Donor-Shifted Phonon-Assisted Magneto-Optical Resonances in *n*-InSb

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We have observed and described new optical transitions between magnetodonor states in InSb, assisted by optic-phonon emission. The phonon-assisted transitions provide a unique opportunity to investigate high excited states of the magnetodonor system (up to principal quantum number $n=13$), which simulates the hydrogen atom in gigantic magnetic fields. High-resolution data reveal the presence of additional excited magnetodonor states related to lower Landau subbands unobserved until the present.

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Magnetoimpurity states in semiconductors, since their discovery in the magnetic freezeout effect by Keyes and Sladek[1] and the pioneering theoretical descriptions of Yafet, Keyes, and Adams,[2] have been the subject of sustained experimental and theoretical interest. The effect of a magnetic field on shallow donor states is particularly important in narrow-gap materials with small effective masses m^* since, in the absence of the field, the donors are always ionized and cannot be observed. Magnetoimpurity investigations have been used to determine the static dielectric constant of a material[3] and its pressure dependence,[4] to identify the chemical nature of impurities,[5] and to study screening properties of the electron gas,[6] the metal-nonmetal transition,[7] etc. Recently, magnetoimpurity investigations proved to be useful in determining the positions of donors in modulation-doped two-dimensional GaAs-GaAlAs structures,[8,9] which is important for device applications.

The importance of the magnetodonor system goes beyond semiconductor physics, however, since a magnetodonor imitates the hydrogen atoms in giant magnetic fields. The problem of an electron subjected to simultaneous Coulomb and magnetic field interactions is characterized by the parameter $\gamma=(\hbar eB/m^*)/2\mathcal{R}^*$, where $\mathcal{R}^*=13.6(m^*/m_0)(1/\kappa^2)$ eV, and κ is the static dielectric contant. The value of γ is of the order of 10^{-5} for the hydrogen atom in vacuum. In narrow-gap semiconductors, however, γ can attain values of 100 or more for available magnetic field strengths. In our experiments we deal typically with $\gamma=70$, which corresponds to the hydrogen atom in a vacuum subjected to a magnetic field of $\simeq 10^7$ T. The above scaling allows one to transpose magnetodonor behavior to that of the hydrogen atom in gigantic magnetic fields.

The problem of atoms in extremely large magnetic fields has attracted a great deal of attention in recent years in both atomic physics and astrophysics.[10] The reason is that white dwarf stars can produce magnetic fields of 10^3 T and accreting neutron stars fields of the order of 10^6–10^8 T. As a consequence, the optical spectra of atoms in such magnetic fields have been observed in the spectra of these stars. For example, very good agreement between all observed spectral features and the computed wavelengths of stationary transitions of the hydrogen atoms in magnetic fields $(1.5–3.5)\times 10^4$ T has been found in the white dwarf GrW+70°8L47.[11] By virtue of scaling laws the energies of heavier atoms can also be gained from those of the hydrogen atom at scaled values of magnetic field.[12] This allows one to study, for example, FeXXVI in gigantic fields, which appears to be prevailing in the vicinity of x-ray pulsars.[13] In addition, the ionization thresholds of the hydrogen atom associated with higher Landau levels is of current interest in astrophysical investigations.[14]

The importance of the magneto-Coulomb system in astrophysics and atomic physics has motivated considerable theoretical work concerned with the behavior of the hydrogen atom at arbitrary magnetic fields.[15] The true atomic spectra are rich and detailed. However, they are limited to the quantum numbers $n=0$ and 1. Using the semiconductor as a medium we have been able to probe for the first time the excited states of the magneto-Coulomb system for n up to 13. This allows one to verify the existing calculations for high excited states.

The effect of optic phonons on optical transitions between magnetodonor states in semiconductors has been observed by Kaplan and Wallis[16] in the form of resonant polaron behavior in cyclotron resonance, and by McCombe and Kaplan in combined cyclotron-spin resonance.[17] In this Letter we report on a new kind of optical transition between magnetodonor (MD) states assisted by the emission of optic phonons.[18] It is the phonon assistance which allows us to observe very high excited states at the magneto-Coulomb system.

