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Photon echo in germanium with shallow donors

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A theoretical study was made of the conditions for observing the photon echo effect in a germanium crystal doped with shallow donors. A numerical calculation of the medium polarization excited by a sequence of two optical pulses at a frequency close to the impurity transition frequency has been made. The effect of excitation pulse parameters, such as the pulse duration, the inhomogeneous broadening of impurity transitions and the relaxation rate of population and coherence in the system on the echo is considered. The key aspect in the experimental implementation of the effect is the control of the crystal temperature under photoexcitation, since the rate of coherence relaxation in the system strongly depends on the temperature of the crystal lattice.

Keywords: germanium, shallow donors, coherent effects, photon echo.

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1. Introduction

Presently, the scientific activity of many researchers is focused on studying the quantum optically-controlled coherent states of various systems and on creating new semiconductor devices based thereon, which are coupled to advanced silicon and germanium technologies [1–6]. One of the candidates is regarded to be Coulomb centers in the semiconductors. The impurity centers in germanium are quite well studied, but up to now there is still no information on their relaxation times of populations of the states and the coherence. The experiment for observation of the photon echo is an effective method of demonstrating that it is possible to create the coherent ensemble of dipoles formed by the donor centers and being in a superposition of the states, and measurement of a cross time of relaxation in the system. The photon echo effect is successfully demonstrated in silicon doped with shallow phosphorous donors when exciting the system at the transition $1s(A_1)-2p_\pm$ [7]. The purpose of this study is to theoretically check the possibility of and determine the conditions of observation of this effect in germanium with shallow donors. It included the numerical modelling of polarization of the medium (germanium with shallow donors), which is excited by a sequence of two optical pulses ($\pi/2$- and π-pulses) at the frequency near the frequency of the transition allowed in an electro-dipole approximation. The medium parameters were simulated by using samples corresponding to real samples of germanium with doping concentrations of $10^{12}-10^{15}\,\mathrm{cm}^{-3}$. The analysis was also aimed at checking the usability of the Novosibirsk free-electron laser (NovoFEL) as an excitement source, whose pulse duration is ~ 100 ps, which is less than typical lifetimes of the Ge donor states in less than 10 times and longer than the reverse width of a heterogeneously widened line of the impurity transitions with doping concentrations $10^{14}-10^{15}\,\mathrm{s}^{-1}$. It has been shown that at these parameters in n-Ge after exposure to the π-pulse, the medium polarization began to increase and in the time approximately equal to the time of delay between the pulses it came to the maximum, thereby corresponding to the photon echo in the studied system.

2. Theoretical model

The theoretical description was made up within a semi-classical model, which regards the impurity atom as a two-level quantum system and classically describes the electromagnetic field (two subsequent pulses of the electromagnetic radiation at the frequency near the frequency of the impurity transition). The approximation of a rotating wave was applied. When neglecting the interaction with the lattice oscillations, the Hamiltonian takes this form

$$H = H_0 + \mu E_1(t)\cos(\nu t) + \mu E_2(t-\tau)\cos(\nu t + \varphi), \quad (1)$$

where H_0 — the Hamiltonian of the unperturbed system, μ — the dipole moment of the transition, $E_{1,2}(t)$ — the time-dependent amplitudes of the fields of two pulses of external radiation, ν — the circular frequency of this radiation, τ — the time delay between the two pulses, φ — the phase radiation difference in the first and second pulses defined by τ.

If neglecting the relaxation both of the population (the longitudinal relaxation) and of the coherence (the transverse relaxation), then in this case it is convenient to make up the description of the interaction of the atom with the field using the method of probability amplitudes [8], which can express the wave function of the systems by the following superposition:

$$\Psi(t) = \alpha_1(t)\varphi_1 e^{-i\omega_1 t} + \alpha_2(t)\varphi_2 e^{-i\omega_2 t}, \quad (2)$$

where φ_1 and φ_2 — the field-unperturbed functions of the atom (the eigen functions of the H_0 operator), ω_1, ω_2 —

the frequencies corresponding to the energies of the atom levels. By substituting the wave function (2) in the Hamiltonian (1), after simple mathematical operations we can obtain a system of the equations in relation to the probability amplitudes, which specifies the dynamics of the system under action of the sequence of the two resonance pulses of the electromagnetic field:

