

Dielectric functions and optical parameters of Si, Ge, GaP, GaAs, GaSb, InP, InAs, and InSb from 1.5 to 6.0 eV

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We report values of pseudodielectric functions $\langle \epsilon \rangle = \langle \epsilon_1 \rangle + i \langle \epsilon_2 \rangle$ measured by spectroscopic ellipsometry and refractive indices $\tilde{n} = n + ik$, reflectivities R , and absorption coefficients α calculated from these data. Rather than correct ellipsometric results for the presence of overlayers, we have removed these layers as far as possible using the real-time capability of the spectroscopic ellipsometer to assess surface quality during cleaning. Our results are compared with previous data. In general, there is good agreement among optical parameters measured on smooth, clean, and undamaged samples maintained in an inert atmosphere regardless of the technique used to obtain the data. Differences among our data and previous results can generally be understood in terms of inadequate sample preparation, although results obtained by Kramers-Kronig analysis of reflectance measurements often show effects due to improper extrapolations. The present results illustrate the importance of proper sample preparation and of the capability of separately determining both ϵ_1 and ϵ_2 in optical measurements.

I. INTRODUCTION

Despite their status as fundamental parameters and the many attempts to determine them accurately, considerable uncertainty still remains in the intrinsic values of the dielectric functions $\epsilon = \epsilon_1 + i\epsilon_2$ of Si, Ge, and the principal III-V semiconducting compounds in the near-infrared to near-ultraviolet spectral range. The most commonly cited values are still those of Philipp and co-workers,¹⁻⁸ obtained by Kramers-Kronig transformations of reflectance data and first reported about 1960. Numerous seemingly equally valid data have since been obtained by transmission, ellipsometric, and non-normal as well as normal incidence reflectance measurements, but discrepancies of the order of 30% in ϵ and 10–20% in the reflectance R are common.⁷

Recently, three advances have occurred that provide an opportunity to obtain demonstrably more accurate spectral data for ϵ for all materials in general and for these semiconductors in particular. The first, and most important, is the development of new etching and cleaning procedures for these semiconductors.^{9,10} These procedures are designed specifically to minimize the thickness of surface transition regions where dielectric properties are changing from their bulk to ambient values. Such surfaces more accurately represent the ideal mathematically sharp interfaces assumed in the two-phase (substrate-ambient) model¹¹ used to calculate pseu-

dodielectric functions $\langle \epsilon \rangle = \langle \epsilon_1 \rangle + i \langle \epsilon_2 \rangle$ from ellipsometrically determined complex reflectance ratios. Consequently, such pseudodielectric functions should more accurately represent the intrinsic bulk responses ϵ .

The second is the development of fast, accurate, automatic spectroscopic ellipsometers¹² that can routinely determine $\langle \epsilon_1 \rangle$ and $\langle \epsilon_2 \rangle$ individually on a wavelength-by-wavelength basis without need to extrapolate measured properties into experimentally inaccessible spectral ranges. In addition to being intrinsically less sensitive to subjective input, the individual functional dependences of $\langle \epsilon_1 \rangle$ and $\langle \epsilon_2 \rangle$ to experimental artifacts such as improper calibration can be used to assess such elusive quantities as systematic errors via the self-consistency of $\langle \epsilon_1 \rangle$ and $\langle \epsilon_2 \rangle$ under Kramers-Kronig analysis.

The third is the development of unambiguous criteria for optically determining the "best" surfaces, that is, those most free of roughness, unintentional overlayers, physisorbed contaminants, and subsurface damage.^{9,10,13} When automatic ellipsometers are used, these criteria can be applied as the sample is being prepared, so spectral measurements can be restricted only to surfaces of the highest quality. This real-time assessment capability, missing in nearly all previous work, is particularly important because the optically most abrupt semiconductor surfaces are not obtained in ultrahigh vacuum, as commonly supposed, but with wet-chemical process-

ing techniques.¹⁴ This is due at least in part to the reconstruction that occurs on atomically clean surfaces, which is known to involve at least several monolayers.¹⁵

In view of the above discussion, it is perhaps not surprising that to lowest order all differences in reported dielectric functions can be understood by assuming that everybody knows how to take accurate data but nobody knows how to prepare and maintain accurate samples. That is, variations resulting from inadequate or improper sample preparation are far more significant than variations resulting from the use of different optical measurement techniques. The importance of sample preparation is well known for metal films, whose optical properties are affected by scattering processes and density deficits at grain boundaries.^{16,17} But since good semiconductor single crystals are readily available, less emphasis has generally been placed on preparation for these materials.

Some exceptions should be mentioned. Vedam and co-worker^{18,19} have shown that single-wavelength null-ellipsometric measurements of Si show systematic differences in the pseudorefractive index values according to preparation method. Archer²⁰ as early as 1958 and Donovan, Ashley, and Bennett²¹ in 1963 stressed the importance of proper sample preparation and of maintaining the samples in a dry-N₂ ambient to minimize atmosphere contamination effects. It is no coincidence that our $\langle\epsilon\rangle$ values for Ge coincide within 3% of the discrete-wavelength values obtained by Archer from 3600 to 7000 Å using null ellipsometry. It is also no coincidence that our calculated reflectance values for Ge coincide within 0.5% over most of the common spectral range with those measured by Donovan, Ashley, and Bennett on carefully electropolished single crystals. In addition, the only previous work on GaP to obtain the full height of the delicate E_1 structure was that of Stokowski and Sell,²² who studied the reflectance properties of GaP also using chemomechanically polished samples. In general, such care has not been taken, so discrepancies result. In some cases, as in reflectance measurements in the vacuum ultraviolet, some degradation is unavoidable owing to delays encountered in the pumpdown process even though the samples are nominally kept in an inert medium.

Upon investigating methods of preparing high-quality surfaces on semiconductors,^{9,10,13,14,23-28} we have noted that different procedures often produce overall agreement among limiting $\langle\epsilon\rangle$ values to within several percent. This indicates that some saturation value has been reached, i.e., that the $\langle\epsilon\rangle$ spectra are approaching their intrinsic bulk values. Based on our experience, there should be little fur-

ther change from these limiting values. Because the variations are now minor compared with those found in the literature, and in particular are much less than differences with respect to the Philipp-Ehrenreich results generally accepted as standard, and because these spectra are useful in many contexts and also bear on such fundamental related properties as band structure and many-body effects, we summarize our results in this paper. That, together with a critical assessment of the previous literature, is the objective of this work. Aside from the ϵ_2 values below the direct thresholds in Si and GaP, for which more accurate absorption data were substituted as discussed later, all results given here are pseudodielectric function data reported as observed with *no* corrections made for possible surface overlayer or substrate damage effects. This procedure is known to give more accurate results than Kramers-Kronig analysis or numerical compensation of overlayer effects.²⁹

II. EXPERIMENT

Pseudodielectric function values were obtained from 1.5 to 6.0 eV with an automatic rotating-analyzer ellipsometer (RAE) described in detail elsewhere.³⁰⁻³² The instrument was designed for accurate determination of $\tan\psi$ and $\cos\Delta$, which are the amplitude and projected phase of the complex reflectance ratio $\rho = r_p/r_s = (\tan\psi)e^{i\Delta}$, where r_p and r_s are the complex reflectances of *p*- and *s*-polarized lights, respectively.

To this end, a 30-mV triangular wave at the first subharmonic of the optical frequency was added to average out digital noise introduced by the analog-to-digital converter, and a light chopper was used to establish zero reference levels and to eliminate component drift and stabilization artifacts.^{31,32} A feedback loop with adjustable nonlinearity was used to establish overall linearity to better than 0.1%.^{31,32} Quartz Rochon prisms were used as combination polarizers-depolarizers to eliminate residual polarization sensitivities that might have remained at the detector.³³ The data were corrected to first order for the optical activity of the prisms.^{33,34} The polarizer azimuth was set at 30° to optimize accuracy for a wide range of reflectance conditions³⁵; the data were invariant to within 0.2% of peak values for azimuths in the range of 30°–40°. A 75-W Xe short-arc lamp provided stable, broad-band illumination with minimum interference from spectral lines. A 0.4-m Cary 14 foreprism-grating monochromator operated at 0.5-mm slit openings provided quasi-monochromatic light at a resolution ranging from 12 Å [full width at half maximum (FWHM)] at

2537 Å to 22 Å at 4915 Å with essentially no scattered light over the spectral range 1.5–6.0 eV.

Samples were optically aligned by maximizing transmission through the system with entrance and exit apertures stopped to their minimum 1-mm values. A previously described calibration procedure was followed after mounting each sample to obtain the necessary polarizer and analyzer reference azimuths.³⁰ The alignment and calibration procedures were accurate to within $\pm 0.03^\circ$ as determined by the overall reproducibility of alignment parameters that are system invariants, such as the azimuth angle of the plane of incidence in the frame of reference of the polarizer. The value $\phi = 67.08^\circ \pm 0.01^\circ$ of the angle of incidence was established by trigonometric measurements with an alignment laser.

Sample surfaces were initially prepared by Syton polishing or chemical etching in 10:1 HNO₃:HF solutions³⁶ for Si, Syton polishing for Ge, and bromine methanol (BrM) chemomechanical polishing for the III-V semiconductors. Final polishing, etching, and stripping procedures depended on the individual materials and surface orientations and are listed in Table I along with the properties of the samples used in this work. The procedures for GaAs, GaSb, and InSb are slightly different from those reported previously.^{9,10} The final stripping procedures were done with optically prealigned samples mounted vertically on a vacuum chuck in a windowless cell that maintained the surfaces in a dry-N₂ atmosphere. The N₂ was supplied by boiloff from a remote holding tank and was further purified by passing it through layers of anhydrous CaSO₄ and zeolite. Using this method of preventing hydrocarbons and water vapor from reaching the chemically cleaned surfaces, we were able to keep total accretion rates (oxidation and contamination) to values of the order of 1 Å/h, somewhat larger than those reported by Archer²⁰ for Ge, but still well within allowable tolerances.

Other surface preparation procedures, including

air cleavage (Si) and ion bombardment and annealing in ultrahigh vacuum (Si and Ge), were also investigated but were found to give thicker transition regions between bulk and ambient dielectric functions. The cleavage results are consistent with previous monochromatic null-ellipsometric measurements by Vedam and So,¹⁸ who also found systematic differences suggestive of damage at cleaved surfaces of Si.

III. THEORY AND ANALYSIS

All data were subjected to four tests: peak $\langle \epsilon_2 \rangle$ value, precision, self-consistency, and comparison to previous data. The peak $\langle \epsilon_2 \rangle$ value is a sensitive measure of transition-region widths.^{9,10,13} Its connection to ϵ , and the analogous connection between the normal incidence reflectances R and R_o obtained with and without a film present at the surface, can be conveniently expressed to first order in film thickness d as³⁷

$$\langle \epsilon \rangle = \epsilon + \frac{4\pi id}{\lambda} \frac{\epsilon(\epsilon - \epsilon_o)(\epsilon_o - \epsilon_a)}{\epsilon_o(\epsilon - \epsilon_a)} \times (\epsilon - \epsilon_a \sin^2 \phi)^{1/2} \quad (1a)$$

$$\cong \epsilon + \frac{4\pi id}{\lambda} \epsilon^{3/2}, \quad (1b)$$

$$R = R_o \left[1 + \frac{8\pi d n_a}{\lambda} \text{Im} \left[\frac{\epsilon_o - \epsilon_a}{\epsilon - \epsilon_a} \right] \right] \quad (1c)$$

$$\cong R_o \left[1 + \frac{8\pi d n_a}{\lambda} \text{Im} \left[\frac{\epsilon_o - \epsilon_a}{\epsilon} \right] \right], \quad (1d)$$

where ϵ_o and ϵ_a are the dielectric functions of overlayer and ambient, respectively, and λ is the wavelength of light. Here d and ϵ_o may be effective values averaged over the transition region.³⁸ The pseudodielectric function $\langle \epsilon \rangle$ is defined formally as that obtained by calculating ϵ from the ellipsometri-

TABLE I. Bulk properties and cleaning procedures for all samples measured in this work. BrM: 0.02 vol % bromine in methanol. BrM pad: polish 20 s on BrM-saturated lens paper, followed by 10-s quench with methanol. HF5: 5 vol % HF in methanol. BHF: Buffered HF, commercial solution. AmH: 1:1 NH₄OH:H₂O. HCl-meth: 1:1 HCl:methanol.

Material	Orientation	Doping (cm ⁻³)	Pretreatment	Final polish
Si	$\langle 111 \rangle$	$2.3 \times 10^{14}, n$	Syton	BrM pad; BrM, HF5, AmH, HF5 (meth rinse)
Ge	$\langle 111 \rangle$	$2.5 \times 10^{14}, n$	Syton	BrM pad; BrM, BHF (H ₂ O rinse)
GaP	$\langle 110 \rangle$	undoped	Bromine-meth	BrM pad; BrM, H ₂ O, AmH (H ₂ O rinse)
GaAs	$\langle 100 \rangle$	$1.7 \times 10^{17}, n$	Bromine-meth	BrM pad; BrM, H ₂ O, AmH (H ₂ O rinse)
GaSb	$\langle 111 \rangle$	$1.5 \times 10^{17}, p$	Bromine-meth	BrM pad; BrM, H ₂ O, HCl-meth (meth rinse)
InP	$\langle 100 \rangle$	undoped	Bromine-meth	BrM pad; BrM, AmH (H ₂ O rinse)
InAs	$\langle 110 \rangle$	$2.7 \times 10^{16}, n$	Bromine-meth	BrM pad; BrM, H ₂ O, AmH (H ₂ O rinse)
InSb	$\langle 110 \rangle$	$8 \times 10^{15}, n$	Bromine-meth	BrM pad; HCl-meth, BRM, H ₂ O

cally measured complex reflectance ratio ρ in the two-phase model,¹¹

$$\langle \epsilon \rangle = \epsilon_a \left[\sin^2 \phi + \sin^2 \phi \tan^2 \phi \left(\frac{1-\rho}{1+\rho} \right)^2 \right], \quad (2)$$

without regard to the possible presence of overlayers. In contrast to R , $\langle \epsilon \rangle$ is a derived, not a measured, quantity, hence the terminology pseudo-dielectric function.

