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Spin quantum beats in InP quantum dots in a magnetic field

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Abstract. In this paper, we studied quantum beats observed in kinetics of photoluminescence of a single layer of InP self-assembled quantum dots in a magnetic field. The reason for the quantum beats is shown to be Zeeman splitting of the exciton radiative level. Studies of behavior of the quantum beats versus the magnetic field magnitude and orientation and versus polarization of the exciting and detected light, have allowed us to identify fine structure of radiative level of the electron-hole pairs and to evaluate g-factors of electrons and holes.

Spin quantum beats is a phenomenon intensively studied in structures with quantum wells, whereas just a few observations of the spin quantum beats in quantum-dot structures is now available in the literature [1, 2]. The reason why the beats are absent in quantum dots, at present, becomes evident. The point is that quantum dots, as a rule, contain excess charge carriers (are charged), and, as a result, fine structure of the energy levels becomes much more complicated [2, 3, 4]. In this paper, we show that discharging quantum dots by means of external electric field gives rise to intense quantum beats in kinetics of photoluminescence (PL) of quantum dots in a magnetic field. Studies of behavior of the beats versus the magnetic field magnitude and orientation and versus polarization of the exciting and detected light have allowed us to identify fine structure of radiative level of electron-hole pairs and to evaluate g-factors of electrons and holes.

We studied a structure with a single layer of the InP quantum dots. A detailed description of the structure is given in [5]. The luminescence was excited by 3 ps pulses of a Ti:sapphire laser, 40 meV above the PL band maximum. The PL kinetics was measured with a time resolution of 6 ps using a 0.25 m subtractive-dispersion double monochromator and a streak-camera. The measurements were made in a cryostat with a superconducting solenoid, in magnetic fields from 1 to 7 T. The design of the cryostat allowed us to excite the sample and to detect its emission only along and across the magnetic field direction (the Faraday and Voigt configurations, respectively). To change the angle ϑ between the growth axis of the sample z and direction of the magnetic field B, we rotated the sample around vertical axis.

It has been established that the quantum beats can be observed only in the presence of electric bias $U_{bias} < -0.3$ V applied to the sample. Mote that the frequency of the beats proved to be independent of the voltage U_{bias}. The beats could be observed at Stokes shifts from 5 to 50 meV, with the form of the beats being virtually independent of the shift value. This is the main difference between these quantum beats and those observed in two-dimensional structures [6].

We have studied PL kinetics at linear and circular polarization of excitation, with co- and cross-polarizations of the detected PL. It was found that the amplitude of the beats is the greatest in circular co-polarization of the PL. It is noteworthy that in none of the experiments have been observed antiphase oscillations in the co- and cross-polarized luminescence, which are typical for quantum beats on Zeeman sublevels [7].

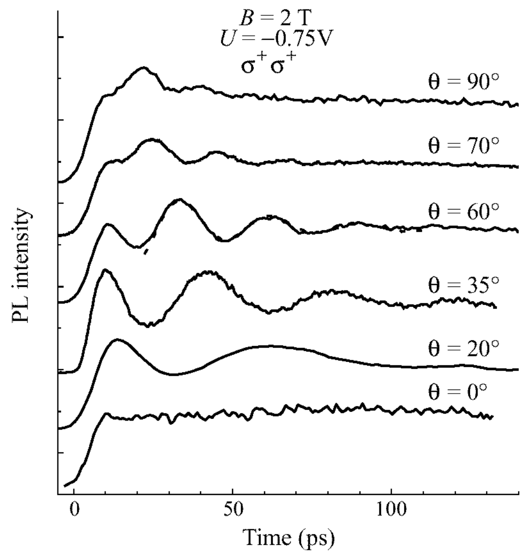

Fig. 1. PL kinetics in the magnetic field $B = 2$ T at different angles ϑ between the magnetic field and growth axis of the structure (solid lines). The dashed line shows fitting the oscillations in one kinetics by formula (1). Polarizations of the PL excitation and detection are σ^+. The PL Stokes shift — $\Delta E = 45$ meV, electric bias — $U_{bias} = -0.75$ V.

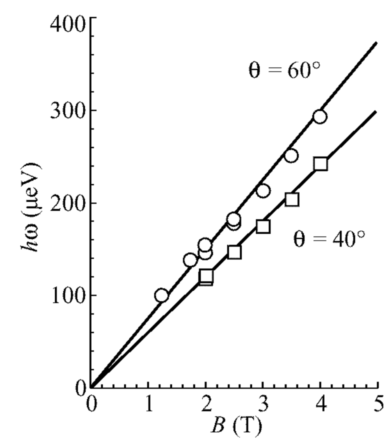

Fig. 2. Dependence of the beat frequency (in energy units) on the magnetic field at different ϑ. Solid lines — calculations by formulas (2) and (3).

Figure 1 shows kinetics of circularly polarized luminescence in the field $B = 2$ T at different ϑ. Intense beats are seen to be observed within the range of angles $\vartheta = 20-70°$, the frequency of the beats being decreased with decreasing ϑ. These beats can be well described by the function

$$y = C \cdot \exp(-t/\tau) \cos(\omega t), \tag{1}$$

where the decay time τ and the beat frequency ω are the fitting parameters.

