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Reflectivity and Optical Constants of Indium Arsenide, Indium Antimonide, and Gallium Arsenide*

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The reflectivities of indium arsenide, indium antimonide, and gallium arsenide were measured at nearly normal incidence from 2050 A to 15 μ at room temperature. The optical constants were computed from these data in the range 0–6.0 ev using the dispersion relation between the phase and the magnitude of the reflectivity. The reflectivities obtained show peaks near 2.5 and 5.0 ev, with splitting of the lower maxima into two smaller peaks whose separations are 0.35, 0.50, and 0.20 ev, respectively, for the three materials. The imaginary part of the reciprocal of the dielectric constant appears to have a maximum beyond 6.0 ev, the magnitudes of this quantity at 6.0 ev being 1.0 for indium arsenide, 1.7 for indium antimonide, and 0.8 for gallium arsenide. These values are uncertain because of the effect of extrapolating the reflectivity to energies above 6.0 ev.

1. INTRODUCTION

It is possible to determine the optical constants of a material from reflectivity data taken at a single incident angle over a large frequency range if a dispersion relation is used between the phase and the magnitude of the reflectivity. This technique was first suggested by Robinson[1] and has been used to obtain the optical constants of germanium,[2–4] barium oxide,[5] and silver.[6] It has further been applied to study the optical effects of the lattice vibrations in lithium fluoride[7] and the free-carrier effects in lead telluride.[8] Use of this technique in the present study of III–V semiconductor materials is described in this paper. Tauc and Abrahám[9] have measured the reflectivities of some of these materials, but no results have been published on the optical constants in the visible and ultraviolet regions.

The reflectivities of indium arsenide, indium antimonide, and gallium arsenide were measured at nearly normal incidence in the wavelength region from 15 μ to 2050 A, corresponding to a photon energy range of 0.1–6.0 ev. An extrapolation of these data to infinite frequencies was necessary in order to apply the dispersion relation. Some of the reflectivity data and calculated results have already been presented by Stern.[10]

2. REFLECTIVITY MEASUREMENTS

The basic instrument used for the measurements was a Perkin-Elmer model 112U spectrometer, modified with attachments for taking both relative and absolute reflectivity data. Interchange of sources, prisms, and detectors permitted measurements over the full spectral region investigated. Measurements in the infrared and at longer visible wavelengths were made with a Perkin-Elmer No. 012-0850 relative reflectance attachment, using an aluminum mirror as a standard beyond 1 μ and a rhodium mirror at shorter wavelengths.[11]

Because of possible lack of stability of mirror standards and the reduction in reflectance of the mirrors themselves at ultraviolet wavelengths, absolute reflectivity data were taken at short wavelengths, using a reflectometer attachment which was fabricated for this purpose.[12] A schematic of this attachment, which was used in the exit beam of the spectrometer, is shown in

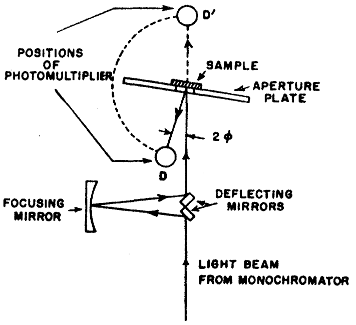

FIG. 1. Optical schematic of the absolute reflectance attachment.

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* Based on a thesis submitted to the Graduate School of the University of Maryland in partial fulfillment of the requirements for the degree of Master of Science.

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[11] The use of a rhodium mirror for short-wavelength relative reflectance measurements was suggested by Dr. Georg Hass, Engineer Research and Development Laboratories, since rhodium films are more stable than aluminum at ultraviolet wavelengths. Dr. Hass graciously calibrated a rhodium mirror for the author, and the mirror standards used in this study were calibrated against the one measured by Dr. Hass. Absolute measurements were subsequently made on this calibrated mirror by the author, the results agreeing with those of Dr. Hass.

[12] The basic design of this reflectometer is the same as that used for reflectivity measurements by Dr. J. Tauc and his associates at The Institute of Technical Physics, Czechoslovak Academy of Sciences, Prague, Czechoslavakia; F. Stern (private communication).

Fig. 1. The aperture plate, containing pairs of matched holes, can be rotated so that the monochromatic beam can either pass through onto the detector at position D', or be reflected from the surface of the sample which is mounted behind an aperture of equal size. The optical paths to D or D' are geometrically equivalent. The detector used for these measurements was a 1P28 photomultiplier, whose mount is fixed to an adjustable arm which can swing into the positions shown.

