

Circularly Polarized Electroluminescence of InGaAs/GaAs/CoPt Spin Light Emitting Diodes Placed in a Strong and Weak Magnetic Field

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A comparative study of the circular polarization degree dependences on external magnetic field was carried out in spin light-emitting diodes including semiconductor InGaAs/GaAs heterostructure and a magnetic CoPt contact and in control non-magnetic structures with an Au contact. In a weak magnetic field, the magnetic field dependence of electroluminescence circular polarization degree is similar to the magnetic field dependence of magnetization: it represents a hysteresis loop with saturation in a field of ~ 0.3 T. In a strong magnetic field, an additional linear contribution to the circular polarization degree is detected. This contribution is associated with the Zeeman splitting of energy levels. The magnitude of the linear contribution depends on the position of the quantum well relative to the ferromagnet/semiconductor interface. The obtained dependence is associated with the influence of the magnetic field of the inhomogeneously magnetized CoPt electrode on the spin relaxation time of carriers.

Keywords: spin injection, quantum well, Zeeman splitting.

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Introduction

Spin light-emitting diodes based on semiconductor heterostructures $A^{III}B^V$ with a ferromagnetic metal injector are a compact and simple version of a circularly polarized light source operating at room temperature. The operation of such devices is based on the injection of spin-polarized carriers from a magnetized ferromagnetic electrode into a semiconductor heterostructure [1–3]. The parameters of spin injection are determined both by the magnetic properties of the electrode (the degree of spin polarization of carriers, the value of residual magnetization) and by the properties of the injected semiconductor structure (spin diffusion length, density of states at the metal/semiconductor interface) [4]. In addition, a study of InGaAs/GaAs heterostructures with a ferromagnetic CoPt contact in [5] has shown that the circular polarization characteristics of electroluminescence were influenced by the magnetic field of the nonuniformly magnetized contact. The possibility of inverting the sign of the circularly polarized radiation due to the effect of precession of spin-polarized carriers in the transverse magnetic field of the CoPt contact was shown in the cited paper [5].

This paper is dedicated to development of studies of the spin transport effect in an inhomogeneous magnetic field. For this purpose, we measured circularly polarized electroluminescence in a strong magnetic field (up to 5 T), which provides a significant Zeeman splitting of energy levels in the active region (InGaAs quantum well). Under such conditions, the dynamics of radiative recombination is

significantly influenced by the processes of spin relaxation of carriers to lower Zeeman levels. Analysis of these processes allows us to draw additional conclusions regarding the spin precession effect discussed in [5].

1. Experimental procedure

The semiconductor diode structure was formed by MOSVD at atmospheric hydrogen pressure [5,6]. The structure was three $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ quantum wells (QWs) of 10 nm width with different composition ($0.1 < x < 0.25$, to separate luminescence from each (QW) by wavelength) located at different distances from the surface (d_s). A scheme of the structure is shown in fig. 1. The distance between the quantum wells was 30 nm, thus the d_s value for each quantum well was also different (20, 60 and 100 nm respectively). The ferromagnetic injection contact to the semiconductor structure was formed at the next technological stage by electron-beam evaporation in vacuum. The contact was a two-layer 8 nm CoPt/(1 nm Al_2O_3), structure in which the magnetic CoPt layer is the source of spin-polarized carriers and the tunnel-thin Al_2O_3 layer is necessary to enhance the spin injection efficiency. The method of production is described in detail in [5,6]. Injection non-magnet Au/ Al_2O_3 -contacts were formed as reference ones. The formation of spin light-emitting diodes (SLEDs) was completed by creating mesastructures of 500 μm diameter by photolithography and chemical etching techniques.

