

## Magnetic Hyperfine Interaction in $\text{Sb}^{121}$ Using the Mössbauer Effect\*

S. L. RUBY AND G. M. KALVIUS  
*Argonne National Laboratory, Argonne, Illinois*  
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The Mössbauer resonance in ferromagnetic MnSb was observed at 4.2°K. Using the ground-state interaction as given by NMR, we obtained the value  $g_{7/2}/g_{5/2}=0.498\pm 0.005$ . Substituting  $+3.359$  nm as the  $\frac{5}{2}$  moment gives  $+2.35\pm 0.03$  nm for the  $\frac{7}{2}$  state.

IN two previous investigations, we have determined the quadrupole splitting<sup>1</sup> and the isomer shifts<sup>2</sup> of the Mössbauer resonance of the 37.2-keV gamma transition in  $\text{Sb}^{121}$  in a variety of antimony compounds. From those data, we extracted the ratio of the quadrupole moments  $Q_{7/2}/Q_{5/2}=1.38\pm 0.02$  and the change in the nuclear-charge radius  $\delta r/r=(8\pm 3)\times 10^{-4}$ . For a complete description of the Hamiltonian for the nuclear hyperfine interaction it was still necessary to know the ratio of the  $g$  factors of the lowest two nuclear states. In the present paper, we determine this previously unknown ratio and use it and the well-known dipole moment of the  $\frac{5}{2}$  ground state to calculate the moment of the first excited  $\frac{7}{2}$  level. This result is then compared with the moment of the similar  $\frac{7}{2}$  ground state in  $\text{Sb}^{123}$ , and also with theoretical predictions made by Kisslinger and Sorensen<sup>3</sup> and by Noya, Arima, and Horie.<sup>4</sup> In the future, Mössbauer measurements can be used for the determination of magnetic fields acting at the Sb nucleus in different environments even if the field is too small to produce a fully resolved hyperfine pattern.

To obtain the ratio of the  $g$  factors, a stoichiometric compound showing ferromagnetic ordering was chosen to ensure that all nuclei would see the same hyperfine field. In addition, it would have been advantageous if the resonant nucleus had been situated in a cubic environment so that no quadrupole interaction could exist. Moreover, the rather short lifetime of the 37.2-keV state ( $T_{1/2}=3.5$  nsec) makes it important to find a substance having the largest possible internal field to gain the necessary resolution.

Our choice was MnSb. Extensive data from NMR<sup>5</sup> and neutron diffraction<sup>6,7</sup> show that this compound has a large internal field; but, unfortunately, having the NiAs structure, the Hamiltonian also contains a quadrupole

term. From Ref. 5, we know that the magnetic field acting at the Sb nuclei is 352.6 kOe. The quadrupole coupling is given as 93 MHz for  $\text{Sb}^{123}$  under the assumptions that the field gradient is axially symmetric (as is expected for the NiAs structure) and that the angle between the field gradient and the spin direction is 90°. The question of the spin direction in MnSb has caused some controversy in the literature; however, the most recent data by Takei, Cox, and Shirane<sup>7</sup> seem to indicate clearly that at low temperatures the spin direction is definitely perpendicular to the  $c$  axis, which is the direction of the field gradient. Taking the ratio  $Q^{123}/Q^{121}=1.275$ , obtained by Barnes and Bray<sup>8</sup>, we get  $\frac{1}{2}eQV_{zz}=18.25$  MHz=0.608 mm/sec, and with  $\mu_{5/2}^{121}=3.359$  nm, we get  $g_{5/2}=\mu^{121}H/I=12.1$  mm/sec. This shows that the quadrupole coupling is small in comparison with the magnetic splitting, so that only a slight deviation from a pure magnetic hyperfine pattern is expected.

The setup used was similar to the one described in our earlier experiments.<sup>1,2</sup> However, since long running times were necessary in the present experiment (e.g., the spectrum shown in Fig. 1, below, took one week of continuous sampling), a glass cryostat<sup>9</sup> was used for keeping source and absorber at 4.2°K. This system has a very low helium consumption of less than 0.8 liter/day. The gamma ray has to pass through 2 mm of glass, but this causes only minor absorption losses at 37 keV. The hyperfine splitting of metallic iron had been used to calibrate the spectrometer.

MnSb was prepared<sup>10</sup> by arc melting a mixture of Sb and Mn metals slightly rich in Sb to compensate for vaporization losses. The alloy was then held at 500°C in vacuum for one week to allow the formation of the ordered compound. X-ray analysis showed that the sample was of single phase and had the desired NiAs structure.

