# ELECTRON-NUCLEAR DOUBLE RESONANCE OF *F* CENTERS IN LITHIUM FLUORIDE*

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In LiF, the spin resonance of the *F*-center electron is partially resolved by its hyperfine interactions with neighboring nuclei.[1] Discrepancies between observed and theoretically predicted values of these interactions[2] are most serious for the lighter alkali halides. This situation motivated a more detailed experimental examination of the electronic structure of this center using the new double resonance technique.[3]

A crystal of LiF was heavily irradiated with 50-kv x-rays thereby inducing a high concentration of *F* centers. The sample, on a long rod, was placed in thermal contact with a metal container filled with liquid helium, at the same time projecting into a rectangular X-band cavity. While partially saturating the electron spin resonance with a microwave field at 9200 Mc/sec, an additional intense radio-frequency field was superimposed on the sample using fine wires suspended inside the metal cavity. As the frequency of this auxiliary field is varied, the spin resonance signal changes whenever the field stimulates nuclear spin transitions between the Zeeman hyperfine levels of the *F*-center electron. The change becomes appreciable as this hyperfine interaction "resonance" of the neighboring nucleus is saturated. A maximum steady state change is approached monotonically by saturating both the electron and nuclear resonances. The rf field intensity for this was around 0.5 gauss and depended somewhat on the particular nucleus involved.

Both the microwave and radio-frequency magnetic fields were perpendicular to a steady magnetic field, $H$, of approximately 3200 oersteds. The crystal [100] axis was along the length of the bar and perpendicular to $H$. On rotation of the sample about this axis, $H$ remained in a (100) plane and varied its angle with respect to a [100] axis. The allowed nuclear spin transitions are $\Delta m_\alpha = \pm 1$. The separations between the connected spin levels of nucleus $\alpha$ are

$$h\nu_\alpha = \left| \tfrac{1}{2}\left[E_\alpha{}^S + E_\alpha{}^{\text{dipole}}\right] \pm \mu_\alpha H / I_\alpha \right| , \quad (1)$$

in the notation of reference 2. For analysis of the experimental data, the dipolar part can be re-expressed as

$$E_\alpha{}^{\text{dipole}} = (3\cos^2\theta_\alpha - 1)\, 2\beta \frac{\mu_\alpha}{I_\alpha} \int \varphi_n(n) \times \frac{[3\cos^2\gamma_{\alpha n} - 1]}{r_{\alpha n}{}^3}\, \varphi_n{}^*(n)\, d\tau_n , \quad (2)$$

where $\theta_\alpha$ is the angle between $H$ and the line, $\alpha$-$n$, joining the $F$ center, $n$, to nucleus $\alpha$, while $\gamma_{\alpha n}$ is the polar angle of integration measured from this axis.

These interactions of (1) give rise to an angle-dependent double resonance spectrum which can be grouped into sets of lines associated with the angle-independent term $E_\alpha{}^S$. The nuclei responsible for any set are crystallographically equivalent with respect to the halide ion vacancy of the *F* center, and form a shell of sites whose distance from this vacancy is $\sqrt{N}$ times the lattice parameter, $N$ being integral. Lines were observed for shells out to $N = 6$. Figure 1 shows the sets of double-resonance lines for the first 3 shells surrounding the *F* center when the direction of $H$ in a (100) plane is varied between 0 and $\pi/4$ with respect to a [100] axis.

The experimental results for the constants, $E_\alpha{}^S/h$ and $E_\alpha{}^{\text{dipole}}/h(3\cos^2\theta_\alpha - 1)$, which are direct measures of the *F*-center electronic wave function $\varphi$, are presented in Table I. The line widths of the double-resonance lines are also included as a measure of the errors inherent in these measurements.

The contact term result for shell 1 agrees very well with its earlier determination. The decline of this term in shell 2 is much faster than expected either from the theory[2] or experiment,[1] suggesting an even greater concentration of $\varphi$ within the vacancy. However, the earlier results rested on the apparently unjustified as-

Table I. Experimental values of hyperfine interaction energies in megacycles per second for the first three shells of nuclei surrounding the *F* center in LiF. The last column contains the half-width of the double-resonance spectral lines associated with the shell.

| Shell | $\dfrac{E_\alpha{}^S}{h}$ | $\dfrac{E_\alpha{}^{\text{dipole}}}{h(3\cos^2\theta - 1)}$ | $\Delta\nu_\alpha$ |
|---|---|---|---|
| 1 | 38.5 | +3.1 | 0.48 |
| 2 | 1.34 | +0.56 | 0.07 |
| 3 | 0.54 | +0.64 | 0.03 |

sumption that the width of each of the 19 partially resolved components was predominantly the hyperfine interaction of the $F^{19}$ nuclei of shell 2. As a contrast, the dipolar interaction shows a remarkable persistence. This fact alone made portions of the outer shell spectra observable, their $E_\alpha{}^S$ being very much smaller.

