PHYSICAL REVIEW B VOLUME 45, NUMBER 20 15 MAY 1992-II

Cyclotron resonance of both magnetopolaron branches for polar and neutral optic-phonon coupling in the layer compound InSe

R. J. Nicholas, M. Watts, and D. F. Howell*
Clarendon Laboratory, Physics Department, Oxford University, Parks Road, Oxford OX1 3PU, United Kingdom

F. M. Peeters and X.-G. Wu†
Department of Physics, University of Antwerp, B-2610 Antwerp, Belgium

J. T. Devreese
Department of Physics, University of Antwerp, B-2610 Antwerp, Belgium
and Eindhoven University, P.O. Box 513, NL-5600 MB Eindhoven, The Netherlands

L. van Bockstal and F. Herlach
Physics Department, K. U. Leuven, Celestijnenlaan 200 D, B-3030 Leuven, Belgium

C. J. G. M. Langerak‡
High Field Magnet Laboratory, University of Nijmegen, Toernooiveld, NL-6525 ED Nijmegen, The Netherlands

J. Singleton
Clarendon Laboratory, Physics Department, Oxford University, Parks Road, Oxford OX1 3PU, United Kingdom
and High Field Magnet Laboratory, University of Nijmegen, Toernooiveld, NL-6525 ED Nijmegen, The Netherlands

A. Chevy
Laboratoire de Physique des Milieux Condensés, 4 place Jussieu, Tour 13, F-75230 Paris, France

(Received 14 January 1992)

Resonant polaron coupling has been observed by cyclotron resonance of two-dimensional electrons in the polar semiconductor InSe. Both upper and lower branches of the magnetopolaron are seen over a wide range of field (B=18–34 T), allowing an accurate test of polaron theories. A homopolar phonon is also observed to give resonant coupling, via a deformation potential, and we present a theoretical treatment of this coupling mechanism. This gives an accurate value of g^2=0.001±0.0005 for the coupling constant.

The most powerful technique for studying electron-phonon coupling in semiconductors is cyclotron resonance. Scanning the magnetic field allows one to induce resonant coupling by bringing the cyclotron frequency ($\omega_c=eB/m^*$) into resonance with the phonon frequency ω_0, where a strongly renormalized magnetopolaron state is formed.[1–6] This causes a splitting of the resonance which is a direct measure of the polaron coupling strength. Several elegant works have been reported in which this coupling has been observed through the transfer of intensity between the upper and lower polaron branches in weakly coupled systems such as the III-V semiconductors InSb, GaAs, and InAs for both bulk[7–9] and two-dimensional[10] (2D) systems. Unfortunately simultaneous observation of both branches has not yet been possible in these systems as even the polar coupling to longitudinal-optic (LO) phonons remains a relatively weak perturbation and other factors such as band nonparabolicity complicate the experimental results. For more strongly coupled systems, such as CdTe and AgBr,[11] the high masses and strong lattice absorption have prevented any successful studies near to the resonance. This paper describes the observation of the resonant Fröhlich coupling in a more polar system, InSe, where the splitting has reached over 40% of the phonon energy due to the large Fröhlich constant (α) of 0.29, and *both branches* of the resonance have been observed simultaneously over a wide field range. This material also shows a second much weaker example of resonant phonon coupling, and we have performed theoretical calculations to demonstrate that this is a spectroscopic observation of *deformation-potential-mediated* coupling. We use this to measure the coupling strength to this second phonon which is shown to be much lower than previously thought.

The material system chosen for study is the layer compound InSe. This has the unusual characteristic that it contains a number of naturally occurring 2D electron gases (2DEG), as first demonstrated by Portal *et al.*[12] The 2DEG systems are formed at stacking faults in the layers, and can exhibit both high mobilities, and well-defined quantum transport phenomena.[13,14] The structural properties allow us to cleave very thin layers of material (a few tens of micrometers) which contain several independent 2D layers giving a large total sheet density of order 2×10^{12} cm^{-2}, and hence a large total absorption. The individual 2DEG's are of low density (of order 1×10^{11} cm^{-2} carriers), and are therefore in the quantum limit at relatively low magnetic fields (>4 T). This is im-

