Spin-selective tunneling of electrons between defects in crystals

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A spin-selective electron tunneling between excited states of radiation-induced defects in CaO crystals has been detected. The electrons tunnel only from single spin sublevels of an excited triplet state of the F_A defects.

In the present letter we report the use, for the first time, of a visually detected, delayed electron spin resonance at $T = 1.2$ K for studying electron tunneling between the F_A and F^+ defects in CaO crystals. The radiation defects are produced by bombarding the crystal by fast neutrons at the dose level of 10^{16} n/cm^2. The energy structure of vacancy-type defects in CaO was studied in Refs. 1–3. The transfer of electrons between the F and F_A^+ defects was reported in Ref. 4.

In the method proposed by us, the delayed magnetic resonance is produced between triplet spin sublevels of photoexcited F_A centers, and the light from spatially separated F centers is detected. This approach allowed us to selectively monitor the tunneling of electrons from single spin sublevels, T_x, T_y, and T_z, of the F_A centers. The energy-level diagram of the singlet and triplet states of the F_A center in a zero magnetic field is shown in Fig. 1. The F_A centers are excited to the ^1E singlet state by a pulse from an Ar^+ ion laser ($\lambda = 458$ nm, $\tau = 20$ ms). The nonradiative intersystem relaxation then causes the spin sublevels, T_x, T_y, and T_z, of the triplet $^3A_1$ state of the F_A center to become populated. The radiative sublevels, T_x and T_y, decay spontaneously in $\tau = 3$ ms, whereas the "dark" spin sublevel, T_z, is a long-lived sublevel with a lifetime $\tau > 400$ ms. Figure 2 shows a series of laser and microwave pulses that bombard the crystal. Also shown in this figure is the light "response" from the F centers. The light from the F centers is detected $t = 250$ ms after the laser pulse, when all radiative states of the defects in the crystal had decayed spontaneously. Light from the F centers is not detected after the indicated delay time. At the time $t = 265$ ms, the crystal receives a microwave pulse ($\nu = 1.369$ GHz, $\tau = 100$ μs), which is at reso-

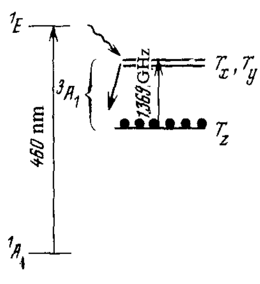

FIG. 1. Energy structure and electronic transitions in a system of singlet levels and triplet spin sublevels in a zero magnetic field for an F_A(Mg) center in CaO.

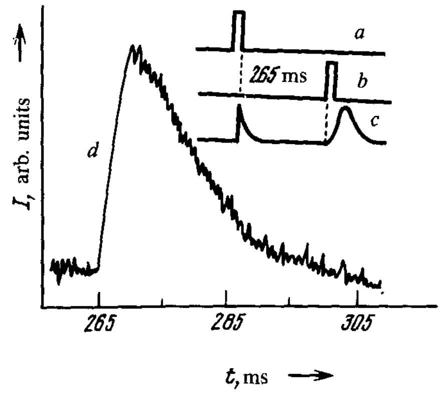

FIG. 2. A series of pulses bombarding a crystal. *a*—Laser pulse; *b*—microwave pulse; *c* and *d*—laser-pulse "response" from the *F* centers, $T = 1.2$ K.

nance with the transition $T_z \rightarrow T_x$, T_y in the F_A center. A magnetic resonance initiates a transfer of electrons to the T_x and T_y states, causing the appearance of an additional phosphorescent pulse of the F_A centers. Of particular interest is the fact that the F centers emit a light pulse (Fig. 2), during which there is no laser pumping.

Because of a large energy mismatch between the levels of these centers, there is no radiative or nonradiative transfer of energy from the F_A center to the F center. At the temperature of the experiment $T = 1.2$ K, the recombination processes involving the conduction band are also ruled out for the F_A center.

Consequently, the magnetic resonance at the triplet spin sublevels of the F_A centers induces electron tunneling from the F_A centers to the F^+ centers which is accompanied by a photochemical conversion of the defects according to the reaction

$$F_A^* + F^+ \rightarrow F_A^+ + F^* .$$

The electron tunneling causes the formation of F centers in the excited state.

Furthermore, these results are direct experimental proof that (a) electron tunneling begins from the excited triplet states of the F_A center and (b) the tunneling probability depends strongly on the initial spin state of electrons; the electrons tunnel from the T_x and T_y spin states but not from the T_z spin state.

The spin-selective electron tunneling we have detected might be attributed to the fact that triplet spin states, T_x, T_y, and T_z, are characterized by different spatial distributions of the wave functions because of the different levels of mixing with the excited singlet states.

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