Measurement of refractive nonlinearities in GaAs above bandgap energy

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We present a technique for single-beam measurement of the optical nonlinearity in GaAs for photon energies above the bandgap. We measured the real and the imaginary parts of the nonlinear refractive index of a bulk crystal by using the change in reflection of dye laser pulses (10 ns, 538 nm). The values obtained, $n_2 = (7.8 \pm 0.6) \times 10^{-8}$ cm^2/W and $\kappa_2 = (-2.8 \pm 0.7) \times 10^{-8}$ cm^2/W, are discussed. © 2000 Optical Society of America

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1. Introduction

The various applications of semiconductors in photonics make them an interesting subject for research in nonlinear optics. Nowadays these materials are widely used in the photonics industry in applications that range from light sources (LED's and laser diodes) to saturable-absorber mirrors[1] for mode locking in laser cavities. The optical properties of structures with low dimensionality, such as quantum wells, quantum wires, and quantum dots, are also an intensely studied subject.[2]

Some possible applications of semiconductors in photonics depend on the semiconductors' third-order susceptibilities. An extensive study of these refractive nonlinearities for photon energies below E_g was presented by Sheik-Bahae *et al.*,[3] who found typical values of $n_2 = -3.3 \times 10^{-12}$ cm^2/W for AlGaAs for the transparent range of the spectrum (810 nm). The absolute value of this nonlinearity increases as we approach the bandgap energy; it changes from a positive value for photon energy $E < 0.65\ E_g$ to a negative value as we approach E_g. Values as high as -2.1×10^{-6} cm^2/W are obtained for GaAs (Ref. 4) at the bandgap energy (1.42 eV). Unfortunately, we are aware of few direct experimental data above E_g.

Techniques such as the transmission Z scan[5] are frequently used for these measurements. In this technique, a focused Gaussian laser beam is transmitted through a thin sample. As we move the sample along the beam-propagation direction (z), this transmitted beam will suffer both amplitude and phase distortions. These distortions come from optical nonlinearities, which are larger at positions close to the waist of the beam, where the beam intensity is higher. From measurement of the phase and amplitude distortions as a function of sample position, we can obtain the value of the nonlinear refractive index. This is an accurate and fast way to measure n_2 by using a simple single-beam method in the transparent regime.

Other studies performed close to the bandgap energy for bulk semiconductors showed[6] a positive change in the refractive index for energies higher than E_g. In this highly absorptive region the change in the refractive index is obtained indirectly from the Kramers–Kronig transformation of the absorption change. This absoption variation is measured in a quasi-cw pump–probe experiment[7] in which the probe absorption is measured for various pump-induced carrier densities.

We propose direct measurement of the nonlinear refractive index in the absorption region of the spectrum for a bulk GaAs crystal. In this case the transmission Z-scan technique cannot be applied because the beam is highly attenuated. The high degree of absorption restricts the measurement of distortions in the transmitted beam, which either will be too feeble or will require a sample thickness that is near the absorption length. This thin sample can be difficult to handle or too fragile or can require the use of a substrate, which could introduce secondary effects

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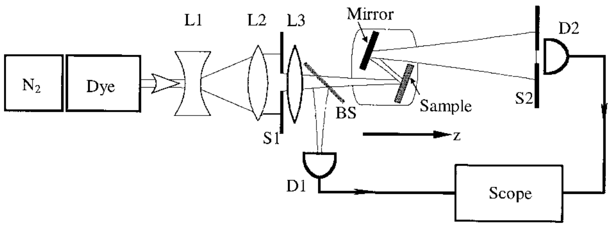

Fig. 1. Experimental setup for the *RZ*-scan measurement.

into our measurement. In this case, new single-beam techniques for measuring surface effects can be used.[8,9] The reflection Z-scan (*RZ*-scan) technique measures the nonlinear terms of susceptibility through the consequent changes in the reflection coefficient of the sample. These changes in reflectivity will produce, as in the transmission case, changes in phase and amplitude of the reflected beam at sample positions close to the beam waist.

