Magnetostriction Due to Rare-Earth Impurities in a Metal

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The magnetostriction of dilute alloys of rare-earth impurities in YPd_3 follows closely the simple H/T dependence of the field-parallel component of a quadrupole moment on the free rare-earth ion moving with the magnetic moment. The saturated magnetostriction per impurity ion is as large as in the magnetically ordered state of rare-earth compounds but does not trace the Stevens factor across the rare-earth series.

The screening of the valence electrons from the nuclear charge is spherically isotropic throughout the Periodic Table except in the rare-earth (RE) elements where it must be distinctly aspherical. In these elements the deep-lying $4f$ shell ($r_{4f} \simeq 0.1 r_I$; r_I is the ionic radius) is very effective in screening the $6s5d$ valence electron shell from the nuclear charge. In contrast to other deep-lying shells, the $4f$ charge distribution is aspherical whenever it carries net orbital angular momentum. One therefore expects a special asphericity of the $6s5d$ RE valence charge distribution due to aspherical $4f$ screening. This screening asphericity (SA) can be distinguished from other asphericities (e.g., that of the $5d$ electron): It must move with the $4f$ magnetic moment $\vec{\mu} = g_J \mu_B \vec{J}$ because the geometrical shape of the $4f$ shell is rigidly tied to its orbital angular momentum vector $\vec{L}$ and this in turn to the spin vector $\vec{S}$ through strong spin-orbit coupling.

A crude estimate of the SA can be extracted from the well-known contraction of the ionic radius, $\Delta r_I \simeq 0.1 r_I$, when the number of $4f$ electrons decreases by one.[1] The difference of the RE radii in two azimuthal directions through a maximum and through a node of the $4f$ charge distribution should be

$$\Delta r_{SA} \simeq \Delta r_I (r_{4f}/r_I)^2 \simeq 10^{-3} r_I . \quad (1)$$

The magnitude and the sign of the SA depend on the $4f$ Hund's-rule ground state and on the electric polarizability of the $6s5d$ shell, which in turn depends on its configuration and environment. In analogy to the role of the ionic radius in determining the lattice constant of RE compounds[1] this SA should cause an *aspherical distortion of the lattice around each RE site* which should be in first order quadrupolar, about 10^{-3} in magnitude, and should move with the RE magnetic moment.

The giant magnetostriction (MS) of RE elements and compounds in the ferromagnetic state, which is of order 10^{-3},[2] and especially the striking correlation of its temperature dependence with that of the magnetization through the so-called single-ion theory[3] seems to support the above picture. Yet to our knowledge the idea of an aspherical RE ion straining the lattice through a permanent quadrupole moment tied to its magnetic moment has never been put to an important test: the MS of a metallic alloy with dilute RE impurities. In such an experiment one can measure without interference from ion-ion interactions the magnitude and sign of the quadrupole moment, if it exists. Knowledge of this quantity and its effect on the lattice would obviously have important implications for crystal-field physics, magnetic order, and electron-phonon coupling in RE metals.

Let us assume that the MS of metal with dilute RE impurities is proportional to the thermal average of a quadrupole moment of the free impurity valence-electron cloud moving with the $2J+1$ degrees of freedom of the $4f$ shell. One then has

$$\left.\frac{\Delta l}{l}\right|_\alpha = x Q_{J\alpha} \frac{\sum_M [3M^2 - J(J+1)] \exp(-M\chi)}{J(2J-1)\sum_M \exp(-M\chi)}$$

$$= x Q_{J\alpha} (2J-1)^{-1} [2(J+1) - 3 m_J(\chi) \coth(\tfrac{1}{2}\chi)]$$

$$\equiv x Q_{J\alpha} q_J(\chi) . \quad (2)$$

Here $\chi \equiv g_J \mu_B H/k_B T$, x is the impurity concentration, α is the angle between Δl and H, and $Q_{J\alpha}$ is the saturated quadrupole moment. The other symbols have their usual meaning. Figure 1 shows the H/T dependence of the normalized magnetization $m_J(\chi)$ and the normalized quadrupole moment $q_J(\chi)$ for free Dy^{3+} ions.

