

goes suddenly from about 40 % of the saturation value to zero. The isomer shift remains essentially constant in passing through this transition (see fig. 2). Since the change in isomer shift between the $4f^6$ and $4f^7$ configurations is about 10 mm/sec, the observed constancy of the isomer shift absolutely excludes the hypothesis that the transition involves a 4f electron configuration change.

Further data on the detailed behavior of the spectrum in the temperature region about 88.6 °K is presented in figure 3. The success of the least-squares fits (described in detail in the figure caption) in describing the data in this temperature region shows that the observed spectra can be considered to arise from the simultaneous existence of two phases in the sample; a magnetically ordered one (with hyperfine splitting) and a paramagnetic one (which appears as the single sharp line in the center of the spectrum). In the temperature region from 88.4 °K to 88.8 °K, the relative proportions of the paramagnetic phase goes from ~ 0 to 100 %.

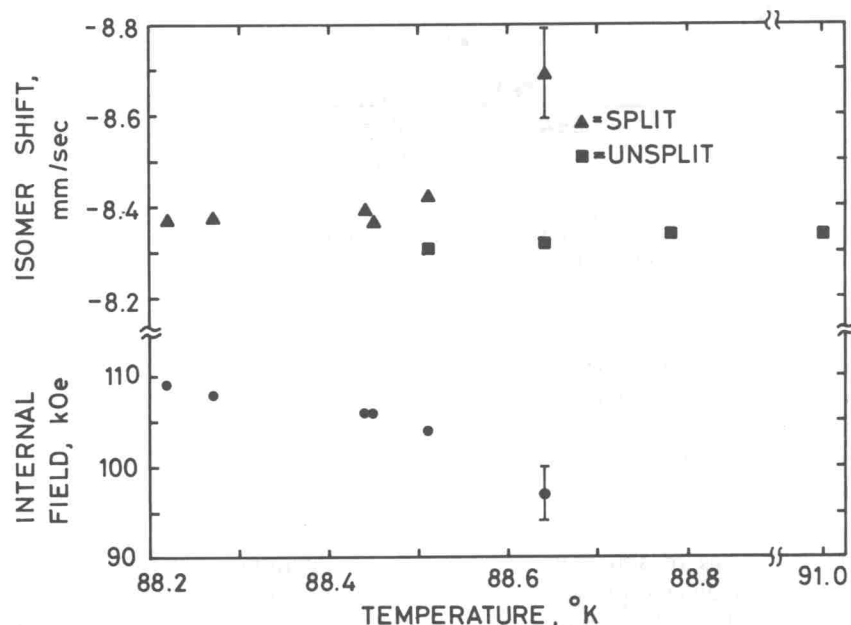


FIG. 4. — Internal fields and isomer shifts obtained from the least-squares fits shown in Figure 3. Error bars shown are 1σ ; where no errors are indicated, they are about the size of the points. Note that the H_{int} of the split spectrum changes only very slightly in the transition region.

Fig. 4 shows some of the results of the least-squares fits to the spectra in this transition region. It is particularly significant here that though the fraction of paramagnetic material decreases toward 88.6 °K, the hf field decreases only very slightly. Thus, the Eu ions transform from a state with a substantial magnetization, to one with no magnetization, discontinuously. In fig. 5, the relative intensities of the two components of the spectrum are shown, with some runs being only ten minutes long. The excellent retracing of the points on increasing and decreasing temperatures shows that, within the accuracy of the measurements, no hysteresis is observed.

Our observation and interpretation of this magnetic ordering as a first order transition is actually supported by the specific heat measurements referred to earlier [2], which observed a sharp peak in the specific heat of Eu metal samples at about 88.6 °K. Both the sharpness and position of the specific heat peaks were observed to be strongly dependent on sample purity.

