DISPERSION OF THE OPTICAL NONLINEARITY IN SEMICONDUCTORS[*]

R. K. Chang

Gordon McKay Laboratory, Harvard University, Cambridge, Massachusetts

and

J. Ducuing

Physics Department, Massachusetts Institute of Technology, Cambridge, Massachusetts

and

N. Bloembergen[†]

Department of Electrical Engineering and Physics, University of California, Berkeley, California

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The dispersion of the modulus of the nonlinear susceptibility $\chi_{14}^{\mathrm{NL}}(2\omega)$ (where NL stands for nonlinear) responsible for second-harmonic (SH) generation has been directly measured for the first time. The crystals investigated were several semiconductors having zincblende ($\bar{4}3m$) symmetry. The frequency range not only includes the absorption-edge region, but also extends well inside this region. A rapid variation of $|\chi_{14}^{\mathrm{NL}}(2\omega)|$ as a function of frequency is observed. Pronounced maxima can be correlated with the presence of the critical points in the joint density of states in the vicinity of the fundamental and/or harmonic photon energy. Such correlations are similar to the ones found in the linear optical data.[1,2] In addition, the nonlinear susceptibility, which is zero when the crystal has inversion symmetry, depends on the antisymmetric potential[3] which mixes the wave functions of different parity. This then leads to marked differences between the linear and nonlinear cases.

Nine different fundamental sources having photon energies in the range from 1.17 to 2.34 eV were used. The lowest photon energy is that of a Nd-doped glass laser, the highest that of its harmonic obtained by prior frequency doubling in a KDP crystal. The other frequencies are those of a ruby laser and its Stokes lines obtained by stimulated scattering from various fluids: H_2 gas gives a Stokes energy of 1.27 eV, cyclohexane 1.43 eV; benzene gives a first Stokes energy at 1.66 eV and a second Stokes at 1.54 eV; nitrobenzene gives a Stokes line at 1.62 eV; and CS_2 at 1.70 eV.

The experimental arrangement is shown in Fig. 1. The nonlinear susceptibility of single crystals was measured by the method of reflected harmonics.[4] Second-harmonic production in a quartz plate is used to monitor the laser intensity.[5] The intensity of the reflected harmonic beam from the semiconductor is measured and compared to that produced in transmission through a KDP crystal by the same source.[6] The nonlinear susceptibility of KDP is assumed to be independent of frequency in this range and taken as a reference.

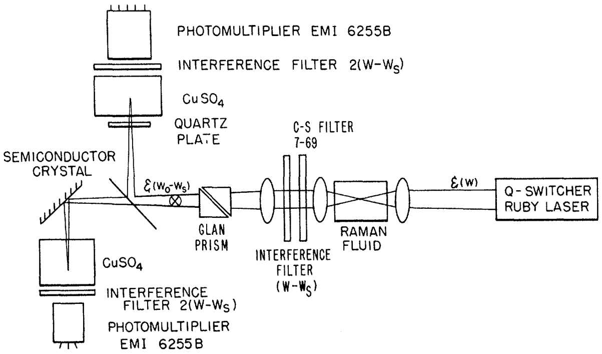

FIG. 1. Diagram of the experimental arrangement for the case when the incident radiation is produced by stimulated Raman scattering.

The nonlinear second-order sysceptibility tensor for a crystal with $\bar{4}3m$ symmetry has only three elements different from zero: $\chi_{14}^{\mathrm{NL}}(2\omega)=\chi_{25}^{\mathrm{NL}}(2\omega)=\chi_{36}^{\mathrm{NL}}(2\omega)$. An expression for the nonlinearity appropriate for a semiconductor has been derived.[7,8] A straightforward rearrangement of this expression yields

$$\chi_{14}^{\mathrm{NL}}(2\omega)=A\lim_{\mathrm{Im}\omega\to0}\sum_{l,l'}\int_{\mathrm{BZ}}d^3k\,f(E_{l,k})\times\left\{\frac{Q_{ll'}^{(1)}(k)}{\omega^2-\omega_{ll'}^2}+\frac{Q_{ll'}^{(2)}(k)}{4\omega^2-\omega_{ll'}^2}\right\}, \quad (1)$$

where A is frequency independent, $f(E_{l,k})$ is the Fermi-Dirac distribution function giving the occupation number of the state with the energy $E_{l,k}$. The summation is over band indexes, and the integration extends over the Brillouin zone (BZ). The quantities $Q_{ll'}(k)$ which involve a summation over intermediate states are real and independent of frequency. They vanish for a material possessing inversion symmetry. The above expression is to be compared with the corresponding one for the linear susceptibility

