JOURNAL OF EXPERIMENTAL AND THEORETICAL PHYSICS, 2024, Vol. 165, No. 6, pp. 828–836

SOLIDS AND LIQUIDS

FEATURES OF POLARIZED LUMINESCENCE OF AN INHOMOGENEOUS ENSEMBLE OF LOCALIZED EXCITONS

© 2024 L. V. Kotova*, V. P. Kochereshko

Ioffe Institute, 194021, St. Petersburg, Russia

**e-mail: kotova@mail.ioffe.ru*

Received February 02, 2024

Revised February 12, 2024

Accepted February 15, 2024

Abstract. The paper considers the features of polarized luminescence induced by a magnetic field in an ensemble of localized excitons. It was found that: 1) in an inhomogeneous ensemble, the splitting of photoluminescence bands in a magnetic field in the right and left circular polarizations may exceed the value of the Zeeman splitting of individual excitons in the ensemble by orders of magnitude; 2) the lower photoluminescence band in terms of energy may have a lower intensity than the upper one, at first glance contradicting the Boltzmann energy distribution; 3) the sign of the circular polarization of photoluminescence may vary along the contour of the radiation band. It is shown that in an inhomogeneous ensemble, all these features are explained by the pendence of the exciton g-factor on its localization energy.

Keywords: *polarized photoluminescence; quantum inhomogeneous ensemble; localized excitons*

DOI: 10.31857/S004445102406e087

1. INTRODUCTION

Magnetic circular polarization of luminescence (MCPL) is an effective method for studying the spin orientation of excitons and carriers in crystals, which is related to the thermal redistribution of carriers and excitons between Zeeman sublevels in a magnetic field. This method was successfully used by Thomas and Hopfield to study bound excitons [1]. Using this method, the fine structure of acceptor impurities was investigated and the acceptor concentration profile was measured [2] in quantum wells. This method also proved to be very effective for studying spin dynamics of charge carriers and excitons [3–5] in nanostructures.

As is known, in a magnetic field, all states split according to the projection of the magnetic moment on the magnetic field direction. Under non-resonant unpolarized optical excitation, these states are populated according to the Boltzmann distribution. The population ratio of these states is determined by the Zeeman splitting value and the temperature factor.

As a result, photoluminescence (PL) becomes circularly polarized. The degree of polarization of this radiation is determined by the population of Zeeman sublevels and a coefficient that accounts for the absence of complete thermodynamic equilibrium.

In strong magnetic fields, the degree of polarization ceases to depend on the magnetic field and is completely determined by the ratio of lifetime to spin relaxation time. In weak magnetic fields, the degree of polarization is proportional to the magnetic field magnitude. This dependence is often used to determine the g-factor and the ratio of lifetime to spin relaxation time. This model describes experimental results well in many cases [5]. However, in some cases, experimental results do not fit into such a simple scheme (see, for example, works [6–8]).

As a rule, emission spectra show not individual objects, but entire ensembles of such objects. This situation occurs during exciton localization on deep levels in bulk crystals [9] or on composition fluctuations in solid solutions [10], on quantum

well thickness fluctuations or quantum wire cross-sectional area, on superlattice interfaces [13], as well as during dimensional quantization of excitons in quantum dots [14–16] or colloidal nanocrystals [17–19], taking into account the variation in their sizes and shapes. Their emission lines under real conditions almost always experience inhomogeneous broadening associated with fluctuations in resonant energies, center concentration, spatial position, or variation in other parameters that determine the energy and width of individual emission lines.

In ensembles of localized states, the MCPL signal can take various forms. Sometimes "inverse population" [20] of Zeeman sublevels is observed, sometimes — non-monotonic dependence of polarization degree [21] on magnetic field, and sometimes — dependence of the exciton g-factor on magnetic field, even with sign reversal. In some cases, giant splitting of emission bands in two polarizations is observed [21,22], while in other cases, conversely, there is no Zeeman splitting despite significant circular polarization of emission [23].

To explain such diversity of MCPL manifestations, various models were proposed, mainly based on the dependence of exciton and carrier capture rate on the distribution of localizing centers [5, 19, 21, 24]. However, some MCPL manifestations cannot be explained within these models, for example, the giant splitting of PL band maxima in two circular polarizations [22].

