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Spin quantum beats of bright and dark excitonic states in neutral InAs quantum dots

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Abstract
The energy eigenvalues and eigenfunctions of heavy-hole (HH) excitons and the expression of spin quantum beats (QBs) in neutral InAs quantum dots (QDs) in an oblique magnetic field have been calculated and derived in terms of a spin Hamiltonian model. It is shown from the oscillating curves of spin QBs that the amplitude and period of QBs are determined by the magnitude and direction of the magnetic field, and that the impact of the coupling component of electrons and holes, δ_0, on QBs should not be neglected even if in an intense magnetic field like $B = 4$ T, so long as the angle $\theta \leqslant 30°$; θ is the angle between the direction of the magnetic field and the z-direction (quantized direction). The influence of anisotropic g factors of electron and hole on QBs has also been studied. The calculated results show that g-factor markedly affected the amplitude and periodicity of QBs and the periodic oscillating of QBs vanishes at $\Delta g_x = g_{e,x} - g_{h,x} = 0.6$, which shows that whether or not the periodic signal of QBs can be observed is related to the 'matching' of the electron and hole g-factors.

1. Introduction

The spin dynamics of carriers and excitons in quantum wells and quantum dots is attracting a wide interest because of their potential applications such as spin logic switches, spin transistors and quantum computer [1–5]. The QB technique has been used for studying the fine structure and Zeeman splitting in quantum wells and superlattices [6–9]. In recent years, QBs of exciton fine-structure states in the absence of the magnetic field [10], spin QBs in microcavities [11, 12], luminescence QBs and biexciton QBs in strain-induced GaAs QDs [13, 14] have been observed and studied. In general fine structures in charged QDs are considerably complex, so the QBs between fine-structure Zeeman components can be observed just after removing the superfluous charges [15–17].

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In the studies cited above, fitting curves and analyses on their experimental results have been given to get the values of the g-factors of electrons and holes, and the QBs between exciton bright states have mainly been studied. The QBs of bright and dark excitonic states are discussed in few of them, and the coupling component of electrons and holes, δ_0, is often neglected [15]. Therefore, how spin QBs of bright and dark excitonic states in InAs QDs are affected by changing the magnetic field B, δ_0 and varying g-factors needs further investigation, which has attracted our interest.

In this paper, the energies and eigenfunctions of HH excitons and the expression of spin QBs, as functions of B, δ_0 and g-factors, have been calculated and derived in InAs QDs in an oblique magnetic field by making use of a spin Hamiltonian model. The oscillating curves of QBs are drawn, which vary with δ_0, different directions and strengths of magnetic fields and anisotropic g-factors of electrons and holes. The influence of them on QBs has been analysed and the conclusions drawn.

2. Theory

The spin Hamiltonian model can be characterized as fine structures of electron–hole pairs in QDs in a magnetic field, which can be obtained from the Hamiltonian of exciton spin states in a bulk semiconductor material after taking the size quantization along the growth axis z into account. Generally speaking, the Hamiltonian of exciton spin states in the bulk can be expressed as [18]

$$H_{\mathrm{ex}} = H_{\mathrm{e}} + H_{\mathrm{h}} + H_{\mathrm{e-h}}. \tag{1}$$

The first term of equation (1) describes the Zeeman splitting of electrons in a magnetic field, which can be written as

$$H_{\mathrm{e}} = \mu_{\mathrm{B}} \sum_{i=x,y,z} g_{\mathrm{e},i} S_{\mathrm{e},i} B_i, \tag{2}$$

where μ_{B} is the Bohr magneton, $g_{\mathrm{e},i}$, $S_{\mathrm{e},i}$ and B_i are the Cartesian components of the electron g-factor, the electron spin and the magnetic field, respectively. The second term of equation (1) describes the Zeeman splitting of holes, which can be expressed as

$$H_{\mathrm{h}} = -\mu_{\mathrm{B}} \sum_{i=x,y,z} (k_i J_{\mathrm{h},i} + q_i J_{\mathrm{h},i}^3) B_i, \tag{3}$$

where k_i and q_i are the Zeeman splitting constants, and $J_{\mathrm{h},i}$ is the Cartesian component of total angular momentum of the holes. The last term of equation (1) describes the exchange interaction of electron and hole spins including exchange, magnetic dipole–dipole and higher multipole interactions, which can be written as

$$H_{\mathrm{e-h}} = -\sum_{i=x,y,z} (a_i J_{\mathrm{h},i} S_{\mathrm{e},i} + b_i J_{\mathrm{h},i}^3 S_{\mathrm{e},i}), \tag{4}$$

where a_i and b_i are spin–spin coupling coefficients.

