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An alternative of spectroscopic ellipsometry: The double-reference method

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We have developed a method conceptually different from ellipsometric techniques which allows the determination of the complex refractive index by simultaneously measuring the unpolarized normal-incidence reflectivity relative to the vacuum and another reference media such as diamond, GaAs, CdTe, etc. From these two quantities, the complex optical response can be directly obtained without Kramers–Kronig transformation. Due to its transparency and large refractive index from the far-infrared to the soft ultraviolet regions, diamond can be ideally used as a second reference over the whole optical spectrum. The experimental arrangement is rather simple compared to ellipsometry. © 2008 American Institute of Physics. [DOI: 10.1063/1.2904623]

Determination of the complex dielectric response of a material is an everlasting problem in optical spectroscopy. Depending on the basic optical properties, whether the sample is transparent or has strong absorption in the photon-energy range of interest, its absolute reflectivity or transmittance is usually detected with normal incidence. Both quantities are related to the intensity of the light and give no information about the phase change during either reflection or transmission. Consequently, the phase shift is generally determined by Kramers–Kronig (KK) transformation in order to obtain the complex dielectric response. However, for the proper KK analysis the reflectivity or transmittance spectrum has to be measured in a broad energy range, ideally over the whole electromagnetic spectrum.

On the other hand, there exist ellipsometric methods^{1,2} capable of simultaneously detecting both the intensity and the phase of the light reflected back or transmitted through a media. The most state-of-the-art one is the time domain spectroscopy but its applicability is mostly restricted to the far-infrared region.^{3,4} Another class of ellipsometric techniques, sufficient for broadband spectroscopy, requires polarization-selective detection of light.^{1,2} (In the following, we will discuss experimental situations in reflection geometry though most of the considerations are valid for transmission as well.) A representative example is the so-called rotating-analyzer ellipsometry (RAE) when the reflectivity is measured at a finite angle of incidence, usually in the vicinity of the Brewster angle.^{1,2} Under this condition the Fresnel coefficients are different for polarization parallel (*p* wave) and perpendicular (*s* wave) to the plane of incidence and the initially linearly polarized light becomes elliptically polarized upon the reflection. By rotating the analyzer, the ellipsometric parameters, i.e., the phase difference and the intensity ratio for the *p*- and *s*-wave components,⁵ are measured and the complex refractive index can be directly obtained.

As an alternative of ellipsometry, we describe a concept, hereafter referred to as double-reference spectroscopy (DRS). It offers a simple way to obtain the complex dielectric function without KK transformation by measuring the unpolarized normal-incidence reflectivity of the sample relative to two transparent reference media. In addition to the

absolute reflectivity, i.e., that of the vacuum-sample interface, we can take advantage from the excellent optical properties of diamond and use it as a second reference. High-quality diamonds such as type IIA optical diamonds are transparent from the far-infrared up to the ultraviolet photon-energy region,⁷ except for the multiphonon absorption bands located at $\omega=0.19\text{--}0.34\text{ eV}$.⁹ Moreover, they have a large refractive index $n_d\approx 2.4$ which shows only 10% energy dependence up to $\omega\approx 5\text{ eV}$.^{6–9}

The significant difference between the refractive index of the two reference media is a crucial point of the method. Thus, optically well-characterized semiconductors with large refractive index, such as Si,¹⁰ GaAs,¹¹ and CdTe,¹² can provide an even better performance for a limited range of energy, typically below $\omega\approx 1\text{ eV}$. Since these materials are popular substrates for crystal growing, DRS can be carried out by the successive measurement of the two sides of the samples.

Since the experimental arrangement is much simpler than that of any ellipsometric technique, this method may find broad field of application, especially when only a narrow spectral range is of interest. A representative example is the class of strongly correlated electron systems when the optical properties are very sensitive to the low-energy excitations. In such a case, due to the nonlocal feature of the KK transformation, the complex dielectric response strongly depends on the low-energy extrapolation of the reflectivity spectrum and it is reliable in a range considerably narrower than that of the measured reflectivity.

In the following, we describe the principles of the DRS and demonstrate its efficiency in comparison with the RAE. The essence of the method is the measurement of the sample reflectivity relative to two media with strongly different dielectric properties, such as vacuum and diamond. For normal incidence the Fresnel equations for the two interfaces have the following form:

$$R_{vs} \equiv R_s = \left| \frac{\hat{n}_s - 1}{\hat{n}_s + 1} \right|^2 \quad \text{and} \quad R_{ds} = \left| \frac{\hat{n}_s - n_d}{\hat{n}_s + n_d} \right|^2,$$

where $\hat{n}_s = n_s + ik_s$ denote the complex index of refraction and $\hat{n}_d(\omega)$ is well documented in the literature for type IIA diamonds.^{6–9} Although the Fresnel equations are highly non-

