Search for Lifshitz points in rare-earth alloys

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Six rare-earth binary-alloy systems have been investigated in a search for Lifshitz points. No such points were found. All systems showed a highest ordering temperature jump when the sample composition went from values favoring helical ordering to values favoring ferromagnetic ordering. It is believed that ferromagnetic nearest-neighbor interactions and antiferromagnetic next-nearest-neighbor interactions are responsible for the observed behavior and that a finite-turn angle jump at the critical concentration will always occur for such systems.

I. INTRODUCTION AND PROCEDURE

The Lifshitz tricritical point is characterized by a generalized susceptibility function $\chi(\vec{q})$ which has vanishing first and second derivatives at $\vec{q}=0$. Here $\vec{q}$ is a vector related to the magnetic structure so that if $\chi(\vec{q})$ has a maximum at $\vec{q}=\vec{Q}$ the magnetic structure is helical with a layer-to-layer turn angle determined by $\vec{Q}$. A general review of the physical properties associated with Lifshitz points was presented by Hornreich[1] at ICM 1979 in Munich and may be found in the conference proceedings. As indicated in that review one might expect certain alloys of rare-earth metals to exhibit Lifshitz points because the temperature at which the change from helical to ferromagnetic ordering occurs has been found to be dependent on the composition of the alloys.[2] We report here an extensive search for Lifshitz points in Gd-Y, Gd-Lu, Tb-Gd, Tb-Th, Tb-La, and Dy-Th alloys.

Samples were prepared by the arc melting of weighed amounts of the high-purity constituents over a water-cooled Cu hearth in an argon atmosphere. Several meltings were used to ensure sample homogeneity. Pieces of the arc-melted button about $3\times3\times5$ mm^3 were cut for the magnetic-ordering temperature measurements. A vibrating sample magnetometer mounted in a cryostat was used to determine the magnetic moment versus temperature at constant applied field of ~ 20 Oe. For Néel points the temperature at the peak of the moment versus temperature was used. For Curie points we used the temperature at which inflection points appeared in the magnetic moment versus temperature curves.

II. EXPERIMENTAL RESULTS

The experimental results for Gd-Lu alloys are shown in the magnetic phase diagram of Fig. 1. It was originally thought a Lifshitz point would be found near 23.3 at.% Lu in Gd. However, the ordering temperatures found show that the 23.4 at.% Lu alloy is ferromagnetic, that the 24 at.% Lu alloy is antiferromagnetic at its highest-ordering temperature and that there is a temperature "gap" at the critical composition of 23.3 at.% Lu. Detailed 20-Oe isofield magnetization data for this system are shown in Fig. 2. The 23.0 at.% Lu sample shows the typical onset of ferromagnetism albeit the transition is fairly broad reflecting an expected statistical composition-fluctuation effect. For the 23.4 at.% sample, the transition is equally broad.

For the 24 at.% Lu sample there is a broad peak in the magnetization at 219.8 K and this is chosen as the Néel point. The very gradual rise in magnetic moment below 217 K leads to an inflection point near 215 K and this is chosen as the T_c' temperature at which ferromagnetic ordering in the sample takes over. For the 25 at.% Lu sample the Néel point is more sharply developed as might be expected since the antiferromagnetic phase becomes more highly favored.

Results for Gd-Y as reported by Legvold *et al.*[3] at ICM 1979 in Munich are shown in Fig. 3. It can be seen that results for Gd-Y and Gd-Lu are almost identical except for the composition at which the multicritical behavior occurs. The 5-K temperature gap is very nearly the same in the two systems.

For pure Tb and Dy the helical structure is favored at the highest ordering temperature. It has been shown by Burgardt *et al.*[4] that small amounts of Th or La will eliminate the helical structure in Tb making these alloys candidates for Lifshitz points. The results for Tb-Th are shown in Fig. 4. It is quite remarkable that the addition of less than 2 at.% Th can eliminate the helical structure. This is a quite different system from Gd-Y in that the helical structure is favored in the pure-Tb host and one moves toward ferromagnetic order as Th is added. Again there is a jump upward in the highest ordering temperature to a Curie point as Th is added. This system therefore

