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Tail absorption in the determination of optical constants of silicon rich carbides

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ABSTRACT

We propose a method to analyze reflectance and transmittance spectra of hydrogenated amorphous silicon-rich carbide alloys, which allows the determination of n–k spectra with very good accuracy. The method is based on the Tauc–Lorentz formulation proposed by Jellison and Modine, in which we introduce an absorption band to take into account the tail states. The method is used to retrieve the n–k spectra of a set of silicon-rich carbide films fabricated in a variety of conditions. The results on oscillator parameters are discussed. We show that by introducing this absorption band we achieve a better accuracy in the determination of the oscillator parameters. The resulting oscillator frequency appears to be well correlated with compositional data.

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1. Introduction

Silicon-rich carbide (SRC) thin films and semiconductor thin films in general, can be considered as the building blocks in a great variety of applications. A rapid, routine characterization of materials represents a requirement whenever the monitoring of the fabrication process is needed. Optical characterization, specifically if based on a widely available, low cost technique as reflectance and transmission (*R&T*) spectroscopy, represents an option, provided that the results are elaborated within a robust framework, able to supply physical information on the investigated material. For materials with optical gap of few eV, spectroscopy in the UV–visible range contains direct information on the density of states, and, if properly analyzed, can be correlated to structural properties such as composition, density, structural order, and hydrogen concentration. In order to obtain such information, a reliable modeling of the optical function is needed.

The determination of the optical constants of silicon-rich carbide, and in general of homogeneous isotropic weakly absorbing amorphous materials, has been addressed by means of a variety of analytical and/or numerical methods, usually applied to spectroscopic ellipsometry [1–10].

Many authors use the Tauc–Lorentz approach as modified by Jellison and Modine [1] (JTL model in the following sections), who showed the need of combining the Lorentz oscillator with the Tauc model [11], in order to supply a comprehensive description of the dielectric constant of amorphous semiconductors over the electromagnetic optical spectrum.

This very popular approach, however, fails in describing the sub-band gap region, where non zero absorption is present due to structural disorder.

Such limitation, already inherent to the Tauc model [3], was noticed by several authors, and alternative approaches were proposed and developed [3,8,10]. An effective improvement for the JTL model was proposed by Ferlauto *et al.* [8], who introduced explicit reference to the Urbach edge [12] to account for the excess absorption at energies lower than the band gap. This Ferlauto et al. model also includes the possibility of selecting between the constant momentum (Tauc approach [2]) and the constant dipole (Cody approach [13]) for the matrix element. The feature is justified by the observation that the Tauc plot does not show a linear behavior, and therefore the Tauc gap E_g obtained by extrapolation at zero ordinate of the quantity $(\alpha E)^{1/2}$, where α is the absorption coefficient and E is the photon energy, depends on the experimentally accessible range of E, which in turn depends on sample thickness [8,14]. The application of the Cody approach in the model requires the introduction of an additional empirical 'buffer' function (Eq. (18) in Ref. [8]) at intermediate energies between the oscillator and the Urbach tail, and of the related demarcation energy [8]. The real part of the dielectric function is then analytically obtained through complete, rather complicated, Kramers–Kronig (KK) integration of the imaginary part. The Urbach edge was also simulated with

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the introduction of one additional parameter with respect to JTL in Ref. [10], however, the model is shown to perform only for one very defective material. An accurate description of the tail absorption can also be found in Ref. [3].

In this paper, we propose an approach to extract the maximum possible information from *R&T* spectroscopy, that is a rapid, non-contact, non-destructive technique, suitable for routine characterization, and whose potential is probably not well understood, with the aim of setting a basis of correlation with the compositional material parameters. *R&T* is specially suitable to probe the medium–low absorption region, which is important in specific optical applications such as photovoltaics or optoelectronics [15,16]. The oscillator energy, except for very thin samples, is normally beyond the accessible range of transmittance, however, the slope and shape of UV reflectance contain sufficient information for the identification of the oscillator parameters.

The approach used within this paper is based on the JTL model [1] further modified to account for the absorption in excess to the Tauc plot. The reason for using the Tauc, rather than the Cody, approach, will be illustrated. The sub-bandgap tail absorption is modeled by making use of an additional Gaussian band (GB), described by three parameters. The Gaussian band is simply added to the imaginary part of dielectric constant. As such function is inherently vanishing with zero derivative for energies apart from its central energy, it does not require the introduction of a demarcation level, and guarantees continuity of the dielectric function and its derivative. The model (JTL-GB in the following discussion) reduces to the basic JTL approach if the amplitude of the Gaussian band is set to zero, which guarantees ready comparison to other oscillator-based literature models. The real part of the dielectric constant is determined through KK numerical integration of the imaginary part; see Appendix A for details. However, for small amplitudes of the Gaussian band the analytical JTL function can be safely applied. The model is applied to *R&T* spectra of a set of silicon rich carbide samples, with Tauc gap E_g spanning from 1.8 eV to 3 eV, that is, up to the stoichiometric case. As the *R&T* technique does not have the dynamics needed to determine the Urbach region, compared to techniques such as photothermal deflection spectroscopy and constant photocurrent method, the latter being limited to photoconducting materials, we do not attempt to correlate the Gaussian band parameters to the Urbach edge; however, we show that if the excess absorption is simply disregarded in the JTL model, the determination of the oscillator parameters, and in particular the oscillator energy, may be severely affected, thus missing valuable information inherent to the experimental data, that can be associated to structural parameters and in particular to the silicon-to-carbon ratio.

