Linear and nonlinear optical properties of $(GaAs)_m/(AlAs)_n$ superlattices

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We report a full-band-structure calculation of frequency-dependent second-harmonic generation in short-period $(GaAs)_m/(AlAs)_n$ superlattices. We use a linear combination of Gaussian orbitals technique, in conjunction with a linearized sampling method, to obtain the dielectric function $\vec{\epsilon}(\omega)$ and the second-order optical response coefficient $\vec{\chi}^{(2)}(-2\omega;\omega,\omega)$. Our results show that the effect of the unique superlattice transitions is more pronounced in $\vec{\chi}^{(2)}(-2\omega;\omega,\omega)$ than in $\vec{\epsilon}(\omega)$.

In recent years there have been many theoretical and experimental investigations of electronic properties of $(GaAs)_m/(AlAs)_n$ superlattices.[1-6] Yet, to our knowledge, there are only three full-band-structure calculations of the dielectric function over a wide range of frequencies.[2,3,6] And there are *no* such calculations for the nonlinear optical properties of these superlattices. In fact, although the field of nonlinear optics is over 30 years old, in general there are few full-band-structure calculations of nonlinear optical properties of semiconducting materials, be they bulk or superlattice. In the past we have carried out such calculations for second-harmonic generation (SHG) in bulk semiconductors.[7] More recently we have reported on calculations of frequency-dependent third-harmonic generation in bulk semiconductors[8] and a full-band-structure calculation of SHG $[\chi^{(2)}(-2\omega;\omega,\omega)]$ in a series of odd-period strained $(Si)_n/(Ge)_n$ (001) superlattices.[9]

In the present work, we present full-band-structure calculations of frequency-dependent SHG in short-period $(GaAs)_m/(AlAs)_n$, with $(m,n)=(1,1)$, (2,1), and (2.2), along with the corresponding linear dielectric functions. We compare our calculated linear dielectric functions to experimentally obtained results.[3,4] Our results show that SHG is a much more sensitive measure of the contributions of the "pure superlattice" effects than the dielectric function.

Since the full details of our approach for calculating electronic band structures and evaluating optical response functions are given in the literature,[8-10] here we only briefly describe our method. We use a linear combination of Gaussian orbitals technique in conjunction with the $X\alpha$ method for constructing the potentials of the constituent *bulk* materials. The local single-site effective potentials and basis functions for bulk GaAs and AlAs are constructed by adjusting the α's for bulk materials to produce the correct two lowest bulk band gaps. We then use these bulk orbitals and potentials to construct the superlattice basis and effective potentials. Therefore our approach involves four fitting parameters. In contrast, the empirical-pseudopotential calculations of Caruthers and Lin-Chung[2] employed nine fitted form factors. Alouani *et al.*[3] added external potentials to their self-consistent potentials. The strengths of these potentials were fitted such that the band gaps of each of the constituent bulk materials at three symmetry points agree with experiment (i.e., a total of six points). Xia and Chang[6] employed 48 fitting parameters (nineteen parameters for the band structure of each bulk material, nine parameters for optical matrix elements, and one parameter for valence-band offset).

Note that in our approach we do not carry out any fitting to the superlattice properties and, unlike in self-consistent local-density-approximation (LDA) calculations, our band gaps are *not* underestimated. Our calculated values of the direct band gaps, which are of importance in calculating optical properties, are given in Table I along with the corresponding experimental values;[3,4] our results are in very good agreement with experiment.

We use a density-matrix formalism, within the independent-particle approximation, and the minimal-coupling interaction Hamiltonian[7-10] to derive expres-

TABLE I. Theoretical and experimental (Ref. 3) results for direct band gaps (eV).

(m,n)	(1,1)	(2,1)	(2,2)
Theory	2.04	1.82	2.01
Experiment	2.07	1.85	2.08

sions for the linear and nonlinear optical response coefficients. We evaluate these expressions utilizing a linearized sampling method[8,9] to integrate over an irreducible segment of the Brillouin zone (IBZ), first evaluating the imaginary parts of the linear and second-order optical response tensors,[8–10] and then using Kramers-Kronig relations to obtain the real parts. Our past experience[10] in using these types of band structures leads us to expect that the actual magnitudes of the dielectric functions are about double what we have calculated. Therefore, in the present work we have multiplied our results for the dielectric functions by a factor of 2 for comparison with experiment.