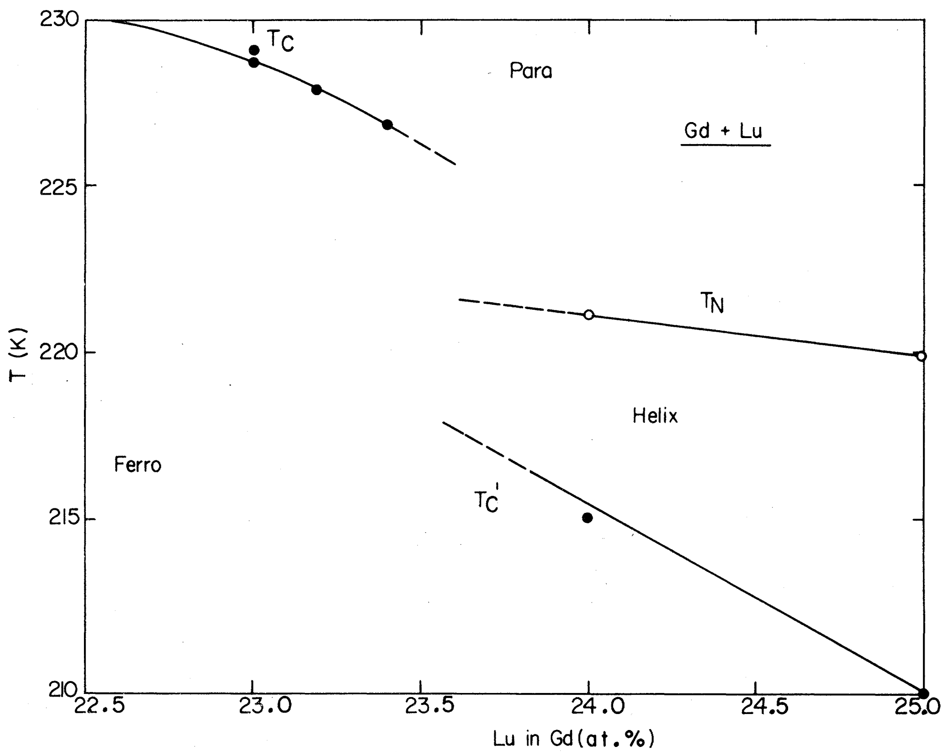

FIG. 1. Partial magnetic phase diagram for Gd-Lu alloys.

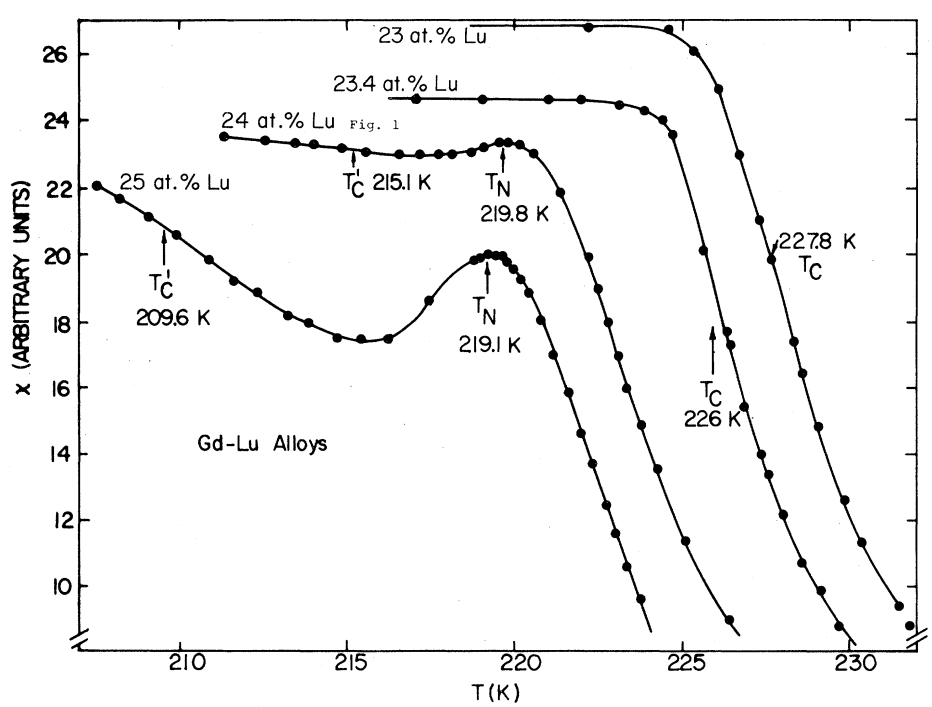

FIG. 2. Magnetic susceptibility vs temperature for four Gd-Lu alloys in a 20-Oe applied field. Additional data (not shown) at 40 Oe were used to find the ordering temperatures of the 24 at.% Lu in Gd sample.

