

PARAMETERISATION OF THE MODEL OF DISPERSION DEPENDENCES OF SOLID STATE OPTICAL CONSTANTS¹

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In this paper the new model of dispersion of the optical constants of the amorphous solids is presented. This model adds to the Lorentz model of the classical oscillator the quantum-mechanical concept of the gap. Moreover, this model respects the existence of the tail in absorption of the amorphous material. It is necessary to point out that this new model is acceptable from all the point of physics. It can be employed for solving the inversion problem within the fields of both optics of thin films and optics of solids. The results obtained for a thin film of hydrogenated amorphous silicon using our new model are introduced as well.

1 Introduction

Good knowledge of the optical constants of solid state materials is important for new technologies in practice. Two basic groups of the methods can be utilized for determining the dispersion dependences of the optical constants of solids. The methods of the first group are based on the separate interpretation of the optical quantities of the system containing the material studied at the individual wavelengths, i. e., the optical quantities measured are treated at every wavelength separately (these methods can be called the single-wavelength methods). The latter methods are based on interpreting the entire spectral dependences of the optical quantities of the system measured within a certain spectral region, i. e., on curve fitting with the aid of dispersion relationships (these methods can be called dispersion methods). The dispersion relationships are given by the physical models of the material forming the system studied.

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The single-wavelength methods are the basic ones enabling us to obtain the optical constants of the materials under investigation without knowledge of the model of the dispersion relationships of these materials. The disadvantage of these methods is the fact that the optical constants obtained by the methods mentioned need not satisfy the Kramers-Kronig relations in consequence of systematic errors of these methods. These systematic errors can be caused by the fact that the model of the system containing the material studied employed for determining the optical constants do not respect the perturbations, for example, roughness of boundaries, inhomogeneities of layers, existence of interlayers etc. Thus, these methods are useful for systems which can be described by simple models, i. e., flat surface model, ideal single layer model etc.

The advantage of the latter methods is given by the fact that these methods enable us to obtain the optical constants of the materials forming more complicated systems. In this case the correlations between the parameters describing these more complicated systems are smaller for the dispersion methods than for the single-wavelength methods. The disadvantage of the dispersion methods is implied by the fact that it is necessary to know an appropriate parameterisation of the optical constants of the materials forming the system studied. All the present dispersion relations used in the broad spectral region (i. e., for example, the formulae of Cauchy, Sellmeier, Lorentz, Forouhi etc.) do not describe correctly the behaviour of the optical constants of the materials in the vicinity of the gap. This fact causes difficulties if it is necessary to express the spectral dependences of the optical constants of thin films formed by absorbing materials within the broad spectral region. We encountered with these difficulties at our optical studies of hydrogenated amorphous silicon layers. Therefore we had to propose a new model of the dispersion dependences of the optical constants of this material fulfilling the Kramers-Kronig relations. In this paper the description detailed of this model will be presented. The spectral dependences of the optical constants of the hydrogenated amorphous silicon determined by means of our new dispersion model will be introduced as well.

2 Description of the models of optical constants of amorphous solids in the interband region

The basic model of the optical constants n (refractive index) and k (extinction coefficient) of amorphous solids in the interband region is based on the idea of the classical damped Lorentz oscillator. Within this model the complex dielectric function $\hat{\epsilon}(E)$ is expressed as follows:

$$\hat{\epsilon}(E) = \epsilon_1(E) + i\epsilon_2(E), \quad (1)$$

where

$$\begin{aligned} \epsilon_1(E) &= 1 + \frac{A(E_0^2 - E^2)}{(E_0^2 - E^2)^2 + B^2E^2}, \\ \epsilon_2(E) &= \frac{ABE}{(E_0^2 - E^2)^2 + B^2E^2}, \end{aligned} \quad (2)$$

$E = \hbar\omega$ is photon energy, A , B and E_0 are the parameters corresponding to the model (A , B and E_0 are the strength, linewidth and resonance energy of oscillator, respectively). The optical constants n and k of materials are connected with ϵ_1 and ϵ_2 as follows:

$$\epsilon_1 = n^2 - k^2 \quad \text{and} \quad \epsilon_2 = 2nk. \quad (3)$$

The model of the Lorentz oscillator is satisfactory for the description of the optical constants of the materials in the region of the interband transition. However, this model does not incorporate the concept of an optical energy band gap E_g . Thus, the optical energy band gap of semiconductors and dielectric can not be directly determined from this approach.