Equations (1b) and (1d) follow in the limit that $|\epsilon| \gg |\epsilon_o| \gg |\epsilon_a|$. To gain some insight into the effect of thin overlayers on $\langle \epsilon \rangle$ and R , we show in Figs. 1 and 2 the results of exact calculations within the three-phase (substrate-overlayer-ambient) laminar model¹¹ for the specific cases of oxides and microscopic roughness for a typical semiconductor GaAs. In this calculation the present data were used for GaAs along with previously determined spectra for its oxide.²⁶ Also shown explicitly for R is the effect caused by microscopic roughness, here modeled in the Bruggeman effective-medium approximation³⁹ as a uniform film consisting of 60% GaAs and 40% voids. These fractions are typical of rough surfaces on crystalline or amorphous semiconductors.^{13,40} The corresponding effect of 14 Å of microscopic roughness on $\langle \epsilon \rangle$ is identical on the scale of Fig. 1 to that of the 10-Å oxide, and is not shown for that reason.

Qualitatively, it is clear that the largest effect in $\langle \epsilon \rangle$ occurs near the absolute maximum in $\langle \epsilon_2 \rangle$ (the E_2 peak), and that R is much less sensitive to the

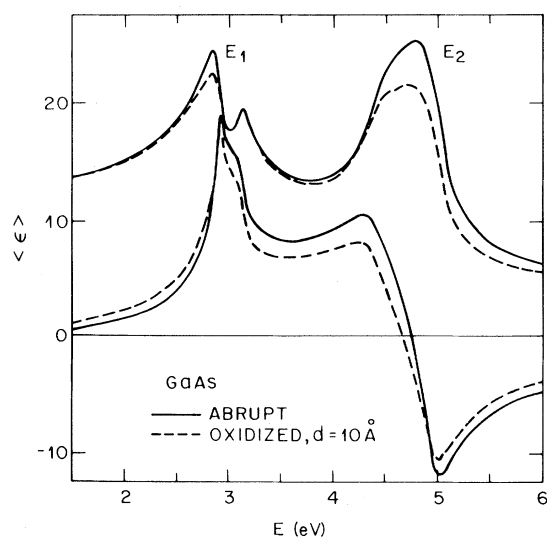


FIG. 1. Differences between pseudodielectric functions $\langle \epsilon \rangle$ for a nominally abrupt GaAs surface (—) and one covered with a 10-Å oxide (---).

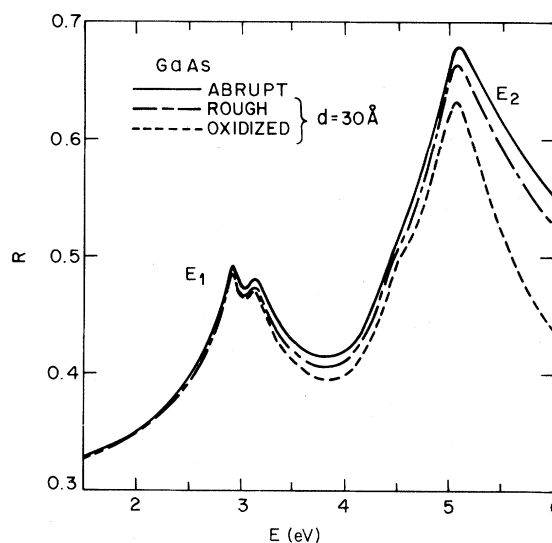


FIG. 2. Differences among calculated reflectances R for a nominally abrupt GaAs surface (—) and surfaces covered with 30 Å of microscopic roughness (---) or oxide (-·-·-).

presence of overlayers than $\langle \epsilon \rangle$. That a connection exists between sample quality and the value of R at the E_2 peak is well known.^{41,42} This connection can be understood qualitatively in that the penetration depth of light at the E_2 peak is near its minimum, so the surface region is relatively more important at this energy. Equations (1) show that R is less sensitive to surface conditions than $\langle \epsilon \rangle$ by a factor of about $\text{Im}[2\epsilon_a(\epsilon_o - \epsilon_a)/\epsilon]$, or about 3 at the E_2 peak of GaAs. While this indicates that reflectance measurements are more tolerant of surface problems than ellipsometric measurements, this insensitivity is not an advantage if the objective is to obtain accurate ϵ values simply because it means that R must be measured more accurately than $\langle \epsilon \rangle$. Also, surface problems are less likely to be recognized in a reflectance measurement until the data are analyzed.

At low energies where all semiconductors are transparent, finite overlayers in first order only affect $\langle \epsilon_2 \rangle$. This is a consequence of the phase retardation Δ of the p wave relative to the s wave, an effect which is commonly used in null ellipsometry as an indicator of the presence of surface films.^{18,19,43-46}

Precision was estimated by numerically calculating the third joint-density-of-states (JDOS) derivatives of the real part of the dielectric functions using the equation

$$D_n(E) = \frac{1}{E^2} \frac{d^n}{dE^n} [E^2 \epsilon_1(E)], \quad (3)$$

where $n = 3$. The calculation was performed by re-

peating the usual three-point algorithm

$$f'(x_1) = [f(x_{i+1}) - f(x_{i-1})] / (2\Delta x)$$

three times. The precision was considered satisfactory if the signal-to-noise ratio for the dominant E_1 derivative structure was at least 200, or if the signal-to-noise ratio for the E'_0 derivative structures (where applicable) was at least 2.

Self-consistency was investigated by calculating the Kramers-Kronig transform of the $\langle \epsilon_2 \rangle$ spectrum over the measured range $E_i \leq E \leq E_f$, then subtracting the result from the measured $\langle \epsilon_1 \rangle$ spectrum. This procedure yields a curve

$$S(E) = \epsilon_1(E) - \frac{2}{\pi} \mathcal{P} \int_{E_i}^{E_f} \frac{E' dE'}{E'^2 - E^2} \epsilon_2(E') \quad (4a)$$

$$\cong 1 + \frac{2}{\pi} \mathcal{P} \int_0^{E_i} \frac{E' dE'}{E'^2 - E^2} \epsilon_2(E') + \frac{2}{\pi} \mathcal{P} \int_{E_f}^{\infty} \frac{E' dE'}{E'^2 - E^2} \epsilon_2(E'), \quad (4b)$$

which in the absence of inconsistencies is smooth because $S(E)$ arises entirely from absorption processes lying outside the measurement range. Information about these absorption processes can be obtained by the curve fitting procedure described previously,⁴⁷ although we use this procedure here only to suppress the slowly varying background. This is done by assuming that ϵ_2 varies linearly between pairs of energies $E_1 - E_i$, $E_f - E_2$, $E_2 - E_3$, and $E_3 - E_4$, giving four amplitudes A_i , A_f , A_2 , and A_3 to be determined. Here $E_i = 1.5$ eV, $E_f = 6.0$ eV, and the higher-energy values E_2 , E_3 , and E_4 are arbitrarily set equal to 7.0, 9.5, and 14.0 eV, respectively. The amplitude A_4 is set equal to zero, and E_1 is adjusted so that A_1 is also zero. The distortions that remain after the best-fit curve has been subtracted from the $S(E)$ data indicate local inconsistencies.

The self-consistency test is not sensitive to overlayers such as oxides or microscopic roughness, but it is sensitive to experimental artifacts. The dominant artifact of a RAE involves ϵ_2 when $\epsilon_2 \cong 0$. The difficulty arises because ϵ_2 is proportional to $\sin\Delta$ for small ϵ_2 , which in a RAE is calculated from $(1 - \eta^2)^{1/2}$, where η is the ratio of ac to dc amplitudes. Suppose a small uncertainty δ exists in η such that $\eta = 1 - \delta$, i.e., $\epsilon_2 \cong 0$. Then it follows, with the use of Si at 2.27 eV as an example, that

$$\epsilon = 16.85 - 13.0\delta + i36.1\sqrt{\delta}.$$

Thus even if δ is as small as 1×10^{-4} , ϵ_2 is still uncertain to 0.36.

Measurements on Si, GaP, $\text{Al}_x\text{Ga}_{1-x}\text{As}$, and other bulk materials where ϵ_2 is known to be zero over at least part of the spectral range have shown that

the intrinsic ac/dc ratio decreases in our system by about 0.002 at 1.5 eV from the calibration value at 3000 Å because of decreased dispersion in the rotating-analyzer prism. This decrease begins to become important below 2.0 eV and results in increased amounts of nominally rejected light appearing at the detector. To approximately compensate for this RAE ratio artifact, we have linearly scaled all ac/dc ratios throughout the entire spectral range by a factor that varies linearly from about 0.998 at 1.5 eV to 1.000 at 3000 Å. For Si we have applied in addition a point-by-point scaling factor to bring $\epsilon_2 \cong 0$ in the region below the onset of direct transitions, then replaced the ϵ_2 data in these spectral ranges with the more accurate values calculated from the results of transmission measurements by other workers. This procedure will be discussed in more detail in the following section. Comparison of these data with those previously reported in the literature will comprise most of the remainder of the paper.

IV. RESULTS AND DISCUSSION

A. Silicon

Pseudodielectric function results for Si are shown in Fig. 3 and listed in Table II. The original $(\tan\psi, \cos\Delta)$ data were corrected for the RAE ratio artifact discussed in the preceding section. Because of the poor accuracy of $\langle \epsilon_2 \rangle$ for $\langle \epsilon_2 \rangle \cong 0$, we have replaced our $\langle \epsilon_2 \rangle$ data below 1.93 eV with values calculated from our results for n and from the absorption coefficient data of Dash and Newman (DN).⁴⁸ Between 1.93 and 2.84 eV, the data are a

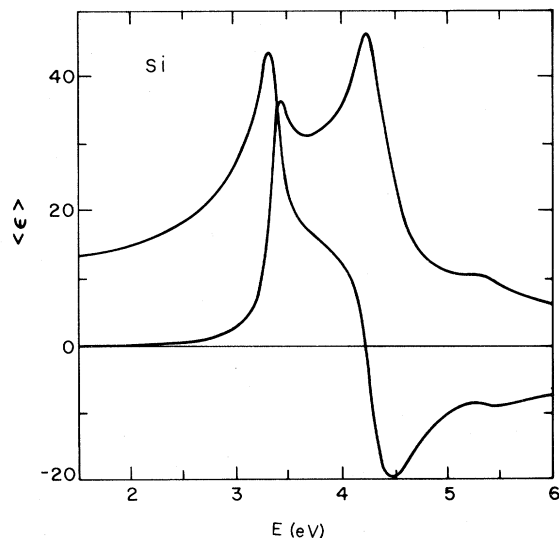


FIG. 3. Pseudodielectric function of crystalline Si, obtained as described in the text.

TABLE II. Optical properties of Si.

E (eV)	$\langle \epsilon_1 \rangle$	$\langle \epsilon_2 \rangle$	n	k	R	$10^3 \alpha$ (cm $^{-1}$)
1.500	13.488	0.038	3.673	0.005	0.327	0.78
1.600	13.793	0.057	3.714	0.008	0.331	1.25
1.700	14.079	0.078	3.752	0.010	0.335	1.80
1.800	14.413	0.099	3.796	0.013	0.340	2.38
1.900	14.797	0.126	3.847	0.016	0.345	3.15
2.000	15.254	0.172	3.906	0.022	0.351	4.47
2.100	15.754	0.236	3.969	0.030	0.357	6.32
2.200	16.334	0.260	4.042	0.032	0.364	7.17
2.300	16.994	0.396	4.123	0.048	0.372	11.19
2.400	17.761	0.508	4.215	0.060	0.380	14.65
2.500	18.661	0.630	4.320	0.073	0.390	18.48
2.600	19.724	0.803	4.442	0.090	0.400	23.81
2.700	20.987	1.193	4.583	0.130	0.412	35.63
2.800	22.565	1.548	4.753	0.163	0.426	46.22
2.900	24.574	2.017	4.961	0.203	0.442	59.75
3.000	27.197	2.807	5.222	0.269	0.461	81.73
3.100	30.874	4.321	5.570	0.387	0.486	121.62
3.200	36.355	7.636	6.062	0.630	0.518	204.28
3.300	43.264	17.717	6.709	1.320	0.561	441.68
3.400	35.224	35.282	6.522	2.705	0.592	932.13
3.500	22.394	33.818	5.610	3.014	0.575	1069.19
3.600	19.124	31.632	5.296	2.987	0.564	1089.90
3.700	17.231	31.527	5.156	3.058	0.563	1146.67
3.800	15.531	32.229	5.065	3.182	0.568	1225.46
3.900	13.965	33.567	5.016	3.346	0.577	1322.69
4.000	12.240	35.939	5.010	3.586	0.591	1454.11
4.100	9.364	39.947	5.020	3.979	0.614	1653.60
4.200	2.371	45.348	4.888	4.639	0.652	1974.84
4.300	-12.404	44.095	4.087	5.395	0.703	2351.38
4.400	-18.818	33.350	3.120	5.344	0.726	2383.23
4.500	-19.815	24.919	2.452	5.082	0.740	2317.99
4.600	-17.931	18.601	1.988	4.678	0.742	2181.15
4.700	-15.190	15.094	1.764	4.278	0.728	2038.07
4.800	-13.087	13.193	1.658	3.979	0.710	1936.06
4.900	-11.507	11.974	1.597	3.749	0.693	1862.11
5.000	-10.242	11.195	1.570	3.565	0.675	1806.67
5.100	-9.291	10.776	1.571	3.429	0.658	1772.70
5.200	-8.724	10.655	1.589	3.354	0.646	1767.66
5.300	-8.751	10.586	1.579	3.353	0.647	1801.26
5.400	-9.168	9.907	1.471	3.366	0.663	1842.63
5.500	-9.106	8.846	1.340	3.302	0.673	1840.59
5.600	-8.726	7.999	1.247	3.206	0.675	1820.07
5.700	-8.325	7.400	1.186	3.120	0.673	1802.31
5.800	-7.987	6.898	1.133	3.045	0.672	1789.99
5.900	-7.721	6.460	1.083	2.982	0.673	1783.51
6.000	-7.443	5.877	1.010	2.909	0.677	1769.27

linear interpolation between our $\langle \epsilon_2 \rangle$ data and those calculated from Ref. 48, with a weighting factor that changes from 0 to 1 over the range. The remaining optical parameters, the refractive index, reflectivity, and absorption coefficient, were all calculated from these results.

