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# THEORY OF ELECTRICALLY DETECTED MAGNETIC RESONANCE OF TRIPLET CENTERS IN SILICON

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A theoretical model of spin-dependent recombination (SDR) through excited triplet states of defects in the forbidden band of a semiconductor is proposed. The theory is developed on the basis of quantum equations for the density operator in the space of spin states of a two-electron system. With the help of these equations, the position and form of experimental resonance peaks of nonequilibrium conductivity are analyzed. The consideration is carried out taking into account the Zeeman and hyperfine interactions, crystalline anisotropy, and the processes of generation, recombination, dissociation, and spin relaxation of localized electrons. The comparison of the theoretical and experimental results is performed.

transitions, where a center in the equilibrium state is filled with two electrons with opposite spins. In this case, the act of recombination starts with the capture of a hole and ends with the electron transition from the conduction band to the center. The cross section of the capture of a conduction electron by a center depends on the mutual orientation of spins of free and localized electrons. In the case of the triplet configuration of these spins, the capture cross section is equal to zero due to the Pauli principle, and the recombination is forbidden. Thus, the recombination rate depends on the spin state of free and localized electrons.

## 1. Introduction

It is generally known that the rate of electron-hole recombination in irradiated, plastically deformed, or porous silicon depends on the spin state of free and localized charge carriers [1–5]. The investigation of the SDR phenomenon during 15 years after its experimental discovery was only of fundamental importance. Since the mid-1980s, the SDR phenomenon was used in the studies of real semiconductors or semiconductor structures (see, e.g., [2–4, 6–10]). In these investigations, the magnitude of nonequilibrium conductivity depending on the recombination rate was measured. In turn, the recombination rate depends on the spin state distribution of electrons that can be changed under the conditions of electron paramagnetic resonance. Thus, by measuring the conductivity of a sample, one performs the electric detection of magnetic resonance (EDMR). The EMDR method was used for investigation of defects in irradiated silicon [2, 6], amorphous silicon [3], GaP:N-based structures [4], etc. [11–14].

The first theoretical model of SDR was proposed by D. Lepine and I. Prejean [1]. According to this model, recombination is realized through a center that had one unpaired electron in the equilibrium state (was paramagnetic). The act of recombination starts with the capture of a free electron by a paramagnetic center and ends with the electron transition to the valence band. There also exists a possibility of spin-dependent

The basic point in the Lepine–Prejean model is the fact that band electrons and holes directly pass to one localized level in the forbidden band without creation of an intermediate state. In this case, only one level in the forbidden band is considered, where two electrons with opposite spins can be located.

The Lepine-Prejean model didn't describe a lot of experimental results, that's why other mechanisms of SDR in semiconductors were considered [15–18]. In all these mechanisms, a pair of electron states in the forbidden band is considered. The act of recombination takes place in three rather than two stages. First, an electron from the conduction band passes to the first localized state. After that, the transition between the first and second states in the forbidden band is realized. On the last stage, the electron passes from the second state to the valence band, and the act of recombination comes to the end. There exists a sequence of electron transitions, where a hole is captured from the valence band on the first stage. The basic fact in these models is that a band electron can pass only to the first state, while the transition to the valence band takes place only from the second state. If the spin represents a "good quantum number", the probability of all three transitions will be spin-dependent. However, the greatest change of the recombination rate is concerned with the transition between localized states [15,19].

The existing mechanisms differ in the nature of two localized states and in factors that influence the spin

state of localized electrons. In the Kaplan-Solomon-Mott mechanism [15], localized electron states are associated with two spatially close defects. According to the spin-dependent Shockley-Reed recombination (SDSRR) [17], they represent the ground and excited levels of one defect. In both mechanisms, the spin state of electrons changes only due to an external magnetic field, moreover,  $g$ -factors of the first and second states differ. The theory of SDR with these mechanisms is well developed (see [20,21]).

