QUADRUPOLE MOMENT OF $^{57}Fe^{m}$

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The quadrupole splitting induced by a magnetic field at low temperatures has been measured by Mössbauer spectroscopy for Fe^{2+} in MgO; a value of +0.21 ± 0.03 barn for the quadrupole moment of $^{57}Fe^{m}$ is obtained.

The value of the quadrupole moment of the 14.4 keV first excited state in $^{57}Fe^{m}$, determined in various investigations, is usually quoted with errors of about 10%, but the values differ from each other by more than 50%. This disparity is due to the various approximations made in calculating the appropriate electric field gradient. Two somewhat independent approaches have been used: namely, to calculate the electric field gradient for ferrous ions [1,2] and for ferric ions [3]. In the ferrous case the major contribution comes from the crystal field and spin-orbit split ^5D term, while in the ferric case, because of the ^6S ground state, the electric field gradient arises from net charges in the lattice. Ingalls [1] used ferrous data to calculate $Q = +0.29 \pm 0.02$ b; his calculation included covalency effects, but not the lattice electric field gradient. Later, Nozik and Kaplan [2] reviewed the problem, and included a calculation of the lattice electric field gradient; they obtained +0.20 b. Artman et al. [3] calculated the lattice electric field gradient for ferric ions in Fe_2O_3 and obtained $Q = +0.283 \pm 0.035$ b.

Because of the cubic symmetry, a quadrupole splitting for Fe^{2+} in MgO would not normally be expected. However, Leider and Pipkorn [4], using Mössbauer techniques with doped MgO absorbers, observed a quadrupole splitting of 0.33 mm/s below 14°K (above 14°K a single line is observed) which was explained by Ham [5] as the result of random strain splitting and slow relaxation. From a comparison of the electronic strain splitting with the splitting produced by a magnetic field [6], the strain splitting was estimated to be about 10^{-2} cm^{-1}. Ham showed that if $\langle r^{-3} \rangle (1-R) = 3.3$ a.u. and the orbital reduction factor $k = 0.8$, then the observed quadrupole splitting is consistent with a value of $Q = +0.21$ b, but not +0.29 b.

An applied magnetic field may also produce a quadrupole hyperfine splitting (in addition to a magnetic hyperfine splitting) from which it is possible to obtain a value for Q. In the cubic crystal field, the ^5D term is split into an orbital doublet and an orbital triplet, with the triplet lying 10^4 cm^{-1} below the doublet. Spin-orbit coupling leaves an electronic triplet state lowest. It is the strain splitting of this triplet that gives rise to the quadrupole splitting observed by Leiden and Pipkorn [4]. An applied magnetic field linearly splits the triplet; if the temperature is low enough so that the three states are not equally populated , a combined magnetic dipole and electric quadrupole hyperfine interaction is observed. The quadrupole interaction arises because $J = 1$ and the magnetic field lowers the effective symmetry. For larger magnetic fields (~ 10 kOe) the electronic splitting is large with respect to the strain splitting and the latter may be neglected. Thus, a measure of the quadrupole interaction in an external field gives a value of Q, which al-

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though related to the value derived from the strain splitting, is independent of the strain splitting. Although the strains are large enough to split the electronic state, they are too small to produce an appreciable lattice electric field gradient and hence there is no uncertainty due to the calculation of the lattice electric field gradient.

The theory of the field induced quadrupole interaction has been given by Ham [5]. For the magnetic field oriented along [100] one has

$$\Delta E_Q = e^2 qA = -\frac{3}{35} k\langle r^{-3}\rangle (1-R) \frac{e^2 Q}{I(2I-1)} ;$$

while for the field along [111], one obtains quadrupole interaction of the same magnitude but opposite sign; R is the Sternheimer shielding factor and k is the orbital reduction factor. Our ΔE_Q is twice that defined by Ham.

The Mössbauer source samples were prepared by depositing $^{57}CoCl_2$ solution on the [100] and [111] surfaces of single crystals of MgO and heating in air for about 24 hours at 1500°C. This treatment produces Fe largely in the 2^+ state.

The experiments were performed in a 50 kOe applied field at 4.2°K; the electronic splitting is 9 cm^{-1}, and only the lowest member of the triplet is appreciably populated. The observed field at the nucleus is -70 kOe, in agreement with the saturation hyperfine field of -120 kOe [6]. For the field oriented along [100], we find ΔE_Q = = -0.32 ± 0.05 mm s^{-1}, while along [111] we find ΔE_Q = +0.32 ± 0.05 mm s^{-1}. Using the value [1,5] of 3.3 a.u. for $\langle r^{-3}\rangle (1-R)$, and 0.8 for k, we find Q = +0.21 ± 0.03 b, where the error reflects only the experimental measurement.

A Jahn-Teller effect in the ground-state triplet would affect the relative magnitudes of the quadrupole interaction observed along [100] and [111]. The next excited electronic state above the ground triplet should come at 2λ i.e. $\sim 200\,cm^{-1}$, where λ is the spin-orbit splitting constant; however, it is observed [7] at 95 cm^{-1}. Using a suitable model, Ham [8] has shown that the reduction of the splitting is due to a Jahn-Teller effect, but that the effect in the ground triplet is small and does not affect the value of Q quoted above.

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