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Authors
Quitmann, D.
Jaklevic, J.M.
Shirley, D.A.

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LAWRENCE RADIATION LABORATORY
UNIVERSITY of CALIFORNIA BERKELEY

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OBSERVATION OF NMR IN AN ISOMERIC STATE FOLLOWING A NUCLEAR REACTION*

D. Quitmann,† J. M. Jaklevic and D. A. Shirley

Lawrence Radiation Laboratory
University of California
Berkeley, California 94720

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The isomeric state in As^{73} with $I^{\pi} = 9/2^{+}$ at 426 kev was produced and aligned by the $Ga^{71}(\alpha,2n)$ reaction in a liquid Ga metal target. The resulting γ-ray anisotropy was attenuated by inducing NMR transitions with a 1 MHz rf field. We obtained $g = +1.146 \pm 0.007$ for the 426-keV state.

Nuclear magnetic resonance transitions in the 5.8-μsec, 426 keV state of As^{73} have been induced by an external rf magnetic field and observed through their effect on the angular distribution of the gamma rays emitted. The isomeric state was produced and aligned by the $Ga^{71}(\alpha,2n)$ reaction on a liquid metal target. This result extends the classical NMR experiments to isomeric states with lifetimes in the 10^{-6} - 10^{-3}sec range. Using this technique, NMR studies of a large number of nuclear states (and of elements) should be possible, providing accurate values of the magnetic moments. It could also help elucidate the dynamics of environmental effects with characteristic times in the 10^{-6} - 10^{-3}sec range, and serve to determine hyperfine constants and Knight shifts for solute atoms.

The radiative detection of magnetic resonance in nuclei (NMR/RD) began with the positronium experiments of Deutsch and Brown.[1] Bloembergen and Temmer suggested combining magnetic resonance with nuclear orientation (NMR/ON),[2] and Abragam and Pound proposed the use of perturbed angular correlations (NMR/PAC).[3] These two methods as well as the alignment by nuclear reactions, have attracted increasing interest.[4] From the standpoint of the applicability of NMR to metastable nuclear states in condensed matter, the present experimental situation can be summarized as follows. States with lifetimes longer than 10^3 sec are generally accessible to NMR/ON, usually employing lattices of ferromagnetic metals. For states with lifetimes of the order of 10^{-6} sec, NMR/PAC may be applicable, again using ferromagnetic hosts. However, since neither method is generally feasible for states in the $10^{-6} - 10^{+3}$ sec range, nuclear reactions must be used to provide the oriented nuclei. Several experiments have been reported on light beta - emitting nuclei with lifetimes in the upper half of this range (i.e. 10^{-2} sec to 10^2 sec).[5] The purpose of the present work was to apply the NMR/RD method to one of the many γ-decaying isomeric states in the $10^{-6} - 10^{-3}$ sec range.

Preliminary measurements were made on several targets using (α,xn) reactions to produce the aligned isomers. In order to decide whether a NMR/RD experiment would be feasible, two criteria were employed: (1) with no external fields a time-integral γ-ray anisotropy of magnitude comparable to the one predicted for the expected alignment must be exhibited, and (2) this anisotropy must disappear when a (small) dc magnetic field is applied perpendicular to the beam-detector plane.

Metallic targets are attractive for such experiments, since their spin-lattice relaxation times (T_1) are sufficiently long at room temperature. When employing nuclear reactions, the alignment may still be lost during the stopping of the recoil nucleus or through quadrupole interactions. Liquid metals, however, reduce these effects while still having[6] relaxation times of the order 10^{-4} to 10^{-3}sec. Liquid gallium metal was recently used by Christiansen et al. in their stroboscopic measurement of the magnetic moment of the 5-μsec isomer of Ge^{69}.[7] Similarly our experiments showed that As^{73} in liquid gallium would be a suitable system to attempt NMR/RD.

