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1989 J. Phys.: Condens. Matter 1 7493

(http://iopscience.iop.org/0953-8984/1/40/024)

J. Phys.: Condens. Matter 1 (1989) 7493–7498. Printed in the UK

LETTER TO THE EDITOR

Two-dimensional magnetopolaron coupling to both homopolar and longitudinal optic phonons in the layer compound InSe

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Received 13 July 1989

Abstract. We have studied cyclotron resonance of the two-dimensional electron gas (2DEG) found in the layered semiconductor InSe at magnetic fields up to 30 T. The results show the first direct observation of resonant polaron coupling to a homopolar optic phonon, coupling via a deformation potential. We have also observed a large increase in the cyclotron effective mass as the LO phonon energy is approached, indicating the strongest polaron effects yet seen in any 2D system ($\alpha \simeq 0.3$).

Polaron coupling in semiconductors and insulators has excited the interest of both theoreticians [1–5] and experimentalists [6–11] for a number of years. Recent developments include extensive studies of two-dimensional (2D) systems in semiconductor heterostructures. To date these studies have primarily involved polaron coupling to weakly polar optic modes in zincblende structure materials where the Fröhlich coupling constant [1] α is of order 0.05.

In this Letter we report evidence for polaron coupling in a 2D electron system formed in the layer compound InSe. Polaron effects arise due to coupling with both conventional longitudinal optic (LO) phonons with a large electric polarisation and a coupling constant of $\alpha \simeq 0.3$ and non-polarised homopolar optic (HO) phonons that modulate the layer thickness and couple via a deformation potential. The importance of the latter, short-range interaction with HO modes polarised along the c axis was first suggested by Fivaz and Mooser [12, 13] for strongly anisotropic layered structures. Subsequently Schmid [14, 15] developed a theoretical model for this interaction in layered structures with a weakly anisotropic band, such as InSe. In contrast, in the case of conventional III–V and II–VI semiconductors with carriers in s-like bands, symmetry arguments suggest that optical deformation potential coupling will vanish in the long-wavelength limit [16].

The presence of a 2D electron gas in bulk InSe was first deduced by Portal and co-workers [17] from magnetotransport studies. Subsequent cyclotron resonance (CR) work by Kress-Rogers and co-workers [18] confirmed the model proposed by Nicholas and co-workers [19] in which the electrons form accumulation layers by binding to impurities

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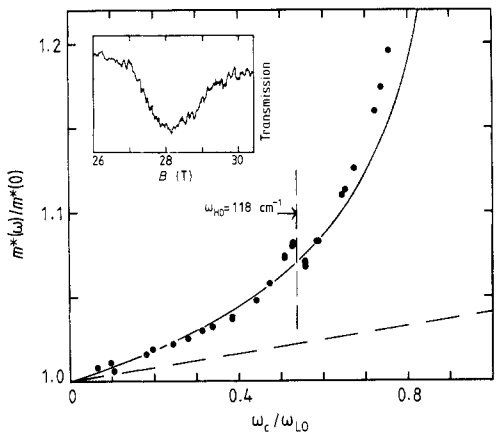

Figure 1. The frequency dependence of the cyclotron effective mass for 2D electrons in a typical InSe sample (taken from ingot 626/180), normalised to $m^*(0) = 0.132m_e$ and $\omega_{LO} = 220\ \text{cm}^{-1}$. The position of the homopolar phonon at $118\ \text{cm}^{-1}$ is indicated. The full curve is from the Devreese and Peeters [5] calculation for bulk material with $\alpha = 0.3$, with the estimated band non-parabolicity added (indicated by the broken line). The inset depicts a typical high-field recording of the magneto-transmission (in arbitrary units).

adsorbed into defect planes. The InSe thus acts as a naturally occurring analogue of the δ-doped structures, widely studied of late [20].

