

hyperfine lines by its field independence and by its behavior when a circular polarizer is placed in the light beam in front of the cavity. A line characterized only by the quantum number $m=0$ should behave the same when either left-handed or right-handed polarized light is used. This is not true of the other transitions.

The experiment has also been attempted in potassium, with negative results. The reason for failure may be the fact that the relatively low hyperfine frequency—462 Mc/sec in K^{39} —is about equal to the optical absorption line width. In the stable rubidium and cesium isotopes, however, the splitting is greater than in sodium, so the success of the experiment in these materials seems assured.

¹ Such an instrument employing a cesium beam is manufactured by the National Company, Malden, Massachusetts.

² N. Ramsey, *Molecular Beams* (Oxford University Press, New York, 1956).

³ T. Carver and R. H. Dicke (private communication).

⁴ H. G. Dehmelt, *Phys. Rev.* **103**, 1125 (1956).

⁵ H. G. Dehmelt, *Phys. Rev.* **105**, 1487 (1957).

⁶ W. E. Bell and A. L. Bloom, *Phys. Rev.* **107**, 1559 (1957).

⁷ This pressure was chosen for reasons other than the present experiment and is probably not optimal.

Electron Spin Resonance of H Centers

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WE have x-rayed KCl crystals at 20°K and investigated their paramagnetic resonance spectra at this temperature, first without warming up, and then after pulse annealing at various temperatures. Before warming up, the spectra of two types of trapped-hole centers are predominant. The first type is an electron deficiency equally shared by two equivalent halide ions. Its basic hyperfine spectrum, which was extensively discussed in previous work,¹ is represented schematically in Fig. 1. The hyperfine splitting of the second-type center, which is the subject of this Letter, can be derived qualitatively from the hyperfine splitting of the first center by splitting every single line into a miniature pattern of the same kind. This is illustrated in Fig. 2, which shows a recording of that part of the "primary" spectrum which is encircled in Fig. 1. It can be con-

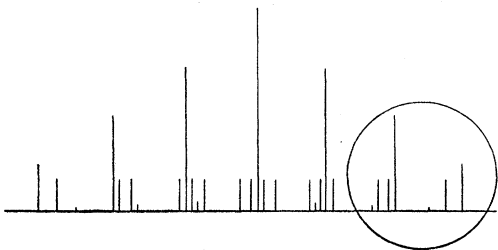


FIG. 1. Basic hyperfine splitting of Cl_2^- molecule ion.

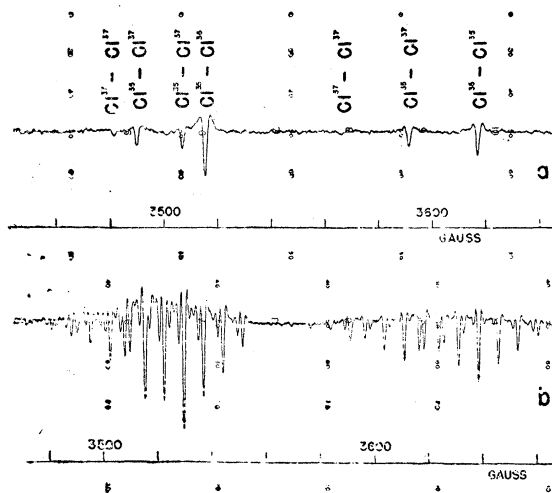


FIG. 2. Hyperfine splitting in the part of the basic spectrum encircled in Fig. 1. Derivative of dispersion measured at 20°K and 9.28 kMc/sec. (a) First-type center (V center of reference 1); (b) second-type center (H center).

cluded that the electron deficiency in the second center interacts strongly with two equivalent halogen nuclei, $\alpha=1$ and 2, but in addition interacts weakly with two more halogen nuclei, $\alpha=3$ and 4, which are again equivalent to each other. To a good approximation the hyperfine interaction with halogen nuclei 1 and 2 is axially symmetric, the axis being exactly a $[110]$ axis of the crystal. The hyperfine interaction with halogen nuclei 3 and 4 is also roughly axially symmetric, and the axis seems to coincide with the axis of the hyperfine interaction between the hole and nuclei 1 and 2. Thus it is most likely that the four nuclei lie in a straight row (with nuclei 3 and 4 at the ends) along a $[110]$ axis. All six $[110]$ axes are equally populated with these centers.

The hyperfine interaction with the four nuclei can be written in the form

$$g_0\beta_0\mathbf{S} \cdot \sum_{\alpha=1}^4 \mathbf{T}^{(\alpha)} \cdot \mathbf{I}^{(\alpha)},$$

where $\mathbf{T}^{(1)}=\mathbf{T}^{(2)}$ and $\mathbf{T}^{(3)}=\mathbf{T}^{(4)}$. The experimentally determined magnitudes of the tensor components are given in Table I. The principal axes $x'y'z'$ are the same for all tensors, and they are oriented with respect to the crystal axes as in our previous work¹ on the first

TABLE I. Absolute values (in gauss) of the principal components of the hyperfine-interaction tensors for the two types of centers. Data for the first-type center are taken from reference 1: $\mathbf{T}^{(1)}=\mathbf{T}^{(2)}$ from Table V, and the upper limits for the principal components of $\mathbf{T}^{(3)}=\mathbf{T}^{(4)}$ from the line width.