In our earlier work²⁴ we performed a similar re-

placement of our ϵ_2 data with those of Hulthén⁴⁹ rather than with those of DN. Upon further analysis, it now appears that the DN data more accurately represent the intrinsic absorption properties of crystalline Si. The DN absorption coefficients are in excellent agreement with similar data obtained by Braunstein *et al.*,⁵⁰ but lower than the Hulthén

data by factors of 2 and 1.3 at 1 and 2.8 eV, respectively. Motivated by these and other discrepancies, Taft⁵¹ very carefully remeasured k in transmission at $\lambda=5461 \text{ \AA}$ on bulk single crystals and found a value 0.031 ± 0.0015 , in agreement with that of DN (Ref. 48) and Braunstein *et al.*⁵⁰ and considerably lower than the value of 0.046 obtained by Hulthén.⁴⁹

Consequently, there seems little doubt that the earlier data are preferred. The origin of Hulthén's discrepancy appears to lie in the epitaxial nature of the silicon-on-sapphire (SOS) films used in his work, which in general show considerable twinning.⁵² Grain boundaries do not possess the full tetragonal symmetry of the single-crystal lattice. It is well known that the absorption coefficient of amorphous Si can exceed that of the crystalline material by a factor of 20 in this spectral region,^{53,54} so it is not unreasonable that the absorption coefficient of an SOS film could rise above that of a bulk single crystal.

The precision of our data as measured by the noise level in the numerically calculated third JDOS derivative of $\langle \epsilon_1 \rangle$ is illustrated in Fig. 4. Essentially no noise is evident over the entire spectrum up to 5.8 eV on the scale of the figure. The representative noise level near 5.3 eV is indicated schematically on the ten-times expanded E'_1 structure, and corresponds to $\pm 0.1\%$ of the peak-to-peak height of the E_1 structure near 3.4 eV. This is in the range of performance achieved by modulation techniques. The residual structure in these curves is in fact not due to random noise but to a small-period oscillation

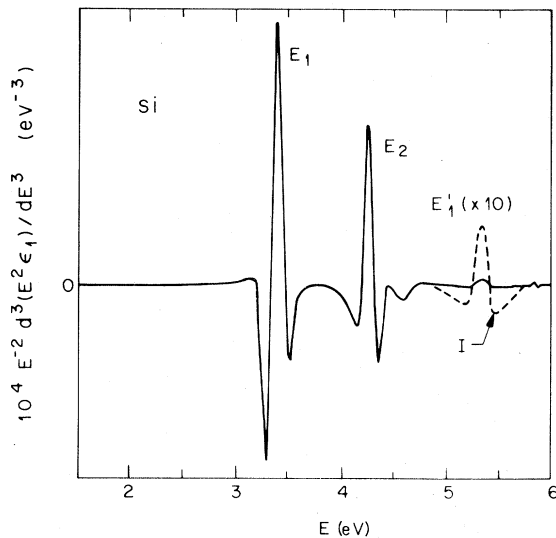


FIG. 4. Numerically calculated third JDOS derivative of $\langle \epsilon_1 \rangle$ for Si. The E'_1 structure near 5.3 eV is shown a factor of 10 larger for clarity. The uncertainty in these data is indicated by the error bar for the $10\times$ expanded section.

apparently resulting from multiple internal reflection within the Rochon prisms.

The self-consistency spectrum is shown in Fig. 5. This spectrum is obtained by removing the slowly varying background from the normalized difference $(\langle \epsilon_1 \rangle - \langle \epsilon_1 \rangle_{\text{KK}}) / \langle \epsilon_2 \rangle_{\text{pk}}$, where $\langle \epsilon_1 \rangle_{\text{KK}}$ is the Kramers-Kronig transform of $\langle \epsilon_2 \rangle$ as described in the preceding section, and $\langle \epsilon_2 \rangle_{\text{pk}}$ is the peak value of $\langle \epsilon_2 \rangle$ (46.80 at 4.25 eV for Si). The $\langle \epsilon_1 \rangle$ and $\langle \epsilon_2 \rangle$ spectra show overall relative self-consistency to within $\pm 0.5\%$. Structures near the E_1 and E_2 critical points at 3.4 and 4.2 eV indicate minor inconsistencies in the data on the scale of 0.2%. The scatter of the points composing the actual relative difference spectrum is indicated by the bars on the figure. The relatively large value near 3 eV is a consequence of the inability of an RAE to determine small values of $\langle \epsilon_2 \rangle$ accurately. The scatter decreases to lower energies because our $\langle \epsilon_2 \rangle$ data were replaced by the Dash-Newman values; the remaining scatter is that intrinsic to $\langle \epsilon_1 \rangle$.

We now compare these data to the results of other experiments. Numerous measurements have been made of the complex refractive index of Si at 5461 \AA , not only because of the technological importance of the material but also because high-quality single crystals are readily available. Moreover, the small extinction coefficient of Si at 5461 \AA possesses a certain attraction and challenge to developers of new methods for determining optical parameters of substrates by reflection or ellipsometric techniques. Considering first for measurements where both n and k are determined, the following results have been reported: $\tilde{n} = 4.13 + i0.04$ by Archer,⁵⁵ measured ellipsometrically on mechanically polished samples etched in HF; $\tilde{n} = 4.05 + i0.03$ by Claussen and Flower,⁵⁶ also measured ellipsometrically but on

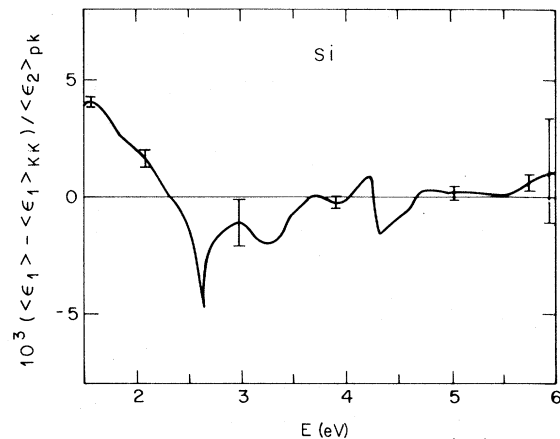


FIG. 5. Self-consistency spectrum for $\langle \epsilon_1 \rangle$ for Si under Kramers-Kronig transformation, as described in the text. The bars represent scatter in the difference spectrum.

chemically polished, hydrogen-annealed surfaces after a correction for an SiO₂ film determined by reflection electron scattering to be 3 Å thick; $\tilde{n}=4.052+i0.029$ by Vedam *et al.*,⁵⁷ measured ellipsometrically on chemically polished surfaces and also corrected for an SiO₂ overlayer; $\tilde{n}=4.075+i0.032$ by Philipp,^{1,5} calculated from a Kramers-Kronig analysis of (0–25)-eV reflectance data and corrected for a (12–15)-Å overlayer of SiO; $\tilde{n}=4.151+i0.210$ by Verleur,⁵⁸ calculated by fitting Lorentz-oscillator line shapes to (0–10)-eV reflectance data; $\tilde{n}=4.004+i0.042$ by Shewchun and Rowe,⁵⁹ calculated from ellipsometric measurements on oxidized and etched surfaces.

For measurements in which k was taken to be determined from transmission measurements, there are the following values: $n=4.05$ by Archer,^{20,60} calculated from ellipsometric data on mechanically polished and chemically etched surfaces; $n=4.09$ by Hulthén,⁴⁹ calculated by interference-fringe spacings observed in transmission measurements on SOS films described above; $n=4.103\pm0.020$ by Picozzi *et al.*,⁶¹ from derivative angle-of-incidence reflectance measurements on oxidized and etched surfaces; $n=4.085\pm0.003$ by van der Meulen,⁶² calculated from ellipsometric measurements also on oxidized and etched surfaces; $n=4.040\pm0.0022$ by Adams *et al.*,⁶³ on samples with a thin oxide present; $n=4.078$ previously reported by us²⁴ and obtained from spectroscopic ellipsometric data on oxidized and stripped surfaces after correcting for 7 ± 2 Å of SiO_{0.4±0.2}; $n=4.086\pm0.003$ by Taft,⁵¹ calculated from ellipsometric data on oxidized surfaces; $n=4.097$ in the present work, measured by spectroscopic ellipsometry on chemically stripped surfaces. Other results include ellipsometric measurements on air-cleaved surfaces by Vedam and co-workers, which yielded values of $\tilde{n}=4.13\pm i0.195$ (Ref. 64) from immersion analysis and $\tilde{n}=(4.140\pm0.02)+i(0.034\pm0.01)$ (Ref. 57) from standard corrections to SiO₂ films growing in air. In Ref. 64 the systematically larger n values for cleaved surfaces were attributed to defects, dislocations, and other imperfections generated in the cleavage process. A comparison of the various Archer data also shows that results on mechanically polished and presumably damaged samples⁵⁵ yield slightly higher values of n than results on chemically etched samples.^{20,60} However, Meyer *et al.*⁶⁵ also obtained data on vacuum-cleaved samples which showed values of n that would correspond approximately to 4.077 if converted to 5461 Å. It is clear that the quality of cleaved surfaces can vary significantly on the microscopic scale.

In a review of the literature, So and Vedam⁶⁶ selected a best bulk value of $\tilde{n}=4.056+i0.028$ at

5461 Å. Recent measurements have tended to show larger values for n . Most probably, the best data at 5461 Å are those of Taft,⁵¹ who reported $\tilde{n}=(4.086\pm0.003)+i(0.031\pm0.0015)$. We note that nearly all recently determined values for n fall within 0.5% of this number.

A similar situation occurs for 6328 Å (1.959 eV). Single-wavelength null-ellipsometric measurements where both n and k were determined independently include $\tilde{n}=3.91+i0.035$ by Algazin *et al.*⁶⁷ on atomically clean surfaces measured in 10⁻⁹ Torr and $\tilde{n}=3.75+i0.025$ by Moy⁶⁸ using an index-matching immersion technique. Spectral determinations evaluated at 6328 Å include $3.86+i0.029$ from Hulthén,⁴⁹ $3.86+i0.018$ from Philipp,⁵ and $3.899+i0.150$ from Verleur.⁵⁸ Transmission determinations of k include 0.018 from Dash and Newman⁴⁸ and 0.029 from Hulthén.⁴⁹ Discrete-wavelength measurements of n for which k is determined independently include $n=3.857$ from van der Meulen and Hien,⁶⁹ 3.87 from Hopper *et al.*,⁷⁰ 3.909 from Picozzi *et al.*,⁶¹ 3.89 and 3.92 from Brillouin scattering measurements by Sandercock,⁷¹ and 3.865 and 3.881 from our previous²⁴ and present works, respectively. The scatter here is considerably less than at 5461 Å.

Continuous ϵ spectra have been determined from 0 to 25 eV (Refs. 1 and 3–5) and 0 to 10 eV (Ref. 58) by analysis of reflectance data, from 0.5 to 4.0 eV by reflectance measurements on Ta₂O₅-coated wafers,⁷² from 2.0 to 3.7 eV by a modified polarimetric method,⁷³ and over various ranges in the visible near uv by spectroscopic ellipsometry.^{24,74–76} The Thutupalli-Tomlin data⁷² were obtained on <111> samples that were polished with alumina on a beeswax lap, and show slightly large values of k and slightly small values of n . This difference is characteristic of polishing damage in the surface region. Similar differences occur in the data of Schmidt,⁷³ although in this case the data were obtained using films of epitaxial Si on Si, where the films were stripped with HF before measurement.

The other data can be conveniently compared by examining the ϵ_2 values at the 4.25-eV E_2 peak. Dielectric function values calculated from a Kramers-Kronig analysis of the uncorrected broad-range reflectance data³ with a peak R value of 0.73 show a peak ϵ_2 value of about 41. Interestingly, the Lorentz-oscillator approach of Verleur yielded an ϵ_2 value of 27.8 at 4.25 eV on reflectance data that showed a higher E_2 peak of 0.82. A comparison of the results of Philipp and Ehrenreich and of Verleur provides a good example of the difficulties of extrapolation procedures and of the importance of determining both ϵ_1 and ϵ_2 in the same measurement.

Generally, published spectra have been corrected

in some way for the presence of an overlayer. Thus, Jellison and Modine⁷⁶ obtain a peak value of 43.92 for ϵ_2 after correcting for 20 Å of SiO₂. Daunois and Aspnes⁷⁴ show an E_2 peak of 52.35 at 4.25 eV by making a similar correction where the oxide thickness was determined by the requirement that ϵ_2 should approach zero in the spectral region below 2.5 eV. The uncorrected original Daunois-Aspnes data show a more realistic $\langle\epsilon_2\rangle$ peak value of 40.46 at 4.22 eV. In view of the present results, the correction in the latter work was unquestionably overdone, probably because of a combination of RAE inaccuracies in determining $\langle\epsilon_2\rangle$ at low energy and the overlayer not being stoichiometric SiO₂. In more recent work Aspnes and Theeten²⁴ calculated a peak value of 47.87 for ϵ_2 at 4.25 eV after correcting for a 6-Å interface of SiO_{0.4±0.2} found by analysis of samples covered with thick SiO₂ overlayers. Philipp⁵ also revised an initial value of 41.17 (Refs. 6 and 8) to 48.21 at 4.25 eV after correcting his original reflectance data for a (12–15)-Å overlayer of SiO. The overlayer was taken to be SiO because the characteristic SiO₂ absorption-edge structure at 10 eV was not present in the data, though the surfaces were known to be slightly oxidized. These results call into question the common practice of correcting optical data by assuming that overlayers are simple oxides. The errors that can be introduced if this assumption is not valid are discussed more fully by Jellison and Modine.⁷⁷ In general, less uncertainty results if one works at preparing high-quality surfaces instead of correcting existing data.