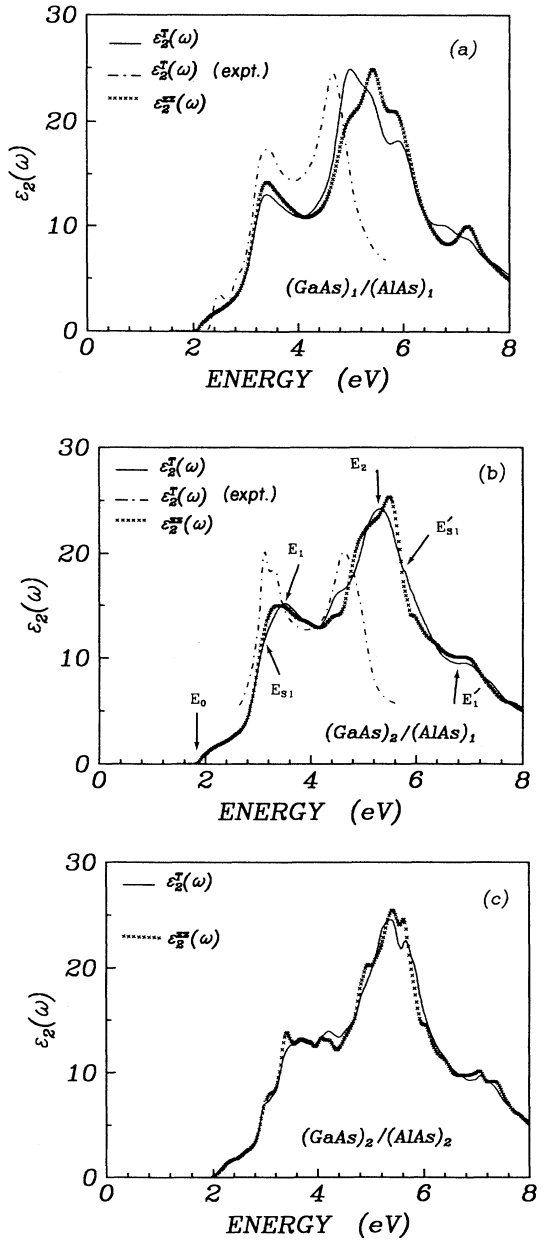

FIG. 1. $\vec{\epsilon}_2(\omega)$ for (a) (1,1); (b) (2,1); (c) (2,2) superlattices. The theoretical results have been multiplied by a factor of 2 (see text). The experimental results are from (a) Ref. 4 and (b) Ref. 3.

We present the results of our full-band-structure calculation of the imaginary part of the dielectric tensor, $\vec{\epsilon}_2(\omega)$, for $(m,n)=(1,1)$, (2,1), and (2,2), and the corresponding experimental results of Garriga *et al.*[4] $[(m,n)=(1,1)]$, and Alouani *et al.*[3] $[(m,n)=(2,1)]$ in Fig. 1. Although some of the peaks in our calculation lie at slightly higher energies than the measured ones, overall there is good agreement between theory and experiment, particularly in view of the following facts. (a) The experimental results were extracted from ellipsometric data using a two-phase model which, as discussed by the authors, is not completely valid.[4] (b) Difficulties in growing uniform superlattice samples[5] could contribute to a discrepancy between theoretical results and *any* experimental measurements. However, part of the problem is due to the fact that in our calculations some of the higher conduction bands lie at energies that, compared to the results of more accurate band structures,[1] are slightly high. A more detailed discussion of the likely source of discrepancy between experiment and calculations of this sort has been presented elsewhere.[9]

We would also like to point out that our results are in good overall agreement with calculated values of Alouani *et al.*[3] For the purpose of comparison we have used their labeling scheme to identify different peaks of $\vec{\epsilon}_2(\omega)$ for $(m,n)=(2,1)$. As seen from Fig. 1(b) (cf. Fig. 1 of Ref. 3), there is one-to-one correspondence between the peaks from their calculation and ours. Also, for $(m,n)=(1,1)$, we find in agreement with Alouani *et al.*[3] that the E_{SL} shoulder is absent. For a complete analysis of the source of different peaks in the dielectric functions, we refer the reader to their work. Both from our results and those of Alouani *et al.* it is evident that there is only a small difference between $\epsilon_2^{zz}(\omega)$ and $\epsilon_2^T(\omega)\equiv\epsilon_2^{xx}=\epsilon_2^{yy}$, where z is the growth direction of the superlattice. This is in contrast to the calculated results of Caruthers and Lin-Chung,[2] which show a large anisotropy. In a recent publication we analyzed the source and size of anisotropy of $\vec{\epsilon}(\omega)$ for strained $[(\mathrm{Si})_n/(\mathrm{Ge})_n]/\mathrm{Si}$ (001) superlattices.[10] We showed that most of the anisotropy is due to strain in Ge, rather than to the lower symmetry of the superlattices. Since the strain in AlAs layers is negligible, we expect that the anisotropy in $(\mathrm{GaAs})_m/(\mathrm{AlAs})_n$ superlattices should be very small, in agreement with the results of Alouani *et al.* and our own.

