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Theory of quasiequilibrium nonlinear optical absorption in semiconductor superlattices

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Quasiequilibrium nonlinear optical absorption spectra are computed for semiconductor superlattices. The theory generalizes the semiconductor Bloch equations to describe anisotropic structures. The equation for the interband polarization is solved numerically and the carrier-density dependent optical nonlinearities are computed. Starting from excitonic absorption, with increasing density exciton saturation and the development of gain is observed. The dependence of the gain spectra on structural parameters of the superlattice is discussed. © *1995 American Institute of Physics.*

Semiconductor heterostructures have received interest for potential applications in optoelectronic devices, since many of their properties can be designed by suitable tailoring of the structure, see e.g. Ref. 1. The operation of resonant nonlinear optoelectronic devices relies on the large nonlinearities in the spectral vicinity of the fundamental absorption edge. For feasibility studies, design, and optimization of possible devices, a fundamental model for the nonlinear optical material response is needed. Any realistic modeling has to include the many-body Coulomb-interaction effects.[2] Such a theory, which is based on the semiconductor Bloch equations, exists for bulk semiconductors[2] and quantum wells.[3] However, for semiconductor superlattices up to now only the linear optical properties, i.e. the excitonic absorption in unexcited systems, have been analyzed in detail.

Early investigations[4] were based on an effective medium approach, which allows approximate analytical solutions of the Wannier equation, but does not fully account for the anisotropy of the underlying semiconductor structure. A more recent approach, which fully takes into account the anisotropy, concentrated on electric-field induced phenomena, i.e. excitonic Wannier-Stark ladders.[5,6] Although there are a number of experimental investigations of nonlinear optical absorption spectra of superlattices,[7–9] up to now no systematic calculation of optical spectra of excited or inverted systems has been published.

In this letter, we analyze the nonlinear optical properties of semiconductor superlattices by numerically solving the semiconductor Bloch equations, which we have generalized to be valid also for anisotropic systems. The reduction of the Coulomb potential due to the screening of the attractive electron-hole (e-h) interaction, the band gap renormalization with increasing carrier density, and bandfilling effects are taken into account. The plasma screening is treated within the single-plasmon-pole approximation.[10]

Choosing the growth direction of the layers as the z axis, the carriers are unconfined in the xy plane and subject to the periodic confinement potential in the z direction. Accordingly, we factorize the wave functions as products of a plane wave for the free xy motion and a superlattice periodic z dependent function of Bloch type. Based on the Kronig-Penney model, the electronic states of the superlattice are computed within the effective mass approximation. The periodic potential is modeled as an infinite sequence of identical quantum wells of width w, which are separated by rectangular barries of width b, respectively.[11] The crystalline potential in the superlattice has the period, $d=w+b$, in the growth direction. The three-dimensional wave vector consists of the parallel component k_z, which varies in the Brillouin zone between $\pm\pi/d$, and the in-plane component $\mathbf{k}_\perp$.

For the analysis of quasiequilibrium optical nonlinearities in this letter, we do not need to solve the full semiconductor Bloch equations since the distribution functions are assumed to be Fermi-Dirac like for given carrier density and plasma temperature. Hence it is sufficient to consider the equation for interband polarization $P_{\mathbf{k}}$, which determines the optical spectrum. For a two-band semiconductor the polarization equation is

$$i\hbar\frac{\partial}{\partial t}P_{\mathbf{k}}=\hbar P_{\mathbf{k}}|_{\text{scatt.}}+\hbar(e^e_{\mathbf{k}}+e^h_{-\mathbf{k}})P_{\mathbf{k}}+(1-n^e_{\mathbf{k}}-n^h_{-\mathbf{k}})\times\left[\mu_{\mathbf{k}}E(t)+\sum_{\mathbf{k}'}V_s\left(\begin{smallmatrix}e,&h,&h,&e\\ \mathbf{k},&-\mathbf{k}',&-\mathbf{k},&\mathbf{k}'\end{smallmatrix}\right)P_{\mathbf{k}'}\right]. \tag{1}$$

The factor $\mu_{\mathbf{k}}$ is the optical dipole matrix element between conduction and valence-band states, $E(t)$ is the probe field needed to measure the optical response and V_s is the screened Coulomb potential. $\hbar P_{\mathbf{k}}|_{\text{scatt.}}$, describes polarization decay processes, which we model by a dephasing time. The single-particle energies $\hbar e^e_{\mathbf{k}},\hbar e^h_{-\mathbf{k}}$ are renormalized by the screened-exchange and the Coulomb-hole term.[2] $n^e_{\mathbf{k}}$ ($n^h_{-\mathbf{k}}$) are the electron (hole) distribution functions.

