

Population inversion of the spin states of Cr^{3+} in silicon due to unpolarized optical pumping

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Population inversion of the $-\frac{5}{2} \longleftrightarrow -\frac{3}{2}$ and $\frac{1}{2} \longleftrightarrow \frac{3}{2}$ transitions of the Cr^{3+} ion ($S = \frac{3}{2}$) in silicon due to interband pumping was observed at helium temperatures. The effect can be explained by allowing for the spin-dependent capture of the current carriers and the formation of intermediate paramagnetic states.

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The interaction of current carriers with the paramagnetic systems (electronic and nuclear) may effectively alter the spin polarization of the latter.⁽¹⁻³⁾

In this work we report the first observation of population inversion in a spin system of paramagnetic centers (PC) in a semiconductor due to unpolarized optical pumping.

The EPR of PC implantation of Cr^{3+} in Si was investigated in the illuminated samples at $T = 1.8-10$ K. The Cr^{3+} centers in concentrations $N_0 \approx 2 \times 10^{15} \text{ cm}^{-3}$ were obtained by thermal diffusion of chromium into p -type, boron-doped original silicon samples.

The EPR spectrum of Cr^{3+} is comprised of five lines of the fine structure produced by splitting in a cubic crystal field with the constant $a \approx 32 \text{ Oe}$ (Fig. 1a).⁽⁴⁾ Their intensities in the dark correspond to the equilibrium Boltzmann values. The spectrum changes substantially when the sample is illuminated (Fig. 1b). The important feature

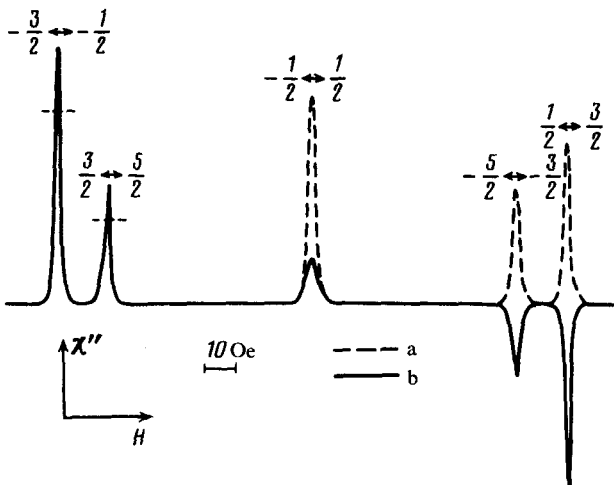


FIG. 1. EPR spectrum of Si:Cr^{3+} in darkness (a) and under illumination (b). $\mathbf{H} \parallel [100]$, $T = 4.2 \text{ K}$, CE concentration $n_c \approx 5 \times 10^7 \text{ cm}^{-3}$.

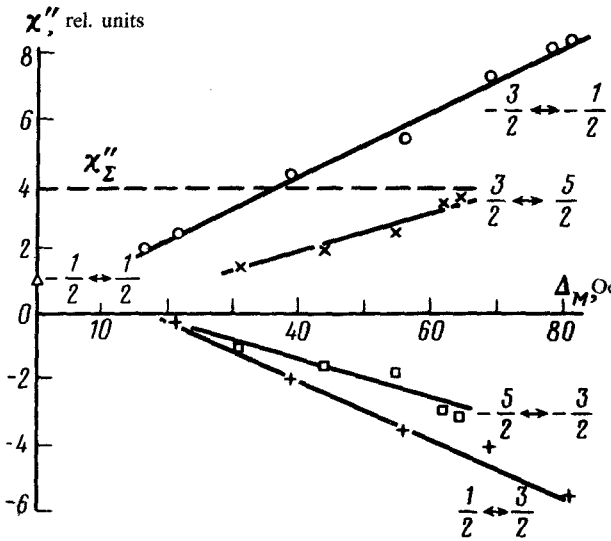


FIG. 2. Dependence of the relative intensity of the different Si:Cr⁺ transitions on detuning Δ_M in illuminated samples. $T = 4.2$ K.

is the inversion of the high-field lines as a result of unpolarized pumping. The rotation of the magnetic field H in the [100] plane produces a linear decrease of the intensity of the side lines with decreasing detuning with respect to the center of the spectrum, and the intensity of the central line remains constant (Fig. 2). The total intensity of the lines χ''_{Σ} , which is independent of the direction of H , is lower than the equilibrium (dark) intensity by a factor of approximately three. When the light is turned off, the intensities of the spectral lines are restored to the equilibrium Boltzmann values with the spin-lattice relaxation time τ_1 in the dark, which corresponds to the spin-polarized nature of its modification.

A part of the signal drop (not shown in Fig. 1) due to charge transfer of Cr⁺ to other deep centers (presumably Cr⁰), which cannot be recovered in darkness at low temperatures, also occurs.

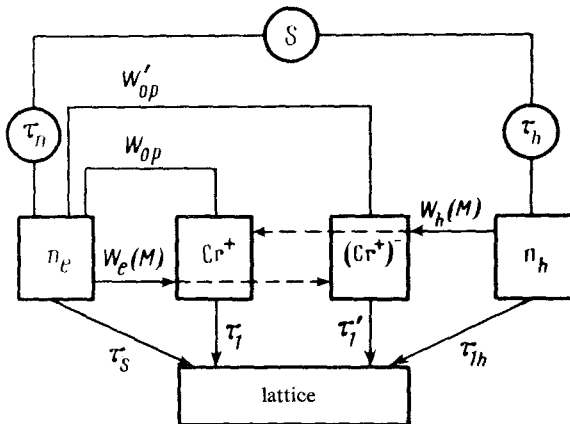


FIG. 3. Schematic diagram of energy-exchange processes in Si:Cr⁺ as a result of illumination. S is the light source, n_h and $W_h(M')$ is the hole concentration and their capture velocity in $(Cr^{+})^{-}$, respectively, W_{op} , W'_{op} are the capture rates of ES of CE by Cr⁺ and $(Cr^{+})^{-}$, τ_s , τ_{1h} , τ'_1 are the spin-relaxation times of the Ce, of the holes, and of $(Cr^{+})^{-}$, τ_n , τ_h are the lifetimes of the CE and the holes, respectively.

