Conference paper UDC 538.958 DOI: https://doi.org/10.18721/JPM.171.105

# Dynamics of electron-nuclear spin system in GaAs:Mn epitaxial layers

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**Abstract.** In this paper we present experimental study of electron-nuclear spin dynamics in GaAs bulk layers doped with Mn ions at temperature 4.2 K. The electron spin dynamics is experimentally investigated by measuring the degree of polarization of photoluminescence in a transverse magnetic field (Hanle effect) and the recovery of the electron spin polarization in a longitudinal magnetic field (polarization recovery curve). To study nuclear spin dynamics, we use two-stage experimental protocol including optical cooling of nuclear spin system and measuring change of the polarisation degree of photoluminescence in different transverse magnetic fields. We show dependence of electron spin relaxation times on excitation power for three samples with different concentrations of shallow donors and acceptors. Electron spin relaxation times have been obtained as at the exciton transition as at the deep acceptor Mn transition. Also we show dependence of nuclear spin-lattice relaxation times T1 on value of external transverse magnetic field.

Keywords: semiconductors, gallium arsenide, spin, spin dynamics, spin relaxation, optical orientation, Hanle effect, polarization

Funding: The study was supported by the St. Petersburg State University grant 94030557.

**Citation:** Berdnikov V.S., Kuznetsova M.S., Kavokin K.V., Dzhioev R.I., Dynamics of electron-nuclear spin system in GaAs:Mn epitaxial layers, St. Petersburg State Polytechnical University Journal. Physics and Mathematics. 17 (1.1) (2024) 31–36. DOI: https://doi.org/10.18721/JPM.171.105

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Материалы конференции УДК 538.958 DOI: https://doi.org/10.18721/JPM.171.105

## Динамика электронно-ядерной спиновой системы в эпитаксиальных слоях GaAs:Mn

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Аннотация. В настоящей работе представлено экспериментальное исследование динамики электронно-ядерной спиновой системы в объемных слоях GaAs, легированного ионами Mn при температуре 4,2 К. Получена зависимость времен электронной спиновой релаксации от мощности оптической накачки для трех образцов с различной концентрацией мелких доноров и акцепторов. Времена электронной спиновой

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релаксации определялись как для экситонного перехода, там и для перехода глубокого акцепторного центра Mn. Получена зависимость времен ядерной спин-решеточной релаксации T1 от величины внешнего поперечного магнитного поля.

Ключевые слова: полупроводники, арсенид галлия, спин, спиновая динамика, спиновая релаксация, оптическая ориентация, эффект Ханле, поляризация

Финансирование: Работа выполнена при финансовой поддержке Санкт-Петербургского государственного университета в рамках гранта № 94030557.

Ссылка при цитировании: Бердников В.С., Кузнецова М.С., Кавокин К.В., Джиоев Р.И. Динамика электронно-ядерной спиновой системы в эпитаксиальных слоях GaAs:MN // Научно-технические ведомости СПбГПУ. Физико-математические науки. 2024. Т. 17. № 1.1. С. 31–36. DOI: https://doi.org/10.18721/ JPM.171.105

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#### Introduction

The idea of using spin states of carriers for processing and storing information has been actively discussed in recent decades. It is important for spintronics to study the characteristics of an electron-nuclear spin system, such as electron and nuclear spin relaxation times.

GaAs is a well-studied semiconductor, but adding a magnetic impurity of Mn leads to unexpected results. In Mn-doped GaAs one can see suppression of the Bir-Aronov-Pikus spin relaxation mechanism, which is significant in regular p-type GaAs [1,2].

In this paper, we present measurements of electron spin relaxation times  $\tau_s$  as function of excitation power for 3 samples with different concentrations of impurities. Also in this work we have obtained the dependence of the nuclear spin relaxation times  $T_1$  on the external magnetic field.

### **Structures and Methods**

The sample under study consists of  $36-\mu m$  layers of bulk GaAs:Mn grown by liquid-phase epitaxy on a (001)-oriented GaAs substrate. Samples are doped with Mn ions. Shallow donors and acceptors are also present. Parameters of doping concentrations are shown in Table.

