Long spin relaxation time of holes in InGaAs/GaAs quantum wells probed by cyclotron resonance spectroscopy

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In this paper, we report on a long, ms range, hole spin relaxation time in InGaAs/GaAs quantum wells probed by cyclotron resonance spectroscopy in pulsed magnetic fields. In our experiments, we found strong hysteresis in the spectral weights of cyclotron resonance absorption lines when rapidly changing magnetic field is used for the experiment. The hysteresis vanishes when a much slower changing magnetic field is used. We attribute this behavior to a long energy relaxation time between the two lowest spin-split hole Landau levels, i.e., a long hole spin relaxation time. We also present transition frequencies calculated using a 4×4 Luttinger Hamiltonian, which confirm our findings.

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I. INTRODUCTION

Strained InGaAs/GaAs multilayer structures are an interesting research subject both fundamentally and in terms of applications. They can be used as a basis for microwave transistors, solar cells, and ir lasers. In addition, during the past few years, the rapidly growing area of spintronics has stimulated interest in these structures due to the demonstration of efficient spin injection[1,2] and circular-polarized electroluminescence in InGaAs/GaAs Schottky diodes, as well as the discovery of the anomalous Hall effect in Mn δ-doped InGaAs/GaAs quantum wells.[3,4] For spintronic applications, a particularly crucial parameter is the carrier spin relaxation time, which preferably should be as long as possible. In this paper, we show the existence of an extra long, ms range, spin relaxation time of holes in strained InGaAs/GaAs quantum wells, using high-field cyclotron resonance spectroscopy.

Cyclotron-resonance (CR) spectroscopy is one of the direct methods to probe the band structure of semiconductors.[5] The cyclotron resonance of holes in InGaAs/GaAs quantum-well (QW) heterostructures was studied in a number of works (see, e.g., Refs. 6–9). In Ref. 6, a set of $In_{0.2}Ga_{0.8}As$/GaAs QWs was used to show that the cyclotron mass of holes at the Fermi level increases from $0.123m_0$ to $0.191m_0$ as two-dimensional hole concentration changes from 0.54×10^{11} cm^{-2} to 8.5×10^{11} cm^{-2} (m_0 is the free-electron mass). In Refs. 7 and 8, a single CR line was observed under quantizing magnetic fields of 17 T corresponding to Landau-level (LL) filling factors $\geqslant 2$, although the width of this line was smaller than the estimated splitting due to the difference in the energies of transitions from the two lower LLs. This effect was viewed as manifestation of hole-hole (hh) interaction. Splitting of the CR line was found in our work[9] for an $In_{0.14}Ga_{0.86}As$/GaAs heterostructure in high pulsed magnetic fields above 20 T. In that work, we calculated the Landau levels of holes in a strained $In_{0.14}Ga_{0.86}As$ QW and compared the estimated transition energies from the two lowest Landau levels $0s_1$ and $3a_1$ (see below) with the position of the lines observed in magnetoabsorption spectra. An excellent agreement was achieved between the calculated transition energies and the experimental data. At the same time, the spectral-weight ratio of the two CR lines observed in Ref. 9 was found to be opposite to the calculated result. The Landau-level filling factor in resonant magnetic fields was $\nu \leqslant 1$, so the upper of the two lowest Landau levels, $3a_1$, should be populated only through thermal activation of holes from the lower level $0s_1$. Hence, the CR line corresponding to the transition from the level $0s_1$ should have a larger spectral weight than the line for the transition from the level $3a_1$, whereas the measured spectra exhibited an inverted relation. We attributed this effect to a renormalization of the Luttinger parameter κ due to hh interaction (similar to the exchange enhancement of the g factor for electrons[10]). Finally, in Ref. 11 we reported a strong hysteresis in the spectral weights of the CR spectra measured on the rising and the falling slopes of the magnetic field pulses. Previously, a similar type of hysteresis in CR spectra was reported by Arimoto *et al.* for electrons in InAs/AlSb quantum wells[12] when fast, microsecond range, magnetic field pulses were used. These authors have used this effect to study the electron spin relaxation time. They have concluded that the hysteresis is caused by the long electron spin relaxation time, found to be comparable to the magnetic field rise time, which was of the order of several microseconds. In the present work, we extend our previous CR studies of holes in InGaAs/GaAs heterostructures to magnetic fields of different rise times ranging from 12 to 34 ms. We found the hysteresis to exist when the magnetic field rise time is of the order of 12 ms, while it vanishes when the magnetic field rise time increases up to 34 ms. Similarly to the previous work of Arimoto *et al.*, we attribute this effect to the long energy relaxation time of holes between the lowest spin-split Landau levels. We argue that in our case the hole spin relaxation time is comparable with a magnetic field pulse length, which is of the order of 10 ms. We also present calculations of the spin relaxation rates as a function of magnetic field for different Luttinger parameters κ.

