UNIVERSAL DISPERSION AND BAND-GAP SCALING OF N_2 IN SOLIDS

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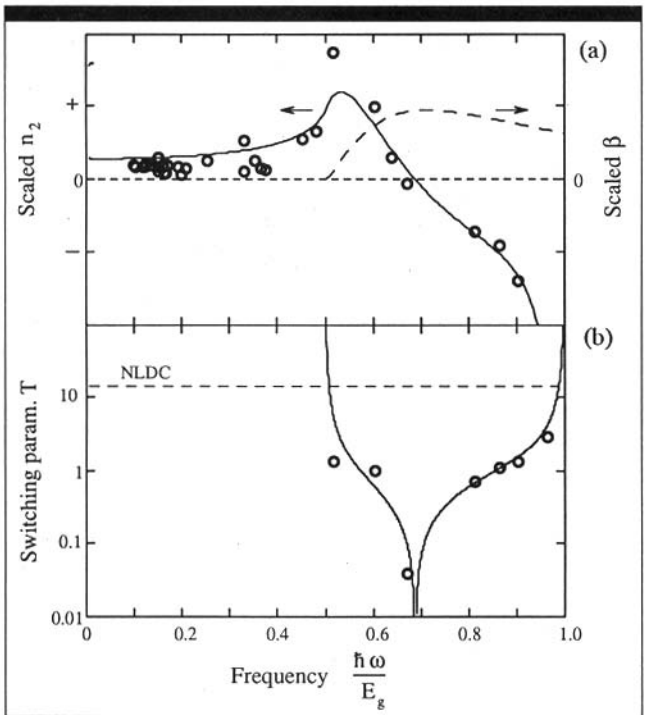
Using the "Z-scan" technique,[1] which can separately and accurately measure nonlinear absorption (NLA) and nonlinear refraction (NLR), two-photon absorption (2PA) and the bound electronic Kerr effect (n_2) have been measured in a large number of inorganic solids.[2] As a result of this large database and detailed characterization, a universal predictive capability for both the NLA and NLR of solids has been developed over a range of band-gaps from 0.2 to 10 eV. It is found that n_2 displays strong dispersion and sign reversal as $\hbar\omega$ approaches the band-gap energy E_g.



(a) UPPER TRACE, DISPERSION OF THE NONLINEAR REFRACTION WITH APPROPRIATELY SCALED DATA (*I.E.*, MULTIPLIED BY $N_0^2E_g^4$) INCLUDING DATA FROM REFS.3,4 (LEFT SCALE) AND THE 2PA SPECTRUM $\beta(\hbar\omega/E_g)$ SCALED BY $N_0^2E_g^3$ (RIGHT SCALE)
(b) FIGURE-OF-MERIT, T, VERSUS $\hbar\omega/E_g$. THE DASHED LINE INDICATES THE MINIMUM VALUE OF T NECESSARY IN A NONLINEAR DIRECTIONAL COUPLER (NLDC).

This universal dispersion and band-gap scaling law for n_2 were calculated using a Kramers-Kronig (KK) transformation of the NLA spectrum.[2] The KK transformation relates nondegenerate NLR to the nondegenerate NLA (*e.g.*, a change in absorption at ω_1 due to the presence of ω_2). To third order, 2PA, AC-Stark, and electronic Raman effects contribute to this NLA and were calculated using a "dressed" state Keldysh formalism from a two parabolic band model. The resulting n_2 varies as $G_2n_0^{-2}E_g^{-4}$, where n_0 is the linear index and G_2 is a dimensionless dispersion function that is an analytical expression, depending only on the ratio $\hbar\omega/E_g$.

The significant features of this theory are depicted in the top part of the figure, where our Z-scan experimental data, scaled by the predicted E_g^{-4} dependence, are compared with theory. Also included are measurements of, Ref.3 for large gap dielectrics. As evident from the dispersion curve, n_2 is positive and nondispersive at long wavelengths (small $\hbar\omega/E_g$). A resonant enhancement due to 2PA is exhibited at half the band-gap energy where n_2 reaches its positive maximum value and then exhibits anomalous dispersion, becoming negative near the band edge due primarily to the band-gap resonance of the AC-Stark effect.

The remarkable agreement between this simple theory and the vast amount of experimental data that spans over four orders of magnitude in n_2, points out the simple and fundamental physics underlying the fast electronic nonlinear processes in bulk semiconductors and large gap dielectrics. In addition, the KK transformation of the NLA spectrum yields, for the first time, a unified relationship between n_2 and 2PA, AC-Stark, and electronic Raman effects.[2] Such a relationship is crucial in evaluating a generalized figure of merit for all-optical switching applications where 2PA imposes serious limitations.[4]

Given both the 2PA coefficient β that scales as $n_0^2E_g^{-3}$ and n_2, one can calculate the all-optical switching figure-of merit $T = \omega|n_2|/\beta c$.[4] The theory shows that T is material independent, being only a function of $\hbar\omega/E_g$ as shown in the lower part of the figure. Also shown are data obtained through a combination of β and n_2 measurements, verifying this material independence. For efficient switching, T must exceed some value (typically of the order of unity) that weakly depends on the particular switching scheme. As seen in the lower part of the figure, for most applications, switching must be performed at frequencies below the 2PA edge.

REFERENCES

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