In this Letter we wish to report measurements of the MS of a series of dilute alloys of the type $RE_x Y_{1-x} Pd_3$. We chose YPd_3 as the matrix for the following reasons: (i) YPd_3 is a simple diamagnetic metal[4] in which RE impurities dissolve

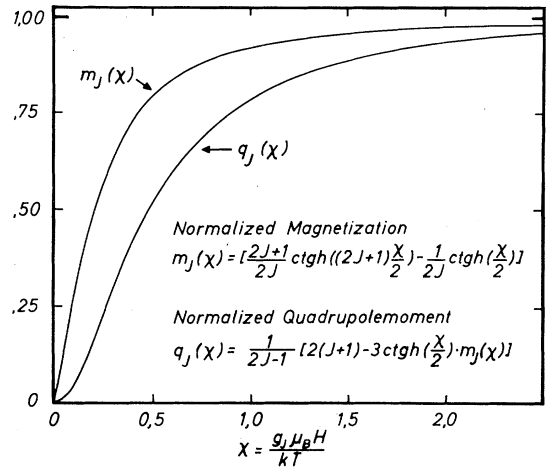

FIG. 1. The H/T dependence of the normalized field-parallel magnetization $m_J(\chi)$ and quadrupole moment $q_J(\chi)$ for free Dy^{3+} ions.

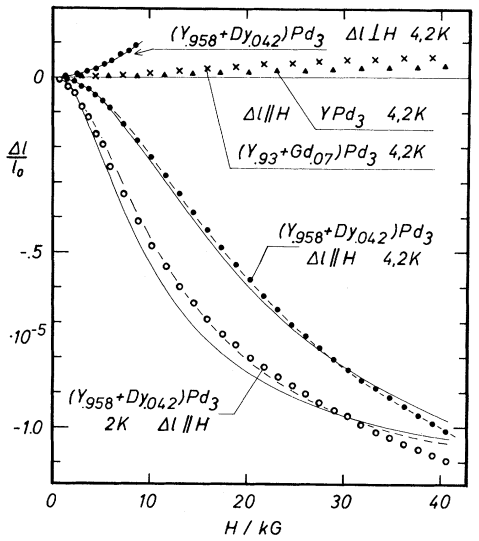

FIG. 2. Magnetostriction of the matrix YPd_3 and alloys with Dy and Gd. The data are taken at the indicated temperatures. The full lines are fits according to Eq. (2) with $T = 4.2$ and 2.0 K and the dashed lines with effective temperatures $T^+ = 4.6$ K and $T^+ = 2.4$ K, respectively.

easily. (ii) Judging from the interaction temperatures of the RE-Pd_3 compounds[4] the impurity-impurity interaction temperatures should be negligible above 1 K for concentrations of a few percent. (iii) The expected reduction of the field-parallel component of the SA quadrupole moment (and the ensuing MS) caused by electrostatic interactions with the environment (crystal-field effects) should be calculated in analogy to that of the magnetization. Some information on crystal-field schemes has been extracted from magnetization measurements on all RE-Pd_3 compounds[4] and from inelastic neutron diffraction on those with RE = Pr, Nd, and Tb.[5] To estimate crystal-field effects, we hoped to be able to extrapolate from these data to the other RE compounds and to high dilution (see, e.g., Ref. 6).

The polycrystalline samples ($Y_{1-x}RE_xPd_3$ with all RE except Ce, and $La_{0.94}Ce_{0.06}Pd_3$) were prepared from 99.99%-pure Y and La, 99.9%-pure RE (Rare Earth Products), and Specpure Pd (Jonston-Mattey) by melting in an argon atmosphere. They were vacuum annealed for 7 days at 900°C and cut into rods with a diamond saw ($8 \times 1 \times 1$ mm^3). Magnetostriction measurements were made using an adjustable-gap capacitance dilatometer[7] in a He^4-bath cryostat. Our resolution was $\pm 2.5 \times 10^{-6}$ pF on a typical working capacity of 35 pF, resulting in a sensitivity of $\Delta l/l \simeq 2 \times 10^{-9}$. Longitudinal MS was measured in fields up to 40 kG using a superconducting magnet and transverse MS up to 8 kG using a standard electromagnet.

