

$\text{EuH}_2$ . The spectra are very sensitive to the presence of such impurities, since all are paramagnetic at the temperature used in this experiment (and therefore they have sharp intense, single lines). They also have higher Debye temperatures and, therefore, higher recoil-free fractions than the metal, so that their lines appear relatively stronger. The Mössbauer spectra are less sensitive to the existence of such possible dissolved impurities as Ca, Yb, and H. However, the sharpness of the transition observed (see Figs. 3 and 4) and the good agreement between our  $T_i$  and the temperature of the specific-heat peak for the high-purity sample in

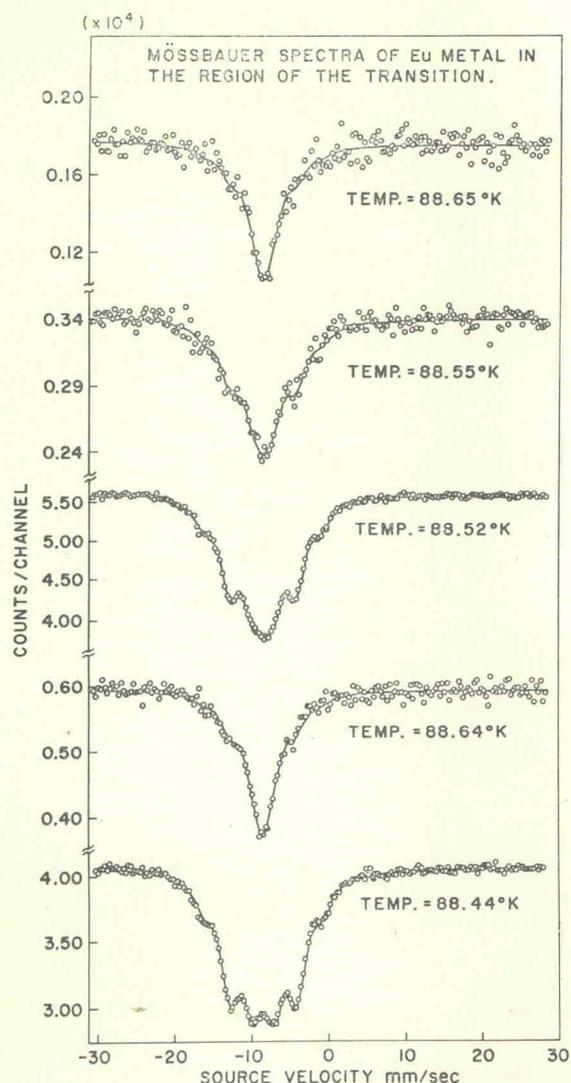


FIG. 2. Spectra of Eu metal in the temperature region of the phase transition. The spectra are shown (from the top) in order of the time at which they were taken. The solid line is the result of a least-squares fit assuming that the absorption resulted partly from a "split" spectrum like those in Fig. 1 and partly from a single line. This is equivalent to the assumption that paramagnetic Eu (unsplit spectrum) and ordered Eu (split) coexist over a narrow temperature range. Parameters evaluated from the least-squares fits are plotted in Figs. 4 and 5.

