Polaron Zeeman effect in GaAs/Al_xGa_{1-x}As multiple-quantum-well structures

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A three-level ($1s$, $2p^+$, and $2p^-$ donor states) Wigner-Brillouin perturbation theory is used to study the resonant polaron Zeeman effect for an impurity placed at the center of a GaAs/Al_xGa_{1-x}As quantum-well structure in the presence of magnetic field. The resonant splitting caused by the polaron coupling between $1s$ and $2p^+$ states and $2p^-$ and $2p^+$ states as a function of well width is calculated. The results agree with experimental data if the GaAs LO-phonon energy is lowered to 250 cm^{-1} , which is believed to be caused by the zone folding of LO phonons in multiple-quantum-well structures.

The interaction between electrons and longitudinal-optical (LO) phonons in polar semiconductors is of importance both fundamentally and technologically. It is well understood for three-dimensional (3D) systems.¹ Recent advances in crystal growth technology, especially molecular-beam epitaxy (MBE), has made possible the fabrication of very thin semiconductor layers. Thus the electron-LO-phonon interaction in two-dimensional or quasi-two-dimensional systems has also been studied extensively.²⁻¹³ Most works, thus far, have been concentrated on the resonant polaron effect of free electrons. In this paper, we will study theoretically the polaron effect of a shallow hydrogenic donor at the center of a GaAs quantum well in a GaAs/Al_xGa_{1-x}As quantum-well structure, with magnetic field normal to the interfaces. The field plays the role of shifting donor state energy levels. When the separation between two levels is equal to the LO-phonon energy of GaAs, resonance occurs. For example, assuming the LO-phonon has an energy of 290 cm^{-1} , a 138-Å quantum well with an Al mole fraction $x=0.3$, and an impurity donor placed at the center of the well, there is resonance between $1s$ and $2p^+$ ($m=1$) states at 15.8 T . For resonance between $2p^-$ ($m=-1$) and $2p^+$ states, it is at 20.8 T . Therefore, the energy of the $2p^+$ state is strongly affected by resonant polaron effect. These magnetic fields are achievable in laboratories. Photoconductivity experiments can be performed to find the $1s$ - $2p^+$ transition energy and compared with our results. Because of the single-particle nature of the system, possible complication from many-body effects (e.g., screening,¹¹⁻¹³ final-state occupation,⁴ etc.) does not come in. Furthermore, the effect of finite thickness of the GaAs quantum well is incorporated in the Hamiltonian so that the form factor correction¹² is not required. Here we briefly summarize our results. The splittings due to LO-phonon coupling between $1s$ and $2p^+$ states and $2p^-$ and $2p^+$ states are calculated. For both $1s$ - $2p^+$ and $2p^+-2p^-$ coupling the splitting is around 15 and 10 cm^{-1} at their respective resonant field for typical well width of 140 Å . As the well width increases, the splittings decrease, which is consistent with the argument that polaron coupling is stronger in a quasi-two-dimensional system than in a three-dimensional system. Although at first

sight these results appear to be inconsistent with experimental findings, when the LO-phonon spectrum in a multiple-quantum-well structure is considered, the results agree quite well with existing experimental data.

Our Hamiltonian has the following form:

$$H = H_e + H_{LO} + H_I. \quad (1)$$

H_e is the electronic part. The potential energy includes that from a quantum well consisting of layers of GaAs and Al_xGa_{1-x}As, and from a hydrogenic donor at the center of the well. A magnetic field is also applied. Within the frame of effective-mass approximation,

$$H_e = \frac{1}{2m^*} \left[\mathbf{P} - \frac{e}{c} \mathbf{A} \right]^2 + V_B(z) - \frac{e^2}{\epsilon_0 r}, \quad (2)$$

where the effective mass has the following empirical expression:

$$\frac{m^*}{m_e} = 0.067 + 0.083x. \quad (3)$$

In our calculation, $x=0.3$. $V_B(z)$ is the square-well potential of the quantum well,

$$\begin{aligned} V_B(z) &= 0, \quad |z| < L/2 \\ &= V_0, \quad |z| > L/2, \end{aligned} \quad (4)$$

z is the crystal growth direction, and L is the well width. V_0 is the well depth which is taken to be 60% of the band-gap difference: $\Delta E_g = 1.155x + 0.37x^2\text{ eV}$. ϵ_0 is the static dielectric constant. We assumed it to be 12.5 for both GaAs and Al_xGa_{1-x}As.