The results of our full-band-structure calculation of the magnitude of $\vec{\chi}^{(2)}(-2\omega;\omega,\omega)$, for $(m,n)=(1,1)$, (2,1), and (2,2) are presented in Fig. 2. Our calculated values of $\vec{\chi}^{(2)}(0)$ for these superlattices and bulk GaAs and AlAs are given in Table II. Note that for the superlattices considered in this work, the tensor $\vec{\chi}^{(2)}(-2\omega;\omega,\omega)$ has only two independent components, which can be chosen to be $\chi^{(2)}_{xyz}(\omega)$ and $\chi^{(2)}_{zyx}(\omega)$.

Since the peaks for the linear response of $(\mathrm{GaAs})_2/(\mathrm{AlAs})_1$ have already been identified,[3] here we shall concentrate on identifying the nonlinear features of

this superlattice. The first peak (1) at around 0.93 eV is due to the 2ω resonance with the E_0 optical peak. The shoulder (2) at around 1.55 eV arises from a 2ω resonance with the superlattice E_{SL} optical peak. The three peaks (3) in the energy range 1.55–2.3 eV are due to contributions from ω resonance with E_0, and 2ω resonances with E_1 and E_0' optical peaks. The major feature (4) at about 2.8 eV is due to 2ω resonances with E_2 and E_{SL}' peaks. Finally, the last prominent peak (5) at around 3.3 eV is due to ω resonances with E_{SL} and a 2ω resonance with E_1'. Note that, for $(m,n)=(1,1)$ superlattice, as in the linear case the E_{SL} optical peak is absent.

In order to clearly distinguish the superlattice features from the bulklike features, we use an effective-medium model to calculate the SHG in these superlattices from the corresponding response of their bulk constituent materials. The model is essentially an extension of an effective-medium model for the linear response.[10] By following the same procedure as for the linear response,[10] the x component of the second-order effective polarization in a material which consists of m macroscopic layers of bulk material A and n macroscopic layers of bulk material B is given by

$$P_x^{(2)\text{eff}} = \left[\frac{\epsilon_{zz}^{\text{eff}}}{m+n} \left(\frac{m\chi_{xyz}^{A}}{\epsilon_{zz}^{A}} + \frac{n\chi_{xyz}^{B}}{\epsilon_{zz}^{B}} \right) \right] E_y^{\text{eff}} E_z^{\text{eff}} ,$$

where E_y^{eff} (E_z^{eff}) is the electric field perpendicular (parallel) to the crystal axis and ϵ_{zz}^{A} is the zz component of the dielectric tensor. The quantity in the large square brackets gives χ_{xyz}^{eff}.

To evaluate this expression, we first obtain $\vec{\vec{\epsilon}}(\omega)$ and $\vec{\vec{\chi}}^{(2)}(-2\omega;\omega,\omega)$ for bulk AlAs and GaAs from full-band-structure calculations. We then use these results to calculate $\vec{\vec{\epsilon}}_{\text{eff}}(\omega)$ and $\vec{\vec{\chi}}_{\text{eff}}^{(2)}(-2\omega;\omega,\omega)$. In Fig. 3 we have plotted the magnitude of $\chi_{xyz}^{(2)}(\omega)$ for $(m,n)=(2,1)$, from our full-band-structure calculation along with the corresponding response from the effective-medium model. As expected, the model fails to predict the contributions of the E_{SL} and E_{SL}' optical resonances to the SHG and this leads to substantial differences at certain energies between the model and the superlattice full-band-structure calculations. In particular note that, due to contributions from the zone-folded states, peaks (4) and (5) of the superlattices SHG have been considerably enhanced in size as compared with the model. Also note that the effective-medium model fails to predict the energy of the direct absorption edge. From Table II it is evident that as the

