PROC. 15TH INT. CONF. PHYSICS OF SEMICONDUCTORS, KYOTO, 1980
J. PHYS. SOC. JAPAN **49** (1980) SUPPL. A p. 879–882

TWO-DIMENSIONAL BEHAVIOUR OF ELECTRONS IN BULK InSe FROM SHUBNIKOV-DE HAAS OSCILLATIONS AND CYCLOTRON RESONANCE

J.C. Portal*, R.J. Nicholas†, E. Kress-Rogers†, A. Chevy^x, J. Besson^x, J. Galibert* and P. Perrier*

* I.N.S.A., Dept. de Physique, Avenue de Rangueil, 31077 Toulouse, France.
† Clarendon Laboratory, Parks Road, Oxford OX1 3PU, U.K.
x Université P. et M. Curie, 4 Place Jussieu, 75230 Paris, France.

The first observations of cyclotron resonance and the Shubnikov-Haas effect are reported for the layer compound InSe. The Cyclotron effective mass is found to be 0.13 m_o. Four series of Shubnikov-de Haas oscillations have been observed. The Landau level separations are shown to be determined only by the component of magnetic field perpendicular to the c-axis. This apparently two-dimensional behaviour is thought to be due to the presence of defects which trap the carriers within a few layers.

Electrical transport measurements in the layer compounds InSe [1] and GaSe [2] have shown that electrical conduction in these materials is highly anisotropic. In particular conduction perpendicular to the layer planes exhibits an activated mobility, with a mobility ratio $\mu\perp c/\mu\| c \sim 100$ even at room temperature. This is thought to be due to the presence of large numbers of stacking faults which inhibit motion in the direction of the c-axis. In contrast exciton absorption [3,4], Resonant Raman [5] and time of flight [7] measurements indicate that both the conduction and valence bands exhibit very little anisotropy with the effective mass showing some evidence of an anomalous anisotropy with $m_\| < m_\perp$. Both two and three dimensional band structure calculations have been found to provide a good description of the density of states measured by x-ray photoemission [10,11]. The present paper reports measurements of cyclotron resonance and Shubnikov-de Haas oscillations in InSe, which demonstrate that the cyclotron orbit remains trapped within the layer planes, and thus exhibits an apparently two-dimensional behaviour.

The samples of InSe were grown by the Bridgemann technique and are thought to be mainly of the $C_{3v}(\gamma)$ polytype, with some of the $D_{3h}(\varepsilon)$ polytype also present. These two structures transform very easily into each other in the presence of stacking faults [3].

Cyclotron resonance absorption was measured in the Faraday geometry using 337 μm, 311 μm, 195 μm and 119 μm radiation from far-infrared gas lasers, at lattice temperatures between 10 K and 40 K. Figure one shows typical recordings of the magnetotransmission taken at 10 K in the orientation B||c , and with the magnetic field oriented at 45° from the c-axis. For B||c there is a resonance absorption which narrows rapidly with increasing frequency. The effective mass $m_\perp$, is determined as 0.128 m_o, 0.132 m_o and 0.137 m_o at 337 μm, 195 μm and 119 μm respectively. This is the first direct experimental determination of the electron effective mass using resonant techniques. It compares with values of fitting resonant raman scattering amplitudes [5]. Bourdon et al [12] have estimated $m_\perp$ as 0.18 m_o from k.p perturbation theory. The exciton reduced effective mass m has been found to be 0.115 m_o for direct E_o gap excitations from fitting the excitonic Rydberg [13] and diamagnetic shift [4], while the E_1 direct gap exciton mass was determined as 0.119 m_o from the Rydberg [14] and magnetoabsorption [15]. It is thus clear that the exciton reduced mass is dominated by the electron contribution.

