

EVIDENCE FOR QUADRUPOLE INTERACTION OF  $\text{Fe}^{57m}$ ,  
AND INFLUENCE OF CHEMICAL BINDING ON NUCLEAR GAMMA-RAY ENERGY\*

O. C. Kistner and A. W. Sunyar

Brookhaven National Laboratory, Upton, New York

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The recoil-free emission and resonant absorption<sup>1</sup> of the 14.4-keV nuclear gamma ray of  $\text{Fe}^{57}$ , has been used to determine the quadrupole coupling for the 3/2- excited state of  $\text{Fe}^{57}$  bound in  $\text{Fe}_2\text{O}_3$ , and to measure an energy shift of this nuclear gamma ray which is attributed to effects of chemical binding. This effect is corollary to the effects of chemical environment on internal conversion coefficients<sup>2</sup> and on electron capture disintegration rates.<sup>3</sup> These measurements also yield the value of the internal magnetic field at the position of the  $\text{Fe}^{57}$  nucleus when it is bound in antiferromagnetic  $\text{Fe}_2\text{O}_3$ .

The M1 emission line of  $\text{Fe}^{57}$  bound in ordinary metallic iron is split into six components by the magnetic hyperfine interaction.<sup>4</sup> The resonant absorption of this emission spectrum by  $\text{Fe}^{57}$  bound in  $\text{Fe}_2\text{O}_3$  has been examined, as well as the much simpler absorption pattern which results when the "unsplit" emission line from  $\text{Fe}^{57}$  bound in a stainless steel lattice<sup>5</sup> is used. Because the interpretation of the two sets of measurements agree, only the latter measurements are presented in this Letter. The former measurements, in which a  $\text{Co}^{57}$  source co-plated with iron onto 1-mil copper was used, will only briefly be remarked upon.

The  $\sim 2\text{-mg/cm}^2$   $\text{Fe}_2\text{O}_3$  absorber used in these measurements contained  $\text{Fe}^{57}$  enriched to  $\sim 30\%$ . The source consisted of  $\text{Co}^{57}$  plated onto 0.001-inch stainless steel (25% Cr, 20% Ni). After plating, this source was annealed for one hour at  $900^\circ\text{C}$  in a hydrogen atmosphere. All measurements were made with source and absorber at a temperature of  $25^\circ\text{C}$ .

In order to obtain a Doppler shift of the emission line, a uniform motion was obtained by coupling a pneumatically driven cylinder to another cylinder filled with oil, the ports of which were connected via a needle valve. A wide range of uniform velocities could be selected by adjusting this valve. The direction of source travel was reversed automatically by means of microswitches. Additional microswitches, set to exclude the region of nonuniform motion near the travel limits, were used to provide gate signals for the counters. The distance of travel between the limits of the counting gates was 0.973 cm. Source velocity was determined

by counting the cycles from a 1000 cps tuning fork oscillator during the time between the gate limits.

The 14.4-keV gamma ray was detected with a NaI(Tl) scintillation counter. The phosphor was 2 mm thick and 1.5 inches in diameter. The counter face was located 5.4 cm above the upper limit of vertical travel of the source. The absorber was placed 4.7 cm from the counter face. A single-channel pulse-height analyzer selected the 14.4-keV gamma ray photopeak. The outputs of this analyzer and the 1000 cps clock were switched between two pairs of scalers so as to record counting rate and velocity separately for both directions of source motion.

Figure 1 shows the counting rate (in arbitrary units) as a function of source velocity relative to the  $\text{Fe}_2\text{O}_3$  absorber for the stainless steel source. Absorption of the "unsplit" emission line at each of the six possible absorption energies of  $\text{Fe}^{57}$  in  $\text{Fe}_2\text{O}_3$  is evident. The lack of symmetry of the absorption pattern about zero velocity shows immediately that one is not dealing simply with a magnetic hyperfine splitting pattern.

