Magnetospectroscopy of Bound Phonons in High Purity GaAs

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Far infrared magnetophotoconductivity performed on high purity GaAs reveals the existence of fine structures in the resonant magnetopolaron regions. The fine structures are attributed to the presence of bound phonons due to multiphonon processes. We demonstrate that the magnetopolaron energy spectrum consists of bound phonon branches and magnetopolaron branches. Our results also indicate that different phonons are bound to a single impurity, and that the bound phonon in Si-doped GaAs is a quasilocalized mode. [S0031-9007(97)03828-3]

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The interaction of an electron with longitudinal optical phonons (polaron) has fundamental importance for electronic properties of polar solids. With an unmatched control of the concentration of electrically active impurities, intentionally doped semiconductors are excellent model systems that have been the focus of numerous studies concerned with polaron excitations [1] including bound phonon and resonant magnetopolaron. The bound phonon is a quasiparticle of longitudinal optical (LO) phonons bound to neutral impurities via electron-phonon interactions [2–4]. The bound phonons were observed in Raman spectra, infrared reflectivity, and photoluminescence of bulk semiconductors with impurity concentrations larger than 10^{17} cm^{-3} [5–7], where impurity interactions are not negligible. However, there is no experimental evidence to prove whether different LO phonons are bound to a single impurity or not. In other words, the physical origin of the bound phonons is still controversial considering whether the bound phonons are due to multiphonon processes or not [7,8]. Because of the complication of impurity interactions, high purity semiconductor samples are necessary to clarify this controversy. The bound phonons are quite different from localized phonon modes due to mass defects and changes in force constants. On the other hand, resonant magnetopolaron effects in both bulk systems [9–11] and quasi-two-dimensional systems [12–14] were extensively studied experimentally and theoretically. In these studies, the experimentally yet unproved assumption has been made that multiphonon processes can be neglected. Thus, whether multiphonon processes in the resonant polaron energy region are negligible or not remains an open question. It is well known that the transition energies between impurity levels can be tuned by magnetic field through the LO phonon energy ($\hbar\omega_{\mathrm{LO}}$). If multiphonon processes are considered, it is expected that new resonances, corresponding to the bound phonons, appear in the resonant magnetopolaron region, as predicted more than 20 years ago [2,3].

It is the purpose of this Letter to clarify the long-term controversy about the physical origin of bound phonons and further our understanding of the effects of multiphonon processes on the polaron energy spectrum.

We measured far infrared (FIR) magnetophotoconductivity spectra on high purity GaAs. The sample used in this work is a 5-μm thick epitaxial GaAs layer doped with Si donors at a nominal concentration of 5×10^{14} cm^{-3}. All data were taken at 4.2 K in the Faraday geometry under unpolarized FIR radiation with a Fourier transform spectrometer in conjunction with a superconducting magnet. The impurity concentration of approximately 10^{14} cm^{-3} is sufficiently small such that the interactions between impurities can be neglected at high magnetic field strength. With improved signal-to-noise ratio and resolution, we observed the resonant magnetopolaron processes between LO phonons and the door transitions from 1*s* ground state to donor metastable states. More importantly, we have for the first time clearly observed the fine structures, which are due to bound phonons, in the resonant magnetopolaron regions ($\hbar\omega \approx \hbar\omega_{\mathrm{LO}}$). Our results demonstrate that the magnetopolaron energy spectrum consists of both bound phonon branches and magnetopolaron branches, which have been studied separately for several decades.

Let us briefly present here the outlines of bound phonon microscopic theory. The Hamiltonian for the LO phonon interacting with an impurity electron is the sum of the Hamiltonians of the two noninteracting systems, and a Fröhlich-type coupling term [1–3]. The bound phonon wave function φ can be obtained by expanding it in terms of all uncoupled phonon-electron states [4],

$$\varphi = \sum_{\mathbf{k}} a_{\mathbf{k}}^{+}|0\rangle c_{\mathbf{k}} + \sum_{j>0} |j\rangle d_j + \frac{1}{2} \sum_{\substack{\mathbf{k}\mathbf{k}' \\ j>0}} a_{\mathbf{k}}^{+} a_{\mathbf{k}'}^{+} |j\rangle f_{\mathbf{k}\mathbf{k}'j} + \dots, \tag{1}$$

