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Universal dispersion model for characterization of optical thin films over wide spectral range: Application to magnesium fluoride

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ABSTRACT

Optical characterization of magnesium fluoride thin films is performed in a wide spectral range from far infrared to extreme ultraviolet (0.01–45 eV) utilizing the universal dispersion model. Two film defects, *i.e.* random roughness of the upper boundaries and defect transition layer at lower boundary are taken into account. An extension of universal dispersion model consisting in expressing the excitonic contributions as linear combinations of Gaussian and truncated Lorentzian terms is introduced. The spectral dependencies of the optical constants are presented in a graphical form and by the complete set of dispersion parameters that allows generating tabulated optical constants with required range and step using a simple utility in the *newAD2* software package.

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1. Introduction

So far the Sellmeier formula has been extensively employed as a standardized approach for the description of optical constants of films and substrates in optics industry. However, the Sellmeier formula is only valid in the range of transparency of the materials forming these films and substrates [1]. The characterization of films in complex multilayer systems requires extending of the spectral range of the measurements to the IR region, which contains phonon absorption, and to the UV region, which contains the electronic excitations. Therefore, a dispersion model suitable for the description of the dielectric response in the wide spectral range must be employed. Such model must take into account the physical conditions following from the theory of dispersion, particularly Kramers–Kronig relations and integrability imposed by sum rules [2–6]. The recently published universal dispersion model (UDM) [7] fulfills these conditions. The UDM enables us to realize a standardized approach for describing the optical constants of optical films and is implemented in the software *newAD2* [8]. It should be emphasized that that the UDM is computationally efficient because all its parts are described by analytical expressions.

This work presents the application of the UDM to the characterization of magnesium fluoride (MgF_2) thin films that are used as low refractive index layers in vacuum UV optics [9–13]. The optical characterization is carried out using simultaneous processing of experimental data obtained by different optical instruments. Six table-top instruments covering the far IR to vacuum UV spectral range were complemented by the spectrophotometer at Elettra BEAR beamline, extending the spectral range of measurements to the extreme UV (EUV). To obtain the true optical constants of the films, it is necessary to take into account film defects influencing the measured optical quantities. In particular, the defect transition layer taking place at bottom boundary must be considered because it causes the sub-gap absorption on localized states. A model of boundary roughness is also important because this defect influences the optical quantity values through scattering losses, especially in EUV range.

2. Universal dispersion model

The mathematical formulation of the UDM is completely presented in [7,14]. The phonon absorption (infrared vibrational absorption) and absorption on localized electronic states are described by a Gaussian broadened discrete spectrum, *i.e.* the imaginary part of dielectric function is given by Gaussian peaks and the real part of dielectric function is given by Dawson integral functions [15]. The Urbach tail is modeled by a specially constructed

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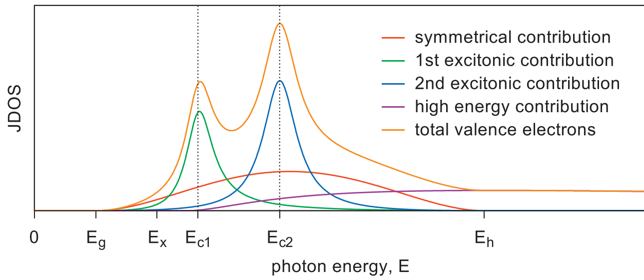

Fig. 1. Schematic diagram of transition density of valence electrons.

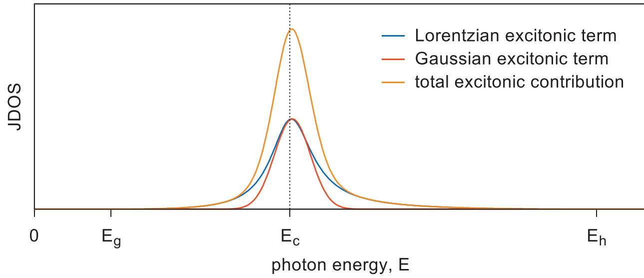

Fig. 2. Schematic diagram of transition density of modified excitonic contribution composed from truncated Lorentzian and Gaussian parts ($G_c \approx 0.4$).

function that contains exponential integral function in the real part of the dielectric function originating in the Hilbert transform of the exponential tail taking place in the imaginary part [14]. Note that in this work the contribution of valence electron excitations was slightly modified in comparison with the UDM described in [7], as described below. The excitations corresponding to core electrons were not taken into account.

