Resonant bipolaron coupling in GaAs quantum wells

S. Huant
High Magnetic Field Laboratory, Max-Planck Institut für Festkörperforschung and Centre National de la Recherche Scientifique, Boîte Postale 166, 38042 Grenoble Cedex 9, France

B. Etienne
Laboratoire de Microstructures et Microelectronique, Centre National de la Recherche Scientifique, Boîte Postale 107, 92225 Bagneux Cedex, France

N. Coron
Institut d'Astrophysique Spatiale, Batiment 121, Universite Paris XI, 91405 Orsay Cedex, France
(Received 27 September 1994)

A resonant polaron effect has been observed for negatively charged donors (D^- centers) in a 100 Å GaAs quantum-well structure. The magnitude of this effect is found to be smaller than predicted theoretically. No dramatic enhancement of the resonant polaron effect on D^- centers over the effect on neutral donors is observed.

In a polar semiconductor, electrons interact with the polarization cloud of longitudinal-optical (LO) phonons which they produce to form a polaron. In a weakly polar material at small magnetic field strengths, nonresonant polaron effects due to the Fröhlich electron-phonon interaction are small and difficult to observe. Polaron effects are best probed in the resonant regime which is reached in a high magnetic field when a given magneto-optical transition approaches the LO phonon energy $\hbar\omega_{LO}$ (36.8 meV in GaAs). This leads to a resonant splitting of this transition (see, e.g., Ref. 1 and references therein).

In the past, resonant polaron effects have been studied in a large number of systems,[1–4] especially shallow neutral donors D^0 in GaAs/$Ga_{1-x}Al_xAs$ multiple-quantum-well (MQW) structures,[1,2] for which there is a good overall agreement between theory and experiment. Simple trial wave functions for D^0 and the use of second-order perturbation theory (PT) to describe the Fröhlich interaction with bulk LO phonons have proved sufficient to describe a large number of experimental data.[1]

Here, we are interested in resonant polaron effects on a *correlated two-electron impurity system* (D^- center), for which there exist no experimental data to the best of our knowledge.[5] A D^- center is a negatively charged shallow donor formed when a D^0 donor traps a second electron.[6] Although difficult to populate in bulk GaAs,[7] D^- states can be made to form in sizable concentrations in QW's by the appropriate modulation doping of a GaAs/$Ga_{1-x}Al_xAs$ MQW structure.[8]

D^- centers in polar crystals can exhibit a large variety of physical properties.[9,10] Depending on the strength of the electron-phonon interaction and on the strength of the Coulomb potential that binds the two electrons in the absence of electron-phonon interaction, D^- centers can be unstable or they can exhibit negative-U behavior among other scenarios.[10] Since GaAs is lying in the weak-coupling–weak-binding limit ($\alpha \ll 1$ and $R^* \ll \hbar\omega_{LO}$, where $\alpha = 0.068$ is the Fröhlich constant in GaAs and $R^* = 5.83$ meV is the effective Rydberg in GaAs), a small decrease of the D^- binding energy is expected at zero magnetic field in the bulk material.[9,10] Indeed, in this so-called polaron regime[10] of D^-, all the interparticle distances in D^- are much larger than the polaron radius (40 Å in GaAs). As a consequence, the two polarons bound to the same impurity ion have little overlap and the D^- binding energy is only slightly affected (the polaron correction to the energy of D^- is comparable to that of a system formed by a D^0 donor plus a free electron).

However, in QW's in a high magnetic field, the situation is expected to change qualitatively[11] because the orbital length of D^- perpendicular to the magnetic field scales down approximately as the magnetic length[12] (57 Å in 20 T) and the orbital length in the field direction is governed by the quantum confinement. As a matter of fact, all characteristic lengths become comparable to (but still larger than) the polaron radius and D^- starts to resemble a bipolaron—strictly speaking a system with two electrons in the same polarization cloud—in the sense that a static polarization starts building up around the impurity ion. In this case, a deepening of D^- can be expected but this is difficult to probe experimentally. More easily accessible to experiment is the resonant polaron effect which, indeed, is predicted to be enhanced over the effect on neutral donors.[11,13] In this paper, we describe such an experiment and we compare our results with available theoretical predictions.

We have used a 100 Å/200 Å GaAs/$Ga_{0.75}Al_{0.25}As$ multiple-quantum-well structure grown by molecular-beam epitaxy. This structure is planar doped with silicon donors in the middle of the wells ($N_w = 1.8\times10^{10}$ cm^{-2}) and in the middle of the barriers ($N_b = 3\times N_w$). This unbalanced double-planar doping geometry favors the conversion of all the D^0 donors in the wells to D^- centers.[8] Far-infrared transmission spectra of this sample were taken using a fast-scan Fourier-transform spectrometer

0163-1829/95/51(8)/5488(3)/$06.00

(Bruker IFS 113v) connected to a 21 T resistive magnet. A high-sensitivity Ge composite bolometer was placed behind the sample.

