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Hyperfine Structure of the 24-kev Transition in Sn^{119}†

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The Zeeman splittings of the two lower levels of Sn^{119} were measured by the method of recoilless emission and resonant absorption of the 24-kev γ radiation emitted in the decay of Sn^{119m}. The source consisted of Sn^{119m} diffused into 0.002-in. iron foil. Grey tin metal at a temperature of ~10°C was used as the absorber. The magnitude of the internal magnetic field at the source nuclei was sufficient to allow resolution of the six absorption peaks resulting from the $\frac{3}{2}^+ \rightarrow \frac{1}{2}^+$ $M1$ transition. Measurements were made with the internal fields aligned by application of a small external magnetic field; in this manner the central line of each triplet could be enhanced or suppressed by observing the radiation emitted either perpendicular or parallel to the external field. The splitting parameters obtained from the measurements yield a value of 0.672 ± 0.025 nm for the magnetic moment of the first excited state of Sn^{119}, and an effective magnetic field at the tin nuclei in the iron environment of 78.5 ± 2.0 koe. The chemical shift is such that the transition energy in the source is less than that in the absorber by $(4.24 \pm 0.04) \times 10^{-8}$ ev. Within the limits of measurement, no quadrupole coupling was observed.

INTRODUCTION

THE hyperfine splitting of nuclear energy levels by magnetic and electric fields may be studied by observing the recoilless emission and resonant absorption of γ radiation between the levels involved. The condition for resonance between different components of the γ transition is obtained by the Doppler shift resulting from a small relative velocity between source and absorber.[1] If the levels in either the source or the absorber are unsplit, the observed absorption as a function of relative velocity consists of a single hyperfine spectrum rather than the resultant of the superposition of two such spectra. For the well-known case of the 14.4-kev radiation from Fe^{57}, magnetic fields sufficiently strong to split the hyperfine components are already present at the iron nuclei in the metal and many of its compounds because of their ferromagnetic properties. For materials such as tin which are not of themselves magnetic, an external field may be applied,[2–4] though generally this cannot be made sufficiently strong to yield resolvable lines. In order to obtain large effective fields, a magnetic compound or alloy may be prepared. Hanna, Meyer-Schützmeister, Preston, and Vincent[5] employed as absorber the ferromagnetic compound Mn_2Sn to study absorption of the 24-kev radiation of Sn^{119}, the source in this case being metallic tin containing Sn^{119m}; from the measured value of -1.041 nm[6] for the ground state magnetic moment they calculated from their absorption spectrum the effective field to be 192 ± 12 koe and the magnetic moment of the first excited state to be 0.75 ± 0.08 nm.[7] Boyle, Bunbury, and Edwards[4] in a similar measurement obtained a value of 195 ± 10 koe for the field in Mn_2Sn, and a value of 0.83 ± 0.03 nm for the excited state magnetic moment. The accuracy with which the Zeeman splitting can be deduced from measurements with Mn_2Sn is limited by the presence of quadrupole coupling.

The present paper describes a series of measurements of the absorption in grey tin metal of the 24-kev γ radiation emitted from Sn^{119m} present in an alloy of 4% tin in iron.[8] It was found that a magnetic field sufficient to resolve the Zeeman components of the radiation existed at the tin nuclei in the iron environment, and that these internal fields could be aligned by the application of an external magnetic field.[9] The resulting polarization made it possible to enhance or decrease the relative intensities of the different components of the hyperfine spectrum by observing the γ radiation emitted either perpendicular or parallel to the direction of magnetization. The absorption spectra so obtained made possible a more precise determination of the hyperfine splitting parameters.

APPARATUS AND EXPERIMENTAL PROCEDURE

The source was prepared by electroplating metallic tin containing Sn^{119m} onto 0.002-in. iron foil. The Sn^{119m} activity was produced by irradiating SnO_2 enriched in Sn^{118} in the Oak Ridge reactor for several weeks. A total of ~2 mg of metal was plated over a circular area 13 mm in diameter, and subsequently annealed for 5 hr at 900°C. The resulting source strength was

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[1] R. L. Mössbauer, Z. Physik **151**, 124 (1958).

[2] V. A. Lyubimov and A. I. Alikhanov, Soviet Phys.—JETP **11**, 1375 (1960).

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[4] A. J. F. Boyle, D. St. P. Bunbury, and C. Edwards (to be published).

[5] S. S. Hanna, L. Meyer-Schützmeister, R. S. Preston, and D. H. Vincent, Phys. Rev. **120**, 2211 (1960).

[6] W. G. Proctor, Phys. Rev. **79**, 35 (1950).