Irradiation with 30-MeV α particles produces the 426-keV, $I^{\pi} = 9/2^{+}$ isomer of As^{73} from $Ga^{71}(I^{\pi} = 3/2^{-})$ via the (α,2n) reaction. A 360-keV(M2) and a 66-keV(M1) γ ray are emitted in the $(9/2^{+}) \rightarrow (5/2^{-}) \rightarrow (3/2^{-})$ cascade depopulating this state. The angular distribution will be described by the expansion

$$W(\theta) = 1 + \Sigma\ A_{\nu}P_{\nu}(\cos\theta), \quad \nu = 2,4 \ . \qquad (1)$$

If one assumes the highest alignment possible in this experiment for the $9/2^{+}$ state (population of the $m = \pm 1/2, \pm 3/2$ substates only, and equal distribution among these), and furthermore transitions of pure multipolarity (see[8]), then: $A_2(360) = +0.417$; $A_4(360) = -0.167$ $A_2(66) = -0.292$; $A_4(66) = 0$. Earlier measurements[9] had given $A_2(360) = +0.34\pm0.04$; $A_4(360) = -0.05\pm0.05$; $A_2(66) = -0.20\pm0.05$, while $A_4(66)$ can be assumed to be small. Therefore, only the P_2 terms will be considered in the following analysis.

The experimental geometry can be described by the relative orientations of four vectors; i.e. the beam direction $\vec{k}_1$, the propagation direction of the observed γ ray $\vec{k}_2$, and the directions of the two magnetic fields acting at the nucleus: the dc field $\vec{H}_0$ and the rf field $\vec{H}_1'(t) = 2\vec{H}_1\cos\omega t$. The geometry employed is illustrated in the insert in Fig. 1. Taking the z-direction along the beam, $\vec{H}_0$ was also along z, $\vec{H}_1$ along ± x, and the two detectors were in the $z(\theta = 0^{\circ})$ and $y(\theta = 90^{\circ})$ directions. The measurements were essentially observations of the time-integral correlations, where the normalized γ-ray intensities are[10]

$$W(\theta) = 1 + A_2\overline{G_2}P_2(\cos\theta) \qquad (2)$$

with

$$\overline{G_2} = (1/4)\ (3\cos^2\beta-1)^2 + 3\sin^2\beta\cos^2\beta/(1+\omega_e\tau)^2) + (3/4)\sin^4\beta/(1+(2\,\omega_e\tau)^2)\quad , \qquad (3)$$

$$\tan\beta = H_1\ /\ (H_0 - \hbar\omega/g\mu_n) \quad . \qquad (4)$$

$$\hbar\omega_e = \mu_n g\ \sqrt{(H_0 - \hbar\omega/g\mu_n)^2 + H_1^{\ 2})} \quad . \qquad (5)$$

The g-factor has been measured earlier[9] as $g = +1.1\pm0.2$, based on[8] $T_{1/2} = \tau\cdot\ln 2 + (5.8\pm0.5)\cdot10^{-6}$ s. To produce a sizeable decrease of $\overline{G_2}$ the quantity $\omega_1\tau = g\ \mu_n H_1\tau/\hbar$ must be of order unity or greater. In this experiment we reached $\omega_1\tau = 0.7$ with the available rf field strength, which was measured <u>in situ</u> with a test coil as $2H_1 = (31\pm3)$ gauss. A pair of coils provided a dc magnetic field up to 1500 gauss, with homogeneity and stability of about one part in 10^3.

The target was liquid Ga^{71} metal, 0.15 mm thick and 3 mm wide, confined between two thin quartz plates. The skin depth a 1 MHz is 0.16 mm. The experiments were performed by stepping H_0, with $H_1'(t)$ held at constant amplitude and frequency (stable to ±0.3 kHz). Of the pulses from two Ge(Li) detectors (1.5 keV FWHM at 122 keV) only those "delayed" events were accepted which occurredin a 0.14 μsec interval between beam bursts (0.16 μsec separation). The timing circuitry was similar to that described elsewhere.[11,12] Four single-channel analyzers selected the 66-keV and 360-keV lines. The ratio of peak to background was about 10:1. The output pulses were registered in a 400-channel analyzer modified to operate as four 100-channel multiscalers which were stepped synchronously with H_0. Data were taken at two frequencies and four values of H_1, sweeping H_0 by between ±30% and ±10%. All runs were consistent with the conclusions discussed below. The data are presented as ratios of counts, e.g. N(66kev, 0°) / N(360 kev, 0°), to correct for beam fluctuations. Figure 1 shows representative curves.

