

## Optical Dispersion by Wannier Excitons

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An analytical expression of the complex dielectric constant of Wannier excitons is obtained, which includes exactly the contributions of *all bound and unbound states*. It allows an improved description of the excitonic influence on the optical properties of semiconductors near the band gap, especially with respect to dispersion effects.

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The importance of excitonic effects on the optical properties of semiconductors in the vicinity of the band edge has been recognized for decades. In his renowned paper [1], Elliott gave the analytical expression of the intensity of optical absorption by Wannier excitons, now reproduced in many textbooks. The excitonic spectrum is similar to that of the hydrogen atom and consists of discrete lines and of a continuum [2], the latter being described by the so-called Sommerfeld or Coulomb enhancement factor. Elliott's formula has been used extensively for the determination of the physical constants of semiconductors and has been convoluted with a Lorentzian to account for the broadening of the energy levels [3]. However, the real part of the dielectric constant was not calculated in Elliott's paper, and dispersion effects were not properly taken into account, the refractive index being considered approximately constant near the absorption threshold.

Such a crude description of dispersion effects is not satisfying any more. Spectral hole burning experiments in semiconductor thin films have demonstrated the importance of reflectivity measurements [4]. Ever more solid-state devices are operating close to the band edge, be it lasers or optical modulators. The field of multilayered structures, such as microcavities, is rapidly growing [5]. An accurate model of the refractive index of semiconductor materials, some of them ternary or quaternary alloys for which scarce experimental information is available, is obviously needed, in academic and engineering

studies alike [6]. Many optical constants [7], along with their temperature or pressure dependence, are linked to the complex dielectric constant and would benefit also from its better description near the absorption threshold.

In this Letter, we complete Elliott's results by giving in Eqs. (6), (11), and (12) the complex dielectric constant  $\varepsilon = \varepsilon_1 + i\varepsilon_2$  of Wannier excitons, taking all bound and unbound states exactly into account, while including a broadening  $\Gamma$  of the energy levels [8–10]. The contribution of the Coulomb enhancement factor to  $\varepsilon_1$  is derived for the first time, and that of all bound states—not merely the fundamental one—as well. The final result for  $\varepsilon$  clearly shows that bound and unbound states are but the manifestation of a single physical effect, namely, the Coulomb interaction between electron and hole. Therefore, they should not be dissociated in the treatment of excitonic effects. We conclude by giving some directions in the application of this newly found expression of  $\varepsilon$ .

The starting point of the calculation is standard. It relies on the linear response theory and Fermi's golden rule for light-matter interaction proportional to  $\mathbf{p} \cdot \mathbf{A}$  [11–12] and assumes that the effective mass approximation with parabolic bands is valid. Weakly bound (Wannier) excitons and a constant dipole matrix element  $|\mathbf{e} \cdot \mathbf{M}_{cv}(\mathbf{0})|$  for allowed transitions are considered [11]. With  $R$  being the energy of the fundamental bound state [13],  $E_g$  the band gap energy, and  $\mu$  the reduced mass of the exciton,  $\varepsilon_2$  reads [3] in SI units

$$\varepsilon_2(E) = \frac{A}{E^2} \left\{ \sum_{n=1}^{+\infty} 4\pi R^{3/2} \frac{1}{n^3} \delta\left(E - E_g + \frac{R}{n^2}\right) + \frac{2\pi\sqrt{R}\theta(E - E_g)}{1 - \exp[-2\pi\sqrt{R}/(E - E_g)]} \right\} \quad \text{for } E > 0, \quad (1)$$

$$A = \frac{\hbar^2 q^2}{2\pi \varepsilon_0 m_0^2} \left( \frac{2\mu}{\hbar^2} \right)^{3/2} |\mathbf{e} \cdot \mathbf{M}_{cv}(\mathbf{0})|^2, \quad (2)$$

where  $\theta$  is the Heaviside function. The advantage of dealing with  $\varepsilon_2$  instead of the absorption is to avoid the complicated energy dependence of the refractive index.