It was expected that the lack of axial symmetry of $\varphi$ about $\alpha$-$n$ axis would be much stronger in the more closely packed LiF cyrstal than in KCl. No evidence of this appeared either as a quadrupole splitting of shell 1 and shell 3 lines, or as an additional $\theta$-dependence of shell 2 lines. Appreciable axial asymmetry would cause the intersecting low-frequency traces in Fig. 1 to be different at 0° and 45°, instead of being practically equal. Work in progress on the intermediate alkali halides may clarify this point.

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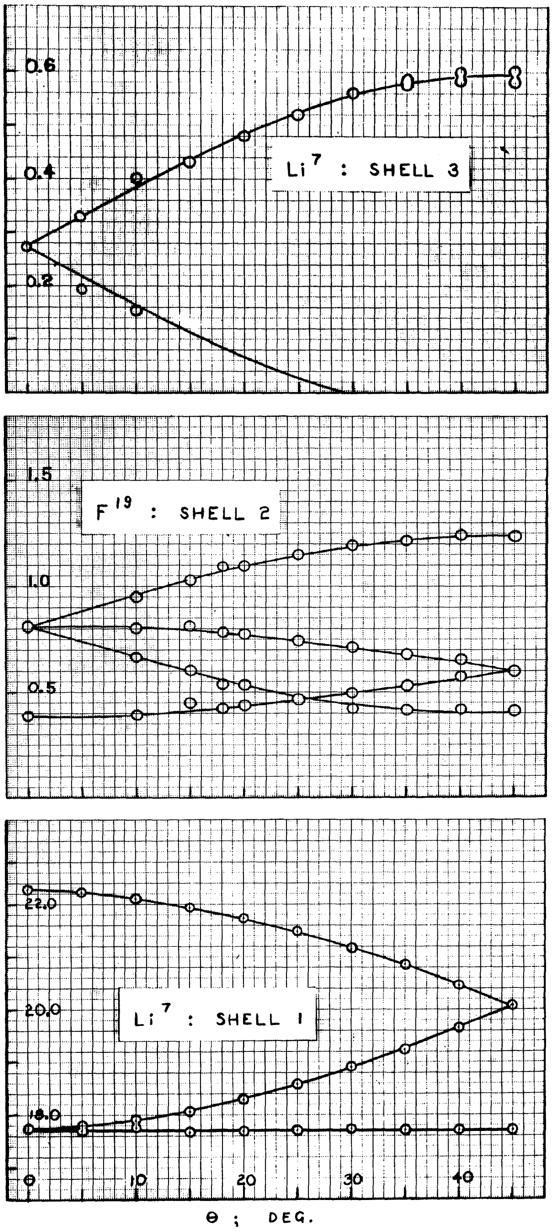

FIG. 1. Variation of double-resonance spectra as a function of the angle $\theta$ between $H$ and the [100] axis in the (010) plane. Splitting in shell 3 is due to slight elevation of $H$ out of this plane.

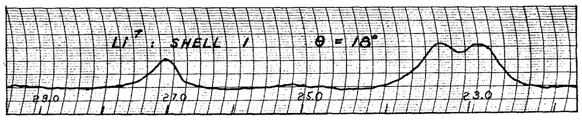

FIG. 2. Portion of double-resonance spectrum due to the first shell, demonstrating the extraordinary line width.

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[1] N. W. Lord, Phys. Rev. 105, 756 (1957).

[2] B. S. Gourary and F. Adrian, Phys. Rev. 105, 1180 (1957).

[3] G. Feher, Phys. Rev. 105, 1122 (1957).

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# "RADIATION BELT" AND TRAPPED COSMIC-RAY ALBEDO*

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The existence of a belt of unidentified ionizing radiation observed by the Explorer satellites[1] has led to speculations concerning the nature and source of this radiation. In a recent Letter Dessler[2] suggests that the radiation is related to auroral particles and is accelerated by hydromagnetic waves. This suggestion runs into several difficulties: (i) Hydromagnetic waves traveling along lines of force are probably the accelerating agent for auroral protons,[3] but only in the auroral zone (60°-70° magnetic latitude), where both waves and particles are trapped and can exchange energy.[4] (ii) Waves probably do not propagate perpendicular to the lines of force because of the strong reduction in conductivity