[7] S. S. Hanna, L. Meyer-Schützmeister, R. S. Preston, and D. H. Vincent, Bull. Am. Phys. Soc. **5**, 429 (1960).

[8] That the tin nuclei would experience a magnetic field under such conditions is suggested by the results obtained for diamagnetic elements dissolved in iron by B. N. Samoilov, V. V. Skylarevskii, and E. P. Stepanov, Soviet Phys.—JETP **11**, 261 (1960).

[9] Preliminary results have been reported by O. C. Kistner and J. B. Swan, Bull. Am. Phys. Soc. **6**, 51 (1961). Measurements of the magnetic field at tin nuclei contained in ferromagnetic metals have also been reported by Boyle, Bunbury, and Edwards (reference 12).

~100 μC. The absorber was prepared by binding a reasonably uniform layer of grey tin powder (20 mg/cm^2) onto a supporting backing of beryllium with acrylic resin. Grey tin, having a cubic structure, is not expected to exhibit quadrupole coupling; it has the further advantage of possessing an appreciably higher Debye temperature than white tin.

The hyperfine spectrum of the 24-kev γ radiation was measured in the usual manner by counting the γ rays transmitted through a resonant absorber as a function of the relative velocity of the absorber with respect to the source. In these measurements the relative velocity was varied continuously, covering the entire range of velocities many times a second. The pulses from the γ-ray detector were modulated with a linear velocity-to-pulse-height converter and then stored in a multi-channel analyzer.

A transducer and velocity detector were conveniently made from a pair of loudspeakers. The source (or absorber) was rigidly mounted to the voice coil of one speaker. This speaker was then driven at its resonant frequency of approximately 30 cps by air coupling to a second speaker which was excited electrically at the desired frequency and amplitude by an audio oscillator. For small excursions of the voice coil of the first speaker about its equilibrium position, the voltage induced across it is directly proportional to its velocity. This voltage, after amplification[10] (without phase shift) determines the envelope of modulation of the velocity-to-pulse-height converter.

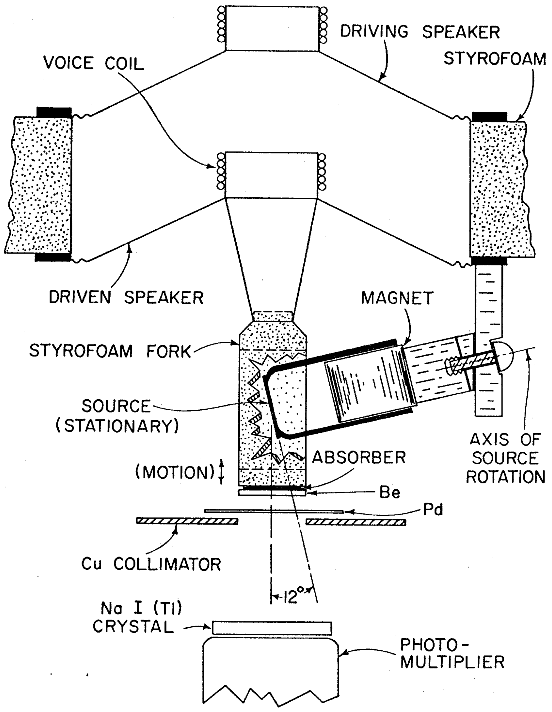

FIG. 1. Arrangement of apparatus used to provide relative motion between the source and absorber, and for detection of γ rays at various angles of emission with respect to the direction of magnetization of the source.

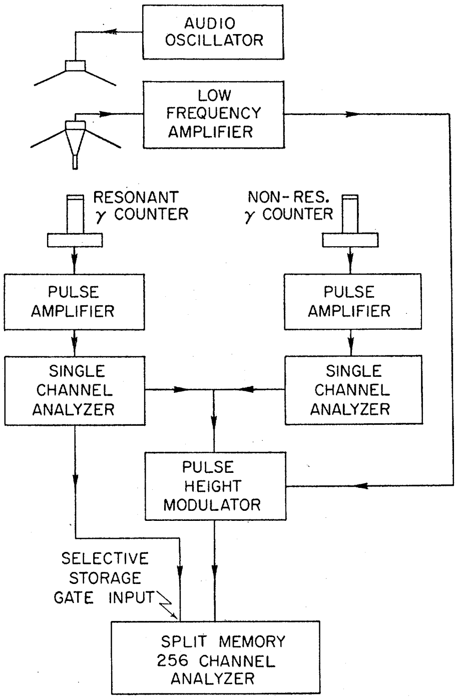

FIG. 2. Block diagram of the circuits used for recording count rate as a function of relative source-absorber velocity.