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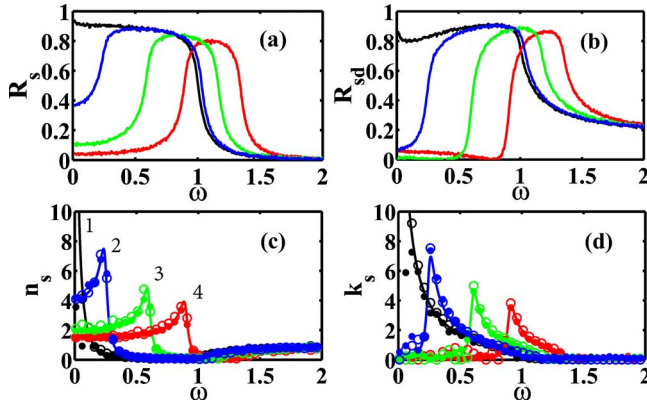


FIG. 1. (Color online) Panels (a) and (b): normal-incidence reflectivity spectra of the vacuum-sample (R_{vs}) and diamond-sample (R_{ds}) interfaces, as calculated from the dielectric function $\hat{\epsilon}_s(\omega) = 1 + (\omega_0^2 - \omega^2 - i\omega\gamma)^{-1}$ for $\omega_0 = 0, 0.25, 0.6, 0.9$, and $\gamma = 0.1$ with the plasma frequency set to unity. Gaussian noise with $\Delta R = \pm 0.005$ standard deviation is introduced to both $R_{vs}(\omega)$ and $R_{ds}(\omega)$. Panels (c) and (d): the refractive index (n_s) and the extinction coefficient (k_s) as obtained from the above reflectivities using the DRS (closed circles) and the RAE (open circles) approaches. The complex refractive index free of noise is indicated by full lines.

linear, \hat{n}_s can be easily expressed in the lack of absorption within the diamond, i.e., for $k_d \equiv 0$,

$$n_s = \frac{1}{2}(n_d^2 - 1) \left(n_d \frac{1 + R_{ds}}{1 - R_{ds}} - \frac{1 + R_s}{1 - R_s} \right)^{-1}, \quad (1)$$

$$k_s = \left(-n_s^2 + 2 \frac{1 + R_s}{1 - R_s} n_s - 1 \right)^{1/2}. \quad (2)$$

We show the effectiveness of the method using the model dielectric function $\hat{\epsilon}_s(\omega) = 1 + (\omega_0^2 - \omega^2 - i\omega\gamma)^{-1}$, where ω_0 is the resonance frequency and γ is the damping of the oscillator. From $\hat{\epsilon}_s(\omega)$ we evaluate both $R_s(\omega)$ and $R_{ds}(\omega)$ by the Fresnel equations while in case of RAE the intensity is calculated for three different orientations of the analyzer.⁵ The resonance frequency is varied in a way that the reflectivity spectra, plotted in the upper panels of Fig. 1, describe both insulating and metallic behaviors, corresponding to $\omega_0 = 0.9, 0.6$, and 0.25 , and $\omega_0 = 0$, respectively. The oscillatory strength common for each is implicitly included since the plasma frequency is chosen as the unit of the energy scale. The typical noise of the detection and the finite energy resolution are taken into account as Gaussian noise superimposed on the intensities with standard deviation of $\Delta R = \pm 0.005$. Furthermore, systematic errors coming from the imperfect experimental conditions, such as sample surface roughness and nonplanarity and misalignment of the light path, are represented as 1° deviation in the angle of incidence for the both cases.

From the given reflectivity spectra the complex refractive index is calculated by the double-reference method using Eqs. (1) and (2) and also by following the more complicated evaluation of RAE. The real and imaginary parts of the respective $\hat{n}_s(\omega)$ spectra are shown in the lower panels of Fig. 1. The exact spectra $\hat{n}_s(\omega) = \sqrt{\hat{\epsilon}_s(\omega)}$ free of experimental errors are also shown for comparison. The precision of the two methods seems comparable.

To classify the range of applicability, the error maps for the two techniques are analyzed in more detail over the plane of the complex refractive index. As Fig. 2 shows, the overall

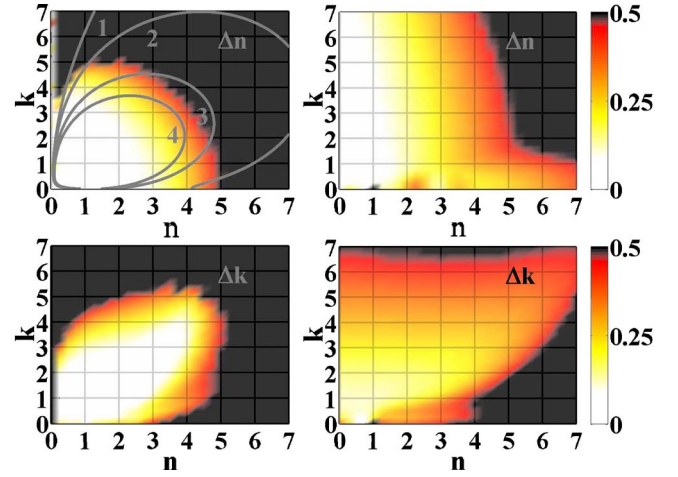


FIG. 2. (Color online) Color map of the absolute error of the complex refractive index, as calculated by the DRS (left panels) and the RAE (right panels). The upper and lower panels show the error for the refractive index (Δn_s) and the extinction coefficient (Δk_s), respectively. Full lines with labels correspond to the spectra shown in Fig. 1.