Typical transmission spectra (normalized to zero-field spectra) are shown in Fig. 1. At low field, two peaks are seen which are the $1s \rightarrow 2p^+$ transitions for D^0 donors in the barriers (peak C) and the $s \rightarrow p^+$ spin-singlet transition of D^- (peak B).[14,15] Upon increasing the magnetic field, both peaks C and B shift to higher energies. It is found that the D^- transition lies in energy below the reststrahlen band for fields less than 16.5 T and goes above $\hbar\omega_{LO}$ for fields in excess of 19.5 T. The field dependence of the D^- peak energy is shown in Fig. 2. A typical anticrossing behavior is observed in the optical-phonon energy range which reveals a resonant polaron effect. The splitting shown in Fig. 2 between the two polaron branches is of the order of $(1.0 \pm 0.1)R^*$. This is only slightly larger than the polaron splitting of D^0 donors in a 100 Å QW, which is $0.9R^*$.[1] This splitting turns out to be less than predicted as discussed below.

The predictions of two recent theoretical treatments of the polaron effect on D^- centers[11,13] are shown for comparison with our results in Fig. 2. It can be seen that these theories differ considerably in the resonant regime. In Ref. 11, a variational approach was used to compute the bare D^- states. The Fröhlich interaction is taken into account within an improved version of Wigner-Brillouin perturbation theory which allows for the inclusion of all the intermediate states. The resonant polaron effect is calculated in the three-level scheme (s, p^-, and p^+ singlet states of D^-). A similar analysis for D^0 donors has shown previously a very good agreement with experiment.[1] Also shown in Fig. 2 are our former data[8] and the predictions of the same theory for the $s \rightarrow p^-$ transition of D^- which is not sensitive to resonant polaron effects. A satisfactory agreement is found

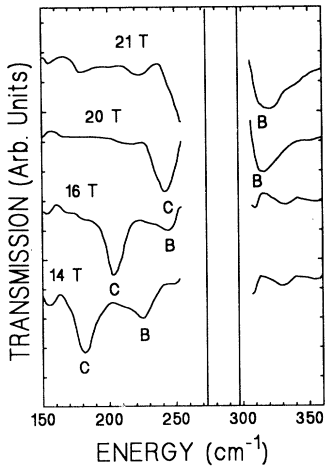

FIG. 1. Transmission spectra of a D^- doped 100 Å QW sample showing the $1s \rightarrow 2p^+$ transition for D^0 donors in the barriers (peak C) and the spin-singlet $s \rightarrow p^+$ transition of D^- (peak B) both below and above the reststrahlen band of GaAs (represented by vertical bars). The magnetic field strength is indicated on each curve.

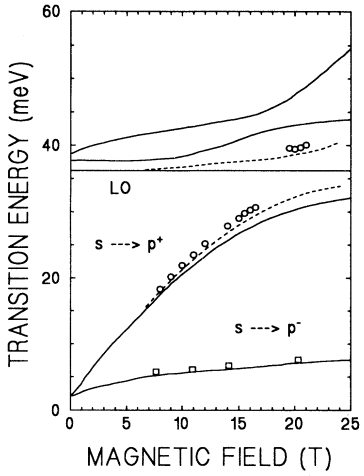

FIG. 2. The field dependence of the $s \rightarrow p^+$ transition energy of D^- centers in a 100 Å QW. The open symbols are the experimental data. The full lines are the predictions of Ref. 11 and the dashed lines are the predictions of Ref. 13.

because the polaron corrections to the s and p^- states compensate each other. Such a good agreement is not found for the transition to the p^+ state. Clearly, the theory of Shi *et al.*[11] greatly overestimates polaron effects in the resonant regime for this transition, i.e., the experimental data are well above ($\approx$ 2.5 meV) the predictions for the lower polaron branch and substantially below ($\approx$ 3.5 meV) for the upper branch.

A closer agreement is found between the present data and the predictions of Ref. 13, although the theory again overestimates polaron effects. In this case, the experiment is above the predictions for both polaron branches ($\approx$ 1 meV for the lower branch and $\approx$ 0.5 meV for the upper branch). In Ref. 13, bare D^- states are obtained by diagonalization of the total two-electron Hamiltonian describing a D^- center in a QW in a magnetic field. This procedure turns out to be more accurate than the approach of Ref. 11 (see below). Five to seven relevant intermediate D^- states are included in a subsequent second-order PT to take into account the Fröhlich interaction. Again, excellent agreement is found for the polaron-insensitive $s \rightarrow p^-$ transition.

