Temperature and many-body effects on the intersubband transition in a GaAs/Al$_{0.3}$Ga$_{0.7}$As multiple quantum well

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Temperature and many-body effects on the intersubband transition in a GaAs/Al$_{0.3}$Ga$_{0.7}$As multiple quantum well are studied using the infrared-absorption technique and the envelope-function-approximation method. In order to explain the measured peak position energy of the intersubband transition and its blueshift observed by decreasing temperature and/or increasing the two-dimensional electron-gas density, a theoretical model is developed which is based on a nonparabolic-anisotropic envelope-function-approximation (NAEFA) method. This model takes into account the many-body corrections, in particular, temperature-dependent electron-electron intrasubband and intersubband exchange and direct Coulomb interaction energies as well as the depolarization and excitonlike shifts within the framework of Zaluzny's implementation of Ando's formalism [Phys. Rev. B **43**, 4511 (1991)]. Temperature-dependent effective masses, nonparabolicity, conduction-band offsets, the Fermi level, and line-shape broadening are also incorporated in the present NAEFA calculations. Our theory provides a qualitative explanation for the magnitude of the measured temperature blueshift. Additional support for many-body effects is obtained by utilizing the persistent photoeffect measurements.

I. INTRODUCTION

Recently, it was shown[1] that the blueshift in the intersubband transition in GaAs/Al$_x$Ga$_{1-x}$As multiple quantum wells (MQW's) observed as the two-dimensional electron-gas density (σ) is increased can be explained in terms of electron-electron exchange and direct Coulomb interactions, depolarization (plasmon shift), and excitonlike shifts. It was also shown[2] that the nonparabolicity alone cannot explain the blueshift in the intersubband transition as the temperature is decreased. The temperature effect on the intersubband transition in GaAs/Al$_x$Ga$_{1-x}$As MQW's for a fixed σ is also studied.[2-4] In this article, we will show that the intersubband-transition blueshift observed as the temperature decreases is increased as a function of σ. This behavior can be explained by incorporating the temperature-dependent electron-electron exchange and direct Coulomb interactions, depolarization, and excitonlike shift in a nonparabolic-anisotropic envelope-function-approximation model. In addition, the peak position energy of the intersubband absorption as well as the total integrated area are decreased by illuminating the samples with white light at 5 K. The latter observation provides additional support that the intersubband transition blueshift—which is observed as σ is increased[1]—is due to many-body effects.

This paper is organized as follows: in Sec. II, experi-

mental techniques and results are presented; Sec. III outlines the theoretical approach used in this study; discussion of the results is contained in Sec. IV; and conclusions are given in Sec. V.

II. EXPERIMENTAL TECHNIQUES AND RESULTS

The samples were grown on semi-insulating GaAs substrates by the molecular-beam epitaxy (MBE) technique and consisted of a 1000 Å undoped GaAs buffer layer followed by 100 periods of 75 Å silicon-doped GaAs quantum wells and 100 Å undoped Al$_{0.3}$Ga$_{0.7}$As barriers This was followed by a 75 Å undoped GaAs cap layer. The samples were doped with different silicon concentrations. Infrared-absorption spectra were recorded at the Brewster angle of GaAs ($\sim73°$ from the normal) using a Bomem DA3 Fourier-transform interferometer. Sample temperatures were controlled between 5 and 300 K to within ±0.5 K and were cooled to 5 K using a continuous-flow cryostat. A secondary white light source was used to illuminate the sample for persistent photoeffect measurements.

