Simultaneous detection of optical constants ϵ_1 and ϵ_2 by Brewster angle reflectivity measurements

N. Dietz and H. J. Lewerenz
Hahn-Meitner-Institut, Bereich Photochemische Energieumwandlung, Postfach 39 01 28, Glienicker Strasse 100, 1000 Berlin 39, Germany

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A new method for determination of the dielectric function ϵ is presented. The experiment is based on the simultaneous measurement of the Brewster angle φ_B and the reflectivity R_p for light polarized parallel to the plane of incidence. ϵ_1 and ϵ_2 as a function φ_B and R_p were calculated and the results plotted as contour plots with the optical constants as parameters. Spectral measurements yield ϵ_1 and ϵ_2 as a function of photon energy. Results obtained on GaAs are evaluated and correspond well to literature data.

The optical properties of semiconductors are usually determined by various types of reflection spectroscopy.[1-5] The evaluation procedure involves Kramers-Kronig analysis with the necessity of introducing appropriate low and high energy tail functions.[1] The validity of the chosen tail function is then to be verified in a self-contained cycle.[6] The possibility to simultaneously determine the optical function ϵ_1 and ϵ_2 at a given wavelength has been recognized quite early. Although the basic mathematical development has existed for a few decades,[7] the experimental development is still to be realized. Led by investigations on deep levels in semiconductors,[8-11] we developed an experimental procedure based on the measurement of the Brewster angle and the reflectivity R_p (for p-polarized light). It turned out that on semiconductors the procedure allows accurate determination of φ_B and R_p and thus of ϵ_1 and ϵ_2 as will be shown below. Starting from the Fresnel equation

$$R_p=r_p r_p^* =\frac{\mu+|\epsilon|^2\cos^2\varphi-\cos\varphi(\mu+\sin^2\varphi)\sqrt{2(\mu+\kappa)}}{\mu+|\epsilon|^2\cos^2\varphi+\cos\varphi(\mu+\sin^2\varphi)\sqrt{2(\mu+\kappa)}}, \quad (1)$$

with $\mu^2=|\epsilon|^2-2\epsilon_1\sin^2\varphi+\sin^4\varphi$ and $\kappa=\epsilon_1-\sin^2\varphi$, the analytical expression for the Brewster angle (generally the minima of the reflectivity R_p) can be developed as an extrema condition[7] from Eq. (1). The third-order equation

$$y^3+\frac{(|\epsilon|^4-3|\epsilon|^2)}{(2|\epsilon|^2+2\epsilon_1)}y^2-\frac{|\epsilon|^4}{(|\epsilon|^2+\epsilon_1)}y+a=0, \quad (2)$$

with $y=\sin^2\varphi_B$ and $a=|\epsilon|^4/(2|\epsilon|^2+2\epsilon_1)$ can be solved analytically by verification of the three solutions.[12] For various given complex dielectric functions the Brewster angle and the reflection minima R_p were calculated. It is simple to verify that only the positive solution can be used. The Brewster angle for a complex dielectric function ϵ is therefore given by

$$\varphi_B=\arcsin\left\{\frac{-|\epsilon|^2}{3(|\epsilon|^2+\epsilon_1)}\times\left[|\epsilon|^2-3+\cos\left(\frac{\chi}{3}+\frac{4\pi}{3}\right)\sqrt{b}\right]\right\}^{1/2}, \quad (3)$$

with

$$\cos\chi=|\epsilon|^4c/\sqrt{b^3};\quad b=|\epsilon|^4+6|\epsilon|^2+12\epsilon_1+9,$$
$$c=|\epsilon|^8+9|\epsilon|^6+27|\epsilon|^4+18|\epsilon|^4\epsilon_1-27|\epsilon|^2+54|\epsilon|^2\epsilon_1+54\epsilon_1^2. \quad (4)$$

For a minute imaginary part of the dielectric function ϵ, Eq. (3) can be reduced to the well-known relation $\varphi_B=\arctan(\sqrt{\epsilon})$.

