

TWO- AND SINGLE-ION MAGNETIC ANISOTROPY IN THE RARE-EARTH METALS

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(Invited paper)

The strong magnetic anisotropy of the rare-earth metals (basically due to the large orbital contributions to the total ionic moments) has normally been attributed to the crystalline field acting on the ions. Within the last few years a number of experiments have revealed that anisotropic two-ion couplings are of comparable importance. A review of the experimental evidence for the presence of anisotropic coupling between the rare-earth ions is given. Such indications are found primarily in the excitation spectra of the metals, but are also implicit in the crystal-field parameters determined in dilute systems.

1. Introduction

The magnetic behaviour of the rare-earth metals is basically understood by making a sharp distinction between the localized 4f-electrons carrying essentially all the magnetic moment and the conduction electrons [1]. The strong spin-orbit coupling implies that the total angular momentum, \mathbf{J} , of the (unfilled) 4f-shell is of constant magnitude, $|\mathbf{J}| = |\mathbf{L} + \mathbf{S}| = L \pm S$, where $\mathbf{S} = (g - 1)\mathbf{J}$ (g is Landé's factor). The exchange between the itinerant and the localized electrons introduces a coupling of the spins, \mathbf{S} , on different sites mediated by the propagation of the itinerant electrons. Neglecting orbital effects the so-called RKKY-coupling is isotropic in spin space, and may be written:

$$\mathcal{H} = -\frac{1}{2} \sum_{i \neq j} \mathcal{J}(\mathbf{R}_i - \mathbf{R}_j) \mathbf{J}_i \cdot \mathbf{J}_j, \quad (1)$$

where $\mathcal{J}(\mathbf{R}_i - \mathbf{R}_j)$ is proportional to $(g - 1)^2$ and the susceptibility of the conduction electrons. The assumption that \mathbf{L} is unimportant can only be justified if the product of the radial extent of the 4f-electrons and the Fermi wavevector is much less than one, which is not the case in the rare-earth metals. Augmented plane wave calculations of the properties of the conduction electrons [2] show that the wavefunctions of both the s- and d-electrons overlap the 4f-electrons, and that the Fermi-surface electrons are of mixed s- and d-character. These considerations imply that a scattering process $(4f, s) \rightarrow (4f^*, d)$ involving \mathbf{L} is very likely to occur. By this scattering process the exchange coupling is augmented by terms depending on the quadrupole moments of the ions. Calculations by Kaplan and Lyons [3] and Specht [4] based on a free-electron model indicate that this modification might be of impor-

tance. The s-d scattering also produces indirect quadrupole-quadrupole couplings effectuated by the Coulomb interaction of the conduction and the 4f-electrons [5]. In principle, both kinds of terms may also be produced by the spin-orbit coupling of the conduction electrons [6] which has a pronounced effect on the band structure [2]. In general all kinds of multipole couplings might be expected [7], restricted in form only by the symmetry. Besides the indirect couplings via the conduction electrons, the electric and magnetic multipoles are coupled directly [8], and the single-ion terms are the most important.

A projection of the couplings on to the ground state J -multiplet of the ions leads to the following general Hamiltonian:

$$\begin{aligned} \mathcal{H} = & \sum_i \sum_{l,m} V_{lm} \tilde{O}_{l,m}(\mathbf{J}_i) \\ & + \sum_{i \neq j} \sum_{l,l' > 0} \sum_{m,m'} K_{lm}^{l'm'}(\mathbf{R}_i - \mathbf{R}_j) \tilde{O}_{l,m}(\mathbf{J}_i) \tilde{O}_{l',m'}(\mathbf{J}_j) \end{aligned}$$

expressed in terms of Racah operators [9]. Only terms for which $l + l'$ are even may appear (neglecting configuration mixing), and only the exchange interaction gives rise to terms for which l (and l') are odd (besides the magnetic dipole-dipole coupling). The single-ion parameters, V_{lm} , describing the effects of the crystalline field might include contributions from the indirect couplings [3, 4]. The coupling parameters (including V_{lm}) in (2) depend on the relative positions of the ions, giving rise to magnetostriction and to interactions between the phonons and the magnetic excitations.