In the SDR mechanism with participation of triplet centers [2,6, 16] as well as in the SDSRR mechanism, recombination takes place with participation of the ground and excited states of one defect. The difference between these mechanisms is concerned with factors that influence the spin state of electrons. Let's consider separate stages of the recombination act through the excited triplet states of point defects. First, the center contains one electron with unpaired spin. On the first stage, the conduction electron is captured to the excited level of the recombination center, so an excited two-electron state of the center is formed. When the center is in the excited state, it cannot capture an electron or a hole. The excited state can have spin 0 (singlet) or 1 (triplet). On the second stage, there takes place the relaxation of the center from the excited two-electron state to the ground one. The relaxation of the center from the excited singlet state to the ground one is allowed by selection rules, while the relaxation from excited triplet states is forbidden if the spin-orbit coupling is absent. In accordance with this mechanism, triplets take part in SDR due to the presence of spin-orbit interaction. It partially allows transitions from the excited triplet state to the ground one, moreover, the rate of transitions from the triplet state with zero projection of the orbital moment onto some specified direction to the ground state differs from the rate of transitions of triplets with other projections of orbital moment [2,6]. This difference depends on the magnitude of spin-orbit interaction and the structure of wave functions of the ground and excited states. On the last stage, one of the electrons of the ground two-electron state passes to the valence band, and the recombination act comes to the end. It is considered that the electron can pass to the valence band only from the ground two-electron state of the center. Thus, the recombination rate depends on the spin distribution of excited two-electron states. If, due to an alternating magnetic field, the transitions between spin sublevels of the excited triplet state begin, the recombination rate and the conductivity of a sample change. The basic point of

this mechanism lies in the fact that the spin dependence of the recombination rate appears due to the difference between the rates of relaxation of excited triplets with different projections of the total spin to the ground state.

The theoretical treatment of experimental data was carried out with the help of classic (not quantum) kinetic equations for the populations of triplet levels [2,16]. The spin dynamics in an alternating magnetic field and the processes of relaxation and recombination were taken into account by means of phenomenological additives to the equations for populations. In the present work, we use another description of SDR in terms of excited triplet states, which is based on quantum-mechanical equations for the density matrix. We introduce the hyperfine interaction of localized electrons and nuclei into consideration, which allows us to analyze the location and the form of lines of an EMDR spectrum. Theoretical results will be compared to experimental data.

## 2. Theoretical Model

The spin Hamiltonian of an electron pair localized at one center can be presented as

$$\hat{H} = -\frac{\hbar}{2}(\gamma_1\hat{\sigma}_1 + \gamma_2\hat{\sigma}_2)\mathbf{H} + \frac{D}{4}(\hat{\sigma}_{1z} + \hat{\sigma}_{2z})^2 + \frac{J}{4}\hat{\sigma}_1\hat{\sigma}_2 + \sum_j \left\{ \frac{1}{4}\hat{\sigma}_j^{(N)} \left( A_1^j\hat{\sigma}_1 + A_2^j\hat{\sigma}_2 \right) - \frac{\hbar}{2}\gamma_N\hat{\sigma}_j^{(N)}\mathbf{H} \right\}, \quad (1)$$

where  $\hat{\sigma}_{1,2}$  and  $\hat{\sigma}_j^{(N)}$  stand for the Pauli matrices of electron and nuclear spins,  $\gamma_{1,2}$  and  $\gamma_N$  denote the gyromagnetic ratios of electrons and nuclei,  $A_{1,2}^j$  are the parameters of hyperfine interaction between electrons and the  $j$ -th surrounding nucleus,  $J$  is the constant of exchange interaction between electrons,  $\mathbf{H}$  is the magnetic field that represents a sum of the static  $\mathbf{H}_0$  and alternating  $\mathbf{h}$  magnetic fields, while the parameter  $D$  describes the crystalline anisotropy. In addition to the Zeeman and hyperfine interactions, this Hamiltonian takes into account the exchange interaction (which is strong in the case of localization of electrons at one center) and the splitting of energy levels of a triplet in the crystal field.

The coordinate axis  $z$  in Hamiltonian (1) is specified by the direction of the axis of crystalline anisotropy and can mismatch the direction of a constant magnetic field. Real experiments [2,15] are carried out under the conditions of slight deviations of the magnetic field from

the anisotropy axis, that's why, in what follows, we consider that the axis of crystalline anisotropy is directed along a constant magnetic field. In the case of strong magnetic fields, where the inequality  $\gamma_{1,2}H_0 \gg A_{1,2}^j$  is satisfied for all nuclei surrounding the defect, accounting for hyperfine interaction in the course of consideration of the electron paramagnetic resonance is reduced to the substitution of the external constant magnetic field  $H_0$  by the effective one [22,23]

$$H_{1,2} = H_0 + \sum_j (\pm) \frac{A_{1,2}^j}{2\gamma_{1,2}\hbar}, \quad (2)$$

where the signs  $\pm$  correspond to different projections of the nuclear spin onto the direction of the field  $\mathbf{H}_0$ . The case of strong coupling between the electron and nuclear subsystems, as well as the nuclear magnetic resonance, will not be considered.