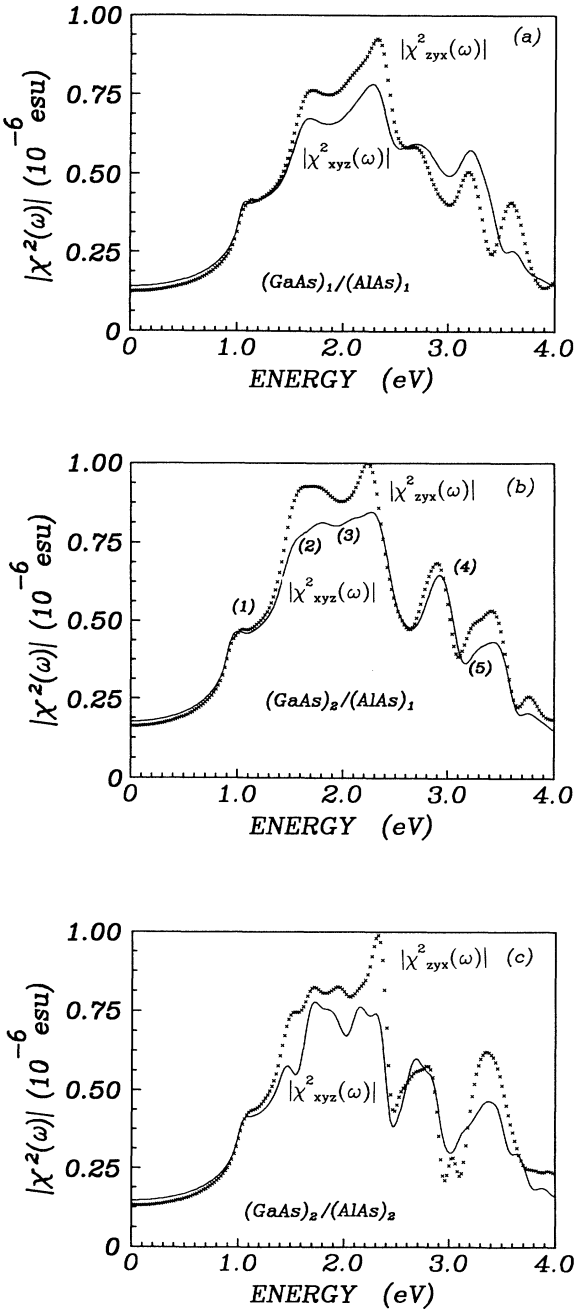

FIG. 2. $|\vec{\vec{\chi}}^{(2)}(-2\omega;\omega,\omega)|$ for (a) (1,1); (b) (2,1); (c) (2,2) superlattices.

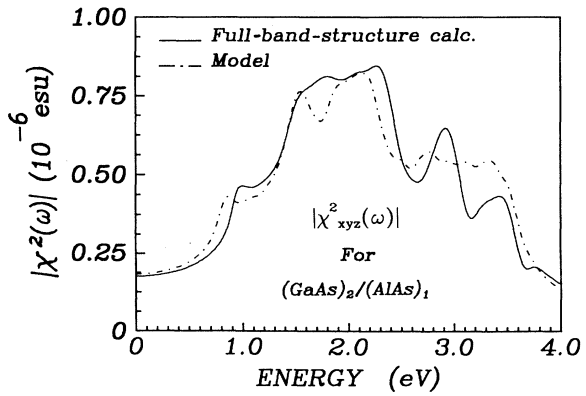

FIG. 3. $|\chi_{xyz}^{(2)}(\omega)|$ for $(GaAs)_2(AlAs)_1$ from the full-band-structure calculation and effective-medium model.

TABLE II. Theoretical results for $\vec{\chi}^{(2)}(0)$ in units of 10^{-7} esu.

	(1,1)	(2,1)	(2,2)	GaAs	AlAs
$\chi_{xyz}(0)$	1.41	1.76	1.47	2.49	0.72
$\chi_{zyx}(0)$	1.26	1.62	1.31	2.49	0.72

number of AlAs layers increases, the nonlinear response decreases. This is due to the fact that $\vec{\chi}^{(2)}(0)$ in AlAs is smaller than in GaAs. Finally, as in the case of linear response the anisotropy in the second-order response is small.

In conclusion, we have carried out a full-band-structure calculation of $\vec{\epsilon}_2(\omega)$ and a calculation of $\vec{\chi}^{(2)}(-2\omega;\omega,\omega)$ in short-period $(\mathrm{GaAs})_m/(\mathrm{AlAs})_n$ superlattices. Our results for dielectric function are in good agreement with experimental measurements and other calculations. Apart from the features at the absorption edge and the features due to E_{SL} and E'_{SL} optical peaks, the linear and second-order optical responses can be thought of as an effective-medium average of the corresponding responses of bulk GaAs and AlAs. Our results show that the effect of the unique superlattice transitions are more pronounced in $\vec{\chi}^{(2)}(-2\omega;\omega,\omega)$ than in $\vec{\epsilon}(\omega)$.

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