In quantum dots, unlike, for example, localized excitons on quantum well width fluctuations, there is no exciton migration between dots. This greatly simplifies the analysis of MCPL spectra, as it does not require introducing a poorly known additional parameter related to exciton migration.

The magnitude of level splitting in magnetic field is determined by g-factor. Quantization of excitons and carriers in nanostructures can lead to changes in their g-factors. For electrons, this change is due to spin-orbital interaction, as was first shown in [25] and confirmed by numerous experiments [26]. The hole g-factor is also sensitive to the shape of the quantization potential, as shown in [27, 28]. For excitons [29, 30], an additional contribution to the g-factor appears, caused by the motion of the exciton center of mass.

This paper discusses a model describing some unusual features of MCPL spectra caused by inhomogeneous emission line widths. The considered model takes into account the dispersion of g-factors of holes, electrons, and excitons in an ensemble of quantum dots of different sizes.

This model does not negate the possible dependence of lifetime, capture probability, and spin relaxation time on exciton localization energy. However, it explains some observed features of emission spectra under conditions of inhomogeneous line broadening. Obviously, the model can be useful not only for describing MCPL in quantum dot ensembles but also for describing polarized PL of impurity centers and excitons under conditions of inhomogeneous broadening of their emission bands.

2. RESULTS

As is known, in a magnetic field, all states split according to the projection of the magnetic moment on the direction of the magnetic field. Under non-resonant, unpolarized optical excitation, these states are populated according to the Boltzmann distribution. The population ratio of these levels is determined by the magnitude of Zeeman splitting ΔE and the temperature factor kT. or a two-level system under equilibrium conditions, this ratio of sublevel populations is described by the relation

$$n_2 n_1 = \exp\left(-\frac{\Delta E}{kT}\right), \quad (1)$$

where n_1 and n_2 are exciton concentrations at the sublevels, $\Delta E(B) = \mu g B$ is the magnitude of Zeeman splitting between sublevels, k is the Boltzmann constant, g is the g-factor, B is the magnetic field, μ is the Bohr magneton.

Radiation from these states has right σ^+ or left σ^- circular polarization depending on the sign of the angular momentum projection on the field direction. The emission line intensities are proportional to the level populations. The degree of polarization equals

$$P_{circ} = \frac{I_{s^+} - I_{s^-}}{I_{s^+} + I_{s^-}}, \quad (2)$$

where $I_{\sigma+}$ and $I_{\sigma-}$ are radiation intensities in right and left circular polarizations.

The splitting of emission lines equals the magnitude of Zeeman splitting of levels ΔE, and the intensity ratio is determined by the Boltzmann

factor. In cases where the emission line width is greater than the magnitude of Zeeman splitting and exchange interaction in the exciton, the degree of polarization at PL line maxima equals

$$P_{circ} = \tau_0/(\tau_0 + \tau_s)\Delta E/2kT, \quad (3)$$

where τ_0 is the lifetime, τ_s is the spin relaxation time, $\tau_0/(\tau_0 + \tau_s)$ is a factor accounting for the fact that complete equilibrium is not reached at finite lifetime.

2.1 Polarized luminescence of an inhomogeneous ensemble

In polarized luminescence experiments, inhomogeneous ensembles of states usually participate. This especially applies to excitons in an ensemble of quantum dots, where there is a large spread of excitonic resonance energies. In this case, polarized luminescence acquires certain features.

For example, let's consider an ensemble of quantum dots. We assume that the excitonic luminescence lines from each single dot have δ-shaped form

$$L(E, E_0) \propto n(E)\delta(E - E_0), \quad (4)$$

where E is the energy of excitonic resonance in a single quantum dot, $n(E)$ is the population of this dot, $L(E)$ is the PL line shape in a single dot.

In an ensemble of quantum dots, there can be dots of different sizes. The size dispersion of E dots leads to a spread in energies of exciton resonances. We assume that the distribution of resonant energies in the ensemble has a Gaussian form:

$$G(E) \propto \exp\left[-\frac{E - E_0}{w}\right]^2, \quad (5)$$

where E_0 the most probable exciton energy in the ensemble, and is the width of this distribution.