The theoretical description of SDR in terms of excited triplet states will be performed on the basis of the system of equations for matrix elements of the density operator  $\hat{\rho}$  in the space of spin states of the two-electron system [20,21,24,25]:

$$\begin{aligned} \partial_t \rho_{\alpha\beta} &= -\frac{i}{\hbar} [\hat{H}_{\text{eff}}, \hat{\rho}]_{\alpha\beta} + \frac{g}{4} \delta_{\alpha\beta} - \\ &- \left( d + \frac{r_\alpha + r_\beta}{2} \right) \rho_{\alpha\beta} - \frac{1}{\tau_s} \left( \rho_{\alpha\beta} - \delta_{\alpha\beta} \frac{\text{Sp}\rho}{4} \right), \end{aligned} \quad (3)$$

where  $g$  and  $d$  denote the probabilities of generation and decay of the two-electron excited state per unit time, correspondingly, while  $\tau_s$  is the time of spin relaxation. The probability of generation and dissociation is considered independent on the spin. The relaxation is calculated in the simplest variant, where the times of longitudinal and transverse relaxation coincide. The constants  $r_{\alpha,\beta}$  describe spin-dependent transitions of the center from excited states to the ground one. The indices  $\alpha, \beta$  number matrix elements of the density operator and the Hamiltonian in the spin space of two-electron excited states of the center,  $\alpha, \beta = S, 0, +, -$  (singlet and triplet with  $S_Z = 0, S_Z = +1$ , and  $S_Z = -1$ , correspondingly). The effective Hamiltonian  $\hat{H}_{\text{eff}}$  has a form

$$\begin{aligned} \hat{H}_{\text{eff}} &= -\frac{\hbar}{2} (\gamma_1 \hat{\sigma}_1 + \gamma_2 \hat{\sigma}_2) h_- \\ &- \frac{\hbar}{2} (\gamma_1 \sigma_1^z H_1^{(i)} + \gamma_2 \sigma_2^z H_2^{(i)}) + \frac{D}{4} (\sigma_1^z + \sigma_2^z)^2 + \frac{J}{4} \hat{\sigma}_1 \hat{\sigma}_2. \end{aligned} \quad (4)$$

The index  $i$  of the effective fields  $H_{1,2}^{(i)}$  takes into account not only the possible orientations of nuclear spins but also the possible locations of nuclei with a non-zero spin with respect to the recombination center. Thus, the recombination rate

$$R = r_s \rho_{ss} + r_0 \rho_{00} + r_1 (\rho_{++} + \rho_{--}) \quad (5)$$

should be averaged over all possible locations of nuclear spins and all their orientations.

For further calculations, it's worth to present the recombination rate (5) as

$$R = r_s (N - N_T) + \bar{r} N_T + r' N' / 3, \quad (6)$$

where  $N_T = \rho_{00} + \rho_{--} + \rho_{++}$ ,  $N' = 2\rho_{00} - \rho_{--} - \rho_{++}$ ,  $\bar{r} = (r_0 + 2r_1) / 3$ ,  $r' = r_0 - r_1$ .

In the case of strong exchange interaction, which is realized if electrons are localized at one center, system (3) is essentially simplified:

$$\partial_t N = g - R - dN,$$

$$\partial_t (N - N_T) = \frac{g}{4} + \frac{N}{4\tau_s} - \frac{N - N_T}{\tau_0},$$

$$\partial_t N' = -\frac{N'}{\tau'} - \frac{2}{3} r' N_T + \frac{3i\gamma}{\sqrt{2}} \times$$

$$\times [h_+ (\rho_{+0} - \rho_{0-}) + h_- (\rho_{-0} - \rho_{0+})],$$

$$\partial_t n = -\frac{n}{\tau_1} + \frac{i\gamma}{\sqrt{2}} [h_- (\rho_{0+} + \rho_{-0}) - h_+ (\rho_{+0} + \rho_{0-})],$$

$$\partial_t \rho_{0+} = -\frac{\rho_{0+}}{\bar{\tau}} + i\gamma \left[ \left( \frac{D}{\bar{\gamma}\hbar} - H_{\text{eff}}^{(i)} \right) \rho_{0+} + \right.$$