$$\chi^{\mathrm{L}}(2\omega)=B \lim_{\mathrm{Im}\omega\to 0} \sum_{l,l'} \int_{\mathrm{BZ}} d^3k f(E_{l,k}) \times \frac{|P_{ll'}(k)|^2}{\omega_{ll'}} \frac{1}{4\omega^2-\omega_{ll'}^2}, \qquad (2)$$

where B is also independent of frequency, the $P_{ll'}(k)$ are suitable reduced-momentum matrix elements,[2] and L stands for linear. The $|P_{ll'}(k)|^2$ are known to be slowly varying functions of k.[9] Were this to be also true of the quantities $Q_{ll'}(k)$, the dispersion of $\chi^{\mathrm{NL}}(2\omega)$ would be almost entirely governed by the joint density of states between bands l, l', as is the case for χ^{L}. If the contribution from the highest valence band and the lowest conduction band is dominant, as in the case of χ^{L}, the nonlinear susceptibility could then be represented to an excellent approximation by a linear combination of the values $\chi^{\mathrm{L}}(\omega)$ and $\chi^{\mathrm{L}}(2\omega)$. In general, such a relation will not hold, as the coefficients $Q_{ll'}(k)$, which involve triple products of the reduced-momentum matrix elements, depend on the phase of these latter quantities. The Q's can vary appreciably over the Brillouin zone, changing in sign as well as in magnitude. Because of this, the dispersion of χ^{NL} will not be determined entirely by the joint density of states. There may also appear additional structure due to the variation of the $Q_{ll'}(k)$ over the Brillouin zone.

The experimental error brackets stem from two causes. First, there is a consistent error of ±10% in determining the ratio of intensity of the reflected SH from the semiconductor and the intensity of the SH from KDP by transmission. The second source of error, which for many cases is the larger of the two, results from not being able to determine exactly the real and imaginary part of the linear dielectric constant, especially when one or both of them are sharply peaked. The Fresnel factors for nonlinear reflectors are a sensitive function of them. The values of the linear dielectric constant for GaAs, InAs, and InSb were obtained from Philipp and Ehrenreich.[2] For ZnTe, a Kramers-Kronig transform was performed using the available data on reflectivity and suitable extrapolation for the high-energy region.[10-12]

The case of ZnTe [Fig. 2(a)] provides a striking illustration of the variation of $|\chi_{14}^{\mathrm{NL}}(2\omega)|$ when the harmonic photon energy penetrates progressively inside the absorption region. Over the investigated range the absorption of the fundamental is negligible, but that of the harmonic increases by more than an order of magnitude to reach a maximum around $2\omega = 3.6$ eV, which corresponds to the peak called E_1 in semiconductor terminology in the joint density of states. Under these circumstances the second term inside the brackets in Eq. (1) ought to predominate entirely; that is, the density of states at $2\hbar\omega$ governs the behavior of $|\chi_{14}^{\mathrm{NL}}(2\omega)|$. Experimentally, as $2\hbar\omega$ penetrates inside the absorption band, $|\chi_{14}^{\mathrm{NL}}(2\omega)|$ increases progressively. Soref and Moos[13] had previously observed a similar behavior at a fixed light frequency when the band gap was varied in (ZnCd) S alloys. However, as they pointed out, their method assumes that the properties of the semiconductor-alloy series are such that varying the band gap by alloying is equivalent to shifting the applied laser frequency.

A different situation exists in InSb [Fig. 2(b)]. Here both the harmonic and fundamental photon energy are absorbed. As the source frequency increases, the fundamental photon energy $\hbar\omega$ gets closer to the E_1 peak, which for InSb appears at a much lower value (1.83 eV) than for ZnTe. On the other hand, there exists no critical point of the joint density of states within the interval covered by the harmonic photon energy, as evidenced by the fact that $\chi^{\mathrm{L}}(2\omega)$ has no pronounced structure. This should result in a strong influence of the first term in Eq. (1) on the dispersion of $\chi^{\mathrm{NL}}(2\omega)$. A pronounced variation of $|\chi_{14}^{\mathrm{NL}}(2\omega)|$ is observed experimentally, which seems to be somewhat related to that of the real part of the linear susceptibility $\chi^{\mathrm{L}}(\omega)$. The maximum value appears at $\hbar\omega = 1.6$ eV, where the $|\chi_{14}^{\mathrm{NL}}(2\omega)| = 5\times 10^{-6}$ esu. This is the highest value measured for a second-order susceptibility, nearly 2000 times higher than the corresponding value for KDP.

