Spin–lattice relaxation of Jahn–Teller Cr^0 centers in Si

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An experimental and theoretical investigation was made of the spin–lattice relaxtion (SLR) of Jahn–Teller Cr^0 centers in Si in the temperature range 1.2 to 8.5°K. The temperature dependence of the longitudinal relaxation rate τ_1^{-1} was described by the expression $\tau_1^{-1} = A \exp(-\Delta/k\tau)$, where $A = (7 \pm 1) \times 10^7$ sec^{-1} and $\Delta = 8 \pm 0.2$ cm^{-1}. The transverse relaxation rate τ_2^{-1} was 1.5 orders of magnitude higher and its temperature dependence was more complex. A theory developed by the authors made it possible to explain the SLR characteristics on the basis of the Jahn–Teller interaction of a triplet state of a paramagnetic center with trigonal and tetragonal vibrations. The energy structure and vibronic wave functions of the center were determined. Inversion splitting $\Delta_i = 13$ cm^{-1} was observed. The vibronic reduction was found to play an important role in the spin–phonon interaction processes.

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1. INTRODUCTION

Investigations of the ESR spectra of Cr^0, Fe^+, Mn^0, and Mn^+ in silicon have played an important role in the development of the theory of the dynamic Jahn–Teller (JT) effect in crystals.[1] However, the information obtained from the ESR spectra of these ions[1,2] is limited to the vibronic reduction of the g factor. This is insufficient for the understanding of the internal structure of the JT centers and of their interactions with the lattice vibrations. However, investigations of the spin-lattice relaxation (SLR) in the case of the static JT effect[3,4] have made it possible to determine the mechanisms of the interaction of the JT centers with phonons and some important internal characteristics of the JT centers. Much less work has been done on the SLR in the case of the dynamic JT effect.[2,5] Therefore, it seems desirable to investigate the SLR of the centers exhibiting the dynamic JT effect, particularly the Cr^0, Fe^+, Mn^0, and Mn^+ ions in Si. The present paper reports an investigation of the SLR of the Cr^0 interstitial centers in Si. These centers can be regarded, because of the magnitude of the vibronic reduction in the g factor, as the model object for investigating the dynamic JT effect in crystals.

We shall show later that the theory of the spin–phonon interactions in the investigated case of the triplet (T) ground state of an impurity should allow for the interactions with tetragonal (e) and trigonal (t_2) deformations of complexes ($T \times e + t_2$ problem).

A study of the dynamic JT effect in this case is difficult.[6–11] The vibronic wave functions of the $T \times e + t_2$ problem have been selected[7] in the form of the wave functions of the $T \times e$ problem, deformed by weak interaction with the t_2 mode. The solution has been obtained by the iteration method using the overlap integral of the e vibrational states. However, the inversion splitting[7] makes it impossible to interpret the ESR spectrum of Cr^0 in Si. A new (orthorhombic) form of minima of the adiabatic potential of the $T \times e + t_2$ problem has been found[8] but the values of the vibronic reduction factors K obtained for this case fail to account for the considerable reduction in the orbit–lattice interaction constants of Cr^0 in Si[12] and, therefore, they do not explain the temperature dependence of the SLR rate. For the same reason, the approximations and results of an analysis of the dynamic JT effect adopted by O'Brien[9] are inadequate. The $T \times e + t_2$ problem has also been solved[10,11] by the perturbation method based on the interaction with the $t_2(e)$ mode. In this case, there is no splitting of the ground state under the action of the perturbation and there is no inversion splitting if allowance is made for the perturbing interaction with the t_2 mode. We shall show later that inversion splitting due to the JT interaction with the t_2 mode is responsible for the SLR mechanism of Cr^0 in Si. It should be noted that the experimental results[12] indicate that the reduction in the orbit–lattice interaction is also due to the strong interaction with the e mode, which cannot be regarded as a perturbation.