 © 1992 The American Physical Society

portant, as occupation effects have been shown to strongly suppress resonant polaron coupling in high-density systems.[4,15,16] The anisotropic crystal structure results in anisotropic band structure and phonon properties, so that the polaron coupling constants are also anisotropic. The Fröhlich constant $\alpha_\perp$ defined as

$$\alpha_\perp = \frac{e^2}{4\pi\epsilon_0\hbar}\left[\frac{m^*}{2\hbar\omega_{LO}}\right]^{1/2}\left[\frac{1}{\epsilon_\perp(\infty)}-\frac{1}{\epsilon_\perp(0)}\right] \quad (1)$$

gives $\alpha_\perp = 0.29$ for motion perpendicular to the c axis, using accepted values for the phonon frequencies,[17] masses,[18] and dielectric constants.[19] In contrast, motion along the c axis would couple with an $\alpha_\parallel$ of only 0.08.

The experiments consist of direct transmission measurements, close to but outside the *reststrahl* region (22.3-27.3 meV), using an optically pumped far-infrared laser, and both continuous and pulsed magnetic fields (up to 40 T). Sample temperatures were typically 10 K. Experimental recordings for the upper polaron branch, taken in single shots of the pulsed-field system are shown in Fig. 1, and the accurate field positions were taken from the average of several different pulses. No difference was detected between the resonance positions measured in dc fields (up to 30 T) and the pulsed-field measurements.

The resonance and effective masses are plotted as a function of field in Figs. 2(a) and 2(b). The latter shows the very large renormalization effects present in this material, with a splitting between the upper and lower branches of 11 meV at resonance (~40% of the LO phonon energy). The splitting of the cyclotron resonance peak is larger than the *reststrahlen* band, in contrast to previous work on weakly polar semiconductors, where its presence has prevented the simultaneous observation of both branches. The effective mass varies by over a factor of 2. The strength of this coupling, and the wide range of fields over which both branches are observed (~16 T), makes this experiment an excellent test of the existing theories for resonant polaron coupling. The data have been fitted using the memory function approach,[6,20,21] starting from the Fröhlich Hamiltonian

$$H=\frac{1}{2m_b}\left[\mathbf{p}+\frac{e}{c}\mathbf{A}\right]^2+\sum\hbar\omega_{\mathbf{k}}a_{\mathbf{k}}^\dagger a_{\mathbf{k}} +\sum(V_{\mathbf{k}}a_{\mathbf{k}}e^{i\mathbf{k}\cdot r}+V_{\mathbf{k}}^* a_{\mathbf{k}}^\dagger e^{-i\mathbf{k}\cdot r}) , \quad (2)$$

where $\omega_{\mathbf{k}}=\omega_{LO}$, and $|V_{\mathbf{k}}|^2=\alpha(4\pi/V)(\hbar/2m_b\omega_{LO})^{1/2}\times(\hbar\omega_{LO})^2/k^2$, with V the crystal volume and m_b the electron band mass. In Fig. 2 the dashed, dotted, and solid lines show the results of theoretical calculations for coupling to the LO phonon in bulk (3D), sheet (2D), and quasi-2D (Q2D) systems with $\alpha=0.29$. The agreement is reasonable for the 3D case, but is better for the quasi-2D system, where account is taken of the finite z extent of the 2DEG in the symmetric planar layer by fitting with a