Then the luminescence band shape of the quantum dot ensemble represents a convolution of δ-shaped luminescence lines from each quantum dot and the Gaussian distribution:

$$I(E_0) = \int_{-\infty}^{\infty} G(E)L(E, E_0)dE. \quad (6)$$

As a result, we obtain

$$I(E_0) \propto \exp\left[-\frac{E_0 - E_0}{w}\right]^2 n(E_0). \quad (7)$$

In a magnetic field, states split in energy according to the projection of the magnetic moment on the magnetic field direction. The energies of these states in a magnetic field have the form

$$E_0 \to E_0 \pm 1/2\mu B g_{eff}(E_0). \quad (8)$$

Here we assume that the effective g-factor g_{eff} depends only on the exciton energy in the quantum dot and does not depend on the dot shape. The dependence of the g-factor on the dot shape will be discussed in section 3.

Split levels will be populated according to the Boltzmann distribution. As a result, for the emission line of the quantum dot ensemble in a magnetic field, we obtain

$$I^{\pm}(E_0) \propto \exp\left[-\left(\frac{E_0 - E_0}{w}\right)^2 \pm \frac{\Delta E(E_0)}{2kT}\right]. \quad (9)$$

In a magnetic field, the PL band splits into two, $I^+(E_0)$ and $I^-(E_0)$, manifesting in two circular polarizations, σ^+ and σ^-, at a given magnetic field direction.

For simplification of formulas, let's assume $E_0 = 0$. The position of emission band maxima is determined from the solution of the equation

$$\left[E_0 \pm \mu B g_{eff}(E_0)\right]' \propto \left[1 \pm \mu B g_{eff}(E_0)dE_0\right]^n$$

$$nw^2/2kT\mu B g_{eff}(E_0)dE_0 = 0. \quad (10)$$

This equation may have several roots. Consequently, the band shape will differ from Gaussian and may have several maxima.

The bands intersect when $I^+ = I^-$, i.e., when the Zeeman splitting value $\Delta E(E_0)$ in the exponential power of equation (9) becomes zero. At these points, the polarization $P_{circ}(E_0)$ changes sign.

Let's consider three possible cases of the exciton g-factor dependence on the exciton quantization energy.

2.2 The value of *g*-factor in the ensemble does not depend on energy

Let g_{eff} not depend on energy E':

$$g_{eff} = g_0.$$

Then

1. The maxima of emission bands in two circular polarizations are located at energies $E^{\pm}_{max} = \pm \mu g_0 B / 2$.

2. The ratio of these band amplitudes equals $e^{\frac{\mu B}{kT} g_{eff}}$.

3. The more intense emission band is lower in energy than the less intense one. This is consistent with the Boltzmann distribution of excitons across sublevels.

4. The half-width of both bands is equal to $2w\sqrt{\ln 2}$.

5. The bands intersect at point $E' = w^2 / 2kT$.

Since $w \gg kT$, the intersection point is far from the band maxima.

6. The degree of polarization does not change sign along the emission band contour.

The spectral dependence of emission intensity in two circular polarizations is shown in Fig. 1. In this case, the choice of values E_0 does not play a role, only the ratios of values $\mu gB / w$ and kT / w are important. The calculation parameters are as follows:

$$w = E_0, \quad 10\mu gB = kT, \quad B = 5T.$$

Since the choice of does not affect anything, in Fig. 1 let's assume $E_0 = 1.0$.

