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Two-level and three-level resonant measurements of impurity-bound magnetopolarons in multiple-quantum-well structures

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Far-infrared photoconductivity has been used to follow the $1s$-$2p_+$ transition of shallow donors confined in GaAs/Al$_x$Ga$_{1-x}$As multiple-quantum-well structures through resonant regions with GaAs optical phonons by magnetic field tuning up to 23.5 T. Both two-level and three-level measurements show anti-level-crossing behavior with the lower branches far below (~ 50 cm^{-1}) the resonant energies for the bulk long-wavelength LO phonons, resulting in extremely large, asymmetric interaction gaps. The results strongly suggest that electrons interact with more than one set of phonons, or a phonon band. The electron–interface-phonon interaction provides a plausible explanation for the experimental observations.

The interaction of an electron with longitudinal-optical (LO) phonons (polarons) has fundamental importance for electronic properties of polar semiconductors. Extensive investigations on this subject have been carried out for many years. Fröhlich's early formulation[1] and the later development of quantum field calculations[2] have been very successful in providing a quantitative understanding of the polaron problem in three-dimensional (3D) systems. For quasi-two-dimensional (2D) systems, such as GaAs/Al$_x$Ga$_{1-x}$As multiple-quantum-well (MQW) structures, the situation is considerably less clear. Theoretical calculations,[3–5] which made use of Fröhlich's Hamiltonian with quasi-2D electronic states (i.e., 2D electron interacting with 3D phonons), have predicted an enhanced polaron effect in systems with reduced dimensionality. These treatments have shortcomings since some of the assumptions inherent to the model, such as the continuum lattice approximation and dispersionless LO-phonon frequency, may not be completely applicable to quasi-2D systems. Recently, more realistic confined-slab phonon modes and interface phonon modes have been calculated numerically[6,7] and analytically.[8] When these phonon modes were used, the calculations demonstrated that the polaron effect is substantially reduced in comparison with results employing 3D phonons.[9,10]

Several experiments[11,12] carried out on 2D systems with large free-electron densities in the presence of external magnetic fields had led to different results for the magnitude of the interaction; both enhanced and reduced polaron effects were observed. These apparently contradictory results were later explained by recourse to screening[13,14] and occupation effects.[3] Recent cyclotron resonance measurements have demonstrated that the magnitude of the interaction is indeed reduced with increasing electron concentration in single GaAs/Al$_x$Ga$_{1-x}$As heterostructures.[15] To avoid the complications of screening and occupation effects, experiments have also been carried out on an alternate quasi-2D single particle system, i.e., shallow donors confined in MQW structures.[16,17] In these investigations, the intra-impurity transition, $1s$-$2p_+$ (in the 3D notation), was tuned by a magnetic field from lower energy to energies nearly in resonance with bulk zone-center (Γ-point) LO phonons. A large sublinear deviation from the unperturbed transition energy and a rapid decrease in intensity in the lower branch of the resonance have been observed at energies well below the LO(Γ) energy.[16] However, these experiments provided no conclusive data for the upper branch which is very significant in extracting the strength of electron–LO-phonon coupling. The existence of the reststrahlen band in this energy region can also complicate the data analysis.[18,19]

We have carried out far-infrared (FIR) magneto-optical measurements on well-center-doped MQW structures to study the impurity-bound magnetopolaron effects. With improved resolution and signal-to-noise ratio we have achieved much more accurate data for the lower branch and clear observation of the upper branch in the two-level resonance region (transition energy of $1s$-$2p_+$ equal to the LO-phonon energy). In addition, and more importantly, we have observed the three-level resonance process (energy difference between $2p_+$ and $2p_-$ equal to the LO-phonon energy) at magnetic fields around 21 T. This provides the most reliable data to explore this problem since the probing photon energy is much higher than the reststrahlen band. Extremely large "interaction gaps" and severely asymmetric lower and upper branches with respect to the resonant energies involving the bulk LO(Γ) phonons have been observed; this strongly suggests that electrons interact with more than one set of phonons, or a phonon band, in contrast to the dispersionless LO phonons in the 3D case.

