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## EVIDENCE FOR DISCOVERY OF CHEMICALLY BOUND NEUTRONS\*

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Evidence has been obtained that neutrons form a weak bond with electrons trapped in LiF at  $4^\circ$ K. The chemistry of the resulting  $n^-$  species has been studied and shows that it decomposes at this temperature with a half-time in LiF of  $\sim 30$  sec. The bond energy is probably about 0.1 eV. Removal of the trapped electrons greatly reduces, if not completely destroys, the neutron retention in LiF.

This communication reports data which we believe demonstrate that neutrons can, under certain conditions, be bonded by electrons in an in- $\alpha$  and conditions, be bonded by electrons in an  $\alpha$  organic solid at  $4^\circ$ K. While it is known<sup>1-3</sup> from theory and scattering experiments that an interaction potential of 3860 eV exists between electrons and neutrons, to our knowledge no one has previously demonstrated existence of a bound state of the  $n<sup>-</sup>$  species.

The set of experiments described herein were carried out with Harshaw optical grade LiF crystals, cut into 2-mm-thick 2-cm squares. The crystals had been extensively irradiated with gamma rays and were characteristically colored' due to the presence of  $10^{17}$ - $10^{18}$  trapped electrons/cm3. Thermal neutrons were generated at an exposure flux of  $2 \times 10^6$  neutrons/cm<sup>2</sup> sec in a paraffin matrix by an electrostatic 14-MeV neutron generator. ' In a typical neutron irradiation, LiF samples were put into a quartz Dewar located inside a larger stainless steel Dewar for temperature control. The whole assembly was then placed in a previously determined irradiation position in the paraffin matrix. The usual bombardment time was fixed at 60 sec, and the neutron flux was monitored by gold or indium foils located in the inner Dewar.

A neutron counter-spectrometer system was

assembled from a thin  $(2 \text{ mm})$  1-in. -diam. Li<sup>e</sup>l(Eu) scintillation crystal integrally bonded to a 2-in. photomultiplier; pulses from this detector were recorded in a multichannel analyzer. The counter was mounted with the photomultiplier base down inside 4 in. of lead shielding lined with 20 mil of cadmium. The scintillation crystal itself was  $~100\%$  efficient for thermal neutrons. Because of a slight sensitivity to beta and gamma radiation, 20 mil of aluminum and 0.9 cm of lead absorbers were placed between the detector and samples being counted; the overall counting geometry for such samples was  $\sim 20\%$  as determined by a paraffin moderated  $Cf^{252}$  neutron source. Background spectra were collected frequently and were unaffected by the presence of nonirradiated LiF crystals placed on the detector. The integrated background under the neutron peak was, on the average,  $\sim 0.1$  count/min.

In series 1 experiments, LiF containing trapped electrons was irradiated with neutrons at room temperature, removed from the Dewar and generator vault, and placed on the counter in 40 sec. A spectrum was obtained from the composite data for 30 runs, each counted for 10 min, in which the events recorded decreased with increasing energy as shown in Fig.  $1(a)$ . The beta-gamma activity recorded is probably due to  $F^{20}$  from



FIG. 1. (a) Summed spectrum (30 runs) from LiF crystals neutron irradiated at room temperature (series 1). (b) Summed spectrum (23 runs) from LiF crystals neutron irradiated at  $4\textdegree K$  [series 2(b)]. Cross-hatched channels indicate a neutron calibration spectrum obtained from a  $CF^{252}$  source in paraffin. The events near 3.5 MeV may be due to the 3.51-MeV gamma of  $Al^{30m}$  formed by a  $(n,p)$  reaction on the silica Dewar used for the liquid helium, which was near the detector in some of these runs. (c) Summed spectrum (15 runs) from LiF crystals neutron irradiated at  $4^{\circ}\text{K}$ (series 3). Trapped electrons had been removed by heating before irradiation. The events near 3.5 MeV may be due to the 3.51-MeV gamma of  $Al^{30m}$ .