We shall solve the $T \times e + t_2$ problem on the basis of an approximation analogous to the adiabatic approach in which the $T \times e$ (or $T \times t_2$) system will be regarded as fast, and $T \times t_2$ (or $T \times e$) as slow. The vibronic states (nonadiabatic in the ordinary sense) found in this way will be used to determine the inversion splitting into a triplet and a singlet because of the interaction with the t_2 mode. This produces a triplet ground spin–orbit state with a total momentum $J = 1$, which agrees with the ESR data.[2] We shall show that the JT effect has a considerable influence on the spin–phonon interaction processes and that the SLR method is sufficiently sensitive for the determination of the inversion splitting, which governs the SLR mechanism. We shall develop a theory which explains the observed unusual nature of the spin–phonon interaction of the JT centers at low temperatures.

2. EXPERIMENT

The SLR of Cr^0 in Si was investigated using a VARIAN E-12 ESR spectrometer at a frequency of 9.2 GHz using a magnetic field modulated at 10^5, 10^4, 10^3, or 270 Hz by the continuous saturation method.[13] The maximum klystron power was 600 mW. A cylindrical working resonator tuned to the H_{011} mode has a Q fac-

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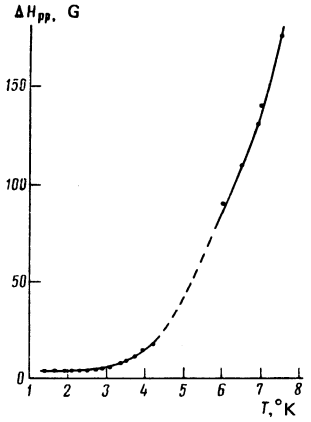

FIG. 1. Temperature dependence of the width of the ESR line of Cr^0 in Si.

tor of 1×10^4. The measurements were carried out at $T=1.2$–8.5 °K. The temperature was determined with a semiconductor thermometer.[14] A sample placed at the center of a resonator in a thin metal holder was immersed in liquid (at $T=1.2$–4.2 °K) or gaseous (at $T=6$–8.5 °K) helium.

Samples of silicon containing Cr^0 interstitial centers were made of a p-type material with a resistivity of 8×10^3 Ω·cm by diffusion annealing and quenching. The dimensions were $3\times3\times8$ mm. The Cr^0 concentration was 1×10^{16} cm^{-3}.

The ESR spectrum of Cr^0 in Si consisted of two lines, the maximum separation between which was 24 G at the frequency of 9.2 GHz when the magnetic field H was oriented along the [100] crystallographic axis. The majority of measurements described below was carried out on the first, narrower and stronger, line in the spectrum, which represented a transition in which the quantum number J_z changed from 0 to +1 and which was the narrowest among the peaks of the first derivative, $\Delta H_{pp}=3.2$ G at $T<1.7$ °K. The temperature dependence of ΔH_{pp} is plotted for this line in Fig. 1. In the range $T=1.2$–4.0 °K the experimental points correspond to the first line. At higher temperatures T, the two lines overlap and, beginning from 6 °K, only one broad line is observed whose ΔH_{pp} is also included in Fig. 1. The values of ΔH_{pp} obtained at $T=1.2$–4 °K are subject to an error not exceeding 3%, and those obtained $T=6$–8.5 °K are subject to an error within 10%. At all temperatures T, the line profile is nearly Lorentzian.

Figure 2 shows typical saturation curves of the ESR signal illustrating considerable changes in the relaxation times τ_1 and τ_2 with T. We can see that, in the interval $T=1.2$–2.5 °K, it is possible to record the complete saturation curve, i.e., it is possible to reach values of H_1 (H_1 is the amplitude of the microwave magnetic field in the resonator) at which the saturation curve bends at the point corresponding to half the intensity of the maximum signal. Therefore, at these values of T, we can use the method[13] which makes it possible to determine τ_1 and τ_2. At $T>2.5$ °K the homogeneous broadening begins to predominate so much that, in the range $T=2.5$–4 °K, we can use the incomplete saturation curve[15] to find $\tau_1\tau_2$. The line broadening in this range T can then be used to find τ_2 and, consequently, τ_1.

