

FIG. 1. Experimental arrangement.

showers in a target which usually was a slab of lead. The shower photons emerging from the target at various angles with the incident beam were detected by the radioactivity induced in copper foils. The 16-mil thick foils were 3-inch squares used whole or cut into halves or fourths and mounted as cylindrical segments with the beam as their axis. The mount was 21 cm from the target for angles  $<31^\circ$ . For data at angles  $>31^\circ$  the mount was 11 cm from the target and only the outer four foil positions were used.

The reaction employed was  $\text{Cu}^{63}(\gamma, n)\text{Cu}^{62}$ , the  $\text{Cu}^{62}$  undergoing decay with a ten-minute half-life. The excitation curve of this reaction has a peak at 17.5 Mev and a full width at half-maximum of about  $5\frac{1}{2}$  Mev;<sup>2</sup> hence the photons detected are those of energy near 17.5 Mev. After a 20-minute bombardment the fractional foils at a given angle were Scotch-taped together to form "standard" 3-inch square foils and were counted along with the monitor foil for 15 minutes using Victoreen 1B85 aluminum walled Geiger tubes.

Primarily this work was carried out with a target of 2.8 shower units of lead (taking 1 s.u. = 0.52 cm of Pb). 2.8 shower units is roughly the depth in lead at which there are a maximum number of gammas which can produce the reaction  $\text{Cu}^{63}(\gamma, n)\text{Cu}^{62}$ , and also it is about the depth at which maximum ionization occurs. Distributions at the shower maximum are easiest to calculate theoretically. The experimental results for 2.8 shower units are shown in curve 1, Fig. 2. The background, which is the relative activity observed when the target was absent, ranged between 15 percent at small angles to 10 percent at large angles. There was no straightforward way of subtracting off this background, but the relative error introduced is probably less than 10 percent. Points on the theoretical curve as calculated by Eyges and Fernbach<sup>3</sup> are indicated by the X's. The two curves are arbitrarily set equal at  $9^\circ$ . The agreement in shape seems good. To further compare the

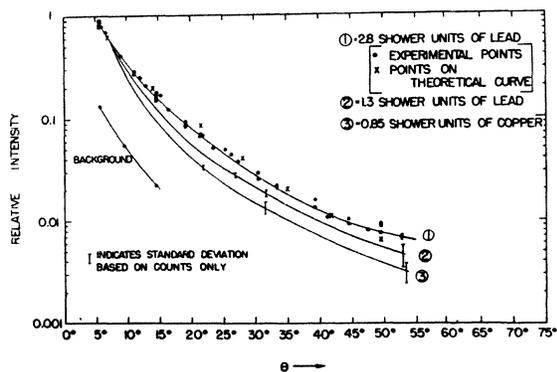


FIG. 2. Angular distribution of photons in showers in lead and copper.

two curves the total flux was integrated experimentally between  $0^\circ$  and  $5\frac{1}{2}^\circ$ , using a copper disk intercepting these angles and normalized to the same geometry as the other detectors. For the curves equal at  $9^\circ$  the experimental integral is 1.3 times the integral from the theoretical curve. This agreement is not bad since the method used by Eyges and Fernbach gives unreliable results at small angles.

Measurements were also made with 1.3 shower units of lead (curve 2) and 0.85 shower units of copper (curve 3). The curves have been set equal to the 2.8 shower unit curve at  $7\frac{1}{2}^\circ$ . No theoretical curves were available for comparison. For curves normalized in this manner the experimental integrals of the total flux between  $0^\circ$  and  $5\frac{1}{2}^\circ$  have the following relative values:

0.85 s.u. Cu	$1.75 \pm 10$ percent
1.3 s.u. Pb	$1.59 \pm 10$ percent
2.8 s.u. Pb	$1.00 \pm 10$ percent
5.9 s.u. Pb	$0.70 \pm 10$ percent.

(Curve for 5.9 s.u. taken at small angles only.)

The writer is indebted to Professor A. C. Helmholtz for helpful discussions of this work.

\* This work was sponsored by the AEC.

<sup>1</sup> J. W. Rose, Phys. Rev. **81**, 649A (1951).

