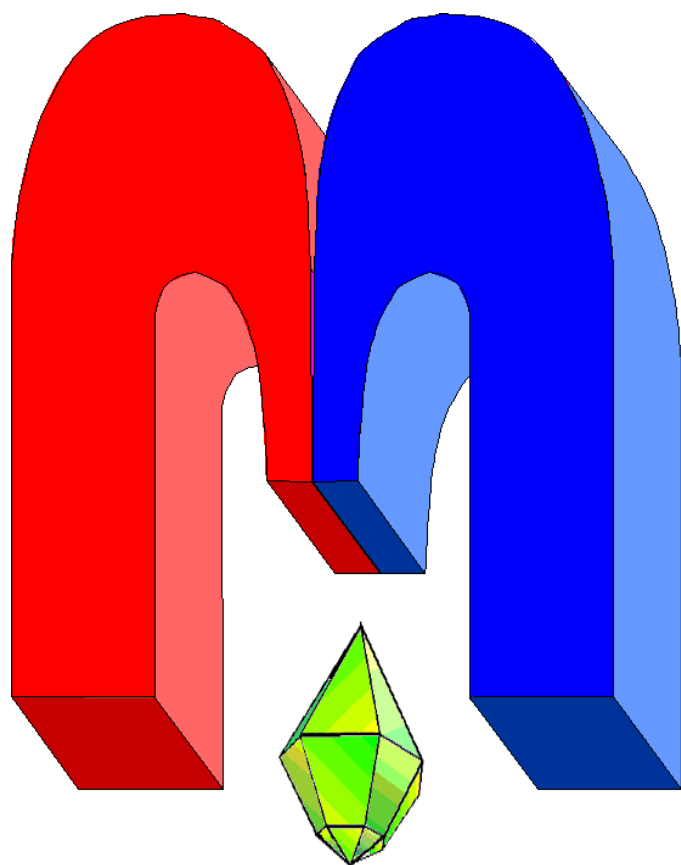


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In Kazan University the Electron Paramagnetic Resonance (EPR) was discovered by Zavoisky E.K. in 1944.

Temperature dependence of the conduction electron g -factor in silicon: theory and experiment

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Temperature dependence of the conduction electron Lande g -factor in silicon has been investigated both theoretically and experimentally. Theoretical consideration is based on the renormalization of the electron energy in the magnetic field by the electron-phonon interaction in the second-order perturbation theory. Interaction with lattice vibrations decreases the conduction electron g -factor. The g -factor was measured in the electron spin resonance (ESR) experiments for n-Si samples. In the high temperature limit the g -factor linearly decreases with temperature in good agreement with the experimental data.

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Keywords: g -factor, electron-phonon interaction, spin flip, renormalization, temperature dependence, conduction electron spin resonance

1. Introduction

Since pioneer demonstration of the spin injection and detection in Si [1], silicon becomes a perspective material for spintronics due to weak spin-orbit interaction, large spin relaxation time and diffusion length in comparison with typical III-V semiconductors such as GaAs. One of the most important characteristics of conduction electrons in various systems with spin-orbit coupling is the spin relaxation rate. Spin relaxation in silicon mainly occurs through the Elliott [2] and Yafet [3] mechanisms, where spin flip processes are caused by electron-phonon interaction. Quantitative theoretical studies of the Elliott-Yafet spin relaxation in silicon [4] yields the spin relaxation rate proportional to a third power of temperature. This fact is in good agreement with the experimental data. The spin relaxation rate can be extracted from the line width of the conduction electron spin resonance (CESR).

