Observation of Optically Detected Magnetophonon Resonance

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(Received 5 September 1990)

We present the first observation of the magnetophonon effect in the frequency-dependent conductivity of electrons in a polar semiconductor. The cyclotron-resonance position and linewidth oscillate as a function of magnetic field and show resonances for $\omega_{LO} \approx N\omega_c$. The resonances grow rapidly with increasing temperature, giving a direct measure of the optic-phonon scattering rate. Good agreement is found with a one-electron theory of the absorption spectrum, showing that the oscillations have the same origin as magnetophonon-resonance oscillations in the resistivity.

PACS numbers: 71.38.+i, 78.65.Fa

Magnetophonon resonance (MPR) occurs when two Landau levels are a phonon energy apart, leading to an increase in the probability of absorption or emission of (usually) optic phonons. The vast majority of authors have investigated the influence of this resonant scattering on the transport properties of semiconductors, usually the magnetoresistance, in what is inevitably a complicated average of the scattering processes. In this Letter we describe a direct measurement of the resonant scattering through a study of the cyclotron-resonance (CR) linewidth, which shows strong temperature-dependent resonances close to the conventional MPR condition:

$$N\omega_c = \omega_{\rm LO}$$
, (1)

where ω_c is the cyclotron frequency (eB/m^*) and ω_{LO} is the longitudinal-optic- (LO-) phonon frequency. In addition, we have developed the theory of this effect in a calculation of the high-frequency conductivity using the memory-function approach. This clearly predicts the oscillatory linewidth, and shows a simultaneous shift in the CR position caused by the interrelation of the real and imaginary parts of the memory function. This is also observed experimentally.

MPR was first predicted by Gurevich and Firsov, who calculated that resonant structure should appear in both the transverse and longitudinal magnetoresistance. It has been extensively studied both in bulk materials (see Refs. 2 and 3 for reviews) and more recently in 2D systems. The oscillations in the magnetoresistance are the result of a combination of scattering and broadening processes which can lead to a quite complicated dependence of the resonance amplitudes on doping, sample structure, carrier concentration, and temperature. Our observation of the optically detected MPR (ODMPR) allows us to make quantitative measurements of the scattering for transitions between specific levels. The theory and experiment also show that the resonance position can provide a very direct measure of the resonant

scattering.

CR was observed in three conventional high-mobility GaAs-(Ga,Al)As heterojunctions [A, B, and C, withspacer layers of 800, 200, and 100 Å, and carrier concentrations $(n_e, \text{ in units of } 10^{11} \text{ cm}^{-2})$ of 0.8 (1.6), 3.4 (5.2), and 6.2 in the dark (light)] over a range of temperatures (4.2-100 K). Data were taken using either a Fourier-transform spectrometer or a far-infrared laser. The linewidths (ΔB) were measured directly as the half width at half absorption, and were uncorrected for the finite carrier concentrations. Previous work on CR linewidths at high temperatures⁶ has shown that at relatively low fields (\sim 6 T), ΔB is dominated by nonresonant optic-phonon scattering for temperatures above 60 K. ΔB was found to be well described by the analytic form of the expression first derived for short-range scatterers,⁷

$$\Delta B = C(B/\tau_0)^{1/2} \,, \tag{2}$$

where C is an experimental constant and τ_0 is the opticphonon-limited scattering time calculated by Walukiewicz *et al.*⁸ These data were modeled by Wu and Peeters, ⁹ who found a good fit when a Landau-level broadening of around $0.1 \hbar \omega_{LO}$ was introduced.