Table

| Sample   | $N_{Mn} \cdot 10^{17} \text{ cm}^{-3}$ | $N_d - N_a \cdot 10^{15} \text{ cm}^{-3}$ |
|----------|--|---|
| Sample 1 | 2.77                                   | 3.44                                      |
| Sample 2 | 3.6                                    | -0.33                                     |
| Sample 3 | 1.2                                    | 0.03                                      |

Concentrations of Mn acceptors and the difference in concentrations of shallow donors and acceptors for different samples

Notations:  $N_{Ma}$ ,  $N_{a}$  and  $N_{a}$  are the concentrations of Mn and shallow donors and acceptors, respectively.

Electron spin dynamics is experimentally investigated by measuring the degree of polarization of photoluminescence (PL) in a transverse magnetic field (Hanle effect) and the recovery of the electron spin polarization in a longitudinal magnetic field (polarization recovery curve, PRC). Electron spin polarization is created by circularly polarized continuous wave excitation using Ti:Sapphire laser operating at 800 nm. Excitation beam is focused on the sample to the 300-µm diameter spot. To avoid the effect of nuclear spins to electron spin dynamics, the polarization helicity of the excitation has been modulated at the frequency 50 kHz. To create the transverse to optical pump magnetic field we use the electromagnet installed outside the cryostat. Pairs of Helmholtz coils create longitudinal magnetic fields with magnitude up to 30 mT.

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Spin relaxation time of electron  $\tau_{a}$  and lifetime of optically excited electrons  $\tau$  are obtained by measuring the series of the Hanle curves and polarization recovery curves. The dynamics of the average electron spin in Voigt geometry manifests itself in the magnetic-field dependence of degree of circular polarization of luminescence [1]:

$$\rho_{c} = \frac{\rho_{0}}{\left(1 + \frac{\tau}{\tau_{s}}\right)} \frac{1}{\left(1 + \left(B / B_{1/2}\right)^{2}\right)},\tag{1}$$

where  $B_{1/2} = \hbar / (g_e \mu_B) T_s^{-1}$  is the characteristic field that gives inverse electron spin lifetime

 $T_s^{-1} = \tau_s^{-1} + \tau^{-1}$ , and *B* is the value of the external transverse magnetic field. To get initial polarization  $\rho_0$ , we model data from experiments in Faraday geometry with the following expression [3]:

$$\rho_{c} = \frac{\rho_{0}}{1 + \frac{\tau}{\tau_{c}^{*}}}, \quad \tau_{s}^{*} = \tau_{s} \left[ 1 + \left( B / B_{c} \right)^{2} \right], \quad (2)$$

where  $B_{c}$  is the correlation field (fitting parameter), and B is the value of the external longitudinal magnetic field.

Optical orientation and the Hanle effect form the basis for experiments to study electron-nuclear spin dynamics. To investigate the nuclear spin relaxation time, we use two stage experiment. At the first stage the nuclear spin system is cooled due to hyperfine interaction with polarized electron spin and application of the longitudinal magnetic field  $B_z = 3$  mT. At this stage the Overhauser field  $B_x$  is generated. At the second stage the field  $B_z$  is turned off and certain transverse magnetic field  $B_x$  with magnitude range from 0.1 mT to 40 mT is turned on.

Precession of the electron spin in the total effective transverse magnetic field  $B_x \pm B_N$  results in the following time dependence of the PL circular polarization degree [4]:

$$\rho_{c} = \rho_{0} \frac{B_{1/2}^{2}}{B_{1/2}^{2} + \left(B_{x} + b + \left(B_{N} - b\right)\exp\left(-\frac{t}{T_{1}}\right)\right)^{2}},$$
(3)

where  $\rho_0$  and  $B_{1/2}$  are parameters extracted from the analysis of the 50 kHz modulated Hanle curve at the same excitation power; t is the laboratory time; b is the effective field appeared due to dynamic nuclear polarization during the optical pumping at the second stage.

### **Results and Discussion**

Firstly, we measure photoluminescence spectra at different excitation power from 0.01 mW to 20 mW. For clarity, only spectra for Sample 1 are presented in Fig. 1, a. The energy of excitation is 1.55 eV. There are peaks corresponding to energy transitions shown in Fig. 1, b with black arrows. We can see a wide Mn-related band with peak at the energy 1.41 eV (labeled as Mn band). The peaks at the 1.51 eV and 1.49 eV correspond to free and donor-bound excitons (labeled as X) and conduction-band-acceptor transitions (labeled as Ac) respectively.