A pure magnetic hyperfine interaction will cause the ground state of  $\text{Sb}^{121}$  to split into six levels and the excited state into eight. Between these levels, the M1 selection rules allow 18 transitions. Figure 2 shows a plot of the calculated line positions for a range of values of  $R=g_{7/2}/g_{5/2}$ . In these calculations, the internal mag-

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<sup>1</sup> S. L. Ruby, G. M. Kalvius, R. E. Snyder, and G. B. Beard, *Phys. Rev.* **148**, 176 (1966).

<sup>2</sup> S. L. Ruby, G. M. Kalvius, G. B. Beard, and R. E. Snyder, *Phys. Rev.* (to be published).

<sup>3</sup> L. S. Kisslinger and R. A. Sorensen, *Rev. Mod. Phys.* **35**, 853 (1963).

<sup>4</sup> H. Noya, A. Arima, and H. Horie, *Progr. Theoret. Phys. (Kyoto) Suppl.* **8**, 33 (1958).

<sup>5</sup> A. Tsujimura, T. Hihara, and Y. Koi, *J. Phys. Soc. Japan* **19**, 1078 (1962).

<sup>6</sup> S. J. Pickard and R. Nathans, *J. Appl. Phys. Suppl.* **30**, 280S (1959).

<sup>7</sup> W. J. Takei, D. E. Cox, and G. Shirane, *Phys. Rev.* **129**, 2008 (1963).

<sup>8</sup> R. G. Barnes and P. J. Bray, *J. Chem. Phys.* **23**, 1177 (1955).

<sup>9</sup> M. Kalvius, in *Mössbauer Effect Methodology*, edited by I. Gruverman (Plenum Press, Inc., New York, 1966), Vol. 1, pp. 163-183.

<sup>10</sup> The sample was prepared by K. M. Myles of the Metallurgy Division, Argonne National Laboratory.

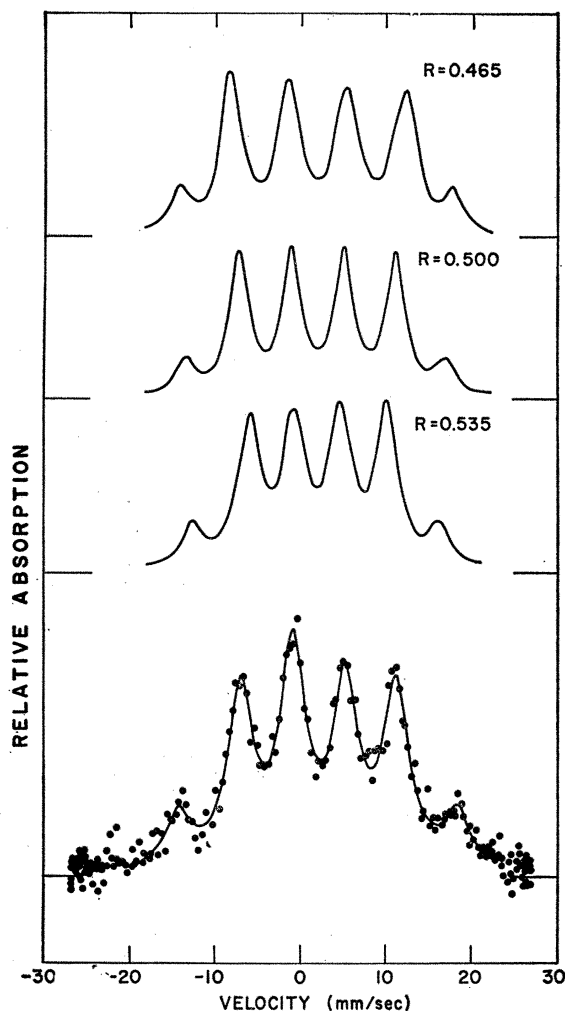


FIG. 1. Magnetic hyperfine splitting of the 37.2-keV gamma ray of  $\text{Sb}^{121}$ . Lower curve: the measured spectrum. The single-line source was  $\alpha$  tin (Sb), the absorber 22 mg/cm<sup>2</sup> MnSb. Both were kept at 4.2°K. Upper curves: calculated Mössbauer spectra for  $\text{Sb}^{121}$  for three indicated values of  $R = g_{7/2}/g_{5/2}$ . The field at the nucleus ( $H_i = 352.6$  kOe), the quadrupole interaction ( $eQV_{zz} = 72.6$  MHz), and the angle between  $H$  and  $q$  ( $\theta = 90^\circ$ ) are taken from Ref. 5.