The setup is shown in Fig. 1. The sample is a bulk GaAs substrate whose refractive and extinction coefficients are $n_0 = 4.3$ and $\kappa_0 = 0.363$, respectively.[10] We used a dye laser pumped by a pulsed nitrogen laser with a 10-Hz repetition rate. From the laser output we had 3-mJ 10-ns pulses (FWHM) at 538 nm, linearly polarized, with a poor Gaussian profile. Because of this poor spatial quality we worked with a top-hat beam, produced with beam-shaping optics. We used a diverging lens (L1; $f_1 = -25$ mm) and a converging lens (L2; $f_2 = 250$ mm) to expand the laser beam and an aperture to select a small region with a reasonably flat intensity profile. The output from aperture S1 was focused by a second converging lens (L3; $f_3 = 150$ mm), producing a top-hat beam with a waist radius of w_0. A beam splitter (BS) was used to produce normalization reference for the incident energy, measured by a silicon P–I–N photodiode (D1). The sample was moved along the direction of propagation simultaneously with a mirror oriented to produce parallel alignment between the output and the incident beams. Detector D2, placed in the far field, can measure either the total reflected power or, with a small aperture (S2), the on-axis intensity of the beam.

2. Reflection *Z*-Scan Technique

In the *RZ*-scan technique, a focused laser beam is reflected from the sample surface. For nonlinear material, the reflectivity of the sample depends on the change in refractive index induced by the incident intensity. This dependence of the reflectivity on the incident intensity produces phase and amplitude distortions in the reflected beam, that are similar to those produced in the transmission *Z*-scan technique. All *Z*-scan-based techniques use the measurement of these distortions in the reflected or transmitted beam to measure the nonlinear refractive index of the sample.

The laser beam is focused by a converging lens, which produces a spot of diameter $2w_0$. As we move the sample along the propagation direction of the beam, we find an increase in incident intensity as we approach the confocal region. Therefore the amplitude of any nonlinear effect will be augmented. If we measure the reflected power as a function of sample position z, we observe that the relative change in the reflected power will increase for the sample positioned at the waist.

The phase change in the beam for the sample placed near the confocal range will produce either an increase or a reduction in the divergence of the beam. A positive phase change will make the sample act as a convex mirror, increasing the divergence of the beam. A negative phase change, however, will act as a concave mirror, causing a convergence in the reflected beam. These phase changes can be measured by the variation of the far-field intensity at the center of the spot of the reflected beam.

To calculate the effect of these nonlinearities on the reflected beam, let us consider an incident beam with a dependence in the axial (z) and the radial (ρ) coordinates, whose amplitude is expressed by $E(\rho, z)$. For a sample with a nonlinear change in the refractive index, the reflection coefficient will depend on the local beam intensity. Expanding the reflection coefficient in first order for small changes in the complex refractive index ($|\Delta\tilde{n}| \ll \tilde{n}$), we can express the reflection as

$$\tilde{r} = \tilde{r}_0 + \frac{d\tilde{r}_0}{d\tilde{n}}\,\Delta\tilde{n} = \tilde{r}_0[1 + \hat{r}(\theta)\Delta\tilde{n}(\rho, z)]; \tag{1}$$

we consider here that the reflection coefficient will depend on the beam polarization and on incident angle θ. $\hat{r}$ is the relative change in the reflection coefficient and can be strongly dependent on the incident angle for parallel-polarized beams.[11] In this case, the linear term is given by the Fresnel reflection[12] and the normalized nonlinear reflection term is given by[11]

$$\hat{r}_p(\theta) = \frac{2\tilde{n}^3\cos(\theta) - 4\tilde{n}\cos(\theta)\sin^2(\theta)}{\tilde{n}^4\cos^2(\theta) - \tilde{n}^2 + \sin^2(\theta)}\,[\tilde{n}^2 - \sin^2(\theta)]^{-1/2}. \tag{2}$$

The reflected beam's amplitude is $E_R(\rho, z) = E(\rho, z)\tilde{r}_0[1 + \hat{r}(\theta)\Delta\tilde{n}(\rho, z)]$. From this amplitude we can calculate the total reflected power, measured by detector D2, as a function of sample position z along the beam. This power is given by the integration of the beam intensity, $R_T(z) = 4\pi \int_0^\infty |E_R(\rho, z)|^2\rho d\rho$.

