The influence of δ-<Mn>-layer’s magnetization on polarization of photoluminescence of quantum well in singular and vicinal InGaAs/GaAs/δ-<Mn> heterostructures

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Abstract. Magneto-optical properties and spin dynamics in a p-type GaAs-based heterostructures containing an InGaAs quantum well and 2 nm thick GaAs layer doped with 5 at. % Mn (GaAs:Mn) on flat and vicinal substrates were studied between 2 and 300 K. A significant enhancement of photoluminescence circular polarization \( P_C = 15 \% \) was found at temperatures \( T < 30 \) K. In the non-magnetic quantum well \( P_C = 0 \% \). The polarization of the InGaAs quantum well photoluminescence follows the temperature dependence of the GaAs:Mn layer magnetization. Two types of ferromagnetism were found. In the heterostructures grown on the flat substrate parallel to the (001) GaAs plane the magnetization obeys the Bloch \( T^{3/2} \) temperature dependence while for the structures grown on the vicinal surface (disoriented by 3°) the magnetization follows percolation dependence. The later is attributed to the formation of strongly disordered islands of GaAs:Mn.

1. Introduction

Heterostructures containing a quantum well and a thin layer of a diluted magnetic semiconductor (DMS) like GaAs:Mn have attracted great attention due to their possible application for injecting spin-polarized carriers in light emitting diodes [1]. The implantation of Mn into the quantum well leads to low mobility of charge carriers and suppresses radiative recombination [2]. The spatial separation of charge carriers and the GaAs:Mn layer results in a considerable increase in the mobility of charge carriers and the luminescence quantum yield [3]. This combination of optically and magnetically active structures appears promising for controlling optical properties by spin-dependent magnetic field control over the luminescence.

The presence of charge carriers in the GaAs:Mn layer facilitates the indirect Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interaction between Mn\(^{2+}\) ions in hybrid structures [4-8]. Measurements of the luminescence polarization [8], the electric resistance and the Hall voltage [4-7] indirectly confirmed the existence of ferromagnetism in GaAs:Mn layers. The interpretation of the magnetization data, however, is difficult and usually requires additional assumptions for the separation of the contributions of the diamagnetic crystal lattice and paramagnetic structural defects, substrate, and interlayer boundaries. This problem can be solved by the electron spin resonance (ESR) technique [9] allowing the separation of contributions of above mentioned subsystems to the magnetic susceptibility of the samples.
It will be shown here that the circular polarization of the photoluminescence of a InGaAs quantum well is sensitive to the ferromagnetic ordering in the GaAs:Mn layer. The combinations of optical measurements with superconducting quantum interference device (SQUID) magnetometry and electron spin resonance (ESR) spectroscopy allowed us to identify fine details of electronic transitions, their dependence on spin polarization and their correlation to the observed increase of the circular polarization in the ferromagnetic ordered state of GaAs:Mn.

Here, we present results on:

a) the ferromagnetic GaAs:Mn layer contribution to the magnetic moment of the samples,
b) the effect of the GaAs:Mn layer magnetization on the polarization of the photoluminescence of the InGaAs quantum well,
c) the effect of different GaAs substrate orientations on the type of magnetic ordering,
d) exchange parameters, magneto-optical properties and spin dynamics in the GaAs-based heterostructures with a InGaAs quantum well and a GaAs:Mn layer.

2. Experimental setup

P-type GaAs-based heterostructures containing a InGaAs quantum well and GaAs:Mn layer (Mn delta-doped GaAs thin layer) (figure 1) were prepared by stepwise epitaxial growth [10].

Figure 1. Scheme of the heterostructure grown on a 0.5 mm GaAs substrate (1). Heterostructure consists of 0.5 μm thick GaAs buffer layer (2), 1 nm thick GaAs:C layer (3), 15 nm thick GaAs separating layers (4), 10 nm thick InGaAs quantum well (5), 3 nm thick GaAs separating layers (6), 2 nm thick GaAs:Mn layer (7) and 30 nm thick GaAs cover layer (8). External magnetic field $B_{ext}$, magnetic field $B_{well}$ in the GaAs:Mn layer with spin-polarized $\text{Mn}^{2+}$ ions, and magnetic field $B_{int}$ in the InGaAs quantum well with spin-polarized holes $p$ are shown.
InGaAs quantum well was grown by metal-organic hydride epitaxy on a GaAs substrate, as described in detail earlier [10]. The GaAs:Mn layer (concentration of Mn at. 5 %) was grown in the same reactor by laser ablation of Mn and GaAs targets [10]. Moreover, in order to compensate the effects of depletion of charge carriers in the quantum well, the acceptor GaAs:C layer was deposited on the side of the buffer layer in all studied heterostructures.

