Magnetic Properties of Heavy Rare Earths Diluted by Yttrium and Lutetium*

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A neutron-diffraction study of the heavy rare earths Tb, Dy, Ho, Er, and Tm diluted with yttrium and of Tb with lutetium is reported. The modulated antiferromagnetic structures of the rare-earth metals are found to exist in the alloys, but the ferromagnetic phases of Tb and Dy are destroyed with small admixtures of Y. The Néel temperatures of the alloys and the pure metals are found to be a universal function of the average of the square of the spin projection on J, given for these heavy-rare-earth alloys by $\xi=c(g-1)^2 \times J(J+1)$, where c is the atomic concentration of rare earth. The value of the interlayer angle ω at T_N which is related to the wavelength of the modulation of the magnetic structure is also found to be a universal function of ξ, and the temperature variation of ω decreases with decreasing ξ so that ω approaches a temperature-independent value of about 50° per layer for small ξ, regardless of the magnetic ion in the alloy.

I. INTRODUCTION

As part of a study of rare-earth systems we have investigated alloys of the heavy rare earths with yttrium. These alloys show the effects of dilution by a nonmagnetic element on the magnetic interactions between the rare-earth constituents. Neutron-diffraction studies of the heavy-rare-earth metals have shown the rather complicated character of their magnetic-structure properties. Data for the rare-earth metals Tb to Tm[1] are summarized in Table I. Despite some differences in detail, the major characteristics of the structures are similar in all five elements. They become antiferromagnetic at a temperature T_N, ordering into some form of a modulated moment distribution with a wave vector τ along the c axis of the hcp crystal. The period of the modulation, which is conveniently characterized by the angle ω between the moments in adjacent planes along the c axis, is temperature-dependent except for Tm. For the modulated moment or antiphase-domain structure, ω is the "equivalent turn angle," $\tau c_0/2$. As the temperature is lowered, another transition occurs either to a classical ferromagnet or to a modification of the antiferromagnetic structure in which a ferromagnetic component is present. The values ω_i, ω_f', and ω_f given in Table I are the values of the turn angle at the Néel temperature, just above the transition to ferromagnetism, and at low temperature, respectively. Considerable theoretical effort has now gone into obtaining a better understanding of the magnetic properties of these metals. There are, however, many aspects of the properties which are not yet well understood, and it was felt that a study of the alloys of the rare earths might help to provide a more complete understanding.

Thoburn *et al.*[2] made magnetization studies on the Gd-Y system some years ago and observed that the magnetization curves for some of the alloys were characteristic of antiferromagnetic substances in contrast to the ferromagnetism of Gd[3] itself. In fact, dilution with Y appeared to produce structures in the alloys similar in many respects to the structures of the pure metals following Gd in the series. Concurrently with portions of the present work, Weinstein *et al.*[4] studied the magnetization of the Dy-Y, Tb-Y, and Ho-Y systems and found that dilution caused a suppression of the spiral to ferromagnet transition of Tb and Dy similar to the suppression of the ferromagnetism of Gd in the Gd-Y system. Schmidt[5] has studied the resistivity of rare-earth-alloy systems, and his results indicate a similar effect.

The $4f$ rare earths are usually regarded as localized $+3$ ions imbedded in a sea of almost-free conduction electrons. At least for the metals with more than half-filled shells in the second half of the series, the small spatial extent of the f shell would suggest that direct-exchange effects are relatively small. Therefore, some form of indirect exchange must be present to explain the observed cooperative magnetic phenomena. This indirect-exchange interaction is generally considered[6] to act through the conduction electrons of the metal and to be the type proposed by Ruderman and Kittel,[7] and extended by Kasuya[8] and Yosida.[9] This Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction is long range, oscillates in sign as a function of distance, and

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TABLE I. Magnetic structures of pure rare earths.[a]