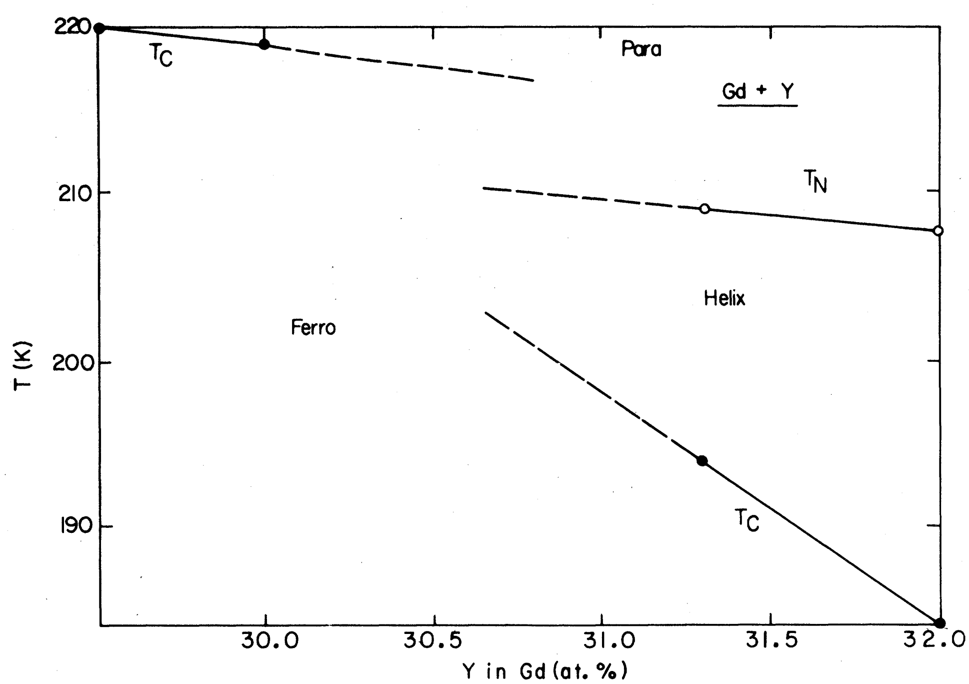

FIG. 3. Partial magnetic phase diagram for Gd-Y alloys.

does not give a smooth change from helical to ferromagnetic ordering and a jump in the magnetic-moment turn angle must take place near the 1.85 at.% Th critical concentration.

The critical concentration of La in Tb is close to 4.7 at.% as can be seen in Fig. 5. Again there is a small jump in the ordering temperature at the critical concentration.

In all the previous systems one element was magnetic and the other nonmagnetic. To investigate an alloy system consisting of two magnetic elements we chose to add Gd to Tb. The magnetic phase diagram

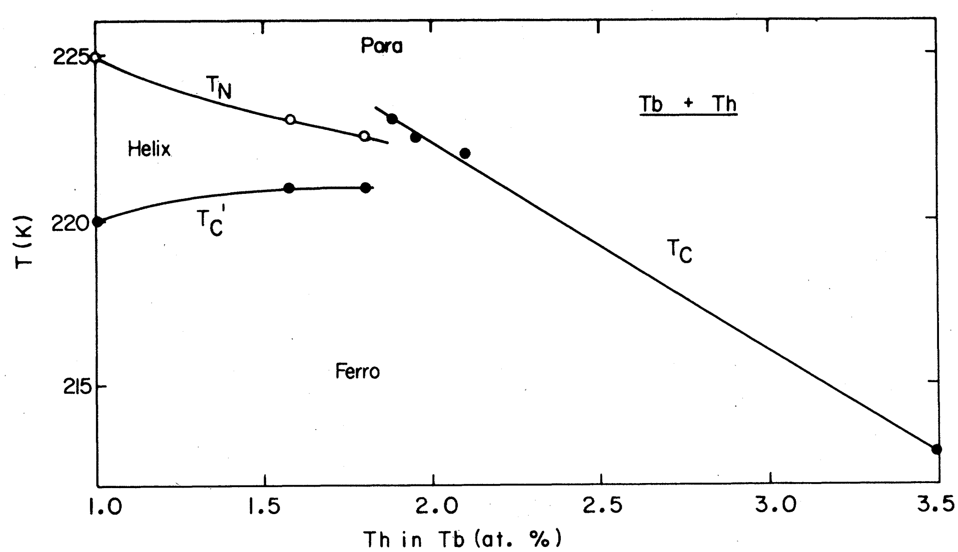

FIG. 4. Partial magnetic phase diagram for Tb-Th alloys.