The velocities at which absorption peaks occur are given in Table I. Experimental values have been corrected by  $-2.5\%$  to take account of the effect of geometry on our velocity scale. These absorption line velocities may be fitted precisely in terms of an energy level diagram as shown in Fig. 2. It is necessary to introduce an energy shift  $\Delta E = \Delta E_1 + \Delta E_2$  between the center of gravity of the absorption lines of  $\text{Fe}^{57}$  in  $\text{Fe}_2\text{O}_3$  and the emission line of  $\text{Fe}^{57}$  in stainless steel. In addition, an energy shift  $\epsilon$ , of positive or negative sign, on the individual  $m$  states of the excited level is required. This is interpreted as being caused by a quadrupole interaction when  $\text{Fe}^{57}$  is bound in  $\text{Fe}_2\text{O}_3$ . A least-squares fit to our data yields the following splitting parameters (in "velocity units") for the two  $\text{Fe}^{57}$  nuclear states when  $\text{Fe}^{57}$  is bound in  $\text{Fe}_2\text{O}_3$ :

$$g_0' = 0.611 \pm 0.005 \text{ cm/sec,}$$

$$g_1' = 0.345 \pm 0.003 \text{ cm/sec,}$$

$$\Delta E = 0.047 \pm 0.003 \text{ cm/sec,}$$

$$\epsilon = 0.012 \pm 0.003 \text{ cm/sec.}$$

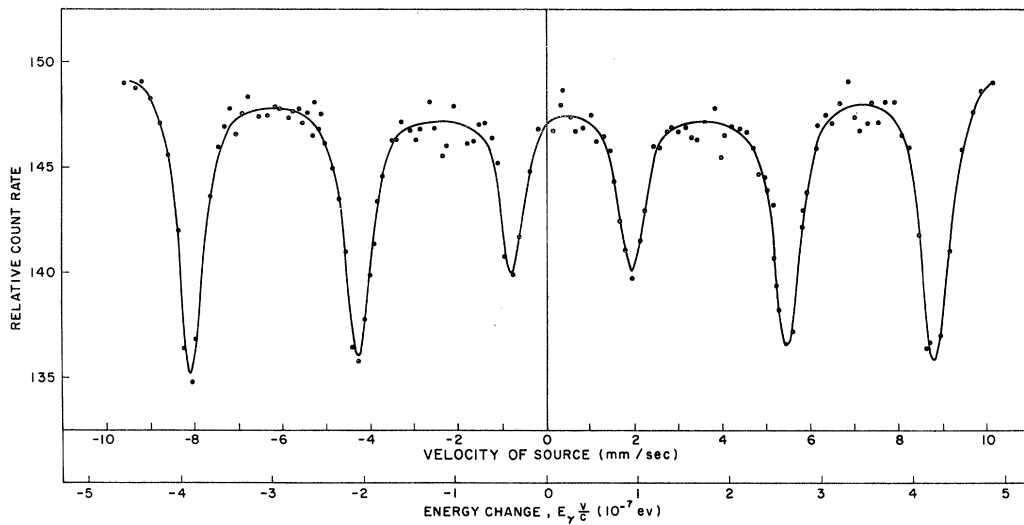


FIG. 1. The absorption by  $Fe^{57}$  bound in  $Fe_2O_3$  of the 14.4-keV gamma ray emitted in the decay of  $Fe^{57m}$  bound in stainless steel as a function of relative source-absorber velocity. Positive velocity indicates a motion of source toward absorber.

