Optical orientation of nuclei in nitrogen alloys GaAsN at room temperature

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The intensity and the giant circular polarization of edge luminescence in a longitudinal magnetic field have been measured in nitrogen alloys GaAsN under circularly polarized pumping. It has been found that these dependences are shifted with respect to zero field by a value \( B_{\text{eff}} \). The magnitude of the internal field \( B_{\text{eff}} \) increases with increase in pumping intensity and reaches saturation \((\approx 250 \text{ Gauss})\) at great densities of excitation. The saturation of the \( B_{\text{eff}} \) field with growth of pumping indicates that this is a field of nuclei, polarized dynamically due to hyperfine interaction with optically oriented deep paramagnetic centers, rather than a field of exchange interaction created on the center by spin-polarized photo-excited conduction electrons. The short time of nuclear polarization by electrons \((<15 \mu \text{s})\), measured under modulation of circular polarization of the exciting light with high frequency, points to a small number of nuclei undergoing hyperfine interaction with an electron localized at a center.

In the last few years the spin properties of nitrogen alloys Ga(In)AsN have been drawing heightened attention, owing to the fact that in such alloys there occurs at room temperature an anomalously great \((\text{up to } 90\%)\) spin polarization of free electrons, which is retained for a long time under optical pumping (see review 1 and references therein).

Recently we have found 2 that the circular polarization degree \( \rho \) and the intensity \( J \) of the edge photoluminescence (PL) excited by circularly polarized light in GaAs\(_{1-x}\)N\(_x\) crystals \((x \sim 1\%)\) at room temperature, increase substantially in a longitudinal magnetic field \( B \sim 1 \text{ kG} \) (for an example see Fig. 1). This increase depends on intensity of pumping and can reach a twofold value under weak or moderate pumping. We considered in 2 the suppression of spin relaxation of deep paramagnetic centers in a longitudinal magnetic field as a possible cause of such a rise in \( \rho \) and \( J \). These centers appear in the process of crystal growing, when nitrogen atoms are introduced into gallium arsenide, and create a dominant channel for recombination of free carriers. The recombination is spin-dependent, since its rate is governed by spin polarization of the electrons localized on centers. In other words, the paramagnetic centers act as a spin filter, blocking the capture from the conduction band of electrons with predominant orientation of spins. Therefore an increase in spin relaxation time of the centers in the magnetic field brings about a strong increase in their polarization \( P_e \), which, in its turn, is accompanied by the growth in the spin polarization \( P \) of free electrons and also in their concentration. The former reveals itself in the rise of \( \rho \) \((\text{since } \rho \propto P)\), and the latter, in the rise of \( J \). We supposed in 2 that the spin relaxation of deep centers of GaAsN in the zero field is due to chaotic magnetic fields, which are generated by spin fluctuations of the nuclei located in the vicinity of a paramagnetic center and coupled with the center by the hyperfine interaction 3 4.

Also in 2 it has been found that the experimental dependences \( J(B) \) and \( \rho(B) \) are shifted with respect to zero field by a value of \( |B_{\text{eff}}| \sim 100 \text{ Gauss} \); besides, the direction of the shifting reverses with reverse of the sign of circular polarization of exciting light (see Fig. 1). It is pointed out in 2 that such a shift is possibly caused by the Overhauser stationary field, \( B_N \), which acts on a localized electron from the optically-oriented crystal-lattice nuclei located near center. The field \( B_N \propto \langle I \rangle \) 3 5 6, where \( \langle I \rangle \) is the mean nuclear spin, and is directed along the external field \( B \) (along the exciting beam). As a result, adding up with the external field, or subtracting from it, the field \( B_N \) brings about the shift of the curves \( J(B) \) and \( \rho(B) \). As the polarization of nuclei is proportional to the polarization of electrons, the sign reverse of the pumping polarization involves a change in the direction of the field \( B_N \) and, consequently, in the direction of the shift of the \( \rho(B) \) and \( J(B) \) curves. At the same time, the shift of the \( \rho(B) \) and \( J(B) \) curves can be also induced by the exchange interaction of a polarized paramagnetic center with other paramagnetic centers or with spin-polarized free electrons in the conduction band. Indeed, each of these exchange interactions creates at the center an effective magnetic field \( B_{\text{ex}} \), which is proportional to