The solution of a reduced fourth-order equation yields an analytical expression for the dielectric constants ϵ_1 and ϵ_2 as a function of the measured Brewster angle φ_B and the reflectivity $R_p|\varphi_B$. This can be obtained by rearranging Eq. (2) with respect to Eq. (1) in a similar way as shown by Humphreys-Owen:[7]

$$\mathbf{g}^4+\mathbf{u}\mathbf{g}^2+\mathbf{v}\mathbf{g}+\mathbf{w}=0, \quad (5)$$

where

$$\mathbf{g}=|\epsilon|^2-z^2/4(4x^2+1);\ y=\sin\varphi_B;\quad x=\cos\varphi_B;$$
$$z=y^2/x^2,\quad P^2=[(1-R_p|\varphi_B)/(1+R_p|\varphi_B)]^2;$$
$$\mathbf{u}=z^3[4y^2x^2+P^2(1+y^2)^2]-3/4z^4(4x^2+1)^2;$$
$$\mathbf{v}=2y^2x^2P^2(1+y^2)-z^6/8(4x^2+1)^3-z^5/2(4x^2+1)[(4y^2x^2+P^2(1+y^2)^2];$$

and

$$\mathbf{w}=y^4x^5P^2+3/256z^8(4x^2+1)^4-z^7/16(4x^2+1)^2[(4y^2x^2+P^2(1+y^2)]-y^2z^6P^2/2(4x^2+1)(1+y^2).$$

It is possible to prove that only the positive part of the complex cubic resolvent of Eq. (5) yields a physically sensible solution with respect to $|\epsilon|$. The explicit solution will

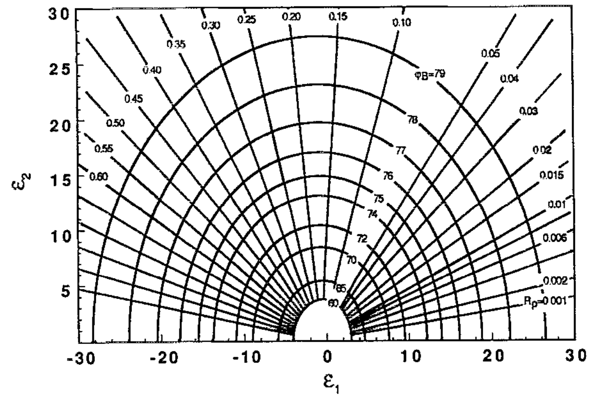

FIG. 1. Families of iso-$R_p|\varphi_B$ contours in the ϵ_1–ϵ_2 plane. $R_p|\varphi_B$ is the reflectivity at the Brewster angle φ_B for light, polarized parallel to the plane of incidence with assumed values from 0.001 to 0.85. φ_B is the Brewster angle with assumed values from 60° to 79°. This nomogram represented a part of the analytical solution of Eq. (5), solving the $(R_p|\varphi_B, \varphi_B) \rightarrow (\epsilon_1, \epsilon_2)$ problem.

be given elsewhere.[13] The real and imaginary part of the dielectric function are given by

$$\epsilon_1 = |\epsilon|^2(1+2\cos^2\varphi_B - |\epsilon|^2\cot^2\varphi_B)/\sin^2\varphi_B$$

and

$$\epsilon_2 = \sqrt{|\epsilon|^2 - \epsilon_1^2}$$

Figure 1 shows a contour plot of ϵ_2 vs ϵ_1 for various values of φ_B and R_p calculated according to Eq. (5). The method allows accurate determination of ϵ_1 and ϵ_2 as a function of the Brewster angle, measured by angular detection of the minimum of the reflected intensity and R_p at φ_B. The calculation $(\epsilon_1,\epsilon_2) = f(\varphi_B, R_p|\varphi_B)$ can be carried out analytically for each Brewster angle and absorption strength and is only limited by the experimental detection of the reflection minima at high absorption.