These data indicate that the ratio  $g_0'/g_1'$  is  $1.77 \pm 0.02$ , in excellent agreement with the results of Hanna et al.,<sup>4</sup> and that the internal magnetic field in antiferromagnetic  $Fe_2O_3$  is larger than in ferromagnetic iron by the factor  $1.547 \pm 0.022$ . If we take  $3.33 \times 10^5$  oersteds as the value of the internal field<sup>4</sup> in Fe, we find a value of  $5.15 \times 10^5$  oersteds at the Fe nucleus in  $Fe_2O_3$ . It is worth noting that the absorption peaks in Fig. 1 (reading from left to right) should have "thin absorber" intensity ratios of 3:2:1:1:2:3. However, the relative intensities found for the absorption peaks clearly exhibit evidence of saturation due to the rather large effective thickness of the enriched  $Fe_2O_3$  absorber. Nevertheless, the experimental intensity ratios are such that an inverted hyperfine pattern is required for

the 3/2- state, providing confirmation for this fact as previously reported.<sup>4</sup>

Let us now consider the effect of quadrupole

Table I. Velocities of maximum absorption (corrected for geometry) for a stainless steel source and  $Fe_2O_3$  absorber. The negative signs indicate motions of source away from absorber.

Absorption line	Relative source-absorber velocity (cm/sec)
$1/2+ \rightarrow 3/2+$	-0.789
$1/2+ \rightarrow 1/2+$	-0.417
$1/2+ \rightarrow 1/2-$	-0.076
$1/2- \rightarrow 1/2+$	+0.192
$1/2- \rightarrow 1/2-$	+0.536
$1/2- \rightarrow 3/2-$	+0.859

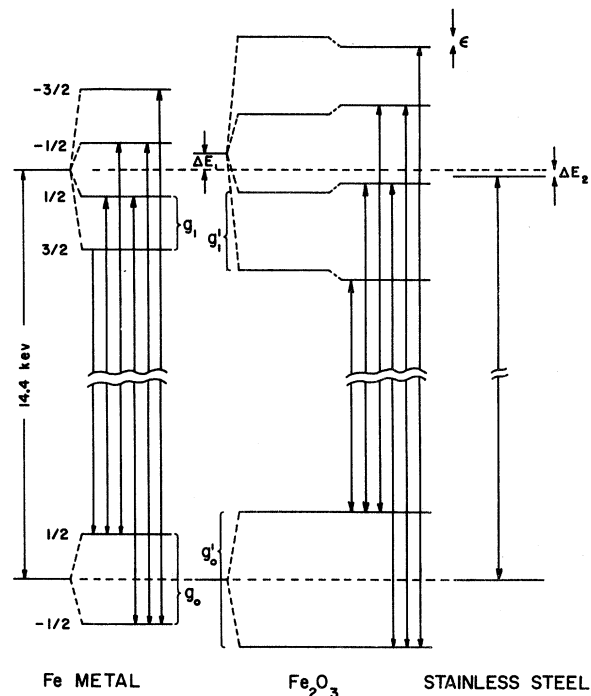


FIG. 2. Schematic representation of the ground and 14.4-keV excited states of  $Fe^{57}$  bound in ordinary iron,  $Fe_2O_3$ , and stainless steel. This diagram illustrates the details of magnetic hyperfine splitting, quadrupole interaction, and energy shifts due to chemical binding effects.

interaction. For a pure quadrupole spectrum, in the case of axially symmetric field gradients, the shift  $\epsilon$  for the substates of a spin  $I$  state is given by<sup>6</sup>