Also, we can use Fresnel diffraction[12] to calculate the on-axis intensity in the far-field region. The measured power with a small aperture in front of detector D2 placed in a position $d \gg z_0$ is given by $R_0(z) = 4\pi|\int_0^\infty E_R(\rho, z)\rho d\rho|^2$.

If saturation effects on the nonlinearity are neglected, the complex change in the refractive index ($\Delta\tilde{n}$) will be linear with the intensity and expressable as $\Delta\tilde{n}(\rho, z) = (n_2 + i\kappa_2)|E(\rho, z)|^2$, where n_2 is the nonlinear refractive index and κ_2 is the nonlinear extinction coefficient.

Normalizing the integrals given above by their linear terms, we have for the normalized reflected power

$$I_T(z) = 1 + 2\,\mathrm{Re}[\hat{r}(n_2 + i\kappa_2)] \frac{\int_0^\infty |E(\rho, z)|^4 \rho d\rho}{\int_0^\infty |E(\rho, z)|^2 \rho d\rho} \quad (3)$$

and for the normalized on-axis intensity

$$I_0(z) = 1 + 2\,\mathrm{Re}\left[\hat{r}(n_2 + i\kappa_2) \frac{\int_0^\infty E(\rho, z)|E(\rho, z)|^2 \rho d\rho}{\int_0^\infty E(\rho, z) \rho d\rho}\right]. \quad (4)$$

For an RZ scan, in a sample with $n_0 \gg \kappa_0$ the amplitude of I_T is proportional to n_2, and the peak-to-valley amplitude of I_0 is proportional to κ_2, as shown in Ref. 9.

3. Top-Hat Beam

Gaussian beams are often used in Z-scan techniques because they are the common output modes (TEM_{00}) of lasers with good spatial profiles. There are situations, however, in which we want to apply these equations to different beam profiles, such as in the case of a top-hat beam.[13,14] This beam profile can be easily obtained if we use a small part of the wave front of a laser beam, small enough that the beam can be considered to have a uniform intensity profile in this region, and focus this uniform intensity profile, using a converging lens.

Using Fraunhofer diffraction to calculate the propagation from the aperture, we have near the focal plane a beam amplitude given by[12]

$$E(\rho, z) = E_0 \exp(ikz) \int_0^1 J_0\left(\frac{\pi}{w_0} \rho\eta\right) \times \exp\left[-i \frac{\pi\lambda}{4w_0} z\eta^2\right] \eta d\eta, \quad (5)$$

where the waist of the beam is $w_0 = \lambda f/d$, f is the focal length of the lens, d is the diameter of the aperture in front of the lens, and $J_0(x)$ is a zeroth-order Bessel function of the first kind. The Rayleigh length is given by $z_0 = \pi w_0{}^2/\lambda$.

If we know the linear term of the refractive index and the extinction coefficient, we can obtain the values of n_2 and κ_2 by fitting Eqs. (3) and (4) to experimental data. The curve obtained with the use of a top-hat beam does not change much from that obtained with a Gaussian beam. We find an increase (2.5 times) in the sensitivity, but we lose much of the laser power in the aperture. If the spatial profile of our laser is poor, the use of a top-hat beam is a convenient way to ensure the beam quality for a Z-scan measurement.

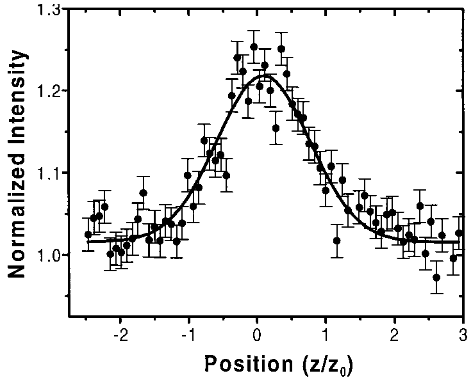

Fig. 2. Normalized reflected power for an RZ-scan, showing the amplitude distortion of the beam.

4. Results

Using the configuration of Fig. 1, we obtained a waist radius $w_0 = 32$ μm close to the focal plane and an average laser energy of 2.8 μJ/pulse. The incident angle was 24°, and, for parallel beam polarization, $\hat{r} = 0.13$, according to Eq. (2). Moving the sample along the beam axis, we measured and average amplitude of ~20 pulses at each position, normalizing the pulse amplitude measured in detector D2 by the value on reference detector D1. Making two series of measurements, one with the integration of the total reflected power and the other with the measurement of the on-axis intensity in the far field, we could observe both phase and amplitude changes in the sample's reflectivity.