Another important characteristic of conduction electrons in magnetic field is their Lande g -factor, which is characterized by the position of the CESR line. First experimental investigations of the conduction electron g -factor were carried out by Wilson and Feher [5]. The measured value for “free” carriers was found to be $g = 1.99875 \pm 0.0001$. In silicon each conduction valley has an axial symmetry, so the g -factor becomes a tensor with two, longitudinal g_{\parallel} and transverse g_{\perp} , principal values. The measured value g is isotropic due to cubic symmetry of the Brillouin zone and has the average form over all 6 conduction band valleys:

$$g = \frac{1}{3}g_{\parallel} + \frac{2}{3}g_{\perp}. \quad (1)$$

In opposite to the spin relaxation rate, which temperature dependence is usually measured in experiments, the g -factor is supposed independent of temperature in a wide temperature range. However, such an assumption, generally speaking, should be verified. In our previous work [6] we first measured the temperature dependence of the electron g -factor in n-type silicon using ESR technique. It was suggested that this effect can be explained by the phonon modulation of the spin-orbit interaction in the system. So, in this work we discuss some theoretical approach to this problem and compare our calculations with experimental results.

2. The model

Theoretical study of the g -factor temperature dependence is based on the renormalization of the electron energy in the external magnetic field by the electron-phonon interaction. Within the framework of a many-body picture the renormalization of the excitation energies by lattice vibrations is represented by [7]

$$\varepsilon(\mathbf{k}) = \varepsilon^{(0)}(\mathbf{k}) + \text{Re}\Sigma^*(\varepsilon, \mathbf{k}), \quad (2)$$

where $\varepsilon^{(0)}(\mathbf{k})$ and $\varepsilon(\mathbf{k})$ are unperturbed and renormalized electron energies, respectively, and $\text{Re}\Sigma^*(\varepsilon, \mathbf{k})$ is the proper self-energy real part, that can be presented in covalent semiconductors, such as silicon, using the Rayleigh-Schrödinger perturbation theory [8]:

$$\text{Re}\Sigma^*(\varepsilon, \mathbf{k}) = \sum_{q\lambda} \sum_{s=\pm 1} \frac{|V_{\mathbf{k}\mathbf{k}+s\mathbf{q}}|^2 (1 - W_{\mathbf{k}+s\mathbf{q}}) (N_{q\lambda} + 1/2 - s/2)}{\varepsilon^{(0)}(\mathbf{k}) - \varepsilon^{(0)}(\mathbf{k} + s\mathbf{q}) + s\hbar\omega_\lambda(\mathbf{q})}, \quad (3)$$

where $V_{\mathbf{k}\mathbf{k}\pm\mathbf{q}}$ is the matrix element describing the electron momentum scattering from the state $|\mathbf{k}\rangle$ to the state $|\mathbf{k} \pm \mathbf{q}\rangle$ accompanied by absorption (upper sign) or emission (lower sign) of the phonon with the wave vector \mathbf{q} and polarization λ , $\omega_\lambda(\mathbf{q})$ and $N_{q\lambda}$ are the frequency and occupation number of the phonon, and $W_{\mathbf{k}}$ is the electron occupation probability for the state $|\mathbf{k}\rangle$. In what follows, we replace the phonon and electron occupation numbers by their equilibrium values describing the Bose-Einstein and Fermi-Dirac statistics, respectively.

If the spin-orbit interaction is taken into account, the electron states become the states with an effective generalized spin that differs from the standard spin moment. We will denote these states as effective spin-up $|\uparrow\rangle$ or spin-down $|\downarrow\rangle$ vectors. In the absence of magnetic field the spin-up and spin-down states have the same energies (the Kramers degeneracy). In the external magnetic field \mathbf{H} oriented along one of the conduction band valleys the electron energy has the additional Zeeman term $\sigma g_i \mu_B H$, where μ_B is the Bohr magneton, g_i is the g -tensor principal value, $i = \parallel, \perp$ for longitudinal and transverse components, respectively, and $\sigma = \pm 1/2$ are effective spin-projections on the magnetic field, and the electron wave vector \mathbf{k} transforms into $\mathbf{K} = \mathbf{k} + e\mathbf{A}/c\hbar$, where \mathbf{A} is the vector-potential of the magnetic field. Generally speaking, one has to consider the single-electron g -tensor as a function of the wave vector as well as the translatory-motion energy. Thereby, in the presence of the external magnetic field renormalization of the total electron energy has the form

