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 Maurer[2]) 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 Maurer[2]) 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 Klick[3] 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 know[1] 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 preferentially the first-type centers. This process also appears to stimulate the transformation of first-type centers into second-type centers.

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 Maurer[a] 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

[a] See reference 2.

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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(Received November 4, 1957)

IT was pointed out by Combrisson, Honig, and Townes[1] 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.

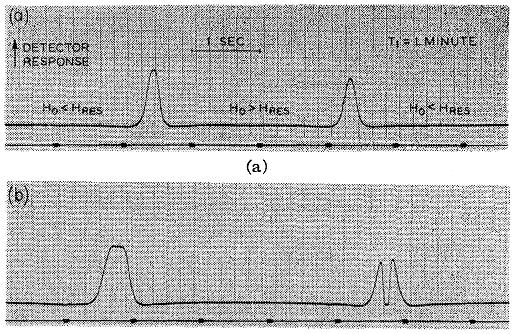
If the magnetization which appears in a spin system in thermal equilibrium in an applied dc magnetic field H_0 is inverted by a 180° rf pulse or an adiabatic fast passage, the energy W put into the system is $W=gN\mu^2H_0^2/kT$, where N is the total number of spins, μ is the Bohr magneton, and g is the electronic g value. The condition for spontaneous reradiation of this energy is[1]

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

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 experiments[1] 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 silicon[3] [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 sample[4]



(a)

(b)

FIG. 1. Adiabatic fast passage through one phosphorus hyperfine line in silicon-28 at 1.2°K and ~9000 Mc/sec. In Fig. 1(a) the dc magnetic field was on for less than the relaxation time; the resulting small magnetization did not satisfy the oscillation condition. In Fig. 1(b) the oscillation condition was satisfied. Note that as a result of the emission the magnetization at the center of the line is destroyed.

or otherwise injecting carriers into it. The cavity was resonant at ~9 kMc/sec, and its loaded Q at 1.2°K was ~20 000.

In Fig. 1 we show the observed signals under adiabatic fast passage conditions. In Fig. 1(a) the oscillation condition *is not* satisfied. Going slowly from a magnetic field below resonance to one above, the magnetization of the sample is inverted and its energy increased. If we now slowly reduce the field to below resonance before the spins relax appreciably, we return the magnetization to its equilibrium value. In Fig. 1(b) the oscillation condition *is* satisfied. The trace shows that the magnetization near the center of the line is destroyed through reradiation which occurs during the first passage.

This immediate emission may be avoided conveniently either by decreasing Q_L or by increasing ΔH during the first passage. We increased ΔH by introducing a field inhomogeneity which was removed after the magnetization was turned over. Then we turned off the microwave signal generator and returned the magnetic field to resonance. Figure 2 shows the pulse of microwave power delivered by the spins to the cavity at this time. The maximum amplitude of the rf field in the cavity during the pulse was of order ΔH. This limit is imposed since the negative susceptibility of the sample decreases at greater field strengths. After the

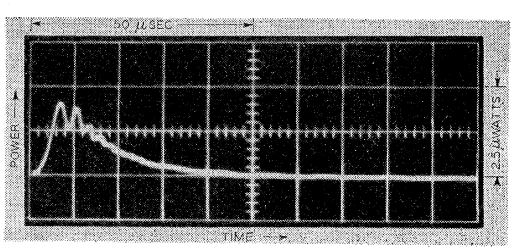


FIG. 2. Power output delivered by the spin system with the microwave oscillator turned off. The area under the curve agrees with calculated energy stored previously in the spin system.

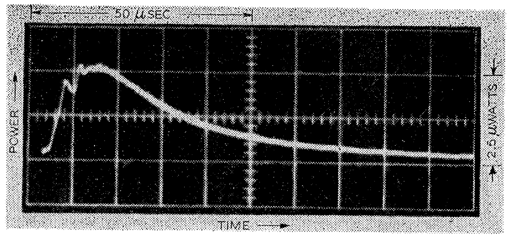


FIG. 3. Same as Fig. 2 with a large magnetization. The spin system was equilibrated at 8000 oersteds before letting it oscillate at 3000 oersteds.

spontaneous oscillation was over, the residual magnetization could be detected by turning on the signal klystron and again observing a fast passage signal. It was found that the magnetization associated with the central portion of the line had inverted itself during the oscillation. The observed energy output obtained from the area under the curves of Figs. 2 and 3, and corrected for the cavity coupling (reflection coefficient =0.94), corresponded roughly to that given up by the spin system. By equilibrating the magnetization at a higher field, a larger output was obtained, as shown in Fig. 3.