II. EXPERIMENT

We measured the CR spectra in an InGaAs/GaAs QW sample no. 5805 (one of the set studied in Ref. 11) under pulsed magnetic fields up to 55 T. The sample was pseudomorphically grown by metalorganic chemical vapor deposition on a GaAs(100) substrate. It consisted of 50 70-Å-wide $In_{0.14}Ga_{0.86}As$ quantum wells separated by 500-Å-wide GaAs barriers. Thin GaAsP layers were built in in the middle of the GaAs barrier layers to compensate the total strain of the structure. We note that these layers do not affect the energy spectrum of the holes in strained InGaAs QWs. The barriers were δ-doped with carbon separated by a 150 Å spacer from each InGaAs/GaAs interface.

The measurements were carried out at the Dresden High Magnetic Field Laboratory (HLD).[13] Two types of coils were used for experiments. The first one delivers magnetic fields up to 51 T with a total pulse duration of the order of 75 ms, while the second type can produce up to 70 T with a total pulse duration of about 150 ms.[14,15] As an excitation source, we used a quantum cascade laser emitting at 75 μm, which was installed with the same cryostat as the sample but far enough from the center of the magnetic field to keep the residual magnetic fields acting on the laser below 5 T. The quantum cascade lasers were proven to be powerful and easy to use radiation sources that were insensitive to magnetic fields below 10 T (depending on the design; see, for example, Refs. 16 and 17 and references therein). The emitted light was guided to the sample by a stainless steel pipe. The light transmitted through the sample was then focused on a Ge:Ga photodetector installed below the sample. The magnetic field was monitored with a Hall sensor.[18] More details of the experimental setup can be found in Ref. 19.

III. RESULTS

Time profiles of the magnetic field pulses for the two solenoids used are shown in Fig. 1. Note that in our earlier work,[9] measurements of the CR spectra were taken only on the falling side of the magnetic-field pulse. In this work, as well as in Ref. 11, the CR spectra were measured at both the falling and the rising sides of the pulse. The data are presented in Fig. 2. The CR absorption exhibits pronounced splitting with two components marked on Fig. 2 by vertical arrows 1 and 2. These transitions were identified in our previous work, where we also performed a detailed calculation of the transition energies as a function of magnetic field and found excellent agreement with experiment.[9] We found that the absorption line 1 corresponds to the transition from the lowest Landau level $0s_1 \rightarrow 1s_1$, and line 2 corresponds to the transition $3a_1 \rightarrow 4a_1$ (see details of the numbering later in the text or in Ref. 9). The inset of Fig. 2(a) shows the energies of four lowest hole Landau levels; vertical arrows mark the corresponding transitions. One can see a pronounced hysteresis of the spectral weights of the two CR lines when a short-pulse magnetic field is used [Fig. 2(a)]: on the rising side of the magnetic-field pulse, the absorption line 1 at about 24 T (corresponding to the transition $0s_1 \rightarrow 1s_1$) is much more pronounced than the absorption line 2 (corresponding to the transition $3a_1 \rightarrow 4a_1$) at around 35 T, whereas on the falling side of the field pulse the absorption line 1 is significantly reduced and weaker than the absorption line 2. On the other hand, the CR spectra measured at the rising and the falling sides of the longer field pulse almost coincide [Fig. 2(b)].

