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Spin-lattice relaxation processes of nuclear spins in GaAs:Mn

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Abstract. This paper presents a theoretical investigation of nuclear spin-lattice relaxation time in epitaxial layers of GaAs doped by Mn ions (the concentration of the deep acceptor Mn $3 \cdot 10^{17}$ cm^{-3}) under optical pumping conditions at the lattice temperature of 4.5 K. Earlier experiments by the optical orientation method showed that the spin-lattice relaxation time depends on the magnitude of the external magnetic field and reaches up to 20 seconds for this sample. Such behavior is not typical for *p*-type semiconductors and is more similar to the nuclear spin system dynamics in *n*-type GaAs. To determine the reasons for the significant difference in the spin-lattice relaxation time between *p*-GaAs and GaAs:Mn, the influence of two possible nuclear spin relaxation mechanisms specific to this material has been evaluated. It is demonstrated that the nuclear spin-lattice relaxation under the action of fluctuating magnetic fields of paramagnetic Mn centers in GaAs does not play a significant role. Relaxation limited by spin diffusion due to hyperfine interaction with Mn holes localized on deep acceptors can make a noticeable contribution to the relaxation rate.

Keywords: semiconductors, gallium arsenide, nuclear spin, spin dynamics, spin-lattice relaxation, optical orientation

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Процессы спин-решеточной релаксации ядер в GaAs:Mn

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Аннотация. В данной работе представлено теоретическое моделирование динамики ядерной спиновой системы в объемном GaAs, легированном ионами марганца (концентрация глубокого акцептора Mn $3 \cdot 10^{17}$ cm^{-3}), в условиях оптического возбуждения при температуре кристаллической решетки 4,5 K. Ранее эксперименты показали, что время спин-решеточной релаксации ядер в экспериментах по оптической ориентации электронных спинов зависит от величины внешнего магнитного поля и достигает 20 секунд для исследуемого образца. Такое поведение нетипично для полупроводников *p*-типа и больше схоже с динамикой ядерной спиновой системы в GaAs *n*-типа. Для определения причин существенного отличия времени спин-решеточной релаксации в *p*-GaAs и в GaAs:Mn проведена оценка

влияния двух возможных механизмов релаксации ядерного спина, специфических для данного материала. Продемонстрировано, что спин-решеточная релаксация ядер под действием флуктуирующих магнитных полей парамагнитных центров Mn в GaAs не играет существенной роли. Ограниченная спиновой диффузией релаксация за счет сверхтонкого взаимодействия с локализованными на глубоких акцепторах Mn дырками может вносить заметный вклад в скорость релаксации.

Ключевые слова: полупроводники, арсенид галлия, ядерный спин, спиновая динамика, спин-решеточная релаксация, оптическая ориентация

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Introduction

In solids, nuclear spin-lattice relaxation time T_1 defines the time of energy exchange between the nuclear spin system (NSS) and crystal lattice. In other words, it is the time of NSS energy relaxation.

It is known from literature that T_1 in *n*-GaAs depends on external magnetic fields and is about hundreds of seconds [1]. At the same time, in non-magnetic doped *p*-GaAs T_1 does not depend on the external field and is 3 orders of magnitude less [1].

In an earlier experimental work [2], it was found that in GaAs doped by Mn ions nuclear spin-lattice relaxation time depends on the magnitude of the external magnetic field and varies in the range from 3 to 20 seconds in presence of optical pumping. But "in the dark" this time is less than 1 second. This behavior is not typical for *p*-type semiconductors.

Mn acceptor impurity is paramagnetic at the doping concentration $n_{Mn} = 2.77 \cdot 10^{17}$ cm^{-3}. Impurity centers Mn can be in two different charge configurations: the neutral acceptor Mn^0 (5d + h), when a bounded hole is present, and the charged acceptor Mn^-(5d), when this hole is absent [3].

We provide a numerical estimation of the effect of two possible nuclear spin relaxation mechanisms, that are specific for such materials. The first is relaxation due to hyperfine interaction with holes localized on deep Mn acceptors. The second is relaxation in fluctuating magnetic fields of paramagnetic impurities.