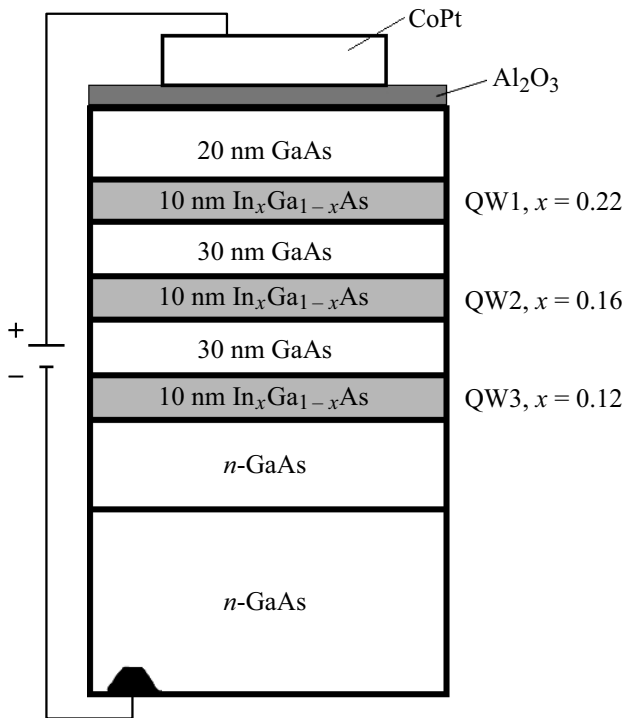


Figure 1. Scheme of studied diode structure with three quantum wells.

The magnetic properties of the CoPt films were analyzed by measuring the magnetic field dependence of the Hall EMF, which, according to [7], is proportional to the magnetization of the film. The measurements were performed in a magnetic field up to 0.3 T at 10 K. The circular polarization of the recombination radiation from produced spin light-emitting diodes (SLEDs) and reference diodes with nonmagnetic contact were also studied in this paper. For this purpose, a forward constant bias was applied to the specimens, resulting in the excitation of electroluminescent radiation in the emission region of the InGaAs core (880–960 nm), which was recorded from the side of the GaAs [5,6] substrate transparent for this range. Specimens were introduced into an external perpendicular magnetic field to produce a circularly polarized radiation component. The degree of circular polarization was measured using the standard [1] technique and calculated by the formula

$$P_{EL} = (I^+ - I^-)/(I^+ + I^-), \quad (1)$$

where I^+ — the intensity of electroluminescence (EL) polarized along the right circle, I^- — the intensity of EL polarized along the left circle.

The studies were performed in two magnetic ranges: in small magnetic fields from 0 to 0.3 T at 10 K in a Janis CCS-300S/202 closed-loop cryostat; in magnetic fields up to 5 T at 2 K in a helium flow cryostat with a superconducting solenoid. In the first case, a MDR-23 monochromator and a FEU-82 photomultiplier tube were used to record the EL spectra. An MDR-23 monochromator was also used for

measurements in the range of strong magnetic fields, and the intensity was recorded with a linear Hamamatsu CCD detector.

2. Results

In accordance with previously obtained results, electroluminescence emission is observed in the forward bias mode of the studied diodes, which is due to the injection of minority carriers (in our case, holes) from the metallic electrode [8]. The main mechanism of injection is the direct „throwing“ of holes into the valence zone by shifting the Fermi level in the metal; other mechanisms of injection, including tunneling of holes from the metal into a quantum well, are much less effective [8]. Fig. 2 shows magnetic field dependences of Hall EMF and degree of diode EL circular polarization in a luminescence region closest to surface QW $d_s = 20$ nm (curves 3 and 1 respectively). The above dependences are similar to each other and describe a hysteresis loop, the saturation of P_{EL} /Hall EMF in a magnetic field above 0.15 T is related to the saturation of the CoPt layer magnetization. The similarity of the magnetic field dependences of Hall EMF and degree of polarization allows us to relate the latter to the injection of spin-polarized carriers from the magnetized CoPt electrode [1–5]. For the reference structure, the magnetic field dependence of circular polarization degree can be described by a linear function, and the highest value of P_{EL} for it (at maximum magnetic field) is much lower (curve 2).