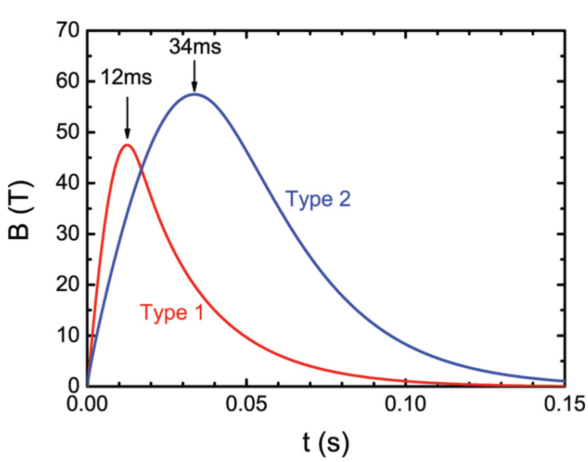

FIG. 1. (Color online) Magnetic-field profiles for the two coils used: Type 1 has a 12 ms rise time and ~75 ms total pulse duration, and type 2 has a 34 ms rise time and 150 ms total pulse duration.[14]

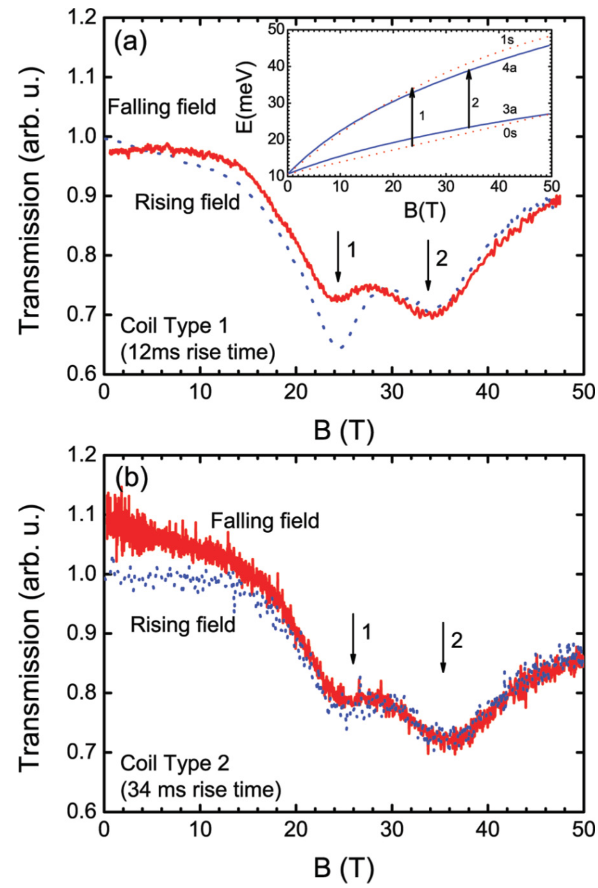

FIG. 2. (Color online) Cyclotron-resonance absorption spectra, taken at $T = 4.2$ K, with excitation wavelength $\lambda = 75$ μm and using (a) fast coil type 1 and (b) slow coil type 2. Dotted curves indicate data taken for rising magnetic field, solid lines for decreasing field. The inset shows the calculated relative position of the two lowest Landau levels; arrows indicate transitions corresponding to the CR lines 1 and 2.