We studied three types of heterostructures:

1. Three samples of singular heterostructures containing an InGaAs quantum well and a 2 nm thick ordered GaAs:Mn layer on a precisely oriented GaAs (001) substrate. The angle between (001) crystallographic plane and the substrate plane is φ < 0.2º.

2. Three samples of vicinal heterostructures containing InGaAs quantum well and disordered GaAs:Mn layer on a vicinal GaAs (001) substrate, with an angle φ = 3 ± 0.2 º between the (001) crystallographic plane and the substrate plane.

3. Two samples of reference heterostructure containing an InGaAs quantum well and a GaAs:C layer instead of the GaAs:Mn layer. GaAs:C layer was used to determine the contribution of boundaries between layers and paramagnetic defects in addition to the ferromagnetic contribution in the GaAs:Mn layer. Thus, reference heterostructures contain two GaAs:C layers.

The temperature dependence of the magnetization was measured using a SQUID magnetometer (MPMS 5XL, Quantum Design) at temperatures T = 2 - 300 K and in the magnetic field B = 0.1 T. ESR measurements were carried out at 9.5 GHz with a modulation frequency of 100 kHz in the temperature range T = 4 - 300 K using a Bruker ESR-500 spectrometer and Oxford Instrument cryostat. The magnetic field was applied along the [001] direction perpendicular to the plane of the heterostructure. Technical details of the experiment have been described elsewhere [11]. The circular polarization, $P_C$, of photoluminescence was measured in a magnetic field of B = 3 T in an Oxford Instruments cryostat.

Polarization of photoluminescence was determined by formula $P_C = (I_+ - I_-)/(I_+ + I_-)$, where $I_+$($I_-$) is the intensity of components with the right (left) polarization obtained by integrating the part of the spectrum corresponding to the optical transition in a quantum well. Photoluminescence was excited with a 632.8 nm He-Ne laser and the magnetic field oriented along the [001] direction perpendicular to the substrate and recorded with a spectral resolution better than 0.05 meV.

3. Experimental results

3.1. Static magnetization of singular heterostructures

We have studied three singular samples to be sure in reproducibility of the experimental data. All three samples possessed same magnetic properties. Typical temperature dependence of the magnetization $M(T)$ for one of the samples is shown on the figure 2. Decrease of the magnetization of δ-<Mn>-layer caused by heating obeys Bloch formula below Curie temperature as it usually observed in the bulk ferromagnetic materials. Curie temperature $T_C$ can be estimated in the frame of Weiss mean field theory. In ferromagnets the magnetization, $M$, is expected to decrease with temperature, $T$, obeying the Bloch $T^{3/2}$ law in the low-temperature range [12]:

$$M(T) = M_0(1 - \beta T^{3/2})$$

(1)

where $M_0$ is the magnetic moment at $T \to 0$ K, and $\beta$ the spin-wave parameter. The Bloch $T^{3/2}$ law has experimentally been found to be applicable for thin ferromagnetic GaMn:As films [13]. We approximated the experimental $M(T)$ dependence for the singular heterostructures using Eq. (1) in the low-temperature range 2 – 10 K. In our case $M_0 \approx M(T = 2 \text{ K})$ is the experimental value while $\beta$ is a fitting parameter. We have found $\beta = (2.1 \pm 0.1) \cdot 10^{-2} \text{ K}^{-3/2}$. In our case $\beta$ value exceeds one obtained in [13] by order of value because Mn$^{2+}$ concentration is higher and J value in our heterostructure is larger at the approximately same Curie temperature $T_C$.

In conventional spin-wave theory [12], the spin-wave parameter, $\beta$, and spin-wave stiffness, $D$, are conjugated by the equation [13]:

$$D \propto J$$
\[ \beta = 2.612 \frac{g \mu_B}{M_0} \left( \frac{k_B}{4\pi D} \right)^{3/2} \]  

(2)

where \( g \approx 2 \) is the g-factor of a Mn\(^{2+}\) ions, \( \mu_B \) is the Bohr magneton, \( k_B \) is the Boltzmann constant. The value of spin-wave stiffness determined from the Eq. (2) is \( D = 1.5 \pm 0.3 \) T \( \cdot \) nm\(^2\). The value of exchange integral, \( J = 0.18 \pm 0.01 \) meV, was calculated using the following expression [14]:

\[ J = \frac{D g \mu_B}{2 S r_s^2} \]  

(3)

where \( r_s \approx 7 \) Å – average distance between Mn\(^{2+}\) ions, \( S = 5/2 \) is the spin of Mn\(^{2+}\) ions. The obtained values of \( \beta, D, J \) are in good agreement with the results reported earlier for similar systems by other authors [15, 16].