In addition to systematic reflectivity measurements over the spectral region studied, the effects on reflectivity of various polishing and chemical etching techniques were explored. The samples were initially polished with aluminum oxide on a pitch and tar lap, and these surfaces were further polished on laps of beeswax, felt, and silk. These tests, carried out principally on indium arsenide samples, showed a strong dependence, at short wavelengths, of the magnitude of the reflectivity on surface treatment. The highest reflectivity data were obtained after polishing on a silk lap. Two different chemical etchants[13] were applied to surfaces of mechanically polished indium arsenide samples for times up to 10 sec, with the result that the detail of the structure in the wavelength dependence of the reflectivity was increased but the over-all magnitude was reduced. It was felt that similar results could be shown for the other materials.

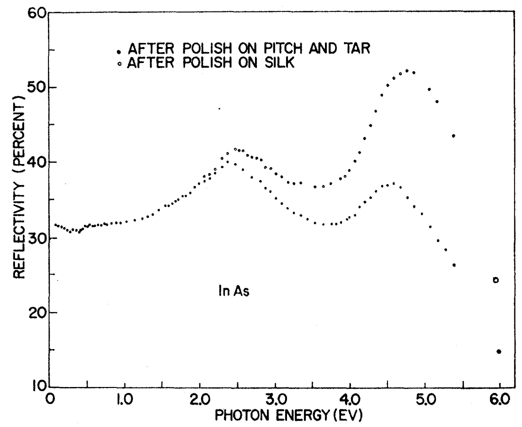

FIG. 2. Reflectivity data from induim arsenide sample from two different surface polishes.

The reflectivities for the materials studied are shown in Figs. 2, 3, and 4, data being included for two different polishes on the same sample of indium arsenide and indium antimonide.

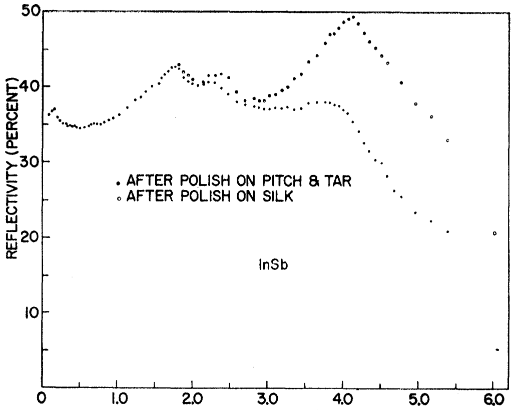

FIG. 3. Reflectivity data for indium antimonide sample from two different surface polishes.

No significant variation in the reflectivity of gallium arsenide resulted after polishing on silk; therefore, only one set of data is reported in Fig. 4.

3. CALCULATION OF THE OPTICAL CONSTANTS

At normal incidence the Fresnel reflection equations reduce to the single well-known form,

$$\rho=[(n-ik)-1]/[(n-ik)+1], \tag{1}$$

where ρ is the complex square root of the reflectivity, and n and k are the real and imaginary parts of the refractive index of the material; the incident and reflected rays are assumed to be in air. One can write ρ as the complex quantity,

$$\rho=re^{i\theta}, \tag{2}$$

so that simultaneous solution of Eqs. (1) and (2) gives

$$n=(1-r^2)/(1+r^2-2r\cos\theta), \tag{3a}$$

$$k=(-2r\sin\theta)/(1+r^2-2r\cos\theta). \tag{3b}$$

Knowledge of both r and θ at any frequency then determines the optical constants for that frequency. The

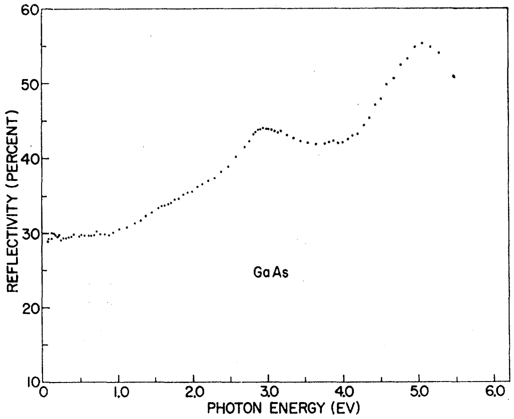

FIG. 4. Reflectivity for gallium arsenide sample after polish on pitch and tar lap.

