Nuclear-Spin, Hyperfine-Structure, and Magnetic-Moment Investigations on 61 Cu, 62 Cu, and 64 Cu[†]

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Atomic-beam magnetic-resonance experiments on the ${}^{2}S_{1/2}$ ground state of three radioactive copper isotopes, ⁶¹Cu, ⁶²Cu, and ⁶⁴Cu, are described. The results are summarized in the following table. The spins of ⁶¹Cu and of ⁶⁴Cu had been measured previously but are included in parentheses for completeness.

Isotope	${T}_{1/2}$	Spin	Hyperfine structure (Mc/sec)	Magnetic moment (nm)
⁶¹ Cu ⁶² Cu	3.3 h 9.9 min	(<u>3</u>) 1	$\Delta\nu(2\rightarrow1)=+11225(200)$	$\mu_{\text{uncorr.}} = +2.13(4)$
^{64}Cu $g_J(Cu) =$	12.8 h -2.00228(2)	(1)	$\Delta \nu \left(\frac{3}{2} \to \frac{1}{2}\right) = -1282.140(8)$	$\mu_{\text{uncorr.}} = -0.216(2)$

The nuclear magnetic moments are calculated from the Fermi-Segrè formula by using known constants of ⁶⁸Cu and ⁶⁵Cu. A 1% error is quoted on the ⁶⁴Cu moment to bracket a possible hyperfine-structure anomaly.

HE radioactive isotopes of copper have been the subject of investigation in this laboratory over a number of years. Previously, preliminary reports of this work have appeared only in abstract form, with author credit being given to the various individuals who helped with the several experiments.¹⁻⁵ In this paper a general review and description of all of the work is given. As background to the results quoted here, a short history of previous work should be given. The spin of ⁶¹Cu (Ref. 1) was found to be $I = \frac{3}{2}$, and a preliminary hyperfine-structure separation $\Delta \nu = +11\ 200(400)\ Mc/$ sec was announced in Ref. 2. For 9.9-min ⁶²Cu the spin value I=1 was reported in Ref. 3. Before work began in this laboratory on ⁶⁴Cu, the spin (I=1) and hyperfine structure $\Delta \nu = \pm 1278(20)$ Mc/sec were known.⁶ Also Stroke⁷ and co-workers at Princeton, in unpublished work, had improved the $\Delta \nu$ to a precision of ± 0.7 Mc/sec; however, the sign of the magnetic moment was still undetermined. The electronic g_{J} factor for the ${}^{2}S_{1/2}$ state of copper had been measured by Ting and Lew⁸ as -2.0025(10). The ⁶⁴Cu constants described here were previously reported in abstract form.⁵

THEORY OF THE EXPERIMENTS

The energy Hamiltonian describing a free Cu atom in the ${}^{2}S_{1/2}$ electronic ground state is

$$\mathcal{K} = ha\mathbf{I} \cdot \mathbf{J} - g_J \mu_0 \mathbf{J} \cdot \mathbf{H} - g_I \mu_0 \mathbf{I} \cdot \mathbf{H}, \qquad (1)$$

where a is the hyperfine-structure dipole-interaction constant, I is the nuclear spin, J is the electronic spin, the g factors are given by $g_J = \mu_J / J \mu_0$, $g_I = \mu_I / I \mu_0$, μ_0 is the magnitude of the Bohr magneton, and H is the externally applied magnetic field. For copper, for which $J=\frac{1}{2}$, the energy levels of this Hamiltonian are given by the Breit-Rabi formula,9

$$V(F,m_F) = \frac{-h\Delta\nu}{2(2I+1)} - g_I \mu_0 m_F H + (F-I)h\Delta\nu \left(1 + \frac{4m_F x}{2I+1} + x^2\right)^{1/2}, \quad (2)$$

in which $h\Delta\nu$ is the zero-field hyperfine-structure separation between the states $F=I+\frac{1}{2}$ and $F=I-\frac{1}{2}$, and $x = (g_I - g_J) \mu_0 H / h \Delta \nu$. The $\Delta \nu$ is related to the interaction constant a by