The first convincing evidence for the importance of anisotropic two-ion couplings in the rare-earth metals was found by Nicklow et al. [10] in 1971. They studied the dispersion relation of the spin waves propagating in the c -direction of the conical phase of Er. At about the same time Lindgård and Houmann [11] discussed the

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first indications of similar couplings in Tb and Pr. The Er results have later been reanalysed [12] and the cases of Tb [13] and Pr [14] have been examined experimentally in great detail. At present it can be concluded that anisotropic two-ion couplings produce dispersive effects on the magnetic excitations in all three metals, which at low temperatures are comparable to those caused by the isotropic two-ion coupling. A similar conclusion may be reached on the basis of the studies of Touborg and co-workers [15, 16], who have determined the crystal-field anisotropy by magnetization measurements on single-crystal alloys of rare-earth ions diluted in Y, Lu, and Sc.

2. Erbium

At low temperatures (below 18 K) the ionic moments of Er are ordered in a conical configuration [17] specified by the wavevector \mathbf{Q} (parallel to the c -direction and equal $0.24 \times 2\pi/c$) making an angle $\theta = 28.5^\circ$ with the direction of the magnetic moments. Neutron diffraction studies [17] showed the ordered moments to have the maximum value of $9.0 \mu_B$.

The dispersion relation, $\epsilon(\mathbf{q})$, of the spin waves propagating along the c -direction of Er has been measured at 4.5 K by Nicklow et al. [10]. The splitting $\Delta_q = \epsilon(+q) - \epsilon(-q)$ was clearly resolved in the experiment. If anisotropic two-ion couplings are neglected, Δ_q is a direct measure of $\mathcal{J}(\mathbf{q})$ leaving only a q -independent anisotropy constant, L_A , to be determined by the mean values of the energies [19]. The dispersion relation in Er cannot be described in this simple way. Instead, Nicklow et al. [10] were forced to introduce a giant axial two-ion term [$m = m' = 0$ in (2)] in order to fit the experimental results.

In a subsequent analysis [12] of the dispersion relations, more satisfactory quantitative results were obtained by introducing an anisotropic coupling for which $m = -m' = 2$. For instance, the L_A deduced ($=20$ meV) compare with an estimate [18] based on the magnetization data of Er showing L_A to lie between 15 and 24 meV. The anisotropic two-ion component was reduced by a factor of 20, becoming of the same order of magnitude as the isotropic coupling. The same result is obtained whether l and l' are both equal to two or both equal to three [18], the anisotropy deduced in Er may be written as an arbitrary $[a(ij)$

undetermined] linear combination of the two kinds of couplings:

$$\begin{aligned} \mathcal{K}_a = & -\frac{1}{2} \sum_{i \neq j} \mathcal{K}_{22}(ij) \left\{ \frac{1 - a(ij)}{8J_1^2} (J_i^+ J_j^-)^2 \right. \\ & + \frac{a(ij)}{21.1J_2^2} [J_z(J^+)^2 + (J^+)^2 J_z]_i [J_z(J^-)^2 + (J^-)^2 J_z]_j \\ & \left. + \text{hermitian conjugate} \right\}. \end{aligned} \quad (3)$$

z is along the c -direction and $J_n = (J - \frac{1}{2})(J - 1) \dots (J - \frac{1}{2}n)$. $\mathcal{K}_{22}(\mathbf{q})$ and the isotropic component, $\mathcal{J}(\mathbf{q})$, are shown in fig. 1.

Lindgård [20] has suggested that the spin-wave approximation might not be adequate in the case of Er, because of the quite large ratio between the anisotropy and the exchange energies. The characteristic expansion parameter appearing in the spin-wave theory [21–23] is this ratio divided by $2J$, which is small. In the random-phase theories [21–23] the conventional spin-wave parameters, appearing in zero order of $1/J$, are replaced by renormalized (effective) values. The isotropic coupling, (1), is scaled by the relative magnetization σ , but this introduces also a q -dependent term [23, 24] which behaves as if originating from an anisotropic two-ion coupling. In Er (and in Tb) this term is at least one order of