In InAs-type semiconductors at low temperature, the ground state is formed by an electron with the spin $S_{\mathrm{e}} = \frac{1}{2}$ and a hole with the angular momentum $J_{\mathrm{h}} = \frac{3}{2}$. Due to the quantum size effect in QDs the top of the valence band splits into a light hole (LH) with $J_{\mathrm{h},z} = \pm\frac{1}{2}$ and a heavy hole (HH) with $J_{\mathrm{h},z} = \pm\frac{3}{2}$. As a rule, the splitting energy is much greater than the value of Zeeman splitting, and the lowest energy state is the HH exciton state. Therefore the fine structures of the LH and HH excitons can be analysed independently. Here, the analysis shown below is restricted to the structure of HH excitons.

Assuming the state of the HH is $|h\rangle = |\pm\frac{3}{2}\rangle$, one can get

$$J_{\mathrm{h},x} |h\rangle = J_{\mathrm{h},y} |h\rangle = 0, \qquad J_{\mathrm{h},z}^3 |h\rangle = \tfrac{9}{4} J_{\mathrm{h},z} |h\rangle\,. \tag{5}$$

By using an effective HH spin $\tilde{S}_{\rm h}$, which has the relation $J_{{\rm h},z} = 3\tilde{S}_{{\rm h},z}$, equation (1) can be reduced as

$$H_{\rm ex} = \sum_{i=x,y,z} \left[\mu_{\rm B}(g_{{\rm e},i}S_{{\rm e},i} - g_{{\rm h},i}\tilde{S}_{{\rm h},i})B_i - c_i S_{{\rm e},i}\tilde{S}_{{\rm h},i}\right], \tag{6}$$

where $\tilde{S}_{{\rm h},z} = \pm\frac{1}{2}$ corresponds to the components $J_{{\rm h},z} = \pm\frac{3}{2}$, $g_{{\rm h},i}$ is the Cartesian component of the hole g-factor, and c_i is the spin–spin coupling constant. The relationship of $g_{{\rm h},i}$ and c_i with the coefficients in equations (3) and (4) is given by the formulae

$$\begin{aligned} &g_{{\rm h},x} = \tfrac{3}{2}q_x; \qquad g_{{\rm h},y} = -\tfrac{3}{2}q_y; \qquad g_{{\rm h},z} = 3k_z + \tfrac{27}{4}q_z;\\ &c_x = \tfrac{3}{2}b_x; \qquad c_y = \tfrac{3}{2}b_y; \qquad c_z = 3a_z + \tfrac{27}{4}b_z. \end{aligned} \tag{7}$$

The projections of total momentum upon the quantum z axis $J_z = S_{{\rm e},z} + J_{{\rm h},z}$ characterize the eigenstates of the fine structures of the HH exciton $\{\varphi_i\}$ $(i = 1, 2, 3, 4)$, which can be denoted as $|+1\rangle$, $|-1\rangle$, $|+2\rangle$ and $|-2\rangle$. The set of the states $\{\varphi_i\}$ can be taken as a basis of matrix representation of the HH exciton spin Hamiltonian. The matrix element of equation (6) under this basis can be expressed as

$$H_{ij} = \langle\varphi_i|\,H_{\rm ex}\,|\varphi_j\rangle, \qquad (i, j = 1, 2, 3, 4). \tag{8}$$