As mentioned above, the LL filling factor at resonant fields is less than unity, but the splitting estimated for the two lowest Landau levels[9] is comparable with k_BT (k_B is the Boltzmann constant, $T = 4.2$ K), and CR transitions from both LLs should participate in the magnetoabsorption spectra. Under thermal equilibrium, the lower-state occupation should be the largest and, hence, the line corresponding to the transitions from this state should have the larger spectral weight. It turns out that the more pronounced line in the CR spectra taken at the falling edge of the "short" magnetic-field pulse [Fig. 2(a)] and at both sides of the "long" pulse [Fig. 2(b)] is line 2 (transition $3a_1 \rightarrow 4a_1$). This means that the initial LL for this transition, $3a_1$, lies below the state $0s_1$ (the initial LL for the transition $0s_1 \rightarrow 1s_1$, line 1), which is in contradiction to the results of the single-particle calculations performed in Ref. 9 (see also the inset to Fig. 2). These findings confirm the suggestion put forward in Ref. 9 that the relative position of the LLs $0s_1$ and $3a_1$ is inverted due to hh exchange interaction. Within this concept, the fact that the spectral weight of line 1 at the rising side of the "short" pulse (12 ms rise time) is larger than the spectral weight of line 2 can be explained as follows: when the magnetic field rises, the carriers are unable, due to a lack of time, to transfer from the second Landau level ($0s_1$, the initial state for the transition 1) to the lower hole state, $3a_1$, but a "long-pulse" field (34 ms rise time) allows for the carrier relaxation to the lower Landau level.

IV. CALCULATIONS AND DISCUSSION

To interpret the measured CR spectra, we have calculated the hole-state energies in the QWs of our heterostructure using the effective-mass approximation. The Hamiltonian for the envelope functions includes the Luttinger Hamiltonian, essentially a 4×4 matrix, the deformation-potential-related diagonal component, and the rectangular QW confinement potential. Luttinger parameters γ_1, γ_2, γ_3, and κ in a solid solution were taken via linear interpolation between their GaAs and InAs values, depending on the In content, as was suggested in Refs. 20 and 21. Note that the Luttinger parameters are related by

$$\kappa = \tfrac{1}{3}(\gamma_1 - 2\gamma_2 - 3\gamma_3 + 2) - \tfrac{1}{2}q. \quad (1)$$

Parameter q was neglected, as usual, because of its smallness (about 0.04 for both GaAs and InAs). We also neglected (as, e.g., in Ref. 9) the nondiagonal terms in the Luttinger Hamiltonian, responsible for the anisotropy of the hole dispersion in the QW plane. In such a model, a good quantum number is the projection of the total angular momentum on the heterostructure growth axis (coinciding with the magnetic field direction) J. The calculated Landau levels were characterized by three parameters ns_i: Landau-level number n, the index defining the symmetry of the state (s is used for symmetric states, a for the antisymmetric ones), and index i to number the level due to size quantization, to which the particular Landau level pertains; see more details on the explanation of numbering in our work.[9] The lowest levels $0s_1$ and $3a_1$ of the two heavy-hole Landau-level ladders differ in the spin direction: $J = \pm 3/2$. The hole transitions between such levels (at the equilibrium distribution setting with a rising magnetic field and the LL filling factor decreasing) can be regarded as transitions with spin flip, i.e., as spin relaxation.

The wave functions of these two states have the form

$$\Psi_{3a_1} = \begin{pmatrix} C_4(z) f_{0,0}(\rho,\varphi) \\ C_3(z) f_{1,1}(\rho,\varphi) \\ C_2(z) f_{2,2}(\rho,\varphi) \\ C_1(z) f_{3,3}(\rho,\varphi) \end{pmatrix}, \quad (2)$$

$$\Psi_{0s_1} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ C_1(z) f_{0,0}(\rho,\varphi) \end{pmatrix}, \quad (3)$$

where

$$f_{n,m}(\rho,\varphi) = \frac{1}{\lambda}\sqrt{\frac{r!}{(r+|m|)!}}\frac{e^{im\varphi}}{\sqrt{2\pi}} x^{\frac{|m|}{2}} e^{\frac{-x}{2}} L_r^{|m|}(x), \quad (4)$$

