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POSSIBLE OBSERVATION OF In¹¹⁵ NUCLEAR ELECTRIC HEXADECAPOLE TRANSITIONS

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This Letter reports the observation of ultrasonically induced $\Delta m = \pm 3 \ln^{115}$ nuclear spin transitions in an InAs single crystal. This was accomplished in the following manner: A $\frac{3}{8}$ -in.diameter X-cut quartz transducer, resonant at 14.926 MHz (at liquid-nitrogen temperature), was bonded on the (1, 1, 1) face of a 12-g cylindrical single crystal of InAs. The sample was then placed inside the coil of a 4.974-MHz pulsed nmr spectrometer, and this assembly was immersed in a Dewar of liquid nitrogen centered between the pole faces of an electromagnet set such that the magnetic field corresponded to a Larmor frequency of 4.974 MHz for the In¹¹⁵ nuclei. Approximately 350 V_{rms} of rf was then applied to the transducer, and the rf frequency was swept through the $\Delta m = \pm 3$ In¹¹⁵ resonance frequency while we periodically sampled the In¹¹⁵ nuclear magnetization. This technique is analogous to the observation of $\Delta m = -2$ quadrupole transitions.¹ The magnetization was displayed by recording the output of a box-car integrator system.² The result of such a recording is shown in Fig. 1. This absorption line represents a maximum decrease of 2.3%in the In¹¹⁵ nuclear magnetization.

Other possible effects which might give rise to apparent $\Delta m = \pm 3$ transitions are (1) a mechanical or electrical resonance in the sample holder of nmr coil assembly, (2) some type of magnetic transitions induced by rf leakage, (3) ultrasonically induced magnetic octupole transitions, and (4) simultaneous $\Delta m = -2$ and $\Delta m = -1$ quadrupole transitions of two nearestneighbor In¹¹⁵ nuclei.

Possible mechanical or electric resonance effects were eliminated by changing nmr coil and transducer, and shifting the magnetic field and corresponding Larmor frequency. No magnetic transitions were observed when a coil excited by the transducer amplifier was placed next to the sample and the frequency swept through the $\Delta m = \pm 3$ frequency range.

One can make a rough estimate of the magnetic octupole interaction energy by assuming that the interaction is $(r_n/a)^2$ times the magnetic dipole interaction energy, where r_n is a nuclear radius and a is a lattice constant. This indicates that the octupole interaction is down by a factor of 10^{-10} . Since we have not been able to observe phonon-induced nuclear dipole transitions in other work³ because of the weakness of the coupling, the present transitions cannot be magnetic octupole transitions.

The possibility of simultaneous flipping of two neighboring In¹¹⁵ nuclei, one by $\Delta m = -2$



FIG. 1. $\ln^{115} \Delta m = \pm 3$ nuclear absorption line.

and the other by $\Delta m = -1$, has been eliminated experimentally in the following two ways. We unsuccessfully tried to observe the simultaneous $\Delta m = -2$ flip of the In¹¹⁵ nucleus along with the $\Delta m = -1$ flip of the As⁷⁵ nucleus. We have calculated that in this case, the interaction of the $\mathrm{In^{115}}$ and $\mathrm{As^{75}}$ quadrupole moments is 10^2 times greater than the simultaneous flipping of two In¹¹⁵ nuclei. We unsuccessfully repeated the original experiment on As⁷⁵ in GaAs. Since the spin- $\frac{3}{2}$ As⁷⁵ nucleus has no hexadecapole moment, we were essentially trying to observe simultaneous $\Delta m = -2$ and $\Delta m = -1$ quadrupole transitions involving nearest As⁷⁵ nuclei. The $\Delta m = -2$ ultrasonic resonance of As⁷⁵ is extremely strong in this crystal (roughly 10 times stronger than the In¹¹⁵ resonance as measured by comparing phonon densities necessary for equivalent saturation due to $\Delta m = -2$ quadrupole transitions). We have thus ruled out simultaneous quadrupole transitions to explain our original experiment.

From a theoretical standpoint, we have calculated the following interaction energies for the processes mentioned above:

$$E_Q \approx E_H \approx 10^3 E_{QQ} \approx 10^8 E_O,$$

where E_Q , E_H , E_{QQ} , and E_O are, respectively, the quadrupole, hexadecapole, simultaneous $\Delta m = -2$ and $\Delta m = -1$ quadrupole, and octupole interaction energies. In this calculation, we have assumed that the In¹¹⁵ quadrupole charge-enhancement factor (γ_Q) is 10³, the hexadecapole charge enhancement factor⁴ is $10^2 \gamma_Q$, the In¹¹⁵ hexadecapole moment (M_{16})

is 10^{-47} (cm)⁴, and that 400 times more energy than required for the $\Delta m = -2$ quadrupole transitions was added to the lattice.