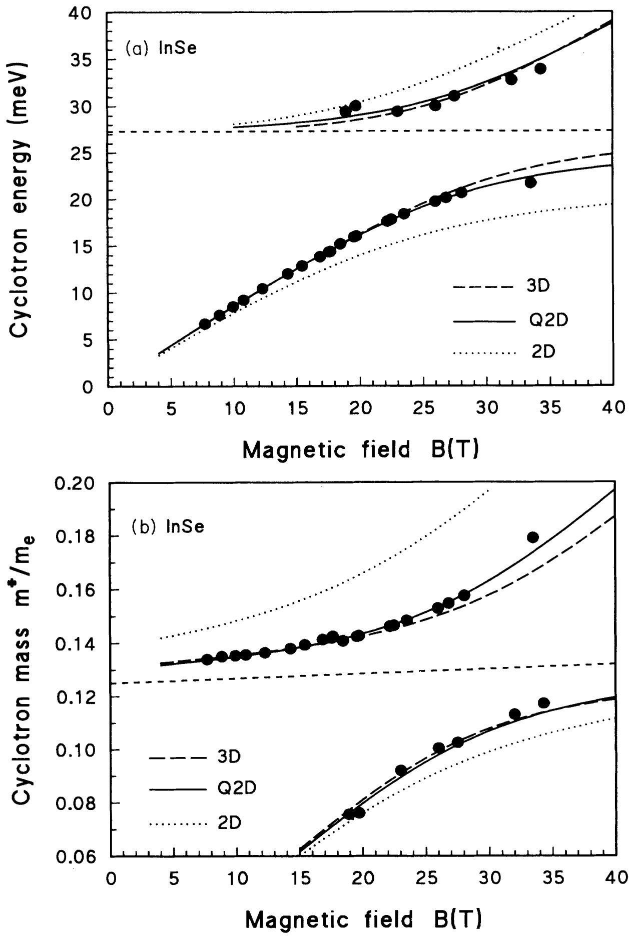

FIG. 2. (a) The resonance position and (b) effective mass plotted as a function of magnetic field. The lines show the results of theory in the limits of 3D and 2D, and after correction for the finite spatial extent in the third dimension (Q2D). The two branches of the polaron are inverted by the effective mass plot.

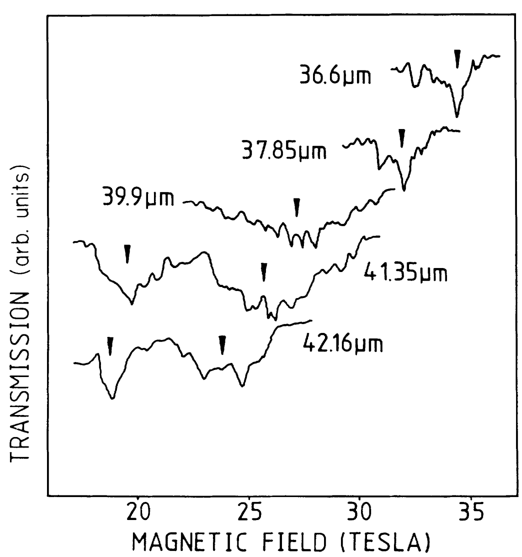

FIG. 1. Typical experimental recordings from single-shot pulsed-field measurements of the upper branch of the magnetopolaron. The transmission is measured using a fast Ge detector, and accurate resonance positions are obtained by averaging several shots.

wave function $|\Psi(z)|^2 \sim e^{-|Z|/a}$, with $a = 36$ Å. This value is in good agreement with the effective Bohr radius for shallow impurities in this material (41 Å), which give binding energies very similar to those found for the 2D layers.[22] A small additional mass increase of 0.14%/T was added to account for the estimated band nonparabolicity, using a conventional Kane model with a band gap of 1.32 eV. The reduction in the splitting from the perfect 2D case is obviously quite large, as can be seen by comparison of the experimental mass splitting ($\Delta m^*/m^* = \pm 0.20$) with either the exact numerical result ($\Delta m^*/m^* = \pm 0.32$) or the analytical formula for the weak-coupling limit[4] of $\Delta m^*/m^* = \pm \frac{1}{2} \alpha^{1/2} \pi^{1/4}$, which gives a splitting of ± 0.31. The reduction in splitting is thus in good agreement with early predictions of this effect.[23]

One further noticeable factor in the resonant-coupling region is a splitting of the resonance into two components very close to the LO phonon frequency, for the traces at 42.16 μm (29.4 meV) and 41.35 μm (30.0 meV). We cannot be definite about the cause of this but it is probably due to the presence of more than one polytype of InSe (Ref. 24) close to the 2D layers with slightly different LO phonon frequencies. The resonance position will be very strongly dependent on the exact phonon frequency at this point. A further possibility which cannot be ruled out is the presence of 2D layers of slightly different carrier concentrations which contribute separate resonances, although this is thought to be less likely as no splittings are observed at lower frequencies.