Figure 1 shows a typical series of resonances, together with plots of the energy dependence of the linewidth and cyclotron effective mass for three temperatures. These show an overall increase in linewidth with temperature, together with the very obvious appearance of a strong peak at an energy (field) of around 140 cm⁻¹ ($B \approx 10.5$ T). This is close to the N=2 MPR condition; MPR is known experimentally to occur at submultiples of a field of 21-22 T.^{5,10} Weaker features are also visible in some cases close to the N=3,4 conditions. The magnitude of the additional magnetophonon contribution to the linewidth appears to be almost independent of sample and carrier concentration; however, the position of the peak moves up in energy as the temperature and carrier con-

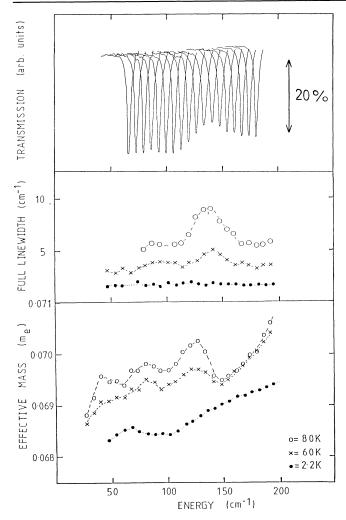


FIG. 1. Upper section shows a series of cyclotron-resonance traces taken at 0.5-T intervals from 14 to 5.5 T at 80 K for sample B. The middle and lower sections show the frequency dependence of the resonance full width and cyclotron effective mass at 80, 60, and 2.2 K.

centration increase. For the lower carrier concentrations the resonance positions appear a few percent lower in field than seen in equivalent MPR experiments in the resistivity,⁵ although the majority of these are performed at higher temperatures. In recent works^{5,11} on MPR in the resistivity of high-mobility heterojunctions, wellresolved series of oscillations were seen up to N=7, which gave an apparent phonon energy of order 5% lower than the normal LO phonon, roughly intermediate between the values of the LO and TO phonons. Calculations suggest 12 that this may be caused by shifts in the resonance position caused by the presence of broadening. The experimental "damping" of the oscillations, which is measured from the relative strength of the successive resonances, is considerably stronger in the ODMPR than the resistivity and thus may produce a larger shift in the apparent resonance position.

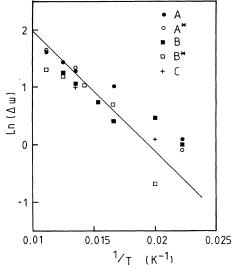


FIG. 2. An activation plot of the resonant peak in the linewidth $(\Delta \omega$, measured in cm⁻¹) as a function of inverse temperature for the three samples studied, A, B, and C, in the dark, and A^* and B^* for the same samples after the excitation of persistent photoconductivity. The solid line corresponds to a dependence $\Delta \omega \propto \exp[-(\hbar \omega_{\rm LO}/2kT)]$.

The overall picture is thus one of a temperature-dependent background, which is only weakly field dependent, with one or more resonant magnetophonon peaks superimposed on top of this. An Arrhenius plot of the linewidth at resonance, with the underlying background subtracted, is shown in Fig. 2. This demonstrates both the sample independence, and the exponentially activated nature of the additional scattering, which has an activation energy of 200 K. This corresponds to half the optic-phonon energy, as found for the nonresonant contribution to the linewidth⁶ and in the calculations below.

The CR effective masses m^* (Fig. 1) also show oscillatory structure, and it is easier to distinguish the higher resonances in plots of the mass, than in the equivalent linewidth data. The resonances are, however, shifted in phase from those in the linewidth. This is due to the dispersive relationship in the field (and energy) dependence of the resonance positions and linewidths, resulting from the Kramers-Kronig type of relationship between the two quantities. 13 The mass can be a more sensitive probe of the oscillations, due to both its derivative-type relation to the linewidth and the greater accuracy in assigning the center of an absorption peak as compared to its linewidth. The MPR positions deduced from the two measurements are nevertheless in good agreement. The underlying temperature-dependent increase in mass agrees with previous data, 11 although its origin remains a matter of controversy. 14-16