2.3 The value of the *g*-factor depends linearly on energy

Let's assume that g_{eff} depends linearly on energy, i.e.,

$$g_{eff}(E') = g_0 + \tilde{g}E'.$$

Let's estimate the value

$$\mu Bd\tilde{g}_{eff}(E')dE'.$$

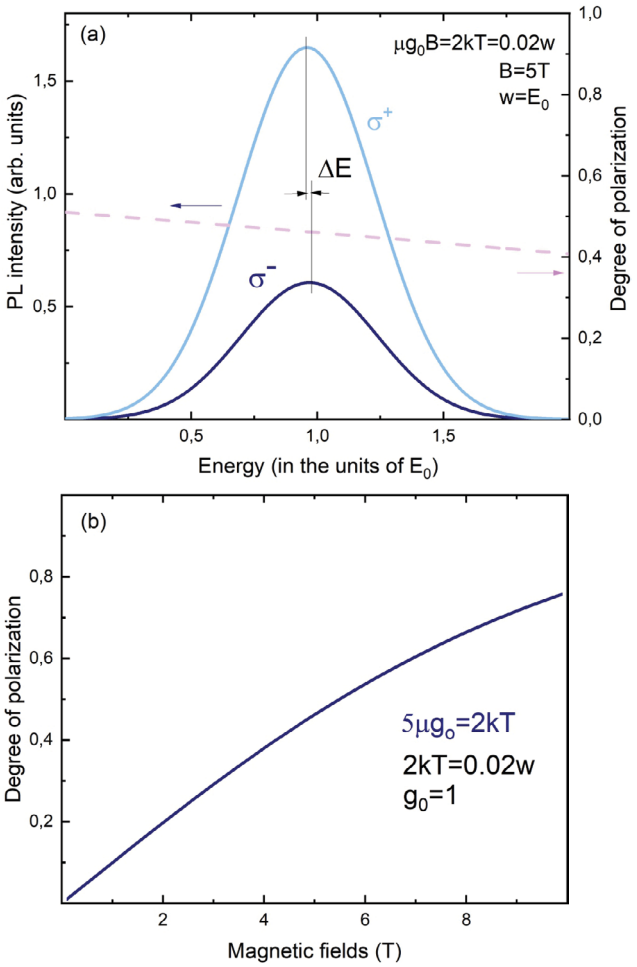

Fig. 1. *a* – Emission spectrum of quantum dot ensemble in two circular polarizations, σ_+ and σ_-, in a fixed magnetic field as a function of exciton quantization energy in the dot (in units of w in equation (5)) assuming that the g-factor does not depend on exciton quantization energy (solid lines). Calculation parameters: $w = E_0$, $\mu gB = kT$, $B = 5$ T. Degree of circular polarization induced by magnetic field as a function of exciton quantization energy (dashed line). *b* – Spectrum-integrated dependence of polarization degree on magnetic field

In the region of 10 T

$$\mu Bdg_{eff}(E')dE' \approx 0.053 \cdot 10\tilde{g}.$$

We assume $\mu B \ll w$. For non-magnetic materials, it is reasonable to assume that g_{eff} can vary no more than from -10 to 10 across the emission band width. Then

$$\mu Bdg_{eff}(E')dE' \ll 1$$

and in expression (10) this term can be neglected compared to unity. From this we obtain the following.