Rare earth	ξ	θ_p(°K)	T_N(°K)	Structure	T(°K)	Structure	T_C(°K)	Structure
Tb	10.5	237	228	Spiral $\mu\perp$ to c $\omega_i=20.5°$		$\omega_f'=18°$	220	Ferromagnetic $\mu\perp$ to c $\omega_f=0°$
Dy	7.08	153	179	Spiral $\mu\perp$ to c $\omega_i=43°$		$\omega_f'=26.5°$	87	Ferromagnetic $\mu\perp$ to c $\omega_f=0°$
Ho	4.50	87	133	Spiral $\mu\perp$ to c $\omega_i=50°$		$\omega_f=36°$	20	Spiral $\mu\angle$ to c Ferromagnetic Component $\parallel$ to c
Er	2.55	40	80	Modulated moment $\mu\parallel$ to c $\omega_i=51°$	53	Modulated moment gradually squaring up $\mu\parallel$ to c axis $\mu\perp$ to c spiral $\omega_f=43°$		$\mu\parallel$ to c ferromagnetic $\mu\perp$ to c spiral
Tm	1.17	19	56	Modulated moment $\mu\parallel$ to c $\omega_i=51°$, $\omega_f=51°$		Gradual change to antiphase domain		Antiphase domain in the sequence $++++---$

[a] See Ref. 1.

leads naturally to the existence of the long-range modulated structures.

However, in contrast to the observation, the calculated value of ω is about 50° for the RKKY interaction and is constant with temperature. This calculation was made for the hcp structure of the heavy rare earths and assumed the free-electron model for the energy bands and the conduction-electron wave functions with three conduction electrons per atom. This discrepancy suggested that further study would be advisable, and diluted rare-earth solid solution with yttrium were selected as the media for this study. Yttrium forms continuous solid solutions with all the heavy rare earths, possesses no f electrons, and has a similar outer-electron configuration. Since the interaction is long-range, structure-dependent, and primarily governed by the nature of the conduction band, yttrium seemed to be an ideal diluent.

II. SAMPLE PREPARATION AND EXPERIMENTAL PROCEDURE

The alloys were prepared from high-purity rare-earth and yttrium metals which were mixed together in the proper proportions and then arc-melted several times to ensure uniformity. If the sample showed any weight loss after melting, it was analyzed to determine its atomic concentration. In all cases complete solid solubility was found, and no evidence for multiphase specimens was detected by either neutron or x-ray diffraction. The alloys were studied as finely machined chips or as solid as-cast rods. In several cases, the rods showed preferred orientation, and one of these, a 10 at.% Tb-90 at.% Y sample, showed such a high degree of preferred orientation that an attempt was made to cut a single crystal grain out of this rod. A grain was obtained which, while not of sufficiently high quality to allow quantitative single-crystal analysis, did substantiate the structure deduced from the powder pattern. The four specimens of Tb-Lu alloys were provided to the authors by Spedding and Daane of the Ames Laboratory in the form of cylindrical solid rods, and the authors' gratitude for their loan is acknowledged.

Powder neutron-diffraction patterns were obtained from the samples at temperatures from 4.2°K to room temperature. A cryostat was used which allowed the sample to be maintained at almost any temperature in this range by using liquid helium or liquid nitrogen as coolants and heating the sample relative to the bath by a small electric heating coil. An exchange gas chamber in the cryostat provided a thermal switch which, when evacuated, would effectively isolate the sample from the coolant. The temperatures were measured by a calibrated copper-constantan thermocouple mounted at the sample.

The presence of antiferromagnetic long-range order of an oscillatory type was indicated by the occurrence of satellite reflections of the allowed nuclear peaks. The presence of a satellite of the origin (000±) occurring at very small angles was taken as evidence for the spiral structure; for a modulated moment structure, it is forbidden.[10] The separation of the magnetic satellites from their respective nuclear positions was used to obtain the values of the turn angle ω. Satellites of the (102) and of the origin were used primarily because these satellites occurred in a region of the pattern which did not have other reflections. The presence of the (102) satellites, combined with the absence of the satellite of the origin, was taken as evidence of a modulated moment structure such as is found in the high-temperature form of Er and in Tm. The values of the transition temperatures T_N were measured by following the in-

[10] W. C. Koehler, Acta Cryst. 14, 535 (1961).

tensity of one or more of the magnetic satellites as a function of temperature. The Curie temperatures T_C were measured from the temperature dependence of magnetic intensity which occurred at the allowed nuclear positions.

