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Intra-acceptor hole relaxation in Be δ-doped GaAs/AlAs multiple quantum wells*

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This paper studies the dynamics of intra-acceptor hole relaxation in Be δ-doped GaAs/AlAs multiple quantum wells (MQW) with doping at the centre by time-resolved pump-probe spectroscopy using a picosecond free electron laser for infrared experiments. Low temperature far-infrared absorption measurements clearly show three principal absorption lines due to transitions of the Be acceptor from the ground state to the first three odd-parity excited states respectively. The pump-probe experiments are performed at different temperatures and different pump pulse wavelengths. The hole relaxation time from 2p excited state to 1s ground state in MQW is found to be much shorter than that in bulk GaAs, and shown to be independent of temperature but strongly dependent on wavelength. The zone-folded acoustic phonon emission and slower decay of the wavefunctions of impurity states are suggested to account for the reduction of the 2p excited state lifetime in MQW. The wavelength dependence of the 2p lifetime is attributed to the diffusion of the Be atom δ-layer in quantum wells.

Keywords: carrier relaxation, multiple quantum well, intra-acceptor dynamics, pump-probe
PACC: 7215L, 7210D, 7320D, 7847

1. Introduction

Terahertz (THz) science and technology have attracted much attention on account of the potential applications of THz radiation in many domains recently.[1−4] The impurity energy spectrum in a semiconductor quantum well (QW) just falls into the THz range, and the energy levels can be tuned in a controlled way by changing the design of the QW structure and the doping mode. Semiconductor QWs are of great interest for far-infrared detectors and solid state THz laser emitters etc.[4−7] For optoelectronic applications, the GaAs/AlGaAs QWs are the most widely used system and beryllium (Be) is a common acceptor impurity in it. The impurity states and optical properties of the GaAs/AlGaAs QWs have been extensively studied both theoretically[4,8−10] and experimentally.[11−14] However, the dynamic properties have been investigated less, though they are very important for device applications,[15,16] such as the population inverse of the laser emitter. In this paper we investigate the intra-level relaxation dynamics of holes bound to Be acceptors in δ-doped GaAs/AlAs mutiple QW (MQW) by means of time-resolved differential pump-probe measurements using a free electron laser for infrared experiments (FELIX).

The rest of this paper is organized as follows. Section 2 introduces the sample preparation and experimental details. In Section 3, the experimental results are presented and discussed. Finally, the conclusions are summarized in Section 4.

2. Sample and experimental details

The samples studied were a GaAs/AlAs MQW and a bulk GaAs:Be, grown by molecular-beam epitaxy on semi-insulating (100) GaAs substrates. The MQW sample was a 200 period QW structure, each comprising a 10 nm wide GaAs well layer and a 5 nm wide AlAs barrier layer, while the centre of every GaAs well layer was δ-doped with Be acceptor atoms to a superficial density of 5×10^{10} atoms/cm^2. The GaAs:Be bulk sample was a single 1-μm-thick epilayer of GaAs with Be doped uniformly at 1×10^{12} atoms/cm^2, as a reference sample. By using the technique of stoichiometric low-

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temperature growth, the QW structure was grown at 550 °C and without interruptions at the QW interfaces, which ensured negligible diffusion of the Be δ-layers. For Fourier-transform infrared absorption and pump-probe experimental measurements, each sample was thinned, polished, and wedged to approximately a 5° angle to restrain optical interference between the front and back faces.

Far-infrared absorption experiments were performed by using a Fourier-transform spectrometer. The samples and Si bolometer detector were placed into the cryostat with liquid helium. The absorption spectra were obtained in a wave number range from 40 to 300 cm^{-1} at 4.2 K.

The time-resolved pump-probe measurements were conducted by using the picosecond FELIX at Rijhuizen.[17] The experimental setup is shown in Fig.1. A free electron laser (FEL) is used as a source of intense far-infrared picosecond pulses. This technique uses two probe pulses, a probe beam and a reference beam; both are split off from the original pump pulse by a beam splitter, so precise time delays can be produced by adjusting the optical path length. The reference beam is delayed by 20 ns. The three beams are focused on the sample at the same position. The transmission signals of the probe and reference beams through the sample are detected by a sensitive Ge:Ga detector. The Ge:Ga detector is biased to 25 MHz, synchronized with FELIX. This results in signals with opposite sign for the probe and the reference beams. The resulting balance between transmissions of two probe pulses is taken as the pump-probe signal. This differential pump-probe technique can remove signal jitter efficiently, and resolve absorption changes due to the pump beam of less than 0.5%.[18] The sample was mounted in a liquid-helium flow cryostat with a polypropylene window. The FEL wavelength was set to the position of the strongest absorption peak (the D line).