The next step, which turns out to be crucial, is to introduce a broadening of the energy levels, while still

preserving the odd parity of  $\varepsilon_2$  with respect to  $E$ , imposed by causality [14]. Using a Lorentzian of width  $\Gamma$ , we thus define  $\varepsilon_2^{(\Gamma)}$  by

$$\varepsilon_2^{(\Gamma)}(E) = \frac{\Gamma}{\pi} \left\{ \int_0^{+\infty} \frac{dE' \varepsilon_2(E')}{(E - E')^2 + \Gamma^2} - \int_0^{+\infty} \frac{dE' \varepsilon_2(E')}{(E + E')^2 + \Gamma^2} \right\}. \quad (3)$$

The second term of the right-hand side of Eq. (3) is almost always unduly discarded because of its nonresonant character. However, one can check that such a definition, after using the Kramers-Kronig relation, gives the broadened dielectric constant for interband transitions [9].

For the continuum states, the change of variable  $u = 2\pi[R/(E' - E_g)]^{1/2}$  allows one to rewrite Eq. (3), using Eq. (1), in the form

$$\varepsilon_{2,\text{unbound}}^{(\Gamma)}(E) \propto \left( \int_0^{+\infty} du F(u^2) \frac{u}{1 - \exp(-u)} - [E \leftrightarrow (-E)] \right), \quad (4)$$

where  $F$  is a rational fraction, and  $[E \leftrightarrow (-E)]$  indicates that the nonresonant contribution must be subtracted, as in Eq. (3). A partial fraction decomposition of  $F$ , the identity  $[1 - \exp(-u)]^{-1} = 1 + [\exp(u) - 1]^{-1}$  and integral 3.415.1 of Ref. [15] lead to

$$\varepsilon_{2,\text{unbound}}^{(\Gamma)}(E) = \text{Im} \left( \frac{A\sqrt{R}}{(E + i\Gamma)^2} (g_{\text{unbound}}(\xi(E + i\Gamma)) + g_{\text{unbound}}(\xi(-E + i\Gamma)) - 2g_{\text{unbound}}(\xi(0))) \right), \quad (5)$$

$$\varepsilon_{1,\text{unbound}}(E > 0) = \frac{A\sqrt{R}}{E^2} \left( \ln \frac{E_g^2}{|E_g^2 - E^2|} - 4\psi\left(\sqrt{\frac{R}{E_g}}\right) + 2\psi\left(\sqrt{\frac{R}{E_g + E}}\right) + 2\psi\left(\sqrt{\frac{R}{E_g - E}}\right)\theta(E_g - E) + 2\text{Re}\psi\left(i\sqrt{\frac{R}{E - E_g}}\right)\theta(E - E_g) - 2\sqrt{\frac{E_g}{R}} + \sqrt{\frac{E_g - E}{R}}\theta(E_g - E) + \sqrt{\frac{E_g + E}{R}} \right), \quad (9)$$

the expression given in Ref. [16] being an approximation of the above equation.

Turning to the contribution of the bound states to  $\varepsilon_2^{(\Gamma)}$ , we perform a partial fraction decomposition with respect to  $n$  and use identities 8.365.1 and 8.365.8 of Ref. [15]. We find an expression similar to Eq. (5), with  $g_{\text{unbound}}$  now replaced by

$$g_{\text{bound}}(\xi) = -2\psi(1 - \xi) - 2\psi(1 + \xi). \quad (10)$$

Again  $\varepsilon_{1,\text{bound}}^{(\Gamma)}$  is obtained by replacing  $\text{Im}(\dots)$  by  $\text{Re}(\dots)$  in Eq. (5).

Summing the contributions of bound and unbound states gives finally

$$\varepsilon_{\text{allowed}}^{(\Gamma)}(E) = \frac{A\sqrt{R}}{(E + i\Gamma)^2} \{g_a(\xi(E + i\Gamma)) + g_a(\xi(-E - i\Gamma)) - 2g_a(\xi(0))\}, \quad (11)$$

$$g_a(\xi) = 2\ln\xi - 2\pi\cot(\pi\xi) - 2\psi(\xi) - 1/\xi. \quad (12)$$

In the case of forbidden transitions [1,11,12], assuming a constant matrix element  $|\mathbf{e} \cdot \nabla_k \mathbf{M}_{c\nu}(\mathbf{0})|$ , the calculation gives a result similar to Eq. (11), with, of course, a new

$$\xi(z) = \sqrt{\frac{R}{E_g - z}}, \quad (6)$$

$$g_{\text{unbound}}(\xi) = 2\psi(\xi) + 2\ln(\xi) + 1/\xi, \quad (7)$$

where  $\psi(z) = d\ln\Gamma(z)/dz$  is the digamma function.