The experiments were performed on high-purity samples of n-InSb with a carrier concentration of 9×10^{13} cm^{-3} and an electron mobility of 7×10^5 cm^2/V sec at 77 K. The output of a grating-tunable cw CO_2 laser was mechanically chopped into 20-μsec-wide pulses and then focused onto a sample in the Faraday configuration. The magnetic field was varied between 0 and 12 T and the temperature between 1.8 and 30 K. Photoconductivity measurements were used to provide a sensitive means of detecting small changes in absorption.[19] Magnetic field modulation and lock-in amplifier techniques were used to record the second derivative of the photoconductive response versus magnetic field.

An example of the magneto-optical spectra is shown in Fig. 1. The observed peaks exhibit a doublet structure in which the higher-field peaks are due to free-electron transitions between Landau levels (LL) and the lower field peaks are related to the corresponding transitions between MD levels (see inset of Fig. 2). This assignment is confirmed by the temperature dependence of the structure (see inset of Fig. 1) because at higher temperatures thermal excitations depopulate the ground MD state, populate the lowest Landau state, and reverse the relative heights of the peaks in the doublets.

The quantum numbers of the final states involved in the transitions are also detailed in Fig. 1. These have been established by correlating the transition energies with theory, as shown in Fig. 2. The free-electron transitions $0^+ \rightarrow 2^+$, $0^+ \rightarrow 2^-$, and $0^+ \rightarrow 3^+$ (where the notation n^+ and n^- denotes LL states of quantum number n with spin up and spin down, respectively) and the associated MD transitions have been previously observed in InSb.[20] For our field orientation $\mathbf{B}\parallel(111)$ and the light polarization $\mathbf{E}\perp\mathbf{B}$ the $2\omega_c$ and the $2\omega_c+\omega_s$ transitions are allowed due to the inversion asymmetry of InSb.[21] The observed transition $3\omega_c$ is, however, not allowed. The same problem was encountered by Favrot, Aggarwal, and Lax[22], who observed the transition $2\omega_c$ for $\mathbf{B}\parallel(001)$, which is not allowed for this field orientation. The current interpretation is that such forbidden transitions become allowed due to the presence of impurities in the crystal.[18,23] Judging by the doublet structure, the corresponding donor-shifted transitions are allowed for the same reasons.

The transitions labeled by the primed numbers are optically induced electron excitations accompanied by the emission of an optic phonon. Because of the momentum transfer caused by the phonon emission, the selection rules are broken, allowing the observation of higher harmonics of the cyclotron resonance.[24] However, as seen from the data in Fig. 1, we observe for the first time the corresponding phonon-assisted MD excitations. This provides a unique opportunity to investigate high excited states of the magneto-Coulomb system. We have been able to observe and describe transitions to final MD states attached to the Landau subbands n up to $n=13$, as seen in Fig. 3.

As indicated in the inset of Fig. 2, the shift between the free-electron and MD transitions is due to the fact that, with increasing n, the binding energies of the corresponding MD states become smaller. For the quantitative description of the MD energies we have used the variational method. Since for all the states in question $\gamma \gg 10$, one may use the one-parameter trial functions of

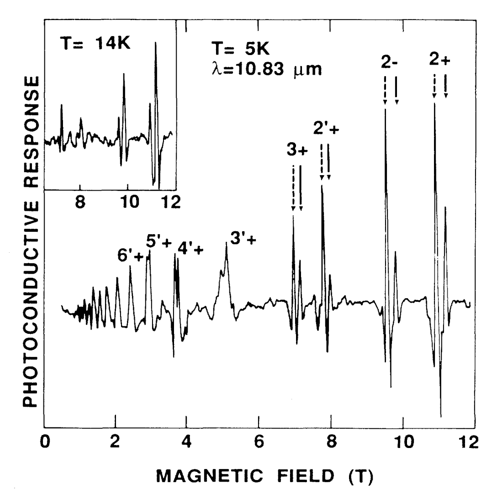

FIG. 1. Photoconductive response of n-InSb vs magnetic field obtained at 5 K using a CO_2-laser wavelength of 10.83 μm. The numbers indicate the final Landau states associated with the free-electron transitions. The primed numbers represent the phonon-assisted transitions. The solid arrows indicate the free-electron resonances and the dashed arrows indicate the MD resonances. Inset: The magneto-optical spectra at higher temperature.