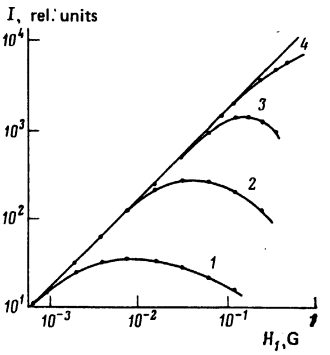

FIG. 2. Saturation curves of the ESR signal of Cr^0 in Si at several temperatures: 1) 1.2°K; 2) 1.75°K; 3) 2.5°K; 4) 3.5°K.

In calculating the absolute value of τ_1, it is necessary to know the field H_1 in the resonator. Our results were obtained on the assumption that $P=200$ MW corresponded to $H_1=0.5$ G, in agreement with the published estimates.[15]

At $T>4$ °K, we were unable to achieve saturation of the signal and estimate τ_1. The value of τ_2 in the range $T=6$–8.5 °K can be deduced from the line broadening (Fig. 1) allowing for the initial splitting observed at $T<4$ °K.

Figure 3 shows the results of the determination of the relaxation rates τ_1^{-1} and τ_2^{-1} obtained for three samples. In the case of τ_1^{-1}, all the points are described by the expression

$$\tau_1^{-1}=A\exp(-\Delta/kT),$$
$$\Delta=8\pm0.2\ \text{cm}^{-1},\quad A=(7\pm1)\cdot10^7\ \text{sec}^{-1}. \tag{1}$$

The values of τ_2^{-1} can be described by

$$\tau_2^{-1}=K+B\exp(-\Delta_1/kT)+C\exp(-\Delta_2/kT), \tag{2}$$

where $K=5.6\times10^5$ sec^{-1}, $B=1.5\times10^9$ sec^{-1}, $C=2.7\times10^{10}$ sec^{-1}, $\Delta_1=8$ cm^{-1}, and $\Delta_2=14$ cm^{-1}.

A study of the saturation of the second line in the ESR spectrum, carried out in the $T=1.2$–2.4 °K range,

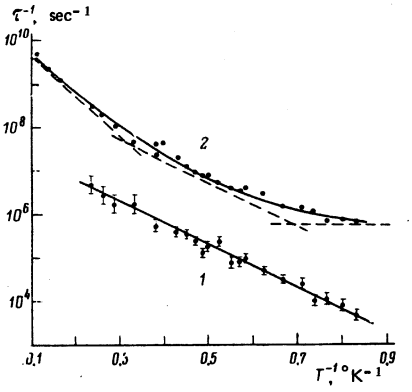

FIG. 3. Temperature dependences of the rates of relaxation of Cr^0 in Si: 1) τ_1^{-1}; 2) τ_2^{-1}. The dashed lines represent the terms in Eq. (2).

gave similar results.

A change in the orientation of H in the (001) plane through an angle of $\alpha = \pm 10°$ from the [100] axis did not alter the results. In the $|\alpha| > 10°$ case, it was not possible to measure τ_1.

We checked whether τ_1 and τ_2 depended on the magnetic field H by an experiment at a frequency of 35 GHz. The experimental conditions were as follows: klystron power 50 mW, resonator Q factor 3000, resonator mode H_{011}, dimensions of sample $0.8 \times 0.8 \times 8$ mm, and $H = 8000$ G (at 9.2 GHz, the field was $H = 2000$ G). Measurements in the range $T = 1.2$–4 °K showed that the temperature dependences τ_1^{-1} and τ_2^{-1} were identical at 9.2 and 35 GHz. Moreover, the absolute values were identical if it was assumed that, at 35 GHz, the power of 50 mW in the resonator corresponded to $H_1 = 0.5$ G.

An attempt was also made to measure τ_1 by a pulse method. At $T = 1.7$ °K and a frequency of 9 GHz, it was found that $\tau_1 \leqslant 10^{-5}$ sec, which was not in conflict with the above results. (It was not possible to reach temperatures below 1.7 °K in the pulse ESR relaxometer employed.)

