Comment on ''Optical properties of $CdTe_{1-x}S_x$ $(0 \leq x \leq 1)$: Experiment and modeling'' [J. Appl. Phys. 85, 7418 (1999)]

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Wei *et al.* [J. Appl. Phys. **85**, 7418 (1999)] performed the room temperature spectroscopic ellipsometry to determine the dielectric function of $CdTe_{1-x}S_x$ films. They have fit the obtained dielectric function using the Holden's model dielectric function [Phys. Rev. B **56**, 4037 (1997)], and derived conclusions about the line shape at the band gap E_0. However, their description of the fitting procedure is ambiguous, and some model parameters in Table I [J. Appl. Phys. **85**, 7418 (1999)] are missing which makes it impossible to reproduce their calculations. Furthermore, the results of Wei *et al.* [J. Appl. Phys. **85**, 7418 (1999)] do not represent conclusive proof of the advantages of their approach over other models available in the literature. © *2000 American Institute of Physics.* [S0021-8979(00)00116-X]

Wei *et al.*[1] have presented their results in modeling the optical functions of $CdTe_{1-x}S_x$ $(0 \leq x \leq 1)$ using Holden's dielectric function model.[2] They conclude that the correct line shape at the band gap E_0 has to include band-to-band Coloumb enhancement effects even if the exciton is not resolved. The concept of a physically sound model which is capable of describing accurately the optical functions over a wide spectral range is certainly very attractive. However, some clarifications on the model equations and fitting procedure are needed in order to enable application of the results of Wei *et al.*[1] to other materials and confirming their conclusions. At present it is not possible even to reproduce their calculations, because parameters A_0, B_0, C_1, C_2, and F_2 are not given in Table I in Ref. 1 [it can be assumed that parameter Γ_2 is actually the γ in Eq. (3)].

Wei *et al.*[1] state that they have used the modified version of analytical form given by Eqs. (A7) and (A12) in the work of Holden *et al.*[2] (though later in the text it is stated that solid line in Fig. 1 are fits to equations given in Holden *et al.*[2] and no modification is mentioned, and in several places model equations are referred to a wrong reference, Ref. 9 instead of Ref. 8). If a modified version was used, it should be given in the paper. Holden *et al.*[2] have eliminated the significant contribution to the imaginary part of the dielectric function ε_2 below the E_0 gap by introducing the linear cutoff for the contributions of E_{ind} (indirect transitions), E_1, $E_1 + \Delta_1$, and E_2 critical points. If a modification of the model was made to enable avoiding significant contributions to the imaginary part of the dielectric function below the E_0 gap in a different way, this important point should have been clearly stated in the article. In any case, the manner of eliminating these contributions should be discussed, and exact equations used for modeling explicitly given if they are different from the quoted reference.

Other serious observation are conclusions of Wei *et al.*[1] about the broadening. They claim that the Lorentzian broadening represents accurate description of the absorption processes in semiconductors. However, Holden's model dielectric function[2] exhibits extended absorption tail below the fundamental band gap (see Fig. 4 in Ref. 2, scale used in Ref. 1 prevents observing any details) which is characteristic for the Lorentzian function and does not describe well the experimental data. Kim and Sivananthan[3] demonstrate that a significant improvement in the region of the E_0 gap is obtained by assuming the Gaussian-like broadening. Wei *et al.*[1] quote the work of Aspnes[4] to support the statement that no evidence of Gaussian broadening has been found in CdTe. This is incorrect, because Aspnes[4] investigated only the E_1 and $E_1 + \Delta_1$ structure, and not the E_0 where the greatest benefit of the Gaussian-like broadening can be observed. It has also been shown that introduction of Gaussian-like broadening eliminates extended absorption tail in the vicinity of E_0 not only for model of Kim *et al.*,[3,5–7] but also for a modification of Adachi's model.[8] Furthermore, there are some serious questions about the validity of Fourier analysis approach used by Aspnes[4] for the determination of the type of broadening. The dependence of a logarithm of Fourier coefficients $\log_{10} C_n$ on the order n is not independent on the false data and baseline correction (false data have been used to isolate the critical point structure, and obtained results are dependent on the choice of energies of boundaries outside which false data are used) and also the number of data points and the width of the spectral region considered, even for a simple Lorentzian or Gaussian function, not to mention the real experimental data. This is demonstrated in Fig. 1, which shows $\log_{10} C_n$ vs n for the same function but with different boundaries of the spectral region considered. In all cases the same number of the data points was generated and the function could be accurately reproduced by Fourier series. Figure 2 shows $\log_{10} C_n$ as a function of n for a range of functions
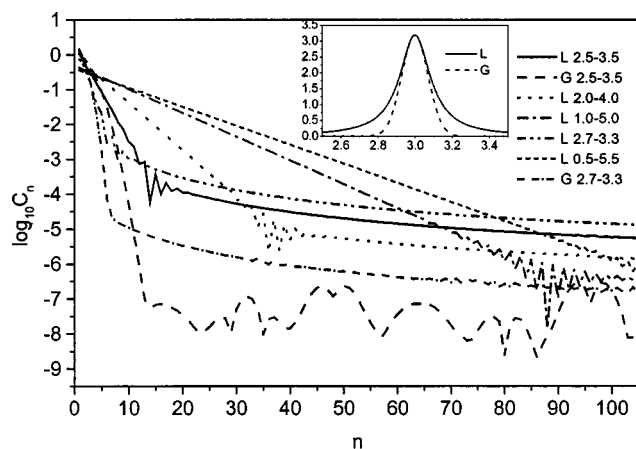
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FIG. 1. $\log_{10} C_n$ as a function of n for the Lorentzian (L) and Gaussian (G) function; curves differ in the width of the spectral region considered. The inset shows the functions (L) and (G) vs energy.