2. Experimental details

All the silicon-rich carbide (SRC) layers presented in this paper were deposited on Corning glass using a plasma enhanced chemical vapor deposition parallel plate apparatus in a variety of conditions. We used either 13.56 MHz rf and 4 W plasma power, or 100 MHz and 50 W plasma power over a 144 cm^2 electrode area. The deposition temperatures span from 220 °C to 450 °C; the gas mixture is composed of $SiH_4 + CH_4$, in some cases diluted by H_2. SiH_4 and CH_4 flow rates are in the range 1.5 to 4 and 8 to 73 sccm respectively. The hydrogen dilution varies from 0% to 95%. Extensive details on sample preparation and properties are reported in Ref. [17]. In this paper, all data are treated together, with no attention to the specific conditions. This guarantees that the applied procedure is robust enough to be applicable to a large variety of sample preparation conditions. The sample Si and C concentration was measured by Rutherford backscattering (RBS). Hydrogen concentration was determined either by elastic recoil detection analysis (ERDA) [18], or by RBS spectra simulation [17]. The carbon ratio, $x = [C]/([C] + [Si])$, varies from 0.2 to 0.63, while the overall hydrogen concentration, which strongly depends on deposition temperature and hydrogen dilution, varies from 20% to 50%.

The UV–visible reflectance and transmittance (*R&T*) spectra were measured either by a diode array HP8452A UV–visible spectrophotometer with deuterium source in the range 190–820 nm equipped with a homemade tool for near-normal reflectance *R*, or by an Avantes fiber optic spectrophotometer with deuterium + halogen lamp sources in the range 200–1000 nm and virtually normal incidence for reflectance *R*. In both cases, *R* is normalized with respect to a calibrated reference. Care was taken to perform *R* and *T* measurements in the same sample position, to make sure to probe the same sample thickness and record perfectly the counter-phase interference spectra.

All calculations were performed using the computer code GTB-fit [19]. The code, that is based on fully open access platform, treats the optical system using the general transfer method [20] and is described in detail in Appendix A. The simulation is based on the optical system film/substrate, with no attention on possible interface or surface layers, which is possibly the cause for rather high residual χ^2 in the simulation in few cases. However, using the criterion described in Ref. [21] the presence of surface layers was not detected. The absorption data used in this work are interference-free, and were extracted by numerical inversion of *R&T* measurements, taking into account the spectral refractive index of the involved media, and reflectance at all interfaces and the overall optical system in general, as allowed by the GTB-fit described in Appendix A. Finally, we note that *R* and *T* are experimentally fully independent, but strictly correlated because they must fulfill theoretical constraints such as the same envelope amplitude at all wavelengths, symmetry with respect of 50% in the region of transparency, and tangency with the signal of bare substrate in the region of transparency [21]. Therefore, the simultaneous simulation of *R* and *T*, together with the averaging nature of χ^2, implies robustness against experimental errors, and allows a higher degree of confidence in the results with respect to the simulation of *R* or *T* spectra only.

3. The JTL-GB model

We start our analysis from the original JTL model. The choice of staying at the Tauc [11], rather than the Cody [13] approach to describe the parabolic decrease of absorption in the near-edge region, as proposed for instance in Ref. [8], is based on the observation that the constant momentum approach shows linearity over a wider spectral range. As an example, in Fig. 1 we show the Cody plot $(\alpha/E)^{1/2}$ and the Tauc plot $(\alpha E)^{1/2}$ versus energy for the same data as obtained by *R&T* spectroscopy on a nearly stoichiometric a-SiC:H sample, and the linear fit of both.

The scales are arranged in order to match the maximum values of the curves. The figure also reports the difference between the experimental data and the fit, showing that for the Tauc plot the linear region

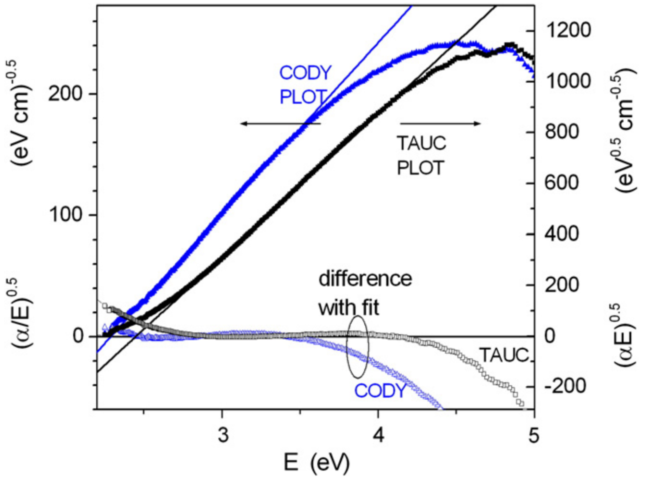

Fig. 1. Cody (left axis) and Tauc (right axis) plots of absorption coefficient measured on a representative nearly stoichiometric a-SiC:H sample.

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