Results from four measurements are listed in Table 1. Each measurement yielded three independent ratios at each of the values of H_0, with H_1 and ω constant. A single seven-parameter least-squares fit was made to the combined three sets of ratios from each measurement, using the appropriate functions derived from Eqs. (2)-(5). The fits were satisfactory in each case. We note the following:

1) The results for the g-factor agree within ±0.3%. This spread is probably due mainly to the error in the field determinations which was independently estimated as ±0.5%. Including the Knight shift constant K explicitly, the weighted average of our results is $g\cdot(1 + K) = 1.1495\pm0.0057$. For As in liquid As, K = +0.00318.[13] Since we estimated that As in Ga has only a slightly different Knight shift, we use as a correction $K = (3.2\pm1.0)\cdot10^{-3}$. Thus

$$g = 1.146\pm0.007 \qquad (6)$$

The large g-factor of +1.146 identifies the 426-keV level of As^{73} quite unambiguously as a $g_{9/2}$ proton state. The ground states in seven odd proton nuclei in the $40<Z<50$ region are also classified as $g_{9/2}$ proton levels, with g-factors ranging somewhat higher, between +1.23 and +1.37. This is the first nucleus with $Z<40$ in which this state has been identified by a magnetic moment measurement.

2) The size of the derived values of A_2 is approximately one-half that observed earlier. The neglect of $A_4(360)$ is not expected to be the reason for this.

However, the possibility of insufficient heating of the target cannot be completely ruled out. Quadrupole interactions in solid gallium (orthorhombic, melting point 29.8°C) or possibly in the liquid when near freezing would destroy the coherent motion of the nuclear spins. If one tries to describe the reduction of A_2 phenomenologically by introducing an effective lifetime τ_{eff} which is shorter than the nuclear mean lifetime $\tau = 8.37 \cdot 10^{-6}$ sec, then $\tau_{eff} \approx (6.5 \pm 0.5) \cdot 10^{-6}$ sec can explain the observed data.

3) The observed lines are distinctly non-Lorentzian. In fact the theoretical lineshapes described by Eqs. (2)-(5) have two maxima and resemble two unresolved Lorentzians split by $\sqrt{2}\ H_1$ when $\omega_1 \tau \gtrsim 1$ (saturation).[10] This shape is expected for experiments in which the observed quantity transforms as a second-rank tensor,[14] but to our knowledge it has not been previously demonstrated in a nuclear magnetic resonance experiment.

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FOOTNOTES AND REFERENCES

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† On leave of absence from Institut für Technische Kernphysik, Technische Hochschule, Darmstadt.

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FIGURE CAPTION

Fig. 1. Effect of NMR on the intensity ratios for the two γ-rays which depopulate the As^{73} isomer ($9/2^{+}$, 426 keV) produced by $Ga^{71}(\alpha,2n)$. The ordinates at the left give the observed ratios. The scales at the right show the change in each ratio ($\varepsilon = 1$ corresponds to $\overline{G_2} = 0$); to within a few percent, $\varepsilon = 1 - \overline{G_2}$ holds. Given with each curve are the rf frequency $\omega/2\pi$, the field strength in the rotating frame H_1, and the identification of the ratio. H_0 is the applied dc field. Statistical errors are shown. The curves drawn are from the least-squares fits. The insert shows the relevant vectors.

Table 1. Summary of results. The errors given parenthetically are statistical errors only

Experimental conditions		Relative effect [a]			Results from fits				
$\omega/2\pi$ MHz	H_1 gauss	$\frac{N(66,0)}{N(360,0)}$	$\frac{N(66,90)}{N(360,90)}$	$\frac{N(66,0)}{N(66,90)}$	$-A_2(66)$ b)	$+A_2(360)$ b)	χ^2	g-factor	
0.9252	14.6(7)	1.123(9) [e]	0.933(7)	1.084(5)	0.136(7)	0.197(14)	0.95	1.1520(10)	c)
0.8535	17.0(7)	1.139(12)	0.941(12)	1.094(6) [e]	0.146(5)	0.193(13)	1.23	1.1467(10)	c)
0.8562	12.4	1.074(15)	0.932(15)	1.050(10) [e]	0.114(8)	0.174(17)	1.19	1.1496(20)	d)
0.8581	8.5	1.056(20)	0.968(20)	1.032(15) [e]	0.126(12)	0.184(22)	0.90	1.1531(30)	d)

a) Ratio (on resonance, i.e., averaged over about ±25 gauss)/ratio (off resonance).

b) The four corrections for background in the energy windows were applied in the fits.

c) The value of H_1 is from the least-squares fit. The H_1 field had been increased for the second measurement by about 10%.

d) The relative values of H_1 had been directly measured for the last three entries. Therefore, H_1 was fixed correspondingly in the last two fits. Also, the ratio $A_2(66)/A_2(360)$ was restricted to be -0.7±0.1.

e) Shown in Fig. 1.

