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Dispersion models describing interband electronic transitions combining Tauc's law and Lorentz model

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ABSTRACT

Five dispersion models of interband electronic transitions in disordered solids based on the combination of the Tauc's law and the Lorentz model (or the Lorentz function) will be discussed. These models are the Campi–Coriasso, Jellison–Modine (Tauc–Lorentz), Ferlauto et al. (Cody–Lorentz) and we also propose two other models. The models that we propose can be considered to be modifications of the Jellison–Modine and Ferlauto et al. models. This modification consists of the fact that these models are based on the Lorentz function, not on the response function of the damped harmonic oscillator. The analytic expressions for the real part of the dielectric function of the Campi–Coriasso model and one of our models will be published. The differences between the presented models will be compared by means of fitting the optical constants (functions) calculated with one model by other models.

1. Introduction

In very rough approximation the interband electronic transitions in disordered solids are described by the Lorentz model representing dielectric response of classical damped harmonic oscillator (DHO) [1]. The dependence of the complex dielectric function on the photon energy E can then be written as

$$\hat{\varepsilon}(E) = 1 + \frac{2}{\pi}\frac{N}{E_c^2 - E^2 - \mathrm{i}BE}\,, \tag{1}$$

where E_c, B and N are the parameters of DHO. The central energy E_c and broadening B in classical model are related to restoring F_r and damping F_d forces:

$$F_r = -k_r r \quad \text{and} \quad F_d = -k_d \dot{r} \tag{2}$$

where r and $\dot{r}$ are deviation of electron from the equilibrium position and its velocity, respectively. The elastic k_r and damping k_d constants are related to E_c and B as [1]

$$E_c = \hbar\sqrt{\frac{k_r}{m_e}} \quad \text{and} \quad B = \hbar\frac{k_d}{m_e}\,. \tag{3}$$

The symbols $\hbar$ and m_e denote the reduced Planck constant and electron mass, respectively. The parameter N is transition strength introduced in [2]. If we assume that only the valence electrons contribute to the interband electronic transitions then the transition strength is proportional to the density of valence electrons $\mathcal{N}_{ve}$:

$$N = \frac{\pi e^2\hbar^2}{2\epsilon_0 m_e}\mathcal{N}_{ve}\,, \tag{4}$$

where e is electron charge and ϵ_0 is vacuum permittivity. Note that the different parameters are often used instead of the quantity N. For example, the oscillator strength (or just strength) $S = 2N/\pi$, plasma energy $E_p = \sqrt{2N/\pi}$ or parameter $A = 2N/(\pi E_c)$ called amplitude. The quantity S is different from the dimensionless quantum mechanical quantity f called oscillator strength.

The real and imaginary part of the dielectric function (1) are expressed as

$$\varepsilon_r(E) = 1 + \frac{2N}{\pi}\frac{E_c^2 - E^2}{\left(E_c^2 - E^2\right)^2 + B^2E^2}, \tag{5}$$

$$\varepsilon_i(E) = \frac{2N}{\pi}\frac{BE}{\left(E_c^2 - E^2\right)^2 + B^2E^2}\,. \tag{6}$$

These functions for chosen dispersion parameters of DHO are plotted in Fig. 1. For an underdamped harmonic oscillator, *i.e.* for $E_c > B/2$, the imaginary part of the dielectric function can be rewritten as

$$\varepsilon_i(E) = \frac{NB}{2\pi E_r}\left(\frac{1}{(E - E_r)^2 + B^2/4} - \frac{1}{(E + E_r)^2 + B^2/4}\right) \tag{7}$$

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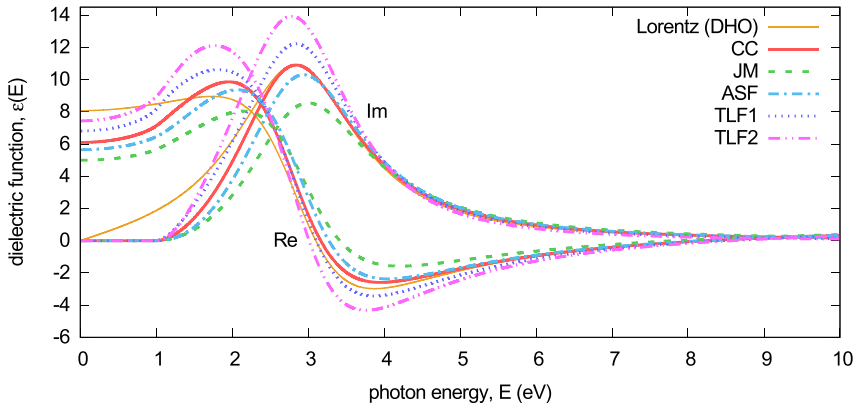