Figure 2. Temperature dependencies of the magnetization of the singular (1) and vicinal (2) heterostructure with the GaAs:Mn layer and the reference heterostructure without GaAs:Mn layer (3) at \( B = 0.1 \) T. The solid lines are approximations (see the text). The inset shows temperature dependencies of the magnetization of a vicinal heterostructure containing GaAs:Mn layer cooled in zero magnetic field (ZFC) and \( B = 1 \) T magnetic field (FC). The magnetic field was applied in the (001) heterostructures plane. The diamagnetic contribution from the GaAs substrate was subtracted. The error bar (± 0.2 \( \cdot \) 10\(^{-4}\) A/m) is equal to the symbol size.

It means that quantum well does not effect on the magnetization of the GaAs:Mn layer. Experimental value of Curie temperature, \( T_C = 35 \pm 5 \) K, was defined by minimum of the d\( M/dT \) derivative. We used mean-field approximation \( M(T) \sim M_0 (1 - T/T_C)^{1/2} \) of the experimental \( M(T) \) dependence in the middle-temperature range 10 – 25 K. In the frame of the mean-field theory, the Curie temperature, \( T_C \), is defined as [16]:

\[ T_C = \frac{zS(S+1)}{k_B} \]  

(4)

This equation allows us to estimate coordination number \( z \). Assuming \( T_C = 35 \) K, one can obtain \( z = 4.38 \pm 0.6 \). This value is very close to expected value \( z = 4 \) typical for tetrahedral symmetry. Coordination number does not mean the number of nearest Mn\(^{2+}\) ions. The \( z \) value corresponds to number of exchange channels including indirect exchange interaction through Ga or As atoms. Because in GaAs indirect exchange is provided by charge carriers, \( z = 4 \) is reasonable value indicating tetrahedral Mn\(^{2+}\) environment.

3.2. Static magnetization of vicinal heterostructures

We have studied three vicinal samples to be sure in reproducibility of the experimental data. All three samples possessed same magnetic properties. Typical temperature dependence of the magnetization \( M(T) \) for one of the samples is shown on the figure 2. In vicinal heterostructures the magnetization, \( M \), decreases gradually with temperature, \( T \), in the low-temperature range 2 – 35 K (in
contrast to a Bloch and mean-field type M vs. T curve in the singular heterostructure). As it was shown previously this type of the temperature dependence is typical for the percolation ferromagnetic ordering [17-19]. The percolation model is based on the assumption that indirect exchange between magnetic centres (Mn$^{2+}$ ions) is carried out by charge carriers (holes in the case of GaAs:Mn). The exchange interaction between holes and magnetic ions leads to the formation of crystal regions with local ferromagnetic ordering. At high temperature, there is no correlation between such individual ferromagnetic regions, their macro spins are oriented arbitrarily and the net magnetic moment obeys the Curie law for superparamagnets. However, as temperature decreases, the radius of ferromagnetic regions increases. The increasing interaction between different ferromagnetic regions leads to macroscopic correlation between magnetic moments of the individual magnetic ions and appearance of the net magnetization. When such correlated clusters of ferromagnetic regions have extended and coupled through the whole sample, the ferromagnetic ordering is complete. From the temperature dependence of the magnetization of the vicinal heterostructures cooled in zero magnetic field (ZFC) and in 1 T magnetic field (FC) (Figure 2) we determine the Curie temperature, $T_C = 30 \pm 5$ K as a point which below the ZFC and FC curves are differ. We approximated the experimental M(T) dependence for the vicinal heterostructures using the expression obtained by the percolation model [17]:

$$M(T) = M_0 \{1 - \exp\left[-\frac{R^3}{r_s^3} \ln \frac{JS(S+1)}{T} \right]\}$$

(5)

where $R$ – radius of indirect exchange interaction. A close correlation between the experimental dependence M(T) and theoretical curve (figure 2), indicates a percolation nature of the ferromagnetism in the vicinal heterostructures.

In our case $M_0 \approx M(T = 2$ K), $r_s = 7$ Å and $S = 5/2$ are fixed values while R and J are variable fitting parameters. The value of exchange integral determined from approximation is $J = 0.72 \pm 0.04$ meV. The parameter R was also extracted from the approximation is $R = (0.21 \pm 0.03) \cdot r_s = 1.47$ Å. This value is practically equal to Bohr radius $a_B = 1.5$ Å of the Mn$^{2+}$ ion in GaAs. This remarkable fact seems to be not casual. Coincidence of the radius of indirect exchange interaction R with localization length indicates contribution of the localized charge carriers to the indirect exchange interaction. Most probably holes being captured by shallow traps near top of the valence band has admixture of d-orbital of Mn$^{2+}$ wave functions. This localization of the holes provides additional channels of exchange interaction between Mn$^{2+}$ ions.