The linewidth falls by a factor of two in going from 337 μm where $w\tau$=6 to 119

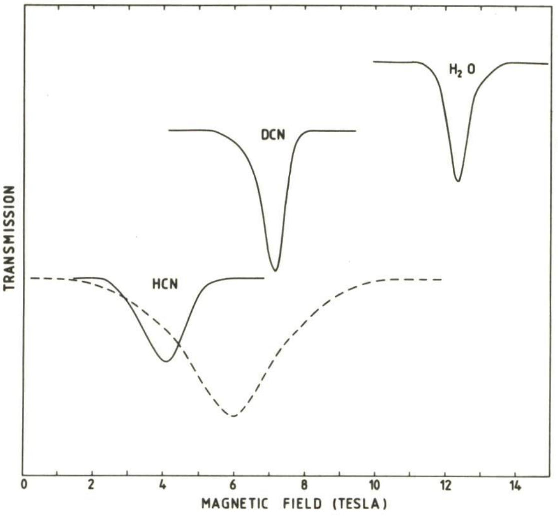

Fig.1 Cyclotron resonance of InSe at a lattice temperature of 10K for 337 μm (HCN-Laser), 195 μm (DCN-Laser) and 119 μm (H_2O-Laser)

Solid lines : $\Theta(\vec{B},\vec{c}) = 0^o$

Dashed line : $\Theta(\vec{B},\vec{c}) = 45^o$

μm where $\omega\tau = 38$, corresponding to an increase in relaxation time from 1 to 2 × 10^{-12} sec. This effect is thought to be related to the decrease in cyclotron radius, which is 132 Å at 4.0 T and 74 Å at 12.4 T, for the two resonance fields respectively.

When the c-axis is rotated from B || c, the resonance field for 337 μm radiation is found to follow a $B*\cos\Theta$ law up to 45°, where Θ is the angle between the magnetic field and the c-axis. There is no strong change $\omega\tau$ upon rotation.

Shubnikov-de Haas oscillations were observed in four samples (A-D) at 4.2 K and 1.5 K (Fig.(2a,c)). The oscillations are complex and Fourier analysis reveals the presence of up to four different oscillatory series (Fig.(2b)). The fundamental fields are summarised in Table 1 for samples A, B and D which all show the presence of four series with fundamental fields in the ratio 1 : 1.6 : 2 : 5.4. The relative amplitudes of the different series varies strongly between the different samples with sample A showing a stronger contribution from series 4 and sample D having a strong contribution from series 1. Sample C, with a lower doping level showed one strong peak thought to be the fundamental of series 4. The effective masses as deduced from the temperature dependence of the amplitudes of the oscillations (m_T^*) are shown in Table 1. The mass for the lowest series, 0.14, is in good agreement with that deduced from the cyclotron resonance data. Using these masses the Shubnikov-de Haas periodicities are consistent with a single Fermi energy for each of the samples, of 2.7, 3.1 and 3.2 meV for A, D and B respectively. This corresponds to a doping level of 1.8 × 10^{11} cm^{-2} for the first ($m_T^* = 0.14\ m_o$) series of sample D in a two-dimensional model, or 4 × 10^{16} cm^{-3} for a three-dimensional model. The alternative explanation for the presence of several series would be due to the occupation of several non-degenerate band edges, possibly separated by a few meV, which would result in differing relative Fermi levels.

Table 1 : The Shubnikov-de Haas Data

	$B_{F_1}(T)$	$B_{F_2}(T)$	$B_{F_3}(T)$	$B_{F_4}(T)$
Sample A	3.2	5.0	6.4	17.0
Sample B	3.9	6.2	7.9	21.1
Sample D	3.7	5.9	7.5	20.1
m_T^* (m_o) (all samples)	.14 ± .02	.26 ± .04	.27 ± .04	.8 ± .05

The presence of four different effective masses and series of oscillations, seen in the Shubnikov-de Haas data, is quite unexpected from band structure considerations [7-10,12]. However, it is possible that small p-type regions are present in the vicinity of the stacking faults, where acceptors have accumulated. These regions could give rise to Shubnikov-de Haas oscillations from holes, which would explain the observation of large effective masses. The hole ($m_\perp$) masses have been calculated by Bourdon [12] to be 0.22 m_o (Γ_6^-), 0.26 m_o (Γ_6^-) and 5 m_o (Γ_1^-). Another possibility is that several non-degenerate conduction band minima exist due to the presence of more than one polytype in the samples investigated. ε- and γ- modifications occur mixed, in the Bridgemann samples [3, 16], and measurements on GaSe have shown that the band edges [16], and exciton binding energies [3], may be shifted by energies of the order of 1 meV between the non-equivalent layers in ε- and γ- modifications. The strong intensity variations of the different series may thus be due to the polytypism of the particular samples studied.