thermal neutrons and  $F^{18}$ ,  $O^{19}$ , and  $N^{16}$  produced by spallation reactions from fast neutrons on  $\mathbf{F}^{19}$ . If a smooth curve is drawn through the spectrum in Fig. 1(a), and this "tailing" subtracted from the events recorded in neutron channels 130-180, then there is no statistical evidence that there are any neutrons being emitted from the irradiated crystals. Such neutrons might have been present from the delayed neutron emitter  $N<sup>17</sup>$  formed by the reaction  $O^{17}(n, p)N^{17}$  on any oxygen impurities in the sample. Separate experiments demonstrated that the low fast-neutron flux  $(6 \times 10^4$  neu $trons/cm<sup>2</sup> sec)$  and 40-sec delay prevented detection of such delayed neutrons from even gram amounts of oxygen. These room-temperature experiments also rule out neutrons from other unknown delayed neutron emitters formed either from spallation reactions on trace elements or from the fission of heavy metals. It is also noted that  $N^{17}$  cannot be formed in the present experiments from the reaction  $\mathbf{F}^{19}(n, \text{He}^3)N^{17}$  because the 17-MeV threshoId energy is not obtainable from the neutron generator. Finally, the room temperature experiments also rule out the secondary reaction  $O^{18}(H^3, He^4)N^{17}$  resulting from thermal capture by  $Li<sup>6</sup>$  in the LiF crystals containing a possible oxygen impurity.

When LiF containing trapped electrons was irradiated with neutrons at 4'K in series <sup>2</sup> experiments, the situation was entirely different. In these experiments a LiF sample was placed in the inner Dewar which had been filled with liquid heIium; the outer Dewar contained liquid nitrogen. After a 1-min neutron irradiation, the Dewars were carried from the neutron generator vault to the neutron spectrometer. The LiF crystal was rapidly removed from the liquid helium and placed on the neutron counter to warm up. In a typical run, at a standard flux, a net of -2 neutrons mere recorded in neutron channels 130-180 [Fig. 1(b)]. There neutrons had been retained in the LiF cyrstal at  $4^{\circ}$ K at least  $40$  sec, the time elapsed between shutting off the neutron generator and removal of the LiF crystal from the liquid helium. Although the shape and location of the peak obtained on summing the data of 23 runs in Fig. 1(b) corresponds closely to that obtained from the  $Cf^{252}$  neutron calibration source, additional verification mas obtained in a separate series of experiments.

In these runs (series 4) LiF containing trapped electrons was irradiated with neutrons at 4'K as before. However, a piece of 20-mil-thick cadmium sheet was substituted for a similar amount of lead absorber placed between the irradiated LiF crystal and the neutron counter. In 14 separate runs, a net  $2 \pm 5$  total events were observed, which amounts to a reduction by a factor of almost 20 from what would be expected from otherwise identical series 2 experiments. The conclusion seems inescapable that thermal neutrons were indeed emitted from the LiF crystals on warming.

Another set of experiments (series 3) were carried out in which a LiF crystal containing

Series	Conditions of neutron irradiation	<b>Irradiation</b> temperature	Net events observed <sup>a</sup> $(channels 130-180)$	Net events observed <sup>b</sup> $(channels 130-180)$ per run
	LiF containing electrons	Room temperature	$9 \pm 6$ (30) <sup>c</sup>	$0.2 \pm 0.2^d$
$2^e$	LiF containing electrons	$4^{\circ}K$	(a) $50 \pm 8$ (30) (b) $43 \pm 8$ (23)	$1.7 \pm 0.3$ $1.9 \pm 0.3$
3	LiF, electrons removed by heating	$4^{\circ}K$	$9.5 \pm 5(15)$	$0.5 \pm 0.3$
4	LiF containing electrons, Cd over detector	$4^{\circ}K$	$2 \pm 5(14)$	$0.1 \pm 0.2$

Table I. Neutron yields from irradiated LiF crystals.

<sup>a</sup>Net events obtained from total observed events in the neutron region of the spectrum minus neutron background and estimated beta-gamma "tailing" (see Fig. 1) taken 40 sec after irradiation.