For GaAs [Fig. 2(c)], the E_1 peak again appears at higher value (2.90 eV), and the main

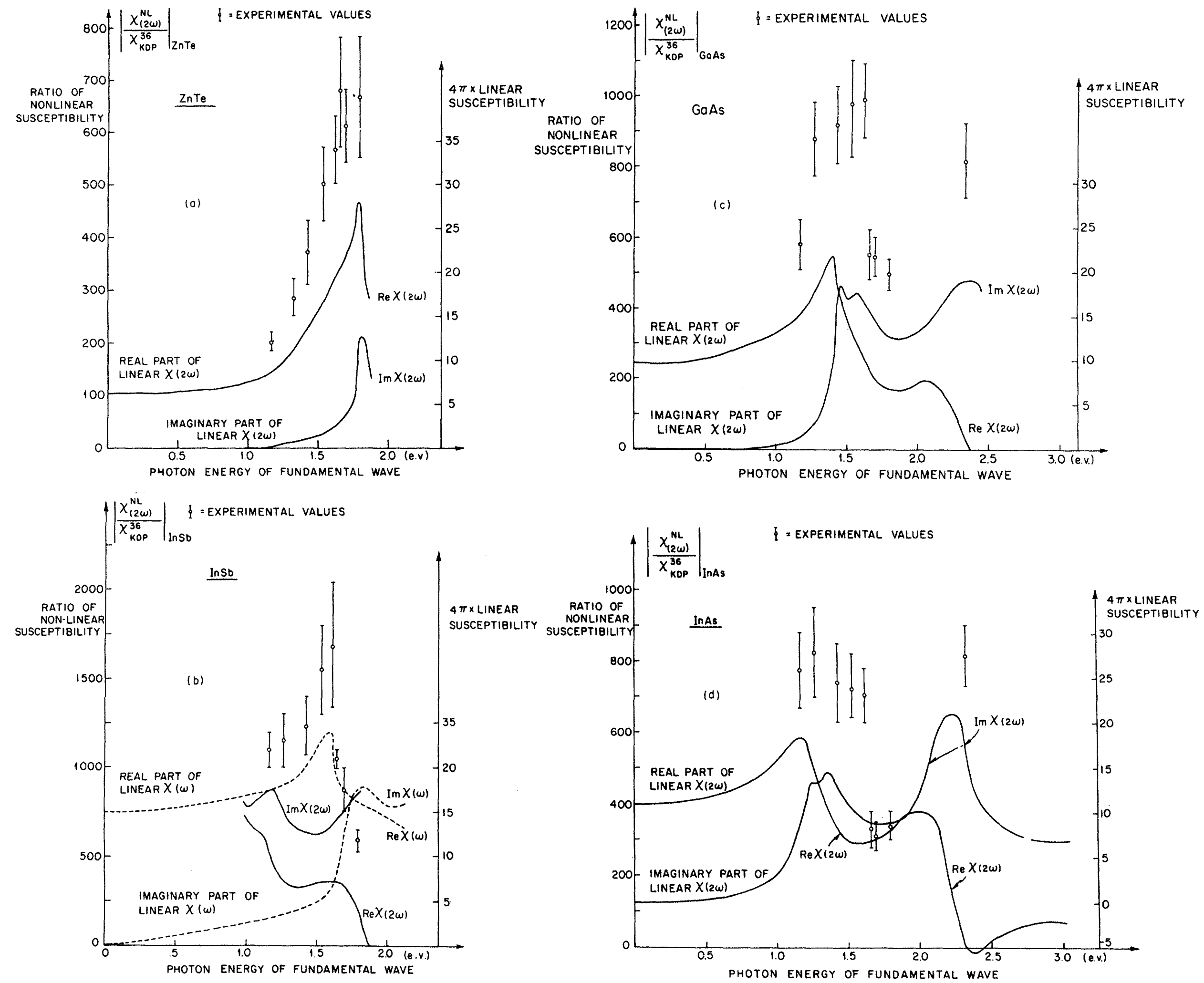

FIG. 2. The modulus $|\chi_{14}{}^{NL}(2\omega)|$ of the nonlinear susceptibility of (a) ZnTe; (b) InSb; (c) GaAs; and (d) InAs. The bar indicates the maximum uncertainty resulting from various sources of error. For the sake of comparison, the real and imaginary parts of the linear susceptibility $\chi^{L}(2\omega)$ have been plotted in all cases, while those of $\chi^{L}(\omega)$ are also displayed in case (b).