$$\epsilon = \frac{e^2 q Q}{4I(2I-1)} [3m^2 - I(I+1)],$$

where  $q$  denotes  $(1/e)(\partial^2 V/\partial z^2)$ . All other symbols have the conventional meanings. When Zeeman splitting is combined with quadrupole interaction, the precise energy shifts for the individual  $m$  states due to quadrupole interaction depend upon the orientation of the magnetic axis relative to the axis of symmetry for the electric field gradient, and are in general not the same for all  $m$  states.<sup>7</sup> However, for a case such as we deal with here, where the quadrupole interaction is small compared to the magnetic interaction, the absolute magnitude of  $\epsilon$  is the same for all  $m$  states of the spin  $3/2$  state within the accuracy of our measurements, independent of the orientation of the crystal axes. Thus, we find  $\epsilon = \frac{1}{4}|e^2 q Q| = 0.012 \pm 0.003$  cm/sec ( $5.75 \times 10^{-9}$  ev). Since our measurements indicate that the shift of magnitude  $\epsilon$  is negative for the  $m = \pm 3/2$  substates and positive for the  $m = \pm 1/2$  substates, the product  $e^2 q Q$  is negative. The absolute value is suggestive of a small quadrupole moment for  $\text{Fe}^{57m}$  although this statement cannot be amplified further without knowledge of electric field gradients in  $\text{Fe}_2\text{O}_3$ .

In our earlier absorption measurements on  $\text{Fe}_2\text{O}_3$  using the emission lines of  $\text{Fe}^{57}$  bound in ordinary iron, we find an energy shift upward (see Fig. 2) of  $\Delta E_1 \cong 0.04$  cm/sec. Within the accuracy of our measurements, the splitting parameters  $g'_0$ ,  $g'_1$ , and  $\epsilon$  remain the same. A separate measurement of the absorption by  $\text{Fe}^{57}$  bound in stainless steel of the emission lines of  $\text{Fe}^{57}$  in ordinary iron confirms the implied existence of an energy shift downward (see Fig. 2) of  $\Delta E_2 = 0.01$  cm/sec in this case.

The existence of an energy shift  $\Delta E = (2.26 \pm 0.15) \times 10^{-8}$  ev between stainless steel and  $\text{Fe}_2\text{O}_3$  has been definitely established. We will now discuss the ways in which differences in chemical environment may produce such a shift. (1) Source and absorber will in general have different Debye temperatures. Since the nucleus in its excited state has a slightly greater rest mass than when it is in its ground state, the energy difference between the nuclear states will decrease from its value for an "unbound" system by different

amounts because of the difference in the zero-point energies. This reduction in energy is larger for the substance having the higher Debye temperature. (2) In addition, when a lattice is at a finite temperature, the energy difference between the nuclear states is reduced further by virtue of the previously reported temperature effect.<sup>8</sup> Since the magnitude of this reduction depends upon the integral of the specific heat of the lattice up to the temperature of the substance, it will be larger for the substance having the lower Debye temperature. We expect this temperature effect to be small, although it should be noted that it acts to reduce the energy shift between different substances which results from zero-point energy differences. The sign and magnitude of the observed shift between stainless steel and  $\text{Fe}_2\text{O}_3$  would require that  $\text{Fe}_2\text{O}_3$  have a considerably lower Debye temperature than stainless steel or ordinary iron. Were this the case, the Debye-Waller factor would materially depress the recoil-free resonant yield in  $\text{Fe}_2\text{O}_3$ , a result which is not indicated by the data. (3) When chemical environment is altered, a nuclear isotope shift may result. This effect has its origin in the change in the electronic wave functions over the region of space occupied by the nucleus.  $s$  electrons may be expected to contribute most to this effect. Since  $s$  electrons are in effect removed in going from Fe in metal to Fe in  $\text{Fe}_2\text{O}_3$ , a smaller charge radius for  $\text{Fe}^{57m}$  than for  $\text{Fe}^{57}$  in its ground state would produce a shift in the observed direction. The direction of the observed energy shift  $\Delta E$  requires the presence of the nuclear isotope shift, since the zero-point energy shift is in the opposite direction. This nuclear isotope shift is similar to the isomeric isotope shift<sup>9</sup> observed in the optical spectrum of  $\text{Hg}^{197}$ .