$$\Delta \nu = a(I + \frac{1}{2}). \tag{3}$$

In an atomic beam flop-in apparatus, the "standard transition" $(F=I+\frac{1}{2}, m_F=-I+\frac{1}{2} \leftrightarrow F=I+\frac{1}{2}, m_F$ $=-I-\frac{1}{2}$ is frequently used for calibration purposes with stable alkali isotopes which can be detected by surface ionization on a hot tungsten wire. This transition is also used to determine the nuclear spin and preliminary values of $\Delta \nu$ in the unknown isotope, since the low-field frequency is given by

$$\nu \approx \nu_0 + (2I/\Delta\nu)\nu_0^2, \qquad (4)$$

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 Phys. Soc. 2, 200 (1957); J. B. Reynolds, R. L. Christensen,
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² B. M. Dodsworth, V. J. Ehlers, W. B. Ewbank, F. R. Petersen, and H. A. Shugart, Bull. Am. Phys. Soc. 4, 353 (1959).
³ B. M. Dodsworth, V. J. Ehlers, W. B. Ewbank, F. R. Petersen, and H. A. Shugart, Bull. Am. Phys. Soc. 4, 452 (1959).
⁴ B. M. Dodsworth (Ph.D. thesis), Lawrence Radiation Laboratory Report UCRL-10780, 1963 (unpublished).
⁵ B. M. Dodsworth and H. A. Shugart, Bull. Am. Phys. Soc. 9, 451 (1964).

^{9, 451 (1964).} ⁶ A. Lemonick and F. M. Pipkin, Phys. Rev. 95, 1356 (1954).

⁷ H. H. Stroke (private communication).
⁸ Y. Ting and H. Lew, Phys. Rev. 105, 581 (1957).

⁹ G. Breit and I. I. Rabi, Phys. Rev. 38, 2082 (1931).

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Calibration isotope and frequency for ⁸⁷ Rb (Mc/sec)	H (G)	⁶¹ Cu frequency (Mc/sec)	Residual frequency (positive g_I) (kc/sec)
40.870(50)	57.39(7)	40.636(30)	+38
72.055(50)	99.85(7)	71.138(120)	-69
80.725(50)	111.46(7)	79.694(60)	+33
91.707 (30)	126.05(4)	90.351(50)	+14
111.800(30)	152.41(4)	109.749(60)	-74
130.150(150)	176.12(19)	127.506(60)	+89
179.950(100)	238.76(12)	174.779 (80)	-31
[For an incorrect negative-moment assumption	$\Delta \nu = 11 \ 225(117) \\ \Delta \nu = -12 \ 077 \ (136)$	$\mu_I = +2.13(4)$ $\mu_I = -2.29$	$\chi^2 = 1.4$ (7 points) $\chi^2 = 4.4$]

TABLE I. 61Cu data.ª

a All resonances consist of the standard flop-in transition. Calibration and comparison information is contained in middle section of Table II.

where

$$\nu_0 = -g_J \mu_0 H / h(2I+1)$$

At low enough fields, where the second term of Eq. (4)may be neglected, the spin may be ascertained from a knowledge of ν , H, g_J , and universal constants. On the other hand, at higher magnetic fields the second term provides an estimate of $\Delta \nu$. Ultimately, various parameters in the Hamiltonian are fitted to the experimental observations by least-squares techniques.

When the experimental data are not precise enough to yield accurate values of μ_I (or g_I) directly from the Hamiltonian, the magnetic moment may be calculated from the Fermi-Segrè relation

$$a_1/a_2 \approx g_{I1}/g_{I2},$$
 (5)

where 1 and 2 refer to isotopes of the same element. The hyperfine-structure anomaly is a measure of the deviation from equality in Eq. (5). For most elements the anomaly is less than 1%, so this value is taken as the limit of accuracy in the magnetic moment computations.