confidence level of the DRS surpasses that of the RAE, especially in case of the extinction coefficient k_s . Furthermore, while the error map for the real and imaginary parts of the refractive index behaves similarly in case of the DRS, the RAE is optimal for the two components in rather distinct regions of the n_s - k_s plane. Although for RAE, the area of applicability is seemingly more extended for the real part of the refractive index in the limit of $k_s \gg n_s$, the extinction coefficient dominating the optical response in this strongly absorbing region exhibits a high error level.

In situations where $|\hat{n}_s| \gg 1$, such as strong resonances or good metals with large extinction coefficient ($k_s > n_s \gg 1$), the Fresnel equations are not numerically independent since the reflection coefficients converge to the unity irrespective of the polarization state, the angle of incidence or the reference media. In this limit, both approaches fail, which is general for any ellipsometry. For the DRS, it means that the difference between the two reference media disappears as $|\hat{n}_s| \gg n_d$. With a realistic noise level specified above, the DRS works with less than $\sim 10\%$ error until the refractive indexes are twice as large as that of the diamond, i.e., almost in the whole range of $|\hat{n}_s| \leq 5$. It is to be emphasized that the large difference in the refractive index of the two reference media highly extends the applicability range of the method and reduces the numerical errors.

Next, we describe a simple procedure for the measurement of R_{ds} applying a wedged diamond piece, as sketched in Fig. 3.¹³ The intensity reflected back from the vacuum-diamond and diamond-sample interfaces (I_{vd} and I_{ds} , respectively) can be detected separately by a few degree rotation; a wedging angle of 2° causes $\sim 10^\circ$ angular deviation between the two reflected beams. Since nearly normal incidence can still be considered for both positions, the reflectivity of the sample relative to the diamond is obtained from the measured intensities as¹³

$$R_{ds}(\omega) = \frac{R_{vd}(\omega)}{[1 - R_{vd}(\omega)]^2} \frac{I_{ds}(\omega)}{I_{vd}(\omega)}, \quad (3)$$

where $R_{vd} (\equiv R_d)$ is the absolute reflectivity of the diamond. The $R_{vd}(\omega)/[1 - R_{vd}(\omega)]^2$ prefactor can be either calculated using the well-documented n_d ,⁶⁻⁹ or checked experimentally

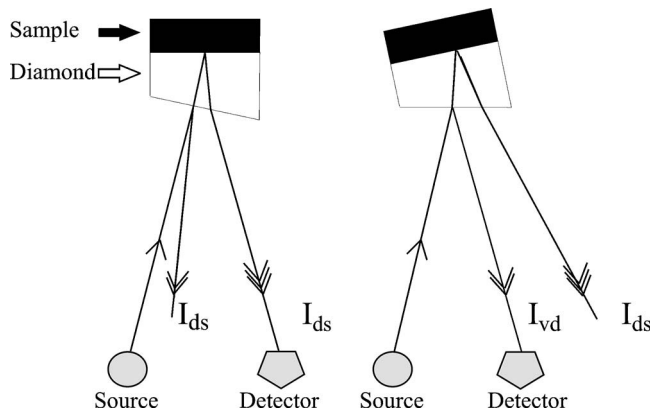


FIG. 3. Experimental condition for the measurement of R_{ds} . Reflection from the vacuum-diamond and diamond-sample interfaces are indicated. Wedging of the window avoids multiple reflections within the diamond and allows for a clean separation of the reflections from the two interfaces and, thus, facilitates reference measurements.

using a standard reference mirror to obtain the absolute value of R_{vd} . The high-energy limit of this method is mainly determined by the roughness of the diamond-sample interface δ_{ds} . Therefore, special care should be taken for the proper matching between the diamond and the sample in order to eliminate interference and diffraction effects inherently appearing for wavelength shorter than δ_{ds} .

In conclusion, we have described a method for the measurement of the complex dielectric response and demonstrated its applicability both for insulators and metals. If the low-or high-energy extrapolation of the reflectivity is ambiguous (as it is the case on the low-energy side for bad metals or narrow-gap semiconductors) it offers a more reliable determination of the complex optical spectrum than the KK transformation, especially close to the cutoff of the detection. The experimental performance, far more simple as

compared with ellipsometric techniques, means the measurement of the normal-incidence reflectivity relative to two reference media, e.g., the reflection from the vacuum-sample and diamond-sample interfaces. The double-reference method may find broad application either in the field of optical spectroscopy or in material characterization due to its numerical precision and simplicity.

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