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Measurements of the nuclear spin of ^{206}Tl , the hyperfine structure splitting of ^{66}Cu , and the nuclear magnetic moments of ^{108}Ag (2.3 min) and ^{110}Ag (24 s)

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Abstract. By using atomic beam magnetic resonance techniques it has been shown that the nuclear spin of ^{206}Tl (half-life 4.2 min) is either zero, or else the nuclear magnetic moment $\mu_I < 10^{-5}$ nuclear magnetons. Measurements on isotopes of copper and silver have yielded the following values for their nuclear spins I and hyperfine interaction constants A , from which their nuclear magnetic moments have been deduced using the Fermi–Segré relation. No diamagnetic correction has been applied.

Isotope	Half-life	I	A (MHz)	μ_I (n.m.)
^{66}Cu	5.3 min	1	-1112.525(4)	-0.282(2)
^{108}Ag	2.3 min	1	+20800(150)	+2.79(3)
^{110}Ag	24 s	1	+21200(300)	+2.84(5)

1. Introduction

The atomic beam magnetic resonance apparatus used in these experiments has already been described by Rochester and Smith (1964 a). Radioactive isotopes formed from stable elements by slow neutron capture in a reactor were transferred rapidly from the pile directly into a hot oven inside the vacuum envelope of the beam machine, thereby allowing the production of an atomic beam less than 30 seconds after irradiation had finished. The oven, which was maintained at temperatures of up to 1800 °K by electron bombardment, consisted of a central molybdenum rod containing a crucible and an outer coaxial molybdenum tube separated from the rod by a carbon sleeve. The hole through which the sample dropped into the crucible could be replaced by a slit by rotating the outer tube. After leaving the oven slit and travelling vertically through a conventional arrangement of focusing and transition fields, refocused atoms adhered to the surface of a thin cold aluminium disk which could be rotated in a horizontal plane and accurately positioned. A rapid 180° rotation of the disk brought radioactive atoms, deposited near the perimeter, close to a solid state β detector placed well away from the axis of the beam. Radio-frequency power, which was fed to the transition field region, and two scalers were switched synchronously with the disk, so that the total count on one scaler represented the signal obtained at one radio-frequency and the total count on the other scaler that obtained at a different radio-frequency.

2. Thallium 206

For the ^{206}Tl experiments the outer molybdenum tube of the oven was fitted with a short carbon snout, with a slit machined in the end. This modification was introduced because thallium alloys with molybdenum but does not attack carbon. The sample temperature during irradiation was kept below 577 °K, the melting point of thallium metal, by circulating nitrogen initially at atmospheric temperature through the sample transfer pipes.

The radio-frequency and counter systems were arranged so that counts recorded on one scaler L always corresponded to the application of the same radio-frequency ν_L to the transition field region, while the counts on the other scaler R corresponded to the application of a variety of different radio-frequencies ν_R . The number of counts C_L obtained from

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each sample thus served to monitor the relative intensities of the active thallium beams obtained after each irradiation. Uncertainties due to background were minimized by choosing for ν_L the frequency for which the transition probability was expected to be a maximum.

Elementary shell model considerations suggest that the spin of ^{206}Tl should be 0. The frequency of the refocused transition in an atom in the $J = \frac{1}{2}$ state with thallium's g_J of $-0.665\,822$ Bohr magnetons (Berman 1952) and $I = 0$ in a field of 2 gauss would be 1.865 MHz, and this was therefore the frequency chosen for ν_L in this field. Figure 1 shows a plot of the function

$$\gamma = \frac{C_R - B}{C_L - B} \quad (1)$$

for different values of ν_R near 1.865 MHz. B is the estimated number of background counts registered in the time it took to accumulate C_L and C_R . Background counts here include counts derived from any source other than the ^{206}Tl beam. A small error is introduced by calculating B from measurements made before and after, rather than during, a run because the background level was affected to a minor degree by the nearby storage of the carriers used to take the thallium to and from the reactor. However, there is first-order cancellation of this effect because it will appear equally in both the numerator and denominator of γ .