The samples used in these experiments were cleaved from a wafer grown by molecular-beam epitaxy; the structure consists of 30-GaAs wells of width 125 Å separated by 125-Å-Al$_{0.3}$Ga$_{0.7}$As barriers. Si donors were doped over the central $\frac{1}{3}$ of the GaAs wells at a nominal concentration of 1×10^{16} cm^{-3}. Two semitransparent chromium

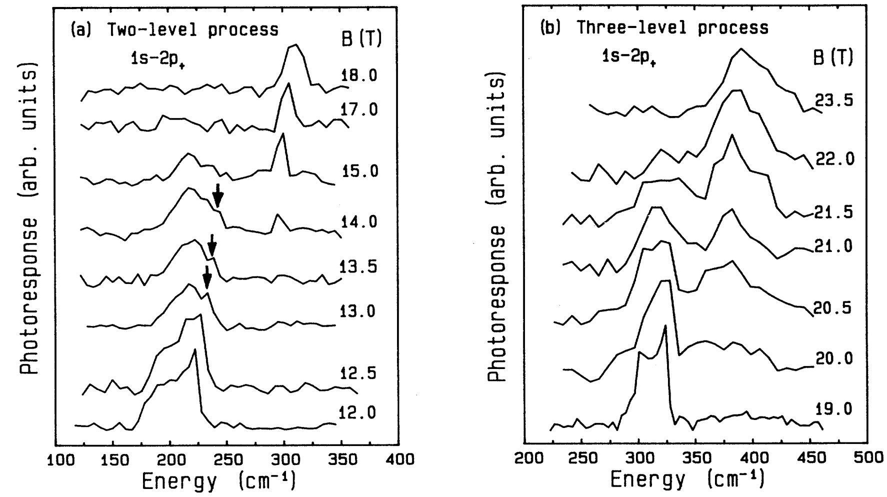

FIG. 1. Capacitively coupled photothermal ionization spectra at several magnetic fields. (a) Two-level resonance process; (b) Three-level resonance process. Arrows in (a) indicate the $1s$-$2p_+$ transitions of well-center-doped impurities. The broad feature at lower frequencies is due to the off-center distribution of impurities.

electrodes separated by a small gap were evaporated on the top surface of the sample. A low-frequency (~100 Hz) ac voltage was applied between the two electrodes coupled capacitively to the resistively conducting quantum well plates, and the ac through the sample was detected. This capacitively coupled photoconductivity technique was relatively insensitive to the reststrahlen band and greatly enhanced the signal-to-noise ratio in comparison to the normal transmission measurement. Far infrared magneto-optical spectra were obtained by using Fourier transform spectrometers in conjunction with a 9 T superconducting magnet (Buffalo) and a 23 T Bitter magnet (Francis Bitter National Magnet Laboratory). All data were taken at liquid-He temperatures in the Faraday geometry (**B** parallel to FIR light propagation direction and normal to the sample surfaces).

Photothermal ionization spectra of the impurity transition $1s$-$2p_+$ for the two-level resonance process are shown in Fig. 1(a) at several magnetic fields. At low magnetic fields ($B < 12$ T) the spectra show a normal line shape with a very sharp peak on the high-frequency side and a tail extending to lower frequencies (~20–30 cm^{-1}). The high-frequency sharp peak is due to well-center impurity transitions while the lower-frequency tail results from the off-center impurity distribution profile.[20] As the field increases near the interaction region, the relative intensity of the sharp peak rapidly decreases. At fields around 14 T, this peak becomes a weak shoulder[21] (indicated by arrows) which is hard to resolve with low resolution and signal-to-noise ratio. At slightly higher field, this line completely disappears (undetectable within the noise); meanwhile a very sharp peak appears at a frequency slightly higher than the LO(Γ) phonon frequency. This line (upper branch of the two-level resonance) increases in relative intensity and slowly moves up to higher frequency as field is increased. The three-level resonance process data are shown in Fig. 1(b). At a magnetic field of 19 T, the spectrum shows nearly the same line shape as those in the low-field region ($B < 12$ T). In the vicinity of 21 T, a typical intensity variation for an antilevel crossing process can be clearly observed.