H_{LO} is the LO-phonon Hamiltonian,

$$H_{LO} = \sum_q \hbar \omega_{LO} (a_q^\dagger a_q + \frac{1}{2}), \quad (5)$$

where ω_{LO} is the LO-phonon frequency. Experimental results¹⁴ show that at zone center (Γ point) $\hbar \omega_{LO} = 290\text{ cm}^{-1}$. Here we have made an approximation that it is dispersionless. H_I is the electron-LO-phonon interaction part. This is what causes the so-called polaron Zeeman effect. The effective Bohr radius of donor states is

about 100 Å.¹⁵ It is much greater than atomic spacing. Electron-phonon interaction is a long-range Coulomb interaction. Therefore, we chose the Fröhlich model¹⁶ for H_I :

$$H_I = \sum_{\mathbf{q}} (V_{\mathbf{q}} a_{\mathbf{q}} e^{i\mathbf{q} \cdot \mathbf{r}} + V_{\mathbf{q}}^* a_{\mathbf{q}}^\dagger e^{-i\mathbf{q} \cdot \mathbf{r}}), \quad (6)$$

where

$$V_{\mathbf{q}} = \frac{ie}{|\mathbf{q}|} \left[\frac{2\pi}{\Omega} \right]^{1/2} (\hbar\omega_{\text{LO}})^{1/2} \left[\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0} \right]^{1/2}. \quad (7)$$

Here Ω is the total volume of the system and ϵ_∞ is the dielectric constant of GaAs in the high-frequency limit. In actual calculation, we took the coupling constant

$$\alpha \equiv \frac{e^2}{2\hbar\omega_{\text{LO}}} \left[\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0} \right] \left[\frac{2m^* \omega_{\text{LO}}}{\hbar} \right]^{1/2} \quad (8)$$

to be 0.07.

We started by solving H_e for electron wave functions. The method was proposed by Greene and Bajaj.¹⁵ It is a variational method with a Gaussian basis. The results agree quite well with experimental data.¹⁷ Once the wave functions of $1s$, $2p^-$, and $2p^+$ donor states were obtained, they were used in Wigner-Brillouin perturbation theory¹⁶ (WBPT) to account for the effect of H_I on $1s$ - $2p^+$ transition energy. The wave functions of donor states were written as products of an envelope function and Gaussian-type functions:

$$\Psi(\mathbf{r}) = f(z)G(\mathbf{r}), \quad (9)$$

where $f(z)$ is the envelope function,

$$f(z) = \begin{cases} \cos(kz), & |z| < L/2 \\ Ae^{-p|z|}, & |z| > L/2. \end{cases} \quad (10)$$

The parameters k , p , and A were determined by matching conditions, i.e., $f(z)$ and effective flux $(\partial f / \partial z) / m^*$ are continuous¹⁸ at GaAs/Al_xGa_{1-x}As interfaces. They result in

$$\frac{\hbar^2 k^2}{2m_1^*} = V_0 \left[1 + \frac{m_2^*}{m_1^*} \tan^2(kL/2) \right]^{-1}, \quad (11)$$

where m_1^* and m_2^* are the effective masses of electrons in GaAs and Al_xGa_{1-x}As, respectively. $G(\mathbf{r})$ describes the internal state of the donor. If we consider the case where the applied magnetic field is in the z direction and choose the gauge

$$\mathbf{A} = \frac{1}{2} \mathbf{B} \times \mathbf{r}, \quad (12)$$

then the azimuthal symmetry is preserved and a great deal of numerical calculation is saved. In our calculation, $G(\mathbf{r})$ has one adjustable parameter and 13 basis functions for the $1s$ state and 10 for $2p^+$ and $2p^-$ states. We refer interested readers to Green and Bajaj¹⁵ for details.

The donor electron interacts with LO phonons. As we increase the magnetic field, the separation between $1s$ and $2p^+$ levels also increases. When their difference is equal to the LO-phonon energy, resonance occurs. The $1s$ - $2p^+$ transition energy is strongly influenced by LO phonons near resonance. The same thing happens for $2p^-$ - $2p^+$ transition energy at a higher magnetic field. To account for electron-LO-phonon interaction, we use the WBPT, which is better suited for studying resonance than, say, the Rayleigh-Schrödinger perturbation theory. Since only one LO phonon can be created or destroyed by H_I , WBPT is equivalent to a modified Koonce and White's method.¹⁹ In the system we have only one electron and one LO phonon. Suppose there are M electronic states and N degenerate LO-phonon states (different momentum $\mathbf{q}$), then the Hamiltonian matrix is

$$\begin{pmatrix} E_0 & g_{1,01} & g_{2,01} & \cdots & g_{N,01} & g_{1,02} & \cdots & g_{N,02} & \cdots & g_{N,0M} \\ g_{1,10} & \epsilon_1 & & & & & & & & \\ g_{2,10} & & \epsilon_1 & & & & & & & \\ \vdots & & & \ddots & & & & & & \\ g_{N,10} & & & & \epsilon_1 & & & & & \\ g_{1,20} & & & & & \epsilon_2 & & & & \\ \vdots & & & & & & \ddots & & & \\ g_{N,20} & & & & 0 & & & \epsilon_2 & & \\ \vdots & & & & & & & & \ddots & \\ g_{N,M0} & & & & & & & & & \epsilon_M \end{pmatrix}, \quad (13)$$

where ϵ_i 's are electronic state energies plus phonon energy and

$$g_{j,i0} = V_{\mathbf{q}_j} \int \Psi_i(\mathbf{r}) e^{i\mathbf{q}_j \cdot \mathbf{r}} \psi_0(\mathbf{r}) d\mathbf{r}. \quad (14)$$