III. RESULTS AND DISCUSSION

Thirty-three alloy specimens from the systems Tb-Y, Dy-Y, Ho-Y, Er-Y, Tm-Y, and Tb-Lu have been studied in this investigation, and the most significant results are given in Table II. The concentrations of the alloys are given in atomic percent. The transition temperatures above 30°K are accurate to about ±3°K, those below 30°K to about ±5°K, and the interplanar turn angles are accurate to about ±1 degree except when otherwise noted. The quantity ξ listed in the table is $\xi=c(g-1)^2J(J+1)$, the average of the square of the projection of S on J, where c is the atomic concentration of the rare earth with angular-momentum quantum number J and Lande splitting factor g. This quantity is therefore the effective S_J^2 for the alloy.

At the temperature denoted by T_N, the alloys order into an oscillatory antiferromagnetic structure which is the same type as that of the parent rare earth. It is modulated along the c axis of the crystal, and the Tb, Dy, and Ho alloys exhibit spiral structures, while Er and Tm alloys have modulated c-axis component structures. Furthermore, the direction of the major component of the moment, insofar as it is either parallel or perpendicular to the c axis, found in the pure metals, is maintained in the alloys within the accuracy afforded by the powder-diffraction technique. By this method, it is not possible either to prove that the moment direction in the base plane is the same or to rule out the possibility of a small difference in the angle between the cone containing the moments and the c axis.

Figure 1 shows the Néel temperatures of the alloys and the pure metals plotted versus the parameter ξ.

TABLE II. Observed parameters for the magnetic structures of rare-earth yttrium and lutetium solid solutions.

Sample	ξ	T_N (°K)	T_H (°K)	T_C (°K)	ω_i (deg)	ω_f' (deg)	ω_f (deg)
Terbium[a]	10.5	228		218	20.5	18.0	0.0
90% Tb-10% Y	9.45	211		175	28.0	22.0	0.0
80% Tb-20% Y	8.40	196		99	32.0	24.0	0.0
75% Tb-25% Y	7.88	188			34.0		24.0
70% Tb-30% Y	7.35	183			40.0		31.0
60% Tb-40% Y	6.30	169			42.0		35.5
50% Tb-50% Y	5.25	149			43.5		41.5
40% Tb-60% Y	4.20	129			47.0		47.0
30% Tb-70% Y	3.15	111			49.0		49.0
20% Tb-80% Y	2.10	85			48.0		48.0
10% Tb-90% Y	1.05	50			49.0		49.0
5% Tb-95% Y	0.53	25±5			50±2		50±2
95.3% Tb-4.7% Lu	10.0	220		180±10	16.5	19.0	0.0
66.7% Tb-33.3% Lu	7.00	166			36.0		29.5
46.7% Tb-53.3% Lu	4.90	129			41.5		40.0
23% Tb-77% Lu	2.41	80			45.5		45.5
Dysprosium[a]	7.08	179		87	43.0	26.5	0.0
95% Dy-5% Y	6.73	168			43.0		28.0
90% Dy-10% Y	6.37	163			44.0		34.0
80% Dy-20% Y	5.66	152			41.0		33.5
70% Dy-30% Y	4.96	134			42.0		37.0
60% Dy-40% Y	4.25	117			44.0		41.5
50% Dy-50% Y	3.54	108			46.0		44.0
40% Dy-60% Y	2.83	94			48.0		45.5
20% Dy-80% Y	1.42	59			48.0		49.0
Holmium[a]	4.50	133		20[b]	50.0		36.0[c]
70% Ho-30% Y	3.15	103			48.0		46.0
50% Ho-50% Y	2.25	80			48.0		47.0
30% Ho-70% Y	1.35	57			50.0		50.0
10% Ho-90% Y	0.45	26±5			49±2		49±2
Erbium[a]	2.55	80	53[d]	20[b]	51.0		43
90% Er-10% Y	2.30	78±5	40±10[d]		50±3		50±3
70% Er-30% Y	1.79	65	44±5[d]		50.5		50.5
50% Er-50% Y	1.28	56	27±5[d]		51.5±2		50.5
30% Er-70% Y	0.77	30±5	15±8[d]		50±3		50±2
Thulium[a]	1.17	56			51.0		51.0
55% Tm-45% Y	0.64	37			51.0		51.0
14.9% Tm-85.1% Y	0.17	20±8			51±3		51±2

[a] See Ref. 1.
[b] Transition temperature for ferromagnetic component along c axis only.
[c] 30.0° in one sample.
[d] Transition temperature for helical structure of base plane components.