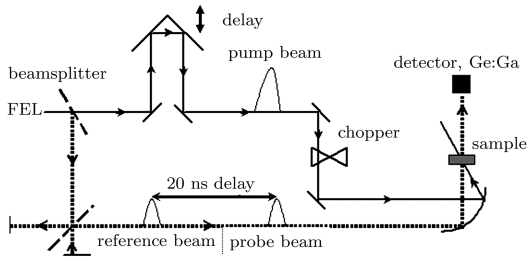


Fig.1. The FEL differential pump-probe experiment.

The differential pump-probe signal was measured directly as a function of optical delay between pump and probe pulses.

3. Results and discussions

3.1. Far-infrared absorption spectra

Figure 2 shows the far-infrared absorption spectra of the samples in the region 120–240 cm^{-1}. For the Be δ-doped MQW sample with well width 10 nm, two principal absorption lines, labelled G and D, are clearly observed at 173 and 222 cm^{-1}. For the GaAs:Be bulk sample, the G, D, and C absorption lines are at 135, 167, and 183 cm^{-1}, respectively.

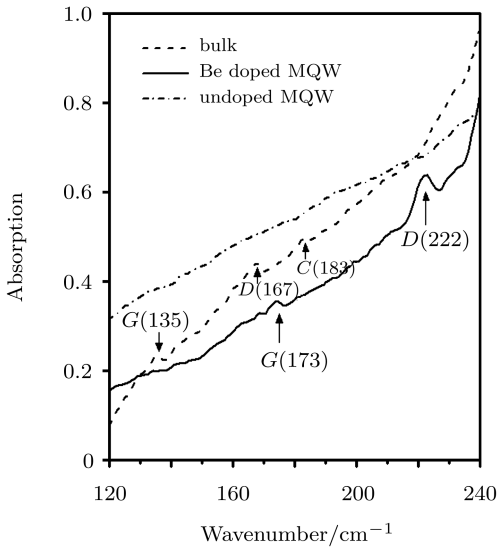


Fig.2. Absorption spectra at 4.2 K of the GaAs:Be bulk and GaAs/AlAs MQW samples with well width 10 nm.

Infrared absorption selection rules only allow the observation of odd parity transitions. For the bulk GaAs, the positions of G, D, and C lines are in good agreement with what have been reported in Refs.[12] and [19], and the calculation in Ref.[20]. These lines are due to acceptor state transitions from the ground state, $1s_{3/2}\Gamma_8$, to the first three excited odd-parity states, $2p_{3/2}\Gamma_8$, $2p_{5/2}\Gamma_8$, and $2p_{5/2}\Gamma_7$, respectively.

In order to identify the origins of the absorption lines in the Be δ-doped MQW sample absorption spectrum, the far-infrared absorption spectrum of an undoped GaAs/AlAs MQW sample was measured. This undoped MQW sample had the same structure as the Be δ-doped MQW sample, but was not doped in both wells and barriers. It contained 100 periods of QW structure with a 10 nm wide GaAs well layer and a 5 nm wide AlAs barrier layer. The absorption spectrum of the undoped MQW sample is indicated by

the dot-dash curve in Fig.2. No absorption peak was found in the region 120–240 cm^{-1}. This result shows that the absorption lines of the Be δ-doped MQW sample originated from the acceptor dopants.

The QW potential reduces the crystal point group symmetry from T_d to D_{2d}.[14] At the Γ point, the valence band states of $J = 3/2$ are split into the heavy hole band of $M_j = \pm 3/2$ and the light hole band of $M_j = \pm 1/2$. Additionally, the $1s_{3/2}\Gamma_8$ acceptor ground state and the $2p_{3/2}\Gamma_8$, $2p_{5/2}\Gamma_8$ excited states are split accordingly into $1s_{3/2}(\Gamma_6 + \Gamma_7)$ and $2p_{3/2}(\Gamma_6 + \Gamma_7)$, $2p_{5/2}(\Gamma_6 + \Gamma_7)$, respectively.[12,14]