3. THEORY

The observed SLR behavior and the broadening of the ESR line of Cr^0 in Si at low temperatures are unusual for the following reasons. Firstly, the values of τ_2^{-1} and τ_1^{-1} are of spin–phonon origin, which is confirmed by their temperature dependence; they differ considerably in magnitude, so that $\tau_2^{-1} \gg \tau_1^{-1}$, and in their temperature dependences. Secondly, the temperature dependence of τ_1^{-1} has no linear region associated with the one-phonon processes at the microwave frequency of the kind that usually predominate at low temperatures. Moreover, the values of τ_1^{-1} and τ_2^{-1} are independent of the magnetic field. Thirdly, the absolute values of τ_1^{-1} and τ_2^{-1} are anomalously high.

In view of these observations, we may expect the behavior of τ_1^{-1} and τ_2^{-1} to be governed by different spin–phonon processes which are not of the one-phonon kind at the microwave frequency.

The exponential dependence $\tau_1^{-1}(T)$ may be associated with the Orbach process, but the energy of the corresponding excited state ($\Delta \approx 8$ cm^{-1}) does not agree with the energy of the first excited electronic state with $J = 2$, equal to 14 cm^{-1}.[2] Moreover, we cannot identify Δ with the energy of the first excited vibrational state because the rate of the Orbach process involving the vibrational states is considerably less than that observed for Cr^0 in Si. Moreover, the value $\Delta \approx 8$ cm^{-1} is far too low for the vibrational excitation energy.

The JT nature of the paramagnetic centers is an important factor in the explanation of the observed behavior of τ_1^{-1} and τ_2^{-1}. The employed model of a JT impurity in a triplet state involves allowance for the interaction of this center with the trigonal (t_2) and tetragonal (e) deformations of the complex. We shall assume that the "electron + e deformation" system is fast compared with the "electron + t_2 deformation" system. Applying the adiabatic (in the above sense) approximation, the problem can be solved in two stages. First, we find the vibronic eigenfunctions and eigenvalues for the JT effect allowing only for the interaction with the e deformation. We then use the wave functions of a vibronic triplet found in this way as the basis in considering the JT effect resulting from the interaction with the t_2 deformation of the complex. In this way, the second stage of the calculation implies that the JT effect is due to the interaction of the t_2 deformation with the vibronic triplet resulting from the JT interaction with the e deformation. It is assumed that the interaction with the t_2 deformation occurs when the JT effect is very strong (almost static) and we can use the tunneling model. Consequently, we obtain a singlet ground vibronic state ($S = 2$) and an excited vibronic triplet state when the tunnel matrix element is $\Gamma < 0$ and the distribution of the levels is inverted for $\Gamma > 0$.

The spin–orbit interaction lifts the fivefold spin ($S = 2$) and the threefold vibronic degeneracy of the triplet, which gives rise to three spin-vibronic multiplets with $J = 1$ ($E = -3\lambda$), $J = 2$ ($E = -\lambda$), and $J = 3$ ($E = 2\lambda$), where λ is the spin–orbit interaction constant. Since, in the case of Cr^0 in Si, the ground state deduced from the ESR data is characterized by $J = 1$, this implies the inequality $3\lambda > \Delta_i$ if $\Gamma < 0$. For the ground state $J = 1$, we find that

$$\begin{aligned}
\tilde{\Psi}_{1-1} &= \frac{1}{\sqrt{20}}(\Phi_x - i\Phi_y)\Psi_{20} + \sqrt{\frac{3}{10}}\Phi_z\Psi_{2-1} - \sqrt{\frac{3}{10}}(\Phi_x + i\Phi_y)\Psi_{2-2},\\
\tilde{\Psi}_{10} &= \sqrt{\frac{3}{20}}(\Phi_x - i\Phi_y)\Psi_{21} + \sqrt{\frac{2}{5}}\Phi_z\Psi_{20} - \sqrt{\frac{3}{20}}(\Phi_x + i\Phi_y)\Psi_{2-1},\\
\tilde{\Psi}_{11} &= \sqrt{\frac{3}{10}}(\Phi_x - i\Phi_y)\Psi_{22} + \sqrt{\frac{3}{10}}\Phi_z\Psi_{21} - \sqrt{\frac{1}{20}}(\Phi_x + i\Phi_y)\Psi_{20}.
\end{aligned} \tag{3}$$

where

$$\begin{aligned}
\Phi_x &\approx {}^1/_2(\Phi_1 - \Phi_2 + \Phi_3 - \Phi_4),\\
\Phi_y &\approx {}^1/_2(\Phi_1 - \Phi_2 - \Phi_3 + \Phi_4),\\
\Phi_z &\approx {}^1/_2(\Phi_1 + \Phi_2 - \Phi_3 - \Phi_4),
\end{aligned} \tag{4}$$