The most sensitive energy range of the method is that, where the reflectivity R_p is less than 10^{-3}, here, materials exhibit a pronounced minimum in R_p at the Brewster angle. For direct-gap semiconductors, this corresponds to $h\nu \Leftarrow E_g$. The method is therefore also particularly suited for the determination of optical constants in amorphous and/or highly compensated semiconductor material.

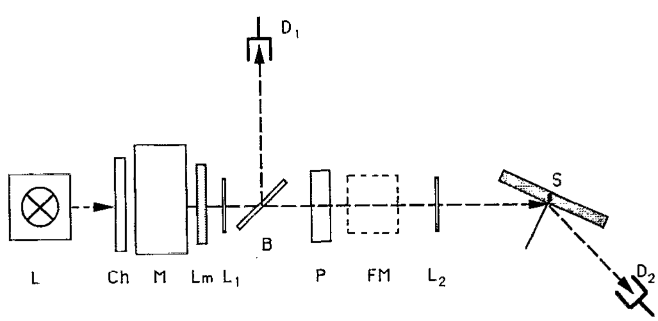

FIG. 2. Schematic diagram of the experimental setup; L: lamp; Ch: chopper; M: monochromator; L_m: achromatic lense system; L_1, L_2: slits; B: beamsplitter; P: polarizer; FM: Faraday modulator; D_1, D_2: detectors; S: sample.

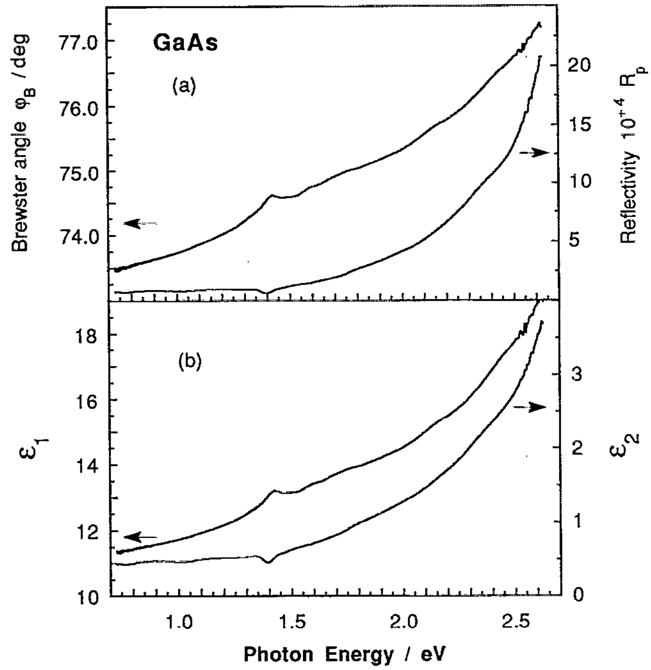

FIG. 3. (a) The measured spectral dependence of the Brewster angle, φ_B and the reflectivity R_p at φ_B for GaAs. (b) The real and imaginary part of the dielectric function, ϵ_1 and ϵ_2, are calculated using Eq. (5).

The experimental principle is shown in Fig. 2 for a mirror-type specularly reflecting sample. A tungsten iodine lamp as light source with a Kratos monochromator was used. The light beam is split into a reference and a signal channel, detected at D_1 and D_2, respectively. The signal beam is polarized parallel to the plane of incidence, using a Glan-Thompson polarizer P. The polarized light is focused onto the sample held at an angle φ close to the Brewster angle φ_B. The reflected intensity is detected by a cooled Si (0.4–1 μm) or Ge detector (0.8–1.7 μm). For analysis of the reflected intensity and the Brewster angle position, the signal at D_2 was measured as a function of the angle φ. The minimum was determined by a least-square fit and the according reflectivity R_p was determined by comparison with