Below a threshold value of excitation power,  $P_{th} \approx 1$  mW, the X peak is absent, while the Mn band is still visible in the spectra. It also can be seen in inset of Fig. 1, a, which demonstrates the ratio  $I_X/I_{Mn}$  of X and Mn intensities.

Fig. 2, *a* demonstrates results of analysis of experimental data of series Hanle curves and PRC, measured for 3 samples from Table. These results are presented for energy of detection 1.41 eV (corresponding to electron recombination with hole at Mn acceptor). In each case the spin relaxation time increases with increasing the excitation power until some value which matches the value  $P_{th} = 1$  mW that we have seen from the analysis of PL spectra. The values of the spin relaxation time in GaAs: Mn are 2 orders of magnitude greater than in p-GaAs, similar to [3].



Fig. 1. Photoluminescence spectra of Sample 1 at different excitation powers. Intensity is shown in logarithmic scale. Inset shows the ratio of X and Mn peak intensities depending on excitation power (a). Energy diagram of GaAs:Mn (b)

All measurements were also performed at the energy of detection 1.51 eV (free and donor bounded excitons). As evident from comparison in Fig. 2, *b*, the behavior of electron spin relaxation time measured at energies 1.41 eV and 1.51 eV is strongly different. The value of electron spin relaxation time measured for energy 1.51 eV monotonously decreases with the increase of excitation power. This difference is a clear indication that it is the Mn impurity which causes such unexpected behavior of spin relaxation time.



Fig. 2. Spin relaxation time of electrons vs excitation power for 3 samples (*a*). Dependences of spin relaxation time on excitation power at different energy of detection for Sample 1 (red points correspond to at Mn peak, blue points to X peak) (*b*). Solid lines are spline fits plotted as guides to the eye

To study nuclear spin dynamics, we measured the time dependences of the PL polarization in the two-stage experiment at different values of the transverse magnetic field. As seen from Fig. 3. the degree of circular polarization of luminescence drops at the moment of field switching. Then we can see partial recovery of the polarization with time.

Curves were modelled with Eq.3, where  $B_N$  and  $T_1$  are fitting parameters. Thus, we obtained the Overhauser field  $B_N$  and nuclear spin relaxation time  $T_1$  as functions of external transverse magnetic field. Results are presented in Fig. 4, *a*, *b*, respectively.

The Overhauser field increases with growing external magnetic field from 27 mT to 60 mT.

Spin-lattice relaxation time depends on external field and rises from 3 s at small magnetic field  $B_{\rm x}$  to 20 s in saturation. This value is 2 orders greater than in nonmagnetic doped *p*-GaAs [5].



Fig. 3. Degree of circular polarization of luminescence in two-stage experiment for Sample 1. Cooling time  $\Delta t = 600$  s, excitation power  $P_{exc} = 3$  mW. Curves are presented as waterfall



Fig. 4. Dependences of Overhauser field (a) and nuclear spin relaxation time (b) on the external transverse magnetic field for Sample 1

We suppose that it is related to suppression of quadrupole nuclear relaxation. However the mechanisms of nuclear spin-lattice relaxation in GaAs:Mn requires further investigation.

### Conclusion

We have confirmed that electron spin relaxation time in epitaxial layers of GaAs: Mn demonstrate an unusual behavior compared to nonmagnetic doped p-GaAs. It can be explained by suppression of Bir–Aronov–Pikus spin relaxation mechanism.

Spin-lattice relaxation time in GaAs:Mn demonstrates the behavior untypical for p-type GaAs.

A two-stage experiment does not allow us to confidently determine the nuclear spin relaxation time T1 because of the effects of optical pumping during the measurement stage. To verify our results, a three-stage experiment including "dark times" [5] needs to be used. In this experiment, different time periods without optical pump are added between optical cooling and measurement stages.

### Acknowledgments

We are grateful to V.M. Litvyak for valuable discussions and support.

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Received 14.12.2023. Approved after reviewing 15.01.2024. Accepted 02.02.2024.