The transition energy as a function of magnetic field has been plotted in Fig. 2. Data taken during different runs are indicated by different symbols; results are quite reproducible (the scattered data points in the high-field region are partly due to the lower resolution). For fields

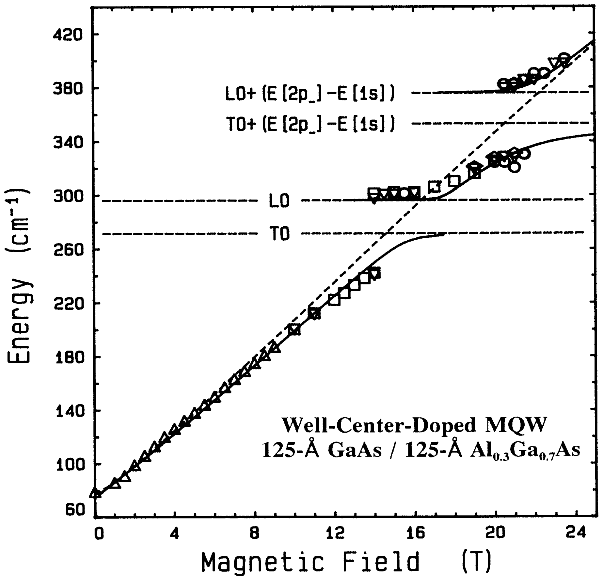

FIG. 2. Transition energy of $1s$-$2p_+$ as a function of magnetic field. Data taken during different runs are indicated by different symbols. The dashed line is the calculated transition energy without electron–LO-phonon interaction. The solid line is the calculation for the impurity-bound magnetopolaron with electron–interface-phonon and electron–confined-LO-phonon interactions involved (see text).

near 14 T, the positions of the shoulder at the high-energy edge of the line profile, indicated by arrows in Fig. 1(a), are used in the plot. The dashed curve is the theoretical calculation for the $1s$-$2p_+$ transition without electron-LO-phonon interaction.[22] A nonparabolicity correction has been made to this calculation. Three branches separated by two gaps can be seen over the spectral region studied. The lower branch is clearly depressed from the unperturbed theoretical curve and disappears completely in the region above 245 cm^{-1} (above 14.5 T) which is well below (~ 50 cm^{-1}) the bulk LO(Γ)-phonon energy, although it has *not* at this point approached the horizontal. The middle branch is actually the upper branch of the two-level resonance ($E[2p_+]-E[1s]\simeq$LO phonon energy) and the lower branch of the three-level resonance ($E[2p_+]-E[2p_-]\simeq$LO phonon energy), and represents the overall effect of these two resonant interactions. Its energy increases very slowly with magnetic field, crossing the calculated unperturbed curve at approximately 17 T and approaching an energy of ~ 330 cm^{-1}. This is again about 50 cm^{-1} below the energy for pinning with a three-level process involving the bulk LO(Γ) phonon. The upper branch for this process starts at the energy $\hbar\omega_{\mathrm{LO}}+E[2p_-]-E[1s]$, as indicated in Fig. 2. Its position increases with magnetic field and approaches the theoretical unperturbed curve at the highest magnetic fields.

As a complementary measurement, another impurity transition, $1s$-$3p_+$, has also been used to study the resonant magnetopolaron effect. This transition is much weaker than the $1s$-$2p_+$ transition, but moves approximately twice as fast with magnetic field; thus it can be tuned through the resonance region at roughly half the magnetic field of the $1s$-$2p_+$ transition. Behavior very similar to that of $1s$-$2p_+$ has been observed in the two-level resonance region ($E[3p_+]-E[1s]\simeq$phonon energy), i.e., an extremely large and asymmetric interaction gap with a sublinear behavior for the lower branch at energies around 245 cm^{-1}, and complete disappearance at slightly higher energies.