The mechanical arrangement is shown in Fig. 1. The magnet assembly which supported the source foil was rigidly attached to the stationary frame of the speaker. The absorber was suspended between the source and the scintillation counter and rigidly attached to the voice coil of the speaker by means of a "fork" of Styrofoam which passed around the pole pieces of the magnet. The magnet assembly was mounted with the plane of the source foil at an angle of 12° with respect to the line between source and counter. The angle of emission of the γ rays incident upon the counter, with respect to the direction of magnetization of the source, could be varied from ~12° to 90° with little change in geometry by rotating the magnet assembly about an axis perpendicular to the plane of the foil. A palladium filter (60 mg/cm^2) was placed between the absorber and counter in order to critically absorb the tin x rays. A collimator placed between the palladium filter and the counter reduced considerably the background due to Compton scattered photons. The detector, a NaI(Tl) crystal 2 mm thick and $1\frac{1}{2}$ in. in diameter, was situated $3\frac{1}{2}$ in. below the source.

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[10] The amplifier used was a chopper-stabilized model USA-3 by Philbrick Researches, Inc.; gain control was by adjustable negative feedback.

The entire system was housed in an insulating enclosure of Styrofoam blocks and maintained at a temperature of $\sim 10°C$, just below the transition temperature of grey tin. Cooling was conveniently accomplished by admitting into the Styrofoam enclosure the cold exhaust from a Dewar of liquid nitrogen in which was immersed a resistive heating element. The temperature was controlled by varying the current flowing through the heater. Styrofoam adequately fulfills the requirements for a rigid supporting and insulating material having a small γ-ray scattering cross section.

It should be noted that it is advantageous to maintain the grey tin absorber at a temperature not much below its transition temperature for the following reason. Grey tin usually contains an appreciable admixture of white tin, the recoilless absorption spectrum of the latter being significantly shifted with respect to that of the former due to different chemical shifts. Because of the much higher Debye temperature of the grey form of tin, the Debye-Waller factor at 10°C will favor the cross section for resonant absorption by grey tin with respect to that by white tin by a factor of 12. At lower temperatures the Debye-Waller factors for the two forms of tin will approach each other; e.g., at liquid nitrogen temperature this ratio is reduced to 1.2.

The block diagram in Fig. 2 illustrates the electronic circuitry used to obtain the spectra of counts vs absorber velocity. The approximately sinusoidal motion of the absorber results in a larger accumulation of counts per velocity interval at the higher speeds, giving a U-shaped distribution symmetric about zero velocity. In order to normalize the velocity spectrum and to correct for analyzer dead time the following procedure was used.

Random pulses were obtained from a separate scintillation detector (nonresonant counter) and arbitrary source and were treated in as near identical a manner as possible to the pulses from the detector (resonant counter) counting γ rays from the Sn^{119m} source. The output from each photomultiplier was amplified and passed through a single-channel analyzer, the resonant channel being set on the 24-kev line and the nonresonant channel being set somewhere on the spectrum of the