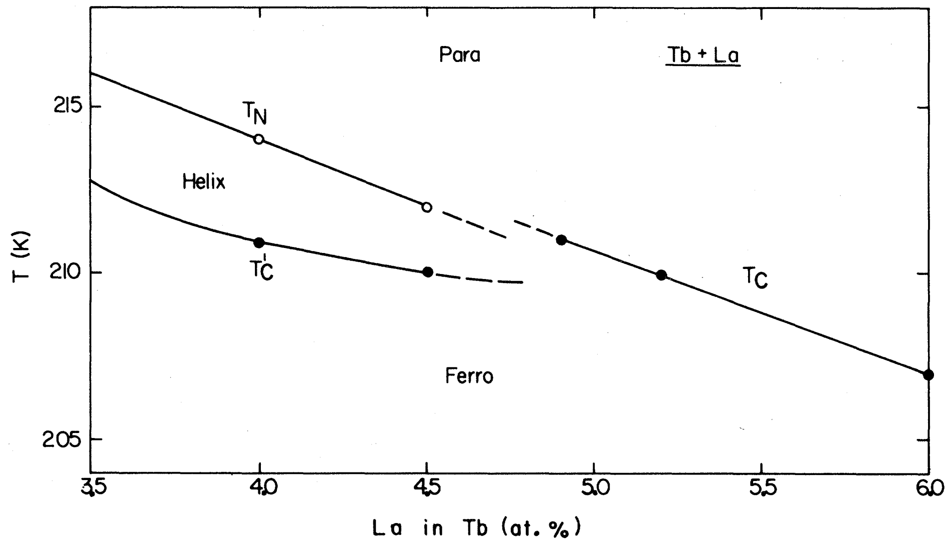

FIG. 5. Partial magnetic phase diagram for Tb-La alloys.

for this system is shown in Fig. 6. Here a temperature "gap" of nearly 1 K occurs near a concentration of 7 at.% of Gd in Tb. The results here are consistent with those for the other alloys.

One alloy system with Dy as the major constituent was investigated. The results for Th in Dy, as shown in Fig. 7, are very much like those for Tb-Th with the basic difference related to the need for about 12 at.% Th to suppress the much broader helical temperature range of pure Dy (219.5 to 89 K). The jump in ordering temperature is indicated to be over 1 K near the 12 at.% Th critical composition.

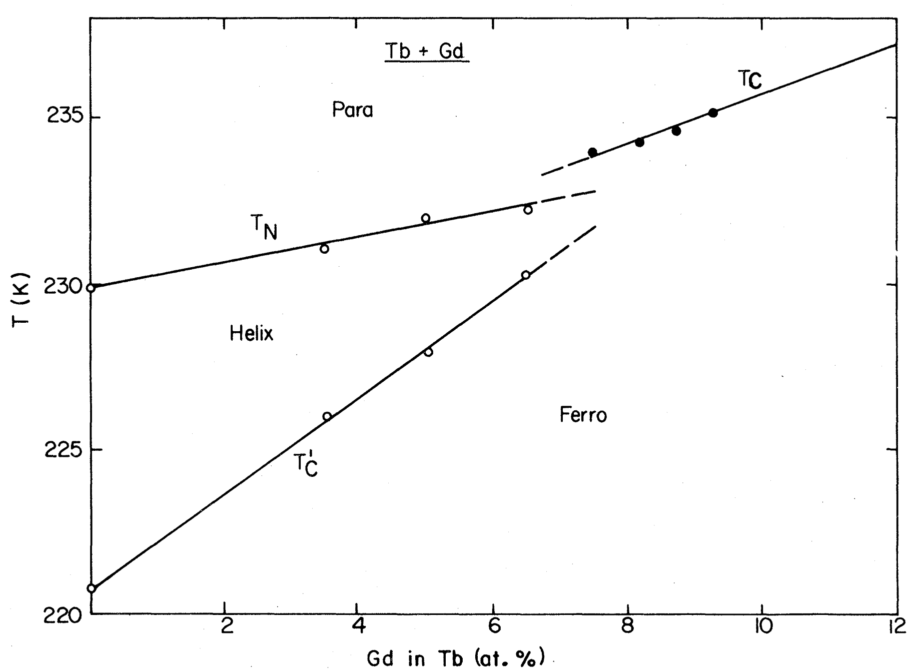

FIG. 6. Partial magnetic phase diagram for Tb-Gd alloys.