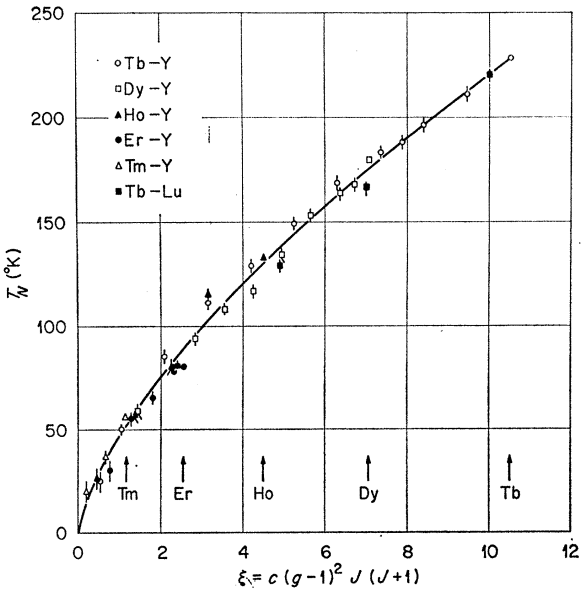

FIG. 1. Néel temperatures of rare-earth yttrium and lutetium alloys as a function of the effective spin variable ξ. The solid curve represents a $\xi^{2/3}$ dependence normalized at pure Tb.

These ordering temperatures follow a universal curve as a function of ξ to a very good approximation. The solid curve represents a $\xi^{2/3}$ dependence normalized at pure Tb, and the fit with experiment is quite good. Figure 2 shows a plot of the initial turn angles ω_i (values at T_N) versus ξ for the alloys and the pure metals. These values also lie on a single curve as a function of ξ but with considerably more scatter of the points. This scatter appears to be greater than the experimental error in the data, although ω_i must be measured just below the transition temperature where the magnetic intensities are weak, and may be partially due to a variation of cold work or lattice defects in the samples. Nevertheless, the general trend is evident. The initial turn angle ω_i saturates at a value of about 50° per layer at small ξ and drops off from this value as ξ increases. Pure Ho and Dy and the Dy-Y alloys with large ξ appear to have values of ω_i higher than the average, but they join the curve as the concentration of the rare earth is decreased. The Tb-Lu alloys approach a saturation value of ω_i two or three degrees smaller than the others for small ξ, but the same qualitative behavior is present in this system.

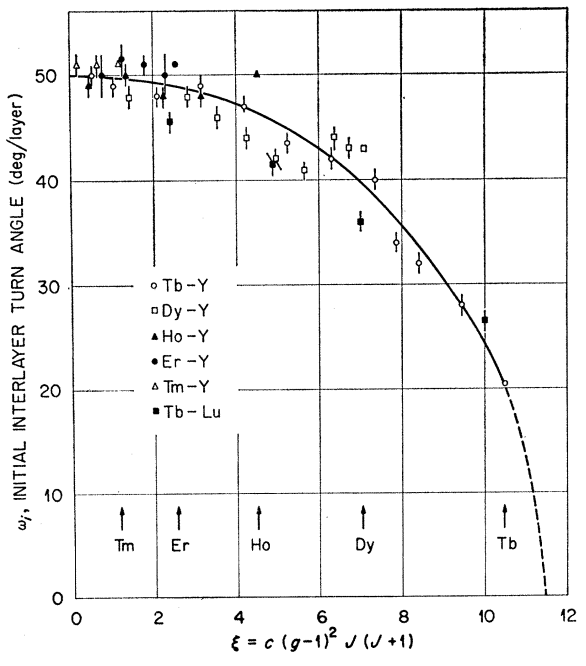

FIG. 2. Initial interlayer turn angle of the oscillatory magnetic structure of the alloys versus ξ.