Figure 1 shows the infrared-absorption spectra for a sample with $\sigma=2.3\times10^{12}$ cm^{-2}. Spectrum (a) was taken after cooling the sample in the dark to 5 K, spectrum (b) was taken at 5 K after illuminating the sample with a secondary white light for 2 min, and spectrum (c) was obtained at 298 K. We will discuss the temperature effect on the intersubband absorption, i.e., spectra (a) and (c),
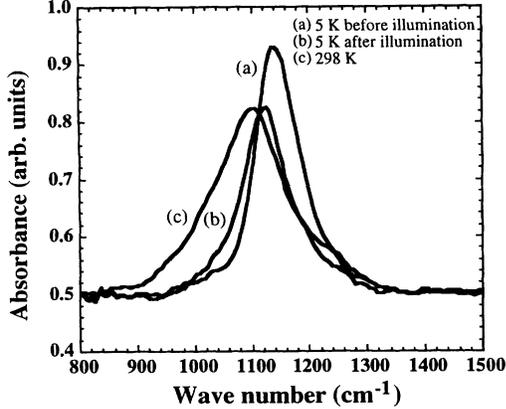
FIG. 1. Optical absorption of the intersubband transition in a sample doped in the well with $\sigma \approx 2.3 \times 10^{12}$ cm^{-2}. Spectrum (a) was taken at 5 K after cooling the sample in the dark. Spectrum (b) is the same as spectrum (a) but after illuminating the sample with white light for 2 min. Spectrum (c) is taken at 298 K.
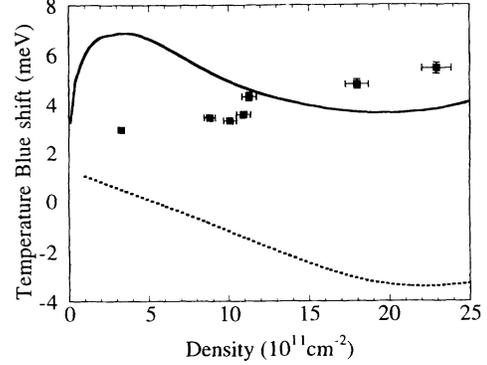

FIG. 2. Temperature blueshift of the PPE as a function of 2DEG concentration. The solid squares were obtained by taking the difference between the PPE measured at 5 and 298 K. The solid line represents the temperature blueshift calculated from Eqs. (1) and (2) with all interaction described by Eqs. (4) and (5) as well as direct Coulomb interaction. The dashed line represents the single-particle calculation.

first and discuss the illumination results (persistent photoeffect) later. It is observed that the total integrated absorption of the intersubband transition of the MQW samples doped in the well remains constant between 5 and 298 K. The peak position energy (PPE), on the other hand, is shifted toward higher energy as temperature is decreased [we will refer to difference between the PPE's of spectra (a) and (c) as a temperature blueshift]. The temperature blueshift of this particular sample is ~ 5.5 meV and is plotted along with temperature blueshifts obtained from samples doped with different concentrations in Fig. 2 as solid squares. It is clear from this figure that the temperature blueshift is decreased as σ decreases.

III. THEORY

In order to calculate the absorption spectrum, we first obtained the electronic structure of the quantum wells as a function of the in-plane wave vector $k_\parallel$. We employed the quartic single-particle Hamiltonian of Ekenberg[5] which is known to reflect accurately the nonparabolic bands structure of bulk GaAs a few tenths of an eV above the bottom of the conduction band. In these calculations, the effective masses in the well and the barrier were obtained from their dependence on the band gap,[6] which was calculated using Varshni parameterization,[7] and the conduction- to valence-band offset ratio was taken[8] at 57/43. For the 75 Å, $x = 0.30$, wells used in this study, the calculation shows only two bound states, $C1$ and $C2$, in the well, between which optical transitions are allowed for light polarized in the growth direction.[2,3] As the result of the nonparabolicity of the original bulk bands, the curvatures of the $C1$ and $C2$ subbands are sufficiently different so that the calculated absorption spectra acquire

a homogeneous width which increases with the doping of the wells.[2,3]

As explained in a series of our earlier papers,[1–4,9–11] we successively improved the accuracy of our calculation by taking several collective effects into account. First, following the suggestion of Bandara et al.,[12] we incorporated the first-order Hartree-Fock (HF) exchange and the direct Coulomb contributions to the calculated single-particle energies. We derived the form of the HF energies for $k > k_F$ for both the ground and excited states.[9] Later, we did the same for the case of multiple band occupancy.[11] For completeness, these expressions are provided below.