$$\varepsilon(\mathbf{K}) + \sigma g_i(\mathbf{K}) \mu_B H = \varepsilon^{(0)}(\mathbf{K}) + \sigma g_i^{(0)}(\mathbf{K}) \mu_B H + \sum_{q\lambda\sigma'} \sum_{s=\pm 1} \frac{|V_{\mathbf{K}\sigma\mathbf{K}+s\mathbf{q}\sigma'}^{(i)}|^2 (1 - W_{\mathbf{K}+s\mathbf{q}\sigma'}^H) (N_{q\lambda} + 1/2 - s/2)}{\varepsilon^{(0)}(\mathbf{K}) - \varepsilon^{(0)}(\mathbf{K} + s\mathbf{q}) + (\sigma g_i^{(0)}(\mathbf{K}) - \sigma' g_i^{(0)}(\mathbf{K} + s\mathbf{q})) \mu_B H + s\hbar\omega_\lambda(\mathbf{q})}, \quad (4)$$

where $g_i^{(0)}$ and g_i are \mathbf{k} -dependent unperturbed and renormalized principal values of the g -tensor, respectively, $W_{\mathbf{K}\sigma}^H$ is the electron distribution function in the $|\mathbf{K}, \sigma\rangle$ state in the presence of the external magnetic field, $V_{\mathbf{K}\sigma\mathbf{K}\pm\mathbf{q}\sigma'}^{(i)}$ is the spin-dependent momentum scattering matrix element for the electron in the i -th valley.

The renormalized term in the expression (4) can be expanded into the Taylor series using standard approaches:

$$\mu_B g_i(\mathbf{K}) H \ll \varepsilon(\mathbf{K}),$$

and

$$W_{\mathbf{K}\sigma}^H \approx W_{\mathbf{K}} + \frac{dW_{\mathbf{K}}}{d\varepsilon^{(0)}(\mathbf{K})} \sigma \mu_B g_i^{(0)}(\mathbf{K}) H \quad (5)$$

for a case of a weak magnetic field. Thereby, one can obtain an explicit expression for the renormalized g -tensor principal values:

$$\begin{aligned} \sigma g_i(\mathbf{K}) = & \sigma g_i^{(0)}(\mathbf{K}) - \sum_{\mathbf{q}\lambda\sigma'} \sum_{s=\pm 1} \left(\sigma g_i^{(0)}(\mathbf{K}) - \sigma' g_i^{(0)}(\mathbf{K} + s\mathbf{q}) \right) \frac{|V_{\mathbf{K}\sigma\mathbf{K}+s\mathbf{q}\sigma'}^{(i)}|^2 (1 - W_{\mathbf{K}+s\mathbf{q}}) (N_{\mathbf{q}\lambda} + 1/2 - s/2)}{(\varepsilon^{(0)}(\mathbf{K}) - \varepsilon^{(0)}(\mathbf{K} + s\mathbf{q}) + s\hbar\omega_{\lambda}(\mathbf{q}))^2} \\ & - \sum_{\mathbf{q}\lambda\sigma'} \sum_{s=\pm 1} \sigma' g_i^{(0)}(\mathbf{K} + s\mathbf{q}) \frac{|V_{\mathbf{K}\sigma\mathbf{K}+s\mathbf{q}\sigma'}^{(i)}|^2 (N_{\mathbf{q}\lambda} + 1/2 - s/2) dW_{\mathbf{K}+s\mathbf{q}}/d\varepsilon^{(0)}(\mathbf{K} + s\mathbf{q})}{\varepsilon^{(0)}(\mathbf{K}) - \varepsilon^{(0)}(\mathbf{K} + s\mathbf{q}) + s\hbar\omega_{\lambda}(\mathbf{q})}. \end{aligned} \quad (6)$$