The matrix $H_{4\times4}$ can be divided into two parts: one of them comprises Zeeman components related to the magnetic field, and the other is related to the exchange interaction of electron and hole spins. They can be expressed as

$$H_{\rm Zeeman} = \frac{\mu_{\rm B}B}{2} \times \begin{pmatrix} -(g_{{\rm e},z}+g_{{\rm h},z})\cos\theta & 0 & g_{{\rm e},x}\sin\theta & -g_{{\rm h},x}\sin\theta \\ 0 & (g_{{\rm e},z}+g_{{\rm h},z})\cos\theta & -g_{{\rm h},x}\sin\theta & g_{{\rm e},x}\sin\theta \\ g_{{\rm e},x}\sin\theta & -g_{{\rm h},x}\sin\theta & (g_{{\rm e},z}-g_{{\rm h},z})\cos\theta & 0 \\ -g_{{\rm h},x}\sin\theta & g_{{\rm e},x}\sin\theta & 0 & -(g_{{\rm e},z}-g_{{\rm h},z})\cos\theta \end{pmatrix}, \tag{9}$$

and

$$H_{\rm e-h} = \frac{1}{2}\begin{pmatrix} \delta_0 & \delta_1 & 0 & 0 \\ \delta_1 & \delta_0 & 0 & 0 \\ 0 & 0 & -\delta_0 & \delta_2 \\ 0 & 0 & \delta_2 & -\delta_0 \end{pmatrix}, \tag{10}$$

where $\delta_0 = \frac{c_z}{4}$, $\delta_1 = -\frac{c_x+c_y}{4}$, and $\delta_2 = \frac{c_y-c_x}{4}$.

The values of the coefficients b_i in equation (4) are much less than those of a_i, so δ_1 and δ_2 in equation (10) can both be neglected based on equation (7) [19]. Then the energies of the HH exciton E_i $(i = 1, 2, 3, 4)$ can be calculated analytically from equations (9) and (10). The expressions of the energies are too verbose to present here and the eigenfunctions are linear combinations of the basis functions $\{\varphi_i\}$, namely

$$\psi_i = \sum_{j=1}^{4} a_{ij}\varphi_j. \tag{11}$$

Here, the expansion coefficients a_{ij}, which are related to the probability in the state φ_j, can be calculated by solving the stationary Schrödinger equation with Hamiltonian (9) and (10). As a rule, InAs QDs are negatively charged, and the superfluous charges can be removed using an external electric field, with which the curves of the photoluminescence (PL) spectra vary [7]. In this paper, we restrict the following analysis to neutral QDs on the supposition that we have applied an appropriate external electric field. When the InAs QDs are excited by a right-circularly polarized laser beam σ^+ the coherence pulse creates a linear superposition of the eigenstates $\{\psi_i\}$, whose evolved equation can be expressed as

$$\Psi(t) = \sum_{i=1}^{4} C_i^{\sigma^+} \psi_i \exp(-\mathrm{i}E_i t/\hbar). \tag{12}$$

The coefficients $C_i^{\sigma^+}$ are time independent, and their values are determined by the initially excited conditions. According to the selection rules, a right-circularly polarized laser beam can be used to excite the states related to $\varphi_1 = |+1\rangle$ only; then the relation can be easily obtained, $C_i^{\sigma^+} = a_{i1}^*$.

In general, the PL profile can be divided into smoothing and oscillating parts. The intensity of PL spectra under right-circularly polarized light can be written as

$$I_{\sigma^+} = I_\mathrm{s} + I_\mathrm{o}, \tag{13}$$

where I_s and I_o are the intensities of the smoothing and oscillating components, respectively. The PL intensity is proportional to the square of the matrix element of optical transition [15], namely

$$I_{\sigma^+} \propto |\langle \Psi(t)|\hat{d}|0\rangle|^2, \tag{14}$$

where $\hat{d}$ is the dipole moment operator and $|0\rangle$ is the ground state of the system. From equations (12)–(14) and $C_i^{\sigma^+} = a_{i1}^*$, the expressions of I_s and I_o are obtained, respectively:

$$I_\mathrm{s} = \sum_{i=1}^{4} |a_{i1}|^4, \tag{15}$$

and

$$I_\mathrm{o} = 2\sum_{i=1}^{k} \sum_{k=2}^{4} |a_{i1}|^2\, |a_{k1}|^2 \cos(\omega_{ik} t), \tag{16}$$

where ω_{ik} is the frequency of the transition from ψ_i to ψ_k,

$$\omega_{ik} = (E_i - E_k)/\hbar. \tag{17}$$

Then the expression of the oscillating part of PL spectra normalized to I_s, i.e. the intensity of the QBs, can be written as

$$I_{\mathrm{beats}}^{\sigma^+}(t) = \frac{2R\sum_{i=1}^{k}\sum_{k=2}^{4} |a_{i1}|^2\, |a_{k1}|^2 \cos(\omega_{ik} t)}{\sum_{i=1}^{4} |a_{i1}|^4} \exp\left(-\frac{t}{\tau}\right), \tag{18}$$

where R is the amplitude factor and τ is the decay time of the oscillations.