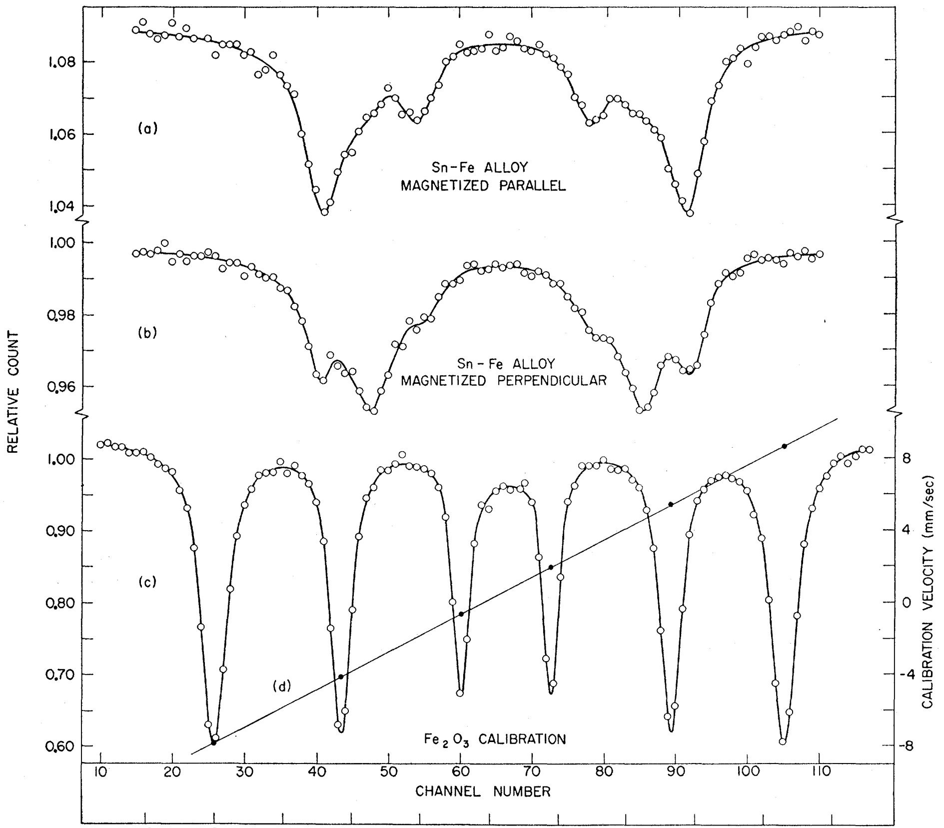

FIG. 3. Experimental absorption spectra obtained by dividing the resonant spectrum by the nonresonant spectrum. Positive velocity indicates the source and absorber approaching each other. (a) and (b) Spectra of magnetized Sn^{119m} in iron source vs grey tin absorber, for γ rays emitted near-parallel and perpendicular to the direction of magnetization, respectively. (c) Calibration spectrum of Co^{57} in stainless steel source vs Fe^{57} enriched Fe_2O_3 absorber. (d) Linear calibration line drawn through the points obtained from the peak positions of curve (c) and the known velocities (right-hand ordinate scale) listed in Table I.

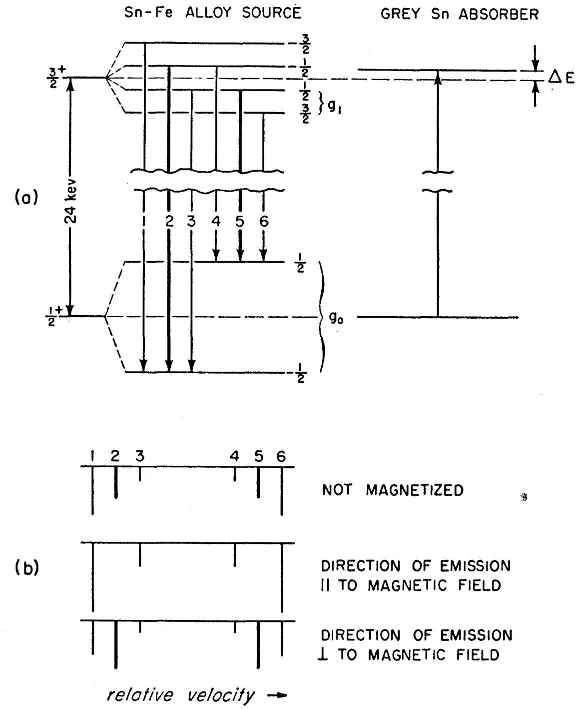

FIG. 4. (a) Zeeman splitting of the 24 kev and ground state levels in Sn^{119} contained in an iron environment and the chemical shift relative to the unsplit levels in grey tin, as deduced from the absorption spectra. The six components corresponding to $M1$ radiation are shown. (b) Theoretical relative intensities of the six absorption lines as they appear in the absorption spectrum for an unmagnetized source and for the two directions of emission from a magnetized source. The lines are numbered according to the corresponding emission components in (a). The angular dependence of the radiation, either $\sin^2\theta$ or $1+\cos^2\theta$, is denoted by the use of heavy or light lines, respectively.

arbitrary source so as to provide the desired count rate. The outputs of these two channels were mixed at the input to the modulator. The output of the modulator, consisting of pulses proportional in height to the instantaneous velocity, was fed into a 256-channel pulse-height analyzer. Selective storage was used to store the "non-resonant" pulses in channels 0-127 and the "resonant" pulses in channels 128-255. Each half of the memory thus displayed a U-shaped spectrum with channel number directly proportional to velocity, zero velocity being at the center. The normalized velocity spectrum was obtained by dividing the number of counts stored in each resonant channel by the number in the corresponding nonresonant channel.

