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Citation: J. Appl. Phys. **41**, 910 (1970); doi: 10.1063/1.1659014
View online: http://dx.doi.org/10.1063/1.1659014
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v41/i3
Published by the American Institute of Physics.

JOURNAL OF APPLIED PHYSICS VOLUME 41, NUMBER 3 1 MARCH 1970

Mössbauer Study of Some 2–17 Lanthanide–Iron Compounds*

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We have studied the iron-rich, rare-earth–iron intermetallic compounds R_2Fe_{17} (R = Pr, Gd, Tm, Lu), using the Mössbauer effect in ^{57}Fe. The measured average hyperfine field $H(T/T_c)$ acting on the ^{57}Fe nuclei is a unique, monotonically decreasing function of T/T_c, with no observed irregularity at temperatures corresponding to a suspected ferromagnetic–antiferromagnetic phase transition in these compounds. The onset of paramagnetism occurs at temperatures close to the *upper* transition points measured by Strnat *et al.*, namely at $T_c = 282°$, 472°, 271°, and 263°K for Pr_2Fe_{17}, Gd_2Fe_{17}, Tm_2Fe_{17}, and Lu_2Fe_{17}, respectively. Low-temperature ($\simeq$90°K) spectra indicate the presence of at least four magnetically nonequivalent Fe sublattices, while for $T > T_c$ broadened 2-line absorption spectra typical of ^{57}Fe at noncubic sites in paramagnetic material are observed.

In recent investigations[1–3] of the magnetic properties of iron-rich lanthanide–iron compounds (stoichiometry[4] R_2Fe_{17}, where R denotes a rare earth) an ordered, possibly antiferromagnetic phase has been postulated to exist at temperatures above the magnetic Curie point for R = Ce, Pr, Nd, Gd, Tm, and Lu. Low-field ac permeability[1,2] or neutron diffraction[3] measurements have indicated the presence of two transition temperatures in these materials, the lower transition temperature T_{CL} being identified[1–3] with the disappearance of ferro- or ferrimagnetic order. The significance of the upper transition temperature T_{CU} is unclear. Strnat *et al.*[1] tentatively suggested that an antiferromagnetic phase might be present for $T_{CL} < T < T_{CU}$.

We have used the Mössbauer effect in ^{57}Fe to examine the magnetic behavior of Pr_2Fe_{17}, Gd_2Fe_{17}, Tm_2Fe_{17}, and Lu_2Fe_{17}. In particular, we wished to test whether impurity effects could be a possible explanation for the appearance of two transition temperatures in the latter three compounds. (One could interpret T_{CL} as the order–disorder transition of the material proper, and ascribe the small permeability peak observed at T_{CU} to the presence of a ferromagnetic impurity.) The alloys used had been prepared for previous investigations. Preparation and analysis are described in the Refs. 1, 2, and 5.

Our Mössbauer measurements indicate that Gd_2Fe_{17}, Tm_2Fe_{17}, and Lu_2Fe_{17} become paramagnetic at temperatures T_c close to the previously determined[1] upper transition points T_{CU} (see Table I). Since the Mössbauer spectra correspond to the main phase, we may conclude that T_{CU} is a bulk property and cannot be associated with impurity effects. The newer permeability measurements[2] confirm this: The most likely contaminants are pure iron or other R–Fe compounds. None of these have their Curie point, or any known transition, near T_{CU}. Moreover, we find that the hyperfine field and spectral shape are continuous with no unusual anomaly near T_{CL}. This fact is rather difficult to reconcile with the postulated[1] ferromagnetic–antiferromagnetic transition at T_{CL}, though we feel that the possibility of such a transition is perhaps not completely ruled out by our data.

Pr_2Fe_{17} becomes paramagnetic at $T_c = 282°$K, and the antiferromagnetic phase postulated by Weik *et al.*[3] for $T > T_{CL}$ was not observed. Strnat *et al.*[1] also observed only one transition temperature (denoted arbitrarily T_{CL} in Table I) for this compound, and their value agrees well with our Mössbauer results.