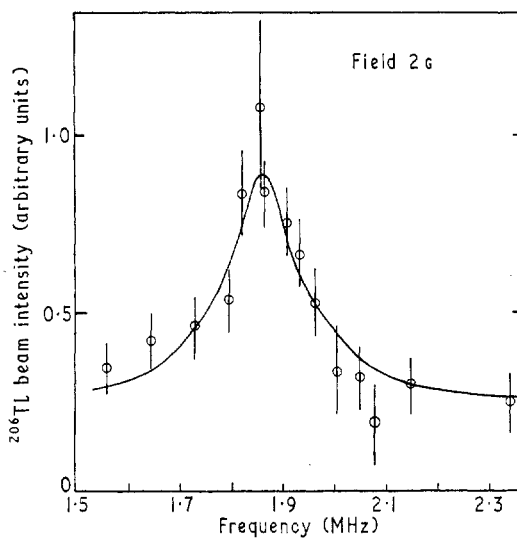


Figure 1. Resonance observed with ^{206}Tl .

It was found that with a typical background of 8 counts per minute the optimum signal-to-noise level was obtained if the beam and detector were run for twenty minutes, or 4.7 half-lives, after the arrival of each freshly irradiated thallium ball. Owing to the low beam intensity it was necessary to add together the total number of counts obtained from three or four thallium balls for each value of ν_R in order to achieve the statistical accuracy shown in figure 1. The error bars associated with each γ are given by

$$S = \frac{C_R - B}{C_L - B} \left\{ \frac{C_L + B(T/t)}{(C_L - B)^2} + \frac{C_R + B(T/t)}{(C_R - B)^2} - \frac{2B(T/t)}{(C_L - B)(C_R - B)} \right\}^{1/2} \quad (2)$$

where t is the time for which the background was counted and T is the time taken to accumulate C_L and C_R .

The full curve in figure 1 is a resonance line of the theoretical shape, but of variable height and width, which has been fitted to the experimental points by the method of least squares. It will be seen that the radio-frequency corresponding to the peak of this curve

is very close to that expected if $I = 0$. Further evidence in favour of this spin assignment lies in the fact that the maximum signal intensity on resonance was approximately half that which could be obtained from the full beam with the deflecting magnets unenergized. The observed resonance could not belong to the $J = \frac{3}{2}$ state because the population of this state at the oven temperature used is a factor of 100 lower than that of the $J = \frac{1}{2}$ state, and in any case the g_J value of the levels involved in the observed transition was confirmed by measuring the transition frequency in a field of 30 gauss as well as in a field of 2 gauss. The linear dependence of frequency on field also rules out the possibility that the refocused signal observed in 2 gauss was due to a $\Delta F = 1$ transition.

If the hyperfine structure splitting were large and $I = 1$ or more, the observable resonance frequencies in a field of 2 gauss would be at 0.622 MHz or less, which rules out this interpretation of the experimental result. However, there is a possibility that the hyperfine interaction constant A in ^{206}Tl may be so small that the nuclear spin and electronic angular momentum are already fully decoupled even in a field as low as 2 gauss. In this case we can follow the argument of Cohen *et al.* (1959), who suggested that such a single line observed to have a width α may be composed of the $2I+1$ superimposed lines $\Delta m_J = \pm 1$, $\Delta m_J = 0$, with $A \leq \alpha(2I+1)^{-1}$. Hence, for $I \neq 0$, $A_{206} < 163$ kHz and the associated moment $\mu_{206} < 10^{-5}$ n.m. This follows since $\mu_{203} = 1.61169$ n.m. (Berman 1952), $I_{203} = \frac{1}{2}$ and $A_{203} = 21\,105.4$ MHz (Lurio and Prodel 1956), and

$$\frac{\mu_{206}}{\mu_{203}} = \frac{A_{206}}{A_{203}} \frac{I_{206}}{I_{203}} \quad (3)$$

if the hyperfine anomaly is neglected.