features of the dispersion are given by the second term in Eq. (1). Here a pronounced peak in $|\chi_{14}{}^{NL}(2\omega)|$ also exists in the neighborhood of the E_1 peak. Two novel features appear in the results for GaAs. First, at the highest source frequency, the harmonic photon energy is near the E_2 peak $(X_{5v}\rightarrow X_{3c}, X_{1c})$. A corresponding increase in the nonlinearity is observed. The contribution from the first term of Eq. (1) might also be significant at this point. Second, a very sharp decrease in $|\chi_{14}{}^{NL}(2\omega)|$ is observed around 1.67 eV. As seen from the linear data, the joint density of states does not exhibit such a fast variation and thus cannot be held responsible for this sharp decrease. Such data must probably be accounted for by a pronounced structure in the Q's. InAs [Fig. 2(d)] yields, as expected, results very similar to those obtained for GaAs. Both these features were observed again.

Our dispersion results clearly show the influence of the critical points in the density of states. They also bring out some features related to the Q's and hence to the hybridization of the wave function. The dispersion of the real and imaginary[14] part of $|\chi^{NL}(2\omega)|$ should provide some information not available from the study of linear properties alone.

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MÖSSBAUER EFFECT RESULTING FROM THE REACTION $Fe^{56}(d,p)Fe^{57}$†

D. A. Goldberg, P. W. Keaton, Jr., Y. K. Lee, L. Madansky, and J. C. Walker

Department of Physics, The Johns Hopkins University, Baltimore, Maryland

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We have observed a large Mössbauer effect in γ rays resulting from the reaction $Fe^{56}(d,p)Fe^{57}$ with a natural iron target. From the present result, together with the previously reported Mössbauer effect[1] from Coulomb-excited levels in Fe^{57}, one reaches the conclusion that the conditions for recoilless emission are relatively insensitive to the reaction process leading to the excitation, and to the amount of recoil momentum imparted to the nucleus during such reaction.

A Mössbauer effect with γ rays resulting from a nuclear reaction was previously observed,[2] but observation of a large effect with resolved hyperfine structure induced by (d,p) reaction is of particular interest in that the measurement of the hyperfine splitting in the target environment would shed additional insight into the still unknown process in which a recoilling nucleus with a large momentum is rapidly restored into a lattice before a recoilless γ emission occurs.

In the present experiment a 20-mg/cm^2 target of natural iron was bombarded with a 0.025-μA beam of 2.8-MeV deuterons from the Johns Hopkins University Van de Graaff accelerator. Due to the low beam currents employed, the target holder was merely cooled by means of room air convection.

The 14.4-keV γ rays were detected in coincidence with the preceding 122-keV γ rays in order to discriminate against the high background which was mainly due to bremsstrahlung and Compton-scattered γ rays. The experimental setup is similar to the one described earlier.[1] Both γ rays were detected using thin NaI(Tl) crystals, and the coincidence-circuit resolving time was approximately 100 nsec. With the beam current used, this gave rise to a coincidence counting rate of 2 counts per second, $\frac{1}{6}$ of which were accidental coincidences. Despite these precautions, the 14.4-keV coincidence spectrum exhibited a background counting rate roughly equal to that of the 14.4 γ rays.

The Mössbauer apparatus consisted of a loudspeaker driven by a parabolic wave form, and a multichannel analyzer operated in a multiscaling mode. A 91%-enriched Fe^{57} coil with a split absorption pattern was used as an absorber in this experiment, since such an absorber gives a very large central absorption peak in conjunction with the split emission pattern from a natural iron target.[3] The velocity range was chosen to exhibit just the central three peaks for the convenience of rapid accumulation of statistics for this initial experiment.

The width and depth of the absorption dips indicate that the difference in hyperfine splitting between source and absorber is less than 10%. The central absorption dip, uncorrected for background, is 15% of the nonresonant transmission (see Fig. 1). Comparison of the (d,p) results with those obtained using a standard split Co^{57} source placed in the same geometry