### 3.3. High-frequency dynamical magnetic properties of singular and vicinal heterostructures

The ESR spectrum of the singular heterostructure shows: an non-angular dependent (isotropic) line near the zero magnetic field (line a), isotropic lines b, d, e and angular dependent anisotropic line c. Isotropic lines b, d, e are also observed in heterostructures without the GaAs:Mn layer (figure 3) and in a GaAs substrate. Consequently lines b, d, e corresponds to the paramagnetic centres of an uncontrollable paramagnetic impurity in all samples.

The intensity of line c drastically decreases with increasing temperature, disappearing at temperatures above 40 K. Resonance line c was observed only in samples with the GaAs:Mn layer (figure 3). The numbers of magnetic moments was determined from the calculated area under the absorption curve (second integral) of ESR spectrum of the line c. The spin number normalized to the known number of spins of a reference CuSO$_4 \cdot 5$H$_2$O sample was 100 times higher than the number of spins corresponding to noninteracting Mn$^{2+}$ ions at 4 K. Therefore, line c corresponds to the enhanced susceptibility due to the long-range ferromagnetic order, that is ferromagnetic resonance in the GaAs:Mn layer. This fact is supported by the temperature dependence of line c, which disappears above $T_C \approx 35 \pm 5$ K. This Curie temperature was determined by ESR technique. Coincidence of the ESR determined $T_C$ value with one determined by SQUID magnetometer confirms ferromagnetic long ranged nature of this magnetic transition, because in the ensemble of superparamagnetic clusters these temperatures are different and frequency dependent [20].
Figure 3. ESR spectra in the singular (1) and vicinal (2) heterostructure with the GaAs:Mn layer and the reference heterostructure without GaAs:Mn layer (3) at T = 4 K. The dc magnetic field was applied in the (001) heterostructures plane. The error bar is equal to the symbol size.

Figure 4. Photoluminescence spectra at T = 2 K in the: singular (1) and vicinal (2) heterostructures with the GaAs:Mn layer and reference (3) heterostructure without GaAs:Mn layer. The magnetic field B = 3 T was applied perpendicular to the (001) heterostructures plane. The error bar is equal to the symbol size.

This conclusion is also confirmed by the axial anisotropy of line $c$ with the easy magnetization axis lying in the (001) plane, which is consistent with the results of magneto-optical studies [8, 10]. The intensity of line $a$ drastically decreases with increasing temperature, and they are not observed at temperatures above 40 K. Line $a$ locates near zero magnetic field. This line can not correspond to ferromagnetic resonance because 0.06 T anisotropy field determined from the angular dependence of the anisotropic line $c$ should control resonant field of the all ESR lines. Isotropy observed in our experiments indicates another origin of the line $a$. Similar lines were observed in [21] where it was shown that this line corresponds to magnetoresistance. In our opinion, line $a$ is caused by the microwave magnetoresistance [11].

ESR spectra of vicinal heterostructures differ from the ones in singular heterostructures. First, nonresonant line $a$ is not observed in vicinal heterostructures (Figure 3). In vicinal heterostructures quasi-one-dimensional conducting channels are formed [22], while in singular heterostructures the electron gas is quasi-two-dimensional. Therefore, dimension limitations suppress the magnetotransport properties of the GaAs:Mn. Similar results were reported earlier for Ge:Mn films [23].

Second, ESR spectra of the line $c$ in the vicinal sample are broader than spectra in singular samples. Reason of broadening of the line $c$ is the distribution of resonant fields corresponding to separated ferromagnetic GaAs:Mn islands of different sizes and orientations in local crystal fields. The observed ferromagnetic resonance line is the sum of the magnetically inhomogeneous local components. Thus line $c$ confirms the assumption of magnetic disorder in the GaAs:Mn layer in vicinal heterostructures. Because isotropic lines $b, d, e$ were the same in the singular and vicinal heterostructures we used these lines as reference signals.