For the amorphous solids the valence and conduction bands exhibit simple forms on contrary to forms of the bands of crystalline dielectrics and semiconductors. The forms of these bands of the amorphous solids are namely similar to the form of the bands of free electrons (i. e., similar to the parabolic bands). Thus, quantum-mechanical treatments imply that the expression of ϵ_2 of amorphous solids for photon energies in the vicinity of E_g is given in the following way (see e. g., [1]):

$$\epsilon_2(E) = \alpha (E - E_g)^\beta, \quad (4)$$

where α and β are constants.

The fundamental quantum-mechanical approach (see e. g., [2]) do not lead to equations for the optical constants enabling us to carry out reasonable parameterisation of the inverse problem (within the inverse problem one determines the values of the optical parameters characterizing solids and thin solids film on the basis of the measured values of the optical quantities such as the reflectance, ellipsometric parameters etc.). Forouhi and Bloomer [3] therefore developed the approach enabling them to derive formulae for the optical constants of the amorphous solids expressing the dependences of these optical constants on photon energy and the band gap E_g , i. e.,

$$\begin{aligned} n(E) &= n(\infty) + \frac{B_\alpha E + C_\alpha}{E^2 - BE + C}, \\ k(E) &= \frac{A(E - E_g)^2}{E^2 - BE + C}, \end{aligned} \quad (5)$$

where

$$\begin{aligned} B_\alpha &= \frac{A}{Q} \left(-\frac{B^2}{2} + E_g B - E_g^2 + C \right), \\ C_\alpha &= \frac{A}{Q} \left[(E_g^2 + C) \frac{B}{2} - 2E_g C \right] \end{aligned}$$

and $Q = \sqrt{C - B^2/4}$.

They utilized these formulae for interpreting the spectral dependences of the optical constants of the thin films formed by amorphous silicon and the other amorphous materials (the measured spectral dependences of the optical constants of the amorphous

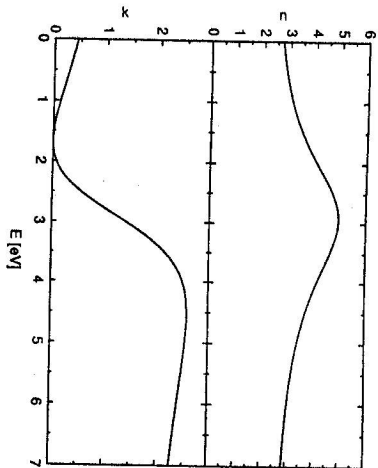


Fig. 1. The spectral dependences of the refractive index n and extinction coefficient k of the hydrogenated amorphous silicon layer calculated using the Forouhi and Bloomer model for the values of the parameters presented in Ref. [3].

silicon-films were taken from Ref. [4]). If the values of the parameters A , B , C , $n(\infty)$ and E_g found for amorphous silicon by these formulae are used for calculating the spectral dependences of n and k (see Ref. [3]) one obtains the curves plotted in Fig. 1. From this figure it is evident that the spectral dependence of k exhibits a peculiar behaviour, i.e., that the values of k are different from zero in the range of E obeying the following relations: $E < E_g$ (Note that Forouhi and Bloomer [3] determined the values of the constants mentioned above by means of the spectral dependences of the optical constants of amorphous silicon corresponding to the range 1.6–3.2 eV). One can see that the spectral dependences of k evaluated by the Forouhi and Bloomer model do not exhibit a reasonable physical behaviour within the broad spectral region.