$$\left. + \frac{h_+}{2\sqrt{2}} (n - N') - \frac{h_-}{\sqrt{2}} \rho_{-+} \right],$$

$$\partial_t \rho_{0-} = -\frac{\rho_{0-}}{\bar{\tau}} + i\gamma \left[ \left( \frac{D}{\bar{\gamma}\hbar} + H_{\text{eff}}^{(i)} \right) \rho_{0-} - \right.$$

$$\left. - \frac{h_-}{2\sqrt{2}} (n + N') + \frac{h_+}{\sqrt{2}} \rho_{+-} \right],$$

$$\partial_t \rho_{+-} = -\frac{\rho_{+-}}{\tau_1} + i\gamma \left[ 2H_{\text{eff}}^{(i)} \rho_{+-} + \frac{h_-}{\sqrt{2}} (\rho_{0-} - \rho_{+0}) \right], \quad (7)$$

where  $n = \rho_{++} - \rho_{--}$ ,  $1/\tau_0 = r_s + d + 1/\tau_s$ ,  $1/\bar{\tau} = \bar{r} + d + 1/\tau_s + r'/6$ ,  $1/\tau' = 1/\bar{\tau} + r'/6$ ,  $1/\tau_1 = 1/\bar{\tau} + r'/2$ ,  $\gamma = (\gamma_1 + \gamma_2)/2$ ,  $H_{\text{eff}}^{(i)} = (\gamma_1 H_1^{(i)} + \gamma_2 H_2^{(i)})/2\gamma$ .

Equations (7) differ from those obtained in [28] at the point that they take the hyperfine interaction into account. It is just the fact that allows one to use them for the treatment of the real EMDR spectra of triplet recombination centers in silicon and the theoretical estimation of the most important parameters of such centers.

The exchange constant  $J$  doesn't enter into Eqs. (7). It entered only into the equation for the quantities  $\rho_{s\alpha}$ ,  $\alpha = 0, +, -$ . In the limiting case  $J \rightarrow \infty$ , these quantities approach zero, as the strong exchange interaction between electrons forbids singlet-triplet transitions. As  $\gamma_1 \approx \gamma_2$ , the effective field  $H_{\text{eff}}^{(i)}$  represents simply a sum of the external constant magnetic field and hyperfine fields of the first and second electrons.

With the help of the substitutions

$$\rho_{+0} - \rho_{0-} = (x_1 + ix_2) \exp(i\omega t),$$

$$\rho_{+0} + \rho_{0-} = (x_3 + ix_4) \exp(i\omega t),$$

$$\rho_{+-} = (x_5 + ix_6) \exp(2i\omega t),$$

$$h_{\pm} = h_x \pm h_y = h \exp(\mp i\omega t),$$

we can pass from the system of differential equations (7) to the system of linear equations with real coefficients

$$\sum_{k=1}^6 c_{ik} x_k = b_i, \quad (8)$$

where

$$c_{11} = c_{22} = c_{33} = c_{44} = \bar{\tau} \left( \gamma H_{\text{eff}}^{(i)} - \omega \right),$$

$$c_{55} = c_{66} = \tau_1 \left( \gamma H_{\text{eff}}^{(i)} - \omega \right),$$

$$c_{12} = -1 - 3(h\tau'\gamma)(h\bar{\tau}\gamma),$$

$$c_{13} = c_{31} = c_{24} = c_{42} = -D\bar{\tau}/\hbar,$$

$$c_{15} = c_{26} = \sqrt{2}h\bar{\tau}\gamma,$$

$$c_{51} = c_{62} = \sqrt{2}h\tau_1\gamma/4,$$

$$c_{21} = c_{43} = -2c_{56} = 2c_{65} = 1,$$

$$c_{34} = -\left[ 1 + (h\bar{\tau}\gamma)^2 (\tau_1/\bar{\tau}) \right],$$

$$b_1 = \frac{\sqrt{2}}{3} (h\tau'\gamma)(r'\bar{\tau}) \times$$

$$\times \left[ \frac{g}{d} \left( 1 - \frac{\tau_0}{4} \left( d + \frac{1}{\tau_s} \right) \right) - \frac{R}{d} \left( 1 - \frac{\tau_0}{4\tau_s} \right) \right],$$

and all other coefficients  $c_{ik}$  and  $b_i$  are equal to zero. In order to close system (8), it's necessary to express the recombination rate in terms of the unknown  $x_i$ :

$$R = \frac{g}{d} \left\{ r_s + \left[ 1 - \frac{\tau_0}{4} \left( d + \frac{1}{\tau_s} \right) \right] \left[ \bar{r} - r_s - \frac{2}{9} r'^2 \tau' \right] \right\} - \sqrt{2}h\tau'\gamma r' x_2. \quad (9)$$