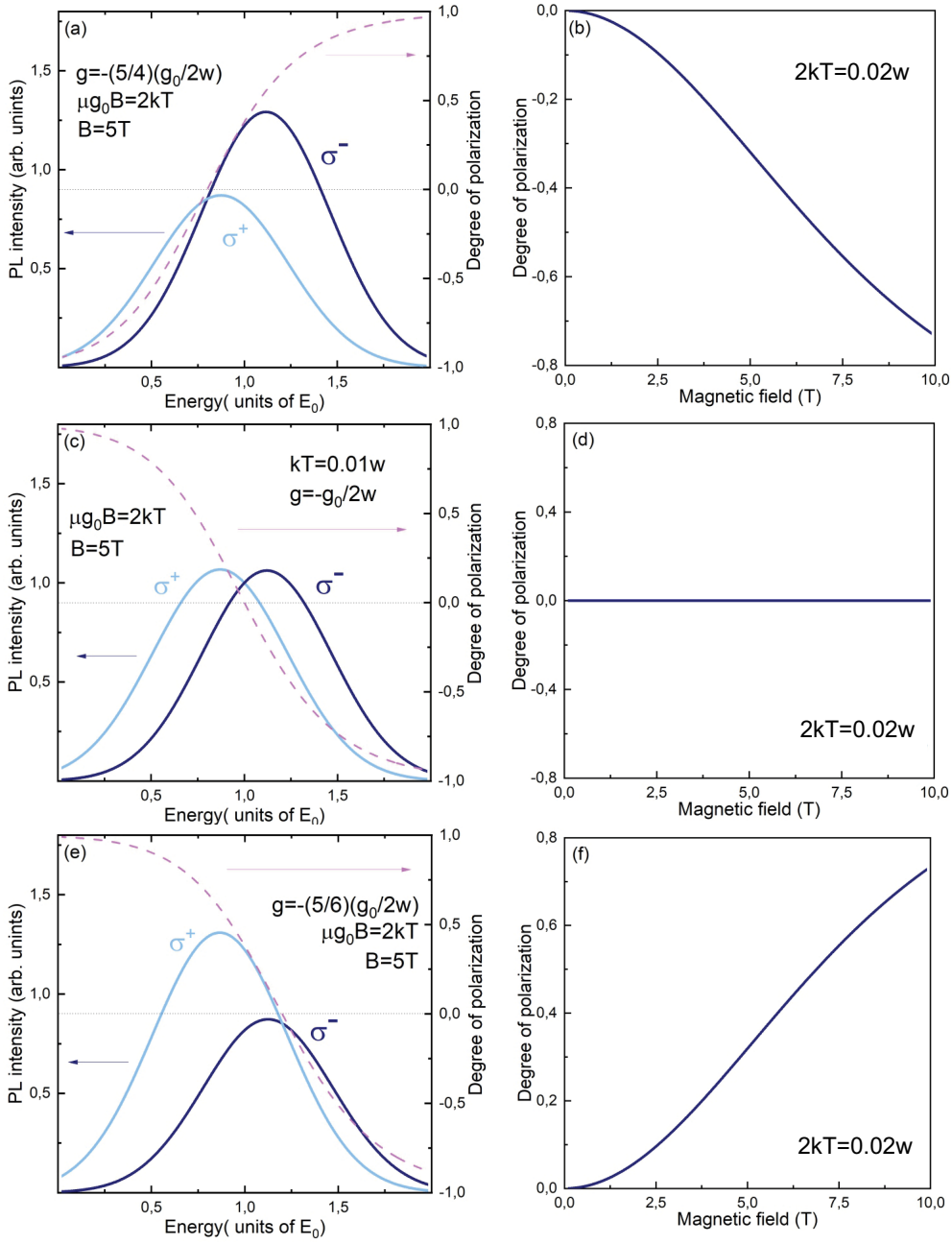

Fig. 2. Emission spectra of quantum dot ensemble in two circular polarizations at fixed magnetic field as a function of exciton quantization energy (in units of w in equation (5)) assuming that g-factor depends linearly on exciton quantization energy (solid lines). Degree of circular polarization induced by magnetic field as a function of exciton quantization energy (dashed lines). Calculation parameters: $\mu g_0 B = 2k\mathrm{T} = 0.02\mathrm{w}$, $\mathrm{g} = -54\mathrm{g}_0 2\mathrm{w}$ (*a*), $\mathrm{g} = -\mathrm{g}_0 2\mathrm{w}$ (*c*), $\mathrm{g} = -56\mathrm{g}_0 2\mathrm{w}$ (*e*). *b*, *d*, *f* – Corresponding spectrum-integrated dependencies of polarization degree on magnetic field

1. The emission band maxima in two circular polarizations are at energies

$$E^{\pm}_{\max} = \frac{\mu B\left(\mu B g_0 g \pm \tilde{g}_0 \pm \dfrac{w^2}{2kT}\tilde{g}\right)}{(1 \pm \mu B\tilde{g})^2}. \qquad (11)$$

Note that the ratio $w^2 g / kT$ can reach values ~ 10^2–10^3. Consequently, approximately $E^{\pm}_{\max} \approx \pm mB\dfrac{w^2}{2kT}g$. In this case, the splitting of emission bands of the quantum dot ensemble can be very large compared to the Zeeman splitting of exciton emission lines in a single quantum dot.