The main features of the apparatus used in this work are described by Hobson et al.¹⁰ Calibration techniques, collection, and normalization procedures followed closely those described by Ewbank et al.¹¹

⁶¹Cu EXPERIMENT

⁶¹Cu (3.3 h) was produced at the Berkeley 60-in. cyclotron by the ⁵⁹Co(α ,2n)⁶¹Cu reaction, with 34-MeV α particles. A chemical separation of the copper from the 4-mil cobalt target was begun shortly after removal of the target from the cyclotron. The foil was dissolved in 12 N HNO₃ with approximately 20 mg of stable copper carrier. The resulting solution was boiled to dryness and the residue redissolved in 3 N HCl, from which the copper was selectively precipitated from the cobalt by H₂S. The CuS precipitate was dissolved in a few drops of concentrated HCl and the copper metal electroplated out of the solution.

A series of resonances of the "standard transition" $(F=2, m_F=-1 \leftrightarrow F=2, m_F=-2)$ were taken at fields up to 239 G. A typical resonance is shown in Fig. 1. The radioactive beam atoms were collected on sulfur-coated buttons which were counted in sodium iodide crystal scintillation counters. A least-squares fit to all the data was performed, with first a positive and then a negative value of g_1 assumed. The case with a positive value for g, gave the best fit to the data, as shown in Table I, and resulted in $\Delta \nu = 11\ 225(200)$ Mc/sec. The error is taken as twice the standard deviation resulting from the least-squares analysis. From the Fermi-Segrè formula and constants for the stable Cu isotopes,^{8,12} the uncorrected magnetic moment of 61Cu is calculated to be $\mu_1(\text{uncorr}) = +2.13(4)$ nm.



FIG. 1. A ⁶¹Cu resonance of the standard transition $(2, -1 \leftrightarrow 2, -2)$.

¹² H. L. Cox and D. Williams, Bull. Am. Phys. Soc. 2, 30 (1957); R. E. Sheriffs and D. Williams, Phys. Rev. 82, 651 (1951); R. V. Pound, ibid. 73, 523 (1948).

¹⁰ J. P. Hobson, J. C. Hubbs, W. A. Nierenberg, H. B. Silsbee, and R. J. Sunderland, Phys. Rev. **104**, 101 (1956).
¹¹ W. B. Ewbank, L. L. Marino, W. A. Nierenberg, H. A. Shugart, and H. B. Silsbee, Phys. Rev. **120**, 1406 (1960).

The nuclear spins of all the measured odd-A isotopes of copper are $\frac{3}{2}$, which is explained on the simple shellmodel picture by assigning the 29th proton to the $p_{3/2}$ shell. The magnetic moment of ⁶¹Cu lies within the Schmidt limits for a $p_{3/2}$ proton, and extends the monotonic decrease in the uncorrected magnetic moments from +2.38 for ⁶⁵Cu and +2.22 for ⁶³Cu to +2.13(4) for ⁶¹Cu.

⁶²Cu EXPERIMENT

⁶²Cu (9.9 min) was produced as a daughter isotope from the β^+ decay of 9.3-h ⁶²Zn. The zinc was made by the ${}^{60}Ni(\alpha,2n){}^{62}Zn$ reaction by bombarding 10-mil natural nickel with 40-MeV α particles. Simultaneously, some ⁶¹Cu (3.3 h) is formed from several reactions, but this isotope is isolated in the first precipitation of copper from the nickel-zinc solution. A portion of the ⁶¹Cu is subsequently added to each later precipitation to provide a long-lived component in the beam for normalization purposes. The chemical procedure for ⁶²Cu was similar to that previously described for ⁶¹Cu except that the nickel foil was first dissolved in hot aqua regia. After each copper precipitation the ⁶²Cu activity grows to a maximum in about 45 min. Hence samples were taken at about 60-min intervals. The copper metal from the chemistry could be produced in 10 to 15 min after the precipitation phase. Most of the metal was placed in the atomic beam oven; however, a small portion was deposited on a chemistry button, which was decayed along with the sample collected in the atomic beam apparatus. Because of the short half-life of the isotope, the entire oven load was emptied on one button at a frequency corresponding to a resonance of a particular spin. This spin sample, along with the corresponding chemistry sample, was decayed to determine the ratio of ⁶²Cu to ⁶¹Cu activity. If the spin sample contains no resonance, the background on the spin sample should have the same ⁶²Cu/⁶¹Cu ratio as the chemistry sample. On the other hand, if the 62Cu undergoes a resonance which is deposited on the spin sample, then the ⁶²Cu/⁶¹Cu ratio on the spin sample increases over that on the chemistry sample. Two of the seven runs on ⁶²Cu are shown in Fig. 2. All samples taken at frequencies corresponding to spin 1 gave significant increases in the ⁶²Cu component on the spin sample. Two resonance curves were taken, at 8 and 16 Mc/sec, in an attempt to determine a preliminary value of the magnetic moment. Owing to the lack of higher field data, only a lower limit of 1000 Mc/sec for the $\Delta \nu$ could be established.

