in favor of the singlet ($S=0$) ground state. Assuming the Landé interval rule given by Eq. (1), the intensity of transitions belonging to the $S=1$ state is proportional to $[\exp(-J/kT)]/(TZ)$, where Z is the partition function for the six spin states. Comparison of the experimental intensities over a range of temperature with this expression gives the value 28 ± 4°K for both J_{nn}/k and J_{nnn}/k. A direct comparison of the temperature dependence of the intensities of nn, $S=1$ and nnn, $S=1$ transitions yields $J_{\mathrm{nnn}}/J_{\mathrm{nn}}=1.0\pm0.1$.

When comparing these results with the bulk properties of MnO we first assume that the value found above for the important ratio $r=J_{\mathrm{nnn}}/J_{\mathrm{nn}}$ is approximately applicable to MnO, despite the small change in lattice spacing in the mixed crystal. A value of 11.3°K for J_{nn}/k and J_{nnn}/k can then be derived for MnO by using the measured Curie-Weiss constant, $\theta=610°\mathrm{K}$,[4] in the relation $\theta=4J_{\mathrm{nn}}(1+r/2)S(S+1)/k$ with $S=5/2$ and $r=1$. The value of about 28°K found in the present experiments is surprisingly large compared with this susceptibility value. The discrepancy may be due to the Mn-Mn separation being slightly smaller in the mixed Mn:MgO crystal. It also seems possible that the true (high temperature) value of θ may be larger than 610°K, since this experimental value appears to refer to the small temperature range 120 to 300°K just above the antiferromagnetic transition at $T_N=116°\mathrm{K}$. Finally, concerning the values found for the anisotropic interaction parameters D and E, that for D can be mainly accounted for by the expected dipole-dipole interaction as can be seen from Table I. The value found for $E(\mathrm{nn})$, on the other hand, is unexpectedly large, and may be partly due to crystal field effects arising from distortion in the mixed crystal.

We are grateful to the General Electric Research Laboratory, Schenectady, and to the British Board of Admiralty for supporting this work. We would like to thank Dr. B. R. Judd and Dr. W. Marshall for helpful discussions, and Professor J. E. Wertz for kindly lending us the crystals used in the experiments.

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ELECTRONIC STRUCTURE OF THE "ANTIMORPH" OF THE *F* CENTER

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(Received January 4, 1960)

The electronic structure of the F center in alkali halides has been investigated extensively in the past, and a well-founded model has emerged from that work. The F center can be described as an electron trapped at a halide vacancy, and it has the symmetry appropriate to the vacancy.[1] Seitz[2] proposed in 1946 the existence of a center which should consist of a hole trapped at an alkali vacancy and thus would be the counterpart of the F center. He called it the "antimorph" of the F center.[3] In this Letter we report on the observation of a paramagnetic resonance spectrum in x-rayed LiF that is probably due to such a center, namely to a hole trapped at a Li^+ vacancy. The analogy with the F center, however, does not go any further as the symmetry of the new center differs considerably from that of the F center.

The total spread of the hyperfine spectrum is what one expects for a hole on fluoride ions.[4] The basic hyperfine spectrum consists of four equally intense lines, and for specialized orientations of the crystals in the dc magnetic field it degenerates into a three-line pattern with the intensity ratio 1:2:1. This indicates that the hole is localized on but two fluoride ions which are nonequivalent with respect to a general orientation of the applied dc magnetic field, but become equivalent for particular orientations. Thus this center can be regarded as a F_2^- molecule-ion, just as the previously investigated self-trapped hole.[4] One principal difference consists therein that in the latter case the two fluorine nuclei are equivalent for every orientation of the crystal in the dc magnetic field.[2] From a study of the anisotropy of the hyperfine interaction we can infer that the symmetry of the new center is that of an isosceles triangle. Its base, which coincides with the internuclear axis, is along [110], and its plane is a (001) plane. Equivalent orientations are equally abundant, but at liquid nitrogen temperature an individual center can be regarded as frozen in.

The principal axes of the tensors $T^{(1)}$ and $T^{(2)}$ that describe the hyperfine interaction with the two fluorine nuclei are tilted by an angle $\delta \approx 4°$ with respect to the internuclear axis as is illustrated in Fig. 1. Otherwise the tensors are identical. This corresponds to a "bending" of the molecular bond. The values for the principal components of the tensors are listed in Table I, and are compared to the corresponding quantities for the self-trapped hole. The hyperfine interaction in the new center is larger, indicating that the internuclear distance is slightly reduced. This fact, together with the tilt and the symmetry, suggests that the F_2^- molecule-ion is associated with a Li^+ vacancy. The perturbation potential of the Li^+ vacancy is, however, small compared to the spacing of the electronic energy levels of the F_2^- molecule-ion. Consequently, the spectroscopic splitting factor is very close to that of the self-trapped hole (Table II), and the same will be true for the optical absorption band (probably ~3400 A).

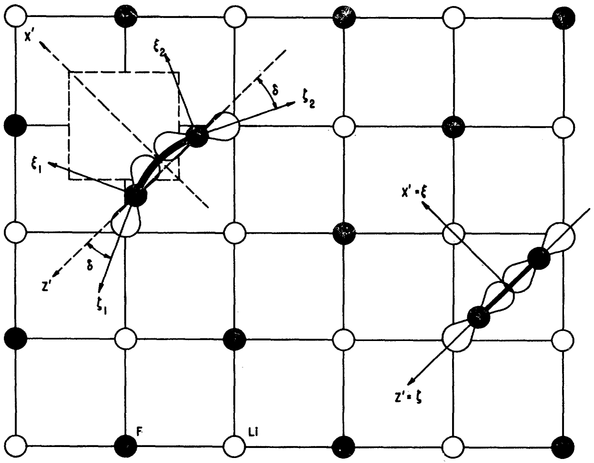

FIG. 1. Schematic representation of the new color center (left) and comparison with self-trapped hole (right). ξ_i, η_i, ζ_i are the principal axes of the hyperfine tensors $T^{(i)}$. The tilt δ is drawn exaggerated. The p functions are symbolized by figure eights.

Table I. Principal components of hyperfine tensors in gauss measured at 9.3 kMc/sec. (For designation of axes see Fig. 1.)

	$\lvert T_{\xi\xi}\rvert$	$\lvert T_{\eta\eta}\rvert$	$\lvert T_{\zeta\zeta}\rvert$
New center	~0	~0	915
Self-trapped hole (V center of reference 4)	59	59	887

Table II. Principal components of g tensors measured at 9.3 kMc/sec. (For designation of axes see Fig. 1.)

	$g_{x'}$	$g_{y'}$	$g_{z'}$	Error
New center	2.023	2.023	2.001	±0.002
Self-trapped hole (V center of reference 4)	2.0227	2.0234	2.0031	±0.001

The thermal stability of the new center is considerably higher than that of the self-trapped hole. This confirms the association of the hole with a negatively charged lattice defect. The self-trapped holes become unstable and migrate above about 125°K.[6] A fraction of them (order of magnitude 10%) become trapped at Li^+ vacancies so that the new, more stable center results, which does not anneal out until the crystal is warmed up to about 230°K.[7]

A search for the corresponding center in NaCl, KCl, and KBr was without results. In particular, the optical V_1 band does not seem to be due to this type of defect.

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