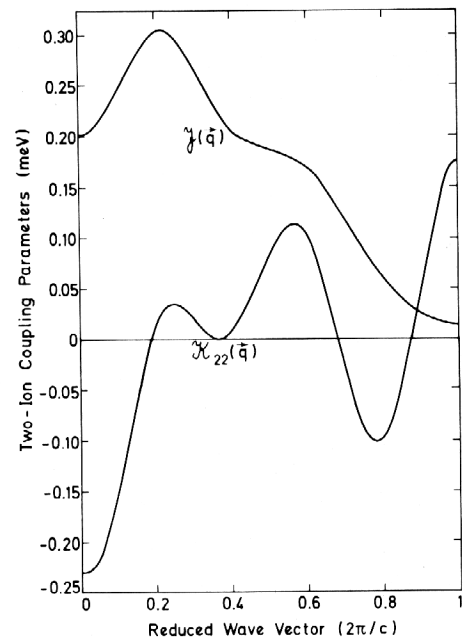


Fig. 1. The q -dependence of the two-ion coupling parameters, defined in the text, in the c -direction of Er (after refs. 12, 18).

magnitude smaller than $J\mathcal{F}(\mathbf{q})$, [24], and can be neglected. The modification considered by Lindgård [20] is produced by the molecular-field Hamiltonian. However, we find [24] that it is caused by a transformation of the Bogoliubov components which leaves the excitation energies unchanged. The applicability of the spin-wave theory in the case of Er (and Tb) at zero temperature is verified [24] by a numerical calculation of the energies of the molecular-field excitons [25]. These energies agree within a few per cent with the prediction of the spin-wave theory (when adjusting L_A ; the molecular-field parameters deduced in [18] were used).

3. Terbium

Tb is ferromagnetically ordered below $T_c = 216$ K, with the moments lying in the basal plane along a b -axis. The moments can be oriented along a hard a -direction by the application of an external field of 32 kOe at 4.2 K. The spin-wave dispersion relation of a ferromagnet does not allow any immediate differentiation of the isotropic and anisotropic two-ion couplings. Only the lifting of the degeneracy of the energies along the K-H edge in Tb provides evidence of such a coupling [11]. Further information on the two-ion coupling in Tb has however been obtained from the field dependence of the spin-wave energies [13, 26].

The dispersion relation is determined by the two Bogoliubov parameters [27]:

$$\epsilon(\mathbf{q}) = \{[A(\mathbf{q}) + B(\mathbf{q})][A(\mathbf{q}) - B(\mathbf{q})]\}^{1/2}. \quad (4a)$$

An external field applied parallel to the magnetization contributes mainly to the $A(\mathbf{q})$ -part:

$$\alpha(\mathbf{q}) = \frac{1}{g\mu_B} \left. \frac{\partial \epsilon^2(\mathbf{q})}{\partial H} \right|_{H \rightarrow 0} = 2A(\mathbf{q}) + \text{corrections} \quad (4b)$$

making a distinction between $A(\mathbf{q})$ and $B(\mathbf{q})$ possible. $\epsilon(\mathbf{q})$ and $\alpha(\mathbf{q})$ have been measured for the magnons propagating in the c -direction of Tb [13, 26] at three different temperatures (4.2, 53, and 134 K), the direction of magnetization being both along the easy and hard planar axes.

In the analysis two complications appeared. The first one is the disturbance caused by the magnon-phonon interactions [28]. The largest one involves directly the spin-orbit coupling of the conduction electrons [28] as proposed by Liu [29]. The main effects of the magnon-phonon

interactions are predictable [28] and were subtracted. The other complication is the corrections in (4b) because the renormalized spin-wave parameters are field dependent. In the analysis the field and temperature dependence of the parameters were assumed to be related through their dependence on the relative magnetization $\sigma(T, H)$ giving rise to an important correction only at 134 K (of about 25%). Although Tb is a well defined spin-wave system at low temperatures, σ is reduced slightly from unity ($\approx 0.5\%$) at $T = 0$ K, and a zero-point change of the renormalized parameters, when applying the field, might be expected. A molecular-field calculation [24] indicates the correction to be less than half the uncertainty by which $\alpha(\mathbf{q})$ was measured. The neglect of this effect might affect slightly the quantitative results deduced but not the qualitative behaviour.