For the angle $\vartheta = 0°$ no oscillations are observed at any magnetic field and any light polarization. Dependence of the oscillation frequency ω on magnetic field, measured at two values of the angle ϑ is shown in Fig. 2. As is seen from the figure, the experimental points well fit straight lines coming out of the coordinate origin.

We suggest that the oscillations observed are related to quantum beats between the fine-structure states of electron-hole pairs coherently excited by laser pulses via higher-energy states. To quantitatively describe the fine splitting, we used the spin-Hamiltonian of electron-hole pair in a magnetic field similar to that given in [8]. This Hamiltonian contains 6 free parameters - 2 components of the exchange splitting, two components of the electronic g-factor, and two components of the hole g-factor (assuming axial symmetry of a quantum dot). Analysis of the above experimental data allowed us to significantly simplify the Hamiltonian. Since, as follows from Fig. 3, no deviation from linear dependence of the splitting is observed in the fields $B > 1$ T, the exchange splitting can be neglected. The magnetic field $B \parallel z$ splits the state of the electron-hole pair into 4 components, two of them being optically active. This splitting is proportional to the difference between z-components of the electron and hole g-factors: $\delta E \sim (g_{e,z} - g_{h,z})$. It follows from the aforementioned absence of antiphase oscillations at the angle $\vartheta = 0°$ in any magnetic field that splitting of the radiative doublet, in such a field, is small, i.e., $g_{e,z} \approx g_{h,z}$. In addition, we suppose, for simplicity, that the electron g-factor tensor is spherical, i.e., $g_{e,x} = g_{e,x}$. In the framework of the these approximations, the spin-Hamiltonian depends on two free parameters only ($g_{e,z}$ and $g_{h,x}$), and the values of the fine-structure state energies can be calculated analytically:

$$E_{1,2,3,4} = \pm\frac{1}{\sqrt{2}}\mu B g_{e,z}\left\{\cos^2(\vartheta) + \frac{1}{2}(g_{e,z}^2 + g_{h,x}^2)\sin^2(\vartheta)\right. \\ \left.\pm\left[(\cos^2(\vartheta) + g_{e,z}^2\sin^2(\vartheta))\cdot(\cos^2(\vartheta) + g_{h,x}^2\sin^2(\vartheta))\right]^{1/2}\right\}^{1/2} \tag{2}$$

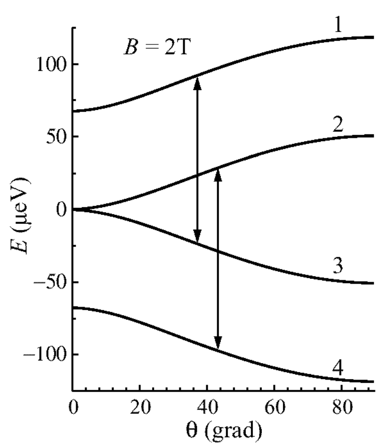

Fig. 3. Splitting of the electron-hole pair fine structure in a magnetic field at different ϑ. The states mixed by the magnetic field responsible for quantum beats in circular co-polarizations are bridged by arrows.

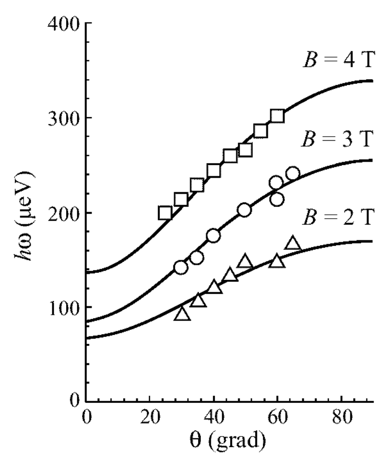

Fig. 4. Dependence of the beat frequency (in energy units) on the angle ϑ in different magnetic fields. Solid lines — calculations by formulas (2) and (3).

Figure 3 shows variation of the energy levels with the angle ϑ for the magnetic field $B = 2$ T, plotted using experimental values of the parameters (see below). At the angle $\vartheta = 0°$, the optically active state is not split, and transitions into the split states are forbidden by the selection rules. Thus, no quantum beats can be observed in these conditions. At finite angles ϑ, the magnetic field mixes all the split states, and possibility to observe the beats is determined by the degree of their mixing. In the conditions of the experiment, the beats were mainly observed at a single frequency. A satisfactory agreements between calculated curves and experimental data proved to be possible under assumption that the mixed states are those bridged by arrows in Fig. 3.

Figure 4 shows dependence of the beat frequency (in energy units) on the angle ϑ in different magnetic fields. As is seen, the experimental dependences well agree with the calculated curves

$$\Delta E = E_1 - E_3, \tag{3}$$

where E_i are calculated by formula (2). The theoretical curves are plotted for the fitting parameters $g_{e,z} = g_{e,x} = g_{h,z} = 0.6$ and $g_{h,x} = 1.5$. The same parameters were used to calculate dependences of the beat frequencies on magnetic field (solid curves in Fig. 3).

In summary, the proposed model can describe fairly well behavior of the quantum beat frequencies in a magnetic field. To describe amplitudes of the quantum beats, a more detailed analysis is needed, which will make it possible to refine the model.

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