Considering that the different sizes, shapes and strains in each direction in QDs cause the g-factors of the electron and hole to be anisotropic [20–23], different curves of the QBs in variation with the magnetic field can be obtained by selecting teams of anisotropic g-factors under different magnetic fields.

3. Calculation and discussion

Based on the theory above, it can be found that the parameters of the energy E_i comprise δ_0, $g_{\mathrm{h},z}$, $g_{\mathrm{h},x}$, $g_{\mathrm{e},z}$, $g_{\mathrm{e},x}$ and $\mathbf{B}(\theta)$, whereas the QB intensity consists of a_{ij} and E_i.

The coefficients a_{11} and a_{31} in equation (18) are not equal to zero when $g_{\mathrm{h},x} = 0$, and the energies of HH excitons can be written as $E_{1,3} = -[g_{\mathrm{h},z}\cos\theta \pm (g_{\mathrm{e},z}^2\cos^2\theta + g_{\mathrm{e},x}^2\sin^2\theta)^{1/2}]\mu_\mathrm{B}B/2$, from which it is easily found that the value of $(E_1 - E_3)/\hbar$ does not

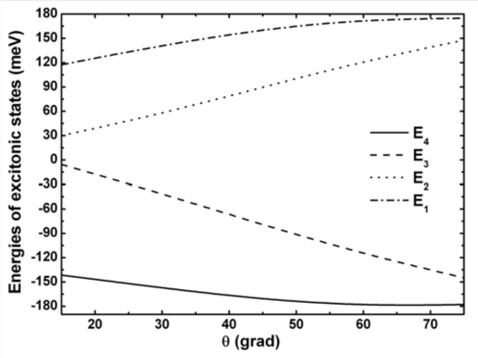

Figure 1. Variation of the HH exciton energies with the angle of the magnetic field for $B = 4$ T, $R = 0.75$, $g_{e,x} = 1.43$, $g_{e,z} = 0.53$ and $\delta_0 = 15$ μeV.

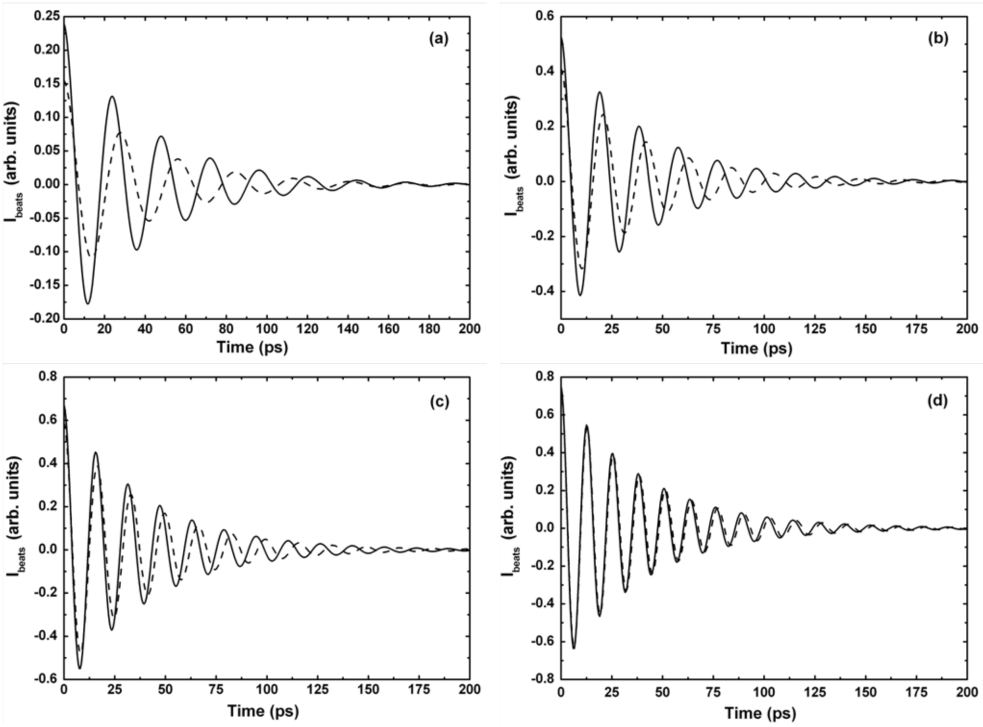

Figure 2. The variation of QBs with different angles of magnetic field: (a) $\theta = 15°$, (b) $\theta = 30°$, (c) $\theta = 45°$ and (d) $\theta = 75°$. The solid lines are at $\delta_0 = 15$ μeV and the dashed lines are at $\delta_0 = 0$ μeV.