In the analysis of the field experiment four different two-ion parameters were defined [13]:

$$\begin{aligned} A(\mathbf{q}) + B(\mathbf{q}) - [A(\mathbf{0}) + B(\mathbf{0})] &= \mathcal{F}(\mathbf{q})\sigma^{j(\mathbf{q})} + \mathcal{H}(\mathbf{q})\sigma^{k(\mathbf{q})} - \mathcal{C}(\mathbf{q})\sigma^{k(\mathbf{q})} \cos 6\phi, \\ A(\mathbf{q}) - B(\mathbf{q}) - [A(\mathbf{0}) - B(\mathbf{0})] &= \mathcal{F}(\mathbf{q})\sigma^{j(\mathbf{q})} - \mathcal{H}(\mathbf{q})\sigma^{k(\mathbf{q})} - \mathcal{D}(\mathbf{q})\sigma^{k(\mathbf{q})} \cos 6\phi, \end{aligned} \quad (5)$$

where ϕ is the angle between the a -direction and the magnetization. The parameters appearing in (5) cannot be classified by the parity of l but only according to the parity of $l + m$ [13]. If only the isotropic component were of importance the three parameters $\mathcal{H}(\mathbf{q})$, $\mathcal{C}(\mathbf{q})$, and $\mathcal{D}(\mathbf{q})$ should all vanish. This is not the case as shown in figs. 2 and 3. The magnetization exponent $j(\mathbf{q})$ varies between two (at $\mathbf{q} = \mathbf{0}$) and zero, whereas $k(\mathbf{q})$ is much larger (between 27 and 11). $\mathcal{F}(\mathbf{q})$ is the parameter which is most closely related to the isotropic coupling, and assuming $J[\mathcal{F}(\mathbf{0}) - \mathcal{F}(\mathbf{q})]$ to renormalize proportional to σ produces the estimate given by the broken line in fig. 2.

4. Praseodymium

The ions of dhcp (double-hexagonal-close-packed) Pr constitute two non-magnetic singlet-ground-state systems [30] which are only weakly coupled. At zero field the paramagnetic excitations of the hexagonal ions correspond to transitions between the ground-state singlet $|J_z = 0\rangle$ and the excited doublet $|1\rangle_s$ and $|1\rangle_a$ [14, 31]. The

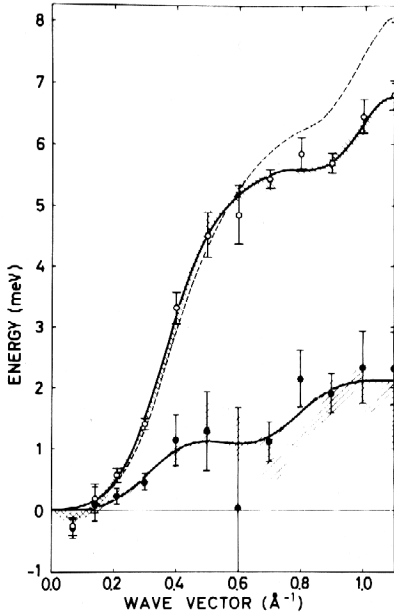


Fig. 2. Coupling parameters $J(\mathbf{q})$ and $K(\mathbf{q})$ in the c -direction of Tb, as defined by eq. (5), represented by open and closed circles, respectively. The broken line shows an estimate of the isotropic part, $J(J(0) - J(\mathbf{q}))$. The hatched areas give the standard deviation regions of the (zero-temperature) parameters (after ref. 13).

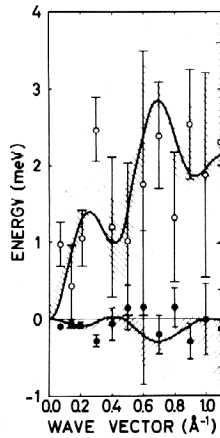


Fig. 3. Basal-plane anisotropy parameters $\mathcal{C}(\mathbf{q})$ (open symbols) and $\mathcal{D}(\mathbf{q})$ (closed symbols) representing the dependence at zero temperature of the two-ion anisotropy in Tb on the orientation of the moments in the basal plane, see eq. (5) (after ref. 13).

dispersion relation of these excitons has been measured at 6.4 K in zero field [14] and as function of field [32]. At zero field (and temperature) the system is equivalent to a spin $S = 1$ system. If anisotropic two-ion couplings were unimportant the doublet excitations should be degenerate. In fig. 4 is shown the dispersion