The traces of Figs. 2 and 3 show a superimposed amplitude modulation which gradually diminishes, and has a frequency of approximately the line width. The exact origin of this effect is not clear at present. It presumably arises from an interference between different spin packets within the line.

We wish to thank Mr. M. Kowalchik for his assistance in preparing the sample.

[1] Combrisson, Honig, and Townes, Compt. rend. **242**, 2451 (1956).
[2] Fletcher, Yager, Pearson, and Merritt, Phys. Rev. **95**, 844 (1954).
[3] We are indebted to the Isotope Division, Oak Ridge, Tennessee, for supplying us with 5 grams of an equal-mole mixture of Si and SiO_2 (99.98±0.02% Si^{28}). This was purified by us by making use of an aluminum reduction step, followed by a solution-precipitation step with molten tin. This procedure permitted 90% of the silicon to be recovered as fusible crystallites. This material was subsequently zone refined and doped with phosphorus to the desired concentration.
[4] G. Feher and R. C. Fletcher, Bull. Am. Phys. Soc., Ser. II **1**, 125 (1956).

Electrical Resistivity of Separated Lithium Isotopes*†

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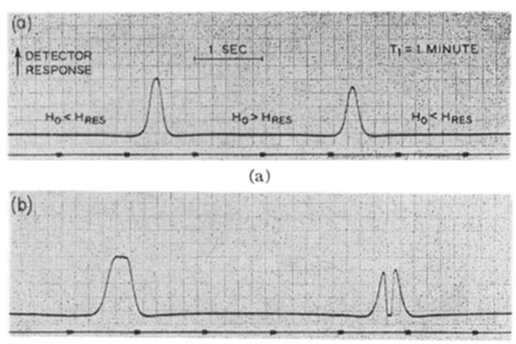
IN the theory of electrical conduction in solids, the resistivity is a function of temperature and of atomic mass, and an involved functional of the atomic fields. The dependence of resistivity on temperature has been studied extensively by many investigators, but the dependence on atomic mass has been little



(a)

(b)

FIG. 1. Adiabatic fast passage through one phosphorus hyperfine line in silicon-28 at 1.2°K and ~9000 Mc/sec. In Fig. 1(a) the dc magnetic field was on for less than the relaxation time; the resulting small magnetization did not satisfy the oscillation condition. In Fig. 1(b) the oscillation condition was satisfied. Note that as a result of the emission the magnetization at the center of the line is destroyed.

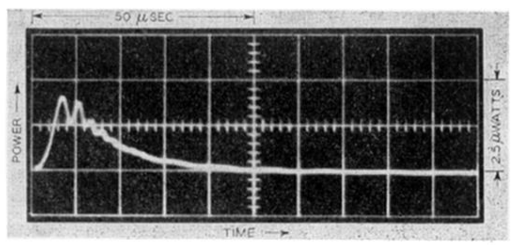


FIG. 2. Power output delivered by the spin system with the microwave oscillator turned off. The area under the curve agrees with calculated energy stored previously in the spin system.

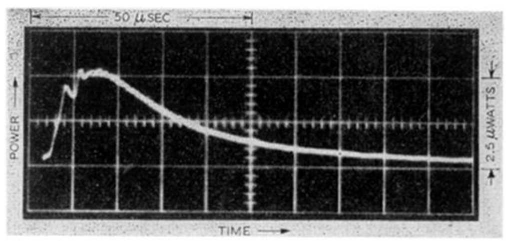


FIG. 3. Same as Fig. 2 with a large magnetization. The spin system was equilibrated at 8000 oersteds before letting it oscillate at 3000 oersteds.