The velocity range could be varied by adjusting the amplitude of the driving speaker, and the number of channels over which the spectrum was spread could be varied by adjusting the gain of the voice coil voltage amplifier. The channel number corresponding to zero velocity was set at 64 by adjusting the lower level setting of the analyzer, and could be conveniently checked at any time by shorting the voice coil of the driven speaker. Linearity and calibration of the recording system were checked periodically using a source of Co^{57} diffused into 0.001-in. stainless steel and an Fe_2O_3 absorber enriched in Fe^{57}, the spectrum of which is known.[11]

RESULTS AND DISCUSSION

The experimental absorption spectra, obtained for two directions of emission from a magnetized source, are shown in Figs. 3(a) and 3(b). The Zeeman splittings of the 24-kev and ground state levels of the Sn^{119} nucleus deduced from these spectra are shown in Fig. 4(a). The theoretical intensity ratios of the six $M1$ emission lines as they appear in the absorption spectrum are shown in Fig. 4(b); for an unmagnetized source these ratios are 3:2:1:1:2:3, while for a magnetized source they are 3:0:1:1:0:3 for parallel emission and 3:4:1:1:4:3 for perpendicular emission. The agreement with the experimental spectra is good and indicates approximately 80% polarization.

A typical calibration spectrum is shown in Fig. 3(c) and the calibration velocities corresponding to each peak are listed in Table I. These velocities were calculated from the splitting parameters for Fe_2O_3 obtained in previous work.[11] The calibration points lie on a straight line, Fig. 3(d), to within the accuracy of measurement, thereby establishing the degree of linearity of the system. Periodic calibrations indicated that the slope of this line varied by less than ±1%.

The velocities of the absorption lines for the Sn^{119m} in iron source and the grey tin absorber determined from the data are listed in Table II. A least-squares fit to these velocities yields the following parameters:

$$g_0=0.644\pm0.010 \text{ cm/sec},$$
$$g_1=0.139\pm0.005 \text{ cm/sec},$$
$$\Delta E=0.053\pm0.006 \text{ cm/sec},$$
$$\epsilon=0.001\pm0.005 \text{ cm/sec}.$$

The errors are based on an estimated maximum error in peak location of ±0.5 channels, or ±0.01 cm/sec. The magnetic splittings of the ground and excited levels are denoted as g_0 and g_1, respectively. The line shift, ϵ,

TABLE I. Relative velocities (in cm/sec) used for calibration with the six absorption peaks from a source of Co^{57} in stainless steel and an Fe_2O_3 absorber, obtained from the splitting parameters determined in reference 11.

-0.788 ± 0.005
-0.419 ± 0.005
-0.074 ± 0.005
$+0.192\pm0.005$
$+0.537\pm0.005$
$+0.858\pm0.005$

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[11] O. C. Kistner and A. W. Sunyar, Phys. Rev. Letters 4, 412 (1960).

TABLE II. Observed velocities and corresponding $M1$ hyperfine components of the absorption lines obtained with the source of Sn^{119m} in iron and the grey tin absorber.

Hyperfine component	Relative source-absorber velocity (cm/sec)
$-\frac{3}{2} \rightarrow -\frac{1}{2}$	-0.477 ± 0.010
$-\frac{1}{2} \rightarrow -\frac{1}{2}$	-0.337 ± 0.010
$\frac{1}{2} \rightarrow -\frac{1}{2}$	-0.201 ± 0.010
$-\frac{1}{2} \rightarrow \frac{1}{2}$	$+0.303\pm0.010$
$\frac{1}{2} \rightarrow \frac{1}{2}$	$+0.449\pm0.010$
$\frac{3}{2} \rightarrow \frac{1}{2}$	$+0.581\pm0.010$

due to a possible quadrupole coupling, is zero within the limits of error. The chemical shift,[11] ΔE, is in the direction which indicates a larger energy for the 24-kev transition in the grey tin absorber than in the source.

These values, together with the value of -1.0411 ± 0.0002 nm[6] for the magnetic moment of the ground state of Sn^{119}, give for the magnetic moment of the 24-kev state,

$$\mu_e = +0.672\pm0.025 \text{ nm},$$

and an effective magnetic field at the tin nucleus in the iron environment of 78.5 ± 2.0 koe. The former is in agreement with the value of 0.75 ± 0.08 nm reported by Hanna *et al.*[7] but lies below the value of 0.83 ± 0.03 nm reported by Boyle *et al.*[4] The excessive width of the absorption lines observed in the measurements with Mn_2Sn may be due to the presence of a quadrupole coupling of the same order as the Zeeman splitting of the excited level. The angle between the electric field gradient giving rise to this coupling, and the magnetic-field producing the Zeeman splitting, may not be unique; this would produce asymmetric line broadening and unequal shifts in the apparent line position.

The value of 81 ± 4 koe for the effective magnetic field reported by Boyle *et al.*[12] for a 1% Sn-Fe alloy agrees with that obtained in the present work.

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[12] A. J. F. Boyle, D. St. P. Bunbury, and C. Edwards, Phys. Rev. Letters 5, 553 (1960).