Theoretical model

Spin precession in random fluctuating magnetic fields leads to spin relaxation. Fluctuating spins of Mn acceptors may be the reason for appearance of such fields in GaAs:Mn. The spin-lattice relaxation time can be found using the following expression:

$$T_1 = \frac{1}{\langle \gamma \rangle^2 \langle B^2 \rangle \tau_c}, \quad (1)$$

where $\langle \gamma \rangle$ is the mean gyromagnetic ratio of isotopes with non-zero magnetic moment; $\langle B^2 \rangle$ is the mean square of magnetic field fluctuation, and τ_c is the correlation time of this field. To estimate the efficiency of this mechanism, we need to calculate these values.

Firstly, we calculate an averaged gyromagnetic ratio, taking into account the abundance of magnetic isotopes in the GaAs sample using a weighted average:

$$\langle \gamma \rangle = 0.3\gamma_{Ga^{69}} + 0.2\gamma_{Ga^{71}} 0.5\gamma_{As^{75}}. \quad (2)$$

Secondly, we estimate the magnitude of fluctuating magnetic fields using the classic dipole-dipole interaction model. Every Mn acceptor with a non-zero magnetic moment creates a magnetic field that can be found as:

$$\vec{B} = \frac{\mu_{\text{Mn}}}{Ir^3}\left(\vec{I} - 3\frac{\left(\vec{I}\cdot\vec{r}\right)\vec{r}}{r^2}\right), \tag{3}$$

where μ_{Mn} is the nuclear magnetic moment of Mn, I is the total spin, and r is the radius-vector from the acceptor. Since this field is random, its mean value is zero. But the square of the fluctuation differs from zero:

$$B^2 = \frac{\mu^2_{\text{Mn}}}{I^2 r^6}\left(I^2 + 3\frac{\left(\vec{I}\cdot\vec{r}\right)^2}{r^2}\right). \tag{4}$$

To get the mean square of the field produced by Mn centers in a spherical layer with thickness dr and radius r at the center of the corresponding sphere, we need to average it by angle:

$$\left\langle B_r^2 \right\rangle = \frac{\mu^2_{\text{Mn}}}{I^2 r^6}\left(I^2 + 3\frac{\left\langle\left(\vec{I}\cdot\vec{r}\right)^2\right\rangle}{r^2}\right)4\pi n_{\text{Mn}} r^2 dr = \tag{5}$$

$$= \frac{\mu^2_{\text{Mn}}}{I^2 r^6}\left(I^2 + 3I^2\left\langle\cos^2\theta\right\rangle\right)4\pi n_{\text{Mn}} r^2 dr = \frac{8\pi\mu^2_{\text{Mn}}}{r^4} n_{\text{Mn}} dr,$$

where θ is the angle between radius-vector and nuclear spin, and n_{Mn} is the concentration of Mn acceptors.

To get the mean squared fluctuation at the center of the sphere with radius R, produced by all Mn centers outside the sphere, we need to integrate this value from R to infinity as shown in Fig. 1:

$$\left\langle B_{\text{R}}^2 \right\rangle = \int_R^\infty \frac{8\pi\mu^2_{\text{Mn}} n_{\text{Mn}}}{r^4}\text{d}r = \frac{8\pi\mu^2_{\text{Mn}} n_{\text{Mn}}}{3R^3}. \tag{6}$$

The magnetic moment of the Mn acceptor can be found as $\mu_{\text{Mn}} = \sqrt{I(I+1)}g\mu_{\text{B}}$, where I is the total moment, g is the g-factor, and μ_{B} is the Bohr magneton.

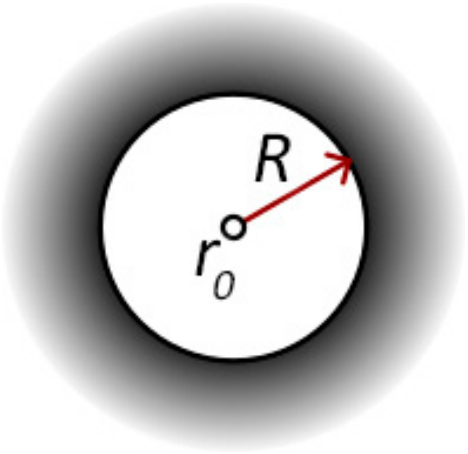

Fig. 1. Diagram of integration limits:
r_0 is the center of the sphere;
R is the radius of the sphere
Gradient shading shows the decreasing effect from distant Mn acceptors