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where $c_{\mathbf{k}}$, d_j, and $f_{\mathbf{k}\mathbf{k}'j}$ are expansion coefficients, the first term is the free one-phonon state with the electron in the impurity ground state $|0\rangle$, the second term is the impurity electronic excited state $|j\rangle$ without phonon, and the third term is the state with two phonons in the impurity electronic excited state. One can show that there are as many bound phonon states close to LO phonon energy as there are impurity excited states [3,4]. It is to be remarked that no bound phonon state would appear at all if only the single-phonon processes were considered. In general, both bound phonons and resonant magnetopolarons are polaron states, quasiparticles of electron and phonon. The resonant magnetopolaron states are formed by the superposition of zero-phonon and single-phonon impurity states in the ratio of approximately 0.5:0.5, while bound phonon states are approximately α:1 (the Fröhlich coupling constant α is 0.068 in GaAs), i.e., only a small fraction of the electron wave function contributes to bound phonons [2].

The experimental transition energies for several impurity transitions have been plotted in Fig. 1 as a function of magnetic field. Zero field hydrogen atom quantum numbers *nlm* and high field quantum numbers (N, m, ν) are used to distinguish between donor bound and donor metastable states, respectively [15]. The metastable state (N, m, ν) is associated with the Landau level N of a free electron, modified by the Coulomb interaction between the donor ion and the electron; as such it is observed only in finite magnetic fields [15–18]. Here m is the magnetic quantum number, and ν is equal to the number of nodes of the eigenstate in the direction of the magnetic field. In the resonant regions, many branches are observed. The solid curves are theoretical calculations for impurity transitions without resonant electron-phonon interactions [16,17]. Band nonparabolicity and nonresonant polaron effect corrections were included in the calculations. All transitions below 280 cm^{-1} can be identified as $1s \rightarrow (210)$, $1s \rightarrow (212)$, $1s \rightarrow (310)$, $1s \rightarrow (410)$, and $1s \rightarrow (510)$ [15,17,18]. However, the transitions within the reststrahlen band were not observed. The six branches at the higher frequency side above the LO phonon frequency can be identified, respectively, as the upper branches of resonant magnetopolaron ($E_{(210)}$-$E_{1s} \approx \hbar\omega_{\mathrm{LO}}$, $E_{(212)}$-$E_{1s} \approx \hbar\omega_{\mathrm{LO}}$, $E_{(310)}$-$E_{1s} \approx \hbar\omega_{\mathrm{LO}}$, $E_{(410)}$-$E_{1s} \approx \hbar\omega_{\mathrm{LO}}$, $E_{(510)}$-$E_{1s} \approx \hbar\omega_{\mathrm{LO}}$, and $E_{(610)}$-$E_{1s} \approx \hbar\omega_{\mathrm{LO}}$) [16]. A typical antilevel crossing behavior of the above mentioned branches with magnetic field has been observed at frequencies around the LO phonon due to resonant electron-phonon interactions.

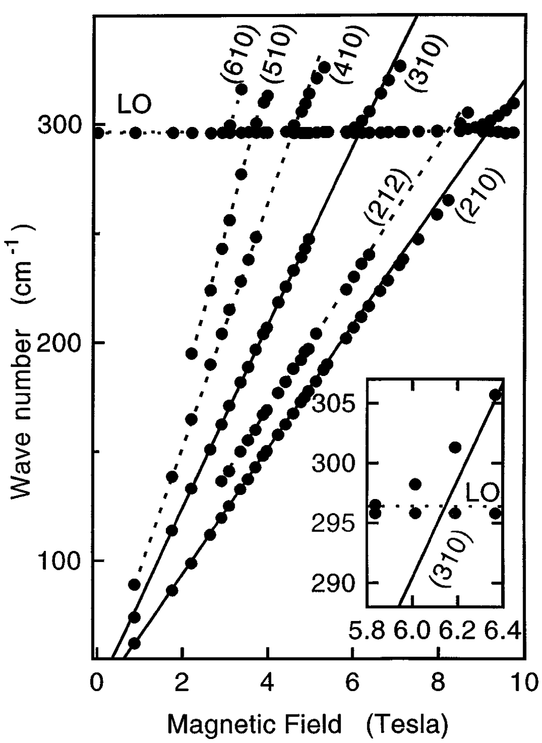

FIG. 1. Transition energy as a function of magnetic field for impurity transitions from $1s$ ground state to final states. The solid dots are experimental data; the solid curves are calculations for donor transitions without resonance electron-phonon interactions. The dashed curve is a guide to the eye. Inset: the enlarged resonant region involving the $1s \rightarrow (310)$ transition.

