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Broadening of dielectric response and sum rule conservation

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ABSTRACT

Different types of broadening of the dielectric response are studied with respect to the preservation of the Thomas–Reiche–Kuhn sum rule. It is found that only the broadening of the dielectric function and transition strength function conserve this sum rule, whereas the broadening of the transition probability function (joint density of states) increases or decreases the sum. The effect of different kinds of broadening is demonstrated for interband and intraband direct electronic transitions using simplified rectangular models. It is shown that the broadening of the dielectric function is more suitable for interband transitions while broadening of the transition strength function is more suitable for intraband transitions.

1. Introduction

The dielectric response can be equivalently described by various functions of photon energy E, such as the imaginary part of the dielectric function ε_i, the transition probability function J and the transition strength function F:

$$\varepsilon_i \approx \frac{J}{E^2} = \frac{F}{E}, \tag{1}$$

where symbol $\approx$ represents the dipole approximation [1]. The function F is defined as a continuous condensed-matter equivalent of the oscillator strength known for discrete transitions in atomic spectra [1]. It was shown that this function satisfies the integral form of Thomas–Reiche–Kuhn sum rule [2,1]:

$$\int_0^\infty F(E)\mathrm{d}E = N, \tag{2}$$

where the constant N is called the total transition strength. Note that J in (1) is proportional to the transition density $\mathcal{J}$ [1] or the joint density of states J_{vc} [3,4] if the transition probability is constant but it is not equivalent in general. In the frame of the dipole approximation the real part of conductivity is related to the transition strength function according to

$$\sigma_r \approx \frac{\epsilon_0}{\hbar} F, \tag{3}$$

where ϵ_0 is the vacuum permittivity and $\hbar$ is the reduced Planck constant. The functions F or J can be used to construct dispersion models fulfilling the sum rule (2) [1,5,6]. In the case of crystalline materials, direct interband transitions and two-phonon absorption are modeled using a piecewise function exhibiting sharp features, i.e. Van Hove singularities [1,7].

In reality, more complex phenomena, such as electron–phonon interaction and isotopic effects, cause broadening of the dielectric response that have to be taken into account in the dispersion models. The broadening is introduced phenomenologically as the convolution with a normalized broadening function. The broadening function usually has the form of a Lorentzian or Gaussian [3,4,8–14] and its selection depends on the broadening phenomena [4]. The most applicable, from our experiences, is the Gaussian broadening function. The Lorentzian broadening or an approximation of Gaussian broadening is often chosen to avoid computational difficulties.

The convolution is applied to different quantities, usually J or ε_i. Kim et al. showed that the broadening of ε_i is preferable because it does not lead to spurious static conductivity in the broadened dielectric response [4].

The broadening needs to be judged also from the sum rule preservation point of view. This work discusses the broadening of ε_i and σ_r that correspond to the Fermi golden rule and the Kubo formula, respectively. These two approaches represent the two starting points for the expression of the dielectric response for quantum systems. It will be shown that the broadening of ε_i is more suitable for interband transitions while the broadening of F or σ_r is better for intraband transitions.

2. Dielectric response

This section will summarize the approaches to the construction of the electronic part of the dielectric response. The incorporation of the nucleonic part of the dielectric response is described in [1]. Within the dipole approximation, the dielectric function can be obtained using different approaches leading to the same unbroadened ε_i. One possibility is to start from the Fermi golden rule that expresses the transition