In the range of magnetic fields up to 5 T for the studied magnetic diodes there is an additional field-linear increase in the degree of circular EL polarization, which on the scale of 0–0.3 T is practically not noticeable (fig. 2–4). It was found that the slope of the linear part of $P_{EL}(B)$ depends

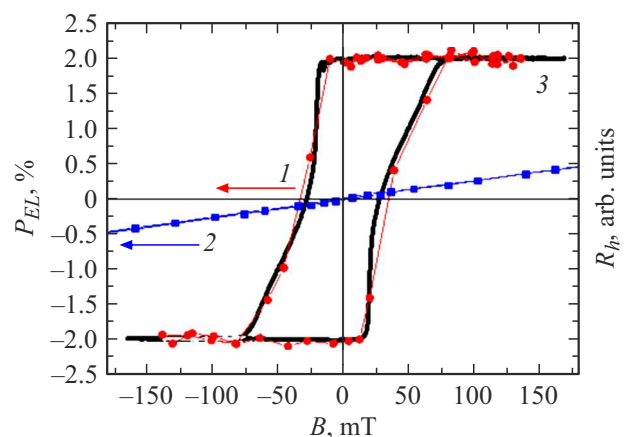


Figure 2. Magnetic field dependences of Hall resistance (curve 3) for CoPt layer used as contacts, and degree of circular EL polarization for the studied structure with CoPt-contact (curve 1) and reference structure with Au-contact (curve 2) in the spectral region corresponding to the luminescence nearest to surface QW (wavelength 960 nm). The measurement temperature was 10 K, the diode current — 10 mA.

most significantly on the type of contact and the value of d_s . Thus, fig. 3,*a* shows the magnetic field dependences of P_{EL} in the range 0–5 T, measured in the radiation region closest to surface QW ($d_s = 20$ nm) for specimens with CoPt contacts and reference specimens with Au contact (curves 1 and 2 respectively). In the low magnetic field region, the circular polarization degree of the Au-contact structure is much lower than that of the CoPt-contact structure (fig. 3,*b*). As the magnetic field increases, for the reference structure there is a linear increase of P_{EL} in both cases, nevertheless the slope of the linear dependence is higher for the structure with Au contact, which leads to a higher value of polarization degree for it in the 5 T field.

For the second and third QWs located at a distance of 60 and 100 nm from the surface (fig. 1), respectively, a similar situation is registered, but the divergence of the dependences $P_{EL}(B)$ in a strong magnetic field for Au- and CoPt-contacts is reduced as compared to the first QW (fig. 4, it a and 5, it a, respectively). The difference in

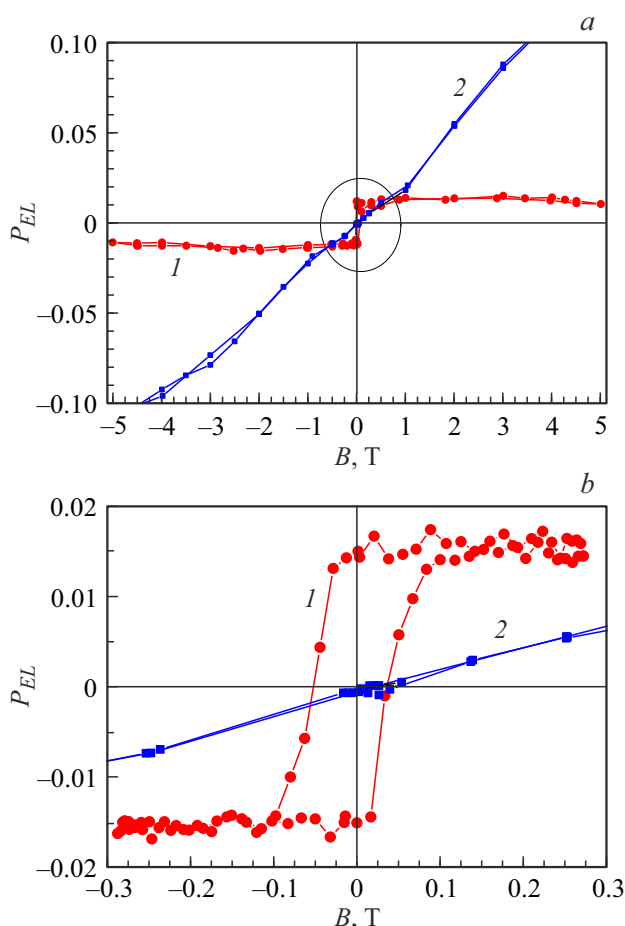


Figure 3. Magnetic field dependences of circular polarization degree measured at 2 K in a helium cryostat for the studied structure with CoPt-contact (curve 1) and reference structure with Au-contact (curve 2) in the spectral region corresponding to the luminescence nearest to surface QW (wavelength 960 nm). Region of strong fields up to 5 T (*a*) and region of small magnetic fields up to 0.3 T (*b*).