<sup>2</sup> B. C. Diven and G. M. Almy, Phys. Rev. **80**, 407 (1950).

<sup>3</sup> L. Eyges and S. Fernbach, Phys. Rev. **82**, 307A (1951).

## Nuclear Magnetic Resonance Fine Structure in Liquids

H. S. GUTOWSKY AND D. W. MCCALL

Noyes Chemical Laboratory, University of Illinois,\* Urbana, Illinois

(Received April 16, 1951)

MULTIPLE nuclear magnetic resonance lines with separations of about 0.3 and 0.7 gauss, respectively, have been observed for  $\text{F}^{19}$  and  $\text{P}^{31}$  in several liquid phosphorus halides. The ratio of the  $\text{P}^{31}$  line splitting to that for  $\text{F}^{19}$ , in the same compound, equals  $\mu(\text{F}^{19})/\mu(\text{P}^{31})$ , indicating that the effect is associated with the local nuclear magnetic-dipole fields in the molecule. In the simpler, rigid-lattice solids, these local fields may split the resonance line into several components separated by as much as 5 to 10 gauss.<sup>1</sup> In liquids, molecular rotation ordinarily produces a zero time average for the local fields and single, narrow lines result.<sup>2</sup> The appearance of multiple nuclear magnetic resonance lines in particular liquids suggests an appreciable restriction of molecular rotation<sup>3</sup> in the liquid state.

Observations were made on  $\text{POCl}_3$ ,  $\text{POCl}_2\text{F}$ ,  $\text{POClF}_2$ , and  $\text{CH}_3\text{OPF}_2$ , at room temperature in an applied field of 6365 gauss, using the equipment and general procedures described previously.<sup>4</sup> The  $\text{F}^{19}$  magnetic resonances in all of the fluorine containing compounds are doublets with components of equal intensity. The  $\text{P}^{31}$  magnetic resonance is a singlet in  $\text{POCl}_3$  and a doublet in  $\text{POCl}_2\text{F}$  with components of equal intensity. In  $\text{POClF}_2$  and  $\text{CH}_3\text{OPF}_2$  the  $\text{P}^{31}$  resonance is a triplet in which the central line is double the intensity of the two equal, symmetrically placed satellites. The separations in gauss between adjacent components are summarized in Table I. The widths of the individual components

TABLE I. Multiple  $\text{F}^{19}$  and  $\text{P}^{31}$  nuclear magnetic resonance lines in liquids at room temperature.

Structure <sup>a</sup>	$\text{F}^{19}$		$\text{P}^{31}$		$\text{P}^{31}$ splitting <sup>c</sup>
	Structure <sup>a</sup>	Splitting <sup>b</sup>	Structure <sup>a</sup>	Splitting <sup>b</sup>	
$\text{POCl}_3$	—	—	singlet	0.000 gauss	—
$\text{POCl}_2\text{F}$	doublet(1-1)	0.294 gauss	doublet(1-1)	0.684	2.328
$\text{POClF}_2$	doublet(1-1)	0.279	triplet(1-2-1)	0.658	2.356
$\text{CH}_3\text{OPF}_2$	doublet(1-1)	0.320	triplet(1-2-1)	0.739	2.308

\* The numbers refer to the relative intensities of the component lines.

<sup>b</sup> Probable errors are  $\pm 0.003$  gauss for the  $\text{P}^{31}$  resonance and  $\pm 0.001$  gauss for  $\text{F}^{19}$ .

<sup>c</sup>  $\mu(\text{F}^{19})/\mu(\text{P}^{31}) = 2.324$ .

were determined in all cases by the magnetic field inhomogeneities and modulation effects, which gave apparent half-maximum widths of about 0.05 gauss.

The single  $P^{31}$  line in  $POCl_2$  reflects the absence of other important magnetic nuclei in the molecule. The  $P^{31}$  doublet in  $POCl_2F$  corresponds to an  $m_I$  for  $F^{19}$  of  $\pm\frac{1}{2}$ . In  $POClF_2$  and  $CH_3OPF_2$ , the two  $F^{19}$  nuclei form a singlet and a triplet nuclear state. For the singlet, the total nuclear spin component from the two  $F^{19}$  nuclei is  $M_I(F^{19})=0$ ; in the triplet,  $M_I(F^{19})=0, \pm 1$ . The central component of the  $P^{31}$  resonance line corresponds to  $P^{31}$  transitions in which  $M_I(F^{19})=0$ . The satellites correspond to  $M_I(F^{19})=\pm 1$ . The double intensity of the central line is a result presumably of the existence of two  $M_I(F^{19})=0$  states.