netic field was taken as 353 kOe in accordance with Ref. 5, and the electric field gradient was assumed to be zero. The addition of a small quadrupole interaction will shift the transition energies only slightly. As one sees from Fig. 2, there are special values of  $R$  at which several of the absorption lines can have the same energy. The observed hyperfine spectrum is shown as the lowest curve in Fig. 1. Comparison with Fig. 2 reveals immediately that the value for  $R$  is close to 0.5. For this value, Fig. 2 indicates four prominent lines of about equal intensity and two pairs of weaker satellites, one pair on each side of this group. Since the outermost line of each pair is quite weak, the velocity sweep was reduced to sample only the six inner lines.

To obtain a more precise value for  $R$ , it is necessary to

take the quadrupole interaction into account. Therefore, we have calculated the Mössbauer spectra for  $R = 0.465$ , 0.500, and 0.535 with a computer program.<sup>11</sup> The ground-state Hamiltonian was taken from the NMR data given above and the ratio of the quadrupole moments was taken from Ref. 1. In Fig. 1, the calculations are compared with the experimental data. One sees that significant changes in the spectrum accompany this small variation of  $R$ . For  $R < 0.5$ , the two strong lines at negative velocities are closer together than the ones on the positive side, and the intensities of the four strong lines decrease from left to right. Both features are reversed if  $R$  becomes larger than 0.5. In the experimental spectrum, the inner lines are all of practically equal height except for the resonance at  $-0.9$  mm/sec. This again indicates that  $R$  has to be close to 0.5. From the relative positions of the four inner lines we extract six values for  $R$ . Averaging gives

$$R = g_{7/2}/g_{5/2} = 0.498 \pm 0.003,$$

where the error quoted is twice the standard deviation obtained from the scatter of the six values. The additional intensity of the second line from the left could be due to a slight oxidation of our absorber, since its position roughly agrees with that of the resonance for  $\text{Sb}_2\text{O}_3$ . From the area, we can conclude that this contamination is less than 3%.

The dipole moment of the  $\frac{5}{2}$  ground state is known from nuclear magnetic resonance.<sup>12,13</sup> The quoted

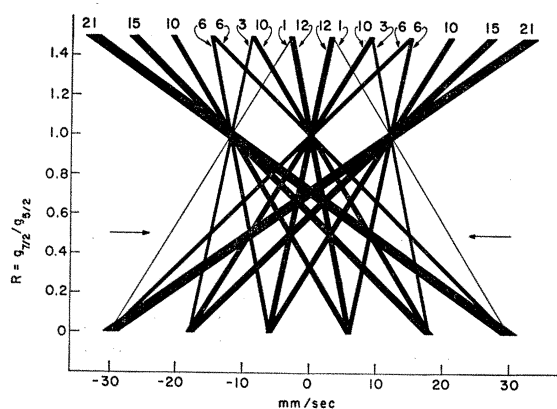


FIG. 2. Calculated spectra for  $\text{Sb}^{121}$  for values of  $R = g_{7/2}/g_{5/2}$  in the range  $0 \leq R \leq 1.4$ . The 18 allowed hyperfine transitions between the  $\frac{7}{2}$  excited state and the  $\frac{5}{2}$  ground state were computed for an internal magnetic field  $H_i = 353$  kOe. The width of the line for each transition is proportional to its relative transition probability, which is also given numerically above each line. The arrows indicate the experimental value of  $R$  obtained by comparing this figure with the measured spectrum in Fig. 1.

<sup>11</sup> J. R. Gabriel and S. L. Ruby, Nucl. Instr. Methods **36**, 23 (1965).

<sup>12</sup> V. W. Cohen, W. D. Knight, T. Wendnik, and W. S. Koski, Phys. Rev. **79**, 191 (1950).