Ψ_{2m} ($m = 2, 1, 0, -1, -2$) are the spin wave functions. The singlet vibronic state

$$\Phi_s \approx {}^1/_2(\Phi_1 + \Phi_2 + \Phi_3 + \Phi_4) \tag{5}$$

is separated from the vibronic triplet by the energy gap Δ_i, which represents the inversion (tunnel) splitting:

$$\begin{aligned}
\Delta_i &\approx {}^8/_3[K^{\text{tetr}}(T_2)]^2 E_{\text{JT}}^{\text{trig}}\,\gamma,\\
\gamma &= \exp\left(-\frac{4[K^{\text{tetr}}(T_2)]^2 E_{\text{JT}}^{\text{trig}}}{3\hbar\omega_{\text{trig}}}\right),
\end{aligned} \tag{6}$$

where $E_{\text{JT}}^{\text{trig}}$ ($E_{\text{JT}}^{\text{tetr}}$) and K^{trig} (K^{tetr}) are, respectively, the JT energy and vibronic reduction factors for the $T \times t_2$ ($T \times e$) problems, and ω_{trig} is the frequency of the t_2 deformations. It should be noted that the appearance of the inversion splitting is associated with the $T \times t_2$ interaction.

The four "single-well" vibronic states to which, as assumed, the tunnel approximation can be applied (the overlap integral is $\gamma \ll 1$) are represented by

$$\Phi_1=\frac{1}{\sqrt{3}}(\Psi_{yz}F_\theta^{yz}F_\varepsilon^{yz}+\Psi_{xz}F_\theta^{xz}F_\varepsilon^{xz}+\Psi_{xy}F_\theta^{xy}F_\varepsilon^{xy})F_{T(xy)}^{(1)}F_{T(xz)}^{(1)}F_{T(yz)}^{(1)},$$
$$\Phi_2=\frac{1}{\sqrt{3}}(-\Psi_{yz}F_\theta^{yz}F_\varepsilon^{yz}-\Psi_{xz}F_\theta^{xz}F_\varepsilon^{xz}+\Psi_{xy}F_\theta^{xy}F_\varepsilon^{xy})F_{T(xy)}^{(2)}F_{T(xz)}^{(2)}F_{T(yz)}^{(2)},$$
$$\Phi_3=\frac{1}{\sqrt{3}}(\Psi_{yz}F_\theta^{yz}F_\varepsilon^{yz}-\Psi_{xz}F_\theta^{xz}F_\varepsilon^{xz}-\Psi_{xy}F_\theta^{xy}F_\varepsilon^{xy})F_{T(xy)}^{(3)}F_{T(xz)}^{(3)}F_{T(yz)}^{(3)}, \quad (7)$$
$$\Phi_4=\frac{1}{\sqrt{3}}(-\Psi_{yz}F^{yz}F^{yz}+\Psi_{xz}F_\theta^{xz}F_\varepsilon^{xz}-\Psi_{xy}F_\theta^{xy}F_\varepsilon^{xy})F_{T(xy)}^{(4)}F_{T(xz)}^{(4)}F_{T(yz)}^{(4)}.$$

Here, $F_{T(j)}^{(i)}$ and $F_{\theta,\varepsilon}^{(i)}$ are the wave functions of the T and E vibrations, respectively, the indices i label the adiabatic potential minima, Ψ_{xy}, Ψ_{yz}, and Ψ_{xz} are the electronic wave functions of the triplet.

Similarly, we can find the vibronic eigenfunctions and states in the opposite limiting case when the "electron+t_2 deformation" system is fast compared with the "electron+e deformation" system.