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the full spin of localized or free electrons \[7, 8\] and hence must change the direction when the sign of circular polarization of excitation reverses. The exchange interaction between deep centers in the studied GaAsN crystals can be neglected on account of the small concentration of centers \((N_c \approx 10^{15} \text{cm}^{-3} \[1\]) and, accordingly, the small overlap of their wave functions \[7, 8\]. One cannot neglect the exchange interaction with conduction electrons beforehand, since in GaAsN the exchange interaction constant is unknown \[9\], while the experimental observation of the field \(B_{\text{eff}}\) requires the use of a strong, \(\sim 50 \text{mW}\), pumping. Under such a pumping the polarization of free electrons approximates 100% \[1\], and their concentration, according to our evaluation, amounts to a considerable value of \(n \sim 10^{15} \text{cm}^{-3}\), which can bring about a great value of the exchange field.

The present work describes an experiment unambiguously indicative of the nuclear nature of the field \(B_{\text{eff}}\). On the qualitative level, we can discriminate between the nuclear field \(B_N\) and the field of exchange interaction with free electrons \(B_{\text{ex}}\) by their dependence on pumping intensity. Indeed, the hyperfine contact \((A_{\text{sc}} \cdot \mathbf{s})\) and exchange \((Q_{\text{sc}} \cdot \mathbf{s})\) interactions create the fields \(B_N \propto (I) \propto P_c\) and \(B_{\text{ex}} \propto S = P n/2 \[7\], which act upon the spin \(s_c\) of an electron localized on the center. Here \(I\) and \(s\) are the spins of a nucleus and a free electron, \(A\) and \(Q\) are the hyperfine and exchange constants, \(S\) is the total spin of free electrons, \(P\), and \(P\) is the polarization of localized and free electrons, respectively. Under sufficiently large pump intensity \(W\) (in our case at \(W \gtrsim 75 \text{mW}\)) the spin filter effect in GaAsN leads to practically full polarization of both localized \((P_c \approx 1)\) and free \((P \approx 1)\) electrons \[1\]. With further increase of pumping the field \(B_N\) has to remain unaltered, while the field \(B_{\text{ex}}\) has to grow linearly as the excitation intensity increases, because \(n \propto W\): \(B_N = \text{const}, B_{\text{ex}} \propto W\). This serves as a main criterion for separation of the nuclear and exchange fields.

We have investigated an undoped alloy GaAsN with a nitrogen content of 2.1%, grown as a film 0.1 \(\mu\text{m}\) thick on a semi-insulating GaAs substrate \[10\]. Polarization of free electrons \(P\) was induced by interband absorption of circularly polarized light \[5, 6\]. Measurements were taken of the intensity \(J\) and the degree of circular polarization of the edge PL \(\rho = P^P P^P\), where the numerical factor \(P^P \leq 1 \[5, 6\]. The degree \(\rho\) is defined as \(\rho = (J^+ - J^-)/J\), where \(J^+\) and \(J^-\) are PL components polarized on the right (\(\sigma^+\)) and the left (\(\sigma^-\)) circle, \(J = J^+ + J^-\). A continuous-wave (CW) titanium-sapphire laser was used to excite photoluminescence, which
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Fig. 2: The shifts of the curves $I(B)$ (solid circles) and $\rho(B)$ (open squares) with respect to zero of the magnetic field in GaAs$_{0.979}$N$_{0.021}$ at room temperature under different powers of right-circularly polarized excitation. Triangles show the dependence $\rho(J)$ taken at $B = 0$; solid line is a guide for the eye. $\hbar\nu_{\text{exc}} = 1.393$ eV, $h\nu_{\text{det}} = 1.159$ eV.

was registered by means of a photomultiplier with InGaAsP photocathode. The values of $\rho$ and $J$ were measured using a high-sensitive polarization analyzer [11] comprising a photoelastic polarization modulator [12] and a lock-in two-channel photon counter. The measurements were carried out at 300 K under perpendicular incidence of the laser beam onto the sample and PL recorded in opposite direction. The magnetic field was directed along the exciting beam (Faraday geometry).