The exchange interaction energy is calculated from an expression for the mth subband, which is based on the Hartree-Fock approximation and is given by[13]

$$E_{\text{exch}}^{(m)} = - \sum_n \int d\mathbf{r} \int d\mathbf{r}' \frac{e^2}{4\pi\varepsilon} \frac{1}{|\mathbf{r}-\mathbf{r}'|} \Psi_n^*(\mathbf{r}')$$

$$\times \Psi_m(\mathbf{r}')\Psi_n(\mathbf{r})\Psi_m^*(\mathbf{r}) , \qquad (1)$$

where the summation is over all occupied subbands. In order to render the model analytical, following Bandara et al.,[12] we used wave functions for a simple parabolic band and an infinite square well. With these simplifications, for the ground-state energy, Eq. (1) reduces (in first order) to

$$E_{\text{exch}}^{(0)}(k) = - \frac{e^2 k_F}{4\pi\varepsilon} \left[\frac{2}{\pi} \mathbf{E}\left[\frac{k}{k_F} \right] - \left[\frac{\pi}{6} - \frac{5}{8\pi} \right] \frac{k_F}{k_L} \right]$$

$$\text{for } k < k_F \qquad (2\text{a})$$

and

$$E_{\text{exch}}^{(0)}(k) = - \frac{e^2 k_F}{4\pi\varepsilon} \left\{ \frac{2}{\pi k_F} \left[k\mathbf{E}\left[\frac{k_F}{k} \right] + \frac{(k_F^2 - k^2)}{k} \mathbf{K}\left[\frac{k_F}{k} \right] \right] - \left[\frac{\pi}{6} - \frac{5}{8\pi} \right] \frac{k_F}{k_L} \right\} , \quad \text{for } k > k_F . \qquad (2\text{b})$$

Equation (2a) is identical to the expression derived by Bandara *et al.*[12] For our densities and well widths, higher-order terms in Eqs. (2a) and (2b) would change the calculated exchange energies by about 10% (see Ref. 14). For the first excited state, Eq. (1) reduces to

$$E_{exch}^{(1)}(k) = -\frac{e^2 k_F}{4\pi\varepsilon}\left[\frac{5}{9\pi}\right]\frac{k_F}{k_L} . \tag{2c}$$

In these equations k is the magnitude of the $k_{\parallel}$ wave vector, k_F is the Fermi wave vector ($k_F = \sqrt{2\pi\sigma}$), $k_L = \pi/L$, $E(x)$ is an elliptic integral of the second kind, and $K(x)$ is an elliptic integral of the first kind. The fact that the ground-state exchange energy is k dependent, while the excited-state exchange energy is not, is due to the orthogonality of the ground- and excited-state wave functions. The Hartree-Fock exchange energy suffers the well-known defect of an infinite slope at the Fermi wave vector. This gives rise to an unphysical "dip" in the absorption spectrum at $k = k_F$ and to a zero density of states at the Fermi energy. A further use of screened exchange would improve the calculation, especially at the higher concentrations used in this work. Note that at low densities $E_{exch}(k=0) \approx -\sqrt{\sigma}$, whereas at very high densities $E_{exch}(k=0) \approx +\sigma$, which is the result of the long-wavelength approximation used in deriving Eqs. (2a) and (2b).

Initially, depolarization and exciton shifts were dealt with using Ando's expression,[15] which strictly is only valid for single-frequency transitions (parallel bands). More recently,[10] we adopted the newly-worked-out implementation by Zaluzny[16] of Ando's formalism for the calculation of the linear absorption coefficient. Therefore we calculate the band structure within the HF formalism whereas the exciton and depolarization shifts are obtained from temperature-independent local-density approximation (LDA) expressions for the exchange-correlation potential. Unlike Ando's expression, which assumes the intersubband density of states to be a δ function, Zaluzny's implementation of Ando's formalism allows for different curvatures of the ground and excited subbands, which gives a finite width for the interband absorption. As a result, we obtained much improved absorption line shapes and a very good agreement between the measured and calculated peak positions as a function of electron sheet density at 5 K. Moreover, the shape of the calculated curve for the peak positions as a function of electron sheet density at 5 K agrees with the corresponding curve (Fig. 4) in a self-consistent Hartree-Fock calculation of Sengers, Tseng, and Kuhn[17] (120 Å wells), which, however, does not take into account the different curvatures of the ground and excited subbands. According to Sengers, Tseng, and Kuhn, the effect of self-consistency in their treatment is less than 1 meV (see their Fig. 1). In another recent paper, Luo *et al.*[18] performed an all-Hartree-Fock calculation of the intersubband spin- and charge-density excitation spectra of n-type GaAs/Al$_x$Ga$_{1-x}$As wide (250 Å) quantum wells. This calculation treats the exchange, exciton, and depolarization effects on the same footing. The shape of the curve for the peak positions as a function of electron

sheet density calculated by Luo *et al.* (their Fig. 7) agrees very well with that obtained by us.[10] Therefore these recent results confirm the validity of our earlier low-temperature calculations.