It is possible to explain the resulting spin for ⁶²Cu on the basis of the simple shell model. The odd 29th proton is assigned to a $p_{3/2}$ level. The five neutrons beyond the closed shell at 28 are divided by the shell theory between the $2p_{3/2}$ and $1f_{5/2}$ states, which lie close together in energy. Occupation of these two levels depends on the magnitude of the difference in



FIG. 2. Two spin searches show the enrichment of ⁶²Cu on the spin-1 samples.

pairing energies $(P_{f_{5/2}} - P_{p_{3/2}})$ relative to the level separation $(E_{f_{5/2}} - E_{p_{3/2}})^{.13}$ Three possible configurations result for the neutrons: (a) $(p_{3/2})^1(f_{5/2})^4$, (b) $(p_{3/2})^3(f_{5/2})^2$, and (c) $(p_{3/2})^4(f_{5/2})^1$. The Brennan and Bernstein rules¹⁴ for coupling between the odd proton and these neutron configurations yield possible spins of 0 and 3 for configuration (a), spin 2 for configuration (b), and the measured spin 1 for configuration (c).

64Cu EXPERIMENT

⁶⁴Cu was produced by the ⁶³Cu (n,γ) ⁶⁴Cu reaction in natural copper at the General Electric reactor at Vallecitos. The resulting activity, which was followed through 8 half-lives, showed a one-component decay of ≈ 12.8 h. Because previous work had identified the spin⁶ and approximate hyperfine structure,^{6,7} the experiments undertaken here were designed to obtain the sign of the nuclear magnetic moment, as well as improved values of the hyperfine-structure separation and the copper g_J factor. In this endeavor various resonances of the $\Delta F = 0$ and $\Delta F = \pm 1$ type were observed at magnetic fields up to 3734 G. Although all resonances were used in the least-squares analysis, particular resonances play a dominant role in determining certain constants in the Hamiltonian, Eq. (1). For example, the hyperfine structure is best established by narrow $\Delta F = \pm 1$ resonances taken at low magnetic fields. A resonance of this type at a field-independent point is

¹³ M. G. Mayer and J. Hans D. Jensen, *Elementary Theory of Nuclear Shell Structure* (John Wiley & Sons, Inc., New York, 1955).

¹⁴ M. H. Brennan and A. M. Bernstein, Phys. Rev. **120**, 927 (1960).



FIG. 3. A $\Delta F = \pm 1$ resonance in ⁶⁴Cu at a field-independent point. The central dip is due to two separated transition regions 90 deg out of phase.

shown in Fig. 3. Because this $\sigma(\Delta m_F = 0)$ transition was induced at the two ends of a $\pi(\Delta m_F = \pm 1)$ type of hairpin, the pattern is one caused by two separated oscillating rf fields 90 deg out of phase. As another example, the g_J factor and sign of the nuclear magnetic moment are best established by certain resonances at high magnetic fields. A sweep of the $(\frac{3}{2}, -\frac{1}{2} \leftrightarrow \frac{1}{2}, \frac{1}{2})$, $(\frac{3}{2},\frac{1}{2}\rightarrow\frac{1}{2},-\frac{1}{2})$ doublet at 1100 G showed only one resonant line, even though two were expected. (In labeling resonances the quantum numbers are taken from a diagram for a positive magnetic moment.) The frequency separation of the components of this doublet is often used to determine g_{I} , and is given by $2 g_{I} \mu_0 H/h$, which is about 362 kc/sec for ⁶⁴Cu at 1100 G (see Fig. 4). The absence of one component can be explained as follows: At zero magnetic field (for either a positive or negative sign of the nuclear magnetic moment) both