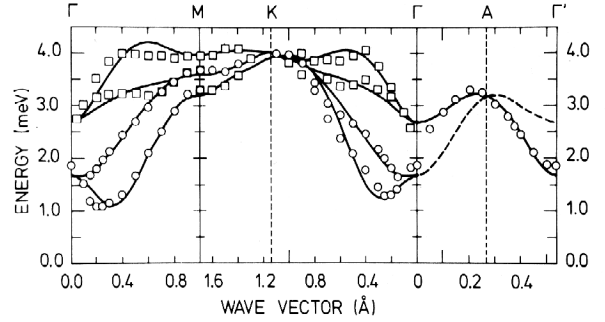


Fig. 4. Dispersion relations for magnetic excitons propagating on the hexagonal sites in Pr at 6.4 K. The double-zone representation is used in the Γ A-direction. A ratio between the isotropic and anisotropic components is given by the difference between two branches (denoted either by squares or circles corresponding to acoustic or optical excitons) divided by the total width of the dispersion relations (after ref. 14).

relations at zero field, and it appears that the doublet excitons are non-degenerate at finite wavevector perpendicular to the c -axis. The large splittings observed can only be explained by introducing an anisotropic coupling (with $m = m' = 1$) of the same order of magnitude as the isotropic coupling. Quadrupole couplings induced by the phonons should be entirely negligible at zero field [33]. In fact, the anisotropy is most likely to be of exchange origin (l and l' both odd) as this assumption leads to a very consistent model for the behaviour of the excitons as function of field [32] and as function of temperature [34].

5. Crystal-field parameters of the rare-earth ions

The crystal-field anisotropy for dilute rare-earth ions in Y, Lu, and Sc have been measured by Touborg and co-workers [15, 16]. The three metals act as hosts which simulate closely the situation of the rare-earth ions in the pure metals (with respect to matching ionic size and the behaviour of the conduction electrons), except that the magnetic two-ion coupling is negligible. The crystal-field parameters, defined in terms of Stevens operators [35]

$$B_{20}O_2^0 + B_{40}O_4^0 + B_{60}O_6^0 + B_{66}O_6^6 \quad (6)$$

are found, when divided by the Stevens factors, to be independent of the rare-earth solute. B_{40}/β , B_{60}/γ , and B_{66}/γ are the same in Y and Lu, and the point charge relation in case of an ideal c/a -ratio, $B_{66} = -\frac{77}{8}B_{60}$, is approximately

satisfied. The B_{20} , scaled according to the deviations from the ideal c/a -ratio, agree reasonably well with the parameters of the pure metals determined by paramagnetic measurements at room temperature [16]. The remarkable consistency of the crystal-field parameters of the rare-earth ions, when placed in the different systems, indicates that they give a reliable estimate of the crystal-field anisotropy of the magnetic metals. A deviation between this deduced anisotropy and that measured experimentally may occur because of two-ion contributions or because the indirect couplings contribute to the single-ion terms. In both cases a discrepancy would indicate the presence of anisotropic two-ion couplings.

In table I the effective single-ion parameters of Pr and Er are compared with the crystal-field parameters deduced by Touborg et al. [16], and in table II is given a comparison of the basal-plane anisotropy parameters at zero temperature:

$$2JP_2 = 6J_1B_{20} - 60J_3B_{40} + 210J_5B_{60} \quad (7)$$

Table I

The crystal-field parameters determined in dilute Pr and Er [16] compared with the effective single-ion parameters of the pure metals (in units of meV). (a) after ref. 32 and (b) after ref. 18.

	Praseodymium		Erbium	
	Dilute	Metal ^(a)	Dilute	Metal ^(b)
$B_{20} \times 10^2$	9.0	19 ± 4	-2.4	-1.8 ± 0.4
$B_{40} \times 10^4$	-4.3	-5.7 ± 5	0.26	-0.48 ± 0.2
$B_{60} \times 10^5$	7.2	10 ± 1	0.24	0.11 ± 0.02
$B_{66} \times 10^5$	-69	-96 ± 10	-2.3	-0.7 ± 0.2

Table II

The anisotropy parameters $2JP_2$, eq. (7), and $36J_5B_{66}$ (in units of meV) calculated from the crystal-field parameters [16] of dilute Tb, Dy, and Ho, and compared with the experimental values of the metals. The values in the brackets include approximately the renormalization effects on the spin-wave parameters (of the order of 50% for B_{60} and B_{66} [23]). (c) after ref. 36 and (d) after ref. 37.