We propose that the $\Delta m = \pm 3$ transitions observed in this experiment are due to the In¹¹⁵ nuclear hexadecapole moment. The existence of the hexadecapole moment has been inferred by Wang⁵ from shifts in the pure quadrupole spectra of Sb¹²¹ and Sb¹²³. The hexadecapole interaction allows $\Delta m = \pm 1, 2, 3$, and 4 transitions for a spin- $\frac{9}{2}$ nucleus in a crystal of $\overline{4}3m$ symmetry.⁶ Thus far attempts to observe $\Delta m = \pm 4$ transitions have been unsuccessful, probably because of the fact that both the magnitude and the angular dependence of the Δm =±3 and Δm =±4 transitions are different⁶ and we observed the maximum $\Delta m = \pm 3$ saturation at a crystal orientation where the nmr signal to noise was a maximum. Both the observed $\Delta m = \pm 3$ saturation and signal-to-noise decreased upon rotation from this angle of maximum effect.

Two comments should be made about our results. First, the maximum saturation observed was only 2.6%, which is extremely small. Second, the line shift shown in Fig. 1 is due to our method of observing the saturation, namely sampling the magnetization before phononspin-system equilibrium has been reached. This shift was shown to be dependent on the phonon energy density and its direction reversed upon reversing the direction of the frequency drive.

We would like to thank Dr. W. G. Proctor for suggesting that we look for the existence of a nuclear hexadecapole moment by attempting to observe $\Delta m = -4$ nuclear spin transitions,

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and also for his continuous encouragement during the last several years. We also thank Dr. R. A. Ruehrwein of Monsanto Chemical Company for supplying the InAs crystal.

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²L. W. James, "A Sensitive System for Observing Phonon-Nuclear Interactions," National Bureau of Standards Technical Report (to be published).

³<u>Nuclear Magnetic Resonance and Relaxation in Solids</u>, Proceedings of the XIIIth Colloque Ampére (North-Hol-

land Publishing Company, Amsterdam, 1965), p. 202. ⁴R. M. Sternheimer, Phys. Rev. <u>123</u>, 870 (1961).

⁵T. Wang, Phys. Rev. <u>99</u>, 566 (1955).

⁶R. J. Mahler, "Nuclear Hexadecapole Interactions" (to be published).

MOTION OF POSITRONS*

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This Letter presents the results of an experiment¹ which was designed to observe directly whether or not positrons in a metal were thermalized. The results were surprising to us. They led to the conclusion that the positron is thermalized but has an effective mass in sodium metal approximately twice the rest mass.

Lee-Whiting² and Adler³ have calculated that a positron in a metal thermalizes, by scattering with the conduction electrons, at a rate much faster than the annihilation rate. In fact, they estimate that a positron should have an energy of order $k_{\rm B}T$ in about 10^{-12} sec whereas the mean life of a positron in a metal is about 10^{-10} sec. Ever since the early results of Stewart⁴ showed a sharp cutoff at the Fermi momentum in the momentum distribution of annihilation photons, it has been known that the positron average energy at annihilation was much less than an electron volt. With our present higher resolution apparatus, it was decided to attempt to observe directly the positron motion.

First of all we must estimate the size of the thermal broadening Δk of the Fermi surface. It is given by

$$(\hbar^2/2m)(k_{\rm F}\pm\Delta k)^2 = E_{\rm F}\pm k_{\rm B}T,$$

from which

$$\frac{\Delta k}{k_{\rm F}} = \frac{k_{\rm B}T}{2E_{\rm F}} \approx \frac{1}{200}$$

for $T = 600^{\circ}$ K. This thermal broadening is one order of magnitude beyond present detectability.

However, the usual experiment measures $k_z = (\vec{k}_+ + \vec{k}_+)_z$, the sum of both electron and positron momentum. The positron momentum k_+ , being its total momentum, is given by

$$(\hbar^2/2m)k_{\perp}^2 = \frac{3}{2}k_{\rm B}T$$

whence

$$\frac{k_{\rm H}}{k_{\rm F}} = \left(\frac{3k_{\rm B}T}{2E_{\rm F}}\right)^{1/2} \approx \frac{1}{10}$$

for $T = 600^{\circ}$ K. This is just observable at present. The effect of temperature upon positron motion should therefore be detectable. Because of the positron's random motion, the effect should be equivalent to a worsening of apparatus resolution.

Sodium was chosen as the material in which to attempt to measure positron motion for two reasons: The Fermi surface is known to be spherical and the higher momentum components therefore probably small, and secondly, even at high temperature the mean free path of electrons is large. There is, therefore, little smearing of the Fermi surface due to electron scattering.

Sodium specimens maintained at 110, 300, 400, and 600°K were bombarded with positrons and the annihilation photons detected in the usual long slit detector arrangement. The slits subtended an angle of 0.2×10^{-3} rad at the specimen. For sodium the "Fermi angle," $\theta_{\rm F}$, is 3.5×10^{-3} rad and thus the optical resolution of the apparatus is 6% of $k_{\rm F}$. The optical res-