Radiation Effects & Defects in Solids, Vol. 155, pp. 159–163
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MAGNETIC RESONANCE INVESTIGATION OF F CENTERS IN LIF CAUSED BY IONIZING RADIATION

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(Received 7 April 2000)

Damage caused in lithium fluoride crystals by γ-rays, electrons, and heavy ions includes, besides other point defects, F centers and their agglomerates. We have studied the paramagnetic F centers by measurements of the nuclear spin relaxation (NSR) of ^{19}F and ^{7}Li, the return of the magnetic circular dichroism of absorption (MCDA) after switching off the resonant microwave field and by optical absorption. The magnetic resonance experiments were performed over a range of defect concentrations to measure the longitudinal spin lattice relaxation times T_{1e} of the F centers. Over several orders of magnitude, the NSR rate T_{1n}^{-1} is proportional to the concentration of F centers. Measurements at different frequencies and temperatures suggest a spin diffusion limited NSR process. Raising the temperature of the crystal initiates an annealing process that can be observed as a decrease of the NSR rate.

Keywords: F center; Nuclear spin relaxation; MCDA, Spin-diffusion limited; Annealing

Ionizing radiation creates in lithium fluoride crystals defects consisting primarily of F centers and their agglomerates. The electron spin of the F centers couples to neighboring nuclei via hyperfine interaction. Fluctuations of the F center electron therefore induce flips of the nuclear spins, which drive the nuclear spin polarization towards thermal equilibrium.

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Hence, measurements of the nuclear spin relaxation (NSR) provide information about the dynamics of the F center. The defects studied in this work were created by irraditation with swift heavy ions (^{208}Pb 29 MeV/u, ^{32}S 11.4 MeV/u. ..., $\Phi = 1 * 10^{10} - 3 * 10^{13}\,\mathrm{cm}^{-2}$) electrons (0.5–20 MeV, $D = 0$–20 MGy) or γ-rays (1.17 and 1.33 MeV, $D = 1$ kGy–5 MGy) at room temperature. While the irradiation with electrons or γ-rays causes a homogeneously distributed defect concentration, the damage from the swift heavy ions is inhomogeneously distributed along the ion tracks. The correlations between these centers, their relaxation times T_{1e} and the NSR rates $1/T_{1n}$ are the objective of this work.

T_{1e} of the paramagnetic F centers was measured via the return of the F center MCDA after switching off the resonant microwave field [1]. For three different F center densities the correlation times are, $T_{1e} = 2.25$ s for D = 1 kGy, $T_{1e} = 650$ ms for D = 126 kGy and $T_{1e} = 35$ ms for D = 1.122 MGy. All values were measured in a magnetic field of 930 mT at 1.5 K (Fig. 1). The NSR rate T_{1n}^{-1}, which was measured by the standard saturation-recovery technique, is determined by the density and by the

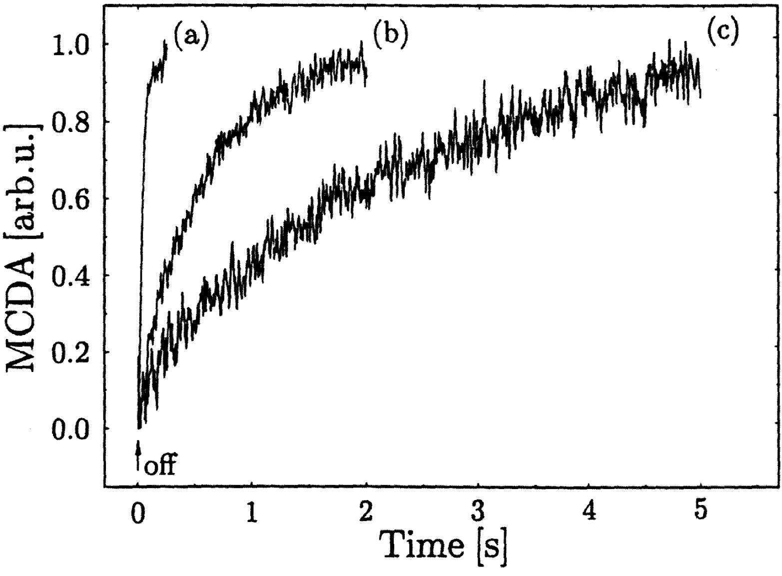

FIGURE 1 Return of the F center MCDA in γ-irradiated LiF after switching off the microwave (25.5 GHz), recorded at $T = 1.5$ K, $\lambda = 265$ nm and B = 930 mT. The applied γ-dose was (a) 1.122 MGy, (b) 126 kGy and (c) 1 kGy.