As seen, the g -factor correction consists of two parts. The first part comes from the denominator of the self-energy, while the second one comes from the expansion of the electron distribution function into the Taylor series. Due to the weakness of the g -factor dispersion in semiconductors, it is possible to neglect the contribution of the term $\sigma' = \sigma$ in the first sum. On the contrary, in the second sum we neglect the term $\sigma' = -\sigma$ because of small intensity of spin-flip scatterings compared to spin-preserving processes [2]. Finally, introducing the g -factor correction $\delta g_i(\mathbf{K}) = g_i(\mathbf{K}) - g_i^{(0)}(\mathbf{K})$, one obtain

$$\begin{aligned} \delta g_i(\mathbf{K}) = & - \sum_{\mathbf{q}\lambda} \sum_{s=\pm 1} \left(g_i^{(0)}(\mathbf{K}) + g_i^{(0)}(\mathbf{K} + s\mathbf{q}) \right) \frac{|V_{\mathbf{K}\uparrow\mathbf{K}+s\mathbf{q}\downarrow}^{(i)}|^2 (1 - W_{\mathbf{K}+s\mathbf{q}}) (N_{\mathbf{q}\lambda} + 1/2 - s/2)}{(\varepsilon^{(0)}(\mathbf{K}) - \varepsilon^{(0)}(\mathbf{K} \pm \mathbf{q}) \pm \hbar\omega_{\lambda}(\mathbf{q}))^2} \\ & - \sum_{\mathbf{q}\lambda} g_i^{(0)}(\mathbf{K} + s\mathbf{q}) \frac{|V_{\mathbf{K}\uparrow\mathbf{K}+s\mathbf{q}\uparrow}^{(i)}|^2 (N_{\mathbf{q}\lambda} + 1/2 - s/2) dW_{\mathbf{K}+s\mathbf{q}}/d\varepsilon^{(0)}(\mathbf{K} + s\mathbf{q})}{\varepsilon^{(0)}(\mathbf{K}) - \varepsilon^{(0)}(\mathbf{K} + s\mathbf{q}) + s\hbar\omega_{\lambda}(\mathbf{q})}, \end{aligned} \quad (7)$$

where $V_{\mathbf{K}\uparrow\mathbf{K}\pm\mathbf{q}\downarrow}^{(i)}$ and $V_{\mathbf{K}\uparrow\mathbf{K}\pm\mathbf{q}\uparrow}^{(i)} = V_{\mathbf{K}\downarrow\mathbf{K}\pm\mathbf{q}\downarrow}^{(i)}$ are spin-flip and spin-preserving scattering matrix elements, respectively. The first term in (7) describes the renormalization of the g -factor caused by the spin-flip processes in the second-order perturbation theory according to the Elliott-Yafet theory of the spin relaxation [3,4]. The second term appears in Eq. (7) from the second-order spin-preserving scattering in the presence of a weak magnetic field due to the difference in the electron occupation numbers for the spin-up and spin-down states (such a difference provides paramagnetic properties for some materials [9]).

In the ESR experiments the g -factor value is extracted from the maximum of the resonance line. The average value of the absorbed photon energy $\hbar\Omega$ can be found as the relationship: $\hbar\Omega = E_{abs}/N$, where N stands for the number of absorbed photons. The denominator of the last fraction coincides with the difference between electron occupation numbers for spin-down and spin-up states: $N = \sum_{\mathbf{K}} W_{\mathbf{K}\downarrow}^H - W_{\mathbf{K}\uparrow}^H$, while the full absorbed energy can be computed as the multiplication of the photon energy on the occupation number difference: $E_{abs} = \sum_{\mathbf{K}} g_i(\mathbf{K}) \mu_B H (W_{\mathbf{K}\downarrow}^H - W_{\mathbf{K}\uparrow}^H)$. Using the second expression in (5), the difference $W_{\mathbf{K}\downarrow}^H - W_{\mathbf{K}\uparrow}^H$ can be expanded into series. The final evaluation can be performed with replacing \mathbf{K} by \mathbf{k} [3]. Thus, the observable i -th principal value of the electron g -tensor, $g_i(T) = \hbar\Omega/\mu_B H$, can be calculated as

$$g_i(T) = \frac{\sum_{\mathbf{k}} g_i^2(\mathbf{k}) dW_{\mathbf{k}}/d\varepsilon}{\sum_{\mathbf{k}} g_i(\mathbf{k}) dW_{\mathbf{k}}/d\varepsilon}. \quad (8)$$