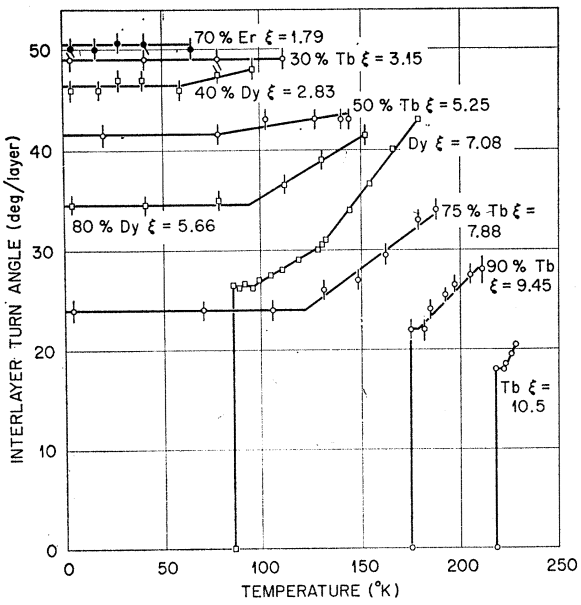

FIG. 3. The temperature variation of ω for some rare-earth yttrium alloys and pure metals at selected values of ξ.

Figure 3 illustrates the temperature variation of ω for some of the alloys and pure metals for certain selected values of ξ which were chosen so as to illustrate the regions in which significant differences exist. At large ξ, the turn angles first decrease linearly with temperature down to an apparently well-defined temperature then remain constant to low temperature or until the material becomes ferromagnetic. As the ξ gets smaller, the same trend is present, but the change in ω decreases until finally, at sufficiently small ξ, ω remains constant at the value of about 50° per layer. This qualitative behavior is observed regardless of the rare earth present in the alloy, although the value of ξ which is small enough to produce a temperature-independent ω does depend to some extent on the rare earth. For Tb alloys, a value of ξ less than about 5.0 produces an ω which is constant within the experimental accuracy, whereas the 40%

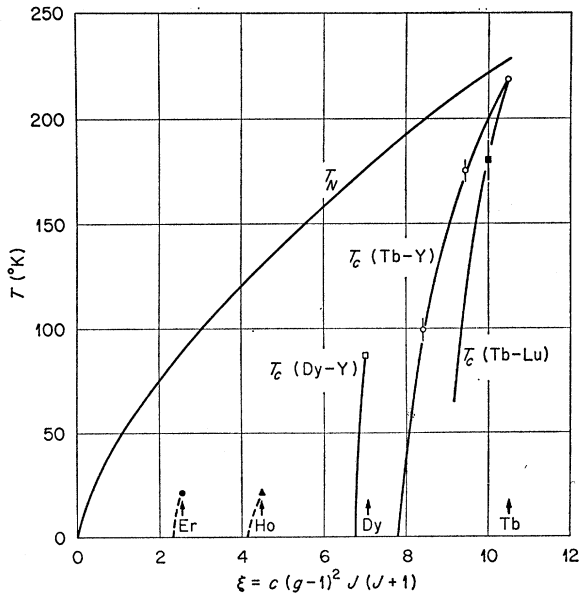

FIG. 4. T_C versus ξ for the alloys and pure metals. T_C for Ho and Er represents the appearance of ferromagnetic components only.

Dy alloy with $\xi=2.83$, shown in the figure, still has a visible change in its turn angle as the temperature is lowered. Although Ho and Er do have a temperature variation of turn angle, these materials, as well as Tm, have such small values of ξ that no variation of ω with T is observed in the alloys of these metals with Y which have been examined. No correlation between the final values of the turn angle of the modulated structure ω_f (or ω_f' for those alloys which become ferromagnetic) and the ξ value is apparent, except insofar as ω_f becomes equal to ω_i for small ξ. For example, a 75% Tb alloy with $\xi=7.88$ has a final turn angle of 24° per layer, and a 70% Tb alloy with $\xi=7.35$ has a final turn angle of 31°, while Dy with $\xi=7.08$ has a final turn angle lying between these two at 26.5°.

