Helifan: A New Type of Magnetic Structure

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The magnetic phases which have earlier been observed when a magnetic field is applied in the plane of the helical magnetic structure of Ho are identified with structures, intermediate between the helix and the fan, which we call helifans. A number of helifan structures have been calculated by a self-consistent mean-field method, and one of them accounts very well for the observed neutron-diffraction pattern. Different sequences of helifans may, in principle, be produced by varying the magnetic field and temperature, and modifying the exchange by alloying.

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In the course of a theoretical investigation of the magnetic phases of Ho in a field, we have discovered a new type of structure, intermediate between the helix and the fan, which we call a helifan. Such intermediate structures should be a rather general feature of periodically ordered systems which display transitions in which the period doubles. Indeed it is apparent that helifans have previously been observed experimentally, without being identified as such. Although the particular system considered here is a rare-earth metal, helifan structures do not require anisotropic interactions, but may occur in the simple Heisenberg system in an applied field. An infinite number of helifan structures exist, and a series of these may be realized in practice by varying the magnetic field. In principle, different sets may be obtained by changing the temperature, or by modifying the exchange, for instance, by alloying different rare-earth metals. Sequences of structures with different periodicities, devil's-staircase phenomena, and possibly chaotic states may therefore be attainable by varying the experimental conditions. Analogs to helifan structures may also occur in nonmagnetic systems.

The effect of a magnetic field applied in the plane of a helical structure was first discussed in detail by Nagamiya, Nagata, and Kitano,[1] on the basis of a mean-field model. As the field is increased, the helix first distorts to produce a moment along **H**, and then undergoes a first-order transition to a fan structure, in which the moments oscillate about the field direction. A further increase in the field reduces the opening angle of the fan which, in the absence of magnetic anisotropy, goes continuously to zero, establishing a ferromagnetic phase at a second-order transition. Hexagonal anisotropy may modify this process into a first-order transition or, if it is large enough, eliminate the fan phase entirely. The magnetization curves of Ho measured by Strandburg, Legvold, and Spedding[2] and by Feron[3] behaved in accordance with this description at low temperatures but, above about 40 K when the fan phase was first observed, a further phase also appeared, manifested by a plateau corresponding to a moment about one-half of that attained in the fan phase. This extra phase was clearly apparent in the magnetoresistance measurements of Mackintosh and Spanel,[4] and later experiments by Akhavan and Blackstead,[5] in which the field was changed continuously, revealed as many as five different phases at some temperatures. The structures in a magnetic field were investigated with neutron diffraction by Koehler *et al.*,[6] who identified two intermediate phases which they called fans and characterized by the intensity distribution of the Bragg peaks. The precise nature of these extra phases has remained a mystery, although they have generally been associated with the very large hexagonal anisotropy in Ho.

In order to elucidate these phenomena we have calculated the effect of a magnetic field on the magnetic structures of Ho by means of a numerical self-consistent mean-field method.[7] The measurements of Gibbs *et al.*[8] have demonstrated that the periodicity of the helix in Ho tends to lock in to the lattice period, and we have therefore performed our calculations on such commensurable structures, allowing for the distortions produced by the large hexagonal anisotropy and the field. Our starting point is the model used earlier[9] to describe the structure and excitations in Ho at low temperatures. The Hamiltonian is

$$\mathcal{H} = \sum_{ilm} B_l^m O_l^m(\mathbf{J}_i) - \tfrac{1}{2}\sum_{ij}\sum_{\alpha\beta}\mathcal{J}^{\alpha\beta}(ij)J_{\alpha i}J_{\beta j} - g\mu_B\sum_i \mathbf{J}_i\cdot\mathbf{H}\,.$$

The first term is the single-ion crystal-field contribution, involving the Stevens operators O_l^m. The crystal-field parameters B_l^m were determined[9] primarily from the magnetic structures and magnetization curves at low temperatures and remain unchanged throughout the calculation, although the effective anisotropy decreases with increasing temperature, roughly as $\sigma^{l(l+1)/2}$, where σ is the relative magnetization, due to the temperature dependence of $\langle O_l^m\rangle$. The second term, the two-ion coupling, comprises an isotropic Heisenberg exchange and the dipolar interaction. The initial values for the former were taken from an earlier analysis of the spin waves[10] in Ho, and depend explicitly on the temperature. They were adjusted slightly during the calculation, to reproduce correctly the transition fields from the helical phase, but

TABLE I. The arrangement of blocks of spins in the helifan structures. The first row shows the relative number of (−) blocks in the different structures.