In general, for  $AX_n$  with structurally identical X's, the A magnetic resonance should have  $(2nI_x+1)$  equally spaced components with relative intensities given by the binomial coefficients. Proctor and Yu have reported<sup>6</sup> a five-component antimony resonance in aqueous solutions of  $SbF_6^-$ . Inspection of their experimental curve,<sup>5</sup> however, suggests that two additional weak satellites may be present; moreover, the relative intensities follow the proper sequence for seven components.

The splitting of the  $F^{19}$  resonance appears to arise solely from the  $P^{31}$   $m_I=\pm\frac{1}{2}$  states. The absence of any observable splitting by other  $F^{19}$  nuclei in molecules such as  $POClF_2$ ,  $CH_3OPF_2$ , and  $PF_3$  contrasts with the fine structure observed in rigid-lattice solids as a consequence of magnetic dipole interactions between like nuclei.<sup>1</sup> The difference may arise from the coupling of the nuclear and rotational angular momenta and the nature of the selection rules. Additional experiments and a more detailed analysis are in progress.

We are indebted to Drs. C. P. Slichter and L. M. Slifkin for several interesting conversations and suggestions regarding the interpretation of the results. Also, we wish to thank Dr. D. R. Martin for furnishing some of the compounds. Equipment was provided mainly by a Grant-in-Aid from Research Corporation.

\* Supported in part by ONR.

<sup>1</sup> Gutowsky, Kistiakowsky, Pake, and Purcell, *J. Chem. Phys.* **17**, 972 (1949).

<sup>2</sup> Bloembergen, Purcell, and Pound, *Phys. Rev.* **73**, 679 (1948).

<sup>3</sup> H. S. Gutowsky and G. E. Pake, *J. Chem. Phys.* **18**, 162 (1950).

<sup>4</sup> H. S. Gutowsky and C. J. Hoffman, *Phys. Rev.* **80**, 110 (1950); H. S. Gutowsky and R. E. McClure, *Phys. Rev.* **81**, 276 (1951).

<sup>5</sup> W. G. Proctor and F. C. Yu, *Phys. Rev.* **81**, 20 (1951).

### Detection of Slow Neutrons\*

R. HOFSTADTER, J. A. MCINTYRE, H. RODERICK, AND H. I. WEST, JR.  
*Department of Physics, Stanford University, Stanford, California*

(Received April 2, 1951)

IN the original publication on alkali halide scintillation counters<sup>1</sup> it was suggested by one of the present authors that a neutron counter could be made with a thalliated lithium halide as a scintillator. Subsequently, with the help of F. B. Harrison at Princeton, single crystals of LiI with a 1 percent TlI impurity were grown,<sup>2</sup> and it was found that they responded to ionizing radiation (gamma-rays and electrons). Since thermal neutrons are captured by  $Li^6$  (7.4 percent relative abundance) with a very large cross section ( $\sim 900$  barns), it may be expected that the alpha-particle and triton, which are released with a combined energy of 4.785 Mev,<sup>3</sup> will produce a large light pulse when slow neutrons are stopped in the  $Li^6$  of the LiI(Tl) crystal. Because the heavy particle pulse ( ${}^2_2He^4$ ,  ${}^3_1H^3$ ) will correspond uniquely to 4.79 Mev, while gamma-rays in a small crystal may be expected to lose a considerably smaller and variable energy, the proposed method of detecting slow neutrons would appear to be natural and simple: energy discrimination will distinguish slow neutrons from  $\gamma$ -rays, electrons, etc. This method was actually tried by one of the authors<sup>4</sup> without success because suitable crystals of LiI(Tl) had not been grown at the time.