$$\begin{cases} \alpha_1' = -\dfrac{i}{2}\alpha_2\left(\Omega_1(t)e^{i\delta t} + \Omega_2(t-\tau)e^{i(\delta t+\varphi)}\right), \\ \alpha_2' = -\dfrac{i}{2}\alpha_1\left(\Omega_1(t)e^{-i\delta t} + \Omega_2(t-\tau)e^{-i(\delta t+\varphi)}\right), \end{cases} \tag{3}$$

where Ω_1 and Ω_2 — the time-dependent Rabi frequencies for the fields correlated to the first and second pulses, respectively, which are expressed as follows:

$$\Omega_{1,2}(t) = \frac{\mu_{21}E_{1,2}(t)}{\hbar}. \tag{4}$$

Here, μ_{21} — the matrix element of the transition, δ — the detuning value. However, it is very difficult to take into account the relaxation in the system in relation to the probability amplitudes. It is convenient to switch to describing the system by real values, such as the populations of the levels and the real and imaginary parts of the dipole moment of the impurity center[1]:

$$\begin{cases} N' = -\Omega_1 R_2 - \Omega_2 T_2 - \gamma_1(1+N), \\ R_2' = N(\Omega_1 + \Omega_2\cos\varphi) + \delta R_1 - \gamma_t R_2, \\ T_2' = N(\Omega_1\cos\varphi + \Omega_2) + \delta T_1 - \gamma_t T_2, \\ R_1' = N\Omega_2\sin\varphi - \delta R_2 - \gamma_t R_1, \\ T_1' = -N\Omega_1\sin\varphi - \delta T_2 - \gamma_t T_1, \end{cases} \tag{5}$$

This system is conventionally supplemented by relaxation summands, which determine rates of the longitudinal (γ_1) and the transverse (γ_t) relaxation (the relaxation of the population and the coherence, respectively). The vectors N_1, R_1, R_2, T_1, T_2 are expressed by the probability amplitudes as follows:

$$\begin{aligned} N &= \alpha_2\alpha_2^* - \alpha_1\alpha_1^*, \\ R_1 &= \alpha_1^*\alpha_2 e^{i\delta t} + \alpha_1\alpha_2^* e^{-i\delta t}, \\ iR_2 &= \alpha_1^*\alpha_2 e^{i\delta t} - \alpha_1\alpha_2^* e^{-i\delta t}, \\ T_1 &= \alpha_1^*\alpha_2 e^{i(\delta t+\varphi)} + \alpha_1\alpha_2^* e^{-i(\delta t+\varphi)}, \\ iT_2 &= \alpha_1^*\alpha_2 e^{i(\delta t+\varphi)} - \alpha_1\alpha_2^* e^{-i(\delta t+\varphi)}, \end{aligned} \tag{6}$$

[1] It is also possible to solve the equations directly in relation to the elements of the density matrix, which is equivalent to an approach used in the present study. However, the non-diagonal elements are fast oscillating functions, while the diagonal elements are smooth time functions, thereby complicating the procedure of numerical counting. If getting rid of the fast oscillating part, thereby reducing the system to the real parameters, which characterize the population of the states and the amplitude of the dipole moment, then in case of an arbitrary relative shift of the phases of the field oscillations in the two oscillations pulses, the system is not strictly reduced to the three real parameters (the Bloch vectors), and it is required to introduce two more parameters taking into account the phase shift.

i.e. N is a difference of the populations of the upper and lower levels, while the values R_1, R_2, T_1, T_2 characterize the dipole moment excited by the sequence of the two mutually phase-shifted excitation pulses. The system (6) is similar to the equations in relation to the Bloch vectors [8]. However, in case of the two excitation pulses with different phases the system is reduced to the five equations, but not to three ones. The real dipole moment of the atom is expressed as follows:

$$P = \mu_{21}(R_1\cos\omega t - R_2\sin\omega t), \tag{7}$$

where ω — the frequency of the atomic transition.

The medium polarizability is found by summing the dipole moments of the donors in a unit volume by assuming that the frequencies of the atomic transitions have the normal distribution with a dispersion corresponding to a width of the heterogeneously widened impurity transition.

Then, in order to avoid solving the space problem, it was assumed the pulses follow one after another along the same path. Consequently, the echo propagation direction coincides with the excitation pulse direction. This assumption does not limit a generality of the physical conclusion on the dependence of the echo on the system parameters in question, but simplifies the calculations.