FIG. 4. Only one line of the doublet could be seen at 1100 G. The expected position for the missing line of low transition probability is indicated by the two arrows (one for a positive magnetic moment and the other for a negative magnetic moment).

components of this doublet have the same transition probability. However, as the field is raised the rf perturbation matrix element of one component (A)increases while that for the other component (B)decreases. The decrease in transition probability is explained by the fact that the B component has allowed changes in quantum number $\Delta F = \pm 1$, $\Delta m_F = \pm 1$ at low fields but has forbidden changes $\Delta m_I = \pm 2$, $\Delta m_J = \pm 1$ at high fields. Calculations of the shifts in frequency of these transitions for a change in the magnetic moment sign show that the A resonance is shifted only 28 kc/sec while the B resonance is shifted 696 kc/sec. In fact, the B resonance for a positive moment is located 362 kc/sec higher in frequency than the A resonance, and for a negative moment it is 362 kc/sec lower in frequency than the A resonance. Therefore the location of the lower probability transition Bin relation to the allowed transition A is a definitive indicator of the sign of the magnetic moment and aids greatly in assigning quantum numbers to the transi-



FIG. 5. Both doublet components were observed at 300 G. The position of the low-probability line on the low-frequency side of the other doublet component establishes the magnetic moment as negative. (The quantum number labels are those appropriate to a positive magnetic-moment diagram.)

tions. When the higher frequency component of the doublet has a lower transition probability, the magnetic moment is positive. When the lower frequency component is less intense, the magnetic moment is negative. An attempt to see the resolved, forbidden-component Bwas finally successful at 300 G, as shown in Fig. 5. This observation demonstrated conclusively that the magnetic moment of 64Cu is negative. This conclusion also results from the over-all least-squares analysis. A collection of data from 24 observations (with quantum labels from a positive moment diagram) and the result of a least-squares analysis of these data appear in Table II. The constants for ⁶⁴Cu are found to be $\Delta \nu(\frac{3}{2} \rightarrow \frac{1}{2}) = -1282.140(8)$ Mc/sec: μ_I (uncorr.) = -0.216(2) nm; $g_J(Cu) = -2.00228(2)$. The errors on $\Delta \nu$ and g_J are taken as twice the standard deviation of the least-squares analysis. The 1% error on

