Effect of low-power nonlinear refraction on laser-beam propagation in InSb

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By use of an expansion in Gaussian functions we solve the problem of the propagation of a laser beam that has been passed through a thin slab of nonlinearly refracting material. We compare the theoretical results for beam profile with experimental measurements for the semiconductor InSb and confirm that self-defocusing has been observed by D. A. B. Miller, M. H. Mozolowski, A. Miller, and S. D. Smith [Opt. Commun. **27,** 133 (1978)]. The deduced value for the nonlinear refractive index at ~5 K is -6×10^{-5} cm^2 W^{-1} at 1886 cm^{-1}. This implies a third-order optical nonlinearity $\chi^{(3)} \sim 10^{-2}$ esu, much larger than any previously reported for a solid.

Miller *et al.*[1] recently reported the first observation of various striking nonlinear optical effects in InSb obtained by using a cw CO laser. In particular, a Gaussian beam, having passed through the sample, was found to have developed a complicated profile, which was power dependent (see Fig. 1). This was tentatively attributed to nonlinear refraction. We here present a simple theory for the effect of nonlinear refraction on beam propagation and show that it can indeed account for observed profiles. We thus establish the sign of the appropriate nonlinear coefficient n_2 and estimate its magnitude, which we find to be exceptionally high. This coefficient is defined by

$$n(\mathbf{r}) = n_1 + n_2 I(\mathbf{r}), \tag{1}$$

where n is the refractive index and $I(\mathbf{r})$ is the beam intensity at position $\mathbf{r}$.

In the geometric-optics approximation, the only effect of the nonlinear refractive index is to produce a *phase shift,* which varies across the beam profile. This is a good approximation for low powers and large beam diameters. According to the theory that follows, the criterion for its validity is

$$\frac{4n_2L^2}{w_0{}^2 n_1} I_p \ll 1, \tag{2}$$

where L is the thickness of the sample, w_0 is the (e^{-2} intensity) beam radius, and I_p is the peak beam intensity.

We also assume that the sample is placed at the waist of the incident beam, as in the case of the measurements with which we make comparison, although this is not essential for the tractability of the theory.

Given these assumptions, we are left with the problem of the propagation in the z direction of a beam that emerges from the crystal at $z = 0$ with an amplitude

$$E(r,0) = E(0,0) \exp \left[\left(-\frac{r^2}{w_0{}^2} \right) + ix \exp \left(-\frac{2r^2}{w_0{}^2} \right) \right]. \tag{3}$$

The phase-shift parameter x is related to n_2 by

$$x = -\frac{\omega}{c} n_2 I_p \left(\frac{1 - e^{-\alpha L}}{\alpha} \right), \tag{4}$$

where ω is the radiation frequency and α is the linear absorption coefficient.

The key to the solution of the problem is to expand Eq. (3) as follows:

$$E(r,0) = E(0,0) \sum_0^\infty \frac{(ix)^m}{m!} \exp \left(-\frac{r^2}{w_m{}^2} \right), \tag{5}$$

where the radius of each of the individual Gaussian terms is given by $w_m{}^2 = w_0{}^2/(2m + 1)$. In this way, we express the initial beam profile as a sum of Gaussian

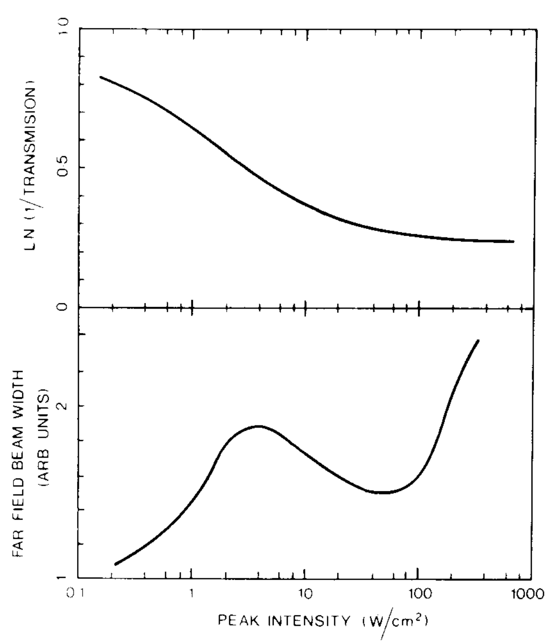

Fig. 1. Experimental transmission and far-field beam width (defined as the diameter at half peak intensity) against peak intensity. Laser line, 1882 cm^{-1}; spot diameter, 208 μm; InSb sample 7.5 mm long, antireflection coated on both ends, $N_D - N_A \simeq 3.8 \times 10^{-14}$ cm^{-3} (n-type); temperature $\simeq$5 K.

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beams of decreasing radius, and the propagation of such beams is described straightforwardly by the theory of Kogelnik and Li.[2] At distance z, the beam becomes

$$E(r,z) = E(0,0) \sum_0^{\infty} \frac{(ix)^m}{m!} \left(1 + \frac{z^2}{d_m{}^2}\right)^{-1/2} \times \exp\left[-\frac{r^2}{w_m{}^2(z)} - \frac{ikr^2}{2R_m(z)} - iP_m(z)\right], \quad (6)$$

In this formula, w_m is the beam radius of the mth Gaussian beam, given as a function of z by $w_m(z) = w_m(0)\,[1 + (z^2/d_m{}^2)]^{1/2}$, and similarly $R_m(z) = z[1 + (d_m{}^2/z^2)]$, $P_m(z) = -\tan^{-1}(z/d_m)$. The parameter d_m is the diffraction length, given by $d_m = [kw_m(0)^2/2]$, where k is the wave vector in free space. [The inequality (2) results from the comparison of sample thickness with such a length *within the sample,* defined for the value of m that makes the largest contribution to Eq. (6)].