When obtaining Eq. (8) some approximations has been used. First, the dispersion of the single-electron g -factor is strong in metals, where electrons are situated near the Fermi surface with a shape, which can be essentially non-spherical. In semiconductor, electrons populate the bottom of the conduction band, so one can neglect the dispersion of the single-electron g -factor in the expression (8). Thereby, the measured g -factor value can be substituted on the g -factor value of the conduction band edge: $g_i(T) \approx g_i(\mathbf{k}_0)$, where \mathbf{k}_0 is the position of the conduction band minima in the Brillouin zone. Besides, as seen from the Eq. (7), the g -factor renormalization becomes greater if the electron energies $\varepsilon^{(0)}(\mathbf{k})$ and $\varepsilon^{(0)}(\mathbf{k} \pm \mathbf{q})$ (as well as the g -factors) have close values. Consequently, we can neglect the zero-order g -factor dispersion in the Eq. (7). Therefore, the measured value of the conduction electron g -factor is

$$g_i(T) = g_i^{(0)} - 2g_i^{(0)} \sum_{q\lambda} \sum_{s=\pm 1} \frac{|V_{\uparrow\downarrow}^{(i)}(\mathbf{q})|^2 (1 - W_{\mathbf{k}_0 + s\mathbf{q}}) (N_{q\lambda} + 1/2 - s/2)}{(\varepsilon^{(0)}(\mathbf{k}_0) - \varepsilon^{(0)}(\mathbf{k}_0 + s\mathbf{q}) + s\hbar\omega_\lambda(\mathbf{q}))^2} - g_i^{(0)} \sum_{q\lambda} \sum_{s=\pm 1} \frac{|V_{\uparrow\uparrow}^{(i)}(\mathbf{q})|^2 (N_{q\lambda} + 1/2 - s/2) dW_{\mathbf{k}_0 + s\mathbf{q}}/d\varepsilon^{(0)}(\mathbf{k}_0 + s\mathbf{q})}{\varepsilon^{(0)}(\mathbf{k}_0) - \varepsilon^{(0)}(\mathbf{k}_0 + s\mathbf{q}) + s\hbar\omega_\lambda(\mathbf{q})}, \quad (9)$$

where $g_i^{(0)} = g_i^{(0)}(\mathbf{k}_0)$, $V_{\uparrow\uparrow}^{(i)}(\mathbf{q}) = V_{\downarrow\downarrow}^{(i)}(\mathbf{q})$ and $V_{\uparrow\downarrow}^{(i)}(\mathbf{q})$ are matrix elements for spin-preserving and spin-flip scatterings from the conduction band minima $|\mathbf{k}_0\rangle$ to the state $|\mathbf{k}_0 \pm \mathbf{q}\rangle$, respectively. In the case of intrinsic or nondegenerate n-type silicon we can neglect the electron occupation number in the expression (9). Therefore, the final expression for the g -tensor eigenvalue is

$$g_i(T) = g_i^{(0)} - 2g_i^{(0)} \sum_{q\lambda} \sum_{s=\pm 1} \frac{|V_{\uparrow\downarrow}^{(i)}(\mathbf{q})|^2 (N_{q\lambda} + 1/2 - s/2)}{(\varepsilon^{(0)}(\mathbf{k}_0) - \varepsilon^{(0)}(\mathbf{k}_0 + s\mathbf{q}) + s\hbar\omega_\lambda(\mathbf{q}))^2}. \quad (10)$$

Considering the zero-temperature limit we should set $N_{q\lambda} = 0$ in Eq. (10). In this case Eq. (10) yields the zero-temperature renormalization of the g -factor by emitting and subsequent absorbing virtual phonon. Evidently, measured low-temperature conduction electron g -factor $g \approx 1.9987$ by Wilson and Feher, includes this correction. Consequently, we will use the low-temperature experimental value as the zeroth approximation:

$$g_i(0) = g_i^{(0)} - 2g_i^{(0)} \sum_{q\lambda} \frac{|V_{\uparrow\downarrow}^{(i)}(\mathbf{q})|^2}{(\varepsilon^{(0)}(\mathbf{k}_0) - \varepsilon^{(0)}(\mathbf{k}_0 - \mathbf{q}) - \hbar\omega_\lambda(\mathbf{q}))^2} = 1.9987. \quad (11)$$