Other corrected spectra include data of Jungk⁷⁵ from 3.2 to 3.6 eV. These data were adjusted for an unspecified amount of SiO₂ on the surface. For convenience, we also compare values at a single wavelength, that of the 3650-Å (3.397-eV) Hg emission spectrum. Jungk's⁷⁵ value $\epsilon=37.5+i36$ and Daunois and Aspnes's⁷⁴ value $\epsilon=42.23+i34.53$ both represent overcorrections when compared, using Eq. (3), to our present result $\langle\epsilon\rangle=35.76+i34.98$. Again, the overcorrection is considerable in the latter case. So and Vedam⁶⁶ determined a single-wavelength value of $\epsilon=(39.43\pm0.44)+i(29.57\pm0.62)$ on an air-cleaved and thermally oxidized sample which is in rather good agreement with the revised Philipp⁵ value of $41.8+i28.5$. The corrected values show considerable scatter, but all lie well on the overcorrected side of present results. More seriously, the Philipp data seem to place the E_1 absorption edge about 50 meV to higher energy in contrast to the more recent work of Jungk⁷⁵ which, aside from amplitude differences, locates the threshold in good agreement with our present data.

Gerhardt⁷⁸ has also reported partial reflectance

spectra, although these were obtained for band-structure analysis and not with the objective of optical accuracy. His peak reflectance value, 0.688 at 4.52 eV, is considerably lower than our calculated peak value, 0.744 at 4.56 eV. Figure 2 shows that a reduced amplitude and a shift to lower energy are both characteristic of an overlying film. Likewise, Zanzucchi and Duffy⁷⁹ find a peak R value of about 0.71, which is essentially equal to the value of 0.717 observed by Philipp and Ehrenreich^{3,6,8} and less than that calculated from our data. Similarly, early work by Tauc and Abraham⁸⁰ on Si samples of unspecified surface treatment showed peak R values of only 0.59.

Ibrahim and Bashara⁸¹ also determined refractive index values at six Hg emission wavelengths from 2967 to 4358 Å by a multiple angle-of-incidence null-ellipsometric method that in principle should reduce sensitivity to overlayers and yield relatively accurate values of bulk dielectric functions. While our values are in reasonable agreement at 4358 Å ($23.50+i1.32$ in Ref. 81 and $23.40+i1.78$ in the present work) the agreement is less satisfactory at shorter wavelengths. At 2967 Å, their value $\epsilon=2.69+i42.92$ shows evidence of an overlayer when compared to our value of $4.51+i44.16$. At 3655 Å, their value of $36.93+i36.69$ cannot be understood in terms of a simple overlayer when compared to our value of $35.77+i34.98$, no matter whose data are assumed to represent the intrinsic bulk value. Independent multiple angle-of-incidence measurements on Si wafers with different thicknesses of thermally grown oxides were also made by So and Vedam⁸² in a procedure that should have yielded equivalent results. Their value of $\epsilon=(39.43\pm0.44)+i(29.57\pm0.6)$ is considerably different from both the present results and those of Ref. 81, indicating the existence of some difficulties with the multiple angle-of-incidence approach.

B. Germanium

Dielectric function data from 1.5 to 6.0 eV are shown in Fig. 6 and listed in Table III. As with Si, these data were taken on $\langle 111 \rangle$ surfaces because we could prepare more abrupt interfaces on these surfaces than on $\langle 100 \rangle$ and $\langle 110 \rangle$ surfaces. Except for the compensation of the RAE ratio artifact as discussed in Sec. III, the data are also as observed.

The $\langle\epsilon_2\rangle$ value at the E_2 peak is 30.63 at 4.26 eV, essentially equal to the value 30.74 at 4.25 eV that we reported previously in our surface-cleaning work.^{9,10} These spectra are self-consistent from 2 to 6 eV to within $\pm 0.1\%$ of the E_2 peak in Kramers-Kronig analysis, with maximum excursions of $\pm 0.5\%$ from 1.5 to 2.0 eV. The relative scatter of

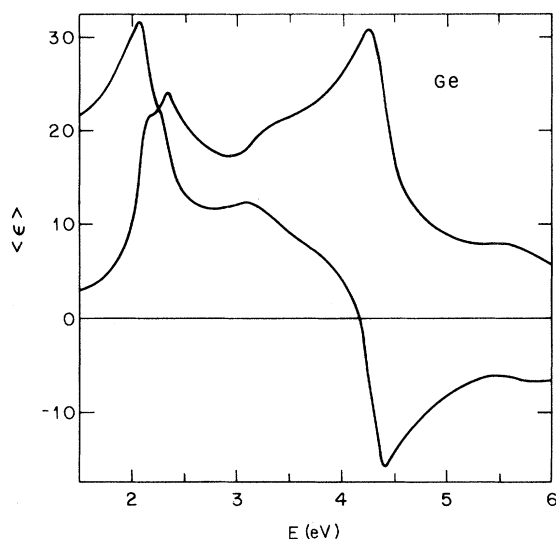


FIG. 6. Pseudodielectric function of crystalline Ge.

$\pm 0.01\%$ from 2 to 5 eV is somewhat better than that for Si.

These data compare well to the absorption coefficient measurements of Dash and Newman⁴⁸ in the near-edge region at 1.5 eV. The DN data in turn are consistent with results reported by Hobden⁸³ below 1.35 eV. Our value of $\alpha = 4.5 \times 10^4 \text{ cm}^{-1}$ at 1.5 eV is within 20% of the DN result $3.9 \times 10^4 \text{ cm}^{-1}$. However, our α values increase more rapidly than those of DN, becoming about 30% larger in the relatively strongly absorbing region near 2.0 eV where ellipsometric measurements are expected to be more accurate. All absorption coefficients are in good agreement at 1.5 eV, where the largest uncertainty occurs in the ellipsometrically determined data.

Unlike the case for Si, there has been very little single-wavelength work on Ge in the strongly absorbing regions of the spectrum. Archer⁵⁵ obtained $\epsilon = 21.34 + i 19.43$ at 5461 Å on mechanically polished samples stripped in HF, in surprisingly good agreement with our current result $\langle \epsilon \rangle = 21.55 + i 22.75$. Two measurements have been reported at 6328 Å. Baklanov *et al.*⁸⁴ obtained $\epsilon = 28.5 + i 8.6$ on thermally cleaned samples in a He atmosphere, while Algazin *et al.*⁶⁷ reported $\epsilon = 29.26 + i 9.65$ on atomically clean surfaces in ultrahigh vacuum. These data also compare reasonably well to our present result $\langle \epsilon \rangle = 29.26 + i 8.50$ at this wavelength.

The remaining data are in the form of reflectance or ellipsometric spectra. Some of these need not be discussed in detail because they were taken with low-accuracy reflectance-immersion techniques⁸⁵ or solely for the purpose of studying details of the

energy-band structure.^{78,86-92} The latter results are generally available over a limited wavelength region, and are uncalibrated or taken on samples not adequately characterized for absolute measurements. However, an early photometric ellipsometric "spectrum" for Ge obtained by Avery and Clegg,⁹³ is of historical interest, even though the resolution is too poor to resolve anything but the gross spectral features E_1 and E_2 . Taken on a natural facet of a pulled single crystal, the approximately correct $\langle \epsilon_2 \rangle$ value of 22 at E_1 together with the exceptionally low $\langle \epsilon_2 \rangle$ value of 8 at E_2 are indicative of a very thick oxide layer.

The first ellipsometric spectrum taken with moderate (120 meV) resolution from 1.77 to 3.44 eV was reported by Archer.²⁰ These data were taken on mechanically polished and chemically etched samples, with particular care being used to maintain the samples in a dry-nitrogen ambient to prevent oxidation and accretion of contaminants from the atmosphere. It is perhaps worth emphasizing that after 25 years of effort we have come full circle: After Archer's correction for a 10-Å oxide layer on his samples, our n and k coefficients are virtually identical ($\pm 5\%$) to his over the common spectral range, although Archer apparently did not resolve (or at least did not report) the E_1 and $E_1 + \Delta_1$ spin-orbit splitting. This splitting may have been destroyed in Archer's samples by the mechanical polishing technique that he used to prepare the surfaces.

Our previous ellipsometric results²³ on atomically clean Ge $\langle 111 \rangle$ samples maintained in UHV are in equally good agreement, although small differences do occur probably because of the difference in polarizability of the interface region. The value of $\langle \epsilon_2 \rangle$ for the E_2 peak of the atomically clean sample is 29.60 at 4.23 eV, indicating the presence of material with different polarizability than the bulk. Similar effects can be seen in the discrete-wavelength null-ellipsometric data of Meyer *et al.*,⁶⁵ taken from 1.3 to 3.3 eV on Ge $\langle 111 \rangle$ surfaces cleaved in UHV. The extinction coefficients agree to within 1% of our data, but the n values show some scatter particularly near the $E_1 + \Delta_1$ transition.

Similar good agreement is obtained with the reflectance measurements of Donovan *et al.*²¹ on electropolished samples also maintained in a dry-N₂ ambient. Reflectance values calculated from our data agree with Donovan's measurements to within 0.5% from 2.2 to 4.4 eV. The Donovan reflectances are 2% and 5% higher at 6000 and 8000 Å, respectively, and lower by about 1.3% at 2650 Å. Good agreement is also obtained with the reflectance measurements of Pajasova⁹⁴ on Ge surfaces etched in 100 ml HNO₃ + 50 ml HF + 5 gm CuNO₃; her measured reflectance peak is 0.71 at 4.5 eV compared to

TABLE III. Optical properties of Ge.

E (eV)	$\langle \epsilon_1 \rangle$	$\langle \epsilon_2 \rangle$	n	k	R	$10^3 \alpha$ (cm $^{-1}$)
1.500	21.560	2.772	4.653	0.298	0.419	45.30
1.600	22.565	3.288	4.763	0.345	0.428	55.97
1.700	23.819	3.925	4.897	0.401	0.439	69.06
1.800	25.426	5.069	5.067	0.500	0.453	91.25
1.900	27.622	6.761	5.294	0.638	0.471	122.96
2.000	30.361	10.427	5.588	0.933	0.495	189.12
2.100	30.373	18.791	5.748	1.634	0.523	347.91
2.200	23.711	21.651	5.283	2.049	0.516	456.93
2.300	20.254	23.466	5.062	2.318	0.519	540.33
2.400	15.224	22.636	4.610	2.455	0.508	597.26
2.500	13.153	20.695	4.340	2.384	0.492	604.15
2.600	12.139	19.307	4.180	2.309	0.480	608.62
2.700	11.642	18.289	4.082	2.240	0.471	613.12
2.800	11.526	17.604	4.035	2.181	0.464	619.05
2.900	11.717	17.282	4.037	2.140	0.461	629.17
3.000	12.065	17.514	4.082	2.145	0.463	652.25
3.100	12.240	18.349	4.141	2.215	0.471	696.14
3.200	11.802	19.450	4.157	2.340	0.481	758.90
3.300	10.944	20.385	4.128	2.469	0.490	825.88
3.400	9.914	20.994	4.070	2.579	0.497	888.81
3.500	9.052	21.442	4.020	2.667	0.502	946.01
3.600	8.268	21.992	3.985	2.759	0.509	1006.86
3.700	7.470	22.664	3.958	2.863	0.517	1073.71
3.800	6.573	23.509	3.936	2.986	0.527	1150.29
3.900	5.521	24.595	3.920	3.137	0.539	1240.23
4.000	4.123	26.056	3.905	3.336	0.556	1352.55
4.100	1.910	27.965	3.869	3.614	0.579	1501.84
4.200	-2.045	30.025	3.745	4.009	0.612	1706.55
4.300	-9.177	30.089	3.338	4.507	0.659	1964.58
4.400	-15.474	23.494	2.516	4.669	0.705	2082.48
4.500	-14.655	16.782	1.953	4.297	0.713	1960.14
4.600	-12.727	13.620	1.720	3.960	0.702	1846.52
4.700	-11.243	11.768	1.586	3.709	0.690	1767.14
4.800	-10.071	10.510	1.498	3.509	0.677	1707.30
4.900	-9.107	9.592	1.435	3.342	0.664	1659.73
5.000	-8.277	8.911	1.394	3.197	0.650	1620.15
5.100	-7.567	8.424	1.370	3.073	0.636	1588.72
5.200	-6.980	8.115	1.364	2.973	0.622	1567.25
5.300	-6.511	7.943	1.371	2.897	0.609	1556.15
5.400	-6.232	7.893	1.383	2.854	0.600	1562.04
5.500	-6.176	7.842	1.380	2.842	0.598	1584.57
5.600	-6.247	7.741	1.360	2.846	0.602	1615.23
5.700	-6.501	7.507	1.310	2.866	0.613	1656.06
5.800	-6.794	6.946	1.209	2.873	0.632	1689.07
5.900	-6.789	6.275	1.108	2.831	0.644	1693.31
6.000	-6.648	5.672	1.023	2.774	0.653	1686.84

our calculated value of 0.714 at 4.47 eV. The significantly lower peak value of 0.61 obtained by Tauc and Abraham⁸⁰ on etched Ge samples is clearly indicative of the presence of overlayer material. The results of Schmidt⁷³ do not extend to sufficiently high energies, but they also show signs of an oxide overlayer.

