman, C. H. Lin, and H. H. Lin, Appl. Phys. Lett. **62**, 2465 (1993).
[3] M. N. Islam, C. E. Soccolich, R. E. Slusher, A. F. J. Levi, W. S. Hobson, and M. G. Young, J. Appl. Phys. **71**, 1927 (1992).
[4] K. Al-hemyari, A. Villeneuve, J. U. Kang, J. S. Aitchison, C. N. Ironside, and G. I. Stegeman, Appl. Phys. Lett. **63**, 3562 (1993).
[5] C. C. Yang, A. Villeneuve, G. I. Stegeman, and J. S. Aitchison, Opt. Lett. **17**, 710 (1992).
[6] B. S. Wherrett, Opt. Soc. Am. B **1**, 67 (1984).
[7] J. H. Lee, Phys. Rev. B **5**, 449 (1972).
[8] E. W. Van Stryland and L. L. Chase (in press).
[9] S. Adachi, Phys. Rev. B **38**, 345 (1988).
[10] E. W. Van Stryland, M. A. Woodall, W. E. Williams, and M. J. Soileau, in the Symposium on Laser Induced Damage, Boulder, Colorado, 1981 (unpublished).
[11] M. Sheik-Bahae, D. C. Hutchings, D. J. Hagan, and E. W. Van Stryland, IEEE J. Quantum Electron. **27**, 1296 (1991).
[12] The linear Urbach tail is discussed in H. Haug and S. W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors* (World Scientific, Singapore, 1990).
[13] V. Mizrahi, K. W. DeLong, G. I. Stegeman, M. A. Saifi, and M. J. Andrejco, Opt. Lett. **14**, 1140 (1989).
[14] M. Sheik-Bahae, D. J. Hagan, and E. W. Van Stryland, Phys. Rev. Lett. **65**, 96 (1990).