Masselink *et al* calculated the Be acceptor states and the complicated valence-band structure in GaAs/Al_xGa_{1-x}As QWs using the variational method in Ref.[8]. Their calculations show that the energy difference between the heavy-hole subband and light-hole subband in 10 nm well is 16 meV, less than 27 meV, the energy difference between the acceptor ground state and the first excited state. The transition energy from the first subband to the second subband calculated by Chang and James is less than 20 meV in 10 nm p-type GaAs/AlAs QWs.[21] The observed two absorption lines of Be δ-doped MQW are 21.45 meV and 27.53 meV, so they are not due to intersubband transitions. The observed absorption energies are consistent with the energies of the $2p_x$-1s and $2p_z$-1s transitions calculated by using the theory described in Ref.[14]. Thus the G and D lines correspond to acceptor state transitions from the ground state, $1s_{3/2}(\Gamma_6 + \Gamma_7)$, to the first two excited odd-parity states, $2p_{3/2}(\Gamma_6 + \Gamma_7)$ and $2p_{5/2}(\Gamma_6 + \Gamma_7)$.

The results show that the energy positions of absorption lines in the Be doped MQW sample are respectively higher than those in the bulk GaAs. This is due to the confinement effect of QWs pushing the hole closer to the negative core, thereby increasing the Be acceptor binding energies and the 1s–2p transition energies.[14]

3.2. Picosecond pump-probe signal at different temperature

In order to study the relaxation of holes bound to the acceptors in QW, the time-resolved pump-probe experiments for the Be doped MQW sample were performed at temperatures of 4, 10, 20, 40, 60, and 80 K, respectively. The FEL wavelength was tuned to the position of the D-line for the sample studied. Figure 3 shows pump-probe signals as a function of delay between the pump pulse and the probe pulse at different temperatures. For the analysis of the experimental data, we used a model of a homogeneously broadened two-level system.[18,22] The rate equation is:

$$\frac{dN_{2p}}{dt} = \frac{\sigma}{\hbar\omega} I(t)(N_{1s} - N_{2p}) - \frac{N_{2p}}{\tau}, \qquad (1)$$

where N_{1s} and N_{2p} are the number of the acceptors with holes in the 1s and 2p state respectively, σ is the absorption cross section, and τ is the lifetime for the transition. $I(t)$ is the intensity of the laser pulse with time. It has a Gaussian shape with pulse duration 15 ps determined by pulse autocorrelation. The absorption of the probe is given by

$$A = \int_{-\infty}^{\infty} \frac{\sigma}{\hbar\omega} I(t - t_d)(N_{1s} - N_{2p}) dt, \qquad (2)$$

where t_d is the delay time between the pump pulse and the probe pulse. The above equations were solved numerically and the parameter τ was adjusted to obtain the best fit with experimental data.

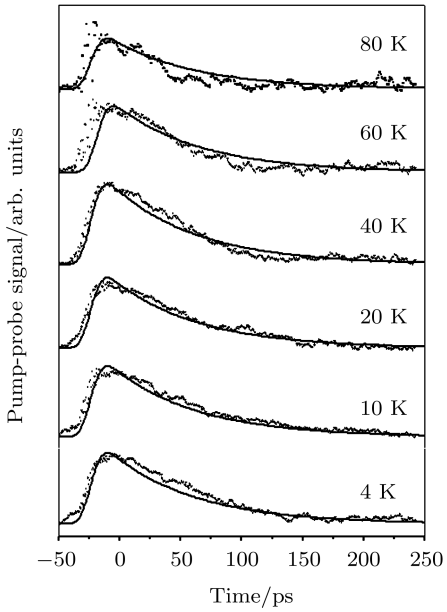


Fig.3. Pump-probe signal from the Be D-line transition in the GaAs/AlAs Be doped MQW sample with the 10 nm well at different temperatures. The dots indicate the experimental data, and the solid curves are generated by the theoretical fit with $\tau = 55$ ps.

In Fig.3, the dotted lines indicate the experimental data and the solid lines are generated by the theoretical fits with $\tau = 55$ ps according to the above two-level model. The relaxation time from 2p excited state to 1s ground state appears to be independent of temperature. For the GaAs:Be bulk sample, the 2p excited state lifetime $\tau = 350$ ps is obtained in the

same manner. This value is a factor of 6 more than that for the 10 nm Be doped MQW, and is independent of temperature also.[18]

The radiative lifetime for the D-line transition can be estimated by assuming that the Be acceptor is hydrogenic. As described in Refs.[6] and [23], the 2p→1s radiative lifetime of a hydrogenic atom is proportional to the square of the effective mass and is inversely proportional to the cube of the wave vector. Taking suitable parameters, we compute the radiative lifetime which is of the order of 0.1 ms, much longer than the 55 ps measured. This indicates that the observed lifetime is nonradiative. The relaxation process is independent of temperature, therefore it is not due to carrier–carrier scattering which changes along with the impurity ionization varying with temperature. Since the transition energy between 2p and 1s state is lower than the LO-phonon energy of 36 meV in GaAs, and the observed lifetime also fits in with typical acoustic phonon scattering time, the 2p→1s relaxation is accomplished mainly by acoustic–phonon scattering.