Fig. 1. Complex dielectric functions calculated by the Lorentz model and different versions of the Tauc–Lorentz model for $N = 100$ eV^2, $E_c = 3$ eV, $B = 2$ eV, $E_g = 1$ eV and $E_q = 1$ eV. Acronyms: DHO – damped harmonic oscillator; CC – Campi–Coriasso; JM – Jellison–Modine; ASF – A.S. Ferlauto et al.; TLF – truncated Lorentz function.

where

$$E_r = \sqrt{E_c^2 - B^2/4} \tag{8}$$

is the resonant energy. In the limit $B \to 0$ the Lorentz functions in Eq. (7) can be rewritten using delta functions as

$$\varepsilon_i(E) = \frac{N}{E_r}\left(\delta(E - E_r) - \delta(E + E_r)\right) \tag{9}$$

which represents the bridge between the classical and quantum theories of dielectric response because this equation can be also derived from the Fermi golden rule [3] for the discrete transitions with energy E_r. Reversely, the function in Eq. (7) can be interpreted as Lorentzian ε-broadened discrete transitions in Eq. (9) [4]. In this case the broadening parameter B represents a full width half maximum (FWHM) value of the normalized broadening Lorentz function:

$$\beta(x) = \frac{B}{2\pi}\frac{1}{x^2 + B^2/4}\ . \tag{10}$$

Note that the critically damped ($E_c = B/2$) or overdamped ($E_c < B/2$) DHO cannot be expressed as Lorentzian ε-broadened discrete transitions.

In practice it is not sufficient to use Lorentz model with only one DHO for the description of interband electronic transitions but it is necessary to consider the model with several DHO:

$$\hat{\varepsilon}(E) = 1 + \frac{2}{\pi}\sum_t \frac{N_t}{E_{c,t}^2 - E^2 - \mathrm{i}B_t E}\ , \tag{11}$$

where the index t is used to distinguish individual DHO. Note that in the limit $E_c \to 0$ the contribution corresponds to the Drude model [1]. The model that combines both the Lorentz and Drude terms is known as the Drude–Lorentz model. The total transition strength of valence electrons N is the sum of individual transition strengths:

$$N = \sum_t N_t\ . \tag{12}$$

The Drude–Lorentz model is well suited for the description of dielectric functions of metals where the Drude terms are used for the contributions from free electrons (intraband transitions) and the Lorentz terms are used to express the interband transitions. In the case of dielectrics the Lorentz model fails because the electronic band structure leads to existence of the band gap and the Lorentz model cannot describe the region below the band gap energy E_g where the absorption is negligible.

The behavior of dielectric response in the vicinity of the band gap can be derived using the parabolic band approximation, which gives the quadratic dependence of joint density of states for energies above E_g:

$$J(E) \propto (E - E_g)^2\ . \tag{13}$$

Therefore, if the momentum matrix element is assumed to be constant, then the imaginary part of the dielectric function in the vicinity of E_g is given by the so called Tauc's law [5]:

$$\begin{aligned}\varepsilon_i(E) &\propto \frac{(E - E_g)^2}{E^2} \quad \text{for} \quad E > E_g\\ \varepsilon_i(E) &= 0 \quad \text{for} \quad E \le E_g\ .\end{aligned} \tag{14}$$

Note that the dielectric function must satisfy three fundamental conditions coming from the classical theory of dispersion. The first condition known as the time-reversal symmetry is [1]

$$\hat{\varepsilon}(E) = \hat{\varepsilon}(-E)^*\ , \tag{15}$$

where the symbol * denotes the complex conjugation. This condition expresses the fact that the complex dielectric function (susceptibility) is a Fourier image of the real response function. The second condition are the Kramers–Kronig relations [1,6,7] which ensure the causality of the response function

$$\varepsilon_r(E) - 1 = \frac{1}{\pi}\int_{-\infty}^{\infty}\frac{\varepsilon_i(X)}{X - E}\mathrm{d}X\ , \tag{16}$$

$$\varepsilon_i(E) = -\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{\varepsilon_r(X) - 1}{X - E}\mathrm{d}X\ . \tag{17}$$

The third condition says that the dielectric function must have convergent sum rule integral

$$\int_0^{\infty} E\,\varepsilon_i(E)\,\mathrm{d}E = N\ , \tag{18}$$

which corresponds to the fact that the system has finite density of charged particles [1,8,9]. The above conditions imply that the imaginary part of the dielectric function must be odd function of E and for high energies it must fall faster than $1/E^2$. The Drude–Lorentz model