The results of Kramers-Kronig analyses of broad-band reflectance data in general show poorer agreement. We have previously²³ compared Philipp's dielectric function spectra⁹⁵ with our UHV results; several discrepancies occur, notably in the steepness of the ϵ_2 edge near E_1 and in the value of $\epsilon_2=32.00$ at 4.22 eV for the E_2 peak. The un-

corrected data^{2,3} may actually agree better over the entire spectral range. Similar remarks apply to the results of Rimmer and Dexter,⁹⁶ who used the Philipp-Taft reflectance values with a different extrapolation above 11.3 eV. The work of Lavilla and Mendelowitz,⁹⁷ a Kramers-Kronig analysis of a composite of several vacuum-ultraviolet spectra,^{2,3,98-100} shows a value of $\epsilon_2 = 22.7$ at the E_2 peak in ϵ_2 and thus is not accurate in the visible-near-uv spectra. Likewise, the Kramers-Kronig calculations of Rustgi *et al.*,¹⁰¹ based on the Philipp-Taft data below 7.6 eV and on data from mechanically polished bulk crystals or thin evaporated films above, lead to anomalously low values of n and k .

Potter^{102,103} has presented refractive index data from 0.5 to 3.0 eV based on a pseudo-Brewster-angle reflectance method. The oxide-corrected data on electropolished samples give n values that agree with ours within 1% if the fine structure is ignored. The k values, however, are low by $\sim 20\%$ from 1.8 to 2.0 eV. Data obtained by Jungk¹⁰⁴ from 1.9 to 4.1 eV on CP-4 etched samples show $\langle \epsilon_1 \rangle$ values that are low at low energies and $\langle \epsilon_2 \rangle$ values that are high at low energies and low at high energies, these being characteristic distortions of oxides or microscopic roughness.

As an example of typical scatter, we list the following refractive index values at 6328 Å: Sandercock,⁷¹ $\bar{n} = (5.55 - 5.61) + i(0.65 - 0.70)$ by Brillouin scattering; Philipp,⁹⁵ $\bar{n} = 5.38 + i0.38$ by Kramers-Kronig analysis of reflectance data; Potter,^{102,103} $\bar{n} = 5.53 + i0.69$ by pseudo-Brewster-angle reflectance measurements; Archer,²⁰ $\bar{n} = 5.43 + i0.82$ by spectroscopic ellipsometry with the sample in UHV; the present work, $\bar{n} = 5.46 + i0.78$. In general, there is much less scatter among the ellipsometric data than among those measured by other techniques.

C. GaP

Dielectric function data for GaP from 1.5 to 6.0 eV are shown in Fig. 7 and listed in Table IV. Like Si, GaP is an indirect band-gap semiconductor, but unlike Si, GaP has a substantial transparency range (1.5–2.24 eV) over this spectral region. Our $\langle \epsilon_2 \rangle$ data are relatively imprecise below the onset of the direct gap, which occurs at 2.76 eV at room temperature.²² For this reason our $\langle \epsilon_2 \rangle$ data were set equal to zero up to the 2.24-eV energy of the indirect threshold. Between 2.24 and 2.76 eV, our data were replaced by ϵ_2 values calculated from absorption coefficient spectra measured on polycrystalline films by Spitzer *et al.*,¹⁰⁵ which are in agreement with similar measurements by Folberth and Oswald¹⁰⁶ and by Welker.¹⁰⁷ Unfortunately, the ac-

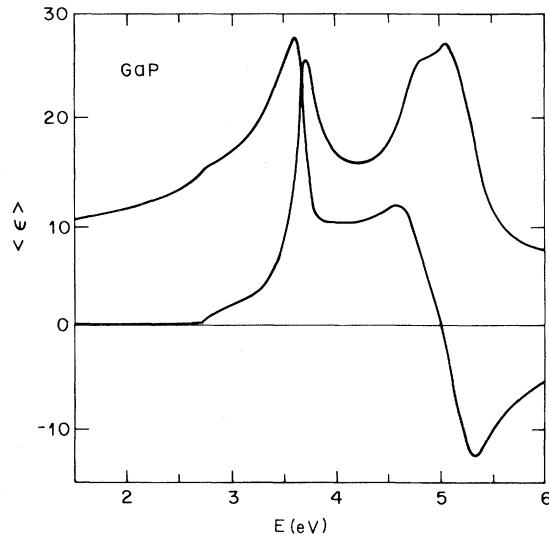


FIG. 7. Pseudodielectric function of crystalline GaP, obtained as described in text.

curate absorption coefficient data of Dean and co-workers^{108,109} are only available at low temperature and are not directly relevant here. At 2.76 eV the transmission-derived ϵ_2 data merge smoothly with our ϵ_2 data, which are used exclusively above this energy.

The data are Kramers-Kronig consistent to within $\pm 0.5\%$ of the $\langle \epsilon_2 \rangle$ peak value of 26.86 at 5.05 eV. These data show structure near E_1 and E_2 of peak-to-peak height 0.3%, indicating a lower degree of self-consistency than that observed for Si and Ge. The scatter is comparable to that of Ge, indicating a relative precision of $\pm 0.01\%$ over most of the spectral range.

Of all materials studied here only GaP has a range of transparency that overlaps the present range of our ellipsometer, so only GaP offers the possibility of a consistency check of ϵ_1 with the highly accurate values calculated from prism-deflection methods. Refractive index data taken by this means have been reported by Oswald and Schade,¹¹⁰ Pikhtin and co-workers,^{111,112} Bond,¹¹³ and Nelson and Turner.¹¹⁴ The data of Nelson and Turner are up to 0.1% larger than those of Bond, but the difference is actually due to uncertainties in the calibration of the divided circle used by Bond.¹¹⁴ The data of Pikhtin and co-workers agree to within the scale of their figures. Our values of n from 5500 to 7000 Å, the overlap range, are between 0.1% and 0.3% larger than theirs. The agreement is quite satisfactory. We take this agreement as indirect support for our values determined for other materials to which prism-deviation methods cannot be applied.

TABLE IV. Optical properties of GaP.

E (eV)	$\langle \epsilon_1 \rangle$	$\langle \epsilon_2 \rangle$	n	k	R	$10^3 \alpha$ (cm $^{-1}$)
1.500	10.102	0.000	3.178	0.000	0.272	0.00
1.600	10.299	0.000	3.209	0.000	0.275	0.00
1.700	10.459	0.000	3.234	0.000	0.278	0.00
1.800	10.639	0.000	3.262	0.000	0.282	0.00
1.900	10.860	0.000	3.295	0.000	0.286	0.00
2.000	11.114	0.000	3.334	0.000	0.290	0.00
2.100	11.390	0.000	3.375	0.000	0.295	0.01
2.200	11.702	0.001	3.421	0.000	0.300	0.04
2.300	12.067	0.011	3.474	0.002	0.306	0.36
2.400	12.496	0.025	3.535	0.004	0.312	0.86
2.500	12.996	0.046	3.605	0.006	0.320	1.63
2.600	13.621	0.082	3.691	0.011	0.329	2.94
2.700	14.475	0.209	3.805	0.027	0.341	7.52
2.800	15.342	0.923	3.919	0.118	0.352	33.42
2.900	15.889	1.463	3.990	0.183	0.360	53.90
3.000	16.601	1.832	4.081	0.224	0.369	68.26
3.100	17.532	2.304	4.196	0.275	0.380	86.28
3.200	18.717	2.923	4.339	0.337	0.394	109.25
3.300	20.231	3.854	4.518	0.426	0.410	142.64
3.400	22.248	5.398	4.751	0.568	0.431	195.80
3.500	24.833	8.268	5.050	0.819	0.458	290.40
3.600	27.354	14.787	5.406	1.368	0.496	499.04
3.700	20.509	25.243	5.149	2.451	0.530	919.21
3.800	11.040	21.496	4.196	2.562	0.500	986.69
3.900	9.826	17.920	3.890	2.303	0.467	910.55
4.000	9.652	16.454	3.790	2.171	0.452	880.10
4.100	9.669	15.756	3.752	2.100	0.444	872.59
4.200	9.833	15.490	3.754	2.063	0.441	878.38
4.300	10.145	15.609	3.792	2.058	0.442	896.99
4.400	10.583	16.163	3.867	2.090	0.449	932.14
4.500	11.073	17.343	3.978	2.180	0.461	994.27
4.600	11.294	19.506	4.113	2.371	0.482	1105.61
4.700	10.124	22.675	4.181	2.712	0.511	1291.94
4.800	6.915	25.149	4.062	3.096	0.539	1506.17
4.900	3.502	25.812	3.844	3.358	0.557	1667.57
5.000	0.218	26.580	3.661	3.631	0.580	1839.99
5.100	-4.628	26.571	3.342	3.975	0.614	2054.74
5.200	-9.408	23.558	2.825	4.170	0.647	2197.82
5.300	-12.316	18.744	2.249	4.168	0.678	2239.10
5.400	-11.967	13.829	1.778	3.889	0.689	2128.87
5.500	-10.266	10.974	1.543	3.556	0.677	1982.53
5.600	-8.785	9.520	1.444	3.297	0.657	1871.33
5.700	-7.669	8.575	1.385	3.096	0.637	1788.84
5.800	-6.790	7.911	1.348	2.934	0.618	1724.82
5.900	-6.096	7.436	1.327	2.803	0.600	1676.14
6.000	-5.521	7.041	1.309	2.690	0.583	1635.71

The value 3.565 determined by Lösche *et al.*¹¹⁵ with the use of ellipsometry at 5461 Å on material polished with NaOCl is about 4% higher.

At energies above the direct edge, the earlier Kramers-Kronig analysis of Philipp and Ehrenreich^{3,6-8} has been augmented by direct ellipsometric determinations from 1.5 to 5.5 eV by Mor-

gan,¹¹⁶ 3.4 to 4.0 eV by Jungk,⁹² and 1.1 to 3.4 eV by Burkhard *et al.*¹¹⁷ In addition, Stokowski and Sell²² have reported high-precision reflectance data in the energy range from 2.4 to 6.0 eV which includes the E_1 and $E_1 + \Delta_1$ transitions. While there is general agreement between our results and the previous data, some significant differences occur. Our E_2 peak

value $\langle \epsilon_2 \rangle = 26.86$ at 5.05 eV is considerably higher than the corresponding values of 22 and 24 for Philipp and Ehrenreich³ and for Morgan,¹¹⁶ respectively; the data for Burkhard *et al.*¹¹⁷ did not extend to high enough energies to make this comparison possible. The lower values are indicative of oxides or other overlayers, as seen in Fig. 1. Our maximum value, $\langle \epsilon_2 \rangle = 25.52$, at the E_1 peak is also significantly larger than the values 17.5 and 18 obtained by Philipp and Ehrenreich³ and by Morgan,¹¹⁶ respectively, but in better agreement with the value of 22 obtained by Jungk.⁹² Although Burkhard *et al.*¹¹⁷ do not express their data in ϵ form, a comparison with their refractive index values indicates a similar discrepancy with their results.

The reason is simple: The E_1 peak in ϵ_2 in GaP is exceedingly sharp and consequently very delicate, and difficult to maintain and to measure. All ϵ_2 values over 20 fall in an energy range only 180 meV wide. Thus either inadequate resolution or sample preparation will fail to reproduce the proper peak value. The major problem with the Philipp-Ehrenreich data³ may have been resolution, although the existence of the oxide overlayer present on their samples would also act to reduce the peak. Morgan¹¹⁶ cleaned his surfaces by ion bombarding and annealing. It is doubtful that such a procedure could yield smooth, stoichiometric GaP surfaces in view of the different volatilities of the two elements. Our previous data on atomically clean (and monoatomic) Ge (Ref. 23) indicate that argon bombardment and annealing do not yield the most abrupt interface regions on semiconductor surfaces. Recent measurements by Gheorghiu and Théye¹¹⁸ on annealed flash-evaporated films are not competitive due to sample quality; however, their objectives involved amorphous, not crystalline, properties.

The room-temperature reflectance data from 2.4 to 6.0 eV reported by Stokowski and Sell²² were taken on GaP(111) single crystals polished with 0.2 vol % bromine in methanol. In contrast to the comparisons discussed above, our agreement with these reflectance data is excellent over the entire spectral range, including the sensitive E_1 and E_2 peaks. Thus Stokowski and Sell measure R values of 0.345 at 2.76 eV, 0.525 at the E_1 reflectance peak at 3.7 eV, and 0.69 at the E_2 reflectance peak at 5.39 eV. The corresponding R values computed from our ellipsometric data are 0.350, 0.530, and 0.689, and the E_1 and E_2 reflectance peaks occur at the same energies. The results obtained by the two independent methods are therefore entirely consistent. In contrast, a reflectance maximum of only 0.43 was reported by Sobolev and Syrbu,¹¹⁹ but they give no indication as to how the surfaces were prepared. These data are suspect for another reason: The au-

thors report a 180-meV spin-orbit splitting at E_1 . The work of Pikhtin and Yas'kov¹¹² on HCl-HNO₃ etched surfaces is an improvement in this regard, showing a peak R value of 0.525 although it is still well below our calculated value of 0.689. Also, Cardona¹²⁰ reported a R peak of 0.31 at the E_1 transition on samples etched in 1:1 HCl:HNO₃, a relatively low value by present standards.

D. GaAs

Pseudodielectric function results for GaAs from 1.5 to 6.0 eV are shown in Fig. 8 and listed in Table V. Except for the usual compensation for the RAE ratio artifact discussed in Sec. III, these values are as observed, with no corrections for surface overlayers. The self-consistency test shows some problems below 2.0 eV, with a maximum relative discrepancy of 1.5% at 1.5 eV. Above 2.0 eV, the data are consistent to within $\pm 0.3\%$ with a relative precision to within $\pm 0.02\%$.