For an exact clarification of the polaron behavior in the resonant regions, the FIR photoconductivity spectra involving the $1s \rightarrow (310)$ transition at magnetic fields of around 6.0 T are taken as an example, as shown in Fig. 2. A very sharp peak, appearing at a frequency of approximately 296 cm^{-1} in the whole magnetic field range (not shown completely), begins to separate into two branches around 5.8 T. The branch on the high frequency side, which is the upper branch of the resonance ($E_{(310)}$-$E_{1s} \approx \hbar\omega_{\mathrm{LO}}$), decreases in relative intensity and moves up to higher frequency with increasing magnetic field. Around 7.5 T, this branch disappears. The second branch on the low frequency side with a pinning frequency of 295.8 ± 0.2 cm^{-1} near LO phonons (Γ) frequency appears at first as a weak shoulder at field of 5.84 T. At slightly higher fields, the relative intensity of the pinning peak increases rapidly. It seems that both branches exchange the oscillator strength with each other.

The inset of Fig. 1 shows the enlarged resonant region involving the $1s \rightarrow (310)$ transition. It is obviously observed that the pinning transitions at fields around 5.8 T appear between the upper branch of resonant magnetopolaron ($E_{(310)}$-$E_{1s} \approx \hbar\omega_{\mathrm{LO}}$) and the theoretical curve of the $1s \rightarrow (310)$ transition energy, where the resonant electron-phonon interactions are not included. This indicates that these pinning peaks, which are called fine structures [2] in the following, cannot be the lower branch of the resonant magnetopolaron involving the $1s \rightarrow (310)$ transition. Of course, the pinning peaks at somewhat higher magnetic fields (such as 6.63, 6.18, and 7.07 T) are mostly due to the lower branch of

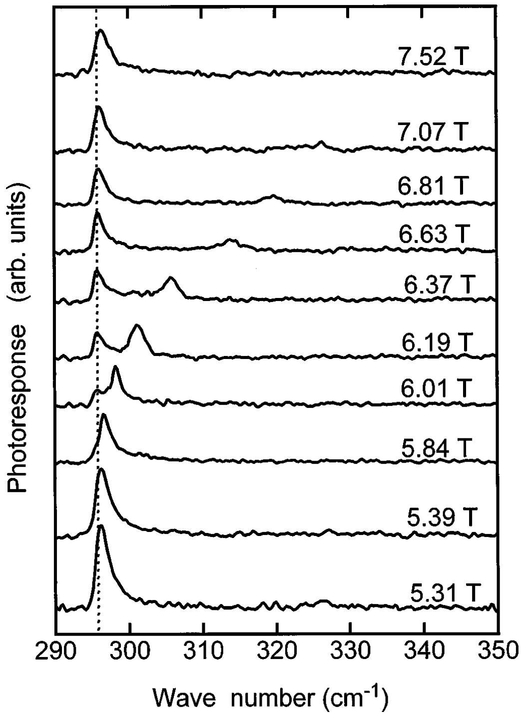

FIG. 2. Photoconductivity spectra in the resonant polaron region involving the $1s \rightarrow (310)$ transition at several magnetic fields. The dashed line is a guide to the eye.

the resonant magnetopolaron involving the $1s \rightarrow (310)$ transition, where the lower branch corresponds to the transition from 1*s* ground state to $1s$ + LO state [note that the $1s$ + LO state is a mixing state of $1s$ + LO state and nominal (310) state].

We believe that the origin of the fine structures in the resonant region are the transitions from 1*s* ground state to the bound phonon states. Other possible origins of the fine structures could be: (1) the transition from $|i, 0\text{-phonon}\rangle$ to $|i, 1\text{-phonon}\rangle$, where $|i\rangle$ could be the excited states, such as 2*s*, $2p_0$, and $2p_-$ state, etc.; (2) the electron-phonon interactions between donor states and local-phonon-modes due to mass defect (force constant); and (3) dielectric artifacts. They can be ruled out for the following reasons:

First, the electron concentration on these excited states is considerably smaller than that on the 1*s* ground state at 4.2 K, otherwise the transitions from the 2*s*, $2p_0$, and $2p_-$ states to high excited donor states should be observed in FIR photoconductivity spectra. However, this is not the case in our experiment. Second, both Si-Ga and Si-As local modes in Si-doped GaAs have a frequency of more than 370 cm^{-1} [19], which is higher than the bulk LO phonons (Γ) frequency of 296 cm^{-1}. Third, the dielectric artifacts, in which the transition line shape could be distorted near the reststrahlen band, are not responsible for the fine structures. This effect can play an important role in multilayer structures due to interference effects [12]. However, it was shown that photoconductivity measurements in bulk samples are insensitive to the dielectric effects [10].