Both $^{205}_{81}\text{Tl}$ and $^{207}_{82}\text{Pb}$ have nuclear spins of $\frac{1}{2}$, so the most likely configuration for $^{206}_{81}\text{Tl}$, based on the elementary shell model, is $(p_{1/2})^n(s_{1/2})^p$. Application of the jj coupling rules of Ames *et al.* (1961) yields an expected value of zero for the nuclear spin of ^{206}Tl , in agreement with the experimental result. This value is also consistent with the simple allowed β spectrum found by Alburger and Friedlander (1951), and the conclusions of Howe and Langer (1961).

One can obtain an empirical estimate of the magnetic moment of ^{206}Tl with $I = 1$ by coupling values of μ_p and μ_n taken from the neighbouring nuclei ^{205}Tl and ^{207}Pb respectively. Since μ_p and μ_n here are both of the order of one nuclear magneton, it is extremely unlikely that a chance cancellation can take place that would give rise to a moment as low as 10^{-5} n.m. This provides good grounds for excluding a spin of unity.

3. Copper 66

Preliminary results for the hyperfine interaction constant A and the nuclear magnetic moment of ^{66}Cu , derived from the observation of some $\Delta F = 0$ transitions, have been reported by Rochester and Smith (1964 b). The frequencies of several $\Delta F = 1$ transitions in this isotope have now been measured, enabling the sign of A to be established and also its magnitude to a far higher degree of accuracy than previously. The frequencies of the observed transitions are given in table 1, from which it will be seen that the ultimate error in the value for A is governed almost entirely by the accuracy to which the frequency of the first-order field-independent transitions

$$(F = \frac{1}{2}, m_F = \frac{1}{2} \leftrightarrow F = \frac{3}{2}, m_F = -\frac{1}{2}) \text{ and } (F = \frac{1}{2}, m_F = -\frac{1}{2} \leftrightarrow F = \frac{3}{2}, m_F = \frac{1}{2})$$

could be determined. The separation of this doublet is 400 Hz in a field of 1 gauss, and the signal obtained as a function of frequency using separated (Ramsey) radio-frequency loops in this field is shown in figure 2, where a curve embodying the linewidths and separation characteristic of the loop system employed has been superimposed on the experimental points. Also given in table 1 are the differences between the measured frequencies and those calculated for the two signs of g_I from the A obtained by least-squares fitting to the data, using the g_I value of $-2.002\,28$ Bohr magnetons given by Dodsworth and Shugart (1966). It will be noted that the best fit occurs when g_I is negative and hence A also is

Table 1. Transitions observed in ^{66}Cu

Field (G)	Frequency (MHz)					Residual frequency (MHz)					
		F_1	m_{F1}	F_2	m_{F2}	$g_I < 0$	$g_I > 0$	F_1	m_{F1}	F_2	m_{F2}
5.000(37)	4.670(50)	—	—	—	—	-28	-26	—	—	—	—
15.000(37)	14.230(30)	—	—	—	—	-22	18	—	—	—	—
30.000(36)	28.991(20)	—	—	—	—	5	13	—	—	—	—
60.000(35)	59.974(30)	—	—	—	—	29	45	—	—	—	—
120.000(33)	128.095(30)	—	—	—	—	-2	27	—	—	—	—
200.000(30)	232.524(44)	—	—	—	—	-34	9	—	—	—	—
350.000(25)	470.875(36)	—	—	—	—	-22	32	—	—	—	—
2.000(37)	1672.605(80)	—	—	—	—	77	84	—	—	—	—
1.000(37)	1668.782(45)	+	+	+	+	-7	0	+	+	+	+
1.000(37)	1668.788(12)	+	+	+	+	-1	6	+	+	+	+
490.000(20)	2131.080(60)	—	—	—	—	13	-112	—	—	—	—
490.000(20)	2131.250(60)	—	—	—	—	-27	-152	—	—	—	—