The samples studied were cleaved from single crystals of InSe grown from a non-stoichiometric melt using the Bridgeman–Stockbarger technique [21]. The crystals are thought to be mainly of the γ-polytype with some ε-polytype present. Individual samples were prepared by 'peeling off' thin layers from the crystals using fine tweezers. In an extensive study of CR [22] we have shown that a typical sample, consisting of a sheet of order 50 μm thick, contains several such 2D layers, each with a carrier density of order $1 \times 10^{11}\ \text{cm}^{-2}$. The pieces used for detailed study at high fields were selected to exhibit narrow linewidths [23] at low fields ($\Delta B_{1/2} < 0.2$ T) and for minimum interference effects.

The experiments consisted of a study of the frequency dependence of the effective mass [22] and linewidth by CR at a temperature of 1.4 K, using an optically pumped far-infrared laser and magnetic fields up to 30.4 T. Figure 1 shows a very rapid increase in the effective mass with frequency, characteristic of the approach to resonant polaron coupling at the appropriate LO phonon frequency [24] of $220\ \text{cm}^{-1}$. In addition there is a weak discontinuity in the mass in the region of $118\ \text{cm}^{-1}$, which we attribute to a new 'polaron' coupling to the HO phonon of this frequency [24], previously seen only as a scattering process [25, 26]. Figure 2 shows that there is also a resonant increase in the apparent linewidth [23] associated with both coupling mechanisms.

The strength of the coupling to phonon modes is defined by a dimensionless coupling constant, which for LO phonons is the Fröhlich constant, α, given by [1]

$$\alpha = (e^2/4\pi\varepsilon_0\hbar)\,(1/\varepsilon_\infty - 1/\varepsilon_0)\,(m^*/2\hbar\omega_{LO})^{1/2} \tag{1}$$

where ε_∞ and ε_0 are the high- and low-frequency dielectric constants, ω_{LO} is the phonon frequency and m^* is the effective mass. Using published values for these parameters [18, 24, 27] gives $\alpha_{\perp c} = 0.29$ for motion perpendicular to the c axis. The Fröhlich polaron interaction leads to a renormalisation of the carrier effective mass at low frequencies, given by $m^*_{pol} = m^*(1 - \alpha/6)^{-1}$, and to the resonant polaron effect at higher frequencies. In the case of homopolar modes polarised along the c axis, Schmid [14, 15] has derived a coupling constant, g^2, given by

$$g^2 = (E_d^2/2\sqrt{2}\pi\hbar MN)\,(m^*_{DS}/\hbar\omega_{HO})^{3/2} \tag{2}$$

where E_d is the appropriate deformation potential, m^*_{DS} the density-of-states mass, M

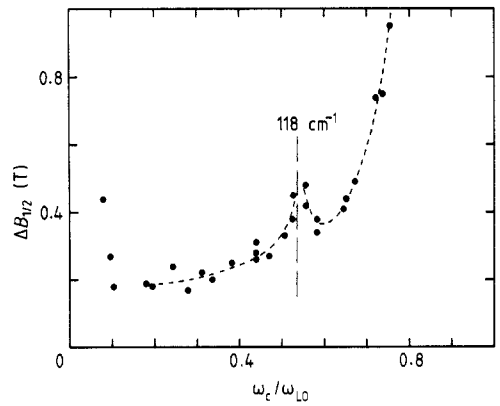

Figure 2. The frequency dependence of the apparent [23] cyclotron resonance linewidth in the same sample. The broken curves are guides to the eye. The large increase in linewidth at low frequencies is the approach to an impurity-related splitting [22] tied to a Landau level filling factor of $\nu = 2$. For ω/ω_{LO} between about 0.35 and 0.5 there is a small increase in the linewidth caused by resonant sub-band–Landau-level coupling [22].

the ionic reduced mass for the phonon mode with frequency ω_{HO} and N is the number of unit cells per unit volume. This leads to a mass renormalisation caused by the homopolar phonon of $m^*_{HO} = m^*(1 - g^2/2)^{-1}$ at low frequencies, analogous to the case of the Fröhlich polaron.