vary with the factor $g_{h,z}$. Choosing $B = 4$ T, $R = 0.75$, $g_{e,x} = 1.43$, $g_{e,z} = 0.53$, and $\delta_0 = 15$ μeV, the variations of the energies and the corresponding QBs with the angles of magnetic field are shown in figures 1 and 2, respectively. A growth of the angle θ is accompanied by increases in the amplitude and the period of the QBs, shown in figure 2, which accords with the results in [15]. After comparing the oscillating part of the QBs at $\delta_0 = 15$ μeV (the solid line) with that at $\delta_0 = 0$ μeV (the dashed line) in figures 2(a)–(c), it can be found that the amplitudes of the former are bigger and the periods are shorter than those of the latter. With a further increase of the angle θ, the transversal effect of the magnetic field exceeds that

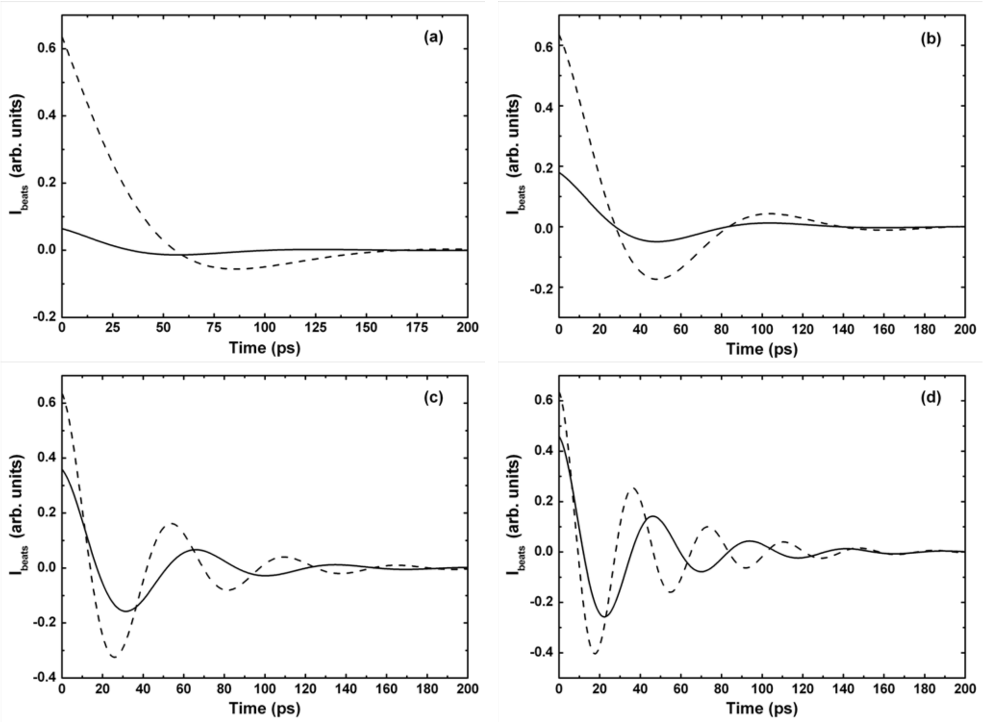

Figure 3. Variation of the QB intensity with the magnetic field for $R = 0.75$, $g_{e,x} = 1.43$, $g_{e,z} = 0.53$, $g_{h,x} = 0$, and $\theta = 60°$: (a) $B = 0.2$ T, (b) $B = 0.4$ T, (c) $B = 0.8$ T, and (d) $B = 1.2$ T. The solid lines are at $\delta_0 = 15$ μeV and the dashed lines are at $\delta_0 = 0$ μeV.

of δ_0, which can be neglected at that time, and the solid and dashed lines gradually superpose, as is shown in figure 2(d). Therefore the impact of δ_0 on QBs should not be neglected in an intense magnetic field when the angle $\theta \leqslant 30°$.