The field magnitude $|B_{\text{eff}}|$ was found through fitting the experimental dependences $J(B)$ and $\rho(B)$ by Lorentzians of the form $y(B) = y_{\text{max}} + (y_{\text{min}} - y_{\text{max}})/(1 + (B - B_{\text{eff}})^2/B_{1/2}^2)$ (solid lines in Fig. 1), where $y_{\text{min}} = y(B = B_{\text{eff}})$, $y_{\text{max}} = y(B \to \infty)$ and $B_{1/2}$ is the half-width of the curve on the half-height. Fig. 2 shows dependences $B_{\text{eff}}(W)$ which obtained as a result of fitting the curves $J(B)$ (solid circles) and $\rho(B)$ (empty squares) measured in GaAs$_{0.979}$N$_{0.021}$ under the change in intensity (from 3 to 300 mW) of right-hand ($\sigma^+$) polarized excitation. We see that $B_{\text{eff}}$ tends to zero with decreasing $W$. This is a result of the decline in efficiency of spin-dependent recombination under weak pumping, in consequence of which the polarization of localized electrons $P_e$ tends to zero [1]. With increasing pumping the field $B_{\text{eff}}$ increases and reaches saturation at $W \approx 75$ mW. The dependence $\rho(W)$ (triangles in Fig. 2) measured at $B = 0$ is saturated at the same intensity, which speaks of practically full polarization of localized electrons, $P_e \approx 1$ [1], at $W \gtrsim 75$ mW. Therefore the saturation of field $B_{\text{eff}}$ at $W \gtrsim 75$ mW permits us to draw an unambiguous conclusion that a dominating role in formation of the field $B_{\text{eff}}$ belongs to the nuclear polarization.

An important parameter of the nuclear spin system is the time of its polarization by optically oriented electrons $T_{1e}$. As a rule, the process of optical orientation of nuclei under interband pumping in semiconductors and semiconductor nanostructures is very slow: according to the concentration of doping impurity and the crystal temperature, the time of longitudinal nuclear relaxation may range from seconds to hours [13]. The inertness of nuclear spin system frequently serves as a means of obliterating its polarization. To this purpose the sign of circular polarization of the exciting light should be varied with high frequency. In that case the nuclear polarization has no time to follow the quick alternation of electron polarization, and the nuclear field $B_N$ comes to zero [5, 6]. We changed the sign of circular polarization of the exciting light with a frequency 35 kHz, passing a linearly polarized laser beam through the photoelastic modulator of polarization [12] working on that frequency. The two-channel photon counter [11] synchronized with the polarization modulator permitted the PL intensity
to be measured separately during adjacent \((\sigma^+ \text{ or } \sigma^-)\) half-periods of polarization changing, by opening each of the channels for the same time equal to 0.47\(^*\) where 7\(^*\) is the oscillation period of the modulator.