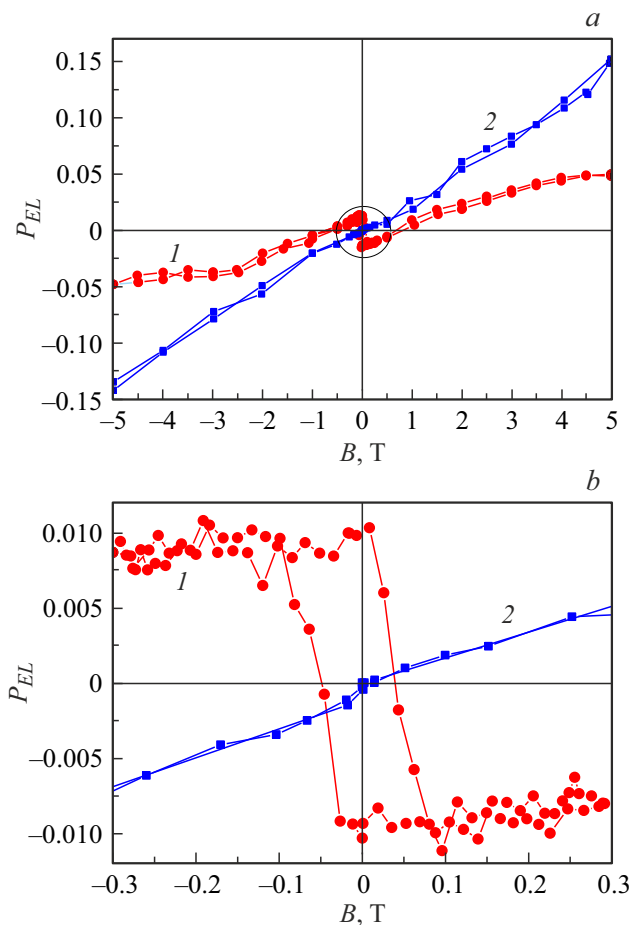


Figure 4. Magnetic field dependences of circular polarization degree measured at 2 K in a helium cryostat for the studied structure with CoPt-contact (curve 1) and reference structure with Au-contact (curve 2) in the spectral region corresponding to the luminescence second to surface QW (wavelength 910 nm). Region of strong fields up to 5 T (*a*) and region of small magnetic fields up to 0.3 T (*b*).

the hysteresis dependence in fig. 3,5 and 4 (curve 1, for P_{EL} from different QWs) is due to the spin inversion discovered earlier in [5] due to precession in the CoPt contact magnetic field, which will be discussed below.

The closest values of polarization degree for Au- and CoPt-contact structures were obtained for the most distant QW from the metal/semiconductor boundary (fig. 5). But even in this case the value of P_{EL} in magnetic field 5 T for structure with Au-contact exceeds that for structure with CoPt-contact.

3. Discussion

Let us proceed to discuss the experimental results obtained. According to a series of papers [1–6,9], the partial circular polarization of EL in structures with CoPt contact can be caused by two factors:

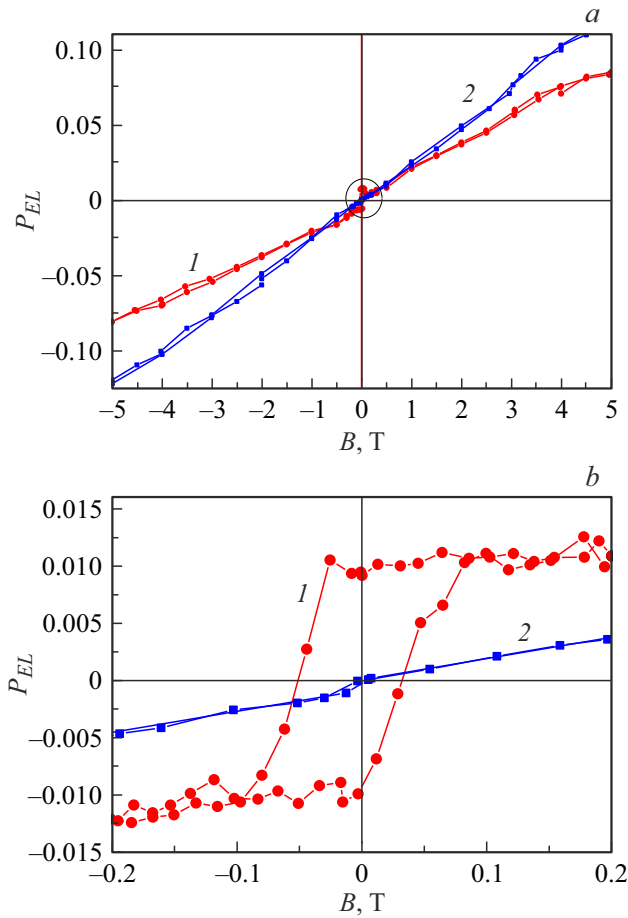


Figure 5. Magnetic field dependences of circular polarization degree measured at 2 K in a helium cryostat for the studied structure with CoPt-contact (curve 1) and reference structure with Au-contact (curve 2) in the spectral region corresponding to the luminescence 3rd to surface QW (wavelength 880 nm). Region of strong fields up to 5 T (a) and region of small magnetic fields up to 0.3 T (b).

1) injection of spin-polarized carriers from a magnetized ferromagnetic electrode into the active region of the light-emitting structure;

2) Zeeman splitting of energy levels in a strong magnetic field.

The first factor depends on the magnetization of CoPt electrode and is responsible for EL radiation polarization of the studied structure in a low magnetic field. Zeeman splitting of the levels causes a field-linear change in the polarization degree in the magnetic field when the ferromagnetic electrode is already magnetized to saturation, and additional spin polarization of the carriers is associated with their relaxation to the lower Zeeman levels [9]. In the Au-electrode structure there is no spin injection (this accounts for the low value of P_{EL} in a weak magnetic field), and the circular polarization observed in the experiment is related only to Zeeman splitting of energy levels and relaxation efficiency of nonpolarized carriers to these levels.

To assess contribution of various mechanisms to the recorded value of circular polarization, let us consider the dynamics of charge carriers in the QW. To this end, we will write down the kinetic equations for the charge carriers distributed at the levels of QWs obtained earlier for similar structures in [10,11]. For a simple qualitative analysis, it is sufficient within this paper to consider a QW with a single spin-split energy level and only one type of charge carrier. For certainty, we consider heavy holes, since it is the spin-polarized holes that are injected from the CoPt contact in the chosen diode configuration [5], and the light hole levels „are displaced“ from the InGaAs QW under the action of elastic stresses [12]. Polarization of the electrons injected into the active region from the n -GaAs substrate is due only to the Zeeman splitting of the levels; taking this polarization into account will not affect the qualitative picture under consideration.

According to [10,11], variation of carrier concentration at levels with different spin is governed by the following equation:

$$\frac{dn_1}{dt} = G_1 - \frac{n_1}{\tau_R} - \frac{n_1}{\tau_s} + \frac{n_2}{\tau_s} \exp\left(-\frac{\Delta E}{kT}\right), \quad (2)$$

$$\frac{dn_2}{dt} = G_2 - \frac{n_2}{\tau_R} + \frac{n_1}{\tau_s} - \frac{n_2}{\tau_s} \exp\left(-\frac{\Delta E}{kT}\right). \quad (3)$$

Here n_1 — concentration of holes with spin $3/2$, n_2 — concentration of holes with spin $-3/2$. The first member in equations (2), (3) describes generation of carriers in QW with speed of $G_{1,2}$, which varies for different spins. The only channel of generation is relaxation of carriers in QW from GaAs-barrier. In the mode of spin injection from GaAs-barrier, polarized spin-injected carriers from CoPt are injected into the QW, so in the first approximation we can write:

$$\frac{G_1 - G_2}{G_1 + G_2} \approx P_{inj}; \quad G_1 - G_2 \approx P_{inj}(G_1 + G_2). \quad (4)$$