Let Mn doping centers distribution obey the Poisson statistics. Then the probability that there are k Mn atoms in the spherical volume with radius R_k can be calculated as:

$$P_{\text{k}} = \frac{\langle N\rangle^k}{k!}\text{e}^{-\langle N\rangle}, \tag{7}$$

where $\langle N\rangle = \frac{4}{3}\pi R_k^3 n_{\text{Mn}}$ is the average number of Mn atoms inside this volume. Let us take the radius of the sphere, where other Mn atoms are absent with 90% probability, as the lower limit of integration:

$$R = \sqrt[3]{\frac{-3\ln(0.9)}{4\pi n_{\text{Mn}}}}. \tag{8}$$

Therefore, we can estimate the mean square of the fluctuating magnetic field as $\left\langle B_R^2 \right\rangle^0 = 3.37\cdot10^{-10}$ T^2 in the case when all Mn acceptors are in the neutral configuration Mn^0, and $\left\langle B_R^2 \right\rangle^- = 7.69\cdot10^{-10}$ T^2 in the case of the

charged configuration Mn^-. Furthermore, we obtain corresponding values of the root-mean-square field fluctuation: $B^0_{mean} = 0.18 \cdot 10^{-4}$ T and $B^-_{mean} = 0.28 \cdot 10^{-4}$ T.

The last component needed for calculation of T_1 is the correlation time of the fluctuating field. For the neutral acceptor Mn^0, the correlation time τ_c is determined by the spin lifetime of holes. It can be estimated from electron spin resonance (ESR) spectra using the following expression:

$$\tau_c = \frac{1}{\Delta H \gamma_{Mn^0}}, \quad (9)$$

where $\Delta H^0 = 0.02$ T [4] is the half width at half maximum (HWHM) of the corresponding resonance. Thus, $\tau_{c0} \approx 1$ ns. Similarly, according to HWHM value of Mn^- resonance: $\Delta H^- = 0.0028$T [5], so one can get the correlation time of charged acceptor $\tau_{c^-} \approx 12$ ns.

Before we complete the calculation of T_1, we need to check the condition $\langle\gamma\rangle B_{mean}\tau_c << 1$. It means that the precession angle of nuclear spin in the fluctuating magnetic field of Mn acceptors during the correlation time is small. In our case, the product $\langle\gamma\rangle B_{mean}\tau_c$ is about 10^{-6} .

Finally, the estimated spin-lattice relaxation time due to random fields of neutral acceptors is of the order of 30000 seconds. At the same time, T_1 time for relaxation by charged acceptor fields is about 1200 seconds. Both values of T_1 significantly exceed experimental data (3–20 s).

Another mechanism of nuclear spin relaxation in GaAs:Mn is hyperfine interaction with holes, localized at deep acceptors Mn. Since Mn impurities play the role of polarization "killer" centers, this channel of relaxation should be limited due to the finite spin diffusion rate to these centers. The efficiency of this mechanism can be estimated using the Khutsishvili – De Gennes formula [6–7]:

$$T_D \approx \frac{1}{4\pi NbD}, \quad (10)$$

where N is the number of impurity centers per unit volume, b is the effective radius of hyperfine interaction, and D is the nuclear spin diffusion coefficient. We use n_{Mn} for N, hole localization radius at Mn acceptor $r_{Mn} \approx 1$ nm for b [3], and take the spin diffusion coefficient in GaAs, $D = 10^{-13}$ m^2/s.

Substituting the values, one can get $T_D \approx 29$ seconds. This value is close to the experimentally measured T_1, but still exceeds it.

Conclusion

We carried out a theoretical estimation of the spin-lattice relaxation time of nuclei in epitaxial layers of GaAs doped by Mn ions. The calculation is based on experimental data and considers two possible relaxation mechanisms.

We conclude that the spin-lattice relaxation of nuclei under the action of fluctuating magnetic fields of paramagnetic Mn centers in GaAs does not play a significant role. Relaxation limited by spin diffusion due to hyperfine interaction with Mn holes localized on deep acceptors can make a noticeable contribution to the relaxation rate, but most likely it is combined with quadrupole relaxation under the action of electric fields created by the charge fluctuations of impurities. Also, additional investigations of spin diffusion effect could help explain the dependence of T_1 time on the magnetic field.

To provide quantitative estimation of efficiency of the latter mechanism, additional experimental data are required, such as the temperature dependence of T_1. We suggest this as a possible direction for further investigations of the electron-nuclear spin system of GaAs:Mn.

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