Figure 2 shows the raw data of the longitudinal and transverse MS of YPd_3 with 4.2 at.% Dy and the longitudinal MS of YPd_3 with 7 at.% Gd and of pure YPd_3. The MS of YPd_3 actually constitutes the cell background for our first runs and is to be subtracted from the other data. This background has some hysteresis, a fraction of the values shown. The MS of Dy impurities is very large, with a quadratic low-field dependence and an approach to saturation at high fields. The full lines through the data are fits according to Eq. (2), with $J = \frac{15}{2}$, $g_J = \frac{4}{3}$, and $T = 4.2$ and 2 K, the dashed lines with $T^+ = 4.6$ K and $T^+ = 2.4$ K. The dashed-line fits have a smaller rms deviation than those made using the actual measuring temperature. With these fits we obtain $Q_{J\parallel} = -3.4 \times 10^{-4}$ and $Q_{J\perp} = +1.7 \times 10^{-4}$ from the 4.2-K data and $Q_{J\parallel} = -2.8 \times 10^{-4}$ from the 2-K data. We have checked the concentration dependence of this MS and found at 4.2 K that $Q_{J\parallel} = -3.1 \times 10^{-4}$ for 2.4 at.% and $Q_{J\parallel} = -3.9 \times 10^{-4}$ for 7 at.% Dy, respectively. Extrapolating against x to zero concentration we arrive at $Q_{J\parallel} = -3.05 \times 10^{-4}$ for Dy.

The magnetization of the alloy with 4.2 at.% Dy was measured between 4.2 and 1.55 K up to 22 kG. Within the scatter of the data, the initial susceptibility follows a Curie-Weiss law ($\Theta \simeq 0.4$ K). The magnetization at higher fields lags behind the

Brillouin function by at most 15% (6% at 22 kG) at 1.55 K and by at most 8% at 4.2 K. This is consistent with an effective interaction temperature of about $\Delta T \simeq 0.4$ K, the same as for the best-fit MS curves. Similar results apply to the magnetization of $Y_{0.95}Tb_{0.05}Pd_3$ ($\Delta T \simeq 0.4$ K). The MS of the sample with Gd in Fig. 2 differs noticeably from the background. Measurements with an improved cell[8] are consistent with a fit with $J = \frac{7}{2}$ and $g_J = 2$, and gave $Q_{J\parallel} = +1.35 \times 10^{-5}$. Magnetostriction measurements like those in Fig. 2 were performed on dilute alloys of all magnetic RE impurities in YPd_3 (Ref. 8) ($LaPd_3$ in the case of Ce). In most cases (except for Pr, Eu, and Sm, where the MS is very small) we can fit the H/T dependence quite well with the free-ion expression Eq. (2). The same is true for the magnetization of all alloys except those with Pr, Sm, and Nd, which show definite and large crystal-field effects.[8] The $Q_{J\parallel}$ values (multiplied by 4 in order to refer them to the RE cell) are listed in the second column of Table I. The same values are shown in the third column, normalized to $Q_{J\parallel}$ for Dy (β_m) to facilitate comparison with the Stevens moments, also normalized to Dy and listed in the fourth column (β_S).[9] The fifth column shows the deviations ΔT of the best-fit temperatures T^+ from the temperature T of the measurement and the sixth, the rms deviations of the best fits from the data taken at 4.2 K.

The $4Q_{J\parallel}$ values are of the order of magnitude expected on the crude estimate in Eq. (1) and close to the giant values of the MS of magnetically ordering RE compounds and elements.[2] Note that $Q_{J\parallel} = -2Q_{J\perp}$ for Dy within experimental error. This implies that we are dealing with a pure shape MS. Note also that the MS from Gd is nearly 2 orders of magnitude smaller than for the other heavy RE (the $4f^7$ shell is spherically symmetric only in first order, but not in second.[10]

TABLE I. The saturated quadrupole moments for RE impurities in YPd_3: Absolute values, $4Q_{J\parallel}$; values relative to Dy, β_m (measured); and β_S (Stevens) (Ref. 9).

RE	$4Q_{J\parallel}$	β_m	β_S	ΔT (K)	rms (%)
Ce	$+4.2 \times 10^{-4}$	−0.31	+0.86	1.6	1.9
Nd	-1.24×10^{-3}	+0.91	+0.35	0.4	1.0
Gd	$+5.4 \times 10^{-5}$	−0.04	0.00	0.6	7.8
Tb	$+2.68 \times 10^{-3}$	−2.00	+1.00	0.4	6.6
Dy	-1.36×10^{-3}	+1.00	+1.00	0.4	0.9
Ho	-1.2×10^{-3}	+0.88	+0.40	0.7	1.7
Er	-1.56×10^{-3}	+1.15	−0.40	0.7	7.5
Tm	-2.44×10^{-3}	+1.79	−1.00	0.8	0.8
Yb	$+1.28 \times 10^{-3}$	−0.94	−1.00	1.6	0.8