The second member in equations (2), (3) is responsible for radiative recombination with time τ_R (recombination lifetime), the third and fourth members describe the spin relaxation to the lower Zeeman level with characteristic time τ_s (spin relaxation time). Other constants in equations (2), (3): k — Boltzmann constant, T — temperature, ΔE — value of Zeeman splitting of heavy hole level, P_{inj} — component of the circular polarization degree associated with the injection of spin-polarized carriers from the CoPt contact.

In stationary experimental conditions at constant excitation

$$\frac{dn_1}{dt} = \frac{dn_2}{dt} = 0. \quad (5)$$

Solutions to the system of equations (2), (3) with conditions (4) and (5) allow us to write down expressions for the degree of spin polarization of the holes:

$$P_{EL} = \frac{n_1 - n_2}{n_1 + n_2} = \frac{P_{inj} + \frac{\tau_R}{\tau_s}(1 - \exp(-\frac{\Delta E}{kT}))}{1 + \frac{\tau_R}{\tau_s}(1 + \exp(-\frac{\Delta E}{kT}))}. \quad (6a)$$

The obtained equation (6a) allows us to qualitatively estimate the influence of the system parameters on the final polarization pattern under Zeeman splitting conditions. Note that the value P_{inj} can enter into equation (6a) with both positive and negative sign. Let us consider the extreme cases for equation (6a).

1) Case $\tau_R \gg \tau_s$ corresponds to the situation of very short spin relaxation time. In case of high Zeeman splitting of levels compared to kT , equation (6a) may be rewritten as

$$P_{EL} \approx \frac{\frac{\tau_R}{\tau_s}(1 - \exp(-\frac{\Delta E}{kT}))}{\frac{\tau_R}{\tau_s}(1 + \exp(-\frac{\Delta E}{kT}))} = \frac{1 - \exp(-\frac{\Delta E}{kT})}{1 + \exp(-\frac{\Delta E}{kT})}, \quad (6b)$$

and if $\Delta E \gg ikT$, then $P_{EL} = 1$. For the long recombination time, complete relaxation along the spin of the charge carriers succeeds, as a result, the information about polarization degree of injected carriers is lost, and polarization is caused only by the Zeeman splitting.

2) Case $\tau_R \ll \tau_s$ corresponds to the situation of very long spin relaxation time. In this case the carriers in QW have no time to relax to the lower Zeeman level before recombination, and equation (6a) is rewritten as:

$$P_{EL} \approx P_{inj}, \quad (6c)$$

i.e., the degree of circular polarization is determined only by the injection of spin-polarized carriers from the magnetized ferromagnetic electrode. Equation (6c) does not depend on the value of Zeeman level splitting.

3) Note also that if the splitting value $\Delta E \ll kT$ is low, $P_{EL} = P_{inj}/(1 + \tau_R/\tau_s)$. This corresponds to the situation observed in a weak magnetic field. Such a case is considered, for example, in paper [13].

Under experimental conditions, there is probably an intermediate situation for which the values of τ_R and τ_s are of the same order. In this case, the final polarization is determined by a combination of factors. Thus, the parameter in formula (6a), which differs for the studied and reference structures, is the value of P_{inj} . For the reference structure with a nonmagnetic Au contact, the value of $P_{inj} = 0$, while for the studied structures with CoPt this value can be either greater or less than zero, which can cause the difference in the values of P_{EL} in a weak magnetic field. However, in the field corresponding to the saturation of CoPt magnetization, the value of P_{inj} stops varying (since it is uniquely related to contact magnetization), and the difference in values of P_{EL} varies in the entire range of magnetic fields, and in the maximum field available is a value up to 0.1 (fig. 3, a). Consequently, the difference in the value of P_{inj} cannot explain the experimentally recorded difference between the polarization degree values of the studied and reference structures.