<sup>b</sup> This figure represents the net number of events observed in the neutron region of the spectrum per run at an average neutron irradiation flux of  $2 \times 10^6$  neutrons/cm<sup>2</sup> sec, taken 40 sec after irradiation.

Parenthetical numbers refer to the number of runs in each series.

<sup>d</sup>This value has been reduced by 20% to the same standard flux of the other series.

<sup>e</sup> Data from the last 23 runs of this series were obtained using a counting arrangement of lower background and with more absorber to reduce beta-gamma counting, and have been calculated separately.

electrons, which had been used successfully in retaining neutrons at 4'K in series <sup>2</sup> experiments, was heated to decolorize the crystal and remove retaining neutrons at 4 K in series 2 experiments<br>was heated to decolorize the crystal and remov<br>trapped electrons.<sup>6,7</sup> This crystal was then neutron irradiated in 15 additional experiments at 4'K and found to be four times less effective in retaining neutrons than before heating.<sup>8</sup> The data from the various series of experiments are summarized in Table I.

A few other observations are pertinent to the retention process. LiF crystals containing trapped electrons are not very effective in retaining neutrons when irradiated at 78'K in liquid nitrogen. It is not possible at present, however, to completely rule out the possibility of retention at this temperature because of the relatively poor background situation prevailing when these liquid-nitrogen experiments were being carried out. It has also been tentatively established that the number of retained neutrons at 4'K under standard experimental conditions increases linearly with the neutron flux, but not the time of irradiation. While neutron irradiations of 60 sec produce more bound neutrons than irradiations of 30 sec, 120-sec irradiations do not increase the yields. These saturation data suggest that an intrinsic process is operating at  $4^\circ$ K which causes LiF crystals to lose neutrons with a characteristic half-time of  $~10$  sec.

We believe these data provide convincing evidence that very slow-moving neutrons can be

captured by electrons in certain kinds of chemical states with a very low bonding energy, characteristic of a 4'K temperature. The zero-point vibrational energy of the  $n<sup>-</sup>$  species trapped in LiF can be estimated<sup>9</sup> to be about  $R\Theta_D$  or 1500 cal (0.1 eV). Consequently the internal bond energy in a LiF enviroment is probably this order of magnitude; conceivably it could be less if there is an appreciable dissociation barrier for  $n<sup>-</sup>$  consistent with a half-time of  $\sim 30$  sec at  $4^{\circ}$ K. It is possible that this bond energy can be identified with the intrinsic electron-neutron interaction arising from the meson dissociation effect. It is also possible that the species can be destroyed by nuclear capture of the neutron, when either diffusion or vibration of the  $n<sup>-</sup>$  brings it too close to a nucleus. In any event, with such a low binding energy, the stability of  $n<sup>-</sup>$  is undoubtedly sensitive to the chemical forces and processes that operate on the electron itself, and it is for this reason that we believe that it may be possible to find even more effective bonding agents than the present system. We also hope to study some of the other interesting questions raised by these observations, particular temperature effects, retention by other paramegnetic ions and molecules, and the kinetics of the formation and destruction of  $n^{-}$ .

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Phys. Rev. 114, 1571 (1959}.

M. Stafne, Phys. Rev. 90, 497 (1953).

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<sup>1</sup>L. L. Foldy, Rev. Mod. Phys. 30, 471 (1958).  ${}^{2}E$ . Melkonian, B. M. Rustad, and W. W. Havens, Jr.,

3D. J. Hughes, J. A. Harvey, M. D. Goldberg, and

 $4$ J. S. Nadeau, J. Appl. Phys. 33, 3480 (1962).

 $5$ Model 111, manufactured by Technical Measurement Corporation, Atlanta, Ga.

 ${}^{6}$ J. J. Gilman and W. G. Johnston, J. Appl. Phys. 29. 877 (1958), and Solid State Phys. 13, 147 (1962).

 $\rm ^7W$ . D. Compton and H. Rabin, Solid State Phys. 16. 121 (1964).

 ${}^8$ In another experiment, the neutron-retaining ability at 4'K was restored in this heated crystal after it was irradiated with gamma rays to once again restore the characteristic trapped electron color.