We consider first the comparison of these data to other optical parameters near 1.5 eV. Because the direct energy gap occurs at 1.425 eV at room temperature,¹²¹ our data do not include any range of transparency. But the absorption threshold is sufficiently weak at 1.5 eV so that transmission data can be taken in spectral regions that overlap our data. Typical absorption coefficient values at 1.5 eV deduced from transmission data are those of Moss and Hawkins¹²² ($0.9 \times 10^4 \text{ cm}^{-1}$), Sturge¹²³ ($1.2 \times 10^4 \text{ cm}^{-1}$), and Sell *et al.*^{124,125} ($\alpha = 1.5 \times 10^4 \text{ cm}^{-1}$). The value of Sturge¹²³ has generally been taken as standard. The RAE ratio correction factor essentially adjusts our absorption coefficient value to agree with that of Sturge, and causes our α values to lie about

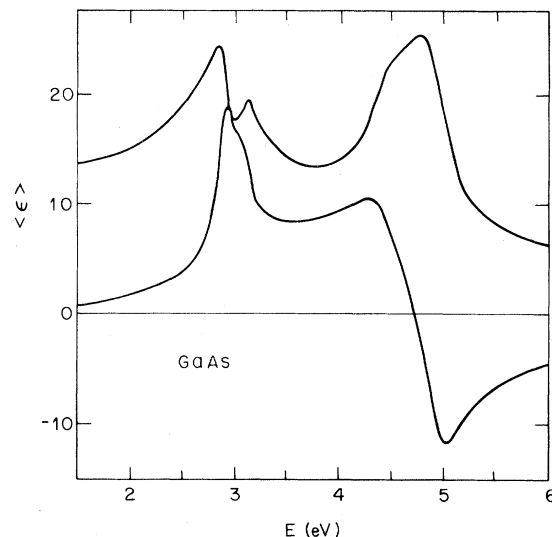


FIG. 8. Pseudodielectric function of crystalline GaAs.

TABLE V. Optical properties of GaAs.

E (eV)	$\langle \epsilon_1 \rangle$	$\langle \epsilon_2 \rangle$	n	k	R	$10^3 \alpha$ (cm $^{-1}$)
1.500	13.435	0.589	3.666	0.080	0.327	12.21
1.600	13.683	0.677	3.700	0.091	0.330	14.83
1.700	13.991	0.838	3.742	0.112	0.335	19.28
1.800	14.307	1.141	3.785	0.151	0.339	27.49
1.900	14.607	1.369	3.826	0.179	0.344	34.45
2.000	14.991	1.637	3.878	0.211	0.349	42.79
2.100	15.463	1.893	3.940	0.240	0.356	51.15
2.200	16.031	2.212	4.013	0.276	0.363	61.46
2.300	16.709	2.622	4.100	0.320	0.372	74.56
2.400	17.547	3.123	4.205	0.371	0.382	90.34
2.500	18.579	3.821	4.333	0.441	0.395	111.74
2.600	19.885	4.841	4.492	0.539	0.410	142.02
2.700	21.550	6.536	4.694	0.696	0.429	190.53
2.800	23.605	9.830	4.959	0.991	0.456	281.33
2.900	22.558	17.383	5.052	1.721	0.490	505.75
3.000	16.536	17.571	4.509	1.948	0.472	592.48
3.100	14.519	18.765	4.373	2.146	0.477	674.17
3.200	10.271	18.022	3.938	2.288	0.468	742.21
3.300	9.086	16.037	3.709	2.162	0.447	723.09
3.400	8.626	14.929	3.596	2.076	0.434	715.28
3.500	8.413	14.216	3.531	2.013	0.425	714.20
3.600	8.355	13.739	3.495	1.965	0.419	717.14
3.700	8.419	13.459	3.485	1.931	0.415	724.14
3.800	8.611	13.365	3.501	1.909	0.414	735.28
3.900	8.890	13.470	3.538	1.904	0.416	752.62
4.000	9.279	13.832	3.601	1.920	0.421	778.65
4.100	9.754	14.538	3.692	1.969	0.430	818.23
4.200	10.235	15.767	3.810	2.069	0.444	880.86
4.300	10.412	17.803	3.939	2.260	0.466	984.86
4.400	9.545	20.582	4.015	2.563	0.494	1143.26
4.500	6.797	22.845	3.913	2.919	0.521	1331.28
4.600	4.163	23.891	3.769	3.169	0.540	1477.66
4.700	1.030	24.835	3.598	3.452	0.565	1644.29
4.800	-3.045	25.196	3.342	3.770	0.596	1834.18
4.900	-8.023	23.393	2.890	4.047	0.633	2009.92
5.000	-11.515	18.563	2.273	4.084	0.668	2069.81
5.100	-11.156	13.677	1.802	3.795	0.676	1961.86
5.200	-9.578	11.143	1.599	3.484	0.661	1836.14
5.300	-8.350	9.758	1.499	3.255	0.644	1748.74
5.400	-7.435	8.806	1.430	3.079	0.628	1685.29
5.500	-6.705	8.123	1.383	2.936	0.613	1636.68
5.600	-6.107	7.593	1.349	2.815	0.599	1597.99
5.700	-5.589	7.182	1.325	2.710	0.584	1565.73
5.800	-5.171	6.882	1.311	2.625	0.571	1543.07
5.900	-4.876	6.587	1.288	2.557	0.562	1528.86
6.000	-4.511	6.250	1.264	2.472	0.550	1503.20

$0.3\text{--}0.5 \times 10^4 \text{ cm}^{-1}$ below those of Sell and Casey¹²⁴ to their limit of 1.8 eV. The latter data were obtained in transmission through $\text{Al}_x\text{Ga}_{1-x}\text{As}$ -GaAs double heterostructures and may be affected by the geometry.

Sell *et al.*¹²¹ also determined refractive index data from 1.2 to 1.8 eV to an estimated accuracy of

0.5%. In fact, these data overlap almost exactly with those of Marple¹²⁶ taken below 1.4 eV with the prism-diffraction method, and therefore can be considered quite accurate. Our values of n are high by about 0.01–0.02 in the (1.5–1.8)-eV region of overlap, and thus fall within the estimated range of error of both experiments. The early data of Oswald and

Schade¹¹⁰ are not given accurately enough for comparison on this scale.

GaAs, like Si, has been a favorite material for monochromatic ellipsometric measurements at 5461 Å, owing to its importance in semiconductor technology and the difficulty of forming stable native oxides. Typical values of n and k for which both are determined independently are as follows: Lukes,¹²⁷ $\tilde{n}=(4.025\pm 0.010)+i(0.030\pm 0.05)$, by air- and toluene-immersion measurements of etched and cleaved surfaces; Dell'Oca *et al.*,¹²⁸ $\tilde{n}=4.00+i0.30$, on peroxide-etched single crystals; Lösche *et al.*,¹¹⁵ $\tilde{n}=4.06+i0.30$, on NaOCl-polished surfaces; Dinges,¹²⁹ $\tilde{n}=4.03+i0.46$, on anodized surfaces after correcting for an interface layer, and Navratil *et al.*,¹³⁰ $\tilde{n}=(3.82\pm 0.01)+i(0.28\pm 0.05)$, on similarly anodized surfaces after correcting for interface roughness. Also, two monochromatic ellipsometric measurements have determined n while assuming $k=0.307$. This value of k comes from Sturge's¹²³ absorption coefficient measurements extrapolated to room temperature. Zaininger and Revesz¹³¹ find $n=3.923$ on chemically etched surfaces, while Adams and Pruniaux¹³² find $n=4.066$, 4.052, and 4.050 for Syton and bromine-methanol polished samples, samples polished with bromine-methanol alone, and epitaxial films.

Our present data, uncorrected for surface layers, show $\tilde{n}=4.073+i0.304$ at 5461 Å. This can be compared to our previous value,²⁶ $\tilde{n}=4.03+i0.35$, calculated by correcting $\langle\epsilon\rangle$ data of GaAs for the presence of a 2-Å overlayer of amorphous As. Similarly, the data of Philipp and Ehrenreich^{3,6-8} show $\tilde{n}=4.05+i0.29$. Perhaps the surprising feature of all these data is their basic agreement. However, it is worth noting that cleaved GaAs surfaces yielded consistently higher values of n at 5461 Å than did chemically etched surfaces,¹²⁷ as was also the case for Si.^{57,64}

Other single-wavelength determinations have been made at 6328 Å by Umeno *et al.*,¹³³ who found $\tilde{n}=3.82+i0.19$ using electroreflectance (sic), and by Moy,⁶⁸ who obtained $3.69+i0.26$ by an immersion approach. Our value of 6328 Å, $3.856+i0.196$, is in good agreement with that of Umeno. Values determined by Adams and Pruniaux¹³² at 4375 Å are $5.096+i1.178$, $5.078+i1.223$, and $5.073+i1.215$ for the same surface preparations described above; our value is $5.05+i1.171$. Except for the results of Moy, who shows similar discrepancies for Si and InP, the monochromatic ellipsometric results are all consistent to within 1%.

We consider next spectral determinations of ϵ and R over the visible-near-uv energy range. Our present values without surface corrections are within

about 5% of our previous values²⁶ that were corrected for 2 Å of amorphous As. Thus our new value of $\langle\epsilon_2\rangle$ at the 4.78-eV E_2 peak is 25.22, compared to our previous value of 23.89 before correction and 24.34 after correction, and our earlier and later "best" values of 24.74 (Ref. 9) and 25.59 (Ref. 10) that were reported in our work on cleaning semiconductor surfaces. Recent ellipsometric data by Burkhard *et al.*¹¹⁷ agree "exactly" (their assessment) with our previous data over the entire (1.5–3.4)-eV range of overlap after correction for an oxide overlayer.

In contrast, the earlier data of Philipp and Ehrenreich³ show clear signs of the presence of inadvertent surface overlayers. The peak reflectance 0.635 at 5.05 eV is close to our calculated value of 0.677, but the Kramers-Kronig value $\epsilon_2=19.4$ at the E_2 peak falls 23% short of our result. Curiously, a number of reflectance spectra have been obtained that yield results in close agreement with the Philipp-Ehrenreich results; Vigil *et al.*¹³⁴ find low values of α near 3.3 eV, and Zanzucchi and Frenchu¹³⁵ find a lower value of R , 0.60, for chemically polished samples used as starting points in an optical degradation study. Early work by Morrison⁴¹ showed an even lower peak value of 0.55 for R , and the data of Tauc and Abraham⁸⁰ at 0.51 are lower yet. Likewise, early measurements of R in the vicinity of the E_1 and $E_1+\Delta_1$ transitions by Lukes and Schmidt⁸⁹ show a peak value of R of 0.408, compared to our calculated value 0.490. The value of ϵ_2 at the E_2 peak determined by Gheorghiu and Théye¹¹⁸ on recrystallized amorphous films was 18, undoubtedly low because of the quality of the recrystallized material.

Sell and MacRae¹³⁶ have reported reflectance data in the vicinity of the E_1 and $E_1+\Delta_1$ transitions in a study of ion-implantation effects. Their measured E_1 peak value of 0.469 is less than our computed value 0.490, even though both occur at the same 2.91 eV energy. More remarkable are the results of Grasso *et al.*,¹³⁷ also obtained in a study of ion-implantation effects. Here, measured E_1 and E_2 reflectance peaks of 0.46 and 0.59, respectively (compared to our values of 0.490 and 0.677), were turned into an ϵ_2 peak of 31 at 4.8 eV. Given the fact that the samples had to be covered with a significant overlayer, this represents a probable error of 50% in the Kramers-Kronig transformation, even though the Ahrenkiel algorithm used¹³⁸ in these calculations is nominally insensitive to extrapolation artifacts.

E. GaSb

Dielectric function data from 1.5 to 6.0 eV are given for GaSb in Fig. 9 and Table VI. After

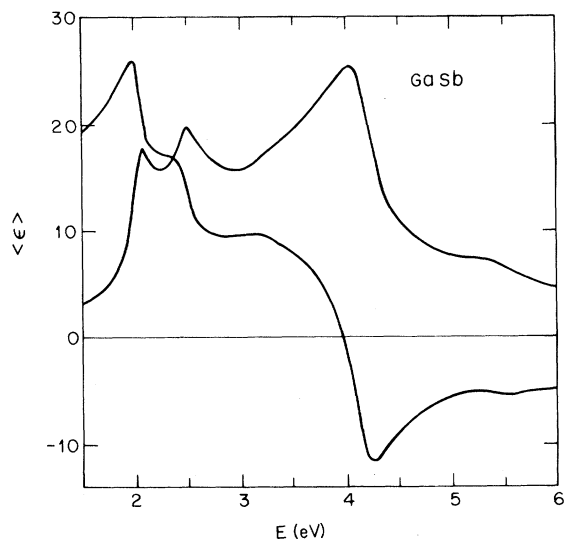


FIG. 9. Pseudodielectric function of crystalline GaSb.

correcting for the RAE ratio artifact, these as-determined spectra are Kramers-Kronig consistent to $\pm 0.5\%$ overall and $\pm 0.2\%$ above 2.5 eV. Scatter indicates a relative precision limit of about $\pm 0.02\%$.

There are almost no previous dielectric function data available for this material, although its differential optical properties have been studied rather thoroughly by modulation spectroscopic techniques. Thus, in the visible–near-uv spectral range, Seraphin and Bennett⁷ list only the reflectance data of Cardona,^{120,139} which reach a maximum value of 0.540 at 4.30 eV. The corresponding value from our data is 0.673 at 4.30 eV. Still lower reflectance values were obtained by Tauc and Abraham⁸⁰; their value for the E_2 peak is 0.36. As seen in Fig. 2, low values are indicative of film-covered surfaces, which is not surprising since GaSb tends to tarnish in air. Lukes and Schmidt⁸⁹ have given reflectance data near E_1 and $E_1 + \Delta_1$ but these spectra are uncalibrated.