It should be remembered that when averaging across the dipole ensemble (the donor centers), the influence of the phase shift of radiation in the second pulse of the excitation in relation to the first one is negated due to the distribution of the natural frequencies of the dipoles (the frequencies of the impurity transitions). In principle, it allows setting $\varphi = 0$ in the expressions (6), whereas the system (5) is reduced to the common Bloch equations in relation to the three parameters only (see, for example, [8]). However, in the present study we will not use such additional assumptions.

When simulating the photon echo effect, it is important to evaluate the rates of relaxation of the population (γ_l) and the coherence (γ_t) in the system. It is assumed that the main mechanism of both the longitudinal and transversal relaxation is the interaction with the phonons. However, the interaction with the phonons also causes thermal transitions between the levels of the impurity centers without changing the state population averaged across the donor ensemble. But they result in loss of the coherence. The rate of the coherence relaxation was evaluated by the following formula:

$$\gamma_t = \sum_i \gamma_{\downarrow i}\big(n_i(T,E_i)+1\big) + \sum_j \gamma_{\uparrow j} n_j(T,E_j), \tag{8}$$

where $\gamma_{\downarrow i}$, $\gamma_{\uparrow j}$ — the rates of various spontaneous transitions in the interaction with the phonons numbered by the indices i and j, downward (the phonon radiation) and upward (the phonon absorption) in terms of the energy, respectively, $n_{i,j}(T,E_i)$ — occupation numbers of the phonons with the energies E_i and E_j (dependent on the lattice temperature (T)), which correspond to the energies of the i-th and j-th transitions. The summing by i and j

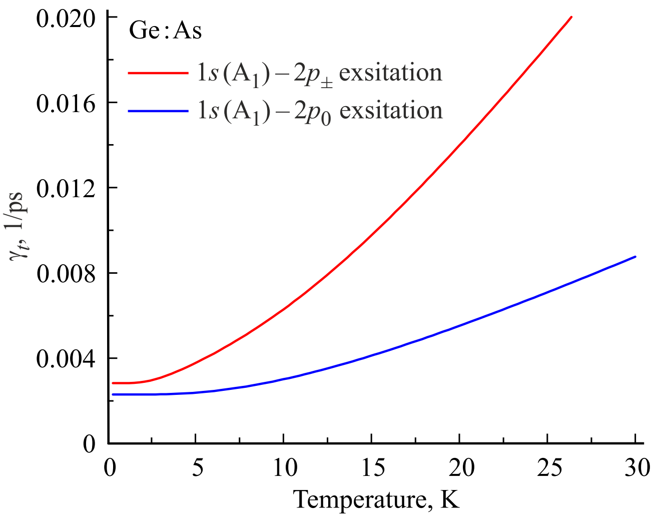

Figure 1. Calculated dependences on the temperature of the coherence relaxation rate in Ge : As for the transitions $1s(A_1)-2p_0$ and $1s(A_1)-2p_\pm$.

also includes the transitions of both from the upper $2p$ and lower $1s$ states forming the superposition (2). In Ge : As, the energy gap between the main and first excited state $1s(T_2)$ is 4.24 meV, and these states also exhibit the thermal transitions therebetween (the $1s(T_2)$ state has significant population at the temperature > 10 K) without changing the average population, but destroy the coherence. In Ge : Sb, the energy gap between the main state and the $1s(T_2)$ state is just only 0.46 meV, and the said processes are significant even at the temperature of liquid helium. That is why it is not promising to use the germanium samples doped with antimony donors to realize the experiment for observation of the photon echo.

Using the rates of the spontaneous transitions calculated in the studies [9,10], and taking into account experimental data on the lifetimes of the excited states of the Sb and As donors in the germanium [11,12], we can evaluate the rate of loss of the coherence by the formula (8), which provides for the higher rates of coherence relaxation in comparison with the longitudinal rate of relaxation (γ_1). Fig. 1 shows the dependence γ_t on the temperature, assuming that the occupation numbers comply with the Bose-Einstein statistics in case of excitation of the coherent superpositions at the two various transitions $1s(A_1)-2p_0$ and $1s(A_1)-2p_\pm$ in Ge : As.