In Fig. 2 we show the result obtained for a Z-scan when we removed the aperture from detector D2. In this situation the detector measures the total reflected power. By fitting Eq. (3) to the experimental data, we obtained a value of $n_2 = (7.8 \pm 0.6) \times 10^{-8}$ cm^2/W. Increased reflectivity at the beam waist is expected for a positive change in the refractive index. As was explained previously,[9] in the reflection Z-scan the real part of the refractive nonlinearity produces an amplitude change in the reflected beam, whereas for the transmission Z-scan the same nonlinearity produces a phase change.

Figure 3 shows the data obtained from the measurement with a small aperture placed in front of the detector. Inasmuch as the aperture diameter is much smaller than the beam diameter at the detector position, we can use Eq. (4) to fit the experimental data. From this fit we measured the imaginary part of the refractive nonlinearity, obtaining a value of $\kappa_2 = (-2.8 \pm 0.7) \times 10^{-8}$ cm^2/W.

There are many processes that can cause this refractive-index change. In presenting the optical nonlinearities for GaAs at room temperature, Peyghambarian and co-workers showed[6,7] that band filling of lower states in the conduction band is a mechanism for the reduction of absorption coefficient in semiconductors. The screening of the Coulomb

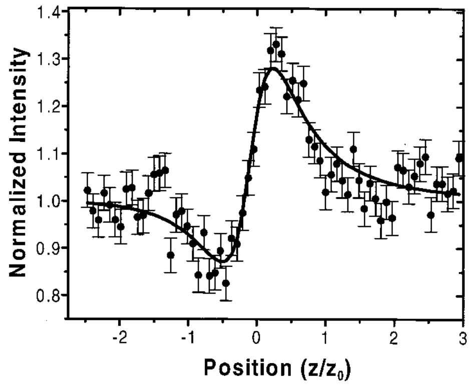

Fig. 3. Normalized on-axis intensity in the far-field region for an *RZ*-scan, showing the phase distortion of the beam.

interaction produced by the photoinduced electron–hole pairs has also been shown to contribute to absorptive nonlinearities.

For the high intensity involved, the contribution of thermal effects must also be considered a source of optical nonlinearities. In our case, as we have a pulse duration larger than the recombination time of the GaAs carriers (i.e., 2.3 ns; Ref. 15), we must take into account the heating of the sample that is due to nonradiative decay. To make a rough estimate of the thermal contribution to the measured change in the refractive index we have to calculate the absorbed power of the beam, converted into heat. Considering that ~23% of the incident energy is converted into heat, the rest being reflected or emitted as luminescent photons, we can estimate the change in the temperature of the sample as[16] $\Delta T \approx E/(\pi w_0{}^2 L_{\text{eff}} C_p)$, where E is the absorbed energy, C_P is the thermal capacity of the sample (1.7 J K^{-1} cm^{-3}), and the approximate heating volume depends on the beam size and the effective absorption length of the sample, $L_{\text{eff}} = 112$ nm. For an approximate value of $\mathrm{d}n/\mathrm{d}T = 18.7 \times 10^{-5}\ \text{K}^{-1}$,[10] we obtain $\Delta n_T \approx 0.2$, much smaller than the value $(n_2 I)$ obtained from the fitting of Fig. 2. Surface expansion can also be a factor for measurements with nanosecond pulses, especially in phase-distortion measurements, and a more precise separation of these effects must be made by use of short laser pulses.

Therefore we have presented a technique for direct measurement of the nonlinear terms of the refractive index in a GaAs sample for a photon energy above the bandgap. We have shown that this simple single-beam technique can be applied to bulk semiconductors. These results can be extended to other semiconductors or to semiconductor structures with low dimensionality. A complete spectroscopy of these values close to the bandgap can also be performed, giving a direct measurement of the results presented in Ref. 7. Using various time regimes, we can separate fast and slow contributions to the nonlinearity, reducing thermal effects.

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