At the fixed value of the angle $\theta = 60°$, using $R = 0.75$, $g_{e,x} = 1.43$, $g_{e,z} = 0.53$, and $g_{h,x} = 0$, the curves of the QBs varying with the magnitude of magnetic field can be obtained, as is shown in figure 3. At $B = 0.2$ T the shapes of the QBs decay almost smoothly, which is shown in figure 3(a). And when the magnetic fields B grow from 0.4 to 1.2 T the amplitudes of the QBs augment and the periods shorten (see the solid or dashed lines in figure 3). After comparing the oscillating part of the QBs at $\delta_0 = 15$ μeV (the solid line) with that at $\delta_0 = 0$ μeV (the dashed line) in figure 3, it can be found the amplitudes of the former are smaller and the periods are longer than those of the latter, which show the contrary phenomena to figure 2. Then from figures 2 and 3, we conclude that the magnitude and direction of the magnetic field B determine the amplitude and period of the QBs, and that with a growth of B the exchange interaction δ_0 reduces the amplitudes of QBs and increases their periods at the beginning, but later on, δ_0 increases the amplitudes and reduces the periods.

In order to compare the influence of $g_{e,x}$ and $g_{h,x}$ on the QBs, the variations of QBs with them are shown in figure 4 by selecting the coefficients in InAs QDs $g_x = g_{e,x} + g_{h,x} = 1.8$ and $g_z = g_{e,z} + g_{h,z} = 0$ [18]. Then at $\delta_0 = 15$ μeV, $\theta = 30°$, $g_{e,x} = 1.0$, $g_{h,x} = 0.8$, i.e. $\Delta g_x = g_{e,x} - g_{h,x} = 0.2$, and $g_{e,z} = -g_{h,z} = 1.2$ we can find the diminution of the amplitudes and shortening of the periods of the QBs with increasing of the magnetic field from 1 to 3 T, as shown in figure 4(a); at $\Delta g_x = -0.2$ and $g_{e,z} = -g_{h,z} = 0.9$ we find similar