Within the UDM the excitations of valence electrons are described using three different contributions schematically depicted in Fig. 1. The number of the excitonic contributions can be arbitrary. The modification concerns the excitonic contributions, whereas the symmetric and high-energy contributions are kept intact. The original excitonic contributions were represented by truncated Lorentzian functions, *i.e.* limited to the interval of photon energies (E_g, E_h), where E_g is the band gap and E_h is the maximum transition energy. The normalized transition density (joint density of states – JDOS) of the truncated Lorentzian term $J_{ex,L}$ was as follows:

$$J^0_{ex,L}(E) = \frac{(E-E_g)^2(E_h-E)^2}{\mathcal{C}_N[(E-E_c)^2+B_c^2]}\Pi_{E_g,E_h}(E) \quad (1)$$

where

$$\Pi_{E_{min},E_{max}}(E) = \begin{cases} 1: & E_{min} < E < E_{max} \\ 0: & \text{otherwise}, \end{cases} \quad (2)$$

E is the photon energy and $\mathcal{C}_N$ is a normalization constant. The modification consists in replacing the truncated Lorentzian term with a linear combination of truncated Lorentzian and Gaussian terms:

$$J^0_{ex}(E) = (1-G_c)J^0_{ex,L}(E) + G_c J^0_{ex,G}(E) \quad (3)$$

where $G_c \in [0, 1]$ is a new parameter (see Fig. 2). The normalized JDOS of Gaussian term has the following form:

$$J^0_{ex,G}(E) = \frac{E^2}{\sqrt{\pi}B_c E_c}\left[\exp\left(-\frac{(E-E_c)^2}{B_c^2}\right) - \exp\left(-\frac{(E+E_c)^2}{B_c^2}\right)\right], \quad (4)$$

where the parameters E_c (peak position) and B_c (width of the peak) are the same as in (1). The parametrization was chosen so that both terms (4) and (1) have the same curvature at the peak maximum for the same parameters. Therefore, B_c in Eq. (4) is not the Gaussian term rms value (the rms value is $B_c/\sqrt{2}$). Otherwise the expression (4) has exactly the same form as the JDOS contribution for localized states [7] and thus the corresponding real part of dielectric function is obtained simply by replacing B_c with $B_c/\sqrt{2}$ in the corresponding formulas.

Note that we described the contributions (3), (1) and (4) as excitonic because they were originally formulated as such. Nevertheless they can be used in practice for the description of many other structures in the joint density of states, not necessarily arising from manybody effects (excitons). Similarly, the terms describing infrared absorption are called phonon contributions even when the material is disordered and long-range lattice vibrations do not occur and the word phonon is not appropriate. Therefore, 'exciton' and 'phonon' must be understood as labels for UDM terms of specific forms rather than exact classification of absorption features.

3. Experiment

The MgF_2 thin films were deposited by vacuum evaporation onto silicon single crystal wafers. For evaporation the commercial SYRUSpro DUV (Leybold Optics) device was employed. The nominal thickness values were controlled by quartz crystal. In this paper the results of the optical characterization will be illustrated for a selected sample with the nominal thickness of 120 nm. The deposition temperature for this sample was 300 °C and the deposition rate was 1 nm/s.

The instruments used to measure the experimental data and the corresponding data sets are summarized in Table 1. It includes six table-top instruments covering the spectral range from the far IR to vacuum UV and the spectrophotometer at Elettra BEAR beamline [16,17] extending the spectral range of our measurements to the extreme UV.

To suppress the influence of the substrate and improve sensitivity to properties of the film, relative reflectance and difference ellipsometry calculated from measured data were added to the data processing. Even though these derived data do not bring any fundamentally new information not contained in the original data, they are less influenced by substrate properties and systematic measurement errors.

The roughness of film upper boundaries was also measured independently by atomic force microscopy (AFM) using a Bruker Dimension Icon microscope in ScanAsyst mode with standard probes.

4. Results and discussion

The spectral dependencies of optical quantities utilized for the optical characterization of MgF_2 film are plotted in Figs. 3–7. The theoretical values of these quantities represented by solid lines were calculated by the matrix formalism [18,19]. All experimental data represented by points were fitted simultaneously in the *newAD2* software [8]. This software can fit together an arbitrary set of optical data using a single consistent model, fitting all parameters simultaneously and equalizing the contributions of individual data sets to the residual sum of squares [7,20,21]. This equalization permits combining individual data sets from different spectral ranges obtained using different instruments exhibiting different systematic errors and acquiring data with different spectral and angular densities while keeping the sensitivity of the method to effects manifested in the individual data sets. One can see that there is a relatively good agreement between theoretical and experimental data.

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