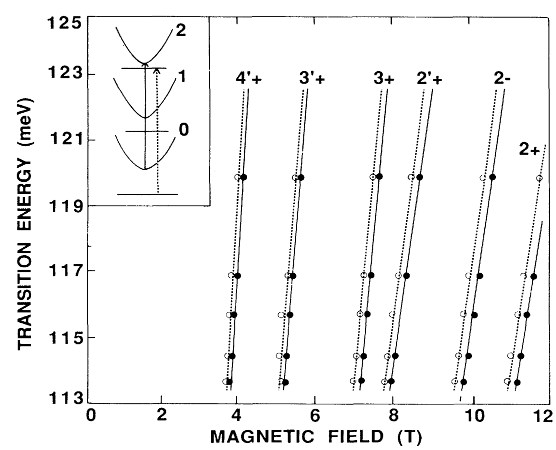

FIG. 2. Energies for the free-electron (solid circles) and magnetodonor (open circles) transitions vs magnetic field for the Landau quantum numbers 1–4. Primed numbers indicate phonon-assisted transitions. The solid lines were calculated using the Pidgeon-Brown model and the dashed lines were obtained by adding the calculated donor shifts. Inset: A diagram of the free-electron (solid arrow) and associated magnetodonor (dashed arrow) transitions labeled by 2^+. The Landau quantum numbers are also indicated in the inset.

Wallis and Bowlden,[25] assuming that the electron motion in a MD state is identical to the free-electron motion in a magnetic field. The trial averages for the kinetic $\langle K\rangle$ and potential energy $\langle V\rangle$ have been derived for an arbitrary MD state $(NM\beta)$, in which $N=0,1,2,\ldots$ is the principle quantum number, $M=\ldots,-1,0,1,\ldots$ quantizes the projection of the angular momentum on the magnetic field direction, and $\beta=0,1,2,\ldots$ quantizes the motion parallel to the magnetic field.[23] The Landau quantum number to which a given MD state "belongs" is $n=N+(M+m)/2$, where $m=|M|$. For the $(0M0)$ states, whose energies are very near the MD ground states related to $n=m$ Landau subbands, the trial averages are (in units of $\mathcal{R}^*$),[23]

$$\langle K\rangle=\gamma(2m+1)+\tfrac{1}{4}\gamma\lambda\,, \tag{1}$$

$$\langle V\rangle=-\frac{2}{m!}\left[\frac{\gamma\lambda}{2\pi}\right]^{1/2}\frac{d^m}{d\lambda^m}\left[\lambda^m\frac{B(\lambda)}{(1-\lambda)^{1/2}}\right], \tag{2}$$

where $B(\lambda)=\ln\{[1+(1-\lambda)^{1/2}]/[1-(1-\lambda)^{1/2}]\}$ and λ is the variational parameter. InSb is a narrow-gap semiconductor with a nonparabolic conduction band which affects the LL and MD energies. The band nonparabolicity is taken into account by writing the total energy in the form[23]

$$E^{\pm}=-\left[\frac{E_g}{2}-\langle V\rangle\right]+\left[\left[\frac{E_g}{2}-\langle V\rangle\right]^2+E_g(\langle K\rangle+\langle V\rangle\mp\alpha\gamma)\right]^{1/2}, \tag{3}$$

where E_g is the energy gap (in units of $\mathcal{R}^*$), $\alpha=\Delta/(2\Delta+3E_g)$, and Δ is the spin-orbit energy. The plus and minus signs correspond to the two spin orientations. Equation (3) has been derived from the three-level $k\cdot p$ model assuming $\Delta\gg E_g$, which is applicable to InSb. Thus we have used it to determine the MD binding energies $E_b(n)$, calculating the free-electron energies from the same model [i.e., putting in Eq. (3) $\langle V\rangle=0$ and $\langle K\rangle=(2n+1)\gamma$]. The difference $\delta^{\pm}(n)=E_b^+(0)-E_b^{\pm}(n)$ gives the upper bound to the energy shift between the LL transitions and the corresponding MD transitions. In Figs. 2 and 3 we compare the experimental transition-energy data with the theoretical predictions. The solid lines for the LL transitions have been calculated using a modified Pidgeon-Brown model[21] with the following band parameters:[26] $E_g=235.2$ meV, $E_p=23.2$ eV, $\Delta=0.803$ meV, $\gamma_1=3.25$, $\gamma_2=-0.2$, $\gamma_3=0.9$, $\kappa=-1.3$, $F=-0.2$, $q=0.0$, and $N_1=-0.55$. The phonon-assisted energies were obtained by adding the LO optic-phonon energy $\hbar\omega_0=24.4$ meV to the calculated LL energies (up to $n=13$). The dashed line for the MD transitions has been calculated by adding the corresponding $\delta^{\pm}(n)$ shifts to the LL transition energies, and using 0.65 meV for $\mathcal{R}^*$. The agreement between experiment and theory is very good (the agreement is seen more clearly in Fig. 4 for the $2'^{+}$ transition at one laser wavelength).