Due to the small difference between $g_i(0)$ and $g_i^{(0)}$ one can replace $g_i^{(0)}$ by $g_i(0)$ in the Eq. (10), so that

$$\frac{\delta g_i(T)}{g_i(0)} = -2 \sum_{q\lambda} \sum_{s=\pm 1} \frac{|V_{\uparrow\downarrow}^{(i)}(\mathbf{q})|^2 N_{q\lambda}}{(\varepsilon^{(0)}(\mathbf{k}_0) - \varepsilon^{(0)}(\mathbf{k}_0 + s\mathbf{q}) + s\hbar\omega_\lambda(\mathbf{q}))^2}, \quad (12)$$

where $\delta g_i(T) = g_i(T) - g_i(0)$ is the temperature-dependent correction to the g -tensor eigenvalue. Thus, the temperature dependence of the g -factor in our approach is formed by phonon-induced spin-flip processes between initial, intermediate and final electron states, where the final state completely coincides with the initial one.

Yafet has shown that, for the spin flip processes in silicon, $V_{\uparrow\downarrow}^{(i)}(\mathbf{q}) \sim \mathbf{q}^2$ in contrast to the spin preserving scattering matrix element that is proportional to \mathbf{q} . As a result, in expression (12) we set

$$|V_{\uparrow\downarrow}^{(i)}(\mathbf{q})| = A_i q^2 \sqrt{\hbar/2\rho V \omega_\lambda(\mathbf{q})}, \quad (13)$$

where the factor $\sqrt{\hbar/2\rho V \omega_\lambda(\mathbf{q})}$ is the normalized coefficient in the electron-phonon interaction definition [10], ρ and V are the silicon density and the sample volume, respectively, A_i are some constants symbolically referred to as spin deformation potentials. Substituting (13) into (12), and counting the electron energy from the conduction-band bottom $\varepsilon^{(0)}(\mathbf{k}_0)$, and then turning the summation over \mathbf{q} into integration, one obtain the T -dependent correction $\delta g_i(T)$ in the following form:

$$\frac{\delta g_i(T)}{g_i(0)} = -\frac{\hbar}{\rho(2\pi)^3} \sum_\lambda \sum_{s=\pm 1} \int d\mathbf{q} \frac{A_i^2 q^4 N_{q\lambda}}{\omega_\lambda(\mathbf{q})(s\hbar\omega_\lambda(\mathbf{q}) - \varepsilon^{(0)}(\mathbf{k}_0 + s\mathbf{q}))^2}. \quad (14)$$

As seen from the expression (14), the scattering with the short-wavelength phonons mainly contributes to the g -factor renormalization. Therefore, we further consider the intervalley scattering. In that case the phonon frequency is almost independent on the wave vector, and can be treated as a constant. Using spherical approximation for the electron energy, it is possible to obtain the following estimation:

$$\frac{\delta g_i(T)}{g_i(0)} = -\frac{A_i^2 q_0^4}{2\rho\pi\hbar} \sum_\lambda N(\omega_\lambda) \left(\frac{2m}{\hbar\omega_\lambda} \right)^{3/2}, \quad (15)$$

where $\omega_\lambda = \omega_\lambda(\mathbf{q}_0)$ is a mean frequency of the intervalley f -phonon, $m \approx 0.26m_0$ is the average electron effective mass, and m_0 stands for the free electron mass. Thus, the temperature dependence of δg_i is caused by the temperature dependence of the phonon occupation numbers $N(\omega_\lambda)$.

Using Eq. (1), numerical values of the acoustic phonon frequencies [11], the spin-flip matrix elements [4], and experimentally observed low-temperature values of the g -tensor components [5] we calculate average (over all 6 valleys) temperature dependence of the conduction electron g -factor in silicon. The results of our calculations are presented in Fig. 1 by the solid line. It is seen that, the g -factor slightly decreases with temperature increasing, and its total variation is about 0.0004 for T ranging within 10-250 K.