In order to calculate the temperature blueshift of the peak absorption, we derived an expression for the HF and direct Coulomb energies for the ground and the first excited states in an n-type quantum well by explicitly incorporating a temperature-dependent Fermi-Dirac distribution into the HF expression. Taking into account the Fermi occupancy of ground-state subband in Eq. (1), the resulting HF exchange energy as a function of in-plane wave vector becomes

$$E_{exch}(k,\beta) = -\frac{e^2}{2\varepsilon}\int\frac{d\vec{k}'}{(2\pi)^2}\frac{1}{e^{\beta[E(k',\beta)-\mu]}+1}$$
$$\times\int dz\int dz'\frac{e^{-q|z-z'|}}{q}$$
$$\times|F(z')|^2|F(z)|^2 , \tag{3}$$

where $F(z)$ is the envelope function for the ground-state subband, β is the Boltzmann factor, ε is the dielectric constant, $q = |\vec{k}-\vec{k}'|$, $\vec{k}'$ is a two-dimensional vector in the plane of the quantum well, and μ is the chemical potential. The first two terms in the expansion of Eq. (3) in powers of q yield

$$E_{exch}(k,\beta) = -\frac{e^2}{2\varepsilon}\frac{1}{\pi}\left\{\int_0^\infty dk'k'\frac{4}{k+k'}K\left[\frac{2\sqrt{kk'}}{k+k'}\right]\right.$$
$$\times\frac{1}{e^{\beta[E(k',\beta)-\mu]}+1}$$
$$\left.-\left[\frac{L}{3}-\frac{5L}{4\pi^2}\right]\pi k_F^2\right\} , \tag{4}$$

which holds for energies below and above the chemical potential energy. At $T=0$, this expression reduces to Eqs. (2a) and (2b). Equation (4) is clearly valid for both degenerate and nondegenerate situations.[19] As can be easily shown, the expressions for the direct Coulomb energies do not change and neither does the HF energy for the first excited state as long as it is unoccupied.

The HF energies are evaluated self-consistently with the equation determining the chemical potential μ, which is obtained from

$$\sigma = \frac{2}{(2\pi)^2}\int d\vec{k}\frac{1}{e^{\beta[E(k,\beta)-1]}+1} , \tag{5}$$

where we used the quasiparticle energies given by

$$E(k,\beta) = E(k)_{single\ particle} + E_{exch}(k,\beta) . \tag{6}$$

IV. DISCUSSION

The linear absorption coefficient was calculated as a function of photon frequency using Zaluzny's implementation of Ando's formalism,[16] including experimentally determined linewidths and theoretically calculated depolarization and exciton shift parameters. The results shown below are based on actual peak positions obtained

in the calculation.

In the absence of many-body effects, the calculated blueshift decreases with increasing carrier concentration (dashed curve, Fig. 2). This can be explained as follows. With increasing density, the Fermi energy rises. As the temperature is increased from 5 to 300 K, electrons begin to occupy higher-energy states along the ground-state subband, which shifts the "center of gravity" of the subband to higher energies. Since the excited-state subband has a lower curvature then the ground-state subband, the effective intersubband separation decreases as the result of raising the temperature; hence the temperature redshift. However, this effect becomes smaller at higher concentrations since the tail of the Fermi-Dirac distribution becomes an increasingly smaller fraction of the Fermi energy, which makes the center of gravity of the band move proportionately less; hence the calculated single-particle blueshift decreases with increasing carrier density.