Compared with the three-dimensional situation, the experimental results reported here show two major anomalies which appear in all the measurements. First, there are apparently extremely large interaction gaps, with the lower branches disappearing at energies far below the resonance energies involving the LO(Γ) phonons; in the bulk, nothing notable happens in these energy regions. Second, the energy separations of the lower and upper branches from the pinning energies are extremely asymmetric if the pinnings are assumed to take place only at LO(Γ) and LO(Γ)$+(E[2p_-]-E[1s])$. These anomalies cannot be explained by the usual Fröhlich formulation[23] and strongly suggest that electrons in the well interact with more than one phonon mode or with a phonon band with the top edge at the LO(Γ) energy and the bottom edge well below the LO(Γ) energy.

Dielectric artifacts,[18,19] in which the transition line shape could be strongly distorted by the internal multiple reflection of FIR light in the complicated multilayer structures near the reststrahlen region, is certainly not the major factor responsible for the anomalies. This effect has been investigated by both an experimental check[16] and a computer simulation[24] for the samples used in this study. The three-level resonance measurement gives strong support for excluding dielectric artifacts in explaining the data since the probing FIR light has much higher frequency than the reststrahlen band in this case.[25]

The calculations with quasi-2D electrons interacting with 3D LO(Γ) phonons exhibit qualitative discrepancies compared with the experimental results. In this treatment, the only pinning frequencies are those involving bulk LO(Γ) phonons; and the separation between upper and lower branches should be centered about these frequencies. This is obviously in contradiction to the present experimental observations. Quantitatively, this model predicts an enhanced polaron effect due to removing the restriction of momentum conservation in the confinement direction. The experimental results, on the other hand, show that the energy splitting is 1 order-of-magnitude smaller than this prediction[26] (the upper branch is used to estimate the interaction strength since it is free from the complications of the reststrahlen band); that is, the interaction is substantially *reduced* compared to the 3D case.

The existence of interface phonons and confined phonons in quasi-2D systems have been verified both theoretically and experimentally.[6-8,27] Very recently a calculation of the impurity-bound magnetopolaron in a quasi-2D system (single GaAs/$\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ QW with donors doped at the center of a 125-Å-well) has been carried out for both two-level and three-level resonances with electron-interface-phonon and electron-confined-LO-phonon interactions involved.[28] The results are plotted in Fig. 2 (solid line), and theoretical treatments are given in Ref. 28. The remarkable characteristics of the calculation are the existence of a gap between bulk TO(Γ) and LO(Γ) frequency, and the lower branch pinning at the TO(Γ) frequency rather than the LO(Γ) frequency. These features are qualitatively in agreement with experimental observations, and the calculation fits the data reasonably well above LO(Γ). However the rapid intensity reduction and strong sublinear behavior at energies around 245 cm^{-1} are still not completely accounted for by this model. The oscillator strength and the line shape of the impurity transitions in this energy region remains to be studied theoretically by including both electron-phonon interactions and the dielectric effects.

Zone-folding effects could also contribute to the anomalies observed in the experiments. The GaAs-LO-phonon dispersion curve overlaps with the $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ GaAs-like LO-phonon dispersion; therefore phonons can propagate in the confinement direction. The superlattice effect causes the dispersion to be folded into a mini-zone, and electrons can interact with LO modes having frequencies near the bulk zone edge (X point), about 250 cm^{-1}. This is the energy near which transitions quickly lose their intensities and the anomalies start. However, at present this is still only a speculation; the strength of this effect will depend crucially on the magnitude of the splitting at the mini-zone boundary and the coherence length of the propagating optical modes. A model calculation has been carried out[23] by assuming that the electron–LO(X) phonon interaction has the same coupling strength as the electron–LO(Γ) phonon interaction; this is certainly a questionable assumption. The conclusive answer must await detailed microscopic calculations and further experimental studies in this direction.

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