The influence of modulation of pumping polarization on the magnitude of the field \(B_{\text{Mm}}\) can be best followed by measuring the magnet-field dependence of luminescence intensity \(J(B)\), applying no other polarization elements but the polarization modulator installed in the excitation channel (the registration channel is not equipped with a quarter-wave plate and linear polarizer of light that are indispensable for measurement of \(\rho\)). Fig.1a shows the dependences of luminescence intensity \(J^+(B)\) (solid squares) and \(J^-(B)\) (open squares), accumulated, respectively, in \(\sigma^+\) and \(\sigma^-\) half-periods of modulation at \(W = 30\) mW. It is evident that these dependences are shifted in opposite directions with respect to the axis of ordinates by the same magnitude equal to \(\approx 165\) G. Within the limit of measurement error this magnitude coincides with the magnitude of shift \(\approx 175\) G measured under the constant circular polarization of pumping with the same intensity (see Fig2). Thus, the change in the sign of circular polarization of pumping with a half-period of 15 \(\mu\)s is not accompanied by suppression of nuclear polarization. It means that the spin relaxation time of nuclei on electrons \(T_{\text{rel}}\) is shorter than 15 \(\mu\)s. The short time \(T_{\text{rel}}\) is a fingerprint of a strong localization of the electron wave function on a center and, hence, of a small amount of nuclei undergoing the hyperfine interaction with the localized electron [5, 13].

The size of the region of electron localization can be evaluated by the order of magnitude on the assumption that the localized electrons lose polarization through relaxation on the chaotic fluctuations of the nuclear field, and the magnitude \(B_f\) of these fluctuations presets the half-width \(B_{1/2}\) of the curves \(J(B)\) and \(\rho(B)\): \(B_f \sim B_{1/2}\).

The characteristic value of the nuclear field fluctuation \(B_f \approx (B_{\text{Mm}}/N)\sqrt{N} = B_{\text{Mm}}/\sqrt{N}\), where \(N\) is the number of nuclei in the localization region, \(B_{\text{Mm}} \propto A/(g_c\mu_0)\) is the field of totally polarized nuclei acting on the spin of localized electron with \(g\)-factor \(g_c\), \(\mu_0\) is the Bohr magneton, \(B_{\text{Mm}}/N\) is the field created by one nucleus, \(A \approx 100\) \(\mu\)eV in GaAs [5]. The \(B_{\text{Mm}}\) value doesn’t depend on the size of the region of electron localization [5]. Using \(B_{\text{Mm}} \approx 53\) kG calculated for GaAs in [13] (see also [5, 6]), where \(|g_c| = 0.44\), and taking into account that in GaAsN \(g_c = 2\) [15], one can find \(B_{\text{Mm}} \approx 12\) kG for GaAsN. Taking typical experimental value \(B_{1/2} \approx 1000\) G (see Fig.1), we obtain that the field \(B_f\) of the same value is created in GaAsN by \(N \approx (B_{\text{Mm}}/B_f)^2 \approx 100\) nuclei.

On the other hand, \(N \approx 2V/v_0\), where \(V\) is the volume of electron localization, \(v_0\) is the elementary cell volume. Assuming \(V \approx 4a_0^3\), where \(a_0\) is the electron Bohr radius, we find \(a_0 \approx 7\) \AA.

So small an extent of electron localization and, as a consequence, a small amount of nuclei, necessitates an alternative explanation to be considered for the above experimental results; this explanation employs the limiting case of hyperfine interaction of a localized-on-center electron with only one nucleus, namely, the nucleus of the center. The Hamiltonian of such an interaction in the presence of an external magnetic field \(B\) has the form [7]: \(A \cdot S_c + g_c\mu_0B\cdot S_c\) (Zeeman energy of the nucleus is assumed to be negligible as compared to the energy of its hyperfine interaction with the electron). For such a case, a theory of optical orientation of electrons and nuclei in a semiconductor was developed in [16] by Dyakonov and Perel. It is based on a partial destruction of the optical orientation of localized electrons in a zero magnetic field due to mixing of eigenstates of the system one electron - one nucleus. The energy of these states at \(B = 0\) is determined by the total spin of electron and nucleus \(M = 1/2 + I\), where \(I\) is the nuclear spin, which for an odd \(I\) takes two values separated by an interval \(\delta = A\) [7]. For example, \(\delta = A\) and \(2A\) for \(I = 1/2\) and 3/2, respectively. The destruction of electron orientation occurs effectively, if the characteristic time of mixing \(\tau^* = \hbar/\delta\) is much shorter than the lifetime of localized electrons \(\tau_c\). At \(A \approx 100\) \(\mu\)eV we obtain \(\tau^* \sim 1\) ps. In GaAsN the time \(\tau_c\) decreases drastically with the excitation intensity increasing [1]; according to our evaluation, however, it extends 50 ps even at the maximum power of pumping \(W = 300\) mW. It means that \(\tau^* < \tau_c\), and the mixing of states and also the loss of electron spin polarization may take place in GaAsN effectively at all employed intensities of excitation. If \(\langle S_0\rangle\) is the mean spin of electrons in a moment of their capture by the center, then at \(\tau^* < \tau_c\) the electron spin decreases down to \(\langle S_0\rangle/2, 3\langle S_0\rangle/8\) and \(\langle S_0\rangle/3\) for \(I = 1/2, 3/2\) and \(I >> 1\), respectively [16].