We wish to emphasize two additional points about recoil-free emission and absorption experiments when source and absorber are chemically different. First, the existence of energy shifts introduces asymmetries into the absorption pattern which make it essential not to combine data taken at equal velocities of opposite sense. Second, any recoil-free absorption at zero velocity is accidental. In general, therefore, a search for this effect by comparing absorption at zero velocity with absorption at a large relative velocity between source and absorber will not yield significant results unless source and absorber are identical chemically. It

is amusing to note that in the absorption by  $\text{Fe}_2\text{O}_3$  of the emission lines of  $\text{Fe}^{57}$  in ordinary iron, two such accidental coincidences do indeed occur to give substantial absorption at zero velocity.

We wish to thank many of our colleagues, particularly M. Goldhaber and J. Wenner, for interesting discussions, and G. K. Wertheim for providing us with a sample of the particular stainless steel used in his measurements.<sup>5</sup>

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<sup>3</sup>R. F. Leininger, E. Segrè, and C. Wiegand, *Phys. Rev.* **76**, 897 (1949).

<sup>4</sup>S. S. Hanna, J. Heberle, C. Littlejohn, G. J. Perlow, R. S. Preston, and D. H. Vincent, *Phys. Rev. Letters* **4**, 177 (1960).

<sup>5</sup>G. K. Wertheim (private communication) has informed us that  $\text{Fe}^{57}$  in stainless steel exhibits a single absorption line. [See G. K. Wertheim, this issue, *Phys. Rev. Letters* **4**, 403 (1960).]

<sup>6</sup>T. P. Das and E. L. Hahn, *Nuclear Quadrupole Resonance Spectroscopy*, Solid State Physics, Suppl. 1 (Academic Press Inc., New York, 1958).

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<sup>9</sup>A. C. Melissinos and S. P. Davis, *Phys. Rev.* **115**, 130 (1959).

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## STRUCTURE OF NUCLEAR MATTER

A. W. Overhauser

Scientific Laboratory, Ford Motor Company, Dearborn, Michigan

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It is usually assumed that the Hartree (or Hartree-Fock) ground state of a Fermi gas is the familiar sphere of occupied momentum states. For a noninteracting gas such is, of course, the ground state; and for an interacting gas, this state is, to be sure, an exact solution of the Hartree-Fock equations. But to regard it as the lowest single-particle state has always been an unproved assumption, and indeed a false one. Explicit Hartree states of lower energy will be displayed below for a Fermi gas with attractive interactions. This problem is of practical interest with regard to nuclear matter, and will be discussed within that context. It will be shown that there are large static density waves in nuclear matter, so that (to an adequate approximation) the total nucleon density has the following spatial variation:

$$\rho = \rho_0(1 + \gamma \cos qx)(1 + \gamma \cos qy)(1 + \gamma \cos qz). \quad (1)$$

In another paper it will be shown that a low-energy Hartree-Fock state of a Fermi gas with repulsive interactions will have large spin density waves. It should not be necessary to emphasize that these new states have not been proved to be the ground state in the single-particle approximation, although they do have considerably lower energy than the "normal ground state." However, it seems clear that any attempt

to improve upon the single-particle approximation, such as the many-body techniques developed in recent years, should begin with the lowest Hartree-Fock state available.

It is not our purpose here to present a general argument, applicable to all conceivable interactions, but rather to illustrate the physical structure of the new low-energy states within the framework of a simple model. We shall treat first a one-dimensional problem, and afterwards a three-dimensional nucleon gas. Consider a large number  $N$  of spinless Fermi particles in a one-dimensional box of length  $L$ . The kinetic energy operator will be the usual one. We shall assume the total interaction energy of the gas to be

$$V = -(\beta L/2N) \int_0^L \rho^2 dx, \quad (2)$$

where  $\rho(x)$  is the total particle density and  $\beta$  a constant. If the difference between  $N$  and  $N-1$  is neglected, the Hartree potential  $U(x)$  experienced by any one of the particles is

$$U(x) = -(\beta L/N)\rho(x). \quad (3)$$

[The results to be derived are by no means limited to the particular interaction (2). For example, any power of  $\rho$  may be used in the inte-