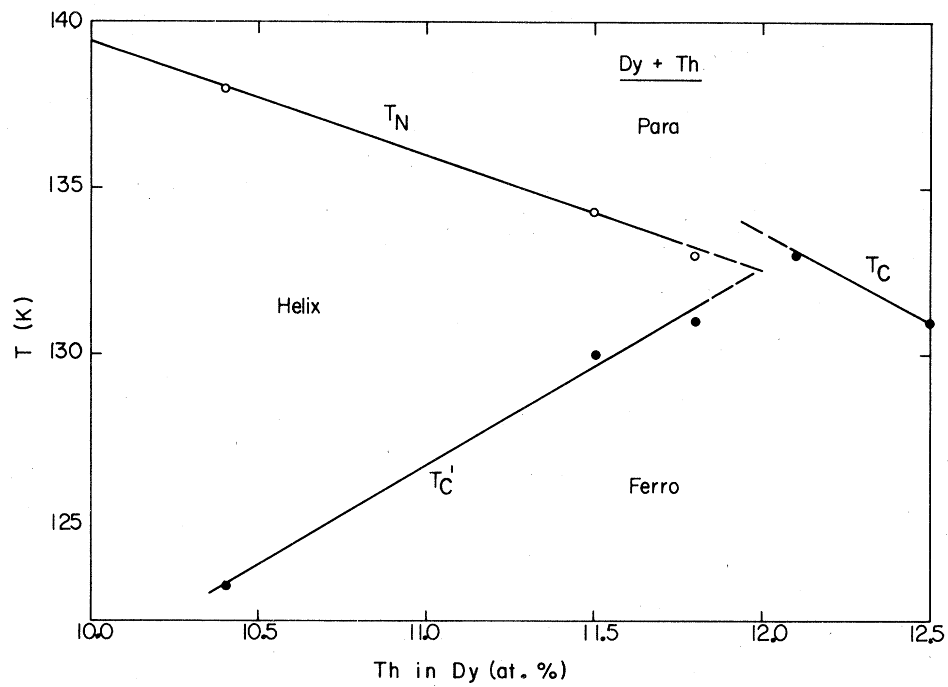

FIG. 7. Partial magnetic phase diagram for Dy-Th alloys.

III. DISCUSSION

The long-range RKKY (Ruderman-Kittel-Kasuya-Yosida) indirect-exchange interaction energy has the form

$$E = (A/R^4)[2\vec{K}_F\cdot\vec{R}\cos 2\vec{K}_F\cdot\vec{R} - \sin 2\vec{K}_F\cdot\vec{R}] ,$$

where A is a constant, $\vec{R}$ is a neighbor-atom position vector, and $\vec{K}_F$ is the conduction-electron wave-vector momentum evaluated at the Fermi surface. When $\vec{K}_F\cdot\vec{R}$ is small the interaction energy is proportional to $-1/\vec{K}_F\cdot\vec{R}$ and ferromagnetism is favored out to the first zero of the oscillatory energy function, E. It has been proposed[1,5] that the nearest-neighbor interaction for rare earths is ferromagnetic while that for next-nearest neighbors is antiferromagnetic. We assume then that for nearest neighbors at $\vec{R}_1$, E is negative and that for next-nearest neighbors at $\vec{R}_2$, E is positive. The two terms must be comparable in magnitude at those alloy compositions under scrutiny here. This being the case we may understand the behavior of the Gd-Lu alloys in a qualitative way as follows: as the temperature is lowered from well above the ordering temperature, the conduction-electron mean free path will be short and increase with decreasing temperature. A short mean free path will favor the nearest-neighbor interaction; i.e., less scattering will take place to spoil the nearest-neighbor interaction than will occur for the longer next-nearest-neighbor interaction. It follows that as the temperature goes down, the magnetic moment grows as short-range ferromagnetic order sets in. This suggests that for those alloys which eventually go antiferromagnetic it is necessary for the positive next-nearest-neighbor interaction term $E(\vec{K}_F\cdot\vec{R}_2)$ to overtake the negative nearest term $E(\vec{K}_F\cdot\vec{R}_1)$ and this makes the Néel point fall below what would have been a Curie point had not nearest-neighbor interactions grown large enough to dominate the ordering. This behavior is illustrated by the data for 24 and 25 at.% Lu in Gd as shown in Fig. 2. For the 24 at.% Lu sample there is an upper inflection point near 223 K which would have been interpreted as a Curie point, T_C, had not antiferromagnetic ordering occurred at about 221 K. The corresponding upper inflection point for 25 at.% Lu falls near 222 K. These upper inflection-point temperatures would fall very close to an extension of the Curie temperature curve, T_C, of Fig. 1. This observation seems to be universal.

It thus appears unlikely that, for systems such as those described here, a Lifshitz point will be found. Results of neutron scattering experiments[2] on rare-earth metals and alloys have not revealed turn angles for helical structures smaller than 18°. This is in keeping with the results and conclusions reported here.

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