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[13] The two etches used were (1) a solution of water and nitric acid at 80°C and (2) a room temperature solution of hydrochloric acid and nitric acid. The composition ratios of these etchants were varied, and the best results were obtained with a 5 sec etch in a 1:4 solution of nitric acid in water at 80°C. These etchants were suggested by H. C. Gorton, Technical Memorandum No. 6, Battelle Memorial Institute (unpublished), p. 3.

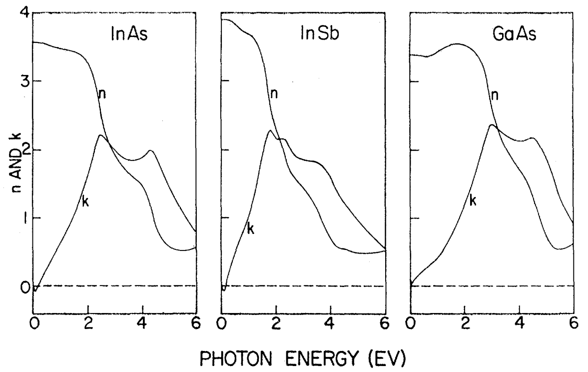

FIG. 5. Calculated values of the optical constants, n and k, from reflectivity data for indium arsenide, indium antimonide, and gallium arsenide.

dispersion relation,[14]

$$\theta(\nu_c)=\frac{2\nu_c}{\pi}P\int_0^{\infty}\frac{\ln r(\nu)}{\nu^2-\nu_c^2}d\nu, \tag{4}$$

where P indicates that the Cauchy principal value is to be taken, can be employed to calculate $\theta(\nu_c)$ if $r(\nu)$ is known for all frequencies. Values for r are obtained from the experimental reflectivity results for frequencies corresponding to photon energies up to 6.0 ev. Extrapolation to infinite frequencies is then required to make use of Eq. (4).

A single-parameter power-law extrapolation was used, the value of the parameter being chosen from a least-squares fit of low-frequency trial values of k with values of k determined from the experimental values of the absorption coefficient in other investigations.[15–17] The integral in Eq. (4) was evaluated by a method similar to that used by Gottlieb.[7] The optical constants were calculated only for the photon energy region 0 to 6.0 ev at intervals of 0.1 ev. At energies near 6.0 ev the calculated optical constants may be in considerable error since this is the limit of experimental data, and the extrapolated reflectivities contribute significantly to the results obtained from Eq. (4).

4. RESULTS AND DISCUSSION

The results of the calculations for n and k are shown in Fig. 5 for the highest reflectivity data for each of the three materials studied. The unphysical negative values for k near zero energy are attributed to the arbitrary choice of extrapolation discussed in the previous section. Peaks in the curves of the extinction coefficient k correspond with peaks at the same positions for the reflectivity data. Similar rises have been observed for germanium.[2] Phillips[18] has identified the one at lower energies with the transition $L_3\rightarrow L_1$ and the upper one with the transition $X_4\rightarrow X_1$. Although the labels for the corresponding states are different for zinc-blende lattices,[19] these same transitions are thought to be related to the peaks shown here for the III–V compounds.

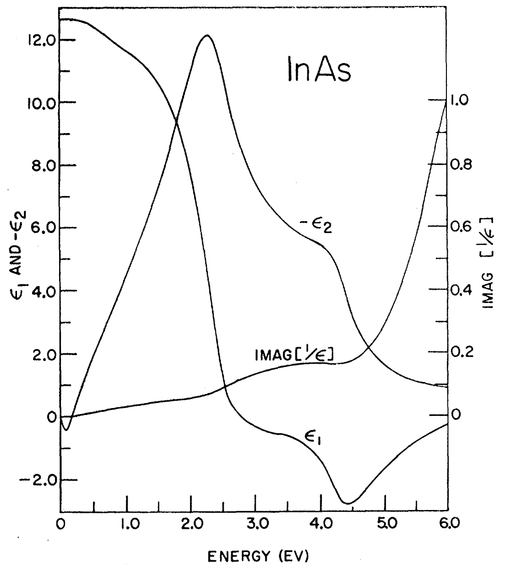

FIG. 6. Real and imaginary parts of the dielectric constant, ϵ_1 and ϵ_2, and $\mathrm{Im}(1/\epsilon)$ for indium arsenide sample as calculated from reflectivity data.