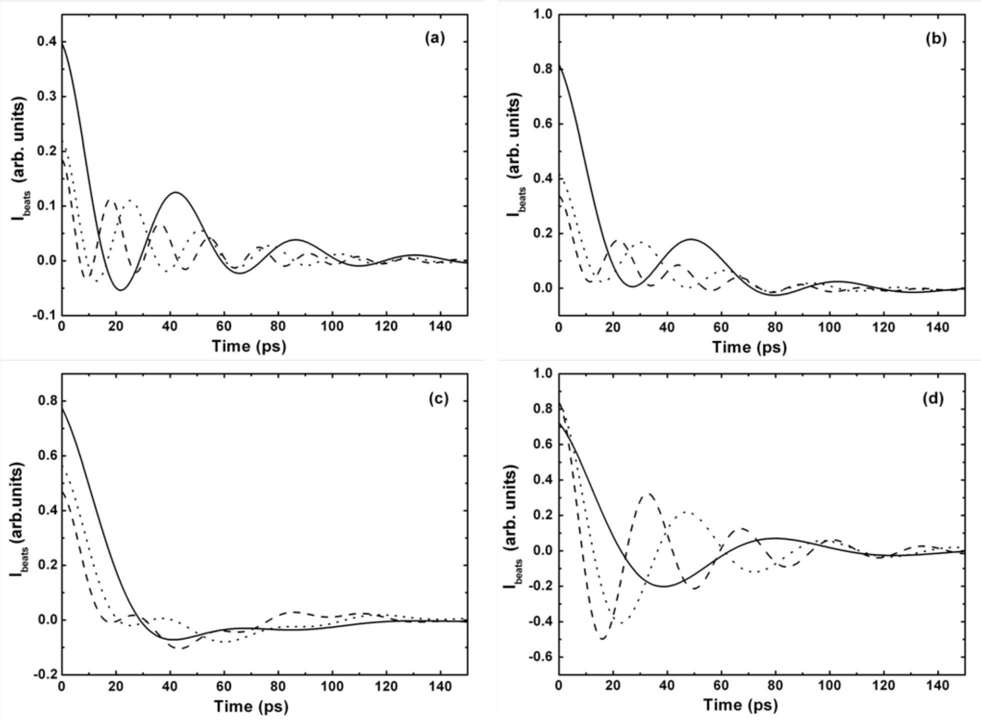

Figure 4. The oscillating curves of the QBs at $R = 0.75$, $\delta_0 = 15$ μeV, and $\theta = 30°$ for different g-factors: (a) $g_{e,x} = 1.0$, $g_{h,x} = 0.8$, $g_{e,z} = -g_{h,z} = 1.2$; (b) $g_{e,x} = 0.8$, $g_{h,x} = 1.0$, $g_{e,z} = -g_{h,z} = 0.9$; (c) $g_{e,x} = 0.6$, $g_{h,x} = 1.2$, $g_{e,z} = -g_{h,z} = 0.5$; (d) $g_{e,x} = 0.1$, $g_{h,x} = 1.7$, $g_{e,z} = -g_{h,z} = 0.2$. The solid, dotted and dashed lines denote the QBs in the magnetic fields $B = 1$, 2, and 3 T, respectively.

changes, besides a faster decaying of the amplitude, as shown in figure 4(b); at $\Delta g_x = -0.6$ and $g_{e,z} = -g_{h,z} = 0.5$ there is no obvious oscillating of the QBs, as shown in figure 4(c); at $\Delta g_x = -1.6$ and $g_{e,z} = -g_{h,z} = 0.2$ the regular signals of QBs appear again with increasing of the magnetic from 1 to 3 T, accompanied by increases of the amplitude and period, as shown in figure 4(d). We chose the different $g_{e,z}$ and $g_{h,z}$ above in order to make it easier to show the regularity of the variations of the QBs. In the same way, the QBs are seen to show similar changes using different $g_{e,z}$ and $g_{h,z}$ with fixed $g_{e,x}$ and $g_{h,x}$.

Whether the transitions between the excitonic states in InAs QDs in an oblique magnetic field can happen is related to the probabilities of the transitions, from which we can find that, when only one frequency $\omega = (E_i - E_j)/\hbar$ is dominant (here i and j are fixed numbers such as 1 and 3), the corresponding probabilistic factor $|a_{i1}|^2|a_{k1}|^2$ in equation (18) is much larger than the others, and then the transition between the states ψ_i and ψ_j is dominant and the oscillating shapes of QBs are periodically decayed, as shown in figures 4(a), (b) and (d); whereas when there are several frequencies in QBs at the same time, i.e. several factors $|a_{m1}|^2|a_{n1}|^2(m, n = 1, 2, 3, 4, m \neq n)$ with similar values, the shapes of the QBs tend to be nonperiodic, as shown in figure 4(c). Because the energies of excitons and the probabilities of optical transitions are both connected to the g-factors of the electrons and holes, we conclude that whether the oscillating signals of QBs can be observed in the oblique magnetic field is related to the 'matching' of the g-factors. And since neutral InAs QDs are obtained by removing the superfluous charges, one should consider the indirect influence of the external electric field on the g-factors besides the size, shape and stress in QDs, which needs further studies.

4. Conclusion

In this paper, we have calculated and derived the energy eigenvalues and eigenfunctions of heavy-hole (HH) excitons and the expression of spin quantum beats (QBs) in neutral InAs quantum dots (QDs) in an oblique magnetic field in terms of a spin Hamiltonian model. Changing the direction and intensity of the magnetic field, the anisotropic g-factors of the electrons and holes, we have plotted the oscillating curves of spin QBs, which show that the impact of δ_0 on QBs in a magnetic field should not be neglected even in an intense magnetic field such as $B = 4$ T when the angle $\theta \leqslant 30°$. The influences of anisotropic g-factors of the electrons and holes on QBs have been studied as well. It is found that the g-factors markedly affected the amplitude and periodicity of QBs, and that the periodic oscillating of QBs vanishes at $\Delta g_x = g_{e,x} - g_{h,x} = 0.6$, which shows that whether the periodic signal of QBs can be observed or not is related to the 'matching' between the g-factors of the electron and the hole, which calls for further studies. The work in this paper is of much importance in understanding the fine structures and spin QBs of the excitons, and promoting corresponding research and application.

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