The Curie temperatures do not appear to be correlated with ξ, except that, for ξ less than about seven, transitions to ferromagnetism are not observed, although a ferromagnetic component does occur for some of the pure metals with ξ less than this value. Figure 4 shows T_C versus ξ for the alloys and the pure metals. The details of the order-order transition from oscillatory to ferromagnetic structure remains uncertain experimentally, but there is a region of temperature in which both types of reflections are simultaneously observed, and the change in ω from ω_f' to 0 is discontinuous with the spiral reflections decreasing in intensity but not changing their positions as the ferromagnetic reflections grow. These facts cause some ambiguity as to just what temperature should be called T_C. The procedure adopted here is that T_C is the temperature at which ferromagnetic intensity appears, even though antiferromagnetic reflections still exist for some range of temperature below T_C. In contrast to the relatively small change in T_N, the Curie temperatures of Dy and Tb are very drastically affected by dilution. As little as 25% Y in Tb lowers the Curie temperature to below 4°K, while only 5% Y in Dy destroys the ferromagnetic transition. Thus, the temperature interval in which the spiral is more stable than the ferromagnetic phase has been increased from 92°K at $\xi=7.08$ for Dy to at least 164°K for the 95% Dy alloy with $\xi=6.73$. In contrast to this reduction, the Néel temperature has been reduced only about 11°K for this change in ξ of 0.35. A 90 and 95 at.% Dy alloy has been studied by Weinstein[4] by magnetization techniques, and he had concluded that these alloys were ferromagnetic. Because of this, the 95% Dy alloy was examined in a diffractometer which allowed an external magnetic field to be applied to the sample. The results confirmed the existence of a ferromagnetic phase in an applied magnetic field and a spiral phase in zero applied field. Furthermore, the ferromagnetic phase showed complete remanence at low temperature when the field was removed. The critical field required to produce ferromagnetism was found to be very temperature sensitive

There is no evidence in any of the alloys of Y with Ho, Er, or Tm that any ferromagnetic component exists. These metals do not become completely ferromagnetic in the pure state, but they all develop a net ferromagnetic component at low temperatures. Although a small ferromagnetic component such as is seen in Ho and Tm would probably not be detectable

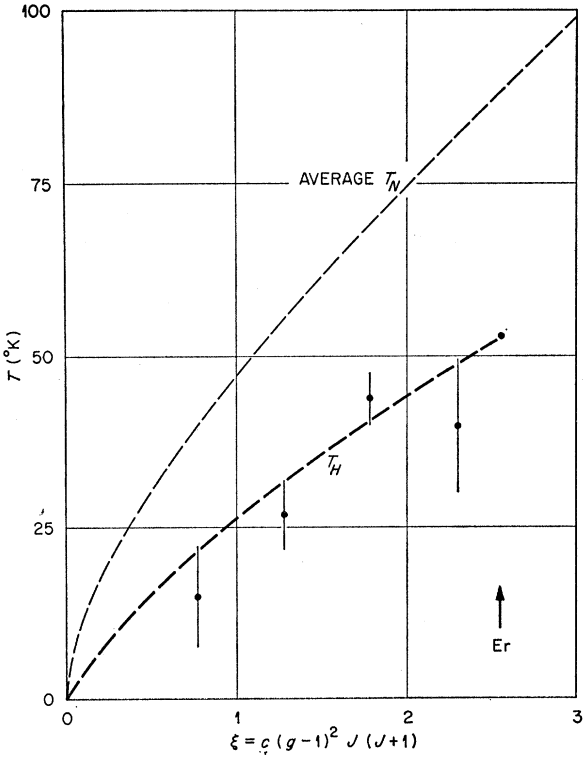

FIG. 5. The transition temperature at which the base plane component orders into a spiral structure for Er and Er-Y alloys.

in these powder-diffraction studies of the alloys, the relatively large one in Er would certainly have been seen if it had been present.

While the ferromagnetic component present in pure Er does not appear in the alloys, the ordering of the base plane components into a spiral structure at a temperature denoted by T_H does occur in the Er-Y alloys. Figure 5 illustrates the variation of T_H versus ξ, and the variation is about the same as that of T_N. When the base plane components order into a spiral in pure Er, the c-axis component begins to square up, and the turn angle begins to change with temperature. Neither of these effects is observed in the alloys, and although it would not be possible to detect the squaring up of the c-axis component, since this would require the observation of the higher odd-harmonic reflections, the change of ω would have been observable if it had occurred.