Further evidence of the two-dimensional behaviour of the carriers is shown in Fig. 3, where the Shubnikov-de Haas oscillations are shown as the c-axis is rotated away from $B \| c$. The oscillations and hence Landau level separations are determined only by the component of B perpendicular to the c-axis, $B{*}\cos\Theta$, up to an angle of 80°. This two-dimensional behaviour is in complete contrast to the exciton, [3, 13], and band structure deductions. The most likely explanation lies in the presence of large numbers of defects, which produce potential barriers between the layers, and thus inhibit motion along the c-axis. The exciton may still be able to behave in a three dimensional manner due to the smaller exciton radius (50Å). In the Drude formalism such an effect could be described in terms of a highly anisotropic relaxation time since $\tau_\perp/\tau_{11} >> 1$ then the two cyclotron resonances will decouple and an exact $B{*}\cos\Theta$ behaviour will be observed.

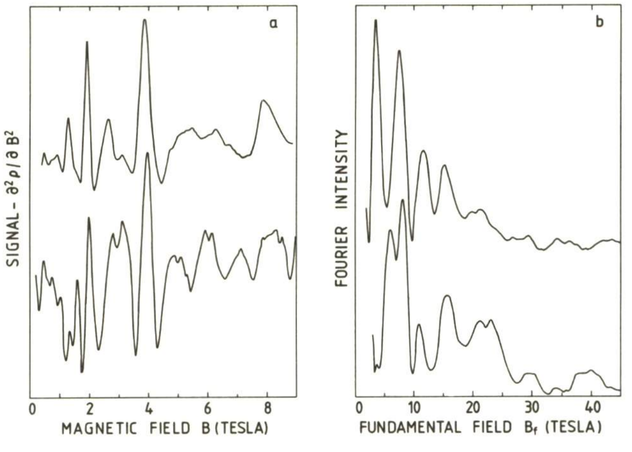

Fig. 2a Shubnikov-de Haas oscillations: second derivative of the resistivity for samples B (lower trace) and D (upper trace) at 1.5K 2b Fourier analysis reveals the periodicities $1/B_f$ of the complex oscillations: A comparison with the original traces shows that in addition to the fundamental periodicities shown in Table 1, higher harmonics are also present due to peak sharpening in the original recordings

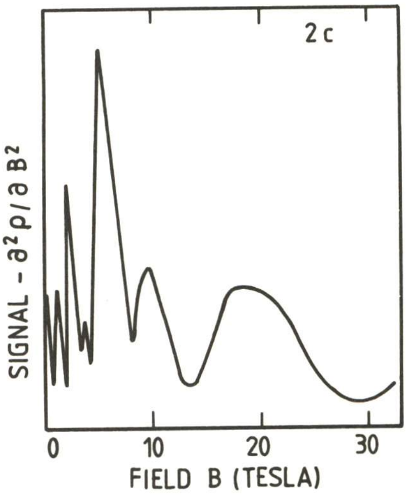

Fig.2c Shubnikov-de Haas oscillations in a pulsed magnetic field for sample B at 4.2K: At 20T and 10T resistivity maxima belonging to series 4 with a fundamental field of B_{f4}=21T are clearly visible. The higher order maxima of this series at lower magnetic fields are strongly dampened due to its high mass.