	$\mathbf{T}^{(1)}=\mathbf{T}^{(2)}$			$\mathbf{T}^{(3)}=\mathbf{T}^{(4)}$		
	$ T_{x'z'} $	$ T_{y'z'} $	$ T_{z'z'} $	$ T_{x'z'} $	$ T_{y'z'} $	$ T_{z'z'} $
1st-type center	~ 9	~ 9	101	< 0.4	< 0.4	< 0.4
2nd-type center	~ 0	~ 0	109	2.7	3.1	7.3

TABLE II. Principal components of the g tensors for the two types of centers. Data for the first-type center are taken from reference 1.

	$g_{x'}$	$g_{y'}$	$g_{z'}$
1st-type center	2.0428	2.0447	2.0010
2nd-type center	2.0227	2.0219	2.0024

center. The true symmetry of both type centers is orthorhombic. The g -tensors are given in Table II.

Since the hyperfine tensors $\mathbf{T}^{(1)}$ and $\mathbf{T}^{(2)}$ of the new second center do not differ appreciably from the corresponding hyperfine tensors for the first center, the structures of the two centers are closely related. It appears that the new center is simply a variant of the Cl_2^- molecule ion in which the wave function of the hole extends appreciably along the molecular axis towards the two nearest halogen nuclei, 3 and 4. Such a model might explain the larger oscillator strength of the second center compared to that of the first center, as well as the difference in their hyperfine interactions.

We have also considered models for the second-type center involving interstitial halide ions or "crowdions." However, we have observed centers of the second type in LiF, where interstitial fluoride ions are extremely unlikely.

If the crystal is warmed up to 42°K (i.e., to the first charge burst observed by Teegarden and Maurer²) and cooled to 20°K again, the first-type centers disappear and the spectrum of the second-type centers becomes stronger. After further warming to 60°K (to the second charge burst observed by Teegarden and Maurer²) and cooling again to 20°K, the second-type centers disappear and centers of the first type appear. Since the optical H band bleaches in the same temperature interval, this suggests that the centers of the second type are H centers. This identification is supported by the experiments of Compton and Klick³ with polarized light, which indicate that the H centers have [110] symmetry.

The influence of the annealing on the relative intensities of the two different spectra is summarized in Table III. Since we know¹ that the first-type centers are thermally stable up to 205°K, we may speculate that the first charge burst (42°K) is due to electrons released from some electron trap which annihilate

TABLE III. Relative total intensities of the dispersion spectra of the two centers measured at 20°K after successive pulse annealing at the three different temperatures at which Teegarden and Maurer² observed charge bursts. No corrections for possible saturation effects have been made.

	After irradiation at 20°K	After warming to 42°K	After warming to 58°K	After warming to 68°K
1st-type center	0.7	0.0	0.12	0.12
2nd-type center	1.0	1.4	0.00	0.00

¹ See reference 2.

preferentially the first-type centers. This process also appears to stimulate the transformation of first-type centers into second-type centers.

We have initiated combined optical and magnetic resonance experiments. A full account of the present investigations will be published in a forthcoming paper.

¹ T. G. Castner and W. Känzig, *J. Phys. Chem. Solids* **3**, No. 3/4 (1957).

² K. Teegarden and R. Maurer, *Z. Physik* **138**, 284 (1954).

³ W. D. Compton and C. C. Klick (to be published).

Spontaneous Emission of Radiation from an Electron Spin System

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IT was pointed out by Combrisson, Honig, and Townes¹ that under certain conditions energy which has been stored in a spin system may be spontaneously and coherently radiated into a resonant cavity at the Larmor precession frequency of the spins. In this note we wish to report the direct observation of such an emission.

If the magnetization which appears in a spin system in thermal equilibrium in an applied dc magnetic field H_0 is inverted by a 180° rf pulse or an adiabatic fast passage, the energy W put into the system is $W = gN\mu^2 H_0^2 / kT$, where N is the total number of spins, μ is the Bohr magneton, and g is the electronic g value. The condition for spontaneous reradiation of this energy is¹

$$N \geq kTV_c \Delta H \langle H_v^2 \rangle_{AV} / (4\pi Q_L \mu^2 H_0 \langle H_s^2 \rangle_{AV}),$$

where Q_L and V_c are the loaded Q and volume of the cavity, ΔH is the full width at half maximum of the spin resonance line, and $\langle H_s^2 \rangle_{AV}$ and $\langle H_v^2 \rangle_{AV}$ are the squares of the microwave fields averaged over the sample and cavity respectively. In previous experiments¹ phosphorus donors in silicon were used, but the above condition was not satisfied, and hence spontaneous oscillations were not observed.

In the present experiments, the spin resonance, which is inhomogeneously broadened by hyperfine interactions of the donor electrons with the Si^{29} nuclei,² was narrowed from 2.7 oersteds in width to 0.22 oersted through the use of a crystal of isotopically purified silicon³ [estimated final isotopic purity (99.88±0.08)% Si^{28}]. As a result the oscillation condition was easily satisfied.

The sample used in this experiment had a volume of about 0.3 cm³ and a phosphorus concentration of 4×10^{16} atoms/cm³. Its relaxation time at the operating temperature of 1.2°K was one minute; however, this can be greatly reduced by shining light on the sample⁴