There is evidence in some of the diffraction patterns for some minor modifications of the antiferromagnetic structures at low temperatures. In many cases, an anomalous small-angle scattering is observed that does not peak at the transition temperature as it would if due to short-range order scattering. Instead, it increases with decreasing temperature. Also, some of the alloys show additional weak reflections which can only be seen near the satellite of the origin because of their small intensity. These reflections appear to be harmonics of the fundamental magnetic period but do not correspond to a simple squaring up of the structure since the 2nd harmonic is observed, not just the odd ones. In a few cases, there appears to be a harmonic inside the fundamental which would correspond to a longer wavelength modulation than the fundamental. These effects may be spurious, due perhaps to double Bragg scattering, but even if real, they represent only minor modifications of the magnetic structure.

Since the structure in the high-temperature ordered phase of the alloys is essentially the same as the parent rare-earth metal, the dilution has apparently not greatly altered the character or range of the interaction. The magnitude of the exchange interaction as measured by the transition temperature is found to be a universal function of the parameter ξ, and it is quite well represented by a $\xi^{2/3}$ dependence, while the magnetic exchange energy due to the RKKY interaction is proportional to ξ to the first power. If the solid curve in Fig. 1 is extended to Gd at $\xi=15.75$, the predicted ordering temperature is 300°K, 3.4% higher than the observed value of 290°K. Furthermore, the transition temperatures of the Gd-Y alloys given in Ref. 2 follow the curve fairly well. Apart from its dependence on ξ to the first power, the RKKY interaction should be independent of the dilution except for neglected second-order or mean-free-path effects, since it is primarily determined by the crystal structure and the number of free electrons per atom, both of which should be unchanged in the alloy. Since the moment directions of the pure metals relative to the c axis of the crystal are retained in the alloys within the accuracy of the experiments, the anisotropy constants apparently have not been changed relative to one another by the dilution. In the approximation used thus far in the theory, the anisotropy energy would be completely independent of the dilution, since this energy was calculated for the rare-earth ion in the crystal field due to +3 point charges at the nearest-neighbor positions.

The initial turn angles ω_i are found to be primarily determined by the value of ξ for the specimen, and the limiting value of 50° is observed regardless of the magnetic ion in the alloy. Extrapolation of the data to ξ values larger than the value of 10.5 for Tb indicates that ω_i would be zero for ξ greater than about 11.5. Therefore, alloys with such values of ξ would be expected to be ferromagnetic, and this is observed for Gd-Y[2] and Gd-Dy[11] alloys and for Gd[3] itself from magnetization data. Alloys containing Gd have not been studied by neutron diffraction because of the very high absorption of thermal neutrons by natural Gd. The temperature variation of ω decreases with decreasing ξ, so that for sufficiently small ξ values ω is constant at the value of 50°. The value of ξ which is small enough to produce a temperature-independent ω depends on the rare earth in the specimen, and, from the limited comparison possible between Y and Lu alloys, the limiting value of ω depends on the diluent to some extent. Since the RKKY interaction predicts a temperature-independent ω of about 50 deg per layer, these data indicate that only when the effective ion spin is small is the ideal RKKY behavior observed.

Although the initial turn angles and transition temperatures of the metals and the alloys are simply related to the values of the effective spin variable ξ, there are distinct differences in their behavior at temperatures below the initial ordering transition. Whereas the pure metals become ferromagnetic (or have a ferromagnetic component) at reduced temperatures, this property is strongly suppressed in the alloys. Even the metals whose magnetic structures contain only a ferromagnetic component lose this component upon dilution by yttrium. The degree of suppression of the ferromagnetic phase per atomic percent diluent is found to depend on the rare earth involved and on the diluent used, since the change in T_C is greater in the Dy-Y system than in the Tb-Y system and greater in Tb-Lu than in Tb-Y. The fact that spiral reflections still exist for some range of temperatures below T_C suggests that two magnetic phases are coexisting in the alloy over this range.

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