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COMMENT

Comment on 'Modelling the optical constants of GaAs: excitonic effects at E_1, $E_1 + \Delta_1$ critical points'

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Abstract

Djurišić and Li [1] have recently presented a calculation of the optical constants of GaAs in which they do not take into account excitonic effects at either E_1, $E_1 + \Delta_1$ or E_0, $E_0 + \Delta_0$ critical points (CPs). They employ band-to-band single-particle expressions with an energy-dependent pseudo-Gaussian broadening function. Their paper states that including excitonic effects at the former CPs has 'dubious physical interpretation'. It is also claimed that (a) 'excitonic effects are usually more pronounced at E_0, $E_0 + \Delta_0$ than at the E_1, $E_1 + \Delta_1$ CPs', (b) 'at room temperature the excitons are severely broadened and *should not* contribute significantly to the dielectric function' and (c) 'excitonic effects at E_1, $E_1 + \Delta_1$ critical points do not represent a significant contribution to the dielectric function at room temperature for materials with low exciton binding energy'. These statements are completely incorrect, as is their fitting scheme.

It has been well known for about 30 years that even at 300 K the '$E_1/E_1 + \Delta_1$' optical features associated with the E_1, $E_1 + \Delta_1$ CPs are primarily excitonic in nature [2–7]. This point is discussed extensively in [2]. As shown in [4], the polarization dependence of the effects of uniaxial stress along [001] on the '$E_1/E_1 + \Delta_1$' optical features could not be explained by band-to-band single-particle (BBSP) transitions. This conclusion was based on symmetry arguments, not any details of a lineshape fit. The fundamental reason why these optical features, even at room and elevated [7] temperatures, are primarily excitonic was first pointed out in [3], a paper that has been largely ignored. The exciton Rydberg energy (R_1) for these CPs is about a factor of ten larger than R_0, the Rydberg energy of the E_0 exciton. This difference is a consequence of the two-dimensional nature of the former CPs plus the larger effective masses (transverse) in relation to the masses at E_0. The excitonic nature of the '$E_1/E_1 + \Delta_1$' optical features is dramatically illustrated in the recent first-principles calculation of [5]. By employing the correct lineshape for the optical features associated with the E_1, $E_1 + \Delta_1$ CPs, the Brooklyn College group has been able to evaluate R_1 for a number of zincblende-type materials, e.g. from 30 meV for $Ga_{0.85}In_{0.15}As_{0.14}Sb_{0.86}$ [8] up to 270 meV for $Zn_{0.53}Cd_{0.47}Se$/InP [9]. The deduced values of R_1 are in reasonable agreement with a simple effective mass/$\vec{k} \cdot \vec{p}$ calculation [8–13]. This lineshape consists of both discrete exciton (DE) and two-dimensional band-to-band Coulomb enhanced (BBCE)/continuum exciton terms.

Excitonic effects at the E_0 CP also must be included, even at room temperature. It has been known for more than 40 years that, in the presence of a DE, the band-to-band E_0 lineshape (within about 6–8 R_0) is changed from the BBSP square root (broadened) term to a three-dimensional BBCE expression, which has a lineshape similar to a step function

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(broadened) and also increases the amplitude of the absorption in relation to the BBSP expression [2, 9, 14, 15]. A question that remained open was the nature of the lineshape if the E_0 exciton was not resolved. This was one of the points addressed in [11] and [13], both of which demonstrated conclusively that even if the exciton at E_0 is not resolved the lineshape is BBCE and not BBSP. The nature of the BBCE lineshape also is clearly illustrated in figure 2 of [16], which shows the experimental (300 K) and fit values of ϵ_2 (imaginary component of the complex dielectric function) and first, second and third derivatives (with respect to energy) in the vicinity of E_0 of ZnSe. The fitting approach of [16] uses both Lorentzian broadening (LB) and Gaussian broadening (GB) terms. The experimental lineshape is clearly a broadened steplike function, i.e. BBCE, while the fit, which is not very good, is BBSP. Similarly the first-derivative experimental curve is almost symmetrical while the fit is asymmetrical on the high-energy side. This result is similar to the point made in figure 2 of [11] and figure 2 of [13]. Furthermore, figure 3 of [16] presents the results of a fit to BBSP plus DE terms, with separate amplitudes, LB and GB for each contribution. Even with the introduction of the DE (and the associated additional fitting parameters) the agreement between calculation and experiment is still not very good. It is clear that a BBCE term would correspond more closely to the data. It should also be mentioned that in the Elliot formalism (DE plus BBCE) the same amplitude factor applies to both terms [2, 14].

Djurišić and Li [1] have replaced the Lorentzian broadening terms in equations (4), (6) and (7) in [1] with an energy-dependent PGB term:

$$\Gamma_j(E) = \Gamma_j \exp\left[-\alpha_j \left(\frac{E - E_j}{\Gamma_j}\right)^2\right] \qquad (1)$$

where the index j refers to a particular CP, Γ_j and α_j are adjustable parameters and E_j is the energy of the CP. If $\alpha = 0$, Lorentzian broadening is generated while for $\alpha = 0.3$ it produces a Gaussian lineshape [17]. There are several problems with this procedure. For example, in [1] a value of α_0 ($E_0/E_0 + \Delta_0$ CPs) of 6.396 is reported. This value is far outside the range of $0 < \alpha < 0.3$ in [17]. There is no discussion of such a high value for this parameter. In addition, the cited values of α are really meaningless since the authors have started with an incorrect lineshape function. Furthermore, by using equation (1) with $\alpha \neq 0$ the real and imaginary components of the dielectric function are no longer Kramers–Kronig transforms of each other.

Djurišić and Li [1] have not even determined the energies of the E_0, $E_0 + \Delta_0$, E_1, and $E_1 + \Delta_1$ CPs from a fit to the data but have used values already reported in the literature.

The work of [1] demonstrates that, even by starting with a physically incorrect model, reasonably good fits to experimental optical data can be achieved by using enough adjustable parameters.

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