Our assignment of the fine structures to the transitions from the 1*s* ground state to bound phonon states, the quasiparticles consisting of both electron and phonon via multiphonon processes, can further be confirmed by the following three facts: (i) the observation of the fine structures by photoconductivity spectroscopy, which indicates that the fine structures are associated with electron states, (ii) the pinning of the fine structures near LO phonons (Γ) energy against magnetic field, which strongly suggests that the fine structures are associated with phonon states, and (iii) the intensity evolution of two branches around LO phonons energy at fields of around 6 T, as shown in Fig. 2. The fine structures with considerable intensity appear in the resonant region. This cannot be explained by second-order perturbation theory neglecting multiphonon processes. On the contrary, the above-mentioned experimental features are in good agreement with the bound phonon theory including multiphonon processes.

Therefore, we conclude that the fine structures in the resonant magnetopolaron region involving the $1s \rightarrow$ (310) transition are due to the bound phonons. Similarly, the fine structures in the resonant region involving the $1s \rightarrow (210)$ transition are also due to the bound phonons. And our magnetospectroscopy of bound phonons demonstrates that the magnetopolaron energy spectrum consists of both bound phonon branches and magnetopolaron branches, where magnetopolaron branches [9] include nonresonant magnetopolaron states in the nonresonant polaron region as well as resonant magnetopolaron states in the resonant polaron region.

As stated in the polaron theory, the sharp difference between the resonant magnetopolaron states and the bound phonon states vanishes at the resonant situation ($\hbar\omega \approx \hbar\omega_{\mathrm{LO}}$) [2]. In the case of resonance, e.g., ($E_{(310)}$-$E_{1s} \approx \hbar\omega_{\mathrm{LO}}$), the upper branch near the LO phonon energy is also a bound phonon to some extent. However, it is not in contradiction to the above mentioned identification of both the fine structures and the upper branch of the resonance involving the (310) state, because the electron part of the upper branch is mostly due to the (310) state, while the fine structures are mostly due to phonon states. As is well known, the donor wave functions are strongly compressed under high magnetic field. The interactions between donors in GaAs ($n_{300k} = 5 \times 10^{14}\ \text{cm}^{-3}$) under high magnetic field are negligible [11]. Thus, the appearance of two bound phonon states in the resonant region at the same time demonstrates that bound phonon multimodes observed in Raman and photoluminescence spectra are associated with different LO phonons bound to a single donor [8] other than a LO phonon bound to different donor pairs [7]. Of course, it can be expected that impurity interactions can have an effect on the bound phonon states.

Finally, note that the bound phonon states corresponding to the fine structure in n-GaAs ($R^* \ll \hbar\omega_{\mathrm{LO}}$, where R^* is Rydberg energy) are resonant with Landau levels. The wave function mixing between bound phonon states and Landau levels can occur to a small extent, since bound phonon states contain nominal donor bound and metastable states, and nonradiative autoionizing transitions may occur between metastable states and the Landau continuum with smaller Landau quantum number [15]. Therefore, the bound phonon in Si-doped GaAs is a kind of quasilocalized mode.

In conclusion, we have observed for the first time the fine structures in resonant magnetopolaron regions associated with the (210) and (310) zero-phonon donor metastable states by high-resolution FIR magnetophotoconductivity. The fine structures are due to bound phonons, indicating that multiphonon processes play an important role in the vicinity of the resonant polaron energy. The magnetospectroscopy of bound phonons demonstrates that the magnetopolaron energy spectrum consists of both bound phonon branches and magnetopolaron branches. The bound phonon is indeed a quasiparticle consisting of both electron and phonon. Different phonons are bound to a single impurity at low temperature. The bound phonon in Si-doped GaAs at low temperature is a quasilocalized mode.

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