Careful examination of the effective mass plots shows that a second resonant coupling also occurs in the region 17–18 T, as shown in the expanded plot of Fig. 3. In our earlier work on this material[25] we have attributed this behavior to a coupling to a homopolar optic (HO) phonon at 14.6 meV. This is a new observation of polaronic behavior as there is no macroscopic electric field associated with this phonon mode, and the only coupling mechanism is via a deformation potential. The coupling is, however,

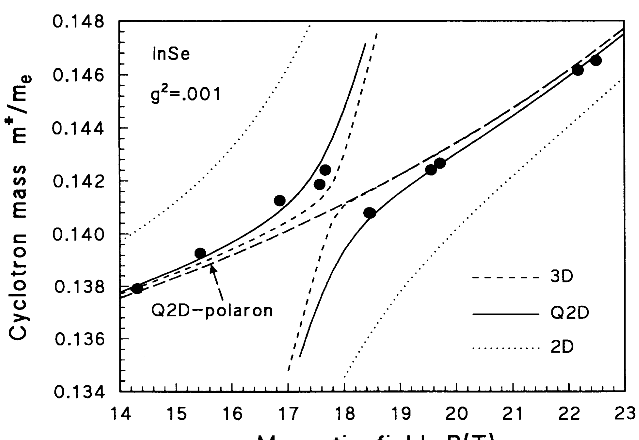

FIG. 3. An expanded portion of the plot of effective mass against magnetic field in the region of the homopolar optic (HO) phonon coupling. The lines show the results of theory for the LO coupling alone (polaron) and using three different values of the HO phonon coupling constant g^2.

thought to be quite strong, as this mode modulates the individual layer thicknesses, and it has been reported[26] that it can make significant contributions to limiting the carrier mobilities at intermediate temperatures. The observation of resonant polaron behavior mediated by a deformation potential offers a different and much more accurate way to assess the importance of this coupling, and is direct evidence of polaron formation via a deformation potential. Deliberate tilted field experiments have shown that this feature cannot be attributed to any sample misorientation and consequent resonant subband-Landau-level coupling,[25] which occurs at lower energies.

The strength of the HO phonon coupling in layer compounds has been a subject of some controversy recently,[25,26] and direct comparison with experiment has been precluded until now due to the lack of a polaron theory which has treated deformation potential coupling in magnetic fields. We have treated the resonant coupling using the memory function formalism and the Hamiltonian [Eq. (2)] in the same way as for the Fröhlich polaron. For the HO phonon interaction[27] the matrix element becomes $|V_{\mathbf{k}}|^2 = g^2(4\pi/V)(\hbar\omega_{\mathrm{HO}})^2/k_{\mathrm{HO}}^3$, where g^2 is the dimensionless electron HO phonon coupling constant, and $k_{\mathrm{HO}} = (2m_b\omega_{\mathrm{HO}}/\hbar)^{1/2}$. We note immediately that this is independent of wave vector k, in contrast to the LO phonon matrix element. The finite wave-function extent was again taken into account using the symmetric wave function fitted to the LO coupling. This factor is very important for the homopolar interaction, as a result of the different wave-vector dependence of $|V_{\mathbf{k}}|^2$ and the larger polaron radius (~45 Å compared to 33 Å for the Fröhlich polaron).

The results are shown in Fig. 3, where a direct fit has been made to the data which includes both the nonresonant LO phonon coupling, and the resonant HO phonon terms. As can be seen the inclusion of both terms provides an excellent fit to the data, using a value for the coupling constant $g^2 = 0.001 \pm 0.0005$. Calculations are also shown for the limits of perfect 2D or 3D coupling to the HO phonon, which clearly demonstrates the much greater sensitivity of the interaction to the finite wave-function extent. If 3D coupling is assumed the value deduced for g^2 is still very small, only increasing to 0.004. In previous work, where g^2 was fitted to the high-temperature mobility, a value over an order of magnitude larger was deduced. This was probably due to difficulties in simultaneously incorporating both the HO and LO phonon scattering, which have rather similar temperature dependences, along with a number of other sample-dependent parameters.[26] For cyclotron resonance in the coupling region the splitting of the resonance is only sensitive to the HO phonon interaction, while the LO phonon coupling and band nonparabolicity only shift the average position of the split resonance.

In summary, we have been able to observe both branches of the resonant magnetopolaron for two different types of optic phonon in the same material allowing a rigorous comparison with theory. The polar LO phonon produces large renormalization of the effective masses due to its stronger coupling, which is well described by existing theory. The deformation potential HO phonon is very weakly coupled, and we present the first theoretical results for a deformation potential mediated magnetopolaron, which allows us to show that its coupling constant is much smaller than previously thought.

One of us (F.M.P.) is supported by the Belgian National Science Foundation, and part of this work is supported by IIKW, FKFO, and IUAP-11(Belgium), and SERC (United Kingdom).