It is clear from the performed calculations that the rate of coherence relaxation has a large temperature dependence. That is why in implementing the experiment for the photon echo effect in germanium, it is necessary to provide for good cooling of the sample in the conditions of the optical excitation. When exciting the donors to the $2p_\pm$ state, the rate of coherence relaxation is rising faster with increase in the temperature. It is correlated to the fact that this state has lesser energy gaps with the nearest states and is located quite near to the conductivity band in terms of the energy.

3. Simulation results

Fig. 2 show the dynamics of the medium polarizability calculated without the system relaxation. The durations of the excitation pulses are taken to be 100 ps (which corresponds to the pulse duration from NovoFEL), the inhomogeneous broadening is 0.01 meV. Based on data on the photo-thermo-ionization spectroscopy of germanium with the impurities of phosphorous, gallium, aluminium [13], for the $2p_\pm$ state, it should approximately correspond to the doping concentration of $\sim 10^{12}-10^{13}\,\mathrm{cm}^{-3}$. The simulation has shown that after exposure to the π-pulse, the medium polarizability excited by the first pulse and relaxed due to dephasing of the oscillations of the various dipoles was fully restored in the time equal to the delay time between the pulses, thereby demonstrating the photon echo effect.

Fig. 3 shows the dynamics of the medium polarizability at the same pulse duration, at the same inhomogeneous broadening, but taking into account the relaxation of the population and coherence in the medium. The following values $\gamma_t = \gamma_l = 5\cdot10^9\,\mathrm{s}^{-1}$ were used. These values for the As donors in Ge approximately correspond to an estimate temperature of the lattice in the optic cryostat

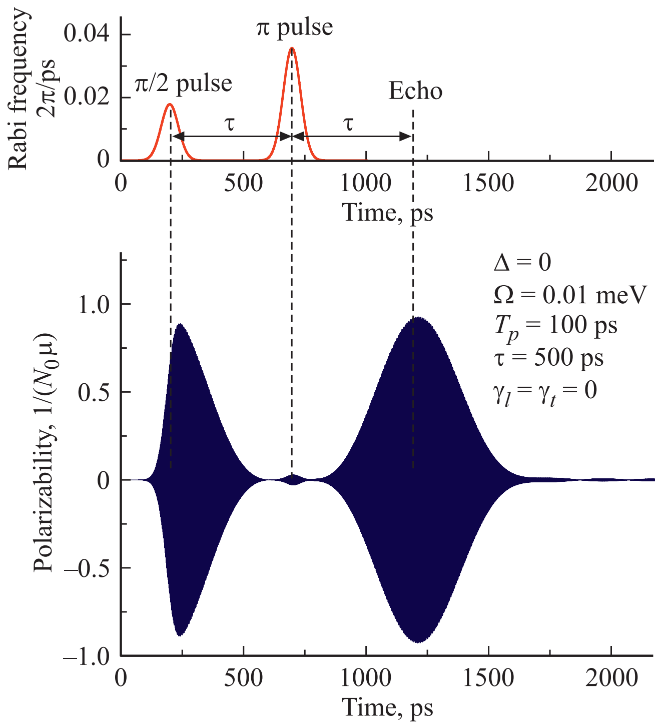

Figure 2. Top — the time dependence of the Rabi frequencies which follow the shape of the excitation pulses; bottom — the dependence of the Ge : As polarizability on time when neglecting the system relaxation. Here, Δ — the value of detuning the radiation frequency from the transition frequency (along the line centers), Ω — the width of the heterogeneously widened line (meV), T_p — the pulse duration (ps), τ — the delay time (ps), γ_l, γ_t — the rates of longitudinal and transverse relaxation, N_0 — the doping concentration, μ — the matrix element of the electro-dipole transition.