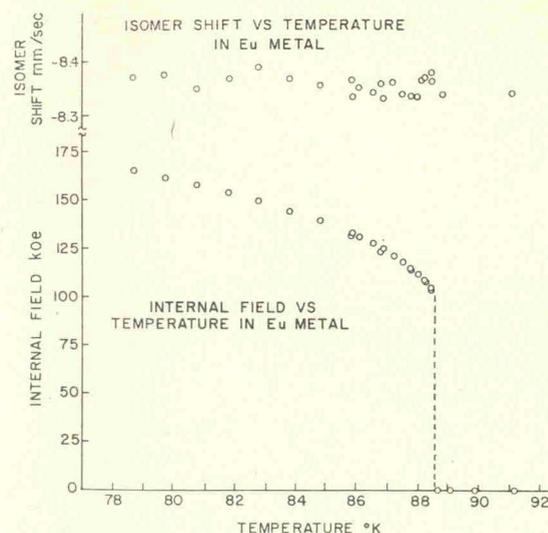


FIG. 3. (a) Observed internal fields plotted as a function of temperature. Note the sudden drop of the field at  $T_i$ . The ( $1\sigma$ ) error flags are about the size of the points. (b) Observed isomer shifts as a function of temperature. Note that there is no substantial discontinuity at  $T_i$ . Isomer-shift values are relative to an  $\text{Eu}_2\text{O}_3$  absorber at 78°K.

Ref. 5 suggest that the evaporated layer consisted of very pure Eu.

The Eu-coated beryllium disc was mounted in a copper ring with clamp screws, using silicone grease to improve thermal contact. The copper ring was surrounded by a radiation shield which was in thermal contact with it and this whole assembly was surrounded by a shield held at about 80°K during the measurements. The absorber holder was thermally connected to a liquid-nitrogen bath through a polyester heat leak, and heated by small resistors to reach the desired temperature. The temperature was measured by a differential thermocouple (Au-2.1%Co versus Cu) with the refer-

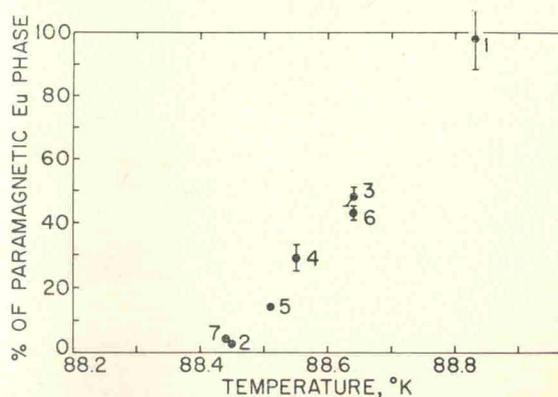


FIG. 4. Ratio of the areas of the ordered and paramagnetic parts of the absorption spectra in the transition region. The points are numbered in the order in which the runs were made. The figure shows clearly the absence of hysteresis. The total transition width is about 0.3°K. The transition width and temperature appear to lie intermediate between those for the two samples used in Ref. 5. (See Fig. 7.)

ence junction in contact with a bath of pure liquid nitrogen. The temperature variations of the reference bath with barometric pressure were taken into account. The thermocouple output was bucked by a voltage corresponding to the desired absorber temperature, and the resulting error signal was amplified and fed to the resistors mounted on the copper ring. This system provided simple, stable, proportional control. The over-all long-term stability and resetability was 0.03°K and was somewhat better for short measurements. These estimates are confirmed by the small scatter of the results. An additional over-all check and an absolute calibration at the boiling point of liquid oxygen were provided by a Cu-Constantan thermocouple (with 273.16°K reference junction). The absolute calibration accuracy of the temperature scale is estimated to be better than 0.3°K.

### B. Results of Mössbauer-Effect Measurements

The resulting spectra taken at 88.45°K and below were analyzed for hf field and isomer shift by least-squares fitting,<sup>12</sup> and the results are shown in Figs. 3 and 5 and in Table I. The calculated hf spectrum was highly constrained by the known properties of the Eu<sup>151</sup> nuclear-level scheme, and contained only the over-all magnetic hf splitting, isomer shift, and linewidth (assumed identical for all 18 lines in the spectrum) as significant free parameters. The good linewidths (~3 mm/sec, FWHM) and generally good fit between calculated and experimental spectra show that relaxation of the Eu-ion moments was fast enough to provide a well-averaged hf field at the Eu nucleus. Thus, the hf

