Spin refrigerator for paramagnetic impurities using electric fields

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Reorienting paramagnetic centers in crystals can be cooled in a steady-state fashion below the lattice temperature by means of nonresonant electric fields.

1. The best-known method for cooling paramagnetic centers in crystals, adiabatic demagnetization,[1] is a one-shot method. There are also cyclic methods for cooling spin systems called "spin refrigerators." Various spin refrigerators for nuclear subsystems are described in monographs.[1–3] In these refrigerators, a steady-state cooling of nuclei is achieved through a rotation of crystals in a magnetic field H.

In the present paper we describe a spin refrigerator of a qualitatively new type, in which the electron subsystem, rather than the nuclear subsystem, is cooled in a steady-state fashion. The work required to cool this subsystem is performed by a nonresonant external electric field E, which acts on the electric dipoles of reorienting paramagnetic centers.

2. The present experiments were carried out on quartz crystals with $Al\text{-}O^-$ and $Ge^{3+}(Na)$ centers. The $Al\text{-}O^-$ centers form during the isomorphic substitution[4] $Si^{4+} \rightarrow Al^{3+}$ in an SiO_4 tetrahedron. A paramagnetic hole (spin $S = 1/2$) localizes at

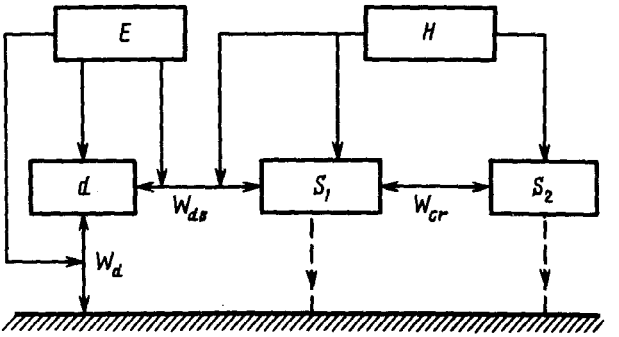

FIG. 1. Diagram of the interactions between different subsystems.

one of the two oxygen ions (we will call them 1 and 2) of a distorted AlO_4 impurity tetrahedron. An Al^{3+} ion and an O^- ion at which a hole is localized form an electric dipole $d = 10$ D (Ref. 4). A hole can execute a tunneling motion between oxygen positions 1 and 2; the effect corresponds to a reorientation of electric dipole d (Ref. 4). Consequently, both electric d dipoles and magnetic S_1 dipoles are associated with Al-O^- centers in quartz. [Fig. 1; the magnetic dipoles S_2 in this figure correspond to Ge^{3+}(Na) centers]. The field E lifts the orientational degeneracy of dipoles d. The Stark splitting is $2\Delta_E = (\mathbf{d}_1 - \mathbf{d}_2)\mathbf{E}$, where $\mathbf{d}_1$ and $\mathbf{d}_2$ correspond to the localization of the hole in positions 1 and 2.

According to Ref. 4, the field E influences the probability (w_d) for the reorientation of electric dipoles associated with Al-O^- centers. At $T = 4.2$ K we have $w_d \sim \Delta_E$, since the probability w_d is set by single-phonon processes, and the phonon density increases with increasing Δ_E. For Al-O^- centers, there may also be a change in the spin projection onto the magnetic field during reorientations of electric dipoles d, by virtue of the spin-orbit coupling. The effect would be a spin-lattice relaxation of these centers.[5] The probability for such joint reorientations of electric and magnetic dipoles, w_{ds}, is greatest for Al-O^- centers when the Stark energy is roughly equal to the Zeeman energy, $2\Delta_E \approx g\beta H$, according to Ref. 5. The direct coupling of the S_1 and S_2 dipoles with the lattice (Fig. 1) is negligibly small.

For paraelectric centers, an isothermal increase in the field E, followed by an adiabatic decrease of this field (as in adiabatic demagnetization), would make it possible to cool the system of electric dipoles associated with these centers to temperatures below the lattice temperature.[1] The electric dipoles would then assume the lattice temperature at a rate w_d.

The entropy of the system of electric dipoles in field E can be written in the form[1]

$$\sigma = Nk\{\ln[2\cosh(\frac{2\Delta_E}{kT})] - \frac{2\Delta_E}{kT}\tanh(\frac{2\Delta_E}{kT})\}. \quad (1)$$

Figure 2 shows the temperature (T) dependence of σ for various values of the field E. Let us assume that the field E acting on a sample is changed in a step fashion from E_1 to E_2 (the inset in Fig. 2) at a frequency ν_E. We also assume $(\mathbf{d}_1 - \mathbf{d}_2)\mathbf{E}_2 \gg (\mathbf{d}_1 - \mathbf{d}_2)\mathbf{E}_1$ and, furthermore, $(\mathbf{d}_1 - \mathbf{d}_2)\mathbf{E}_1 = g\beta H$. Taking into ac-