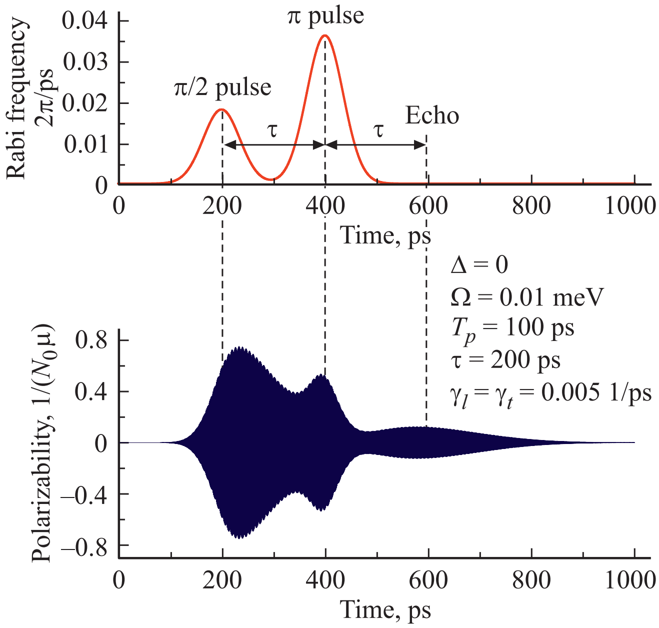

Figure 3. Dependence of the Ge:As polarizability with the system relaxation.

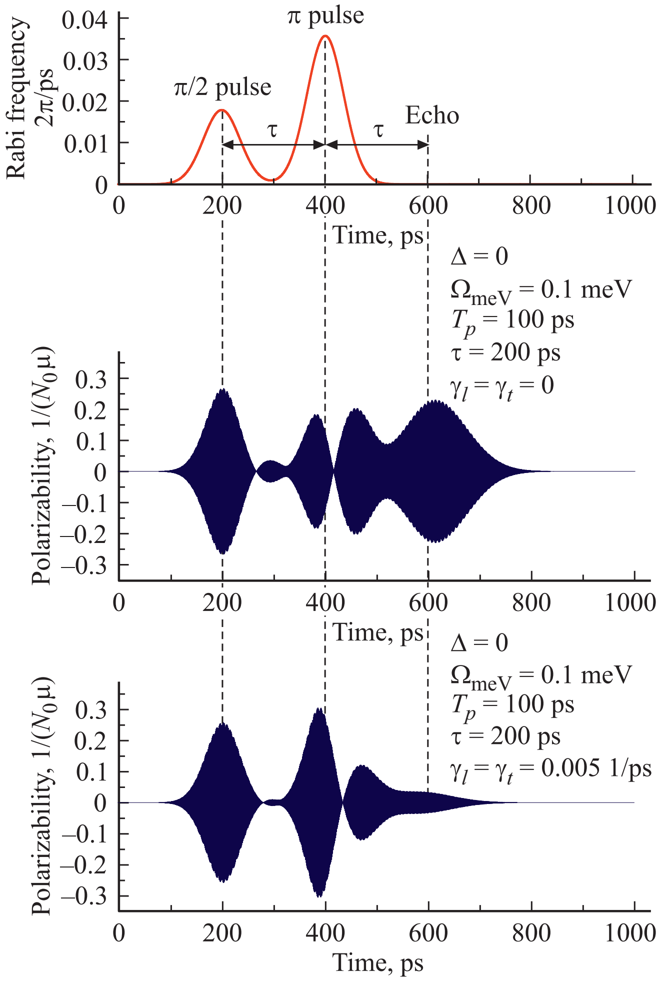

Figure 4. Dependence of the Ge:As polarizability with the quite large inhomogeneous broadening of the impurity transition line without the system relaxation and with it.

cooled to 5 K in case of the resonance excitation by the NovoFEL radiation. With such relaxation rates and laser pulse durations, the pulses should follow after after another with the minimum delay. The calculation shows the significant reduction of the echo amplitude with the substantial relaxation in the medium.

Fig. 4 shows the differences in the polarizability dynamics in *n*-Ge in case of the fast relaxation and without it, in presence of relatively large inhomogeneous broadening of the impurity transition line — 0.1 meV (it approximately corresponds to the doping concentration of $10^{14}-10^{15}\,\mathrm{cm}^{-3}$). It is clear that with the large inhomogeneous broadening there is evidently more complicated dynamics of the medium polarizability, which does not follow the shape of the excitation pulses, but the photon echo effect itself is still fully realized.