Current theories, especially those based on variational calculations of the unperturbed levels, appear to overestimate resonant polaron corrections of the correlated two-electron D^- system in a QW although similar approaches have been quite sucessful for D^0. Perhaps going beyond second-order PT would give some improvement. In Ref. 13, the polaron correction in a 100 Å QW at 20 T is computed to be $\approx$ 19% ($-0.91R^*$) of the total energy of D^- measured from the energy of two electrons in their zero Landau level ($-4.88R^*$). This is a substantial correction (the equivalent number for D^0 is $\approx$ 7%). In Ref. 11, the corresponding numbers are 71%, $-3.10R^*$, and $-4.37R^*$. Clearly, the polaron corrections are here much too large to justify the use of PT. Furthermore, we note that the bare energy is rather poor as compared to the "true" energy of $-5.02R^*$ calculated with diffu-

sion quantum Monte Carlo techniques.[16] Thus we believe that the trial functions are not sufficiently accurate to use them in a subsequent perturbation calculation. It is likely that electron correlation is not correctly described by the theory, or, in other words, the electron repulsion in D^- is not minimized. The two electrons are allowed to stay too close to each other on average, thereby magnifying the polarization cloud around the impurity center and, therefore, leading to an overestimate of polaron effects. To some extent, the wave functions obtained in Ref. 13 appear to be better as suggested by the reasonable bare energy given above and by the better agreement with the present experimental data. It is hoped that the present work will stimulate more studies to reconcile polaron theories of D^- with experiment.

Helpful correspondence with A. B. Dzyubenko is gratefully acknowledged.

[1] J.-M. Shi, F.M. Peeters, G.Q. Hai, and J.T. Devreese, Phys. Rev. B **44**, 5692 (1991).

[2] S. Huant, W. Knap, G. Martinez, and B. Etienne, Europhys. Lett. **7**, 159 (1988).

[3] S. Das Sarma and A. Madhukar, Phys. Rev. B **22**, 2823 (1980); S. Das Sarma, Phys. Rev. Lett. **52**, 859 (1984); F.M. Peeters and J.T. Devreese, Phys. Rev. B **31**, 3689 (1985).

[4] G.Q. Hai, F.M. Peeters, and J.T. Devreese, Phys. Rev. B **48**, 4666 (1993).

[5] A preliminary account of this work has been presented by S. Huant and B. Etienne, in *Proceedings of the Eleventh International Conference on High Magnetic Fields in Semiconductor Physics, Cambridge, 1994*, edited by D. Heiman (World Scientific, Singapore, in press); during the course of the above conference, we learned about a similar work reaching essentially the same conclusions by J. P. Cheng, B. D. McCombe, and W. Schaff, *ibid.*

[6] S. Chandrasekhar, J. Astrophys. **100**, 176 (1944); H.A. Bethe and E.E. Salpeter, *Quantum Mechanics of One- and Two-electron Atoms* (Springer-Verlag, Berlin, 1957).

[7] S.P. Najda, C.J. Armistead, C. Trager, and R.A. Stradling, Semicond. Sci. Technol. **4**, 439 (1989).

[8] S. Huant, S.P. Najda, and B. Etienne, Phys. Rev. Lett. **65**, 1486 (1990); S. Huant *et al.*, in *Proceedings of the 20th International Conference on the Physics of Semiconductors, Thessaloniki, 1990*, edited by E.M. Anastasakis and J.D. Joannopoulos (World Scientific, Singapore, 1990), p. 1369; A. Mandray, S. Huant, and B. Etienne, Europhys. Lett. **20**, 181 (1992).

[9] D.M. Larsen, Phys. Rev. B **23**, 628 (1981).

[10] J. Adamowski, Phys. Rev. B **39**, 13061 (1989).

[11] J.-M. Shi, F.M. Peeters, and J.T. Devreese, Surf. Sci. **305**, 220 (1994); J.-M. Shi, Ph.D. thesis, University of Antwerpen, 1993; J.-M. Shi, F.M. Peeters, and J.T. Devreese, Physica B (to be published).

[12] A. Natori and H. Kamimura, J. Phys. Soc. Jpn. **44**, 1216 (1978).

[13] A.B. Dzyubenko and A.Yu. Sivachenko, Phys. Rev. B **48**, 14 690 (1993).

[14] D.M. Larsen and S.Y. McCann, Phys. Rev. B **46**, 3966 (1992); A.B. Dzyubenko, Phys. Lett. A **165**, 357 (1992).

[15] S. Huant, A. Mandray, J. Zhu, T. Pang, S.G. Louie, and B. Etienne, Phys. Rev. B **48**, 2370 (1993).

[16] T. Pang and S.G. Louie, Phys. Rev. Lett. **65**, 1635 (1990); S.G. Louie and T. Pang, in *New horizons in low dimensional systems*, edited by H. Aoki *et al.* (Kluwer Press, Dordrecht, 1992), p. 445.