Very often DMS crystals consist from two magnetic subsystems: dispersed transition metals ions and ferromagnetic secondary phase in the nanoclaster form [19]. The explanation of
ferromagnetism of the GaAs:Mn layer and corresponding a and c lines in ESR spectra by the presence of the known ferromagnetic secondary phase (MnAs or MnGa clusters) can be excluded, because:

- These materials exhibit different ferromagnetic resonance spectra (another lineshape, resonance field, linewidth) [24].
- The Curie temperatures of these alloys ($T_C = 320$ K for MnAs [25] and $T_C = 600$ K for MnGa [26]) are noticeably higher than the Curie temperature $T_C = 35 \pm 5$ K observed in our experiments. It was found that, as the size of crystals decreases Curie temperature, $T_C$, of ferromagnetic nanoclusters or nanofilms usually decreases no more than 3 – 5 times [27]. Estimation of the size effect on the Curie temperature can be performed by formula: $T_C(d) = T_C(\infty)[1-3\Delta L/2d]$ [27], $T_C(d)$ is the Curie temperature of nanoclusters, $d$ is diameter, $T_C(\infty)$ is the Curie temperature of bulk sample, $\Delta L$ is the thickness of surface layer of nanoclusters, which characterizes the influence of the surface layer on the Curie temperature. If we assume reasonable sizes $d = 1$ nm and $\Delta L \sim 0.5$ nm, Curie temperature becomes $T_C = 80$ K in MnAs alloy clusters. This value is noticeably higher than Curie temperature 35 K observed in our heterostructures.

From the other hand, photoluminescence spectra indicate magnetic long range ordering and Curie temperature coinciding with SQUID and ESR data. Since optical experiments can not correspond magnetic ordering in clusters (because wavelength is higher than particles sizes) one can assume these experiments as an evidence of long range magnetic order.

### 3.4. Magneto-optical properties of singular and vicinal heterostructures

Figure 4 shows the photoluminescence spectra of the InGaAs quantum well for singular and vicinal heterostructures with the GaAs:Mn layer and heterostructures with the GaAs:C layer instead of the GaAs:Mn layer. The emission band of the quantum well in heterostructures with the GaAs:Mn layer was split into two circularly polarized components $\sigma^+$ and $\sigma^-$ due to the Zeeman interaction of the electron and hole states (figure 4). Small difference in concentration and correspondent quantum well depth results in energy shift of photoluminescence line (figure 4). This shift doesn’t influence the results of the magnetic experiments, described in this paper.

In all heterostructures circular polarization, $P_C$, was 0 % in zero magnetic field. Position of the sample was not changed during magnetic field application. For that reason the sample in the absence of the magnetic field was considered as reference one. Circular polarization, $P_C$, was 15 % in the 3 T magnetic field at 2 K (figure 5).

![Figure 5. Temperature dependencies of the circular polarization, $P_C$, of a InGaAs quantum well photoluminescence in a singular (1) and vicinal (2) heterostructure with the Mn-doped GaAs layer and with the C-doped GaAs layer instead of the Mn-doped GaAs layer (3). The solid lines are guides for eyes. The error bar (± 1 %) is equal to the symbol size.](image)

On the contrary, the polarization in heterostructures without GaAs:Mn layer was $P_C \approx 0$ % within the experimental error (figure 4). The circular polarization of the InGaAs quantum well
effected by the GaAs:Mn layer magnetization because ferromagnetism results in spin polarization of the electrons and holes [5]. Same results were obtained in similar heterostructures [28]. Magnetic field dependencies of photoluminescence polarization were discussed in paper [28]. In our work we will discuss temperature dependencies of photoluminescence polarization. Temperature dependencies of photoluminescence polarization, $P_C$, (figure 5) qualitatively reproduce the temperature dependencies of GaAs:Mn layer magnetization (figure 2). In the frame of standard two-level Boltzmann model we suppose polarization of charge carriers in the quantum well due to magnetic field generated by GaAs:Mn layer. Temperature dependencies of photoluminescence polarization, $P_C$, (figure 5) are in a good agreement with temperature dependencies of GaAs:Mn layer magnetization (figure 2). Observed differences in magneto-optical data can be explained by the different types of magnetic ordering in GaAs:Mn layer.

4. Summary

Magnetic field of GaAs:Mn layer induces spin polarization of charge carriers in InGaAs quantum well. The polarization of quantum well photoluminescence follows the variations of GaAs:Mn layer magnetization induced by change of temperature or GaAs substrates orientations. Effects of GaAs substrates orientations on magnetization, spin dynamics and photoluminescence in the heterostructures were found. In singular heterostructures temperature dependence of magnetization follows the mean field theory allowing us to extract reasonable parameters of exchange interaction. In vicinal heterostructures temperature dependence of magnetization follows the percolation theory for disordered ferromagnets. Disorder in vicinal heterostructures leads to broadening of ferromagnetic resonance line and varies of magneto-photoluminescence characteristics.

5. References

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