Thus, the high rate of coherence relaxation is the main obstacle for the experimental realization of the effect in *n*-Ge. It is obvious that in order to overcome it, it is desirable to use the excitation sources with the lesser pulse duration. For example, these include the lasers on free neutrons FELIX (the Netherlands) and FELBE (Dresden–Rossendorf), whose pulse duration does not exceed 10 ps. Fig. 5 shows the calculations of the polarizability dynamics in excitation of the system by the source of the pulse duration of 10 ps with quite large inhomogeneous broadenings (0.1 meV) and relaxation rates $\gamma_t = \gamma_l = 5 \cdot 10^9\,\mathrm{s}^{-1}$. As at such pulse durations, the effect can be realized at significantly lesser times in comparison with the NovoFEL excitation, then the polarizability restored by the π-pulse is substantially higher at the same conditions, thereby positively affecting the power of the recorded photon echo (the spontaneous radiation of the phased dipoles).

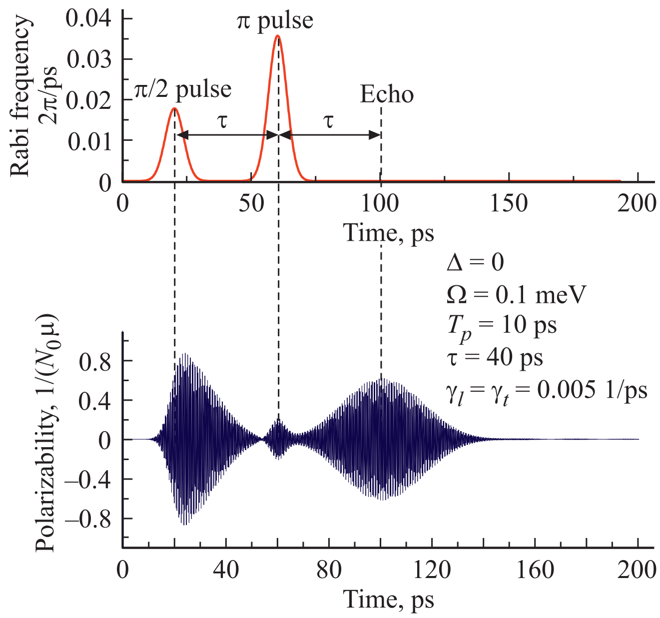

Figure 5. Dependence of the Ge:As polarizability with the quite large inhomogeneous broadening of the impurity transition, the fast relaxation and short durations of the excitation pulses.

4. Conclusion

This study has simulated the dynamics of the *n*-Ge polarizability in the resonance intracenter optic excitation by a pair of pulses with taking into account the relaxation of population and coherence in the center. It has been shown that the sample temperature is a critical factor for observing the photon echo effect as the rate of coherence relaxation largely depends on it. It has been shown that at the inhomogeneous broadenings of the impurity transitions $\sim 0.1\,\mathrm{meV}$ (it corresponds to the doping concentration of $\sim 10^{14}-10^{15}\,\mathrm{cm}^{-3}$ [13])[2], with the excitation pulse durations $\sim 100\,\mathrm{ps}$ (it corresponds to the NovoFEL pulse duration) and in presence of the quite fast system relaxation, the photon echo effect is still observable. However, using sources with the shorter pulses significantly increases the chances of successful observation of the effect. The coherent superpositions in Ge:As excited at the $1s(A_1)-2p_0$ transition have a higher time of relaxation of coherence, thereby increasing the chances of observation of the effect in comparison with a case of excitation of the $1s(A_1)-2p_\pm$ transitions. However, the last transition has a considerably higher value of the matrix element, thereby enabling using lesser powers of the optical pulses. It results in less heating of the sample and, consequently, a lesser increase in the rate of relaxation of coherence. That is why it is difficult to specify a more promising approach for successful realization of the experiment, as it will be determined by a rate of heat removal from the sample, which largely depends on a particular experimantal configuration.

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Conflict of interest

The authors declare that they have no conflict of interest.

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[2] The concentration dependence of the inhomogeneous broadening can be conditioned by various factors, including overlapping of the wave functions of adjacent impurities, the influence of the fields of the ionized impurities, random fields caused by defects related to input of impurities, etc. Although the heterogeneous widening of the lines of the impurity transitions can be defined not only by the impurity concentrations (for example, by an isotopic composition of the material, a number of crystal defects uncorrelated to the impurities, etc.), the concentration dependence is highly important. The study [13] has measured the widths of the impurity transition line for acceptors and donors of phosphorous in germanium depending on the center concentration. It is assumed that this dependence will not substantially depend on a specific kind of the impurity, and as an evaluation, this result will be applied for other donors as well.