Scattering by homopolar phonons through this interaction is well established in layer compounds [12, 15], in particular for holes in GaSe [25] (which has a band structure and phonon properties similar to those of InSe). InSe has two HO modes polarised along the c axis [24], at 118 cm^{-1} and 230 cm^{-1}: these values have been verified by Raman scattering at 77 K for the crystals studied here. Segura and co-workers [26] have used the temperature dependence of the electron mobility in InSe to deduce a value of $g^2 = 0.054$ for coupling to the mode at 118 cm^{-1}. This suggests that the HO phonon interaction is sufficiently strong in layer compounds for the mass renormalisation described above to be significant, and by analogy with the Fröhlich polaron, resonant coupling should also be visible at the HO phonon frequency.

The results in figure 1 show just such an effect, with a pronounced mass anomaly centred at the HO phonon frequency of 118 cm^{-1}, accompanied by the characteristic increase in linewidth associated with the approach of a resonant polaron effect (figure 2). Zero-field transmission spectra at low temperatures [24, 28] show that this mode is not infrared-active for normal incidence. Thus we do not expect to see a discontinuity in the mass at the phonon frequency due to the dielectric response, as seen close to the polar optic phonons in other 2D systems [11, 29]. All the samples studied in detail showed similar discontinuities at 118 cm^{-1}. CR experiments in which the sample was deliberately tilted showed that the discontinuity was not due to resonant sub-band–Landau-level coupling [30], which was found to occur at lower frequencies.

Estimating an effective coupling strength for the homopolar mode is difficult, due both the comparative weakness of the coupling and the lack of any theoretical description for the resonant interaction. One method for estimating g^2 is via the self-energy correction, ΔE, which we expect to disappear above the phonon frequency [4, 6]. Figure 3 demonstrates this effect clearly, with a step of around 2 cm^{-1} at the HO phonon frequency on a plot of the resonance energy against magnetic field. According to Schmid's theoretical calculations [14, 15] the self-energy correction at absolute zero is given by

$$\Delta E = (2 \ln 2) g^2 \hbar\omega_{HO} (I/\hbar\omega_{HO})^{1/2} \tag{3}$$

which I is the conduction band width. This result should be compared with $\Delta E = \alpha\hbar\omega_{LO}$

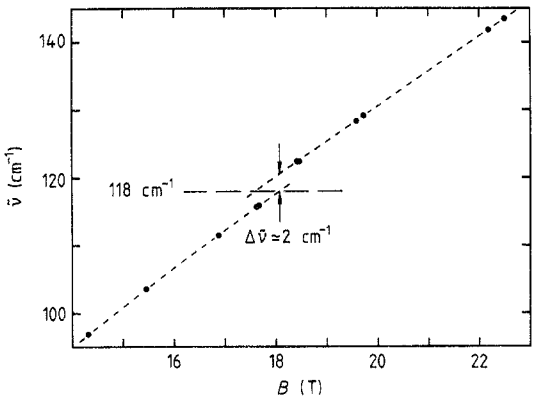

Figure 3. The frequency dependence of the resonant field in the region of the homopolar optic phonon at 118 cm^{-1}. The broken lines are guides to the eye. The disappearance of the 'polaron' self-energy correction ΔE is clearly visible at 118 cm^{-1}.

for the Fröhlich interaction [1]. We estimate I to be around 1 eV from band-structure calculations [31]. Using this value and $\Delta E = 2\,\text{cm}^{-1}$, equation (3) gives a coupling constant of $g^2 \simeq 0.002$. Another rough estimate can be obtained by noting that the resonant coupling is approximately ten times weaker than the LO phonon interaction in GaAs, for which data are available [11] up to $\omega/\omega_{LO} = 0.9$ and the theory is well founded. A linear comparison suggests a coupling constant of around $g^2 \simeq 0.005$. These results are almost two orders of magnitude weaker than the LO phonon coupling in InSe (with $\alpha \simeq 0.3$) and suggest that the polaron coupling to the homopolar mode is in fact rather weak.