To study the influence of Coulomb effects on the superlattice electron-hole excitations we need the Coulomb potential expanded in the basis of the superlattice states. Within the single-plasmon pole approximation,[10] the screened Coulomb potential between the conduction and the valence band states, with $\mathbf{q}=\mathbf{k}_\perp-\mathbf{k}'_\perp$ and $q_z=k_z-k'_z$, is given by

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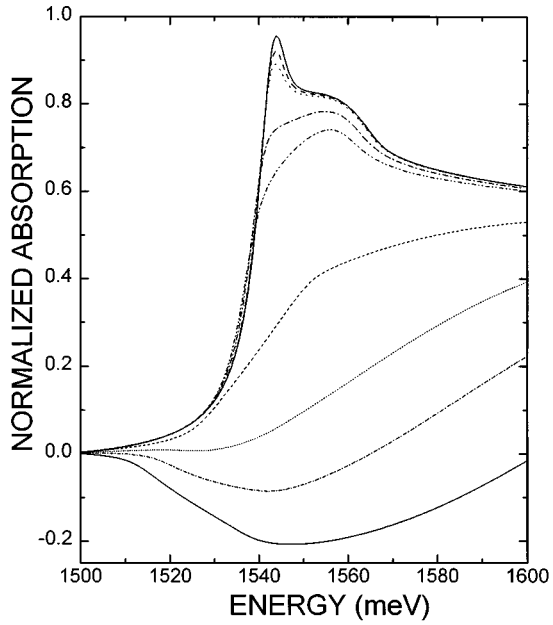

FIG. 1. Normalized absorption spectra α/α_0 of a GaAs/$Al_{0.3}Ga_{0.7}As$ superlattice with well (barrier) width 95(15)Å for various carrier densities at room temperature, dephasing time $T_2=150$ fs and $\alpha_0=10^4$ cm^{-1}. Densities: 0, 5×10^{15}, 1×10^{16}, 5×10^{16}, 1×10^{17}, 5×10^{17}, 1×10^{18}, 1.5×10^{18}, 2.2×10^{18} cm^{-3} (from top to bottom).

$$V_s\left(\begin{smallmatrix} e, & h, & h, & e \\ \mathbf{k}, & -\mathbf{k}', & -\mathbf{k}, & \mathbf{k}' \end{smallmatrix}\right)=\sum_G \frac{4\pi e^2 f_{e,e}^G(k_z,k_z')f_{h,h}^{-G}(-k_z',-k_z)}{\varepsilon} \times\frac{1+x[\mathbf{q}^2+(G+q_z)^2]}{\mathbf{q}^2+(G+q_z)^2+x[\mathbf{q}^2+(G+q_z)^2]^2+\kappa^2}. \tag{2}$$

G is a reciprocal lattice vector of the one-dimensional superlattice potential and $f_{\nu,\nu}^G(k_z,k_z')$ is the superlattice form factor of the product of two Bloch periodic functions.[11] Within the single-plasmon pole approximation $x=(C\kappa^2/4\hbar^2)/(4m_r\omega_{\rm pl}^2)$ and $\omega_{\rm pl}^2=4\pi ne^2/m_r$.[10] Here κ is the inverse of the screening length, n is the total density, $\omega_{\rm pl}$ is the plasma frequency, m_r is the reduced effective mass and ϵ is the background dielectric constant. The constant C is usually treated as a fitting parameter, here taken to be 4. In RPA κ is calculated self-consistently according to:

$$\kappa^2=-\frac{4\pi e^2}{\epsilon Ad}\sum_{\nu=e,h}\sum_{k_z}\sum_{\mathbf{k}_\perp}\frac{dn_{\mathbf{k}}^\nu}{dE}. \tag{3}$$

For the numerical calculations we have used the following structural parameters: we assume a GaAs/$Al_{0.3}Ga_{0.7}As$ superlattice, with 95 Å-well and 15 Å-barrier width. The confinement potentials for the electrons (holes) are 250 meV(123 meV). Effective masses for electrons in the well(barrier) material are 0.067 m_0(0.092 m_0), respectively. The corresponding values for the heavy holes are 0.38 m_0(0.41 m_0). The effective masses for the perpendicular direction have been determined by averaging the inverse well and barrier masses with well and barrier widths. For the electrons (holes) we obtain 0.070 m_0(0.38 m_0). With these parameters the combined miniband width of lowest electron and heavy hole miniband is 23 meV. The light holes have been neglected for simplicity.