$x = \frac{\rho^2}{2\lambda^2}$, $r = n - \frac{|m|+m}{2}$, $L_r^{|m|}(x)$ are Laguerre polynomials, n is the Landau-level number that takes integer non-negative values, and $m = J + 3/2$. Note that the "main" component of wave function (2) is the first component (over 85% by norm). The obtained wave functions of the two lowest hole states were used to calculate the probability of transitions between these levels due to acoustic phonon scattering. The probability of carrier transition between the Landau levels in a magnetic field was sought via Fermi's golden rule:

$$\tau^{-1} = \sum_{e,q} \frac{2\pi}{\hbar} \left|\langle \Psi_{0s_1} | H_d | \Psi_{3a_1} \rangle\right|^2 \delta\left(E_{3a_1} - E_{0s_1} - \hbar\omega_\phi\right). \quad (5)$$

Here, the summing is performed over all polarization directions ($\overrightarrow{e}$) and wave vectors ($\overrightarrow{q}$). The perturbation operator H_d is the "deformation potential" resulting from the correction to the periodic lattice potential caused by an acoustic phonon. This Hamiltonian for the holes near the valence-band edge in a GaAs and InGaAs solid solution is a 4×4 matrix and, according to Ref. 22, has the form

$$H_d = \begin{pmatrix} f & h & j & 0 \\ h^* & g & 0 & j \\ j^* & 0 & g & -h \\ 0 & j^* & -h^* & f \end{pmatrix}, \quad (6)$$

where

$$f = (a-b)\varepsilon_{zz} + \left(a + \frac{b}{2}\right)(\varepsilon_{xx} + \varepsilon_{yy}), \quad (7)$$

$$g = (a-b)\varepsilon_{zz} + \left(a - \frac{b}{2}\right)(\varepsilon_{xx} + \varepsilon_{yy}), \quad (8)$$

$$h = -i\sqrt{3}d(\varepsilon_{xz} - i\varepsilon_{yz}), \quad (9)$$

$$j = \frac{\sqrt{3}}{2}(b+d)(\varepsilon_{xx} + \varepsilon_{yy} - 2i\varepsilon_{xy}). \quad (10)$$

Here $\varepsilon_{i,j}$ are the deformation tensor components, $i,j = x,y,z$. The deformation parameters a, b, and d for InGaAs were taken via linear interpolation between the GaAs and InAs values of these parameters used in Ref. 21. It is seen that the secondary diagonal elements of the deformation potential are zero; such perturbation mixes the wave function (3) with the small components of wave function (2) only, i.e., the acoustic-phonon-induced transition proved to be quasiforbidden. So, the transition times between the levels $3a_1$ and $0s_1$ should be large. Note also that, as in Ref. 9, the calculated energies $0s_1 \rightarrow 1s_1$ and $3a_1 \rightarrow 4a_1$ are in excellent agreement with the experimental absorption positions 1 and 2, respectively. According to the single-particle calculations, $0s_1$ is the lowest state, which is in contradiction with the observed spectral-weight ratio. We found the relative position of states $0s_1$ and $3a_1$ to be sensitive to the value of the Luttinger parameter κ that is analogous to the electron g factor. For the In content $x = 0.14$ in our sample, parameter κ as calculated by use of Eq. (1) is equal to 2.01. As known, many-body effects may cause the g factor to change considerably (see, e.g., Refs. 9,23, and 24). A similar situation is likely to take place for holes, which gives us sufficient grounds for using κ as the fitting parameter in "single-particle" modeling of the LL spectrum and transition frequencies.