Magnetophonon resonances are essentially single-particle effects, and will thus arise directly in a one-polaron theory. Many-particle effects¹⁷ will change the effect

quantitatively, but will not be included in this first description of the theory. The CR absorption spectrum is calculated from the complex conductivity $\sigma_{\mu\nu}(\omega)$, which is the Fourier transform of the velocity-velocity relaxation function $[r_{\mu}(t), r_{\nu}(0)]$. This can be written as

$$\sigma(\omega) = i \frac{e^2 n_e / m_b}{\omega - \omega_c - \Sigma(\omega)} , \qquad (3)$$

where n_e is the carrier concentration, ω_c is the "bare"

cyclotron frequency, and $\Sigma(\omega)$ is the memory function, which contains the effect of the electron-phonon interaction. $\Sigma(\omega)$ is in essence a force-force correlation function, which is well treated by second-order perturbation theory in a weakly polar material such as GaAs. Following Refs. 18 and 19 we find

$$\Sigma(\omega) = \frac{1}{\omega} \int_0^\infty dt (1 - e^{i\omega t}) \text{Im} F(t) , \qquad (4)$$

with

$$F(t) = -\sum_{k} \frac{k^{2}}{m_{b} \hbar} |V_{k}|^{2} f(k) \{ [1 + n(\omega_{LO})] I(k, t) - n(\omega_{LO}) I^{*}(-k, t) \} e^{-i\omega_{LO}t},$$
(5)

where $n(\omega)$ is the LO-phonon occupation number, and f(k) is a form factor to describe the nonzero width of the real 2D gas.²⁰ The Fourier transform of the density-density correlation function is calculated for finite temperature

$$I(k,t) = \exp\left[-\frac{\hbar^2 k^2}{2m_b \omega_c} [1 - e^{i\omega_c t} + 4n(\omega_c)\sin^2(\omega_c t/2)] - \frac{\Gamma^2 t^2}{4\hbar^2}\right],$$
 (6)

with Γ a collisional broadening parameter. Γ is introduced semiempirically to remove the δ -function divergence of the Landau-level densities of states (DOS). Note that Γ is not simply related to the true Landau-level DOS broadening, and will in practice depend on the magnetic field, temperature, and Landau-level index. ²¹ We initially take Γ as constant, and then adjust it to obtain a quantitative agreement with experiment.

The experimental results are simulated using Eq. (3), from which we can see that $\text{Re}\Sigma(\omega)$ is responsible for the shift in the CR frequency, which is governed by the reactive part of the interaction. $\text{Im}\Sigma(\omega)$ gives the width of the CR peak, representing the dissipative part of the interaction. These two components of the memory function are related, as mentioned above, by the Kramers-Kronig relation

$$\operatorname{Re}\Sigma(\omega) = -\frac{1}{\pi} \int_{-\infty}^{+\infty} dx \frac{\operatorname{Im}\Sigma(x)}{\omega - x} , \qquad (7)$$

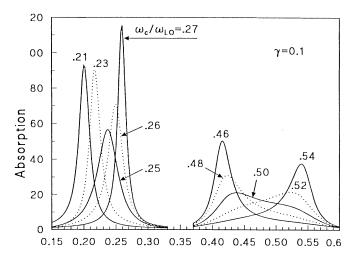
which is exactly satisfied by our theoretical result. 19

The zero-frequency limit of Eq. (3) gives the resistivity directly from $\rho_{xx} = -\text{Im}\Sigma(\omega=0)$, which results in conventional MPR, as studied by Warmenbol, Peeters, and Devreese. ²² This comparison shows us the considerable interest and additional information given by the ODMPR. The oscillations are from a common origin, but the study of the frequency-dependent conductivity now allows us to observe both components of the memory function, in a direct manner which is much less influenced by the other scattering processes present.