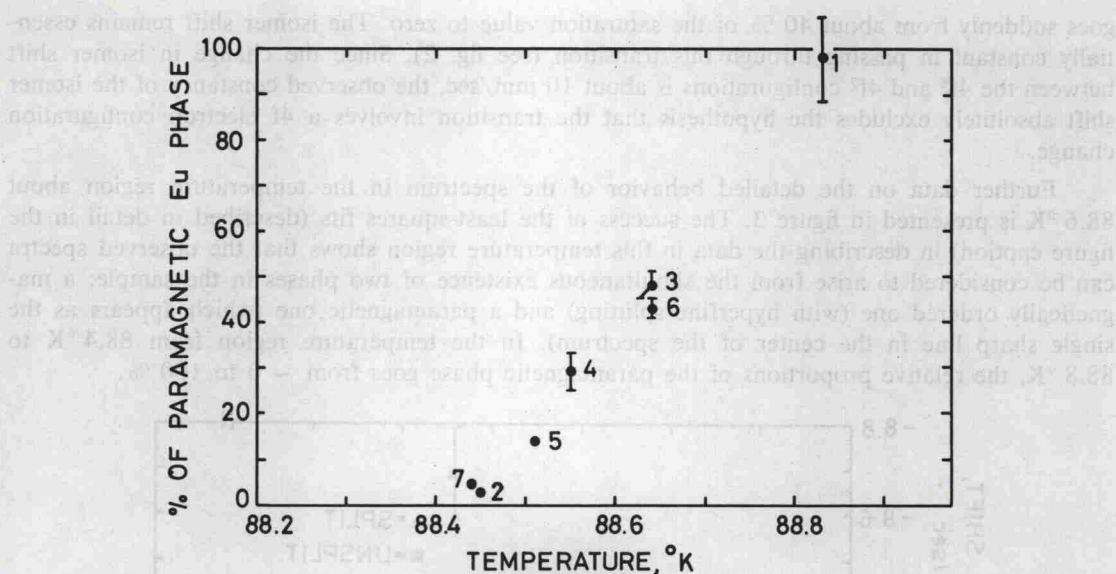


Fig. 5. — Ratio of the areas of the ordered and paramagnetic parts of the absorption spectra in the transition region. The points are numbered in the order in which the runs were made. The figure shows clearly the absence of hysteresis. The total transition width is about 0.3 °K. The transition width and temperature appear to lie intermediate between those for the two samples used in Ref. 2.

We hypothesize that the sharp transition we report was not observed earlier because of sample purity and temperature stability problems; the transition temperature is only about 1 °K below the magnetic ordering temperature obtained by extrapolating the data from 80° to 88° using a power law. Thus, if the sample is slightly non uniform or the temperature drifts slightly during the measurement, the sharp transition is blurred, and looks rather like a normal magnetic ordering.

There are two types of situations discussed in the literature which are used to develop models for first order magnetic phase transitions, which appear to be applicable to the analysis of the results reported here. The simpler of these, the "Isotropic magnetostriction" model developed by Bean and Rodbell [8], simply adds to the free energy of the magnetic spin system a term resulting from the mechanical (isotropic) distortion of the lattice. If the coupling between exchange and lattice constant is strong enough, compared to the stiffness of the lattice, the energy minimum criterion leads to a first order phase transition. For Eu, the appropriate parameters, compressibility and rate of change of " T_N " with lattice constant, are known [9], and it is easy to show that the compressibility is at least one order of magnitude too small to account for the transition with this model. This lack of a large lattice volume change at the magnetic ordering is confirmed both by X-ray studies [5] and by our own dilatometry experiments.

A more complex theory developed by Lines [10] is similar in concept, but considers lattice distortion rather than isotropic compression. We think that in Eu metal the dependence of the exchange on the lattice parameters must be responsible for the occurrence of the first order transition, and that, in principle, a theory as given by Lines and Jones [10] should also describe the present case. Due to the complex magnetic structure of Eu metal [1] no detailed theoretical treatment can be worked out with information presently available.