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probability for transitions from an initial ground state $|i\rangle$ to s final state $|f\rangle$ of a closed system [15]:

$$\varepsilon_{\mathrm{i}} = \frac{\pi}{\epsilon_0 V}\sum_{f}^{f\neq i} |\langle f|\hat{d}_{\mathrm{xe}}|i\rangle|^2 \left[\delta\left(E_f - E_i - E\right) - \delta\left(E_i - E_f - E\right)\right], \tag{4}$$

where V is the volume of the system, E_i and E_f are the energies of the initial and final states, respectively. The symbol $\hat{d}_{\mathrm{xe}}$ denotes the total dipole operator of electrons along the external electric field and can be expressed as follows [15]:

$$|\langle f|\hat{d}_{\mathrm{xe}}|i\rangle|^2 = e^2|\langle f|\hat{x}_{\mathrm{e}}|i\rangle|^2 = \left(\frac{e\hbar}{m_{\mathrm{e}}}\right)^2 \frac{|\langle f|\hat{p}_{\mathrm{xe}}|i\rangle|^2}{\left(E_f - E_i\right)^2}, \tag{5}$$

where m_{e} and e denote the electron mass and the elementary charge, respectively. Operators $\hat{p}_{\mathrm{xe}}$ and $\hat{x}_{\mathrm{e}}$ are the total momentum and position operators of electrons. Therefore, the dielectric function can be written as

$$\varepsilon_{\mathrm{i}} = \frac{(eh)^2}{4\pi\epsilon_0 m_{\mathrm{e}}^2 V}\sum_{f}^{f\neq i} \frac{|\langle f|\hat{p}_{\mathrm{xe}}|i\rangle|^2}{\left(E_f - E_i\right)^2}\left[\delta\left(E_f - E_i - E\right) - \delta\left(E_i - E_f - E\right)\right]. \tag{6}$$

The relation $\varepsilon_{\mathrm{i}} = J/E^2$ is obtained if the transition probability function J is introduced as

$$J = \frac{(eh)^2}{4\pi\epsilon_0 m_{\mathrm{e}}^2 V}\sum_{f}^{f\neq i} \left|\langle f|\hat{p}_{\mathrm{xe}}|i\rangle\right|^2 \left[\delta\left(E_f - E_i - E\right) - \delta\left(E_i - E_f - E\right)\right] \tag{7}$$

and the properties of the δ-function

$$\frac{\delta\left(E_f - E_i \pm E\right)}{\left(E_i - E_f\right)^2} = \frac{\delta\left(E_f - E_i \pm E\right)}{E^2} \tag{8}$$

are utilized.

Another possible starting point is the Kubo formula derived for an open non-interacting system [16,17]. It expresses the real part of conductivity σ_{r} as

$$\sigma_{\mathrm{r}} = \frac{\pi\hbar}{V}\sum_{i,f}^{f\neq i} \exp\left(\frac{\Omega - E_i}{k_{\mathrm{B}}T}\right)\frac{|\langle f|\hat{j}_{\mathrm{xe}}|i\rangle|^2}{E_f - E_i}\left[\delta\left(E_f - E_i - E\right) + \delta\left(E_i - E_f - E\right)\right] \tag{9}$$

using the electric current operator $\hat{j}_{\mathrm{xe}}$. Its matrix element is related to the momentum matrix element as follows:

$$|\langle f|\hat{j}_{\mathrm{xe}}|i\rangle|^2 = \frac{e^2}{m_{\mathrm{e}}^2}|\langle f|\hat{p}_{\mathrm{xe}}|i\rangle|^2. \tag{10}$$

The symbols k_{B} and T denote the Boltzmann constant and temperature, respectively, and Ω is the thermodynamic potential satisfying

$$\sum_i \exp\left(\frac{\Omega - E_i}{k_{\mathrm{B}}T}\right) = 1. \tag{11}$$

The summation over initial states can be avoided for low temperatures, when only the ground state is considered. Then, the expression for the transition strength function is obtained from Eqs. (3) and (9) [1]:

$$F = \frac{(eh)^2}{4\pi\epsilon_0 m_{\mathrm{e}}^2 V}\sum_{f}^{f\neq i} \frac{|\langle f|\hat{p}_{\mathrm{xe}}|i\rangle|^2}{E_f - E_i}\left[\delta\left(E_f - E_i - E\right) + \delta\left(E_i - E_f - E\right)\right]. \tag{12}$$

All three expressions (6), (7) and (12) are equivalent and related by Eq. (1).