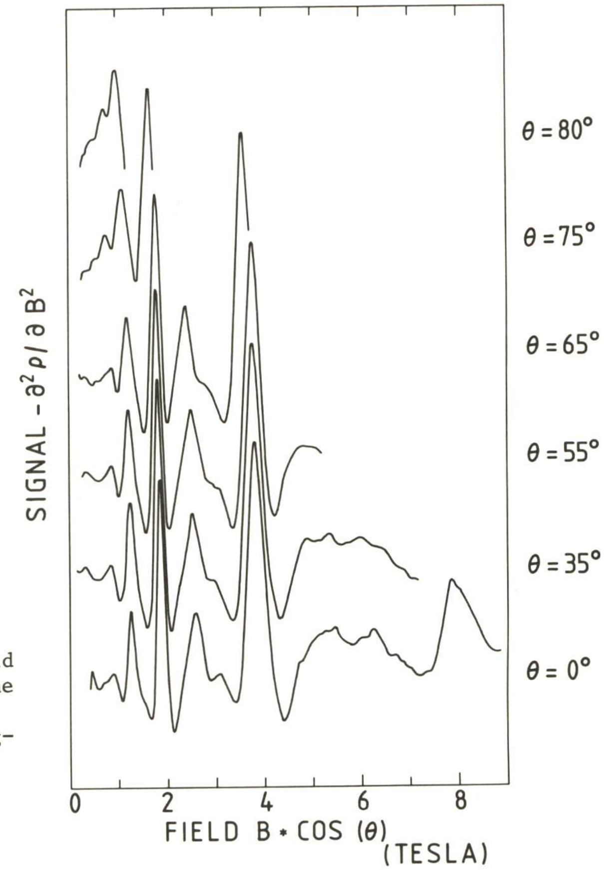

Fig. 3 Angle dependence of the Shubnikov-de Haas oscillations for sample D up to an angle of $\Theta(\vec{B},\vec{c})$ = 80°: The traces show that the conductivity depends only upon the component B*cos Θ of the magnetic field perpendicular to the layers

References

1) S.M. Atakishiev and G.A. Akhundov: Phys. Stat. Sol 32 K33 (1968).
2) R. Fivaz and E. Mooser:Phys. Rev 163 743 (1967).
3) E. Mooser and M. Schlüter:Nuovo Cimento 18B 164 (1973).
4) J.C. Merle, R. Bartiromo, E. Borsella, M. Piacentini and A. Savoia: Solid. State. Commun. 28 251 (1978).
5) N. Kuroda and Y. Nishina: Solid State Commun. 34 481 (1980).
6) G. Ottoviani, C. Canali, F. Nava, Ph Schmid, E. Mooser, R. Minder and I. Zschokke: Solid State Commun. 14 933 (1974).
7) M. Schlüter:Nuovo Cimento 50B 95 (1967).
8) J.V. McCanny and R.V. Murray: J. Phys. C: 10 1211 (1977).
9) A. Balzarotti, R. Girlandi, V. Grasso, E. Doni, F. Antonangeli, M. Piacentini and A. Baldereschi: Solid State Commun. 24 327 (1977).
10)E. Doni, R. Girlanda, V. Grasso, A. Balzarotti, and M. Piacentini: Nuovo Cimento 51B 154, 181 (1979).
11)G. Margoritondo, J.E. Rowe and S.B. Christman: Phys. Rev. B15 3844 (1977).
12)A. Bourdon, A. Chevy and J.M. Besson: Proc. 14th Int. Conf. on Physics of Semicond., Edinburgh (1978) (I.O.P. Conf. Ser. 43) 1371 and priv. comm.
13)J. Camassel, P. Merle, H. Matheiu and A. Chevy: Phys. Rev. B17 4718 (1978).
14)N. Kuroda, I. Munakata and Y. Nishina: Solid. State. Commun. 33 687 (1980).
15)N. Kuroda, Y. Nishina, H. Hori and M. Date: J. Phys. Soc. Japan 47 1373 (1979).
16)E. Aulich, J.L. Brebner and E. Mooser Phys. Stat. Sol. 31, 129 (1969).