In bulk GaAs, the acoustic phonons near the Brillouin-zone centre have energies of only a few meV, less than the energy separation between the 2p and 1s states of the acceptor, so this relaxation of the 2p excited state must be multiphonon scattering. In MQW or superlattices, the existence of artifical periods causes Brillouin-zone folding, and creates additional acoustic phonon modes with energies larger than the acoustic phonon energy in the bulk but less than the LO-phonon energy. The reduction of crystal symmetry in QWs can produce zone-folded acoustic phonon scattering.[24−26] Thus the relaxation observed in MQW is due to the emission of the zone-folded acoustic phonons. The QW potential causes slower decay of the wavefunctions of impurity states in QWs at large wave vectors than that in bulk materials.[27] This slower decay causes faster 2p→1s transitions, assisted by the emission of zone-folded acoustic phonons, and accordingly results in dramatic reduction of 2p excited state lifetimes in MQW.

3.3. Laser wavelength dependence of pump-probe signal

At a temperature of 4 K, the pump-probe experiments for the Be doped MQW sample were repeated at the different FEL wavelengths of 43, 44, 46, 47 μm, respectively. The results are presented in Fig.4. It is shown that the 2p→1s relaxation time is strongly dependent on the laser wavelength. The pump-probe signals at λ = 43, 44, and 47 μm are observed to be two orders of magnitude weaker than that at the D-line position.[26]

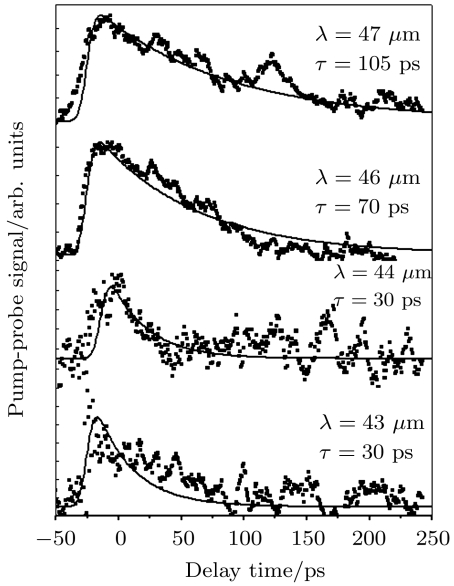


Fig.4. The laser wavelength dependence of pump-probe signal in the 10 nm Be doped MQW sample at 4 K. The dots indicate the experimental data, and the solid curves correspond to the theoretical fits at different laser wavelengths.

The well width fluctuations can affect the excited state lifetime, but much less than the observed lifetime differences at varying wavelengths, so the wavelength dependence cannot be due to well width fluctuations. The acceptor binding energy and hole wavefunction are all functions of the position of impurities in the well.[8,9] Although we used δ-doping Be acceptors, the dopant Be atoms still distribute along the growth direction of the QWs. This changes the impurity energy level and wavefunction whose value at interface determines the rate of transition.[27] Thus the 1s–2p transition energy and the excited state lifetime are changed. Because most impurity atoms are still at the centre of the well, the pump-probe signals at FEL wavelength of the D-line position are stronger than those at other wavelengths. Therefore, the most probable explanation for the laser wavelength dependence of the excited state lifetime is the diffusion of the Be atom δ-layer in QWs.

4. Conclusions

We have investigated the effect of QW confinement on intra-level dynamics of holes bound to acceptors in Be atom δ-doped GaAs/AlAs MQW by using far-infrared pump-probe spectroscopy. The results show that the hole relaxation process of 2p→1s in MQW is dominated by zone-folded acoustic phonons, and the hole relaxation time of 2p→1s decreases in MQW as a result of the slow decay of the wavefunctions of impurity states quickening the intra-acceptor scattering. The diffusion of the Be acceptor δ-layer is likely responsible for the wavelength dependence of the 2p lifetime.

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