 ${}^{9}G$ . T. Furukawa and T. B. Douglas, American Institute of Physics Handbook (McGraw-Hill Book Company, Inc., New York, 1963), 2nd ed. , p. 4-62.

## CYCLOTRON NAVE TRANSMISSION ACROSS A PLASMA\*

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The relativistic Vlasov dispersion relation is used to find the critical density  $n_c$  $=3B_0^2\kappa T/8\pi m^2c^4$  below which the extraordinary wave can propagate freely across a mirror-field plasma into the cyclotron-resonance region.

It is well known that an electron can be accelerated by the extraordinary electromagnetic wave propagating in the direction perpendicular to a constant magnetic field. This wave has its electric-field vector in the plane of the Larmor orbit of the electron. When this single-particle model is extended to an electron in a plasma, the problem is, however, more complicated. Consider a plasma that is confined in a mirror field. If the wave frequency is equal to the electron cyclotron frequency in the interior of the plasma, the wave can in general no longer propagate freely into the resonance region. At some point before reaching the cyclotron-resonance region, the wave will be at least partially reflected in an evanescent region determined by the collective properties of the plasma, for which  $\omega_{\rm uh}$  $<\omega<\omega_{\text{uc}}$ , where  $\omega_{\text{uh}}=(\omega_p^2+\omega_c^2)^{1/2}$  is the upper hybrid frequency and  $\omega_{\text{uc}}=\omega_c/2+[(\omega_c/2)^2+\omega_p^2]^{1/2}$  is the upper hybrid cutoff frequency. The local plasma and cyclotron frequencies are  $\omega_p$  and  $\omega_c$ , respectively.

In this paper, the relativistic dispersion relation for a thermal Vlasov plasma is used to analyze the propagation of these extraordinary waves as a function of density and temperature. The plasma dispersion relation<sup>1</sup> for the waves with  $\vec{E} \perp \vec{B}_0$ ,  $\vec{k} \perp \vec{B}_0$  is

 $N^2 = \left[1-\delta(R_{10}+R_{30})\right]\left[1-\delta(R_{10}-R_{30})\right]\left[1-\delta R_{10}\right]^{-1}$  $(1)$ 

to the lowest order in temperature, where N is the index of refraction,  $\delta = \omega_p^2 / \omega^2$ ,

$$
R_{10} = \left[\alpha^2/3K_2(\alpha)\right]_0^{\infty} \cosh t \exp(-\alpha \cosh t) \sinh^4 t P(t) dt = A_1 + iS,
$$
  
\n
$$
R_{30} = \left[\alpha^2/3\nu K_2(\alpha)\right]_0^{\infty} \exp(-\alpha \cosh t) \sinh^4 t P(t) dt = A + iS,
$$
\n(2)

 $P(t) = [\cosh^2 t - \nu^{-2}]^{-1}$ ,  $\alpha = mc^2/\kappa T$ ,  $\nu = \omega/\omega_c$ , and  $K_2$  is the modified Hankel function. Note that S=0 for  $\nu \ge 1$ .

If  $\nu \neq 1$ , the cosh<sup>2</sup>t term in  $P(t)$  may be replaced by 1 for low-temperature plasmas. Thus,  $R_{10} = (1-1)/$  $v^2$ <sup>-1</sup>,  $R_{30} = v^{-1}R_{10}$  and the dispersion relation reduces to the classical zero-temperature expression. The upper hybrid frequency is found by setting  $1-\delta R_{10}=0$  and the upper hybrid cutoff by setting  $1-\delta(R_{10})$  $+R_{30}$  = 0 in Eq. (1). As the plasma density is reduced, both the cutoff and the hybrid frequencies approach the cyclotron frequency since  $N \propto \delta$ . In order to treat this limit quantitatively, however, it is no longer correct to set cosht = 1 in  $P(t)$ . The integral must instead be evaluated more accurately either by numerical integration in higher temperature plasmas, or by expanding these hyperbolic terms to the first order in  $sinh^2t$ , when the temperature is low.