3. Comparison with experimental results

We studied the electron g -factor in ESR spectra measurements. We have observed dependence of the g -factor of conduction electrons in silicon on temperature and found that in temperature range 80-250 K they are alike for different donors and their concentrations. The measurements are performed on the spectrometer ‘‘Bruker EMX 10/12’’ using helium cryostat with a system of a temperature control (3.8-300 K) ‘‘ER 4112 HV’’. We use natural silicon samples doped with lithium and phosphorus to make some concentration of electrons in the conduction band. Donor concentrations in both samples were very close to minimize difference in the spin flip process intensity.

The experimental results are presented in Fig. 1 together with the theoretical curve. As temperature increases from zero to ~ 80 K, a monotonous rise of the g -factor value takes place. We suppose that in this temperature range electrons are still localized on the donor centres and have the discrete energy spectra. Each of these discrete states can be described by different g -factors, and

changing the electron distribution among these states with the temperature rising can lead to rising of the g -factor. Consideration of this effect in more details goes beyond the scope of the present paper.

However at higher temperatures the donors become ionized and the electrons can propagate over the sample. In this temperature range one can see approximately linear decrease of the g -factor for both samples, which qualitatively agrees with prediction for conduction electrons. The linear approximation of the experimental results for a temperature range over 80 K has shown, that the absolute value of the experimentally observed coefficient of the g -factor linear T -dependence is about $4 \cdot 10^{-6} \text{ K}^{-1}$ independently of the donor type. This is approximately two times greater than the one predicted theoretically. Thus, more accurate calculations are required.

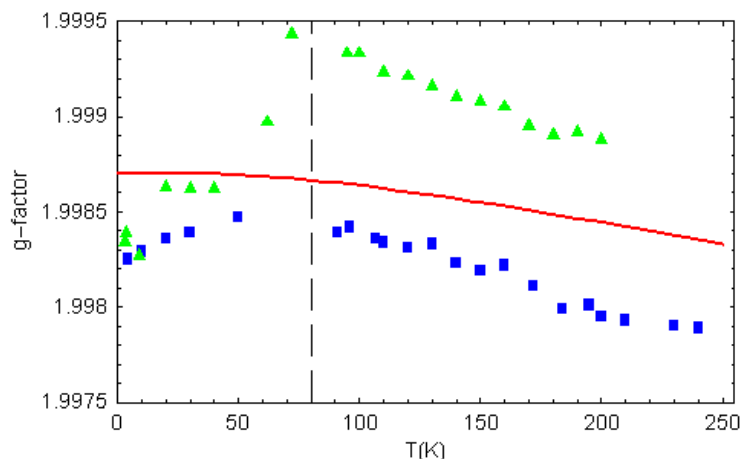


Figure 1. Calculated (red solid line) and measured temperature dependences of the average electron g -factor in Si sample doped with phosphorus (blue rectangles) and lithium (green triangles), whose concentrations are $3.3 \cdot 10^{18} \text{ cm}^{-3}$ and $3.7 \cdot 10^{18} \text{ cm}^{-3}$, respectively. Vertical dashed line at 80 K shows a relative border between localized and propagating electrons.

Also, CESR measurements yield different values of the g -factor for Si samples doped with different donors, especially at T greater than $\sim 80 \text{ K}$. The theory developed in the present paper does not describe this effect. It is, however, clear that, the spin-flip scattering due to the impurity centres is similar to that takes place due to the lattice vibrations, and, therefore, can modify the g -factor as well. Correspondingly, the complete quantitative analysis have to include calculations of the spin-flip matrix element as a function of the donor type. However, this problem goes beyond the frame of our paper, and needs special study.

4. Conclusion

Generally speaking, the g -factor modification in silicon has a similar nature to the spin relaxation process. In this work we have shown, that the Elliott-Yafet mechanism of the spin-flip scattering leads to the conduction electron g -factor renormalization. The generalization of this result onto the other channels of the spin-flip process, such as impurity or nuclear spin scattering, can be considered as a fundamental problem of the solid-state spin physics, which has been waiting for its solving.

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