	Helifan				
Helix	(4)	(3)	($\frac{3}{2}$)	(2)	Fan
$\frac{1}{2}$	$\frac{3}{8}$	$\frac{1}{3}$	$\frac{1}{3}$	$\frac{1}{4}$	1
+	+	+	+	+	+
−	+	+	+	+	+
+	+	+	−	+	+
−	−	−	+	−	+
+	+	+	+	+	+
−	−	−	−	+	+
+	+	+	+	+	+
−	−	+	+	−	+
+	+	+	−	+	+
−	+	−	+	+	+
+	+	+	+	+	+
−	−	−	−	−	+

remained consistent with the spin-wave data, within the experimental error, at the three temperatures considered.

Ho crystallizes in the hcp structure and the atomic moments in any hexagonal plane are all aligned. The first step in the calculation is to assume a distribution $\langle \mathbf{J}_i \rangle$ of the moments at a given temperature. The structure is assumed commensurable, with a repeat distance which may be as high as 50–100 atomic layers for the more complex configurations. The assumed values of $\langle \mathbf{J}_i \rangle$ are inserted into the Hamiltonian and a new set of moments calculated, using the mean-field method to reduce the two-ion term to the single-ion form. This procedure is repeated until self-consistency is attained. The free energy and the net moment in the field direction can then readily be calculated for the self-consistent structure.

We have used such self-consistent calculations to describe the magnetic structures of Ho in zero field,[11] including the spin-slip structures.[8] The repeat distances were determined from experimental data. For the most elementary and stable one-spin-slip structure, with a repeat distance of eleven atomic layers, our results agree within experimental error with the very detailed neutron-diffraction study of Cowley and Bates,[12] both with respect to the bunching of the moments about the magnetically easy axes, and the distortion of the structure in the vicinity of the spin slip. For the other spin-slip structures, there is again good agreement for the mean bunching angle, but the experimental results indicate that there is an extra distortion due to irregularities in the positions of the spin-slip planes in these less stable configurations.

In a magnetic field the hexagonal anisotropy has a decisive influence on the structures at low temperatures, ensuring that a first-order transition occurs from the helix or cone (a helix with an additional ferromagnetic component along the c axis) to the ferromagnet, without any intermediate phase. Above about 40 K, when the hexagonal anisotropy is not so dominant, intermediate stable phases appear between the helix and the ferromagnet. The nature of these phases may be appreciated by noting that the helix can be considered as blocks of moments with components alternately parallel and antiparallel to the field. If we write this pattern schematically as (+ − + −), then the fan structure may be described as (+ + + +). The new phases, which we call helifans, then correspond to intermediate patterns of the type specified in Table I. We use the notation helifan (p) to designate a structure whose fundamental period is p times that of the helix (the single number p is not generally adequate for discriminating between different helifans). The primordial helifan ($\frac{3}{2}$) structure is depicted in Fig. 1. It is clear that these structures represent compromises between the demands of the exchange for a periodic structure and the field for a complete alignment of the moments. They are not due to the hexagonal anisotropy which, on the contrary, tends to suppress them, and occur both when the field is applied along the easy and hard directions in the plane. The energies of the various magnetic phases were calculated as a function of magnetic field and the results for the easy direction at 50 K are shown in Fig. 2. The wave vector $\mathbf{Q}$ was allowed to vary in small, discrete steps, by changing the repeat

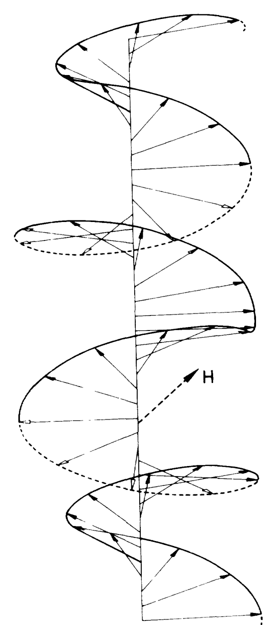

FIG. 1. The helifan ($\frac{3}{2}$) structure in Ho at 50 K. The moments lie in planes normal to the c axis and their relative orientations are indicated by arrows. A magnetic field of 11 kOe is applied in the basal plane and moments with components, respectively, parallel and antiparallel to the field are designated by solid and open arrowheads. This component of the moments has a periodicity which is $\frac{3}{2}$ that of the corresponding helix and the helicity of the structure changes regularly.