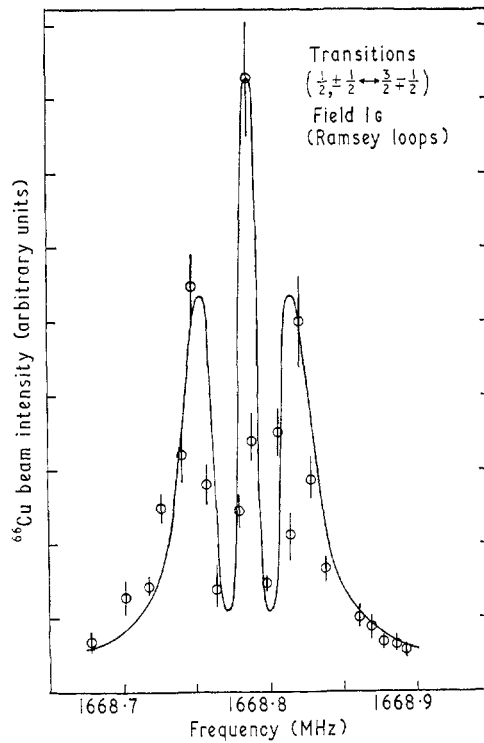


Figure 2. The $\Delta F = 1$ resonance observed with ^{66}Cu .

negative, since according to the Fermi-Segré relation A must have the same sign as g_I for a $J = \frac{1}{2}$ state.

The sign of A was in addition determined by a method previously employed by Dodsworth and Shugart (1966). For both signs of A the doublet component $P(\frac{3}{2}, \frac{1}{2} \leftrightarrow \frac{1}{2}, -\frac{1}{2})$ appears at a lower frequency than the other component $Q(\frac{3}{2}, -\frac{1}{2} \leftrightarrow \frac{1}{2}, \frac{1}{2})$. As the magnetic field at the atom is increased towards the Paschen-Back region in which m_I and m_J become good quantum numbers, the component Q becomes forbidden if $A > 0$ since this transition can only occur if $\Delta m_I = 2$, which is not allowed by the selection rules. Thus, as the magnetic field at the atom increases, the amplitude of the lower-frequency component of the doublet either increases or decreases with respect to the other component, depending

on whether A is positive or negative respectively. The choice of the magnetic field in which to examine the doublet was governed by the need to provide adequate resolution between the two components, whilst retaining a sufficiently high transition probability for the smaller resonance to remain visible. The field was set at 490 gauss, which was calculated to produce a difference of 210 kHz between the lines of the doublet. Rather a high radio-frequency field was required to produce an observable transition in the resonance line which had the lower transition probability, but, even if this field broadened the other line to such an extent that the two were no longer resolved, the doublet was at least expected to show an asymmetry which would indicate the sign of A . The experimental points obtained are shown in figure 3, together with theoretical curves which have widths and centre frequencies determined by the apparatus and maxima adjusted to fit the experimental points. The absence of a peak on the high-frequency side provides further evidence for the negative sign of A .

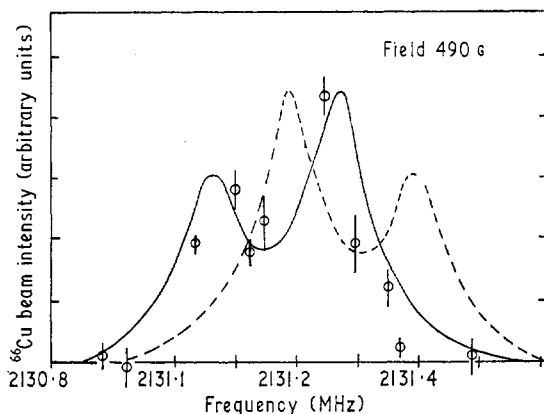


Figure 3. Superposition of theoretical curves for $A < 0$ (full curve) and $A > 0$ (broken curve) and experimental points with ^{66}Cu at 490 G.

By using equation (3) with $A_{65} = 6284.405$ MHz (Ting and Lew 1957), $I_{65} = \frac{3}{2}$, $\mu_{65} = 2.385$ n.m. (Fuller and Cohen 1969) and the computed value for

$$A_{66} = -1112.525(4) \text{ MHz}$$

it is found that $\mu_{66} = -0.282(2)$ n.m. The error in μ_{66} given here arises from the estimated order of magnitude of the hyperfine structure anomaly. The agreement between the experimental result and various theoretical estimates of μ_{66} has already been discussed (Rochester and Smith 1964 b, Kisslinger and Sorensen 1963, Phillips *et al.* 1967).