The good quality of our free-ions fits of the quadrupole and magnetic moments is surprising. We did not expect to detect any significant impurity-impurity interactions.[11] But the near absence of crystal-field effects is quite puzzling. According to the inelastic neutron diffraction data,[5] overall crystal-field splittings in $PrPd_3$, $NdPd_3$, and especially in $TbPd_3$ are at least 5 meV and should be of similar magnitude in the other compounds (except Gd), i.e., large compared to our magnetic Zeeman and thermal energies. On the basis of some test calculations we expect the H/T dependence in such large crystal fields to be distinctly different from that of free ions in our field and temperature range. Since the MS is, on the contrary, very close to free-ion behavior, and since the magnetization of all impurities (except Sm, Pr, and Nd) is that of nearly free ions in H/T dependence and especially in *absolute value*, we are tempted to roughly estimate the overall crystal-field splittings and/or interaction temperatures of our dilute impurities (as seen through magnetization and quadrupole moments) to be of the order of the ΔT's in the table, i.e., at most a few kelvins—much smaller than in $TbPd_3$ especially. Crystal fields must of course be present. We observe, after all a reaction of the crystal to the alignment of the quadrupoles. But apparently the crystal-field energies were reduced drastically by dilution in YPd_3 in most cases. We consider this a fortunate coincidence since it allows us to exhibit the free-ion giant quadrupole so clearly.

In view of the nearly free-ion behavior of MS and magnetization, the absence of any correlation between our Q_J's and those derived from the Stevens factor (Table I) cannot be due to a partial suppression of the free-ion quadrupole effect by crystal fields. It is more likely that the complicated dependence of the SA on the polarizability of the $6s5d$ shell destroys this correlation. Calculations are in progress to clarify this point.[12] In any case, the absence of this correlation is in itself a strong indication that the observed quadrupole is not that of the $4f$ charge distribution itself. The frequent failure of the point-charge model may be due not to the wrong choice of point charges, but of quadrupoles.

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Generation and Propagation of an Intense Rotating Proton Beam

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An annular magnetically insulated diode in a cusplike magnetic field has been used to produce an ~6-kA, ~190-kV rotating proton beam. Diamagnetic signals and magnetically insulated Faraday cups indicate that (35±15)% of the beam traversed the cusp and reached the end of the system magnetic field 35 cm from the diode. Diamagnetism was in excess of 2% of the 8-kG applied field, and the beam was distinctly hollow with a 6-cm average radius.

The theoretically favorable plasma-confinement properties of field-reversed magnetic field configurations have led to many reactor proposals.[1-5] Such configurations have been experimentally realized (i) by the injection of relativistic electron beams[6] to form a reversed-field electron ring,[7] (ii) by plasma currents induced by relativistic-electron-beam injection,[8] and (iii) by reversed-field θ pinches.[9] However, synchrotron-radiation energy losses make a relativistic electron ring unsuitable for a fusion reactor. Indeed, Christofilos amended his original Astron concept[1] by replacing electrons with high-energy protons. Developments in intense-ion-beam[10-13] technology make it possible for a reversed-field ion ring to be produced by single-pulse injection.[2] This has led to two experimental programs[12,13] aimed at the injection and trapping of an ion ring in a mirror magnetic field.

In this Letter, we report experimental results on the production and propagation of a rotating beam of the type required for ion-ring formation. To summarize, an ion beam was produced with up to 40% efficiency using an annular magnetically insulating diode, and was caused to rotate by a cusplike magnetic field. Of the $\sim 4\times 10^{15}$ ions extracted from the diode, (35±15)% traversed the cusp, formed a rotating proton beam, and reached the end of the system magnetic field 35 cm from the diode. The (2–3)% diamagnetic signals (in an 8-kG applied field) are significantly larger than those previously reported.[14,11]

The apparatus used for the present experiments, including a detail of the diode region, is shown in Fig. 1. It is a cylindrically symmetric annular system, the anode surface of which is made coincident with a flux surface by properly positioning coils within the anode itself which are diamagnetic with respect to an externally applied field. The resultant magnetic field in the anode-cathode gap both inhibits the flow of electrons and imparts a rotational velocity to the protons as they cross the radial magnetic field lines both in and after the diode. For magnetic-insulation tests, the