We can now compare the measured and expected strengths of the homopolar interaction in InSe. For holes in GaSe Schmid and Voitchovsky [25] deduced a value for the homopolar deformation potential of $E_d = 6.6$ eV Å^{-1} from $g^2 = 0.25$. Assuming a similar value of E_d for InSe, and scaling this with the relevant masses in equation (2), yields a value of $g^2 = 0.01$. This value is larger than that deduced from our cyclotron resonance measurements, but the factor of two to five discrepancy may reflect a difference in deformation potential between the two materials, as is evidenced by their very different pressure coefficients for the direct band gap [32]. A smaller value of E_d (2–3 eV Å^{-1}), as suggested by our results, is in fact comparable to values deduced theoretically for the conduction band, for deformations in the thickness and separation of the layers [32]. These theoretical values are in agreement with the pressure coefficient of the band gap, but were not calculated explicitly for the HO modes [32]. Alternatively, the discrepancy between the apparent value for electrons in InSe and holes in GaSe may represent a substantial difference in the deformation potentials for the conduction and valence bands in layered semiconductors.

Segura and co-workers [26] deduced a larger value of $g^2 = 0.054$ for electrons in InSe based on an analysis of the temperature dependence of the electron mobility over the range 120 to 500 K, assuming that this is dominated by HO phonon scattering. However, this is almost certainly an overestimate as no account was taken of LO phonon scattering which is very significant in this temperature range. The relative coupling strengths can be gauged from the size of the resonant polaron broadening seen in figure 2, where the LO phonon is clearly dominant despite the fact that the data stop some way from resonance. In fact, calculations of the mobility due to polar optic phonon scattering [22], with $\alpha = 0.29$. $\omega_{LO} = 220\,\text{cm}^{-1}$, are a good fit to both the temperature dependence of the cyclotron linedwidth [22] and the experimental data of Segura and co-workers [26]

above 120 K. Thus the experimental evidence all points to a small value for g^2, of order 0.002–0.005 and a value for the homopolar deformation potential of 2–3 eV Å^{-1}. Note that even the simple theoretical estimate presented above suggests that g^2 is a factor of five smaller than the value deduced by Segura and co-workers [26]. This conclusion is in marked contrast to previous work on layered compounds [12, 15], where coupling to HO phonons is usually the dominant scattering mechanism.

Returning to the Fröhlich interaction, the overall mass enhancement is clearly dominated by coupling to the LO mode (figure 1), reaching 20% by 166 cm^{-1} (corresponding to $\omega/\omega_{LO} = 0.76$). These values are considerably larger than previously found in any 2D system, and are comparable to those seen in bulk CdTe [5, 7], which has a comparatively large coupling constant of $\alpha \simeq 0.3$. There will be a small contribution to the mass increase due to band non-parabolicity. This is estimated to be about +3% at $\omega/\omega_{LO} = 0.8$ from two-band $\boldsymbol{k} \cdot \boldsymbol{p}$ theory although the exact value is uncertain due to the lack of detailed knowledge of the band structure. The estimated band non-parabolicity has been indicated by the broken line on figure 1. In order to provide some comparison with theory we have also plotted on figure 1 the theoretical results of Devreese and Peeters [5] for polaron coupling in bulk materials with $\alpha = 0.3$, modified to incorporate the estimated band non-parabolicity. This theory appears to give a reasonable description of the InSe 2D results. However, at the highest frequencies, there is some indication of an increase in the coupling strength as the LO phonon is approached. This is in good agreement with Langerak and co-workers [11] who found very similar results when comparing bulk and 2D electrons in GaAs, provided that the system was in the low-density limit.

Our conclusion is that by studying the layer compound InSe we have observed a new type of 'polaron' coupling to homopolar optical phonons polarised along the c axis coupling only via a deformation potential. This interaction is allowed in layered materials due to the low structural symmetry which causes the coupling to appear in first order of the atomic displacements. However, we find that the coupling strength is much weaker than anticipated from previous transport data. At high frequencies we observe conventional polaron coupling with the largest mass enhancement yet seen in any 2D system.

We acknowledge the invaluable technical support of K van Hulst, H van Luong, J A A J Perenboom and J Rook. This work was supported by the SERC (UK), FOM and NWO (The Netherlands).

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