4. Silver 108

Single, double and triple $\Delta F = 0$ transitions have been observed in ^{108}Ag , half-life 2.3 min, in magnetic fields up to 500 gauss at the frequencies given in table 2. At 300 gauss the linewidths of the single, double and triple quantum transitions were in the ratio 0.33 : 0.15 : 0.06 MHz, approximately in agreement with theory, and the corresponding intensities were in the ratio 3.3 : 23.0 : 5.3. Theoretically, the transition ($\frac{3}{2}, -\frac{1}{2} \leftrightarrow \frac{3}{2}, -\frac{3}{2}$) is not refocused unless the A and B deflecting magnetic fields are large enough for the effective magnetic moment dE/dH of the level ($\frac{3}{2}, -\frac{1}{2}$) to be equal and opposite to that of the level ($\frac{3}{2}, -\frac{3}{2}$). One finds that because of the large value of the hyperfine structure splitting of ^{108}Ag ,

$$(dE/dH)_{3/2, -1/2} = 1.17 \text{ MHz G}^{-1}$$

and

$$(dE/dH)_{3/2, -3/2} = -1.50 \text{ MHz G}^{-1}$$

in a field of 20 000 gauss. This difference accounts for the observed lower amplitude of the single-quantum transition.

If the value of $g_J = -2.00233$ Bohr magnetons for silver, given by Hayne and Robinson (1960), is used, the closest fit to the experimental frequencies given in table 2 is found to be obtained with $A_{108} = 20\,800(150)$ MHz and $g_{108} = +3.0(3)$ n.m. By employing equation (3) with $A_{107} = -1712.56(4)$ MHz (Wessel and Lew 1953), $I_{107} = \frac{1}{2}$ and

$$\mu_{107} = -0.11358(3) \text{ n.m.}$$

(Fuller and Cohen 1969), it is found that

$$\mu_{108} = +2.79(2) \text{ n.m.}$$

The most likely nuclear configurations for ^{108}Ag have already been discussed by Rochester and Smith (1964 b). It will be noted that the agreement between μ (empirical) and μ (experimental) is now very close for configurations $p(g_{9/2}^{-3/2})_{7/2}$, $n(d_{5/2}^{5/2})$ and $p(g_{9/2})$, $n(g_{7/2})$.

Table 2. The field dependence of the transitions $(\frac{3}{2}, m \leftrightarrow \frac{3}{2}, -\frac{3}{2})$ observed in ^{108}Ag

Field (G)	Transition m	Observed frequency (MHz)	Calculated frequency (MHz)
5.00(5)	$\frac{1}{2}$	9.40(5)	9.32
15.00(5)	$\frac{1}{2}$	28.05(5)	28.00
30.00(5)	$\frac{1}{2}$	56.06(5)	56.02
60.00(4)	$\frac{1}{2}$	112.18(8)	112.14
120.00(4)	$\frac{1}{2}$	224.72(5)	224.70
150.00(4)	$\frac{1}{2}$	281.11(5)	281.10
300.00(3)	$-\frac{1}{2}$	284.95(8)	284.96
300.00(3)	$\frac{1}{2}$	564.69(5)	564.65
300.00(3)	$\frac{3}{2}$	839.63(7)	839.69
510.00(5)	$\frac{1}{2}$	490.53(9)	490.53

The frequencies were calculated assuming $A_{108} = 20\,800$ MHz.