Our highest-resolution data revealed two additional resonances on the lower-field side, as shown in Fig. 4, for

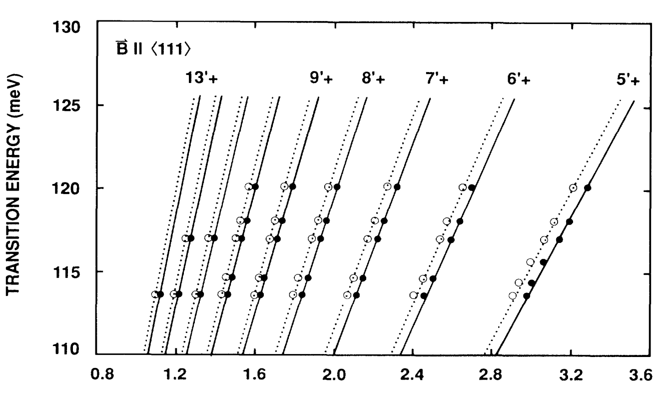

FIG. 3. Energies for the free-electron (solid circles) and magnetodonor (open circles) transitions vs magnetic field for the Landau quantum numbers 5–13. The solid lines were calculated using the Pidgeon-Brown model and the dashed lines were obtained by adding the calculated donor shifts.

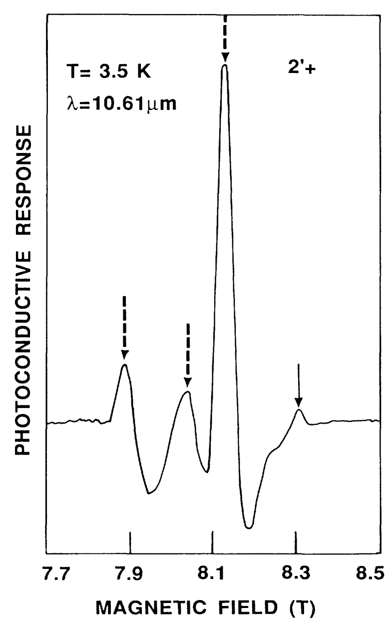

FIG. 4. High-resolution magneto-optical spectra for the phonon-assisted free-electron $0^+\rightarrow2^+$ transition (solid arrow), and the $(000^+)\rightarrow(200^+)$, $(000^+)\rightarrow(2\bar{3}0^+)$, $(000^+)\rightarrow(2\bar{1}1^+)$ magnetodonor transitions (dashed arrows). The magnetic field positions of the above transitions, indicated by the arrows, were calculated using the models discussed.

the $0^+ \rightarrow 2^+$ phonon-assisted excitations. They are present in the phonon-assisted as well as the ordinary MD transitions. This suggests that the resonances are due to additional selection rules caused by the band-structure complications rather than the phonons. The transitions shown in Fig. 4 have been identified as originating from the MD ground state (000^+) to excited MD states associated with the 2^+ Landau subband. The corresponding final states and calculated magnetic field positions are indicated by dashed arrows in Fig. 4. They obey the selection rules calculated by Wlasak.[27] The main transition ($000^+ \rightarrow 200^+$) (or the transition $000^+ \rightarrow 020^+$ having almost the same energy) is possible due to inversion asymmetry, while the weaker transitions ($000^+ \rightarrow 2\bar{1}0^+$) and ($000^+ \rightarrow 201^+$) are possible due to band warping. The weaker resonances and their dependence on light polarization and magnetic field orientation require further investigation.

In summary, we have observed and described new optical transitions between magnetodonor states in InSb. The phonon-assisted transitions allow the opportunity to study high excited states of an electron subjected to simultaneous Coulomb and magnetic field interactions, which is of direct interest not only to semiconductor physics, but also to atomic physics and astrophysics.

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