---

*Present address: G.E.C.-Marconi, Hirst Research Centre, Wembley, Middlesex, United Kingdom.
†Present address: University of Ohio, Department of Physics, Athens, OH 45701-2070.
‡Present address: Physics Department, Nottingham University, Nottingham, United Kingdom.

[1]D. M. Larsen, in *Polarons in Ionic Crystals and Polar Semiconductors,* edited by J. T. Devreese (North-Holland, Amsterdam, 1972).
[2]S. Das Sarma and A. Madhukar, Phys. Rev. B **22**, 2823 (1980).
[3]S. Das Sarma, Phys. Rev. Lett. **52**, 859 (1984).
[4]D. M. Larsen, Phys. Rev. B **30**, 4595 (1984).
[5]G. Lindemann, R. Lassnig, W. Seidenbusch, and E. Gornik, Phys. Rev. B **28**, 4693 (1983).
[6]F. M. Peeters and J. T. Devreese, Phys. Rev. B **31**, 3689 (1985).
[7]D. H. Dickey, E. J. Johnson, and D. M. Larsen, Phys. Rev. Lett. **18**, 599 (1967).
[8]C. J. Summers, P. G. Harper, and S. D. Smith, Solid State Commun. **7**, 1267 (1967).
[9]H. Sigg, P. Wyder, and J. A. A. J. Perenboom, Phys. Rev. B **31**, 5253 (1985).
[10]M. Horst, U. Merkt, and J. P. Kotthaus, Phys. Rev. Lett. **50**, 754 (1983).
[11]J. W. Hodby, G. P. Russell, P. M. Peeters, J. T. Devreese, and D. M. Larsen, Phys. Rev. Lett. **58**, 1471 (1987).
[12]J. C. Portal, R. J. Nicholas, E. Kress-Rogers, A. Chevy, J. Besson, J. Galibert, and P. Perrier, in *Proceedings of the 15th International Conference on the Physics of Semiconductors* [J. Phys. Soc. Jpn. **49**, Suppl. A 879 (1980)].
[13]R. J. Nicholas, E. Kress-Rogers, J. C. Portal, J. Galibert, and A. Chevy, Surf. Sci. **113**, 339 (1982).
[14]G. L. Belenkii, M. O. Godzaev, and V. N. Zverev, Pis'ma Zh. Eksp. Teor. Fiz. **43**, 594 (1986) [JETP Lett. **43**, 769 (1986)].
[15]C. J. G. M. Langerak, J. Singleton, P. J. van der Wel, J. A. A. J. Perenboom, D. J. Barnes, R. J. Nicholas, M. A. Hopkins, and C. T. B. Foxon, Phys. Rev. B **38**, 13133 (1988).
[16]Wu Xiaoguang, F. M. Peeters, and J. T. Devreese, Phys. Rev. B **34**, 8800 (1986).
[17]C. Carlone, S. Jandl, and H. R. Shanks, Phys. Status Solidi B **103**, 123 (1981).
[18]E. Kress-Rogers, R. J. Nicholas, J. C. Portal, and A. Chevy, Solid State Commun. **44**, 379 (1982).
[19]N. Kuroda and Y. Nishina, Solid State Commun. **34**, 481 (1980).
[20]F. M. Peeters, X-G. Wu, and J. T. Devreese, Phys. Rev. B **34**, 1160 (1986).
[21]J. T. Devreese and F. M. Peeters, Solid State Commun. **58**, 861 (1986).
[22]E. Kress-Rogers, G. F. Hopper, R. J. Nicholas, W. Hayes, J. C. Portal, and A. Chevy, J. Phys. C **16**, 2439 (1983).
[23]S. Das Sarma, Phys. Rev. B **27**, 2590 (1983).
[24]A. Chevy, A. Kuhn, and M. S. Martin, J. Cryst. Growth **38**, 118 (1977).
[25]D. F. Howell, R. J. Nicholas, C. J. G. M. Langerak, J. Singleton, T. J. B. M. Janssen, and A. Chevy, J. Phys. Condens. Matter **1**, 7493 (1989).
[26]A. Segura, F. Pomer, A. Cantarero, W. Drause, and A. Chevy, Phys. Rev. B **29**, 5708 (1984); A. Segura, B. Mari, J. Martinez-Pastor, and A. Chevy, *ibid.* **43**, 4953 (1991).
[27]Ph. Schmid, Nuovo Cimento **21B**, 258 (1974).