Spin polarization of noncentral paramagnetic ions induced by a tunneling effect

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A spin polarization of paramagnetic ions which results from a coherent tunneling between noncentral positions has been observed for the first time. This tunneling can occur either with or without a change in the spin state of the ions.

1. Noncentral impurity ions are a good model for studying tunneling in solids. Under the usual experimental conditions the internal fields of defects localize noncentral impurity ions in the various minima of a multiwell potential, and tunneling transitions of these ions are induced by phonons that are inelastically scattered by the ions.[1] Two fundamental questions arise in connection with the problem of studying the tunneling of local centers in a solid. First, is it possible to arrange an experimental situation in which a coherent tunneling of noncentral impurity ions can be observed directly, without a contribution of inelastic scattering of phonons, under conditions such that the ions are localized in different potential wells, and tunneling states do not form? Second, what role would be played by the spin-dependent tunneling for paramagnetic noncentral impurity ions, and what new effects would this type of tunneling generate? We report here a study of these questions. We have found that a spin-dependent (specifically, a spin-orbit) coherent tunneling gives rise to a significant spin polarization of noncentral Co^{2+} impurity ions in SrO upon a change in the external field.

2. In experiments with the model system SrO: 0.01% Co^{2+}, we studied the relaxation to an equilibrium population of the noncentral energy minima and of their spin levels after a static electric field E is turned on or off [$E = (1\text{–}2)\times 10^5$ V/cm]. The predominant orientation of the dipole moments of the impurity centers which is created by the field is manifested experimentally as a change in the intensities of lines in the ESR spectrum and in the appearance of a dichroism in the optical absorption spectrum of the SrO:Co^{2+} crystal. In the ESR and optical measurements, we used the geometry $\mathbf{E}\|[110]$ and measured the change in the number of centers with dipole moment perpendicular to the field E (Ref. 2). The rate at which the field was turned on (or off) was varied by changing the time constant (τ_{RC}) of the circuit of the charge of the sample.

At $T = 4.2$ K, the ion-lattice relaxation time of the Co^{2+} ion is[2] $\tau_0 = 85 \pm 10$ s, but if the field E is turned on or off sufficiently slowly (if the switching time τ_{RC} lies in the range 10^{-3}–1 s), part of the relaxation will occur at a rate $1/\tau = 10\ s^{-1}$, which is three orders of magnitude higher than $1/\tau_0$ (the A_{on} and A_{off} "steps" on the time evolution of the ESR and the optical absorption; see Fig. 1). After the field is switched under these conditions, we also observe a brief increase in the ESR signal (Fig. 1a); the resultant change (A_{on}, A_{off}) in this signal corresponds to an increment in the number of

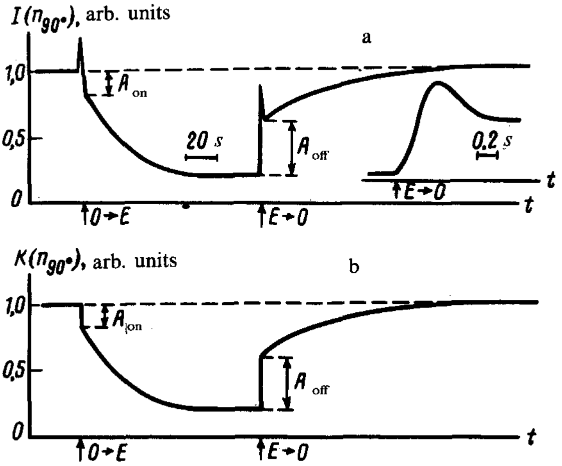

FIG. 1. Time evolution of (a) the ESR signal and (b) the optical absorption signal of SrO:Co^{2+} crystals after an electric field is turned on ($0 \to E$) and off ($E \to 0$). $T = 4.2$ K, $E = 100$ kV/cm, $\tau_{RC} = 0.09$ s. The inset in Fig. 1a shows the shape of the "spike" in the ESR signal (in a larger time scale).

90° centers obtained from the size of the step on the curve of the absorption coefficient $K(t)$ (A_{on}, A_{off} in Fig. 1b).

Figure 2 shows the length and height of the "spike" in the ESR signal versus the rate at which the field is turned off. Curve 2, which shows the ratio (η) of the height of the signal at the top of the spike to the height of the signal after the spike has been subtracted, goes through a clearly defined maximum at $\tau_{RC} \cong 0.1$ s.

These experimental results are evidence that when the electric field is turned on (or off) sufficiently slowly, the change in the orientation of the dipole moment of a significant fraction of the Co^{2+} centers occurs at a rate $1/\tau \gg 1/\tau_0$. The spin polarization that arises exceeds the equilibrium value by a factor of more than two (curve 2 in Fig. 2).