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	Calibration							⁶⁴ Cu residual frequency
Isotope	Frequency (Mc/sec)	Field (G)	F_1	⁶⁴ Cu T <i>M</i> 1	ransitio F ₂	M_2	Frequency (Mc/sec)	(negative magnetic moment) (kc/sec)
⁸⁵ Rb	80.515(40)	152.742(67)	3		3	- 3	177.450(150)	+79
³⁹ K	1088.203(15)	500.028(6)	3	= =	30	- 3	835.750(15)	+8
³⁹ K	10 122.973(100)	3 734.410(36)	3		3	- 3	9 646.785(70)	+13
³⁹ K	10 123.009 (110)	3 734.423 (39)	3	- 1	32	- 3	9 646.760(70)	-48
³⁹ K	10 123.192(100)	3 734.489 (36)	32	$-\frac{1}{2}$	32	$-\frac{3}{2}$	9 646.950(70)	-40
⁸⁵ Rb	80.478(30)	152.680(51)	32		12		1 208.810(20)	-3
⁸⁵ Rb	80.532(30)	152.771(51)	32	$-\frac{1}{2}$	1/2	$-\frac{1}{2}$	1 208.810(20)	-3
³⁹ K	3 308.745(10)	$1\ 300.081(4)$	- 3 2	$-\frac{1}{2}$	12	$-\frac{1}{2}$	3 435.420 (40)	-12
^{39}K	0.217 (50)	0.309(71)	32	$\frac{1}{2}$	- 12		1 282.138(10)	-2
³⁹ K	0.217(50)	0.309(71)	$\frac{3}{2}$	- 1 2	- 1 2	$-\frac{1}{2}$	1 282.140(10)	-0.2
^{39}K	0.217(50)	0.309(71)	32	12	12	$-\frac{1}{2}$	1 282.137(15)	-3
$^{39}\mathrm{K}$	0.562(20)	0.799(28)	$\frac{3}{2}$	12	12	$-\frac{1}{2}$	1 282.148(10)	+6
^{39}K	0.562(20)	0.799(28)	$\frac{3}{2}$	12	12	$-\frac{1}{2}$	$1\ 282.140(10)$	-2
³⁹ K	0.562(20)	0.799(28)	$\frac{3}{2}$	$\frac{1}{2}$	12	$-\frac{1}{2}$	1 282.139(10)	-3
$^{85}\mathrm{Rb}$	1.509(50)	3.225(107)	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$-\frac{1}{2}$	1 282.223(75)	+54
$^{85}\mathrm{Rb}$	2.291(50)	4.890(106)	$\frac{3}{2}$	$\frac{1}{2}$	12	$-\frac{1}{2}$	1 282.226(80)	+20
³⁹ K	428.445(50)	250.229(20)	$\frac{3}{2}$	1 2	12		1 446.636(20)	-4
¹³³ Cs	113.998(10)	300.038(24)	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$-\frac{1}{2}$	1 514.815(10)	+2
^{39}K	$1\ 088.100(25)$	499.991(9)	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$-\frac{1}{2}$	1 872.144(15)	+0.4
^{39}K	1 088.100(25)	499.991(9)	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$-\frac{1}{2}$	1 872.154(15)	+10
³⁹ K	2 750.501(10)	$1\ 100.068(4)$	32	$\frac{1}{2}$	$\frac{1}{2}$	$-\frac{1}{2}$	3 315.090(20)	+7
³⁹ K	3 308.745(10)	$1\ 300.081(4)$	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	<u>1</u>	3 841.025(20)	-2
$^{39}\mathrm{K}$	428.445(50)	250.229(20)	$\frac{3}{2}$	$-\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$1\ 446.574(20)$	+16
¹³³ Cs	113.998(10)	300.038(24)	$\frac{3}{2}$	$-\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	1 514.715(15)	+0.5
Calibrat	tion and comparison	information:						<i>,</i> , ,
		~ •	<i>(</i> 2,				$\Delta \nu$	μ_I (uncorr)
	Isotope	Spin	$g_{J}({}^{2}S_{1/2})$				(Mc/sec)	(nm)
	^{39}K	$\frac{3}{2}$	-2.0022	95(2)			461.719723	+0.3909
	⁶³ Cu	$\frac{3}{2}$					11 733.83(1)	+2.2206
	⁶⁵ Cu	$\frac{3}{2}$					12 568.81(1)	+2.3790
	⁸⁵ Rb	$\frac{5}{2}$	-2.0023	32(2)			3 035.732439	+1.3482
	⁸⁷ Rb	$\frac{3}{2}$	-2.0023	32(2)			6 834.682614	+2.7413
	^{133}Cs	$\frac{7}{2}$	-2.0025	42(2)			9 192.631770	+2.5641
Summa	ry of results:							
	64 Cu $I=1$ $g_J=-2.00228(1)$		$\Delta \nu = -1 \ 282.140(4)$			μ	I = -0.216(2) nm	$\chi^2 = 2.5$ (24 points)
			(1	For an ir	correct	positi	ve-moment assumptio	$\mathbf{n}: \boldsymbol{\chi}^2 = 145)$

TABLE II. ⁶⁴Cu data and results.

netic moment is intended to include a possible hyperfinestructure anomaly.

Coupling the moments of the spin- $\frac{5}{2}$ neutrons of 65 Zn or 67 Zn to the spin- $\frac{3}{2}$ protons of 63 Cu or 65 Cu results in a predicted magnetic moment of -0.50 to -0.65 nm for 64 Cu. This range of values based on a very simple coupling scheme does not agree well with the experimentally determined value of -0.216 nm.

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