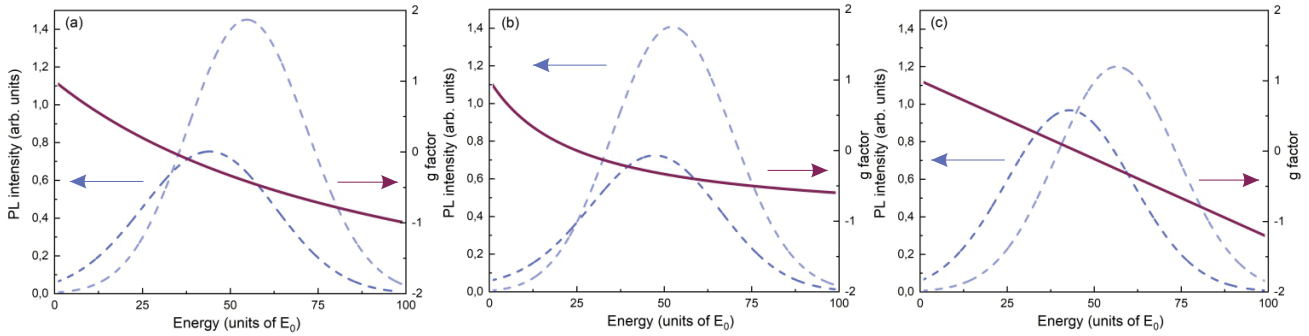

Fig. 3. Emission spectra in two circular polarizations σ_+ and σ_- (dashed curves). Calculated dependencies of g-factor on exciton quantization energy in localizing potential (13) (solid curves) for different cases: $b(E')$ is independent of quantization energy, pendent of quantization energy, $b(E') = 0.5E_0$, $R(E') = 0.25E'$ (*a*); $R(E')$ is independent of quantization energy, $b(E') = 0.5E'$, $R(E') = 0.25E_0$ (b); $b(E')$ and $R(E')$ is independent of quantization energy, $b(E') = 0.45E_0$, $R(E') = 0.45E_0$ (*c*)

2. The ratio of emission line amplitudes is about $(\mu Bgw / kT)^2$.

3. The amplitudes of these lines are

$$I^{\pm}_{\max} \propto \exp\left\{-\frac{\left(E^{\pm}_{\max}\right)^2 n\dfrac{\mu Bw^2 g_{eff}(E^{\pm}_{\max})}{2kT}}{w^2}\right\}. \quad (12)$$

It can be seen that the ratio of band amplitudes depends on the sign of g-factor at the band maximum energy.

2.4. Realistic dependence of g-factor on energy

Let's assume that g_{eff} determined by the formula

$$g_{eff}(E) = g_0 + aE\phi(E\phi) + R(E\phi). \quad (13)$$

Here E' is the exciton quantization energy in the localizing potential, g_0, a, b and R are certain parameters depending on E'.

Similar dependence for g-factor was obtained for excitons localized in quantum dots and for excitons in quantum wells [29, 33].

Depending on the values of $g_0, a, b, (E')$ and $R(E')$ all three cases described above are possible: g-factor changes sign at energy below emission band maximum, at maximum energy, and above maximum energy. This leads to emission spectra similar to those shown in Fig. 3.

Here, three cases are possible.

1. g-factor changes sign at energies below emission band maximum in zero magnetic field. In this case, the low-energy emission band has lower intensity than the high-energy one (Fig.2*a, b*).

2. g-factor changes sign at the energy of PL band maximum in zero magnetic field. Emission bands in both polarizations have equal amplitude (Fig. 2*c, d*).

3. g-factor changes sign at energies above the emission band maximum in zero magnetic field. The low-energy emission band is more intense than the high-energy one (Fig. 2 *e, f*).

Thus, we obtain the following.

1. The widths $\Delta_{\pm}$ of these two emission bands are equal

$$\Delta_{\pm} \approx 2w\sqrt{\ln 2}. \quad (14)$$

The less intense band is slightly wider than the more intense one. The difference in band widths is less than 10%.

2. The intersection of bands and, consequently, the nullification of emission polarization degree occurs at the energy when $g_{eff}(E\phi) = g_0 + gE\phi = 0$.

3. The polarization degree along the emission band contour changes sign at the energy value when $g_{eff}(E\phi)$ changes sign.

The spectral dependence of emission intensity in two circular polarizations is shown in Fig. 2.

Obviously, qualitatively similar dependencies are obtained for any monotonic dependence of the g-factor on exciton energy.