To remove this physical discrepancy we tried to develop a new model of the optical constants of amorphous materials exhibiting the properties acceptable from the physical point of view. We required in order that our model might fulfil the following requirements:

1. The function $\epsilon_2(E)$ must be an odd function according to the photon energy E .
2. The complex function $\hat{\epsilon}(E)$ must satisfy the Kramers–Kronig relations.
3. At the vicinity of E_0 the behaviour of the function $\hat{\epsilon}(E)$ must correspond to the the model of Lorentz oscillator (see eq. (2)).
4. The behaviour of the function $\epsilon_2(E)$ must fulfil eq. (4) for the values of E close to E_g .

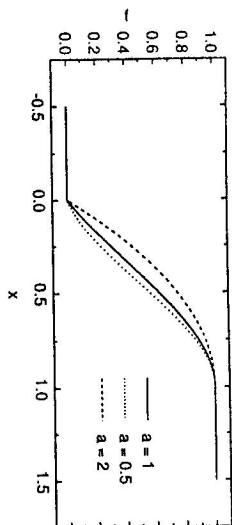


Fig. 2. The courses of the function $f(x)$ for several values of the parameter a .

5. The values of $\epsilon_2(E)$ must exhibit small values different from zero even for the values of E smaller than E_g because amorphous solids have levels of electron energy corresponding to bound states inside of the gap.

From the foregoing requirements concerning $\hat{\epsilon}(E)$ it is evident that the function $\epsilon_2(E)$ can be written in the following form:

$$\epsilon_2(E) = \frac{ABE}{(E_0^2 - E^2)^2 + B^2E^2} G(E), \quad (6)$$

where $G(E)$ is a function which modifies the classical model of the Lorentz oscillator from the point of view of the concept of quantum mechanics, i.e., this function $G(E)$ takes into account the existence of the gap, i.e.,

$$G(E) = \left[f \left(\frac{|E| - E_g}{E_0 - E_g} \right) \right]^b, \quad (7)$$

where the function f is defined in this way:

$$f(x) = \begin{cases} 0 & \text{for } x < 0 \\ 1 & \text{for } x > 1 \\ ax + (3 - 2a)x^2 + (a - 2)x^3 & \text{for } 0 < x < 1. \end{cases} \quad (8)$$

It is clear that the function $f(x)$ is given by a cubic spline for the values of x laying between 0 and 1. In Fig. 2 the courses of the function $f(x)$ are plotted for three values of the parameter a . This cubic spline was constructed by means of the following conditions:

$$\begin{aligned} f(0) &= 0, & f'(0) &= a, \\ f(1) &= 1 & \text{and } f'(1) &= 0. \end{aligned} \quad (9)$$

The symbol f' denotes the derivative of f owing to x .

From the preceding formulae one can easily see that for reasonable values of α (e.g., for $\alpha = 1$) and for the values of E near to the value of E_g , the dependences of the functions $\epsilon_2(E)$ expressed by eqs. (6) and (4) converge each other. The constant α introduced in eq (4) is then expressed by means of the parameters defined in our model through the following equation:

$$\alpha = \frac{ABE_g}{(E_0^2 - E_g^2)^2 + B^2E_g^2} \left(\frac{a}{E_0 - E_g} \right)^b \quad (10)$$

To be able to include the existence of the levels inside of the gap into our model we can express the function $F(E)$ by the following convolution:

$$F(E) = \frac{1}{\sqrt{2\pi}\delta E} \int_{-\infty}^{\infty} G(s) e^{-\frac{(E-s)^2}{2\delta E^2}} ds, \quad (11)$$

where δE represents an extension of the absorption edge connected with the well known tail in absorption of the amorphous solids. From the foregoing it is implied that the function $\epsilon_2(E)$ is given as

$$\epsilon_2(E) = \frac{ABE}{(E_0^2 - E^2)^2 + B^2E^2} F(E). \quad (12)$$

Further, the function $\epsilon_1(E)$ must obey the following equation expressing the Kramers-Kronig relations:

$$\epsilon_1(E) = 1 + \frac{1}{\pi} \int_0^{\infty} \frac{\epsilon_2(E+s) - \epsilon_2(E-s)}{s} ds. \quad (13)$$