These conclusions are similar to those reached in a recent all-LDA calculation by Zaluzny.[20] In his calculation, Zaluzny employed a simplified band-structure model in order to incorporate nonparabolicity and neglected the temperature dependence of all band-structure parameters. By neglecting the temperature dependence of exchange (as well as any temperature dependence of the exciton and depolarization shifts), the blueshift observed by Zaluzny is due to band-population effects alone, as explained above. In particular, Fig. 2 of Zaluzny's paper shows that the temperature blueshift decreases with increasing carrier concentration, which confirms the dashed-curve result in our Fig. 2. We note, however, that the wave-vector dependence of the exchange energies, Eqs. (2)–(4), is another important source of band nonparabolicity which affects the calculated temperature blueshifts. This effect is described next.

The calculated temperature blueshifts (including the temperature-dependent exchange interaction, depolarization, and exciton shifts, and Zaluzny's implementation of Ando's formalism) are shown together with the measured results as the continuous curve in Fig. 2. One notes that the experimental blueshifts are relatively insensitive to the carrier sheet concentration, varying by only 2 meV as the sheet density increases by a factor of 2.5. The calculated spectrum is of about the correct order of magnitude but does not reproduce the experimental trend very well. In this context, it would be desirable to repeat the experiment at several additional low concentrations. In particular, the sample with the lowest concentration is modulation doped, whereas all other samples are doped in the well. In our earlier study,[2,3] that sample exhibited an athermal loss of carriers (as proved by a decrease of the integrated area under the absorption peak), which makes this sample less suitable for comparison.

Before suggesting possible improvements to the model, it is important to understand the behavior of the calculated curve. At very low densities, $\sigma < 0.5 \times 10^{12}$ cm^{-2}, the blueshift increases mainly because of the temperature dependence of the HF exchange energy. For these densities, electrons in the well obey Fermi-Dirac statistics at low temperatures but, at room temperature, they follow Boltzmann statistics. For example, at $\sigma = 0.1 \times 10^{12}$

cm^{-2}, the chemical potential μ at room temperature is some 50 meV below the bottom of the ground-state subband. As a result, electrons in the well are in the long tail of the Boltzmann distribution for the ground-state band, which leads to a weakening of the exchange in comparison to its value at 5 K; hence the large blueshift.

At the intermediate concentrations used in this study, the chemical potential at 298 K is above the bottom of the ground-state subband and the tail of the Fermi-Dirac distribution is progressively a smaller fraction of the chemical potential energy. This leads to smaller differences in the value of the exchange at 5 and 298 K; hence, according to the model, the overall blueshift decreases. At still higher concentrations, $\sigma > 2.0 \times 10^{12}$ cm^{-2}, the blueshift begins to increase again, but in this region we are approaching the limit of the validity of the two-term expansion of the Hartree-Fock energy [i.e., using the first two powers of q in Eqs. (1) and (3)].

The relatively fair agreement between the experiment and theory calls for more experimental measurements as well as for further improvements to the theory. Given the relatively high accuracy of the present model, the one improvement that readily comes to mind is the incorporation of a realistic, temperature-dependent correlation contribution, especially at lower densities. In particular, the trends in the data could be explained by a temperature-dependent correlation-energy contribution to the excited-state subband. For example, in their work on inversion layers, DasSarma et al.[21] have shown a complicated temperature and density dependence of the correlation energy for the excited state. For low densities, they find that the correlation energy for the excited-state subband decreases as the temperature is raised, which would increase the intersubband separation with increasing temperature and lead to smaller blueshifts, in better agreement with experiments. Such trends cannot be obtained with the local-density approximation because of its mean-field character, which prevents it from distinguishing between occupied and unoccupied states. In particular, our LDA expressions for the exciton and depolarization shifts are temperature independent. This may not be a bad approximation, especially for the depolarization shift, as the effect of temperature is to distribute carriers in the ground-state subband, which does not affect their spatial distribution. Finally, the present treatment implicitly assumes that the carrier densities used in this study are above the metal-insulator transition. However, at lower densities, there could be vestiges of the impurity band of silicon donors before the band merges with the conduction-band continuum. This situation would require a simultaneous treatment of both the impurity levels and of the band continuum.