In Fig. 1 we present typical low-temperature Mössbauer absorption spectra of Pr_2Fe_{17} and Gd_2Fe_{17}. Spectra of Tm_2Fe_{17} and Lu_2Fe_{17} in the ordered phase are similar. The spectra in Fig. 1 are complex, and at least four different component subspectra can be discerned. Such complexity is to be expected in view of the rather complicated crystal structure[1,5,6] of the R_2Fe_{17} compounds, there being four crystallographically nonequivalent Fe sites per unit cell. We have attempted a computer analysis of the Pr_2Fe_{17} spectrum, assuming four nonequivalent collinear Fe sublattices, but have been unsuccessful in obtaining a convincing fit to the experimental data. It seems that the actual magnetic structure is more complex.

In Fig. 2 we plot the temperature dependence of the approximate average hyperfine field H as a function of T/T_c. For $T/T_c \gtrsim 0.6$ the component subspectra are not resolved. At lower values of T/T_c we have plotted the weighted average hyperfine field. We stress that for the Gd, Tm, and Lu compounds our disordering temperature T_c, corresponding to the onset of paramagnetism, is close to the temperature T_{CU} reported in Refs. 1 and 2. It is also interesting, that within the accuracy of our measurements, the hyperfine field $H(T/T_c)$ appears to be a unique function of T/T_c for all four compounds studied.

Above T_c, the four compounds have 2-line absorption

TABLE I. Comparison of "Curie" temperatures of some R_2Fe_{17} compounds (R is a rare earth). The temperature T_{CL} corresponds to the disappearance of "ferromagnetism" (Refs. 1–3). At T_{CU} a secondary peak in the permeability is obtained (Refs. 1 and 2), or neutron diffraction (Ref. 3) indicates the persistence of magnetic order. We have used T_c to denote the onset of paramagnetism, as determined from the Mössbauer effect.

	"Curie" temperature in °K						
	Strnat *et al.* (Ref. 1)		Salmans (Ref. 2)		Weik *et al.* (Ref. 3)		This study
Compound	T_{CL}	T_{CU}	T_{CL}	T_{CU}	T_{CL}	T_{CU}	T_c
Ce_2Fe_{17}	93	411	91	403	...	...	...
Pr_2Fe_{17}	283	none	287	none	280	>570	282±1
Nd_2Fe_{17}	327	none	327	none	326	>570	...
Gd_2Fe_{17}	460	472	460	~473	...	...	472±3
Tm_2Fe_{17}	232	277	248	282	...	...	271±3
Lu_2Fe_{17}	238	265	235	268	...	...	263±2

spectra typical of ^{57}Fe in a paramagnetic material. In view of the noncubic symmetry at the various iron sites, a quadrupole splitting is to be expected, average values being 0.62±0.05 mm/sec, 0.59±0.05 mm/sec, 0.58±0.05 mm/sec, and 0.58±0.05 mm/sec for Pr_2Fe_{17}, Gd_2Fe_{17}, Tm_2Fe_{17}, and Lu_2Fe_{17}, respectively. At room temperature these compounds have isomer shifts close to that of Fe metal, the measured values being −0.01± 0.03 mm/sec, −0.19±0.05 mm/sec, −0.07±0.02 mm/sec, and −0.10±0.03 mm/sec, respectively, relative to Fe metal at room temperature.

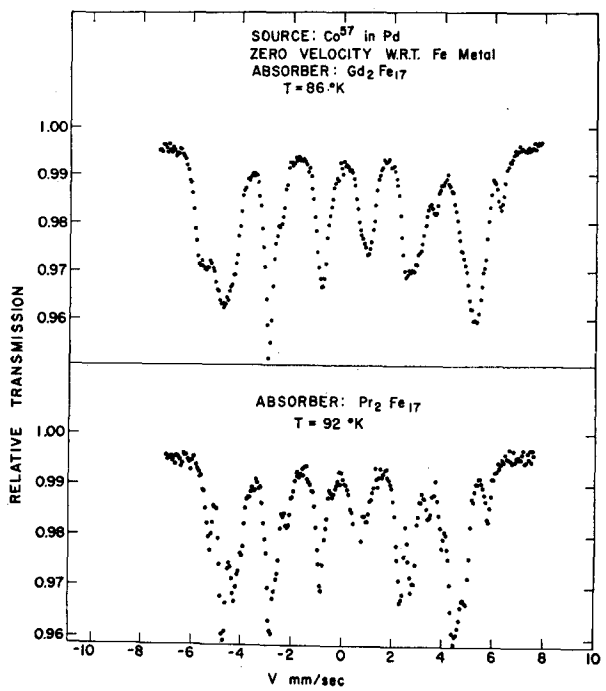

FIG. 1. Mössbauer absorption spectra of Pr_2Fe_{17} and Gd_2Fe_{17} at 92° and 86°K, respectively.