Figure 3 illustrates the magnetic-field dependences of the transition rates between two lowest hole levels due to transverse acoustic phonon scattering, calculated by use of Eq. (5) for κ: $\kappa = 2.01$ and 1.2. It should be noted that at low fields, the rate tends to zero because (i) at $B = 0$ the transition is forbidden [since in Eq. (2) $C_{2-4}(z) \rightarrow 0$] and (ii) the spacing between the states $0s_1$ and $3a_1$ tends to zero, which reduces the number of phonons available for such a transition. For both κ values, the transition frequencies in magnetic fields are of the same order of magnitude. We are interested in the case in which $\kappa = 1.2$, since it reproduces correct relative positions of the levels $0s_1$ and $3a_1$ and ensures correct spectral weight distribution between the CR lines 1 and 2. As seen from Fig. 3(b), in this case the spin relaxation time (transition between the two lowest states) at 24 T (resonant field for the absorption line 1) is about 10 ms, which is comparable with the rise time of the "short-pulse" magnet but is several times smaller than the rise time of the "long-pulse" field. This explains the variation of the CR line intensities measured on the rising and falling sides of the magnetic field pulses, and it indicates a long charge-carrier relaxation time between the two lowest Landau levels. Please note that in this paper we present only a rough estimation of the hole spin lifetime, giving its upper limit. The accurate and systematic determination of the hole spin lifetime is beyond the scope of the current paper, and would probably require different experimental methods, such as, for example, the pump-probe technique.

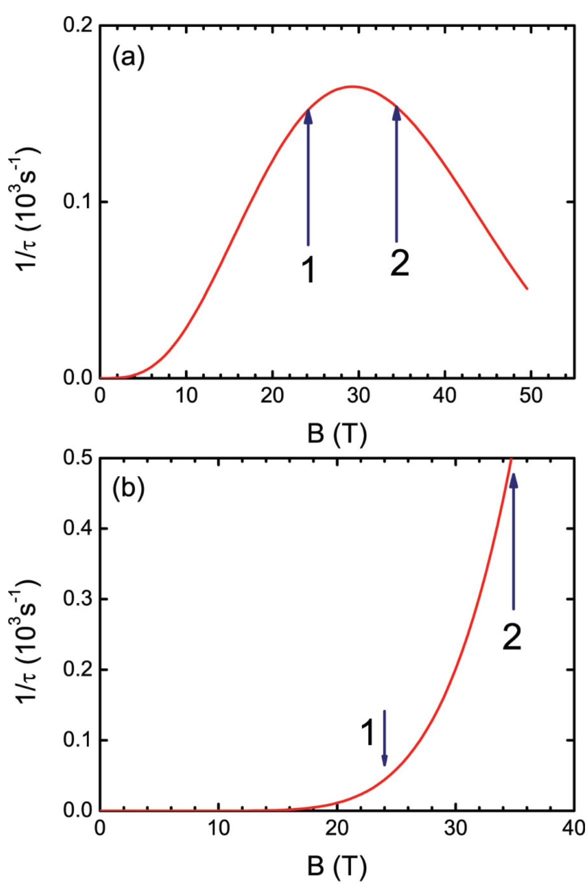

FIG. 3. (Color online) Transition rate $1/\tau$ between the states $0s_1$ and $3a_1$ assisted by longitudinal acoustic phonons as a function of magnetic field for the Luttinger parameter (a) $\kappa = 2.01$ and (b) $\kappa = 1.2$. Arrows indicate the magnetic field values where the transitions 1 and 2 occur.

V. CONCLUSION

In this work, we have studied cyclotron resonance absorption in an InGaAs/GaAs quantum-well sample using pulsed magnetic fields with different pulse durations. We have confirmed our suggestion made in Ref. 9 that the relative position of the two lowest Landau levels $0s_1$ and 3_1 is inverted due to exchange hh interaction. We found a strong hysteresis in the spectral weight of the CR lines when a fast magnetic field pulse with 12 ms rise time was used for the experiment. We also found that the hysteresis vanishes when longer magnetic field pulses were used. We attribute this effect to a long, millisecond range, relaxation time between the two lowest spin-split Landau levels, i.e., to a long spin-relaxation time. We confirmed our findings with calculations using a 4×4 Luttinger Hamiltonian. We also hope that our results will attract the attention of the spintronic community to the hole systems and initiate studies of the hole spin lifetime by direct methods, for example by ultrafast pump-probe techniques. This can be particularly interesting in strained structures, where the reduction of the heavy-hole effective mass due to strain leads to considerable enhancement of the carrier mobility, making the electrical transport properties of these materials comparable to those of usual electron structures.

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