The eigenvalues of the $M \times N$ by $M \times N$ matrix can be

solved for exactly:

$$E - E_0 = \sum_{i,j} \frac{|g_{j,i0}|^2}{E - \epsilon_i}. \quad (15)$$

Since we used dispersionless phonons, the summation

FIG. 1. (a) $1s-2p^+$ transition energy vs applied magnetic field. The three branches are three solutions of Eq. (17) minus E_{1s} (see text). (b) Same as those in (a) except that LO-phonon energy was shifted to 250 cm^{-1} . The circles are experimental data in Ref. 21.

over LO-phonon states can be carried out independent of electronic state summation:

$$E - E_0 = \sum_i \frac{\sum_j |g_{ji,0}|^2}{E - E_i - \hbar\omega_{LO}}. \quad (16)$$

In our case $\psi_0(\mathbf{r})$ is the $2p^+$ wave function. In principle, the intermediate electronic states should include all the donor states and the entire continuum. We took into account only $1s$ and $2p^-$ states because they are to be studied and, as far as we are concerned, their effect is the most important one.²⁰ As a result, Eq. (16) becomes

$$E - E_{2p^+} = \frac{\sum_j |g_{j,1s2p^+}|^2}{E - E_{1s} - \hbar\omega_{LO}} + \frac{\sum_j |g_{j,2p^-2p^+}|^2}{E - E_{2p^-} - \hbar\omega_{LO}}. \quad (17)$$

Using the donor-state wave functions obtained by Greene and Bajaj's method,¹⁵ the matrix element $g_{j,0i}$ can be evaluated and the summation over momenta can be carried out easily. For an impurity placed at the center of a $138\text{-}\text{\AA}$ GaAs quantum well, the three eigenenergies obtained from Eq. (17) minus E_{1s} are plotted in Fig. 1(a) as functions of magnetic field. This should be compared with the measured $1s-2p^+$ transition energy of, say, photoconductivity experiments. The lower splitting at 290 cm^{-1} is due to a $1s-2p^+$ resonant polaron effect where the unperturbed E_{2p^+} is equal to $E_{1s} + \hbar\omega_{LO}$. The applied field there is 15.8 T . The upper splitting occurring at $B = 20.8 \text{ T}$ is due to $2p^- - 2p^+$ resonance.

A recent experiment²¹ found that the lower branch was at 240 cm^{-1} near $B \approx 15 \text{ T}$. This seems to be quite far away from our result. However, the LO phonon is not dispersionless as it was assumed in our calculation. Its frequency is 290 cm^{-1} at Γ point and 250 cm^{-1} at the X point in bulk.¹⁴ In a multiple-quantum-well system, the dispersion curve is folded back many times to give many branches.²² The section near the X point is folded back

FIG. 2. Resonance splitting of the $1s-2p^+$ (solid line) and $2p^- - 2p^+$ (dashed line) transitions at the respective resonance applied field as a function of well width.

to $q \approx 0$ as a part of the lowest branch. Therefore, to compare with experimental data, we took ω_{LO} to be 250 cm^{-1} and redid our calculation. The result is plotted in Fig. 1(b) with circles showing experimental data. We see that there is reasonable agreement.

Finally, in Fig. 2, we present the relation between resonance splitting and well width: the wider the well, the smaller the splitting. The ratio of 2D splitting to that of 3D is about 1.3, which is smaller than the enhanced polaron effect expected for free electrons.¹² This result comes out naturally with our method; no form factor correction is required. On the other hand, the stronger the magnetic field, the denser the orbitals, and the larger the splitting. Since a wide well needs a stronger field to reach resonance, it may not be fair to compare the polaron Zeeman effect between the 2D and 3D systems since the resonant field is higher for a 3D impurity. If the resonance of the 2D and 3D polaron Zeeman resonances had occurred at the same magnetic field, the ratio should have been greater than 1.3.

The $1s-2p^+$ transition at the $2p^- - 2p^+$ resonant field is a so-called three-level system. It may avoid the complication caused by the presence of a reststrahlen band in the two-level system. It would be interesting to find out experimentally the transition energies between $1s$ and $2p^+$ states near $2p^-$ and $2p^+$ resonant fields. The magnetic field needed for this resonance is 20.8 T for a $140\text{-}\text{\AA}$ well. It could be lower due to the presence of zone-folding LO phonons. Apparently, the dispersion of zone-folding LO phonons is essential. Research on this aspect is underway.

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