5. Silver 110

The activity observed when a beam of irradiated natural silver was focused on the detector suggested that the beam contained ^{110}Ag , with a half-life of 24 s, as well as ^{108}Ag , and that the two isotopes have the same spin and similar magnetic moments. A resonance thought to be due to ^{110}Ag was resolved in the following way. Silver balls were irradiated for 1 min and transferred to the oven. The beam was collected for 2 min on the target and then turned off. The counts were recorded separately for the 2 min collection period and for the following 10 min. In addition, the corresponding counts were recorded when the balls were delayed for 2 min between the reactor and the oven, thereby allowing the ^{110}Ag activity to decay before the beam was produced. By using the ^{108}Ag counts obtained from the 10 min periods to normalize the ^{108}Ag count expected in the 2 min period with undelayed balls, the counts due to ^{110}Ag were obtained. A plot of effective ^{110}Ag beam intensity as a function of frequency is shown in figure 4, where the error bars indicate uncertainties due to the estimation of ^{108}Ag activities. The frequencies of $(\frac{3}{2}, \frac{1}{2} \leftrightarrow \frac{3}{2}, -\frac{3}{2})$ transitions in three different fields are given in table 3 together with frequencies calculated assuming that $I_{110} = 1$, $A_{110} = 21\,200(300)$ MHz and $\mu_{110} = 2.84(4)$ n.m. No signal was obtained at frequencies corresponding to $I = 2$. The signals could not be observed at higher fields because of the large radio-frequency power required to induce the double-quantum transitions. The refocusing condition could not be satisfied with single-quantum transitions because the large A value prevented complete decoupling in the deflecting fields. The ^{110}Ag and ^{108}Ag resonances were separated by less than a linewidth at the highest field used.

According to Dzelopov and Zhukovskii (1963), the spin of ^{110}Ag must be 1 if sense is to be made of the β and γ spectra, although this leads to a contradiction which is difficult to explain on the basis of existing experimental data. The reader who is interested is

referred to their discussion of this point. The most probable shell model configurations for ^{110}Ag are thus the same as those for ^{108}Ag , so it is not surprising that the nuclear moments of these two isotopes should be very nearly the same.

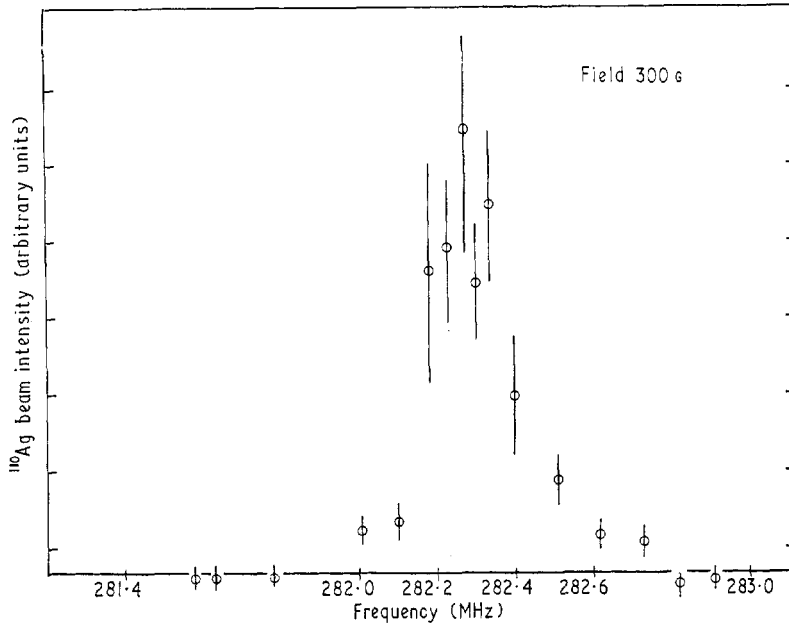


Figure 4. Resonance observed with ^{110}Ag at 300 g.

Table 3. The field dependence of the transition ($\frac{3}{2}, \frac{1}{2} \leftrightarrow \frac{3}{2}, -\frac{3}{2}$) in ^{110}Ag

Field (g)	Observed frequency (MHz)	Calculated frequency (MHz)
30.00(5)	56.04(8)	56.02
60.00(4)	112.16(8)	112.18
300.00(4)	564.56(9)	564.56

The frequencies were calculated assuming $A_{110} = 21\,200$ MHz.

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