It should be pointed out that the integrals taking place in Eqs. (11) and (13) must be solved using an appropriate numerical procedure which can complicate the inverse problem in practice. However, this complication is not too important with respect to an efficiency of contemporary computers and with respect to a possibility of a parallelization of these computers. We employed our model for determining and interpreting the spectral dependences of the optical constants of hydrogenated amorphous silicon-films. Detailed results concerning this study will be presented elsewhere. In this paper we shall only introduce final results expressing the spectral dependences of the optical constants of a chosen film of hydrogenated amorphous silicon determined using the model presented here.

By using a combined method of both spectroscopic multiple angle of incidence ellipsometry and near-normal spectroscopic reflectometry we determined the values of the parameters of our model for the chosen film of hydrogenated amorphous silicon as follows: $A = (11.47 \pm 0.04) \text{ eV}^2$, $B = (2.43 \pm 0.03) \text{ eV}$, $E_0 = (3.891 \pm 0.003) \text{ eV}$, $E_g = (1.728 \pm 0.006) \text{ eV}$, $\delta E = (0.105 \pm 0.005) \text{ eV}$, $a = 2.78 \pm 0.05$ and $b = 1.728 \pm 0.003$. In Fig. 3 one can compare the spectral dependences of the optical constants of the samples of hydrogenated amorphous silicon determined using our model and Forouhi

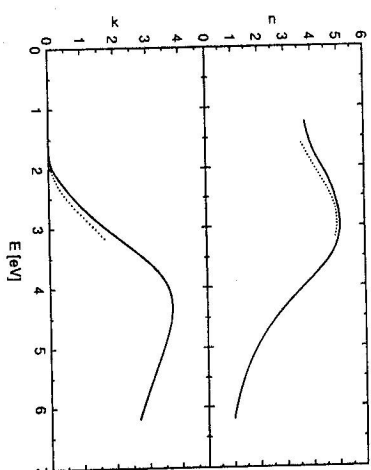


Fig. 3. Comparison of the spectral dependences of the refractive indices n and extinction coefficients k of the hydrogenated amorphous silicon layers found by us (solid curves) and by Forouhi and Bloomer [3] (dashed curves).

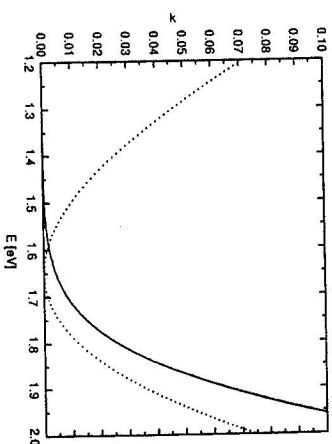


Fig. 4. The spectral dependences of the extinction coefficients k of the samples of hydrogenated amorphous silicon found by us (solid curve) and Forouhi and Bloomer [3] (dashed curve).

and Bloomer model. Note that in Fig. 3 these spectral dependences are only plotted in the spectral regions in which they were measured. The comparison of the curves plotted in Figs. 1 and 3 implies that the Forouhi and Bloomer model is not usable in a broad spectral region (i.e., the Forouhi and Bloomer model can only be used in the relatively narrow spectral region corresponding to photon energies greater than value of E_g).

From Fig. 4 it is evident that the application of the model of Forouhi and Bloomer [3] is not reasonable for photon energies smaller than value of E_g from the physical point of view.

3 Conclusion

In this paper the new model of dispersion of the optical constants of the amorphous solids has been presented. This model adds to the Lorentz model of the classical oscillator the quantum-mechanical concept of the gap. Moreover, this model respects the existence of the tail in absorption of the amorphous material. It is necessary to point out that our new model is acceptable from all the point of physics. It can be employed for solving the inversion problem within the fields of both optics of thin films and optics of solids.

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