The possibility of polarization of PC as a result of their interaction with the conduction electrons (CE) was predicted in Refs. 1 and 3. The exchange scattering (ES) of spin-polarized CE by PC can⁽³⁾ polarize the latter at large values of Δ_M , where $\Delta_M = \hbar(\omega_e - \omega_{M,M-1})$ is the difference between the Zeeman frequencies of CE and PC. However, in the case of Si:Cr⁺ Δ_M cannot explain the magnitude and sign of polarization of the individual components of the Cr⁺ spectrum.

In this paper we propose a new polarization scheme (Fig. 3) that is associated with spin-dependent processes of current-carrier capture, the formation of small hydrogen-like states (Cr⁺)⁻, and polarization of the latter due to ES of CE.⁽³⁾

An electron captured in the large-radius orbit undergoes exchange interaction with the 3d⁵ shell of Cr⁺, as a result of which two spin multiplets with $\tilde{S} = 2$ and $\tilde{S} = 3$, which are separated by the 6J gap, are produced. Since $J > 0.6J \gg kT$,⁽³⁾ the $\tilde{S} = 3$ multiplet is predominantly populated. Because the rate of the CE capture $W_e(M)$ depends on the projection M of the Cr⁺ spin, the spin-state populations of the CE and (Cr⁺)⁻ diverge from the equilibrium. The spin Hamiltonian of (Cr⁺)⁻ has the terms $a'(\tilde{S}_x^4 + \tilde{S}_y^4 + \tilde{S}_z^4)$ corresponding to the cubic crystal field; moreover, at $a' \gg a$ the (Cr⁺)⁻ spectrum is essentially spaced unequally. According to Ref. 3, the ES of CE in (Cr⁺)⁻, which are detuned significantly $\Delta_{M'} \propto a'$, redistribute the populations of the (Cr⁺)⁻ states, which is proportional to the value and the sign of $\Delta_{M'}$, if $n_{e/N} W'_{op} \gg (\tau_1^0)^{-1}$ (N' is the steady-state concentration of (Cr⁺)⁻). This is followed by the capture of holes (predominantly with $S = 1/2$ ⁽⁵⁾), which also depends on the projections M' of the (Cr⁺)⁻ spins. As a result, the Cr⁺ spin system is polarized in such a way that the high-field transitions are inverted.

The steady-state solution of the kinetic equations for the polarizations Cr⁺ (P_M) and CE (P_e), which take into account the indicated processes (Fig. 3), for W_e

$\gg \frac{N}{n_e} \tau_1^{-1}$, W_{op} in the high-temperature approximation has the form

$$P_{M, M-1} = \frac{11}{7} \frac{a'}{a} \frac{\Delta_M/kT}{1+a} + P_e + P_h \frac{a-1}{a+1}, \quad (1)$$

$$P_e = \left(P_h \frac{1-5a/6}{1+a} + P_e^0 \frac{\tau_s^{-1}}{W_e} \right) \left(\frac{8}{3} + \frac{\tau_s^{-1} + \tau_n^{-1}}{W_e} \right)^{-1}, \quad (2)$$

where $P_{M,M-1} = (2S+1)^{-1} \frac{N_{M-1} - N_M}{N}$, N is the Cr⁺ concentration as a result of pumping, P_h is the equilibrium polarization of the p_e^0 $a = W_e/42W'_{op}$, and W_e is the rate of CE capture in Cr³⁺, averaged over M .

Equation 1 shows that $P_{M,M-1} \propto \Delta_M$ is the detuning of Cr⁺, which is amplified by a factor $k = \frac{a'}{a} (1+a)^{-1}$, where $k \gg 1$, if $W'_{op} \gg W_e$. When $W_e \gg \tau_s^{-1}$, τ_n^{-1} and $a \ll 1(2S+1)^{-1} \sum_M P_{M,M-1} = P_{1/2, -1/2} \approx P_e \approx \frac{3}{8} P_h$.

A comparison of Eqs. (1) and (2) with the experiment shows that a quantitative agreement is obtained at $(a'/a) \approx 30$ and $P_h \approx P_h^0$. A linear dependence of $P_{M,M-1}$ on Δ_M and a weak temperature dependence of the effect, which follow from Eq. (1), also agree with the experiment.

We also examined the non-steady-state model which produces an inversion in the EPR spectrum of Cr^+ . The model is based on nonadiabatically rapid switching of the effective magnetic field in the $(\text{Cr}^+)^-$ system as a result of the exchange interaction due to electron capture by Cr^+ , which disappears as a result of thermal emission or recombination. The $(\text{Cr}^+)^-$ system has fast spin-relaxation channels that are associated with the modulation J by lattice vibrations. As a result, spin-state populations of $(\text{Cr}^+)^-$ are redistributed in such a way that, finally the Cr^+ transitions $\frac{1}{2} \longleftrightarrow \frac{3}{2}$ and $-\frac{5}{2} \longleftrightarrow -\frac{3}{2}$ are inverted.

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