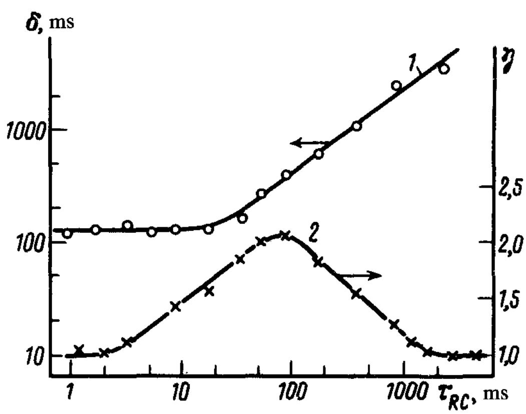

FIG. 2. Duration (1) and amplitude (2) of the spike in the ESR signal as functions of the rate at which the field is turned off. $T = 4.2$ K, $E = 150$ kV/cm.

3. The reorientational relaxation of a noncentral ion at low temperatures is caused primarily by hopping tunneling processes.[3–6] The weak temperature dependence of the characteristics of this effect (measured in the interval 1.6–4.2 K) also indicates a tunneling nature of the "fast" relaxation process. The particular way in which the experiments were carried out in the present study is highly conducive for observing coherent tunneling under conditions such that the external electric field cancels the internal (defect) field with an accuracy to within the natural width of the levels of the single-well states, and the tunneling occurs without the involvement of phonons (a so-called tunneling-controlled process[3,4]). If the external field E exceeds the half-width $\langle \Delta E \rangle$ of the distribution function of the internal electric field which displaces the levels of the adjacent wells of a center by an amount $p \cdot \Delta E$, then over the time over which the external field changes the conditions most favorable for a tunneling-controlled process $[E(t) = -\Delta E]$ are arranged for about half of all the centers.

Using the example of a two-well potential, let us examine the process by which the field is turned off; during this process, well b, which was higher than well a with the field on, descends. The population of well b is increased by four tunneling-controlled channels, which come into play in succession when the spin levels belonging to wells a and b come into coincidence (Fig. 3). Because of the spin-orbit interaction, there should be a significant spin-dependent tunneling effect.[4] Estimates show that the spin-orbit tunneling matrix elements in our case are of the same order of magnitude as the spin-independent tunneling matrix elements. As a result, when channels 1 and 4 come into play, reorientational transitions accompanied by a change in the spin state of the ion occur. As well b descends, the level $b\,|-\rangle$ is initially filled through channels 1 and 2, with the result that a nonequilibrium spin polarization of the particles arises in well b; then the level $b\,|+\rangle$ becomes filled through channels 3 and 4, and the spin

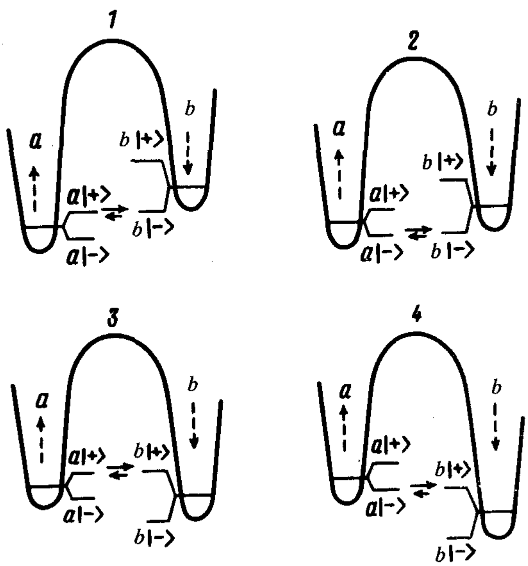

FIG. 3. Four characteristic situations in the coincidence of the spin levels of wells a and b which arise when an external field E is turned off. The g-factors of the minima a and b are not the same: $g_a < g_b$.

polarization decreases. The degree of polarization is also weakened by spin-lattice relaxation within a single potential well.

A comparison of the approximate solutions of the kinetic equations found for this model with experimental data shows that the spin-orbit and spin-independent coherent tunneling can be used to construct a qualitatively correct description of the main features of the observed effect. The shape of the time dependence of the spin-polarization effect and the scale time for the process can be explained (the tunneling matrix elements are on the order of 10^5–10^6 Hz), as can the way in which the duration and amplitude of the spin-polarization process depend on the rate at which the field is switched. With decreasing τ_{RC}, the times at which the four tunneling-controlled channels come into play thus move closer together, with the result that the duration of the overall spin-polarization effect and its amplitude are reduced significantly. For $\tau_{RC} \lesssim \tau$, the duration of the polarization process reaches a minimum and then remains essentially constant as τ_{RC} is reduced further. We should emphasize that the theory agrees much better with experiment when we take into account a spin-orbit tunneling that leads to transitions involving a change in the spin state.

In summary, the spin polarization and "fast" electric-dipole reorientational relaxation of a noncentral Co^{2+} ion in SrO, which is observed when an external field is turned on and off, is due to a coherent tunneling of the ion, and significant part of this tunneling consists of tunneling transitions involving a change in the spin state of the ion.

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