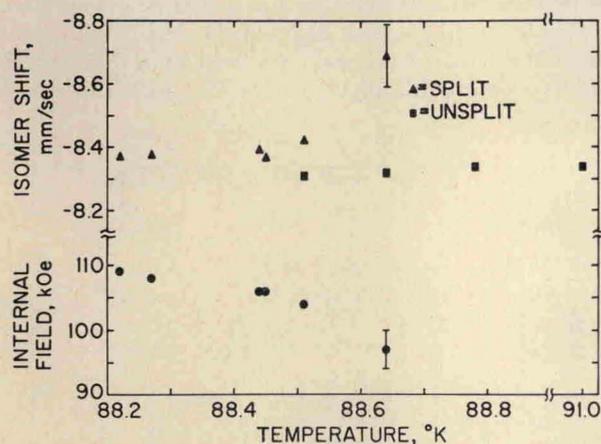


FIG. 5. Internal fields and isomer shifts obtained from the least-squares fits shown in Fig. 2. Error bars shown are  $1\sigma$ ; where no errors are indicated, they are about the size of the points. The fact that in the transition region the paramagnetic and ordered parts of the spectrum have noticeably different isomer shifts is discussed in the text. Note that the  $H_{int}$  of the split spectrum changes only very slightly in the transition region.

<sup>12</sup> We are indebted to Dr. C. S. Roberts and Dr. C. Reinsch for general purpose least-squares fitting programs.

splitting is proportional to the time-averaged hf field and, to a good approximation, to the sublattice magnetization. This fact will be used later in the discussion.

The temperature region just above 88.45°K shows spectra (Fig. 2) which appear to be a combination of "split" spectra from magnetically ordered Eu with approximately the same hf field as the 88.45°K spectrum, and a single line from paramagnetic Eu. This is exactly what would be expected if a variation in  $T_c$  through the absorber (resulting from strain or an inhomogeneous distribution of impurities) resulted in the coexistence of ordered and disordered phases over a narrow temperature range. To attain good fits, it was necessary to allow the "split" and "unsplit" spectra to have independent isomer shifts. This is discussed

TABLE I. Internal fields and isomer shifts in Eu at various temperatures. Indicated errors are  $\sim 1\sigma$  and represent only the precision of the data, arising primarily from statistical effects. Absolute (calibration) error is less than 3%. Isomer shift is relative to Eu<sub>2</sub>O<sub>3</sub> at 78zK.

Temp	$H_{eff}$ (kOe) ( $\pm \sim 0.3$ kOe)	Isomer shift (mm/sec) ( $\pm 0.02$ mm/sec)
5	257.2	8.36
78.74	164.7	8.37
79.74	161.5	8.38
80.78	158.0	8.35
81.82	154.6	8.37
82.81	149.6	8.39
83.82	144.9	8.37
84.83	139.6	8.36
85.86	133.0	8.37
85.89	132.7	8.35
86.07	131.8	8.35
86.56	128.0	8.34
86.83	125.2	8.36
86.90	125.8	8.33
87.22	122.0	8.36
87.53	118.8	8.34
87.84	115.0	8.34
87.85	114.5	8.32
88.02	113.1	8.34
88.22	109.4	8.37
88.27	108.1	8.37
88.44	104.5	8.38
88.45	105.2	8.36

below. As can be seen in Fig. 2, an analysis based on this model is very successful in explaining the observed spectra. This emphasizes the fact that there is a unique smallest field, essentially the value obtained at 88.45°K, from which the hf field goes discontinuously to zero as the material becomes paramagnetic. The plot of the relative intensities of the paramagnetic and ordered components (Fig. 4) shows us the width of the transition; the value of about 0.3°K in our absorber is slightly greater than the width of about 0.2°K observed for the specific-heat peak in the better of the two samples studied by Gerstein *et al.*<sup>5</sup> The curve of paramagnetic fraction versus temperature shown in Fig. 4 does not depend on the thermal history of the sample; thus, to the time scale of the shortest runs (about 10 min long), we can say that no hysteresis was observed. The