crystal, being influenced by the stress field due to the existing imperfections. It seems to be already certain that the observed boundary has a magnetic origin. However, in order to get more direct proof, further experiments such as an attempt to find any field effect will be made.

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# Electric Quadrupole Coupling in $\alpha$-Fe†

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The $^{57}$Fe 14.4-keV-level electric-quadrupole coupling in $\alpha$-Fe has been detected by Mössbauer spectroscopy.

The purpose of this note is to point out that the $^{57}$Fe 14.4-keV-level electric-quadrupole coupling in ferromagnetic $\alpha$-Fe is measurable by Mössbauer spectroscopy. A quadrupole effect was first observed in high-precision spectra of the National Bureau of Standards (NBS) Standard Reference Material (SRM) No. 1541, $\alpha$-Fe foil. Subsequently, 36 high-precision spectra from samples of this material have been measured on the NBS and Livermore absolute-velocity Mössbauer spectrometers at 298 and 4.3°K. These data confirm the presence of a quadrupole effect.

In previous studies[1] quadrupole coupling in $\alpha$-Fe was assumed to be zero because of cubic symmetry. However, even with cubic symmetry, a quadrupole effect can be magnetically induced.[2] Furthermore, $\alpha$-Fe is cubic only in the paramagnetic state. It must undergo a lattice distortion (magnetostriction) upon ferromagnetic ordering.[3] The magnetostriction of macroscopic $\alpha$-Fe samples indicates that the lattice distortion may be too small to be detected by standard x-ray techniques.

The material (SRM No. 1541) examined at both laboratories is low-carbon, high-purity iron, rolled to a thickness of about 20 $\mu$m. The impurities in weight percent are Ni, 0.03; Co, 0.02; Cr, 0.015; and Mn, 0.005. The carbon and oxygen contents are C, 60 ppm and O, 100 ppm. All other impurities are less than 0.001 wt%. The NBS absolute-velocity spectrometer uses an electromagnetic velocity transducer with an optical (Michelson) interferometer feedback loop, in the constant-acceleration mode.[4] The Livermore spectrometer is a mechanical constant-velocity system.[5] The velocity calibration of each spectrometer is derived from moiré-fringe distance measurements. The line positions were obtained by least-squares fitting, with no constraints, six Lorentzian lines to each spectrum (nineteen independent parameters). The $\alpha$-Fe quadrupole effect based on sixteen NBS runs with the sample at 298°K, and on twenty Livermore runs, twelve with the sample at 298°K and eight at 4.3°K, is given below. The 298°K results of the two laboratories are in excellent agreement and the weighted mean is quoted. The accuracies of both the NBS and Livermore results were determined by

the statistical errors only. All other random and systematic errors were negligible in comparison.[6]

If $a$, $b$, and $c$ are the respective $I=\frac{3}{2}$ splittings with quadrupole interaction, then necessarily $b=\frac{1}{2}(a+c)$. Values of $a$, $b$, and $c$ obtained from the NBS and Livermore results satisfy this requirement within experimental error.

If the quadrupole coupling is sufficiently small relative to the magnetic splitting, as it is in $\alpha$-Fe, then the quadrupole shifts in the $Fe^{57}$ excited-state ($I=\frac{3}{2}$) levels are essentially equal in magnitude for any value of the angle $\theta$ between the hyperfine magnetic field and the $Z$ principal axis of the electric-field gradient. Therefore, our results can be quoted in terms of, say, $\epsilon$, the quadrupole shift in the $\pm\frac{3}{2}$ levels. If $eQV_{ZZ}$ is the quadrupole coupling, then $4\epsilon$ varies between $eQV_{ZZ}$ ($\theta=0$) and $-2eQV_{ZZ}$ ($\theta=90°$). Twice the difference between the centers of gravity of lines 1 and 6 and lines 2, 3, 4, and 5 gives the best value[7] of $4\epsilon$:

$$4\epsilon = 2[\tfrac{1}{2}(v_1+v_6)-\tfrac{1}{4}(v_2+v_3+v_4+v_5)]$$
$$= \begin{cases} +0.0045 \pm 0.0012 \text{ mm/sec (298°K)} \\ +0.0088 \pm 0.0070 \text{ mm/sec (4.3°K).} \end{cases}$$

The effect at 298°K is almost four times the standard deviation and is significant. At 4.3°K the effect is comparable to the standard deviation and is only indicative. The 298°K result implies that the quadrupole coupling in $\alpha$-Fe at 298°K is between $-0.0022 \pm 0.0006$ and $+0.0011 \pm 0.0003$ mm/sec.

Our results do not specify which mechanisms are operative for quadrupole coupling in $\alpha$-Fe. We are pursuing this question by experimenting on $\alpha$-Fe foils placed in strong magnetic fields and on single crystals of very high-purity iron.

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