Equation (5) is purposely written in such a form to make the calculation of  $\varepsilon_1$  quite trivial. Indeed  $\varepsilon_1$  and  $\varepsilon_2$  are linked by a Hilbert transform known as a Kramers-Kronig relation [14]

$$\varepsilon_1(E) = \frac{1}{\pi} P \int_{-\infty}^{+\infty} dE' \frac{\varepsilon_2(E')}{E' - E}, \quad (8)$$

where the usual  $+1$  term in Eq. (8) has been discarded, since it can be absorbed in the background dielectric constant.  $\varepsilon_1$  is easily calculated by contour integration in the complex upper half-plane when  $\varepsilon_2(E')$  exhibits no singularity for  $\text{Im}(E') > 0$  and conveniently vanishes for  $|E'| \rightarrow \infty$ , since only the residue at  $E' = E$  is then needed. Consequently,  $\varepsilon_1(E)$  verifies an equation very similar to Eq. (5) with  $\text{Im}(\dots)$  merely replaced by  $\text{Re}(\dots)$ . This is actually the case for  $\varepsilon_{1,\text{unbound}}^{(\Gamma)}(E)$ . By taking the limit  $\Gamma \rightarrow 0$  we find the contribution of the unbound states to  $\varepsilon_1$  for positive energies:

constant  $A'$  and a function  $g_a$  replaced by  $g_f$ :

$$g_f(\xi(z)) = [z - (E_g - R)]g_a(\xi(z)). \quad (13)$$

It is possible to show that  $g_a(\xi(z))$  is, in fact, proportional to the resolvent of the Schrödinger equation associated to the state  $|\mathbf{r} = \mathbf{0}\rangle$ . Indeed Eq. (12) gives the energies  $E_g - R/n^2$  and the oscillator strengths  $\propto 1/n^3$  of the bound states because of the poles and the corresponding residues of  $\pi\cot\pi z$  in the complex plane; Eq. (13) indicates that forbidden transitions start at the  $n = 2$  level only [1]. Not unexpectedly, forbidden transitions do not lead to pronounced features for  $\varepsilon_1$  near  $E_g$ . We will thus limit ourselves to the case of allowed transitions in the following discussion.

The exact expression of the dielectric constant in Eq. (11) turns out to be extremely compact, and the effect of the Coulomb interaction is given by the ratio  $R/E_g$ , as expected. The corresponding  $\varepsilon_1$  and  $\varepsilon_2$  are shown in Fig. 1 along with their usual interband values [9], obtained for vanishing  $R$  [we have in that case  $g_{\text{interband}}(\xi) = -1/\xi$ ]. We see that dispersion effects are important close to  $E_g$ , even for a substantial broadening, when compared to the interband case. Therefore, the latter model should not be used to describe excitonic effects

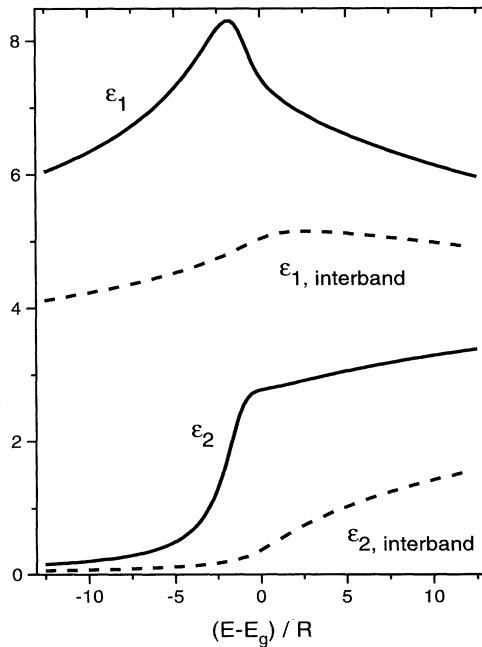


FIG. 1. Real and imaginary parts of the dielectric constant with excitonic effects (solid line) and without excitonic effects (dashed line) for  $E_g = 1.42$ ,  $R = 0.004$ , and  $\Gamma = 0.006$ , in units of  $A\sqrt{R}$ .