The system of equations (8), (9) completely describes SDR in terms of excited triplet states and is applicable to both numerical and analytical calculations. The effective fields  $H_{\text{eff}}^{(i)}$  depend on the location of nuclei, that have a nuclear spin, relative to the recombination center. In the case where these nuclei are chaotically distributed in a sample, obtaining a resonance relative change of the recombination rate requires to average over various values of the fields  $H_{\text{eff}}^{(i)}$ :

$$\begin{aligned} \frac{\Delta R}{R} &= \frac{R(H_0, h) - R(H_0, 0)}{R(H_0, 0)} = \\ &= \frac{\sum_i P_i \left[ R\left(H_{\text{eff}}^{(i)}, h\right) - R\left(H_{\text{eff}}^{(i)}, 0\right) \right]}{\sum_i P_i R\left(H_{\text{eff}}^{(i)}, 0\right)}, \end{aligned} \quad (10)$$

where  $P_i$  is the probability of the realization of a specific value of  $H_{\text{eff}}^{(i)}$ .

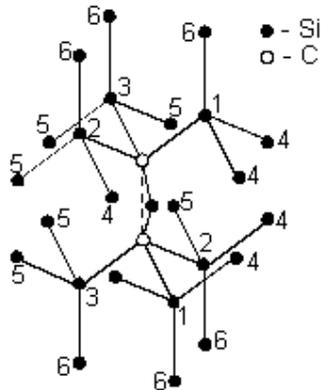


Fig. 1. Diagram of a recombination center consisting of two substitutional C atoms and one interstitial Si atom

### 3. Calculation of Peaks of Nonequilibrium Conductivity

The proposed model can be used for the mathematical treatment of experimental results obtained in the course of investigations of irradiated silicon [2,26,27]. For example, let's consider a Si-PT1 spectrum [27]. The spectrum consists of two intense dominant lines, each of them having several satellites. At low temperatures, one observes the hyperfine structure of the dominant lines. In order to explain the structure of this spectrum, a model was proposed, according to which the recombination center consists of two substitutional carbon atoms and one interstitial silicon atom "pushed" out of the C-C line [2,27]. The diagram of the center is presented in Fig. 1.

The structure of the spectrum is completely explained by the hyperfine interaction of electrons in the excited triplet state and Si-29 nuclei that have a nuclear spin  $I = 1/2$  and the natural abundance of 4.75%. The distance between the dominant lines is determined by the coefficient  $D$  in Hamiltonian (1) that characterizes crystalline anisotropy. The appearance of satellites is concerned with the hyperfine interaction between the localized electrons and the interstitial silicon atom. The structure of the dominant lines is caused by the hyperfine interaction with 24 nearest silicon atoms [2,27]. In the figure, these atoms are marked with numbers 1-6.

Experimentally, one determines a resonance change of the nonequilibrium conductivity  $\Delta\sigma$ :

$$d(\delta\Delta\sigma/\Delta\sigma)/dH_0 = -d(\Delta R/R)/dH_0. \quad (11)$$

When calculating the resonance peaks of conductivity, one can consider that the hyperfine structure is conditioned only by Si-29 nuclei distributed

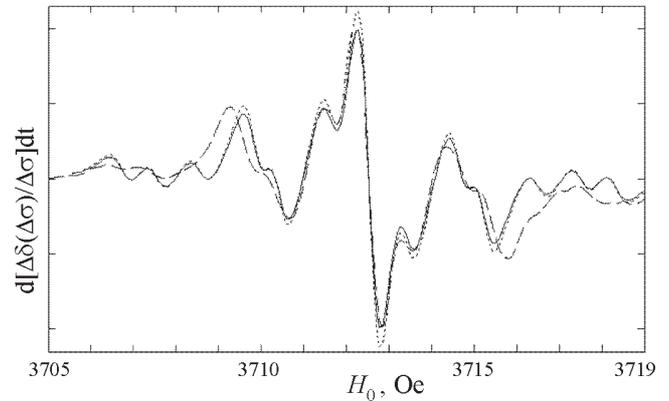


Fig. 2. Experimental (dotted line) and calculated [solid ( $\tau_s = 10^{-3}$  s) and dashed ( $\tau_s = 5 \times 10^{-4}$  s) lines] resonance spectra of nonequilibrium conductivity for a Si-PT1 center

in a sample completely randomly, and the opposite directions of nuclear spins are equally probable.