<sup>13</sup> W. G. Proctor and F. C. Yu, Phys. Rev. **81**, 21 (1951).

value<sup>14</sup>  $\mu_{5/2} = +3.359 \pm 0.001$  nm contains a small diamagnetic correction whose uncertainty is the main contribution to the error. Using this number, we find the moment of the first excited state to be

$$\mu_{7/2}^{121} = +2.35 \pm 0.03 \text{ nm.}$$

This value is indeed close to the moment

$$\mu_{7/2}^{123} = +2.547 \pm 0.001 \text{ nm}$$

of the ground state of  $\text{Sb}^{123}$ . This agreement is to be expected, since the two nuclei differ only by the addition of two neutrons. However, it is noteworthy that although the theoretical framework used by Kisslinger and Sorensen<sup>3</sup> quite successfully explained the magnetic moments of nearly all odd-proton nuclei, it fails rather badly in this case. Where they predict 3.69 and 2.24 nm for the  $\frac{7}{2}^+$  and  $\frac{5}{2}^+$  states in  $\text{Sb}^{121}$ , the experimental

results are 2.35 and 3.36 nm. On the other hand, the computations of Arima and Horie are quite consistent with these moments. For example they give  $\mu_{7/2}^{121}$  as  $+2.45$  nm and  $\mu_{5/2}^{121}$  as  $+3.49$  nm,<sup>15</sup> in excellent agreement with our new experimental result.

Together with an earlier result<sup>2</sup> (namely, that the fractional change in nuclear-charge radius between the  $\frac{5}{2}$  and  $\frac{7}{2}$  states in  $\text{Sb}^{121}$  is much larger than predicted by Uher and Sorensen<sup>16</sup> and is of opposite sign), this is further evidence that the low-lying states in the antimony nuclei are not well represented by the theoretical assumptions used in Refs. 3 and 16.

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<sup>14</sup> G. H. Fuller and V. W. Cohen, in *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington 25, D. C., 1965), Appendix 1.

<sup>15</sup> A. Arima and H. Horie, *Progr. Theoret. Phys.* (Kyoto) **12**, 623 (1954); and private communication.

<sup>16</sup> R. Uher and R. A. Sorensen, *Nucl. Phys.* **86**, 1 (1966); and private communication.

## Effect of Low-Temperature Deuteron Irradiation on Some Type-II Superconductors\*

H. T. COFFEY, E. L. KELLER, A. PATTERSON, AND S. H. AUTLER

*Westinghouse Research Laboratories, Pittsburgh, Pennsylvania*

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To study the effects of low-temperature irradiation on some type-II superconductors, the critical current density  $j_c$ , transition temperature  $T_c$ , upper critical field  $H_{c2}$ , and normal-state resistivity  $\rho_n$  were measured after irradiation with 15-MeV deuterons. The  $10^{17}$ -deuteron/cm<sup>2</sup> irradiations produced about 0.3% atomic displacements. The ribbon samples consisted of Nb-61% Ti, Nb-25% Zr, Nb<sub>3</sub>Sn, Nb, and Pb. These samples were held at about 30°K during irradiation and 5°K between irradiation and measurement. The cryostat contained a 50-kOe superconducting magnet for *in situ* measurements. All effects were studied after irradiation and after annealing at 77 and 300°K. The resistivities were generally increased and the transition temperatures decreased by irradiation. Changes in  $H_{c2}$  (generally reductions) are correlated with changes in  $T_c$ ,  $\rho_n$ , and the Ginzburg-Landau parameter  $\kappa$ . Reductions of  $\sim 20\%$  in  $j_c$  of cold-worked NbTi and NbZr were observed, whereas the only  $j_c$  effect in strain-free NbZr was the production of a peak effect near  $H_{c2}$ . Large, thermally stable  $j_c$  changes were found in vapor-deposited Nb<sub>3</sub>Sn; an increase in low- $j_c$  material and a decrease in high- $j_c$  material. The induced effects in the alloys and pure metals were reduced by 75% or more upon warming the samples to room temperature. For Nb<sub>3</sub>Sn, less than 25% of the induced effects were recovered by annealing at 300°K.

### I. INTRODUCTION

ALTHOUGH many factors affecting it have been studied in detail, the precise mechanism(s) responsible for the large transport currents in type-II superconductors has not been determined. Heaton and Rose-Innes,<sup>1</sup> for instance, have clearly demonstrated that cold working a type-II superconducting alloy

markedly increases its critical current. Which of the many types of imperfections introduced by cold working was responsible for the increased current is not known, however. The controlled introduction of impurities frequently results in a current density peak near the upper critical field in niobium, but the physical mechanism correlating the cause and effect remains unknown.<sup>2,3</sup> Precipitated impurities likewise affect the

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<sup>1</sup> J. W. Heaton and A. C. Rose-Innes, *Appl. Phys. Letters* **2**, 196 (1963).

<sup>2</sup> W. De Sorbo, *Phys. Rev.* **132**, 107 (1963).

<sup>3</sup> W. De Sorbo, *Phys. Rev.* **134**, A1119 (1964).