in the dielectric constant [17]. The logarithmic term in Eq. (12) accounts for most of the discrepancy between  $\epsilon_1$  and  $\epsilon_{1,interband}$  and comes solely from the unbound states. Such a logarithmic singularity has already been proposed as the consequence of the steplike behavior of absorption at  $E_g$  [18,19] or been derived from an effective  $\epsilon_2$  [20,21]. Let us stress, however, that its prefactor corresponds to only half the discontinuity. This comes from the correct asymptotic expansion of the Sommerfeld enhancement factor as given by Dow [22]. Finally, in view of the exact above result, we note that the shell source approximation introduced by Stahl and Balslev in their coherent wave approach is excellent and that  $\epsilon_1(E_g^+)$  remains finite indeed [16].

The new expression Eq. (11) takes the influence of excitonic effects on the optical properties of semiconductors into better account near the band edge and can be used directly in wavelength modulation spectroscopy or ellipsometry [9], thereby contributing to a more accurate determination of optical constants like  $E_g$ ,  $R$ , and oscillator strengths for such materials. The derivatives of  $\epsilon_1$  and  $\epsilon_2$  with respect to energy have been plotted in Fig. 2, which shows that the maximum of  $\epsilon_1$  is reached below  $E_g - R$ , as expected; these quantities may be more interesting because the background dispersion is removed. As a further consequence, the dependence of optical constants with temperature or any external perturbation [7,9] can now be given with greater detail, provided that one keeps

in mind the limitations of the Wannier exciton model [3,23,24].

The calculation of  $\epsilon$  for the two-dimensional Coulomb potential, which can be viewed as the limiting case for quantum wells and used in the description of  $M_1$  edges (saddle-point excitons) [11,12,17,24], has also been performed and will be published elsewhere [25].

Another application of Eqs. (11) and (12) is the determination of the refractive index  $n(E)$  and of  $n'(E) = n + E dn/dE$  [20], of utmost importance in the design of solid-state lasers, optical modulators, multilayered devices, etc. The particular cases of AlGaAs and InGaAsP will be addressed in forthcoming papers [25]. Finally, the method developed in the first part of this Letter should provide an easy way to calculate expressions similar to Eq. (1), like those appearing in Raman scattering or those corresponding to potentials different from the Coulomb interaction [25].

In conclusion, the expression of the complex dielectric constant of Wannier excitons has been calculated exactly. The stage is now set for a better determination of the optical properties of direct band gap semiconductors near the absorption threshold, with dispersion effects fully taken into account.

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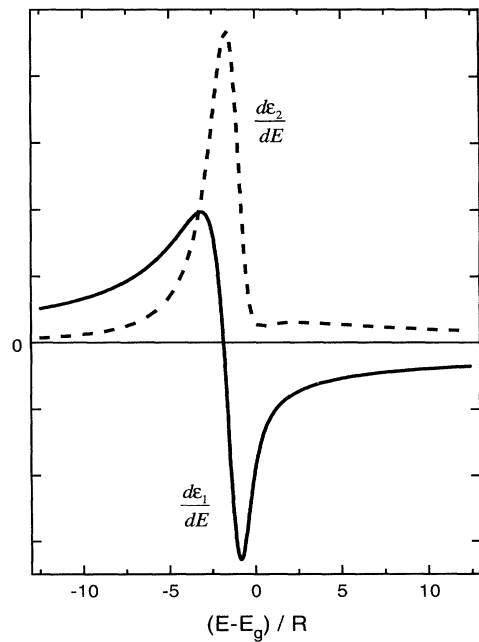


FIG. 2. Derivatives of  $\epsilon_1$  and  $\epsilon_2$  with respect to energy (linear scale). Same parameters as in Fig. 1.

*Note added.*—In a recent work Zimmerman gives an analytic expression for the continuum contribution to the complex susceptibility [26] similar to Eq. (7).

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