In order to explain the structure of the dominant lines of the spectrum, we considered the hyperfine interaction of localized electrons that form the spin  $S = 1$  with six groups of the surrounding atoms (marked with numbers from 1 to 6 in Fig. 1). Each of these groups is characterized with a certain number of nuclei (groups 1-3 consist of six nuclei, and groups 4-6 contain two of them) and hyperfine fields  $H_N^i$  induced by nuclei of the  $i$ -th group at localized electrons. These fields  $H_N^i$  were considered as fitting parameters. That is, their value was chosen to attain the best agreement between theory and experiment.

The spectrum depicted in Fig. 2 is calculated for the following values of the parameters:  $f = 9.213648$  GHz,  $h = 0.35$  Oe,  $\tau_s = 10^{-3}$  s,  $d = 10^3$  s $^{-1}$ ,  $r_s = 10^4$  s $^{-1}$ ,  $r' = 2 \times 10^2$  s $^{-1}$ ,  $\bar{r} = 10^3$  s $^{-1}$ ,  $H_N^1 = 5.81$  Oe,  $H_N^2 = 4.99$  Oe,  $H_N^3 = 3.97$  Oe,  $H_N^4 = 2.7$  Oe,  $H_N^5 = 2.1$  Oe,  $H_N^6 = 0.87$  Oe. In the figure, we give also the experimental spectrum (dotted line) [27]. It is seen that the theory describes the experimental data well enough in the case of realistic values of the parameters of Hamiltonian (4). It's worth noting that the form of the calculated spectrum is extremely sensitive to the amplitude of the alternating field and parameters that characterize the hyperfine interaction. In order to illustrate the influence of dynamic parameters, the spectrum calculated for the above-indicated parameters with the only difference -  $\tau_s = 5 \times 10^{-4}$  s is presented in Fig. 2 (dashed line). One can see that the decrease in the time of spin relaxation results only in the increase of the intensities of maxima.

The obtained set of the constants of hyperfine interaction characterizes the coordinate part of the wave function of electrons that form the excited triplet state. The values of the constants of hyperfine interaction  $A_N^i = 2\gamma_{1,2}\hbar H_N^i$  can be used for clarifying the structure of the crystal field that forms the wave function of electrons of the Si-PT1 center. In a similar way, one can also consider other kinds of paramagnetic centers.

#### 4. Conclusions

The quantum-mechanical equations (3) for the density operator used in [20,21] for the theoretical description of SDR according to the Kaplan–Solomon–Mott mechanism are also acceptable for the theoretical treatment of real EMDR spectra of triplet centers in silicon. Such a treatment was successfully performed only due to the fact that, in contrast to works [2,26,27], the dynamics of electrons within the proposed approach is based on the first principles of quantum mechanics rather than on phenomenological additives to the classical equations.

Thus, we have used the proposed theory for the treatment of experimental data, by studying SDR in terms of excited triplet states. This allowed us to investigate the dependence of the structure of the spectrum on its parameters and to determine these parameters by the form of the measured spectrum, as well as their errors.

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ТЕОРІЯ ЕЛЕКТРИЧНО ДЕТЕКТОВАНОГО  
МАГНІТНОГО РЕЗОНАНСУ ТРИПЛЕТНИХ  
ЦЕНТРІВ У КРЕМНІЇ*О.В. Барабанов, Р.А. Хорошок*

## Резюме

Запропоновано теоретичну модель спін-залежної рекомбінації через збуджені триплетні стани центрів у забороненій

зоні напівпровідника. Теорію побудовано на основі квантовомеханічних рівнянь для оператора густини в просторі спінових станів двоелектронної системи. За допомогою цих рівнянь проаналізовано положення та форму експериментальних резонансних піків нерівноважної провідності. Розгляд проведено з врахуванням зееманівської та надтонкої взаємодій, кристалічної анізотропії, а також процесів генерації, рекомбінації, дисоціації та спінової релаксації локалізованих електронів. Проведено порівняння теоретичних результатів з експериментальними.