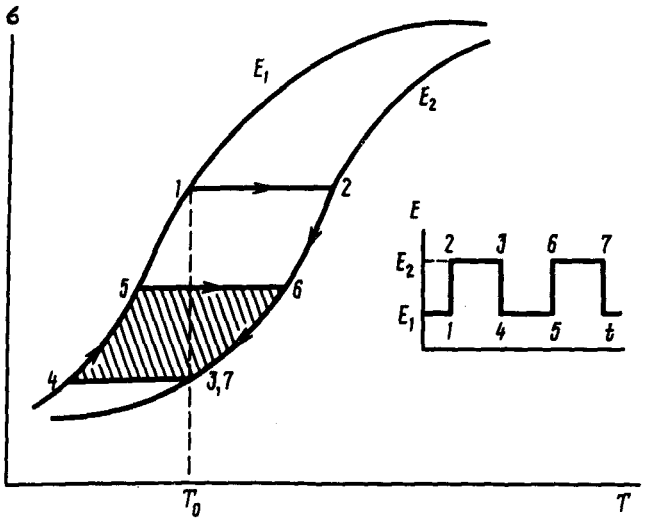

FIG. 2. Explanation of the operation of a spin refrigerator. T_0 is the lattice temperature. The inset shows the time (t) evolution of the field E.

count the comments above regarding the relaxation of the electric and magnetic dipoles, we can then set $w_d(\mathrm{II}) \gg w_d(\mathrm{I})$ and $w_{ds}(\mathrm{II}) \ll w_{ds}(\mathrm{I})$, where the I and II specify the half-periods corresponding to $E = E_1$ and $E = E_2$, respectively. Let us assume that the frequency of field E is such that we have $w_d(\mathrm{I})/2\nu_E \gg 1$ and $w_d(\mathrm{II})/2\nu_E \ll 1$; i.e., during half-periods I, dipoles d do manage to reach equilibrium with the lattice, while in half-period II they do not. The states of the system of electric dipoles at times $1-7$ (see the inset in Fig. 2) will then correspond to the states shown in the diagram in Fig. 2.

The system of electric dipoles d (Fig. 1) is thus periodically (during half-periods I) cooled below the lattice temperature T_0 under the influence of field E. It is during these half-periods that the probability w_{ds} is large; as a result, there will be a cooling of the spin system cycle after cycle. During half-periods II, when dipoles d quickly reach equilibrium with the lattice, the magnetic dipoles are isolated from the electric dipoles (w_{ds} is small), so the system of magnetic dipoles will remain in a steady state at spin temperatures below T_0.

3. Experiments have been carried out at $T = 4.2$ K on a superheterodyne ESR spectrometer in the 3-cm wavelength range. It was found by the method of continuous saturation of the ESR signals that the spin-lattice relaxation of Ge^{3+}(Na) centers in quartz occurs through a diffusion to Al-O^- centers. The rate of the spin-lattice relaxation of the Ge^{3+}(Na) centers is $\sim w_{cr}$ (Fig. 1), where w_{cr} is the probability for cross-relaxation transitions. This probability depends on ΔH, i.e., the difference between the resonant fields for the Al-O^- and Ge^{3+}(Na) centers.

When quartz samples with Al-O^- centers are acted upon by a varying electric field, the intensity of their ESR signals increases,[6] because of a quasisteady increase in the magnetization of these centers by the field E (Ref. 7). The field E has no direct effect on the Ge^{3+}(Na) centers. During the application of a varying field E to quartz samples with Al-O^- and Ge^{3+}(Na) centers (the inset in Fig. 2 shows the form of the

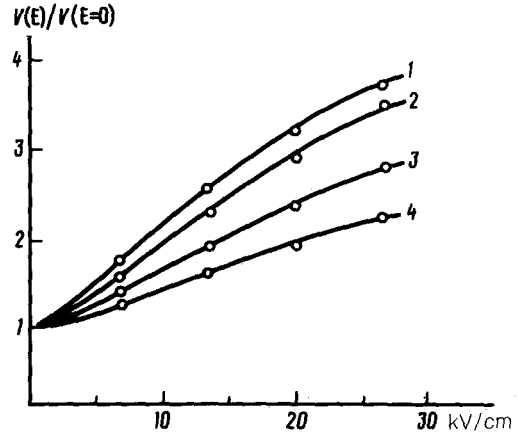

FIG. 3. Intensities of the ESR signals, V, versus the field E. 1-Al-O$^-$ centers; 2,3,4-Ge^{3+}(Na) centers, with ΔH values of 8.5, 15.1, and 26.7 Oe, respectively. Here $V(0)$ is the intensity of the ESR signals at $E=0$ ($\nu_E = 300$ Hz, $T_0 = 4.2$ K).

field E when the internal fields of the defects are taken into account[5]); however, the decrease in ΔH will be accompanied by, in addition to an increase in the ESR signals of Al-O$^-$ centers, a steady-state intensification of the ESR signals of Ge^{3+}(Na) centers (Fig. 3). The rate of increase of the intensities of the ESR signals of the Ge^{3+}(Na) centers after the field E is turned on is determined by w_{cr}; for curves 2–4, this rate is 2.0, 0.6, and 0.3 s^{-1}, respectively. These values agree with the values of the spin-lattice relaxation rates found from saturation curves for Ge^{3+}(Na) centers.

The spin temperatures T_s to which the paramagnetic centers are cooled can be determined, according to Boltzmann statistics, from the ratio of the intensities of the ESR signals with and without the field: $V(E)/V(0) = \tanh(g\beta H/kT_s)/\tanh(g\beta H/kT)$. For curves 1–4, these temperatures turn out to be 0.6, 1.5, 2.2, and 3.5 K, respectively.

In summary, a varying field E cools the system of Al-O$^-$ centers in our experiments. Through cross-relaxation, this cooling is extended to the Ge^{3+}(Na) centers. This cooling of paramagnetic centers might be utilized to control the magnetic characteristics of crystals by means of a field E in magnetooptic, rf spectroscopy, and other experiments.

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