Note that the sum over Gaussian beams is turned inside-out in the far field to become a sum over Gaussian beams of increasing radii.

For a general z, this series must be re-summed numerically to give the new beam profile. However, for the center of the beam, in the far-field limit ($r = 0$, $z \to \infty$), it can be summed analytically, to give

$$\frac{I(0,z)}{I(0,0)} \simeq \frac{\pi z^2}{4d_0{}^2} x^{-1} \,|\mathrm{erf}(\sqrt{-ix})|^2, \quad (7)$$

Here erf is the error function, also expressible in terms of the Fresnel integrals.[3] This expression has pronounced oscillations as a function of x and hence may be a suitable object of future measurements.

[As an alternative to expanding $E(r,0)$ in terms of Gaussian modes of differing spot sizes, Eq. (5), it is possible to make an expansion in terms of the Laguerre–Gaussian modes of equal spot size. These are described by Kogelnik and Li.[2] Such an expansion is, however, less satisfactory than that now given because one must perform integrations to obtain the expansion coefficients and because the series does not converge so well as Eq. (5).]

Before using this propagation theory for InSb we must show that the observed nonlinear absorption[1] cannot readily explain all the beam-shape distortion. We have measured the overall transmission and the gross width in the far field of an initially Gaussian[4] laser beam passed through an InSb crystal (Fig. 1). The absorption clearly saturates,[5,6] and this could change the beam shape by giving higher transmission in the beam center (giving effective narrowing in the near field and consequent broadening in the far field). There is indeed an alteration in the beam width correlated with the change in absorption. However, in the region above ~50 W/cm^2, the beamwidth increases strongly while the transmission shows very little change. This is evidence that the beam-shape distortion in this region is refractive rather than absorptive. Nonlinear absorption *can* be included in the present theory, and an effect ~0.02 cm W^{-1} (corresponding to the observed residual nonlinearity of absorption) gives only a small correction.

We have taken data for relatively low powers, for which the structure in the beam profile is most easily understood and where the validity criterion [Eq. (2)] is satisfied, and compared it with theory in the near and far field (Fig. 2). The initial beam can be considered as consisting of merely two terms in Eq. (5), and it is thus easy to see how Eq. (6) gives the near- and far-field profiles. The best fit has $x = 3.5$ (self-defocusing). Self-focusing ($x = -3.5$) gives an almost identical far-field profile (omitted for clarity), but the near field is in obvious disagreement with experiment.

Using Eq. (4) and noting that the sample has a measured effective *linear* absorption coefficient of ~0.9 cm^{-1} in the regime of interest, we infer a value for n_2 of -6×10^{-5} cm^2 W^{-1} for radiation of wave number 1886 cm^{-1} (12 cm^{-1} from the band gap) at ~5 K, corresponding to self-defocusing.

The conclusion that the effect observed here is self-defocusing has important consequences for the microscopic mechanism. Since in InSb (for photon energies below the band gap) the refractive index increases as the band-gap energy approaches the photon energy and the band-gap energy reduces with temperature, the refractive index should increase with temperature. For a Gaussian beam this should lead to self-focusing if thermal effects are the dominant mechanism. Furthermore, thermal effects would not lead to a functional form for n as in Eq. (1) (because of thermal conduction) so that the propagation theory described here would not apply; therefore the reasonable agreement of experiment and theory (in the near and far field), based on Eq. (1), is further evidence that the mechanism is not thermal. One likely explanation, which does predict strong self-defocusing, is that, just as linear absorption at frequencies above the band edge influences (by

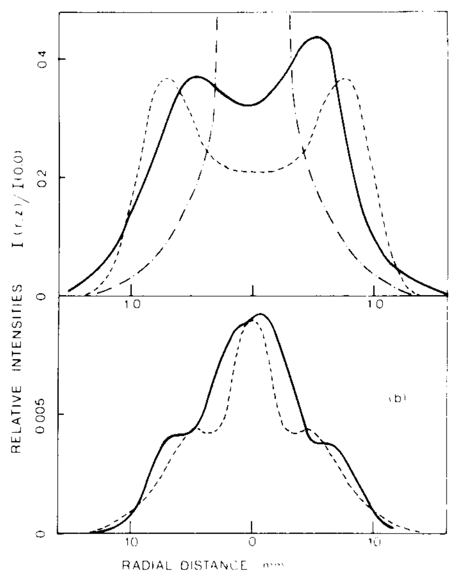

Fig. 2. Experimental (solid lines) and theoretical (broken lines) intensity profiles (a) in the near field at 7 cm from the sample and (b) in the far field at 189 cm. Data for 130-mW beam of 1.67-mm spot diameter on laser line at 1886 cm^{-1}; InSb sample and temperature as for Fig. 1. Theoretical profiles are shown for the self-*defocusing* condition ($x = 3.5$, dashed lines) in the near and far field and for the self-*focusing* condition ($x = -3.5$, dot–dashed line) in the near field to emphasize that the experimental results originate from a *defocusing.* In the far field, the focusing and defocusing results are practically identical. The theoretical and experimental plots are normalized to give the same *power* levels.

causality) the linear dispersion at all frequencies, then nonlinear absorption above the edge influences the nonlinear dispersion at all frequencies, and in particular at frequencies marginally below the edge. Indeed at the power levels used, a measure of saturation is expected above the edge.[7,8] The theory of the resulting band-gap resonant self-defocusing is discussed elsewhere.[6,9] We note that equivalent experimental results have been reported for gases,[10] self-defocusing occurring at frequencies just below absorption lines.

We note also that further evidence that this effect is refractive has been obtained in recent observations of optical bistability and differential gain in nonlinear Fabry–Perot devices using InSb as the nonlinear medium.[11]

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