We have previously reported^{9,10} $\langle \epsilon_2 \rangle = 25.28$ at the 4.04 E_2 peak, in essential agreement with the result 25.24 obtained here. An earlier ϵ spectrum from 1.5 to 6.0 eV was calculated by us⁴⁷ in a simultaneous solution for the dielectric functions of both oxide and substrate with the use of spectroscopic ellipsometric data from two samples, one with a 30-Å natural oxide and one anodized to a thickness of 2400 Å. The peak ϵ_2 value of 27 obtained there indicates that the data were overcorrected and that the oxide layer was thinner than that actually assumed.

F. InP

As with GaSb, there has been relatively little prior work on InP although there is now a revival of in-

terest in the material due to its position as the endpoint of the technologically relevant quaternary series $\text{In}_x\text{Ga}_{1-x}\text{As}_y\text{P}_{1-y}$. Our results from 1.5 to 6.0 eV are given for InP in Fig. 10 and Table VII. These values, again corrected for the ratio artifact but not for possible overlayers, are Kramers-Kronig consistent to within $\pm 0.4\%$ over the entire range and show a relative midrange scatter of $\pm 0.01\%$.

Cardona's measurements,^{42,120,140} listed in Seraphin and Bennett,⁷ provided until recently the only information on the visible–near-uv optical properties of this material. In the first work⁴² the peak in R was reported to be 0.515, but a higher value of 0.553 was obtained in later measurements,^{7,140} presumably because of improved surface preparation techniques. Our present calculated value of 0.621 at 5.07 eV indicates that the surfaces of Cardona's samples were still not as perfect as they could have been. Further support for this conclusion comes from the recent work of Burkhard *et al.*,¹¹⁷ who report n (but not k) data for liquid-phase epitaxial (LPE) InP films from 1.2 to 3.6 eV. The Burkhard spectroscopic ellipsometric data merge smoothly into the prism-deviation refractive index values of Pettit and Turner,¹⁴¹ lending independent support to the Burkhard *et al.* results, even though they are corrected for an oxide overlayer. These latter data also shows that Cardona's n values are low throughout most of the (1.2–3.6)-eV spectral range. Our n values agree almost exactly (our assessment) with those of Burkhard *et al.*¹¹⁷; for example, our peak value of $n = 4.44$ at 3.06 eV is within 0.3% of the value 4.43 of Burkhard *et al.*, and similar good agreement is seen at all lower energies.

Other recent data are generally in good agreement. The single-wavelength value of

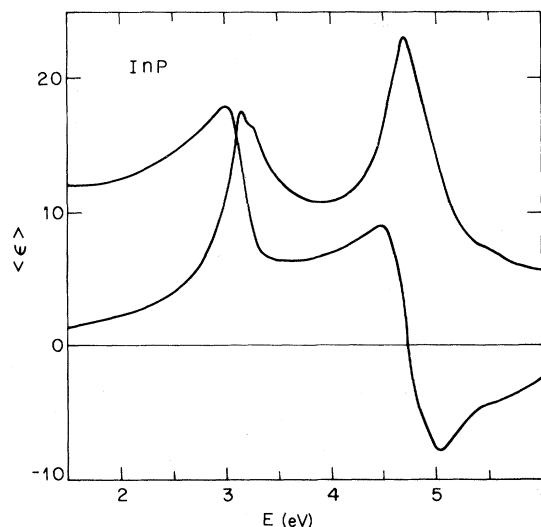


FIG. 10. Pseudodielectric function of crystalline InP.

TABLE VI. Optical properties of GaSb.

E (eV)	$\langle \epsilon_1 \rangle$	$\langle \epsilon_2 \rangle$	n	k	R	$10^3 \alpha$ (cm $^{-1}$)
1.500	19.135	3.023	4.388	0.344	0.398	52.37
1.600	20.137	3.752	4.507	0.416	0.409	67.51
1.700	21.322	4.503	4.643	0.485	0.421	83.56
1.800	22.826	5.889	4.817	0.611	0.437	111.53
1.900	24.836	8.373	5.052	0.829	0.458	159.59
2.000	25.545	14.442	5.239	1.378	0.487	279.43
2.100	18.883	16.963	4.705	1.803	0.474	383.74
2.200	17.386	15.794	4.521	1.747	0.461	389.54
2.300	16.980	16.069	4.492	1.789	0.461	416.97
2.400	16.521	17.708	4.513	1.962	0.473	477.22
2.500	13.367	19.705	4.312	2.285	0.484	579.07
2.600	10.676	18.172	3.984	2.280	0.470	600.94
2.700	9.828	16.966	3.836	2.211	0.457	605.14
2.800	9.484	16.216	3.760	2.157	0.449	612.05
2.900	9.399	15.810	3.728	2.121	0.445	623.34
3.000	9.479	15.738	3.732	2.109	0.444	641.20
3.100	9.628	16.070	3.766	2.134	0.448	670.45
3.200	9.558	16.797	3.800	2.210	0.456	716.82
3.300	9.121	17.658	3.808	2.319	0.465	775.62
3.400	8.490	18.440	3.794	2.430	0.475	837.48
3.500	7.852	19.267	3.785	2.545	0.485	902.86
3.600	7.011	20.306	3.774	2.690	0.497	981.54
3.700	5.853	21.453	3.748	2.862	0.512	1073.44
3.800	4.281	22.719	3.701	3.069	0.530	1182.10
3.900	2.058	24.057	3.620	3.323	0.553	1313.68
4.000	-1.374	25.138	3.450	3.643	0.583	1477.21
4.100	-6.203	24.648	3.100	3.976	0.620	1652.40
4.200	-10.699	20.831	2.522	4.130	0.658	1758.30
4.300	-11.435	15.607	1.989	3.923	0.673	1709.93
4.400	-10.196	12.500	1.723	3.628	0.665	1618.09
4.500	-8.989	10.763	1.586	3.392	0.651	1547.17
4.600	-8.031	9.642	1.503	3.208	0.637	1495.68
4.700	-7.249	8.823	1.444	3.055	0.623	1455.44
4.800	-6.594	8.244	1.408	2.928	0.608	1424.76
4.900	-6.079	7.846	1.387	2.829	0.595	1405.00
5.000	-5.693	7.529	1.369	2.751	0.585	1394.02
5.100	-5.365	7.290	1.358	2.685	0.575	1387.91
5.200	-5.156	7.173	1.356	2.645	0.568	1394.03
5.300	-5.151	7.099	1.345	2.638	0.568	1417.39
5.400	-5.353	6.890	1.299	2.653	0.578	1452.17
5.500	-5.527	6.410	1.212	2.645	0.592	1474.51
5.600	-5.497	5.866	1.127	2.602	0.601	1476.67
5.700	-5.297	5.385	1.062	2.535	0.602	1464.56
5.800	-5.102	5.070	1.022	2.479	0.601	1457.65
5.900	-5.002	4.814	0.985	2.444	0.603	1461.49
6.000	-4.962	4.520	0.935	2.416	0.610	1469.28

$\tilde{n} = 3.65 + i0.38$ at 5461 Å determined by Lösche *et al.*¹¹⁵ on etched samples compares favorably to our calculated value of $3.67 + i0.40$. The value $\tilde{n} = 3.39 + i0.33$ determined by Moy⁶⁸ at 6328 Å is about 4% low with respect to our value

$3.536 + i0.307$ at that wavelength. Our previously reported peak heights of 23.00 obtained in cleaning experiments^{9,10} are in excellent agreement with our present value of 22.98. Our previous results^{25,28} for the InP end point in an investigation of the dielec-

TABLE VII. Optical properties of InP.

E (eV)	$\langle \epsilon_1 \rangle$	$\langle \epsilon_2 \rangle$	n	k	R	$10^3 \alpha$ (cm $^{-1}$)
1.500	11.904	1.400	3.456	0.203	0.305	30.79
1.600	11.972	1.509	3.467	0.218	0.307	35.30
1.700	12.022	1.680	3.476	0.242	0.308	41.64
1.800	12.120	1.889	3.492	0.270	0.310	49.34
1.900	12.284	2.062	3.517	0.293	0.313	56.44
2.000	12.493	2.252	3.549	0.317	0.317	64.32
2.100	12.734	2.488	3.585	0.347	0.322	73.87
2.200	13.026	2.755	3.629	0.380	0.327	84.65
2.300	13.382	3.060	3.682	0.416	0.333	96.89
2.400	13.812	3.425	3.745	0.457	0.341	111.25
2.500	14.313	3.904	3.818	0.511	0.349	129.56
2.600	14.899	4.524	3.903	0.579	0.360	152.71
2.700	15.585	5.337	4.004	0.667	0.372	182.41
2.800	16.365	6.482	4.121	0.786	0.386	223.21
2.900	17.188	8.205	4.256	0.964	0.404	283.32
3.000	17.759	10.962	4.395	1.247	0.427	379.23
3.100	16.483	15.325	4.415	1.735	0.454	545.30
3.200	11.211	17.043	3.976	2.143	0.458	695.23
3.300	7.911	15.797	3.576	2.209	0.446	738.76
3.400	6.639	13.592	3.299	2.060	0.419	709.95
3.500	6.400	12.443	3.193	1.948	0.403	691.21
3.600	6.312	11.731	3.133	1.872	0.391	683.12
3.700	6.330	11.266	3.103	1.816	0.383	680.92
3.800	6.432	10.974	3.095	1.773	0.378	682.96
3.900	6.616	10.841	3.108	1.744	0.376	689.47
4.000	6.874	10.871	3.141	1.730	0.376	701.54
4.100	7.205	11.088	3.196	1.735	0.380	720.91
4.200	7.620	11.539	3.275	1.762	0.387	750.02
4.300	8.119	12.358	3.384	1.826	0.400	795.80
4.400	8.644	13.739	3.527	1.948	0.419	868.69
4.500	8.891	16.161	3.697	2.186	0.449	996.95
4.600	7.484	20.039	3.800	2.637	0.493	1229.49
4.700	2.292	22.948	3.560	3.223	0.543	1535.24
4.800	-3.469	20.989	2.984	3.517	0.577	1711.26
4.900	-5.868	17.894	2.546	3.514	0.591	1745.40
5.000	-7.678	14.896	2.131	3.495	0.613	1771.52
5.100	-7.787	11.483	1.745	3.291	0.620	1701.26
5.200	-6.668	9.399	1.558	3.016	0.601	1589.64
5.300	-5.654	8.308	1.482	2.802	0.577	1505.35
5.400	-4.915	7.717	1.455	2.652	0.554	1451.50
5.500	-4.528	7.308	1.426	2.562	0.542	1428.14
5.600	-4.280	6.832	1.375	2.484	0.534	1410.02
5.700	-3.924	6.317	1.325	2.383	0.522	1376.99
5.800	-3.509	5.924	1.299	2.280	0.504	1340.27
5.900	-3.073	5.680	1.301	2.183	0.483	1305.47
6.000	-2.681	5.644	1.336	2.113	0.461	1285.10

tric properties of LPE quaternary films showed a peak ϵ_2 value of 21.61, but the measurement was performed on an as-grown surface without treatment beyond the removal of the natural oxide. Thus the situation for InP is consistent.

G. InAs

Our results for InAs are given in Fig. 11 and Table VIII. Except for compensation of the RAE ratio artifact, these values are as-observed and are

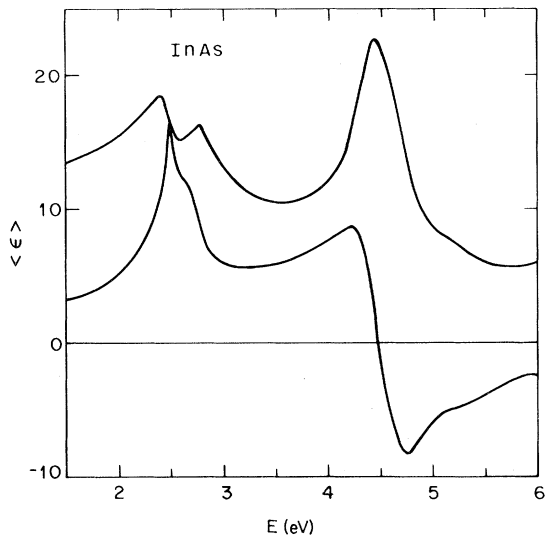


FIG. 11. Pseudodielectric function of crystalline InAs.

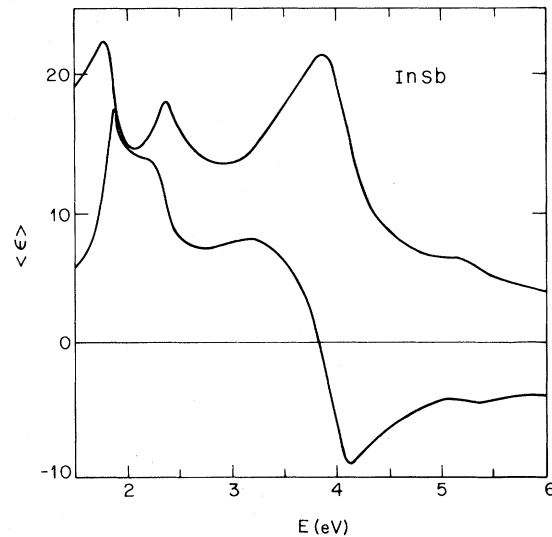


FIG. 12. Pseudodielectric function of crystalline InSb.

Kramers-Kronig consistent below 5.6 eV to a relative accuracy of $\pm 0.3\%$. Relative scatter is about equal to $\pm 0.02\%$.