FIG. 3. The first derivative of the imaginary part of the dielectric function of GaP vs energy. Solid line denotes the experimental values, dashed line is the derivative of modified Adachi's model (Ref. 12).

where the damping constant Γ has been replaced with the energy dependent damping[3] $\Gamma(E) = \Gamma \exp\{-\alpha[(E-E_g)/\Gamma]^2\}$. It can be clearly observed that the linearity of $\log_{10} C_n$ does not necessarily imply that a line shape is pure Lorentzian (note similarity between the Lorentzian curve and curves for $\alpha = 0.1$ and 0.2). Also, it has been shown that the best results in fitting the third derivative of the room temperature dielectric function of GaAs and CdTe are obtained with a linear combination of 75% Gaussian and 25% Lorentzian representations.[9] Therefore, Fourier analysis proposed by Aspnes[4] can be useful for the determination of energies of critical points, but caution is needed in deriving any conclusions on the broadening function and broadening parameter Γ. Also, it should be noted that Kim and Sivananthan[3] obtained excellent fit using their model for their CdTe dielectric function data, which probably correspond to a sample of better quality, according to the criterion of Aspnes and Studna.[10]

Also, model of Kim et al.[3,5–7] is to our knowledge the best model for successfully describing the dielectric function and its derivatives at the same time. The fit of the derivatives
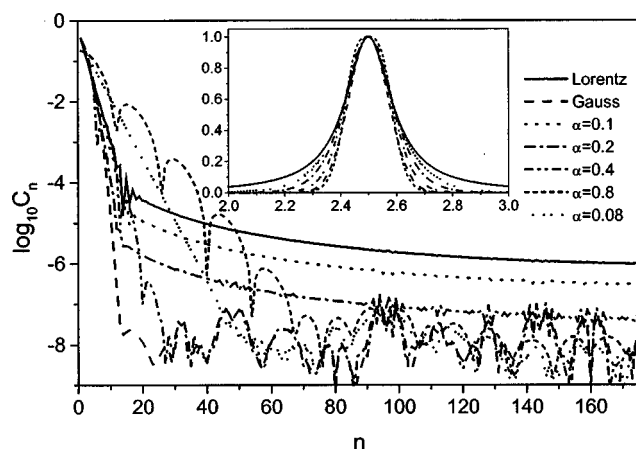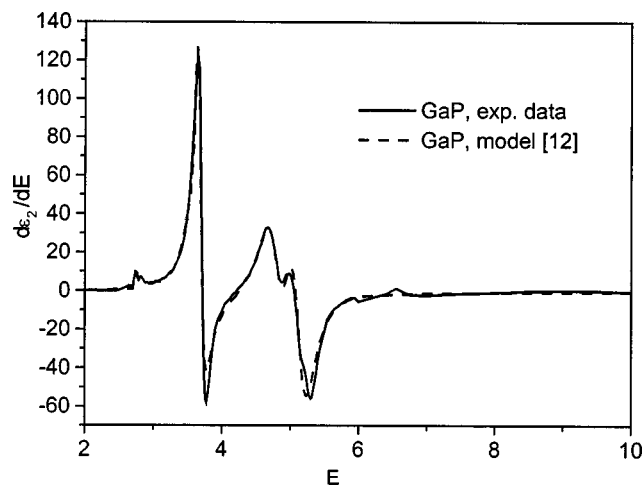

FIG. 2. $\log_{10} C_n$ as a function of n for a range of broadening functions: the Lorentzian, Gaussian, and functions with different α, which are shown in the inset. The broadening parameter $\Gamma = 0.1$ for all functions.

in the work of Kim et al.[3,5–7] exhibits significantly better agreement with the experimental data than fits of Holden et al.[2] or Wei et al.[1] There is additional key difference (except the model employed) in the work of Kim et al.[3,5–7] and Wei et al.[1] in fitting the derivatives. Garland et al.[11] have emphasized the importance of using the numerical derivatives for both the experimental data and the model, and this approach has been used in the work of Kim et al.[3,5–7] to fit at the same time the dielectric function and its three derivatives. Wei et al.[1] fitted the analytical expression for the derivatives, determined initial parameter values and then fitted the dielectric function (some parameters describing the structures which are not pronounced in the experimental data have been kept fixed). Furthermore, the argument of Wei et al.[1] that the line shape is definitely band-to-band Coloumb enhanced (BBCE) one based on comparison with the derivative spectra is definitely inconclusive. They compare the experimental first derivative and calculated derivative of band-to-band single particle (BBSP) function, and the experimental second derivative and calculated BBCE second derivative. The second derivative of BBSP and the first derivative of BBCE are not shown. They have used only the $\text{Im}(E-E_0+i\Gamma_0)^{1/2}$ term as a BBSP function, which is, in our opinion, not a valid argument to label all the BBSP approaches as incorrect. Figure 3 shows the numerical derivatives of the dielectric function of GaP, solid line is the derivative of the experimental data while the dashed line is the derivative of a modified Adachi's model.[12] Good agreement with the experimental data can be clearly observed, so in terms of the agreement with either the experimental dielectric function or its derivative, there is no obvious advantage of BBCE over BBSP approach.

Therefore, Wei et al.[1] did not present conclusive evidence that the BBCE approach is superior over the BBSP approach in terms of the agreement with the experiment. Also, no conclusive proof is given that the broadening is purely Lorentzian. However, BBCE approach enables obtaining some important material parameters such as Rydberg energy R_1, for which BBSP approach including discrete ex-

citon terms is not as accurate. This would undoubtedly be very useful if the authors would clarify the matters concerning the actual model equations used in the fitting procedure.

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