Recently, we have examined single crystals of LiI(Tl) as neutron detectors<sup>2</sup> using the Stanford cyclotron as a source of slow neutrons. Figure 1(a) shows the pulses obtained when slow neutrons are

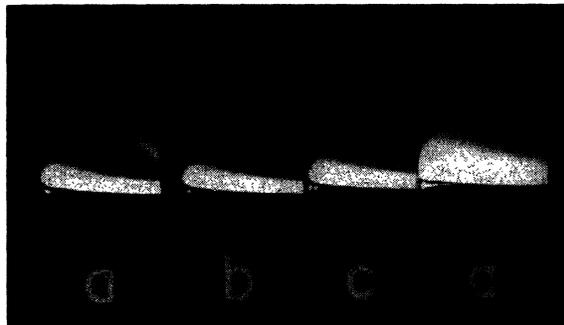


Fig. 1. Time exposures of LiI(Tl) scintillation pulses as observed on an oscilloscope screen. (a) crystal in beam of thermal neutrons, no absorber; (b) same as (a) except for boron absorber; (c) same as (a) except for cadmium absorber; (d)  $Co^{60}$  calibration pulses with double gain in amplifier.

allowed to impinge on the LiI(Tl) crystal which had been set atop a 5819 photomultiplier tube. Figures 1(b) and 1(c) show, respectively, the pulses obtained for a similar exposure, when the crystal was surrounded by thin boron and cadmium shields. The absence in 1(b) and 1(c) of the group of uniform pulses in 1(a) shows that slow neutrons formerly detected by the crystal are now being absorbed by the boron and cadmium and hence do not activate the crystal. The uniformity of the neutron pulses and their relatively large size indicate that most other radiations can be screened out by energy discrimination methods where desirable. The crystal of LiI(Tl) is thus selective to slow neutrons.

Figure 1(d) shows the pulses due to  $Co^{60}$  gamma-rays when the amplifier gain is doubled. By measuring relative pulse heights, and assuming that the light pulse size is proportional to energy, whatever the particle responsible for the energy loss may be, one obtains a value of 4.1 Mev for the neutron pulse heights. Since distortion by the amplifier occurs at larger pulse sizes, and since the crystal was yellow and not uniform, the agreement with the expected energy release must be considered good. In all parts of Fig. 1 the small pulses at the bottom appear to be due to photomultiplier noise and slow-neutron induced radioactivity in the iodine component of LiI(Tl).

A natural crystal of LiI(Tl), of thickness one centimeter, will capture about 0.6 of all the thermal neutrons passing through it. Hence the efficiency of this counter is very high. With enriched  $Li^6$  the efficiency can be made even higher. In these experiments the crystals used were  $4 \times 4 \times 6$  mm<sup>3</sup> and  $2 \times 4 \times 5$  mm<sup>3</sup> in size. The crystals were somewhat yellow, indicating the possible presence of an unwanted impurity. Clear colorless LiI(Tl) crystals should give even better results. The decay constant of LiI(Tl) is close to 1.2 microseconds; thus the speed is high enough to permit coincidence experiments with scintillation detectors for neutrons. The emitted light is blue-green and the density of LiI(Tl) is 4.06 g/cc. Since LiI(Tl) is extremely deliquescent, it must be placed in an air-tight container. This imposes no hardships with respect to detecting slow neutrons. The pulse heights for ionizing particles are of the order of  $\frac{1}{10}$  the size of NaI(Tl) pulses for equal energies.

The authors wish to thank Dr. W. E. Meyerhof for putting the cyclotron at their disposal.

\* This work was aided by the joint program of the ONR and AEC, and at an earlier time by a Signal Corps contract.

<sup>1</sup> R. Hofstadter, *Phys. Rev.* **74**, 100 (1948).

<sup>2</sup> R. Hofstadter, *Research Reviews (ONR)* **1**, 4 (September, 1949); *Nucleonics* **6**, No. 5, 70 (1950).

<sup>3</sup> Hornyak, Lauritsen, Morrison, and Fowler, *Revs. Modern Phys.* **22**, 291 (1950).

<sup>4</sup> R. Hofstadter, Brookhaven National Laboratory Internal Report, BNL-1-7 (November 1, 1949), unpublished.