relaxation time T_{1e} of the F centers. The nuclear spin polarization created by this process is distributed to more remote spins by spin diffusion

$$\frac{\partial m_z}{\partial t} = -(m_z - m_0)\left(\frac{r_0}{r}\right)^6 \omega_{\text{int}}^2 \frac{T_{1e}}{1+\omega_L^2 T_{1e}^2} + D_s \nabla^2 m_z \tag{1}$$

Here, m_z is the longitudinal nuclear spin polarization, r_0 an atomic-scale length, w_{int} denotes the strength of the hyperfine interaction, w_L the Larmor frequency, and D_s the spin diffusion constant. If the process is spin diffusion limited, the solution of this equation is [2, 3]

$$1/T_{1n} = 8.5 N_p \left(\frac{2}{5} S(S+1)\gamma_p^2 \gamma_I^2 \frac{T_{1e}}{1+\omega_L^2 T_{1e}^2}\right)^{1/4} D_s^{3/4}, \tag{2}$$

where N_p denotes the F center concentration, S the electronic spin, and $\gamma_p(\gamma_I)$ the gyromagnetic ratio of electronic (nuclear) spin. The parameters of this equation can be measured independently. For a sample irradiated with γ-rays of a ^{60}Co-source and a dose of 126 kGy, the values are: $T_{1e} = 650$ ms (MCDA at 1.5 K); $N_p = 1.5 * 10^{18}\ \text{cm}^{-3}$ (optical absorption); $D_s = 1 * 10^{-12}\ \text{cm}^2\,\text{s}^{-1}$ (calculated); $w_L = 127$ MHz. Using these values, we obtain from Eq. (2) a ^{7}Li relaxation time $T_{1n} = 144$s, which agrees surprisingly well with the experimental value of 145 s. Moreover, for both ^{19}F and ^{7}Li, the NSR rates obey a $w^{-1/2}$-law in accord with Eq. (2). In consequence, Eq. (2) forms a basis to study the F center dynamics by NSR experiments over a large parameter range, which is not accessible to direct EPR or MCDA measurements. For samples irradiated by heavy ions, measurements of the nuclear spin relaxation rate as a function of dose yielded good agreement with the Lidiard relation [4], $T_{1n}^{-1} = T_{1n\infty}^{-1}\sqrt{1-\exp(-K\Phi)}$. In the case of γ-ray irradiation, we found a linear increase of the NSR rate, with no signs of saturation for medium doses ($D < 5$ MGy). Figure 2 shows, that the NSR rate $1/T_{1n} \propto T$ for $T < 70$ K and $1/T_{1n} \propto T^{1/2}$ above 70 K, or by use of Eq. (2), $T_{1e}(T > 70\,\text{K}) \propto T^2$ and $T_{1e}(T < 70\,\text{K}) \propto T^4$. This indicates a Raman process as the dominant relaxation mechanism.

If the sample is heated to 500 K or higher, an annealing process sets in that eliminates the F centers. We investigated this process by rapidly heating the crystal to the annealing temperature T_a, keeping it there for $\Delta = 20$ min, and cooling the sample to room temperature, where the NSR