Of all materials discussed here, the least work has been done on InAs, and all previous ϵ values have been calculated from reflectance. The reflectance values are uniformly low, indicating the presence of overlayers. This is not surprising since no precautions were taken to ensure that surfaces were properly stripped or maintained out of contact with air. Thus, Lukes and Schmidt⁸⁹ find a peak R value of 0.417 in the vicinity of the E_1 and $E_1 + \Delta_1$ transitions, while our calculated value is 0.454. Tauc and Abraham,⁸⁰ Morrison,⁴¹ and Philipp and Ehrenreich³ report R values of 0.49, 0.52, and 0.559 at the E_2 peak; our calculated value is 0.623 at 4.77 eV. The Lukes and Schmidt reflectance data,⁸⁹ taken over the (2.35–2.85)-eV spectral range, show “hyperfine structure,” but in retrospect this can only be due to experimental artifacts. Our previously reported^{9,10} E_2 peak value of $\langle \epsilon_2 \rangle$ is 22.81 at 4.44 eV, in agreement with the present results which show 22.61 at 4.45 eV, and in contrast to the Philipp-Ehrenreich value of 21. Morrison calculated ϵ_1 and ϵ_2 values by extrapolating his (0.1–6.0)-eV measured reflectances with a power-law relationship above 6.0 eV. It is perhaps indicative of the difficulties inherent in the Kramers-Kronig approach that his value of ϵ_2 at the E_2 “peak” is 5.

H. InSb

Our results for InSb are given in Fig. 12 and Table IX. These values, corrected for the RAE ratio artifact but otherwise as determined, are Kramers-

Kronig consistent to $\pm 0.5\%$ overall and $\pm 0.2\%$ from 2.0 to 5.5 eV. The relative scatter is less than $\pm 0.01\%$ over the major fraction of the spectral range.

Previous data for InSb include spectroscopic ellipsometric as well as reflectance and single-wavelength ellipsometric measurements. Our present E_2 peak value of 21.27 at 3.87 eV is somewhat better than our previous values of 20.89 reported in Refs. 9 and 10 and 19.3 reported in Ref. 27. The difference is due to improved cleaning procedures, motivated in part by our previous inability to find a suitable stripping agent to remove the bromine-methanol residue, and by the recent report of a peak value of 22.0 by Bermudez and Ritz.¹⁴² The Bermudez-Ritz data were obtained on a surface cleaved and maintained in ultrahigh vacuum, and therefore are by conventional standards probably most representative of the true bulk response. However, there are some systematic differences whose origins are not interpretable in terms of dielectric overlayers on our samples. Thus from Eq. (5) the differences between $\langle \epsilon_2 \rangle$ spectra should decrease upon going to lower energies from E_2 , crossing zero at about the energy of the E_1 transition. This trend is seen clearly in the difference between our earlier data^{27,29} and the present work. In contrast, the Bermudez-Ritz difference increases to a maximum of about 1.2 at E_1 and $E_1 + \Delta_1$. Thus the difference does not appear to be simply interpretable in terms of dielectric overlayers.

Generally, the discrepancies between the present results and previous reflectance data can be understood in terms of surface overlayers. The peak re-

TABLE VIII. Optical properties of InAs.

E (eV)	$\langle \epsilon_1 \rangle$	$\langle \epsilon_2 \rangle$	n	k	R	$10^3 \alpha$ (cm $^{-1}$)
1.500	13.605	3.209	3.714	0.432	0.337	65.69
1.600	13.884	3.478	3.755	0.463	0.342	75.11
1.700	14.181	3.744	3.798	0.493	0.347	84.94
1.800	14.545	4.083	3.851	0.530	0.353	96.72
1.900	15.015	4.481	3.917	0.572	0.361	110.16
2.000	15.558	5.062	3.995	0.634	0.370	128.43
2.100	16.205	5.820	4.088	0.712	0.380	151.51
2.200	16.957	6.905	4.199	0.822	0.394	183.33
2.300	17.776	8.582	4.331	0.991	0.411	230.98
2.400	18.298	11.458	4.466	1.283	0.433	312.08
2.500	15.856	15.592	4.364	1.786	0.454	452.64
2.600	12.611	15.160	4.021	1.885	0.441	496.84
2.700	11.229	15.766	3.911	2.016	0.445	551.66
2.800	8.276	16.010	3.626	2.208	0.448	626.53
2.900	6.603	14.211	3.337	2.129	0.428	625.87
3.000	6.083	13.003	3.197	2.034	0.412	618.46
3.100	5.831	12.162	3.108	1.957	0.400	614.80
3.200	5.736	11.540	3.051	1.891	0.389	613.30
3.300	5.735	11.082	3.018	1.836	0.381	614.18
3.400	5.820	10.753	3.004	1.790	0.375	616.84
3.500	5.973	10.550	3.008	1.754	0.371	622.13
3.600	6.197	10.471	3.030	1.728	0.370	630.47
3.700	6.478	10.529	3.069	1.715	0.370	643.29
3.800	6.835	10.754	3.129	1.719	0.374	661.95
3.900	7.254	11.187	3.208	1.743	0.382	689.18
4.000	7.744	11.919	3.313	1.799	0.393	729.23
4.100	8.273	13.130	3.449	1.903	0.411	791.03
4.200	8.663	15.173	3.615	2.099	0.437	893.42
4.300	8.000	18.639	3.761	2.478	0.478	1080.14
4.400	4.024	22.171	3.644	3.042	0.527	1356.76
4.500	-1.663	22.006	3.194	3.445	0.566	1571.19
4.600	-5.509	19.372	2.705	3.581	0.593	1669.74
4.700	-7.921	15.762	2.205	3.575	0.617	1703.11
4.800	-7.961	12.077	1.803	3.349	0.622	1629.16
4.900	-6.905	9.909	1.608	3.081	0.605	1530.10
5.000	-5.923	8.752	1.524	2.871	0.583	1455.26
5.100	-5.264	8.107	1.484	2.732	0.565	1412.38
5.200	-4.942	7.600	1.436	2.646	0.556	1394.86
5.300	-4.665	6.980	1.366	2.555	0.550	1372.78
5.400	-4.278	6.425	1.312	2.449	0.537	1340.56
5.500	-3.851	6.008	1.282	2.344	0.521	1306.62
5.600	-3.424	5.738	1.276	2.248	0.501	1275.94
5.700	-3.006	5.595	1.293	2.163	0.479	1249.73
5.800	-2.642	5.602	1.333	2.102	0.459	1235.70
5.900	-2.430	5.764	1.383	2.084	0.448	1246.25
6.000	-2.403	6.055	1.434	2.112	0.448	1284.15

flectance value reported by Philipp and Ehrenreich,^{3,6-8} 0.579, falls short of our calculated value 0.636. This leads to a substantial reduction of the E_2 peak of ϵ_2 from 21.26 to 17.5. Earlier data are in general less satisfactory. Thus, Tauc and Abraham⁸⁰ find a reflectance peak 0.50, while Morrison⁴¹

reports 0.49 and Kurdani¹⁴³ finds a local maximum of 0.37. The data of Lukes and Schmidt⁸⁹ are not normalized, so no comparison there is possible.

A number of single-wavelength null-ellipsometric values of \tilde{n} have also been reported. Lösckke *et al.*¹¹⁵ find an average value of $3.90 + i2.07$ at

TABLE IX. Optical properties of InSb.

E (eV)	$\langle \epsilon_1 \rangle$	$\langle \epsilon_2 \rangle$	n	k	R	$10^3 \alpha$ (cm $^{-1}$)
1.500	19.105	5.683	4.418	0.643	0.406	97.79
1.600	20.302	6.838	4.568	0.749	0.421	121.39
1.700	21.699	9.019	4.754	0.949	0.441	163.46
1.800	22.148	13.707	4.909	1.396	0.467	254.73
1.900	16.144	16.603	4.433	1.873	0.463	360.65
2.000	14.448	14.875	4.194	1.773	0.443	359.46
2.100	13.974	14.643	4.136	1.770	0.439	376.79
2.200	13.674	15.302	4.135	1.850	0.445	412.62
2.300	12.653	16.936	4.111	2.060	0.458	480.25
2.400	9.377	17.480	3.822	2.287	0.463	556.30
2.500	7.811	15.856	3.570	2.221	0.447	562.77
2.600	7.278	14.787	3.447	2.145	0.434	565.33
2.700	7.069	14.069	3.377	2.083	0.425	570.01
2.800	7.044	13.617	3.345	2.036	0.419	577.71
2.900	7.150	13.395	3.342	2.004	0.415	589.12
3.000	7.354	13.421	3.366	1.994	0.416	606.27
3.100	7.627	13.779	3.419	2.015	0.420	633.20
3.200	7.742	14.572	3.482	2.093	0.431	678.77
3.300	7.507	15.631	3.525	2.217	0.445	741.69
3.400	6.782	16.678	3.520	2.369	0.459	816.31
3.500	5.995	17.673	3.511	2.517	0.474	892.82
3.600	4.830	18.854	3.485	2.705	0.492	987.01
3.700	3.147	20.102	3.427	2.933	0.514	1099.83
3.800	0.534	21.064	3.287	3.204	0.541	1234.25
3.900	-2.838	21.177	3.044	3.479	0.572	1375.21
4.000	-6.722	19.443	2.632	3.694	0.608	1497.79
4.100	-8.911	15.595	2.127	3.666	0.633	1523.33
4.200	-8.580	12.296	1.791	3.433	0.634	1461.59
4.300	-7.678	10.382	1.618	3.209	0.623	1398.45
4.400	-6.910	9.191	1.515	3.034	0.610	1353.08
4.500	-6.297	8.351	1.443	2.894	0.598	1320.24
4.600	-5.788	7.690	1.385	2.776	0.586	1294.36
4.700	-5.324	7.160	1.341	2.669	0.574	1271.51
4.800	-4.912	6.761	1.312	2.576	0.562	1253.17
4.900	-4.534	6.492	1.301	2.495	0.548	1239.36
5.000	-4.250	6.378	1.307	2.441	0.537	1237.01
5.100	-4.190	6.360	1.309	2.430	0.534	1255.97
5.200	-4.359	6.207	1.270	2.444	0.543	1288.04
5.300	-4.505	5.815	1.194	2.435	0.556	1308.25
5.400	-4.487	5.345	1.116	2.394	0.563	1310.58
5.500	-4.325	4.931	1.057	2.333	0.563	1300.55
5.600	-4.126	4.664	1.025	2.275	0.558	1291.44
5.700	-3.995	4.470	1.000	2.235	0.555	1291.22
5.800	-3.945	4.282	0.969	2.210	0.558	1299.21
5.900	-3.925	4.029	0.922	2.185	0.565	1306.73
6.000	-3.835	3.681	0.861	2.139	0.572	1300.85

5461 Å, compared to our value of $4.13+i1.98$. Again, the difference is consistent with the presence of an overlayer on the surface of their sample. Saxena⁴⁵ found 5461-Å values of $4.104+i2.058$, in reasonable agreement with our data. Syoseva and Ayupov¹⁴⁴ performed immersion measurements at

5461 Å on InSb surfaces in equilibrium with 20-ml 0.045% HCl, 5-ml 15% citric acid, and 10-ml 3% H₂O₂. Their results $(4.17 \pm 0.04) + i(1.91 \pm 0.04)$ for $\langle 100 \rangle$, $(4.06 \pm 0.06) + i(1.92 \pm 0.03)$ for $\langle 111 \rangle$, and $(4.12 \pm 0.06) + i(1.91 \pm 0.03)$ for $\langle 211 \rangle$ surfaces, are to within experimental uncertainties in agreement

with our values. Results obtained by Harkness and Young¹⁴⁵ from variations in (ψ, Δ) plots an oxide-covered samples yielded in anomalous value $4.6 + i1.6$, but this is not surprising in view of the poor optical quality of the native oxide on InSb.²⁷

One measurement, $\tilde{n} = 3.8 + i2.25$, has also been reported¹⁴⁶ at 6.328 Å. The comparison with our value $4.252 + i1.900$ shows the effects of a surface overlayer.

V. CONCLUSION

In this paper we have presented pseudodielectric function data and calculated optical parameters of carefully prepared and maintained single crystals of Si, Ge, and the major III-V binary semiconductor compounds. With the exception of low-energy $\langle \epsilon_2 \rangle$ values of Si and GaP, these results are as measured and uncorrected for the presence of surface overlayers. Rather than attempt an analytical solution to the overlayer problem, we have put our efforts into minimizing the thickness of surface transition regions, taking advantage of the "biggest-is-best" rule¹³ of assessing surface quality by spectroscopic ellipsometry.

Because surface layers are unavoidable, and because we have not tried all possible ways of minimizing their effect, further improvements in these results are certainly possible. However, we have observed that the data presented here are limiting values in the sense that they are approached very closely, but not exceeded, by a variety of alternative cleaning techniques. This is indicative of the fact that these $\langle \epsilon \rangle$ data are approaching the intrinsic bulk values. Consequently, we do not anticipate

significant further changes, certainly no more than 5% and probably no more than 1 or 2%.

We mention also that there is now quite good agreement between the various optical functions measured by different techniques by different workers as long as the samples are prepared with techniques such as chemical etching that can provide smooth, undamaged surfaces. Examples include the reflectance measurements of Donovan *et al.*²¹ on Ge and Stokowski and Sell²² on GaAs, and the immersion null-ellipsometric measurements of Syoseva and Ayupov¹⁴⁴ on InSb, among others. And in general, the refractive index data for GaP, GaAs, and InP merge smoothly with the highly accurate corresponding values determined in transmission through prisms. Consequently, future work is likely to take the direction of preparing similar high-quality surfaces in ultrahigh vacuum to extend the present results into the vacuum ultraviolet spectral range and to other temperatures.

One further comment is in order. In many optical measurements, such as studies of oxidation, corrosion, or damage effects, the quantity of importance is the change of optical response after treatment rather than the accurate values themselves. Owing to the large number of effects that can adversely affect the smoothness, cleanliness, and atomic perfection of real samples, it is not possible in general to assume that the data presented here will accurately represent samples in their initial states. While our data will be useful in assessing the initial quality of such samples, they are no substitute for actual measurements even if the materials and surface orientations are identical and the resultant optical data "should be the same."

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