The mixing of states slows down upon application of a longitudinal magnetic field, which ruptures hyperfine coupling thus restoring polarization of electrons [16, 17]. In this case the half-width \(B_{1/2}\) of the curves \(J(B)\) and \(\rho(B)\) determines the order of magnitude for the external magnetic field in the presence of which the Zeeman energy of an electron becomes equal to the energy of hyperfine interaction: \(g_c\mu_0B_{1/2} \sim \delta\). The value of \(\delta\), evaluated from the half-width \(B_{1/2} \sim 1000\) G of experimental curves \(J(B)\) in Fig.1a, is 10 \(\mu\)eV.

The deep paramagnetic center, responsible for the spin-dependent recombination in GaAsN, is the self-interstitial defect Ga\(^{2+}\) [15]. The nucleus of this defect is represented by two isotopes, \(^{69}\)Ga and \(^{71}\)Ga, each
with spin \( I = 3/2 \). The analysis of the EPR spectra of that center has revealed the fact that up to 20\% of an electron is localized on its nucleus [15]. Such a density of the electron on the nucleus corresponds to the energy of hyperfine interaction equaling \( \approx 10 \mu eV \) (we are taking into account that the hyperfine interaction constants for Ga isotopes are \( A_{\text{Ga}} \approx 38 \mu eV \) and \( A_{\text{Ga}} \approx 49 \mu eV \) [14]), which coincides in the order of magnitude with the value of \( \delta \), evaluated above from the half-width \( B_{1/2} \) of the curve \( J(B) \). This indicates that the hyperfine interaction of an electron with one nucleus enables us to describe qualitatively the growth of experimental relations \( J(B) \) and \( \rho(B) \).

The Dyakonov-Perel theory [16] predicted also that the decay of optical orientation of electrons in zero field, involved by the mixture of states, is accompanied by the appearance of stationary polarization of nuclear spins in an ensemble of centers. In the simplest case \( I = 1/2 \) and \( \tau_c/\tau^* \gg 1 \) the mean nuclear spin \( \langle I \rangle = \langle S_0 \rangle \nu_{1c}/(\nu_{1c} + \nu_1) \) [16], where \( \nu_{1c} = f/2 \) is the decay rate of nuclear orientation due to hyperfine interaction with captured nonoriented electron, \( \nu_1 \) is the rate of nuclear relaxation in the absence of illumination, \( f \) is the number of electrons captured on the center per unit time. Since the capture frequency \( f \) and, hence, \( \nu_{1c} \) are proportional to the light intensity \( W \), \( \langle I \rangle \) grows linearly at low intensities and reaches saturation at a level \( \langle S_0 \rangle \) under strong light when \( \nu_{1c} \gg \nu_1 \).