Numerical results for the absorption spectra as a function of frequency are shown in Fig. 3 for magnetic fields (measured in terms of $\omega_c/\omega_{\rm LO}$) close to the N=4 and N=2 resonances for perfectly 2D electrons in GaAs. The broadening is $\gamma=0.1$ ($\gamma=\Gamma/\hbar\,\omega_{\rm LO}$) and the temperature $kT=0.21\,\hbar\,\omega_{\rm LO}$ ($\simeq 90$ K). The cyclotron peak around N=4 shows the expected broadening, while the simulations at $\omega_c/\omega_{\rm LO}=0.48-0.52$ demonstrate very

clearly the nonlinear shift of the cyclotron peak with magnetic field. The resonance has developed an anticrossing behavior, with the intensity switching from the lower- to higher-frequency components. At this temperature over 90% of the carriers are in the lowest electronic Landau level (l=0); however, the finite optic-phonon population leads to a considerable probability of being in an $|l=0\rangle+1$ phonon state. This anticrosses with the $|l=2\rangle$ Landau level, causing the total transition to split, with a splitting which is found to increase in size with increasing temperature.

Quantitative fits are shown in Fig. 4, with the CR linewidth and mass as a function of magnetic field for



Frequency: ω/ω_{LO}

FIG. 3. Theoretical calculations of the absorption (arbitrary units) as a function of frequency for magnetic fields (defined by ω_c) close to the N=4 and N=2 MPR conditions. The temperature is 90 K and the broadening is 43 K. Alternate curves are dotted for clarity.

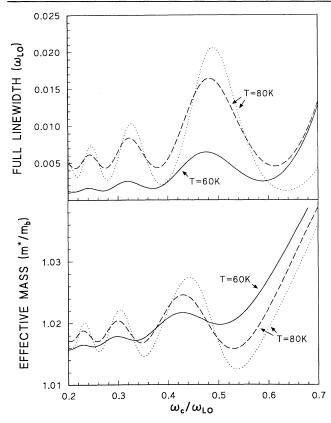


FIG. 4. Theoretical results for the frequency dependence of the cyclotron linewidth and effective mass at 60 and 80 K. The broadening is $\gamma = \Gamma_0(\omega_c/\omega_{LO})^{1/2}$; the dashed and solid curves are for $\Gamma_0 = 0.25$, and the dotted curve is for $\Gamma_0 = 0.2$.

conditions corresponding to those of Fig. 1 ($n_e = 3 \times 10^{11}$ cm⁻²). The broadening was taken⁷ as $\gamma = \Gamma_0(\omega_c/\omega_c)$ $\omega_{\rm LO}$) ^{1/2}, with results shown for $\Gamma_0 = 0.25$ and 0.2. The figures clearly show the derivativelike relation between the mass and the linewidth as found in the experiment. The magnitude of the oscillations in both the linewidth $(\Delta B \approx 0.01 \omega_{LO} = 3 \text{ cm}^{-1})$ and effective mass $(\Delta m^*/m^*)$ \approx 0.01) are in good agreement with the experimental results at 80 K, using $\Gamma_0 = 0.25$. The calculations also show a small shift away from the exact resonance condition (1), as found in the experiment. The amplitude of the oscillations increases rapidly with temperature, as expected, and further calculations show that it is proportional to the square root of the LO-phonon occupation number for a wide range of temperatures. This is also in agreement with the experimental results above (Fig. 2), and is due to the internal self-consistency of the theory. A similar temperature dependence is observed for the nonresonant scattering [Eq. (2)]. The relatively strong damping required to fit the experimental results explains why no strong asymmetry or splitting of the resonance was observed close to the resonance condition N=2.

In summary, we have demonstrated the first experimental detection of and theory for optically detected

magnetophonon resonance (ODMPR). The comparison between theory and experiment shows that the study of the frequency-dependent conductivity will generate much more specific and direct information about the nature of electron-phonon interactions in semiconductors than previous methods for studying MPR. ODMPR should be easily detectable in a variety of both bulk and 2D semiconducting materials.

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