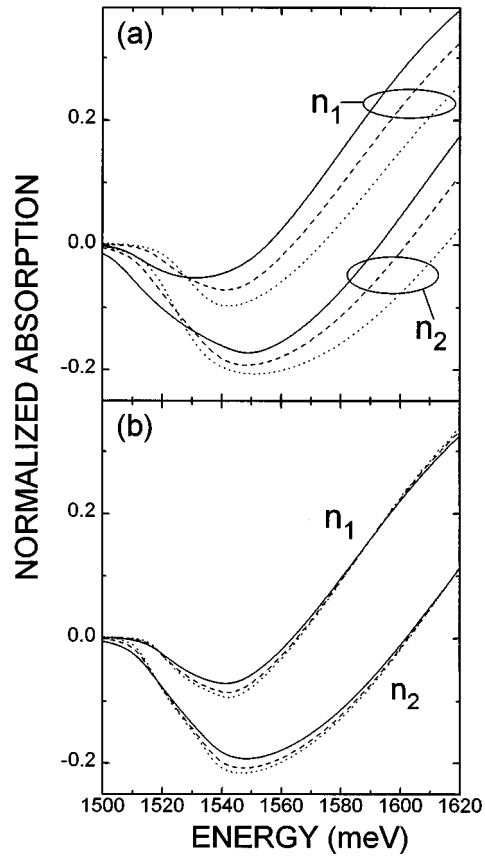

FIG. 2. (a) Normalized absorption spectra α/α_0 of GaAs/$Al_{0.3}Ga_{0.7}As$ superlattices with different barrier widths 8 Å (solid), 15 Å (dashed) and 26 Å (dotted) and constant well width of 95 Å at room temperature, for two different densities: $n_1=1.5\times10^{18}$ cm^{-3}, $n_2=2.2\times10^{18}$ cm^{-3}. (b) Normalized absorption spectra α/α_0 of a GaAs/$Al_{0.3}Ga_{0.7}As$ superlattice with well (barrier) width of 95 (15)Å for two densities (n_1,n_2) and different dephasing times 100 fs (solid), 150 fs (dashed), and 200 fs (dotted).

Numerically, the solution of the integrodifferential equations is performed on a grid in k space. In order to obtain good resolution in the calculated spectra we typically used 32 points for k_z and 64 points for $k_\perp$. This means that we have a k grid with 2048 points, which we use to compute the superlattice dispersion and wave functions, and the optical and Coulomb-matrix elements for the superlattice states. Since the Coulomb-matrix element depends separately on $\mathbf{k}$ and $\mathbf{k}'$ the storage of this calculation requires substantial computer memory. After the initial calculations where we set up the matrix elements, we solve Eq. (1) by a Runge-Kutta integration and obtain the time-dependent total polarization $P(t)$, which is given by $P(t)=\Sigma_{\mathbf{k}}\mu_{\mathbf{k}}^*P_{\mathbf{k}}(t)$.[12] Via a Fourier transform we then calculate the optical susceptibility $\chi(\omega)=P(\omega)/E(\omega)$. The absorption $\alpha(\omega)$ is determined by the imaginary part of $\chi(\omega)$

$$\alpha(\omega)=\frac{4\pi\omega}{n_b c}\chi''(\omega). \tag{4}$$

Here c is the speed of light and n_b is the background refractive index. The typical CPU time on a DEC AXP 3600 workstation is about 5 hours for one spectrum.

Figure 1 shows the absorption spectra of a GaAs/AlGaAs superlattice for various plasma densities at room temperature. For zero density the absorption exhibits a pronounced peak, corresponding to the anisotropic superlattice exciton. The high energy shoulder is related to the miniband dispersion.[5] With increasing carrier density the strength of the excitonic absorption decreases due to phase-space filling and screening of the attractive e-h interaction. Above the Mott-density (here about 5×10^{16} cm^{-3}), the exciton completely disappears from the spectrum. For densities larger than about 10^{18} cm^{-3}, Fig. 1 shows optical gain. The structure of the optical gain curves below the gain maximum reflects the superlattice density of states. At the band gap the frequency dependence of the density of states exhibits a square-root behavior similar to the case of bulk semiconductors. For energies which are more than one miniband width above the band gap the density of states becomes constant, i.e. effectively two dimensional.

In Fig. 2(a) the gain spectra are compared for different barrier widths. For a barrier width of 8 Å the coupling between neighboring wells is larger than for the configuration with a barrier width of 15 Å. Consequently, the combined miniband width increases to about 39 meV and the carriers in the vicinity of the band edge behave more like three-dimensional carriers. This explains the strong blue shift of the gain maximum with increasing carrier density for this structure. On the other hand, for a barrier width of 26 Å, there is only small coupling between neighboring quantum wells and the miniband width is only about 10 meV. In two dimensions the blue-shift of the gain maximum is known to be smaller than in three dimensions, which is in agreement with our results shown in Fig. 2(a). As expected the amplitude of the gain maximum increases with increasing barrier width, i.e. decreasing miniband width.

In Fig. 2(b) we compare the gain spectra for three different choices for the dephasing time T_2, 100 fs, 150 fs, and 200 fs, which are typical values in this regime. It is seen that the amplitude of the gain has a weak dependence on the dephasing time, but the shape of the spectra is not sensitive. This demonstrates that our results and interpretations are not strongly influenced by the specific choice of the dephasing time.

In conclusion we have presented a theory which gives a good description of the nonlinear optical absorption of semiconductor superlattices. The transition between the excitonic absorption to the gain regime, which takes place with increasing density, has been studied. It has been shown that the anisotropy of the superlattice structure strongly influences the gain spectra.

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[12] For light propagating in the growth direction of the superlattice the optical dipole matrix element is given by $\mu_{\mathbf{k}}=\mu_0 f_{e,h}^{G=0}(k_z,k_z)$, f is again the superlattice form factor (Ref. 11). Using the longitudinal-transverse splitting in GaAs $\Delta_{\mathrm{LT}}=0.08$ mV, the exciton Bohr radius of 132 Å and the background dielectric constant $\epsilon=12.9$, we have determined $\mu_0/e=4.5$ Å, e is the electron charge.