We have also undertaken macroscopic magnetic moment measurements on a powder sample of Lu_2Fe_{17}, using a vibrating sample magnetometer. Measurements in a field of 500 G showed a rapid drop of the magnetic

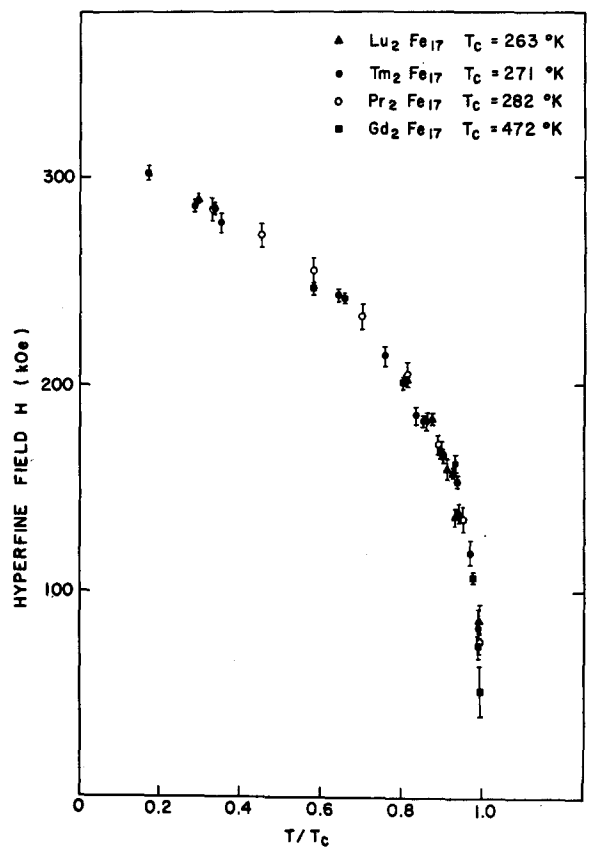

FIG. 2. Hyperfine field $H(T/T_c)$ for Pr_2Fe_{17}, Gd_2Fe_{17}, Tm_2Fe_{17}, and Lu_2Fe_{17}. At low temperatures, where more than one subspectrum can be resolved, we have plotted the weighted average hyperfine field.

moment in the vicinity of T_{CL} and a small peak at T_{CU} in accordance with the ac permeability data. The latter peak vanished upon repeating the measurement in a field of 5 kG. We may speculate that T_{CL} represents a drastic change of the crystal anisotropy while the ferro- or ferrimagnetic order persists up to T_{CU}. Our data does not support the notion[3,7] that magnetic order in some R_2Fe_{17} compounds exists even above ~500°K. More work will be necessary before the magnetic behavior of these substances is fully understood.

* The research reported in this document has been sponsored in part by the Air Force Materials Laboratory, Research and Technology Division, AFSC, through the European Office of Research, U.S. Air Force Contract F 61052-67C-0040.

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JOURNAL OF APPLIED PHYSICS VOLUME 41, NUMBER 3 1 MARCH 1970

Numerical Solution of the Behavior of Helical Spin Structures in Applied Magnetic Fields

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The behavior of helical spin structures and the phase transitions they exhibit in applied magnetic fields are studied theoretically.[1] Approximate solutions have been developed in the cases of high and low fields.[2] We have obtained a numerical solution valid for the case of the planar helix with an arbitrary number of interplanar exchange interactions, an arbitrary form of in-plane anisotropy, and an external magnetic field applied in the plane. We calculated the phase diagram for the case of two interplanar exchange constants and no anisotropy in order to compare with the previous theory and found different behavior in the intermediate field region. As a new result in the case of an in-plane anisotropy the angle dependence of the transition fields has been obtained.

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JOURNAL OF APPLIED PHYSICS VOLUME 41, NUMBER 3 1 MARCH 1970

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Speculations on Magnetic Charge

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