Another parameter determining the recorded value of the circular polarization degree is the ratio τ_R/τ_s , which, generally speaking, may also differ for the studied and reference structures. The fact that in magnetic field 5 T

value $P_{EL}(\text{Au}) > P_{EL}(\text{CoPt})$, may be caused by

$$\frac{\tau_R}{\tau_s}(\text{Au}) > \frac{\tau_R}{\tau_s}(\text{CoPt}). \quad (7)$$

Since electroluminescence intensities for the studied and reference structures are at the same level, and the QW in both structures is identical, it can be assumed that the radiative lifetime for structures with Au and CoPt contacts is the same ($\tau_R(\text{Au}) \approx \tau_R(\text{CoPt})$). Consequently, in order to meet inequation (7), it is necessary that

$$\tau_s(\text{Au}) < \tau_s(\text{CoPt}), \quad (8)$$

i.e., the Au-contact structure is characterized by a lower spin relaxation time as compared to the CoPt-contact structure. Indeed, in this case in the Au-contact structure it has time to establish distribution of carriers on Zeeman levels, which is close to equilibrium, while in the CoPt structure polarization is more determined by spin injection (it is most clearly seen in fig. 3).

Presumably, the cause for inequation (8) is built-in magnetic field of CoPt contact. Earlier in [5] we demonstrated that the magnetized CoPt contact with inhomogeneous composition is the source of a magnetic field whose parallel component causes the precession of spin-polarized carriers in the spin injection mode. Spin precession, in particular, causes the inversion of the P_{inj} sign in the weak magnetic field (such inversion is observed for the second QW in the studied structure, as shown in fig. 4). It can be assumed that the component of this inhomogeneous field perpendicular to the plane of the CoPt film may also influence the spin relaxation processes.

In particular, it is known that the spin relaxation time according to the Diakonov–Perel mechanism increases in the magnetic field [14]. τ_s increases regardless of the direction of the magnetic field. Suppression of the Diakonov–Perel mechanism by magnetic contact field increases the total spin relaxation time, which is observed in the experiment.

Maximum amplitude of internal magnetic field is achieved at the minimum distance from the CoPt electrode. This explains the greatest differences between the P_{EL} values for the studied and reference structures exactly for the first QW. The more distant from the contact, the lower the field amplitude, and the less the influence on τ_s . For the most distant QW from the surface, the values of the polarization degree in the studied and reference structures are close to each other.

Note also that the effect of the perpendicular component of the internal contact magnetic field also affects the value of the ΔE Zeeman level splitting. However, unlike the parameter τ_s whose value increases in any direction of the contact magnetic field, the change in the value of ΔE depends on the field direction: if the internal contact magnetic field is in the same direction with the external one, ΔE and the polarization degree increase, if it is opposite (i.e. it compensates the external field) — both ΔE and P_{EL} decrease. With random distribution of

magnetic heterogeneities in the CoPt contact, the contact field direction should vary from a point to a point, and the value averaged over the contact area will be zero. In a similar system, the effect linear by magnetic field from change of ΔE is absent [5], therefore, modulation of Zeeman splitting cannot account for the significant difference in the degree of circular polarization observed in the experiment for the magnetic and reference structures.

Conclusion

In conclusion, we note that a comparative analysis of the $P_{EL}(B)$ dependences for light-emitting diodes with magnetic (CoPt) and nonmagnetic (Au) contact in the region of small magnetic fields allows us to identify the contribution of spin injection from CoPt to the registered circular polarization of EL. In the region of strong magnetic fields (when Zeeman splitting of levels becomes essential), comparison of the same structures has revealed the influence of the spin relaxation time of the carriers τ_s on the degree of circular polarization. In particular, for the CoPt structure, the above parameter is modulated by the magnetic field of the inhomogeneously magnetized contact, resulting in a lower degree of circular polarization as compared to the reference structure. Thus, the analysis of the static parameters of the spin light-emitting diodes ($P_{EL}(B)$ dependence under constant exposure) allows us to draw qualitative conclusions about the kinetic properties of the system (spin relaxation time τ_s). This conclusion seems valuable from the methodological point of view to assess the influence of the internal magnetic field of the ferromagnetic layer on the spin injection processes.

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Conflict of interest

The authors declare that they have no conflict of interest.

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