Additional support for the many-body effects on the intersubband optical absorption can be provided by varying σ. One method of varying σ is by illuminating the sample with a secondary white light at $T \leq 100$ K; using this method, the electrons can be excited (removed) out of the well and trapped by defects (persistent photoeffect). The results of this test are presented in Fig. 1 for a sample doped in the well with $\sigma \approx 2.3 \times 10^{12}$ cm^{-2}. Spectrum (b) in this figure is the intersubband optical absorption ob-
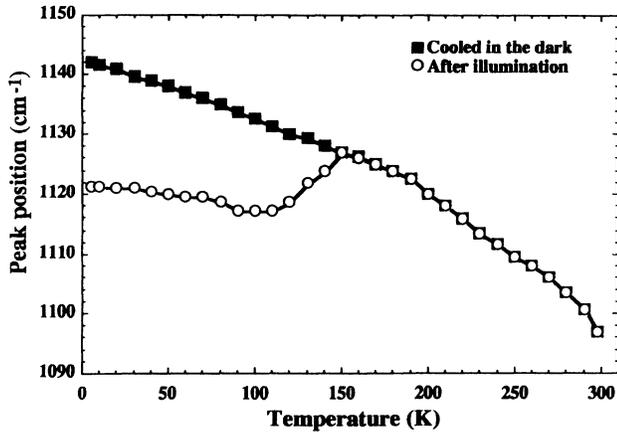
FIG. 3. Effect of the secondary white light illumination on the peak position energy of the intersubband transition as a function of temperature. The solid squares represent the peak position energy of spectrum (a) of Fig. 1, which was taken after cooling the sample in the dark to 5 K. The open circles represent the peak position energy of spectrum (b) of Fig. 1, which was taken after cooling the sample in the dark to 5 K and white light illumination.

tained after cooling the sample in the dark to 5 K and then illuminating the sample with white light for 2 min. It is noted in this figure [spectrum (b)] that the peak position is shifted to a lower energy and that the total integrated absorption area is reduced by about 20% after illumination. This result demonstrates that σ is decreased after illuminating the sample and, consequently, that the many-body effects are reduced, as can be clearly seen by the redshift of the PPE. We have verified that the change in carrier density in the well leads to very small (less than 1 meV) Coulombic shifts of the calculated energies and that it is the density dependence of the exchange that is responsible for the shift of the measured spectrum following white light illumination. It should be noted that the total integrated absorption areas of spectra (a) and (c) are the same within experimental error, suggesting that the electrons are confined in the quantum well in the temperature range of 5–298 K.

The PPE of the intersubband transition observed in the sample used in Fig. 1 is studied as a function of temperature. The results are shown in Fig. 3. The solid squares present the PPE of the intersubband transition spectra taken between 5 and 298 K after cooling the sample in the dark to 5 K [this is the PPE of spectrum (a) of Fig. 1]. The open circles represent the PPE after cooling the sample in the dark, followed by white light illumination [this

is the PPE of spectrum (b) of Fig. 1]. The PPE starts to recover at 110 K, and a complete recovery occurs at ~ 150 K. The temperature at which the recovery occurs is identical to that of the DX center.[22] The latter result is in good agreement with the results reported by Dischler et al.[23] for $GaAs/Al_xGa_{1-x}As$ multiple-quantum-well samples. It should be noted that the amount of the redshift observed after white light illumination is reduced as the dopant concentration is decreased. The exact nature of the electron trapping process is not yet clear. However, we offer the following tentative explanation for this persistent photoeffect. Since the trapping effect is observed in the heavily doped multiple quantum well (where the diffusion of silicon from the well to the barrier is known to occur from the capacitance-voltage and secondary-ion mass-spectroscopy measurements), a DX center is formed in the barrier materials. The electrons, therefore, are excited from the well by the white light illumination and then trapped by the DX center.

V. CONCLUSIONS

In conclusion, it is shown that the temperature blueshift obtained by taking the difference between the peak position energies of the intersubband transition in $GaAs/Al_xGa_{1-x}As$ MQW's at 5 and 298 K is increased approximately linearly as σ is increased. The existence and the magnitude of this blueshift are explained qualitatively by incorporating the exchange and direct Coulomb interactions, depolarization, and excitonlike shifts in the calculation of the peak position energy of the intersubband transition. The persistent photoeffect observed after a secondary white light illumination demonstrates that the peak position energy of the intersubband transition can be shifted and the total integrated absorption can also be changed by varying σ. The latter conclusion provides a strong argument that many-body effects play a significant role in the intersubband transitions in quantum wells.

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