3. DISCUSSION

If g-factor does not change sign, the polarization degree remains of the same sign for all energies. If g-factor changes sign at some energy, the PL polarization degree also changes sign along the PL band contour. The energy at which polarization changes sign corresponds to the energy at which g-factor becomes zero.

In this case, the splitting of emission band maxima is described by formula (10) and can be comparable to the width of the bands themselves, as if the effective exciton g-factor had a giant value $g_{\text{eff}} \sim 100$. This appears very unusual for non-magnetic materials. This result is determined by the dispersion of g-factors and the magnitude of inhomogeneous broadening (formula (10)). In this case, the ratio of band intensities according to formula (12) depends on the sign of the g-factor.

The dependence of polarization degree $P_{circ}(B)$ on magnetic field also appears unusual. In the classical case (3), this dependence is linear with magnetic field in small magnetic fields:

$$P_{circ}(B) \mu \text{ mg}B / 2kT.$$

However, in an inhomogeneous ensemble, we obtain a quadratic dependence of the integral polarization degree in small fields:

$$P_{circ}(B) \propto (\mu\tilde{g}\omega B / 2kT)^2.$$

The presented analysis was conducted for equilibrium PL, when thermodynamic equilibrium is established at Zeeman sublevels. Deviation from equilibrium distribution is accounted for in formula (3) using the depolarizing factor

$$\frac{t_0}{t_0 + t_s}. \qquad (15)$$

Thus, we effectively assume that the spin relaxation time t_s is much shorter than the lifetime t_0. In a real situation, complete equilibrium may not be achieved. The population ratio of Zeeman sublevels in the non-equilibrium case is

$$\frac{n_1}{n_2} = \frac{\exp\left(-\frac{DE}{2kT}\right) + \frac{t_s}{t_0}\text{ch}\left(\frac{DE}{2kT}\right)}{\exp\left(\frac{DE}{2kT}\right) + \frac{t_s}{t_0}\text{ch}\left(\frac{DE}{2kT}\right)}. \qquad (16)$$

Obviously, equation (16) transforms into equation (1) $\tau_s \leqslant \tau_0$. However, accounting for non- equilibrium does not qualitatively change the spectral dependencies (Fig. 2). In the limiting case of absent spin relaxation, $\tau_s \geqslant \tau_0$, the difference between emission spectra in two polarizations disappears.

Thus, the main factor affecting the unusual behavior of magnetic-induced polarized luminescence of quantum dot ensemble is the dependence of exciton g-factor on the size dispersion of quantum dots.

The dependence of hole g-factor on quantum well thickness was experimentally observed in quantum wells based on GaAs [34, 35] and InAs [33].

For electrons, the g-factor also depends on quantization energy [36,37]. These dependencies will either enhance or weaken each other. But typically, the electron g-factor is smaller than the hole g-factor, thus its contribution is small.

In practice, the g-factor value is experimentally determined from the splitting of emission band maxima in magnetic field, and the level population is determined from the ratio of band intensities in two circular polarizations. As seen from the presented work, this can give highly incorrect results.

4. CONCLUSION

In this work, using the example of a quantum dot ensemble, magnetic field-induced polarized luminescence of localized excitons was studied. The following was discovered.

1. The splitting of the luminescence band maxima of the ensemble exceeds the Zeeman splitting of each individual exciton by several orders of magnitude.

2. The low-energy luminescence band has a lower intensity than the high-energy one. This seemingly contradicts the Boltzmann distribution of excitons across Zeeman sublevels.

3. The effects described in points 1 and 2 only occur when the exciton factor depends on the localization energy and changes sign at a certain energy.

4. If the sign change of the g-factor occurs at energies above the emission band maximum, i.e., for small-sized points, then the intensity ratio of the bands becomes consistent with the Boltzmann distribution.

The results of this work are not limited to just inhomogeneous quantum dot ensembles. Emission lines of excitons and impurity centers in crystals are always inhomogeneously broadened. The only question is the ratio between homogeneous and inhomogeneous emission line widths.

The sign change of the *g*-factor in quantum dot ensembles can significantly affect mode-locking effects, spin echo, and spin coherence in quantum dot ensembles [38].

ACKNOWLEDGMENTS

The authors thank D. S. Smirnov and T. S. Shamirzaev for helpful discussions.

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