		Dilute	Metal
Terbium ^(c)	$2JP_2$	0.94 (0.60)	7.0 ± 0.2
	$36J_5B_{66}$	0.79 (0.40)	-0.30 ± 0.02
Dysprosium ^(d)	$2JP_2$	4.8 (5.8)	4.3 ± 1.0
	$36J_5B_{66}$	-3.2 (-1.7)	-1.0
Holmium ^(d)	$36J_5B_{66}$	5.9 (3.0)	2.4

and $36J_5B_{66}$ in Tb, Dy, and Ho.

The differences are quite small in the case of Pr but very significant in Tb. The importance of two-ion contributions to the anisotropy of Tb was also detected directly by the spin-wave measurements at zero wavevector [36] as a strong ϕ -dependence of the axial anisotropy (related to $\mathcal{C}(\mathbf{q})$ in fig. 3). Apart from the good agreement obtained in the case of Dy (which might be accidental) the discrepancies increase with the strength of the exchange coupling, which might suggest that the anisotropic two-ion couplings appearing in the metals mainly originate from the indirect exchange interaction.

6. Discussion

The abnormal dispersive effects observed in the excitation spectra of Er, Tb, and Pr at low temperatures are so large that they can only be convincingly explained by the introduction of anisotropic two-ion couplings. Induced couplings due to the phonons are unimportant in Pr at zero field (and presumably also in Er), and in Tb the main effect were subtracted. The results for Tb suggest the presence of a great variety of two-ion couplings some of which must be of quite high rank in order to explain the large exponent, $k(\mathbf{q})$ [at least $l + l' \geq 6$ for the terms contributing to $\mathcal{C}(\mathbf{q})$ and $\mathcal{D}(\mathbf{q})$]. We shall add that the influence of couplings of high ranks, in general, is limited at higher temperatures. The two-ion couplings deduced in the three metals are all of long range, indicating the importance of the conduction electrons. In Gd the orbital moment is zero and anisotropic effects should be unimportant. The isotropic coupling, $\mathcal{J}(\mathbf{q})$, in Er and Tb is not determined uniquely, however, the appropriate scaled functions are similar, both in magnitude and the general \mathbf{q} -dependence, with the two-ion coupling deduced in Gd [38], as shown in fig. 5. This agreement supports the RKKY-theory in emphasizing the importance of the indirect exchange interaction via the conduction electrons, (1). The differences which appear in fig. 5 are probably mainly due to a variation of the properties of the band electrons as discussed by Lindgård and Liu [39].

The behaviour of Pr and the crystal-field parameters of the dilute alloys both suggest the exchange mechanism to be important for the appearance of the anisotropic couplings [3, 4].

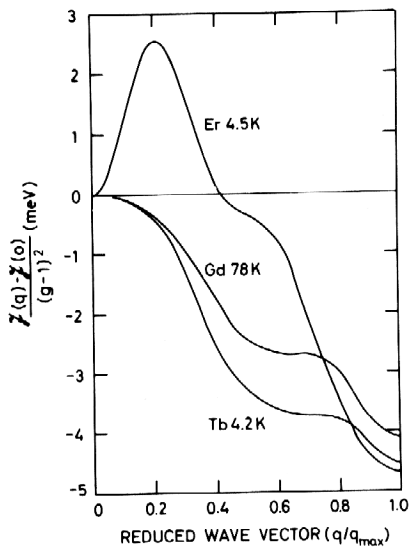


Fig. 5. Exchange function $[\mathcal{F}(q) - \mathcal{F}(0)]/(g-1)^2$ in the c -direction for the heavy-rare-earth metals Gd, Tb, and Er [in the case of Tb is shown $-\mathcal{F}(q)/J(g-1)^2$].

That the spin-orbit coupling of the conduction electrons should also be considered is suggested by the large magnon-phonon interaction observed in Tb [28] (and also in Dy and possibly in Er). A first principles calculation of the magnetic coupling in Gd, which takes into account the band structure and the wavefunctions of the conduction electrons, has been performed by Lindgård et al. [40]. A similar calculation for the case of a non-zero orbital moment would be very valuable.

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