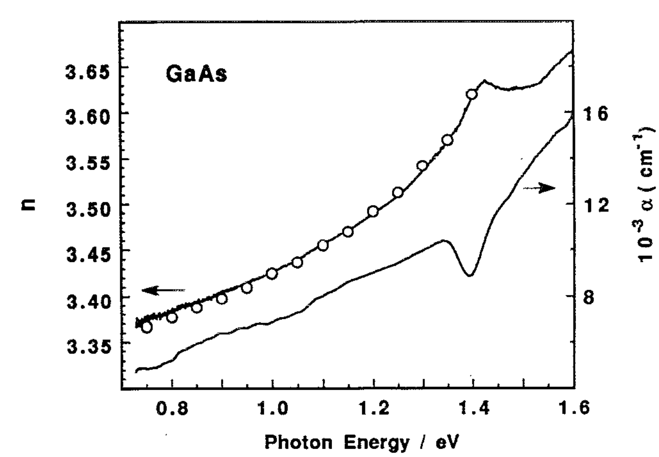

FIG. 4. The refractive index n compared with literature data (open circle) and the absorption coefficient α in the transparent region of GaAs.

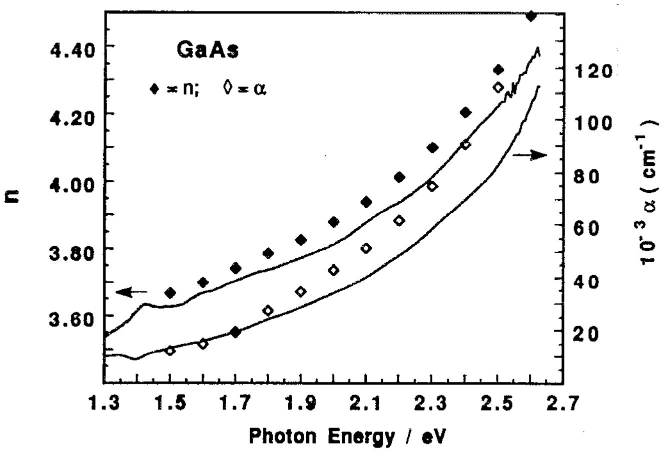

FIG. 5. The spectral dependence of the refractive index n and the absorption coefficient α calculated from the measured Brewster angle φ_B and the reflectivity R_p at φ_B compared with the values (♦$=n$; ◊$=\alpha$) determined by ellipsometry.

the signal at detector D_1. Standard lock-in technique was used for data acquisition. The accuracy of the method depends critically on the angular resolution of the goniometer table on which the sample is mounted. The plane of incidence is adjusted using Faraday modulation. The mechanical specification yields a resolution better than 2×10^{-3} deg. The step motor limitation results in a resolution of 4×10^{-3} deg.

Figure 3(a) shows—for high pressure Bridgmann grown GaAs(100)—φ_B and R_p as a function of photon energy. The real and imaginary part of the dielectric function ϵ_1 and ϵ_2, deduced from the experimental values of φ_B and R_p, were calculated. The results are shown in Fig. 3(b). For the transparent region from 0.7 to 1.5 eV, the calculated refractive index n and the absorption coefficient α are shown in Figs. 4 and 5, and compared with literature values.

For the low energy region in Fig. 4 the refractive index data obtained with the prism-diffraction method[14] (open circles) show excellent agreement with an accuracy of $<\pm0.1\%$, thus demonstrating the outstanding capabilities of the new method.

Figure 5 shows our calculated data n and α above the energy gap, compared with the ellipsometric values obtained by Aspnes.[15] The data given by Aspnes (without surface correction) agree within the given 5% error range for the refractive index n. For the absorption coefficient a much larger difference of up to 25% at 2.5 eV occurs, which cannot be explained by statistical errors. An assumed error of $\pm10\%$ in the reflectivity $R_p|\varphi_B$ (roughness and/or surface contamination) shows differences in $n=n^*(1\pm10^{-4})$ and in $\alpha=\alpha^*(1\pm5\times10^{-2})$. The influence of surface layers on our uncorrected data is presumably low and will be discussed elsewhere in more detail.[13]

The good agreement in the transparent energy region of GaAs with literature data supports the use of the new method if the inherent limitations are considered. The data clearly reveal absorption centers within the energy gap of GaAs and show that the new method allows the identification of defect levels in semiconductors and insulators at room temperature.

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