Using Eq. (4), we determine the vibronic reduction factors in the case of the interaction of the triplet state with the t_2 and e deformations on the assumption that the "adiabatic" procedure is valid. We thus obtain

$$\begin{aligned}K(T_1)&=K_{\text{trig}}(T_1)K_{\text{tetr}}(T_1),\\ K(T_2)&=K_{\text{trig}}(T_2)K_{\text{tetr}}(T_2),\\ K(E)&=K_{\text{trig}}(E)\end{aligned} \quad (8)$$

for the T_1, T_2, and E representations. The resultant JT energy is

$$E_{\text{JT}}=E_{\text{JT}}^{\text{tetr}}+[K^{\text{tetr}}(T_2)]^2E_{\text{JT}}^{\text{trig}}. \quad (9)$$

We shall now consider possible low-temperature SLR processes which may be exhibited by such JT centers.

In the ground state, the eigenfunctions (3) with the total orbital momentum $J=1$ represent mixed spin–orbit–vibrational wave functions. The orbit–vibrational–lattice interaction H_{OVL}, which couples the electronic degrees of freedom and the vibrations of a complex with the lattice deformations, then gives rise to transitions in which the projection of the total orbital momentum changes. Such transitions correspond to the ESR relaxation. There is also a change in the projection of the spin of the center.

Since H_{OVL} is not of spin origin and does not contain spin operators, in this situation the SLR is due to the spin-free interaction. This is usually stronger than the spin–phonon interactions and accounts for the observed anomalously high SLR rate at low temperatures.

The constants of the interaction of the total orbital momentum of a center with lattice vibrations were found experimentally by Ludwig and Ham.[12] The values of these constants were considerably smaller than the usual orbit–lattice interaction constants. This can be explained if allowance is made for the vibronic reduction. Since the constants of the interaction with the t_2 and e lattice deformations are quite small, it follows that, in this case, the important effect is the JT interaction with the t_2 and e modes of the complex, which results in the vibronic reduction. This confirms the adopted model of the JT effect.

The vibronic reduction in the orbit–lattice interaction constant, which reduces the corresponding constants and, particularly, G_{44}, is responsible for the fact that, at sufficiently low temperatures, the SLR rate is not governed by the one-phonon process at the microwave frequency. The vibronic reduction G_{44} is associated with the interaction of a center with the e deformations of the complex. If we use the measured values of G_{44},[12] we find that at $T=4.2$ °K the one-phonon SLR rate at the microwave frequency is two orders of magnitude smaller than that actually observed. This is the reason for the disappearance of the dependences $\tau_1^{-1}\propto TH^2$ typical of the one-phonon process at the microwave frequency.

We shall assume that the low-temperature SLR mechanism involves relaxation under the action of the orbit–vibrational–lattice interaction H_{OVL} due to the Orbach process at the frequency of splitting Δ between the singlet and the ground triplet state ($J=1$). This explains the temperature dependence of τ_1^{-1} and the absence of the magnetic-field dependence of τ_1^{-1} and allows us to identify Δ with the experimentally found value $\Delta=8$ cm^{-1}.

The energy gap between the singlet and triplet is $\Delta=|3\lambda\pm\Delta_i|$ for $\Gamma\gtrless 0$, respectively. Using the reported[2] value $\lambda\approx 7$ cm^{-1}, we find that the experimentally observed Δ can occur only for $\Gamma<0$. Therefore, the first derivative of the potential energy of the t_2 deformation, whose sign is governed by that of Γ, is also negative. The vibronic singlet is the ground state relative to the vibronic triplet of Cr^0 in Si. Therefore, using the above values of λ and Δ, we find that the inversion splitting is $\Delta_i=3\lambda-\Delta=13.2$ cm^{-1}.

A calculation of τ_1^{-1}, measured by the continuous saturation method, will be made using Eqs. (3)–(8) and H_{OVL} in the form

$$H_{\text{OVL}}=\sum_{ij}h_{ij}e_{ij}, \quad (10)$$

where e_{ij} are the components of the deformation (strain) tensor. Symmetry considerations lead to

$$\tau_1^{-1}\approx 2.4\frac{\pi^3\Delta^3|\langle\Phi_1|h_{xy}|\Phi_1\rangle|^2}{\rho v^5h^4}\exp\left(-\frac{\Delta}{kT}\right). \quad (11)$$

The matrix element $\langle\Phi_1|h_{xy}|\Phi_1\rangle$ in Eq. (11) is related to the constant G_{44} determined by Ludwig and Ham.[12] Using this constant, we find that not only the temperature dependence but also the absolute value of τ_1^{-1}, derived from Eq. (11), are in agreement with the experimental results. It should be noted that the matrix element $\langle\Phi_1|h_{xy}|\Phi_1\rangle$, governing τ_1^{-1}, is subject to the vibronic reduction.