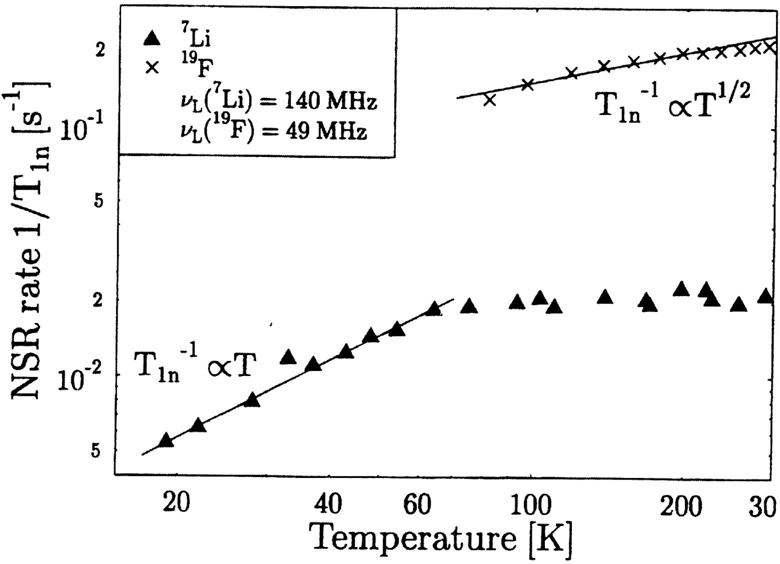

FIGURE 2 Temperature dependence of the NSR rates of ^{7}Li and ^{19}F in single-crystalline LiF irradiated with γ-rays ($D = 525$ kGy).

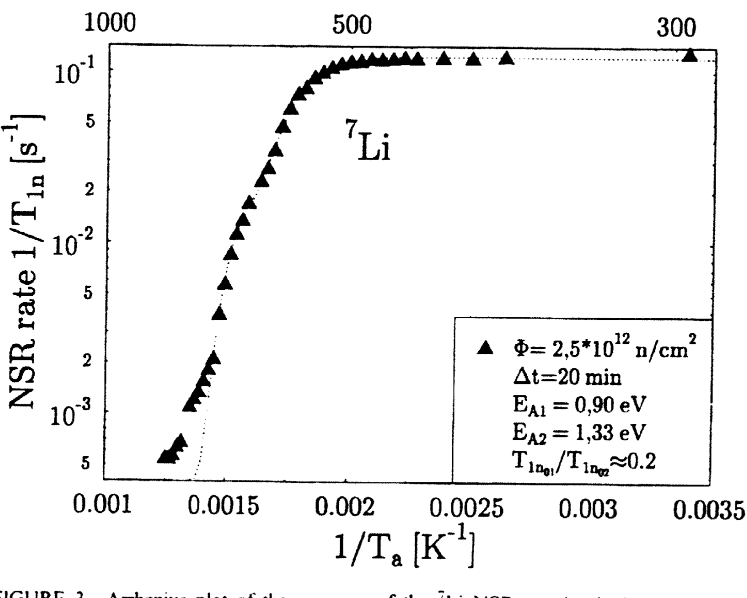

FIGURE 3 Arrhenius plot of the recovery of the ^{7}Li NSR rate in single-crystalline LiF irradiated with ^{208}Pb ($\Phi = 2.5 * 10^{12}$ n/cm^2, E = 29 MeV/u). Data are fitted by Eq. (4) using the parameters $E_{A1} = 0.90$ eV and $E_{A2} = 1.33$ eV and $T_{1n_{01}} = 10$ s, $T_{1n_{02}} = 50$ s as best fit.

rate was measured. For a thermally activated first order process, the density N_p of the F changes as

$$\dot{N}_p(t) \propto -N_p(t) \exp(-E_A/kT) \tag{3}$$

The experimental data (see Fig. 3) indicate two different recovery stages. A reasonable fit is obtained by the corresponding solution of Eq. (3)

$$\begin{aligned} 1/T_{1n} = {} & T_{1n_{01}}^{-1} \ \exp(-B_1 * \ \exp(-E_{A1}/kT_a)\Delta) \\ & + T_{1n_{02}}^{-1} \ \exp(-B_2 * \ \exp(-E_{A2}/kT_a)\Delta) + T_{1n_\infty}^{-1} \end{aligned} \tag{4}$$

using $E_{A1} = 0.90$ eV and $E_{A2} = 1.33$ eV and $T_{1n_{01}} = 10$ s, $T_{1n_{02}} = 50$ s as best fit parameters. B_1 and B_2 are fit parameters with the dimension s^{-1} but have no physical importance.

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