3. Broadening

Broadening of the dielectric response is introduced phenomenologically by replacing the δ-functions in the formulas (6), (7) and (12) above with a normalized broadening functions β, e.g. the Gaussian

$$\delta(x) \to \beta(x) = \frac{1}{\sqrt{2\pi}B}\exp\left(-\frac{x^2}{2B^2}\right). \tag{13}$$

Here $B > 0$ denotes the broadening parameter. However, the previously equivalent formulas become non-equivalent after this transformation. Depending on how the inverse square of energy in the formulas is split between the powers of $(E_f - E_i)$ and E, different types of broadening are obtained:

$$\begin{aligned}
\widetilde{\varepsilon}_{\mathrm{i}}(E) &= \frac{1}{E^2}\int_{-\infty}^{\infty}\beta(E-t)J(t)\mathrm{d}t && J\text{—broadening},\\
\widetilde{\varepsilon}_{\mathrm{i}}(E) &= \frac{1}{E}\int_{-\infty}^{\infty}\beta(E-t)\frac{J(t)}{t}\mathrm{d}t && \mathrm{F}\text{—broadening},\\
\widetilde{\varepsilon}_{\mathrm{i}}(E) &= \int_{-\infty}^{\infty}\beta(E-t)\frac{J(t)}{E^2}\mathrm{d}t && \varepsilon\text{—broadening},
\end{aligned} \tag{14}$$

where $\widetilde{\varepsilon}_{\mathrm{i}}$ denotes broadened ε_{i}. The different broadening types can be written more succinctly if we denote convolution by $*$ and understand that the independent variable is energy E:

$$\begin{aligned}
\widetilde{\varepsilon}_{\mathrm{i}} &= \frac{1}{E^2}(\beta * J) && J\text{—broadening},\\
\widetilde{\varepsilon}_{\mathrm{i}} &= \frac{1}{E}\left(\beta * \frac{J}{E}\right) = \frac{1}{E}(\beta * F) && \mathrm{F}\text{—broadening},\\
\widetilde{\varepsilon}_{\mathrm{i}} &= \beta * \frac{J}{E^2} = \beta * \varepsilon_i && \varepsilon\text{—broadening}.
\end{aligned} \tag{15}$$

Note that the symmetries of J, F and ε imply that β must be an even function so that the broadening preserves the symmetries.

4. Sum rule conservation

In order to study the conservation of the sum rule by different types of applied broadening, it is advantageous to expand F in the following form

$$F(E) = \frac{E^m}{2}\int_{-\infty}^{\infty}\left[\delta(E-t) + (-1)^m\delta(E+t)\right]F(t)t^{-m}\mathrm{d}t, \tag{16}$$

where m is an integer. This identity clearly holds because F is an even function. The factor $(-1)^m$ in the integrand follows from the identity

$$E^m\delta(E \pm t) = (\mp t)^m\delta(E \pm t). \tag{17}$$

When δ in Eq. (16) is replaced with β, different values of the power m lead to different types of broadening introduced above. The broadened transition strength corresponding to the m-th power, denoted $\widetilde{F}_m$, can therefore be written as

$$\widetilde{F}_m(E) = \frac{E^m}{2}\int_{-\infty}^{\infty}\left[\beta(E-t) + (-1)^m\beta(E+t)\right]F(t)t^{-m}\mathrm{d}t, \tag{18}$$

where $m = -1, 0$, and 1 correspond to J-, F- and ε-broadening, respectively. The sum rule is conserved by the broadening if

$$\frac{1}{2}\int_{-\infty}^{\infty}\widetilde{F}_m(E)\mathrm{d}E = \frac{1}{2}\int_{-\infty}^{\infty}F(E)\mathrm{d}E = N. \tag{19}$$

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