The appearance of inversion splitting due to the $T\times t_2$ interaction and the absence of the one-phonon process at the microwave frequency due to the vibronic reduction resulting from the $T\times e$ interaction provide direct confirmation of the adopted $T\times e+t_2$ model.

Investigations of the ESR line broadening support the spin–phonon broadening mechanism not associated with the mechanism responsible for τ_1 (because τ_1 and τ_2

have different orders of magnitude and different temperature dependences). In fact, the SLR processes considered above do not have the rate necessary to explain the spin–phonon broadening of the ESR line. The reason for the increase of τ_2^{-1} relative to τ_1^{-1} may be the broadening due to the spin–phonon processes under the action of H_{OVL} without vibronic reduction, which ensures that $\tau_2^{-1} \gg \tau_1^{-1}$. Similar processes in the case under discussion result from one-phonon transitions between the spin–vibronic multiplets with $J=1$ and $J=2$ at a frequency 2λ. The transitions without vibronic reduction only occur between states with identical projections of the total momentum. Thus, they do not contribute to τ_1^{-1} but may be responsible for the ESR line broadening if the corresponding lifetimes of the states with different total momentum projections are different. Such a situation occurs in the interaction of the totally symmetric deformation of the lattice. Consequently, τ_2^{-1} can be represented in the form

$$\tau_2^{-1} \approx 3.2 \frac{\pi^3 (2\lambda)^3 A_0^2}{h^4 \rho v^5} \exp\left(-\frac{2\lambda}{kT}\right), \qquad (12)$$

where $A_0 = |\langle \Phi_1 | h_{ii} | \Phi_1 \rangle|$. It follows that A_0 and, therefore, τ_2^{-1} do not include the vibronic reduction factor, which may account for the anomalously large thermal broadening of the line. For this ESR line broadening, the rate τ_2^{-1} is an exponential function of temperature and, in the case considered, the argument of this function is $2\lambda = 14$ cm^{-1}. A similar exponential rise of τ_2^{-1} is observed experimentally in the temperature range $T = 3$–8.5 °K. Applying the experimentally determined preexponential factor (2), we find from Eq. (12) the value of the constant representing the interaction of a center with the totally symmetric deformation of the lattice A_0. The result is $A_0 = 1.7$ eV, which is reasonable for the constant H_{OVL} without vibronic reduction. It follows from Eq. (12) that τ_2^{-1} is independent of H, which is again in agreement with the experimental values.

Broadening of the ESR line in the $T = 1.6$–3 °K range, which obeys the dependence $\tau_2^{-1} \propto \exp(-\Delta/kT)$, where $\Delta \approx 8$ cm^{-1}, is clearly due to the process involving the singlet vibronic state which is 8 cm^{-1} above the ground state. Further studies are needed to determine details of this process.

The experimental evidence obtained at low temperatures in the range $T = 1.2$–1.5 °K shows that τ_2^{-1} is independent of temperature, which can be explained allowing for the homogeneous line broadening due to the spin–spin interaction. Estimates indicate that, for the concentrations of paramagnetic centers $n \sim 10^{15}$–10^{16} cm^{-3}, the spin–spin broadening is in agreement with the experimental observations.

Thus, our JT effect model ($T \times e + t_2$ problem) of the deep Cr^0 impurity in Si predicts the appearance of a new singlet vibronic state, which is not predicted by Ludwig and Woodbury. The vibronic states, vibronic reduction, and inversion splitting explain the experimental results of the investigations of the low-temperature SLR and ESR line broadening. They also give the magnitude of the inversion splitting and the energy structure of the investigated centers.

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