The higher values of the reflectivities shown in Figs. 2, 3, and 4 agree well with those obtained by Tauc and Abrahám[9] for these materials. The splitting of the long-wavelength peak shows up well for the indium antimonide samples, although this detail is not so apparent for the indium arsenide and gallium arsenide data. This splitting was displayed in all of the reflectivity curves of Tauc and Abrahám; their indium antimonide results showed it more sharply. This detail, which also appears for indium antimonide in the k curves of Fig. 5, is attributed to the splitting of the state L_3 by spin-orbit interaction.[9,20]

A quantity of interest in studying the rate of energy loss of an electron passing through a material is the imaginary part of the reciprocal of the dielectric constant,[6,21,22]

$$\mathrm{Im}(1/\epsilon)=\frac{-\epsilon_2}{\epsilon_1^2+\epsilon_2^2}\equiv\frac{2nk}{(n^2+k^2)^2}. \tag{5}$$

This quantity was calculated between 0 and 6.0 ev for indium arsenide, using the higher reflectivity data shown in Fig. 2. These results as well as the values of the real and imaginary parts of the dielectric constant ϵ_1 and ϵ_2, are shown in Fig. 6, the latter quantities being

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given by Eqs. (6):

$$\epsilon_1 = n^2 - k^2, \tag{6a}$$

$$\epsilon_2 = -2nk. \tag{6b}$$

$\mathrm{Im}[1/\epsilon]$ appears to be reaching a maximum just beyond 6.0 ev, corresponding to a small value of ϵ_2 when ϵ_1 is nearly zero. At this energy one should expect a collective or plasma oscillation to occur.[6] Similar maxima should appear for indium antimonide and gallium arsenide at energies just beyond 6.0 ev, $\mathrm{Im}(1/\epsilon)$ for these materials at 6.0 ev being 1.74 and 0.77.

CONCLUSIONS

The results of the investigation reported in this paper can be summarized in the following conclusions:

(a) The optical properties of InAs, InSb, and GaAs can be obtained from measurements of reflectivity and analysis of the data using dispersion relations. This technique can be applied to calculated optical constants where transmission measurements are not possible.

(b) The magnitude and spectral detail of the reflectivity of III–V compounds are strongly dependent on the surface quality, although for a given surface the measured reflectivities are reproducible to within ±0.01.

(c) Calculations of $\mathrm{Im}(1/\epsilon)$ for the three materials studied indicate that there should be a plasma energy at about 7 ev.

(d) Although the optical constants calculated are believed to be nearly correct at energies well below the limit of experimental data, more definitive results require extension of the reflectivity data further into the ultraviolet as well as a more refined extrapolation procedure.

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Sublattice Switching in Antiferromagnets

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An experiment is described which enables an antiferromagnetic sublattice assignment to be monitored in the presence of a small applied magnetic field. Using this technique it becomes possible to observe sublattice switching events, should they occur. It is concluded, despite the negative character of the results, that a particular magnetic sublattice assignment in $CuCl_2 \cdot 2H_2O$ can be considered a stable form at 1.4°K.

In principle, a quantum mechanical resonance effect should exist between a given antiferromagnetic spin arrangement and the configuration which follows upon inverting the magnetization direction of each of its spin sublattices. These two spin arrangements are indistinguishable and, in the case of a system of only two magnetic sublattices, are related by an interchange of sublattices.

Theoretical treatments by Anderson,[1] Kubo,[2] and Van Kranendonk and Van Vleck[3] established the long-term stability against rotation of the magnetization axes in the ground state of three-dimensional antiferromagnets, even in the limit of zero anisotropy energy. The tendency for the magnetization direction to wander is further inhibited by the existence of anisotropic interactions in real systems. The resonance effect between the two indistinguishable spin arrangements is extremely weak, in the ground state occurring only in a perturbation expansion carried to an order N, which is of the order of the number of atoms in the crystal. Although the problem becomes more complicated at higher temperatures,[2] the presence of even a small anisotropy energy tends to make rotations of the magnetization axes through large angles highly improbable. The assignment of definite magnetization directions to particular sublattices in antiferromagnetic materials has been likened[3] to heavy molecular isomers, technically metastable, but considered stable for all practical observation times.

Previous experimentally deduced upper limits placed upon the frequency of sublattice exchange events are too high to be significant. Neutron diffraction studies of antiferromagnetic materials are insensitive to the sublattice switching process if the lifetime of a particular magnetic sublattice arrangement is long compared to the time of interaction between an incident neutron and the spin array. Electron and nuclear magnetic resonance linewidth considerations in antiferro-

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