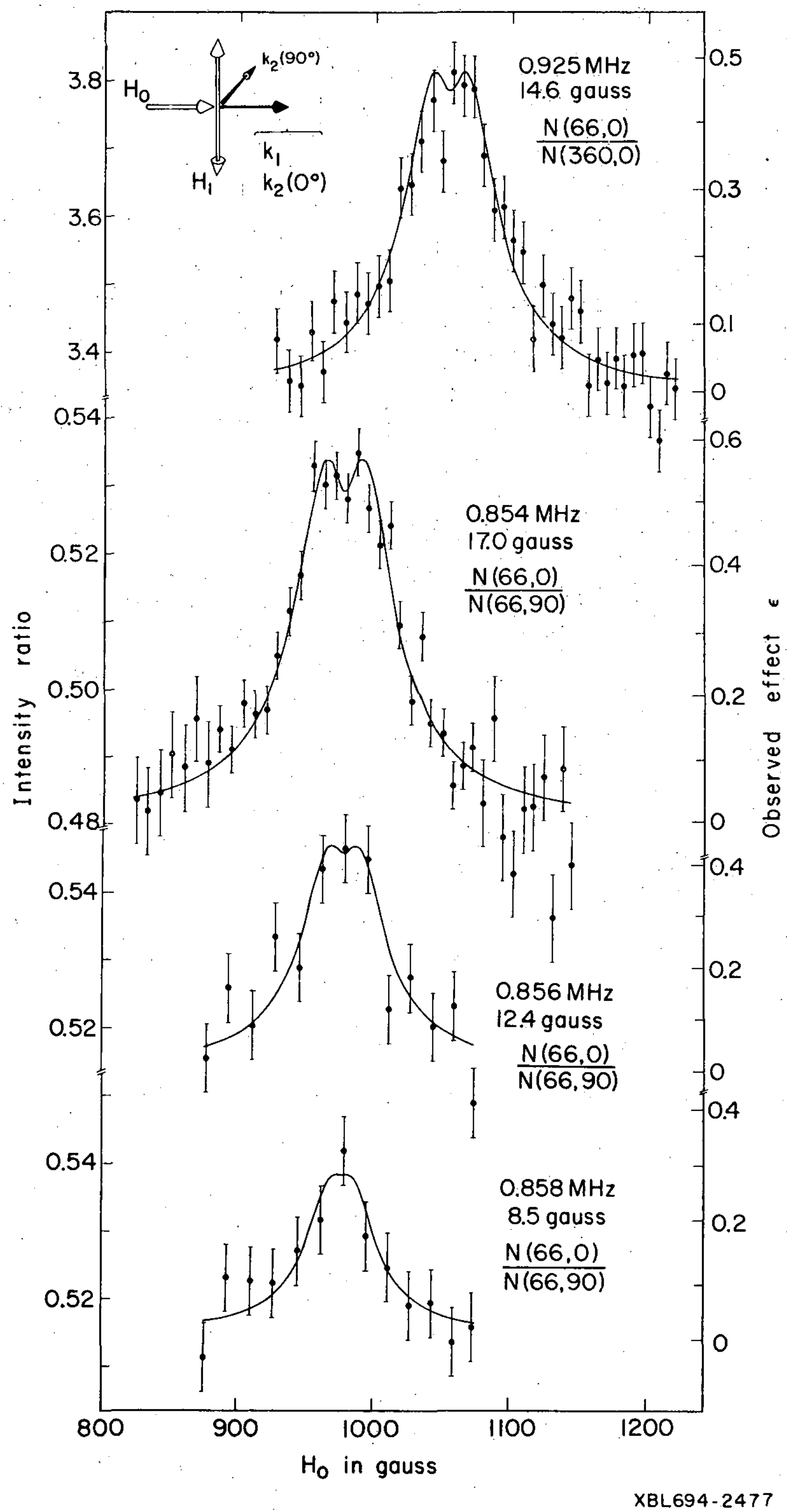
H0
H1
k2(90°)
k1
k2(0°)
0.925 MHz
14.6 gauss
N(66,0)/N(360,0)
0.854 MHz
17.0 gauss
N(66,0)/N(66,90)
0.856 MHz
12.4 gauss
N(66,0)/N(66,90)
0.858 MHz
8.5 gauss
N(66,0)/N(66,90)
Intensity ratio
Observed effect ε
H0 in gauss
XBL694-2477

Fig. 1.

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LAWRENCE RADIATION LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720