As the appearance of stationary nuclear polarization is brought about by the mixing of states, the suppression of mixing in a longitudinal magnetic field \( B \) reduces the value of \( \langle I \rangle \) [16]. This is in line with the decrease in relaxation rate \( \nu_{1c} \), which, according to [16], takes form \( \nu_{1c} = (f/2)(1/(1 + (g\mu_B B/I)^2) \) for \( I = 1/2 \) and \( \tau_c/\tau^* \gg 1 \). In particular, it means that in a longitudinal field a greater intensity of excitation will be required to reach the same polarization of nuclei found at \( B = 0 \). The expression for \( \langle I \rangle \) at \( I > 1/2 \) is nowhere to be found in literature.

The theory [16] infers that the nuclear polarization may cause asymmetry in the dependence of electron polarization on the longitudinal magnetic field if the nucleus of a center alone is taken into account. Such dependence, however, has not been presented explicitly. In the case of a great amount of nuclei in the region of localization \( N \gg 1 \) the influence of nuclear stationary polarization on the orientation of electrons shows up through the effective magnetic field of nuclei \( B_N \) which adds up to the external field \( B \) [3] (see also [5, 6]). At \( N = 1 \) for \( \langle I \rangle \neq 0 \) there exists also an electron spin splitting averaged over nuclear spin projections, which can be interpreted as the effect of the mean nuclear field \( B_N \propto \langle I \rangle \). As with \( N \gg 1 \), the field \( B_N \) attenuates or strengthens the effect of an external magnetic field upon the electron spin and, as a consequence, shifts the curves \( J(B) \) and \( \rho(B) \) relative to zero of the field. It has been noted above that at \( N = 1 \) the nuclear polarization \( \langle I \rangle \) in the field \( B = 0 \) increases linearly with increasing pumping under a weak pumping and gets saturated under a strong pumping. Therefore the magnitude of the shift must also arrive at saturation with growing \( W \). Thus the model of hyperfine interaction with the nucleus of the center can explain not only the increase but also the shift of dependences \( J(B) \) and \( \rho(B) \), that we have discovered in GaAsN.

An additional investigation is required in order to elucidate the relative contributions of the center nucleus and the nearest-to-it nuclei of the crystal lattice into the optical orientation of a localized electron and its asymmetry in the longitudinal field in GaAsN. The observation of forbidden electron magnetic transitions in weak magnetic fields \( B < B_{1/2} \), i.e., in the fields of the order of some hundreds or tens Gauss (e.g. by means of electrically [18] or optically [19] detectable EPR) could give a direct corroboration of the mixing effect.

In the case of \( N = 1 \) the nuclear polarization, being maximal in the zero field, decays at \( B >> B_{1/2} \) [16], while the reverse situation is observed for \( N >\) 1: here the nuclear polarization is at its maximum in a strong magnetic field [3] [5] [6]. Measurement of the \( \langle I \rangle \) magnitude in the weak and the strong field \( B \) could also give arguments in favor of one or the other scenario.

The initial polarization of electrons \( \langle S_0 \rangle \) in both models of optical orientation presented in [3] and [16] is determined only by selection rules under interband absorption. In GaAsN the polarization of an electron captured on the center changes drastically due to the spin-dependent recombination [1]. Therefore, in order to describe quantitatively the experimental results we have obtained in GaAsN, it is necessary to modify the model of optical orientation of electrons in the presence of spin-dependent recombination through the deep paramagnetic center [1]. This can be done by way of taking into account the hyperfine interaction of the localized electron both with the center nucleus and with the field fluctuations of nuclei situated in the vicinity of the center.
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   Theoretical magnitude of the exchange integral depends strongly on the radius of the localized electron and the wave function structure of the deep center, which are also unknown exactly. Assuming that the ground state of the deep center is situated in the middle of the band gap, (the binding energy $\varepsilon_c \gtrsim 0.5$ eV) one can evaluate $B_{ex} \lesssim 10$ G. As the binding energy decreases, the magnitude of the field $B_{ex}$ increases. Hence, not knowing exactly the magnitude of $\varepsilon_c$, we cannot reject a priori the contribution of exchange interaction. The authors are grateful to K. V. Kavokin who has evaluated the magnitude of the field $B_{ex}$.
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