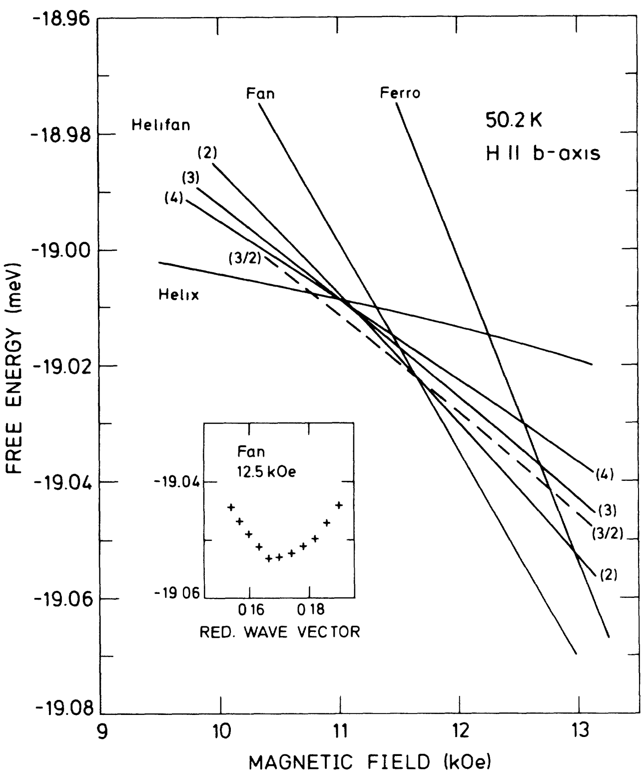

FIG. 2. The magnetic free energy, for different magnetic structures in Ho at 50 K, as a function of the magnetic field along an easy b axis. The free energy is in each case minimized with respect to the wave vector which characterizes the structure, as illustrated for the fan phase in the inset.

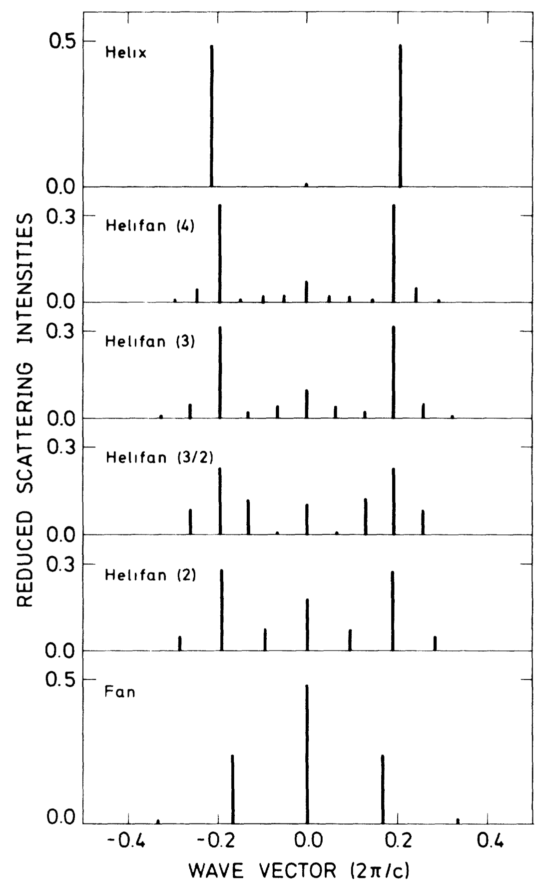

FIG. 3. The neutron-diffraction patterns predicted for the different periodic structures at 50 K. The scattering vector is assumed to lie along the c axis. The structures are calculated with a field of 11 kOe along the b axis.

distance, and the absolute minimum in the free energy for the structure thereby determined, as illustrated in the inset of Fig. 2. These calculations lead to the prediction that the stable magnetic structures follow the sequence helix $\rightarrow$ helifan ($\frac{3}{2}$) $\rightarrow$ fan $\rightarrow$ ferromagnet as the field is increased. In a narrow interval between the helix and the helifan ($\frac{3}{2}$), other stable phases appear, e.g., the helifan (4′) ($+ + - + + - + -$), and similarly the sequence of helifans with m ($+$) blocks followed by a ($-$) ($m \geq 3$) occurs in the close neighborhood of the fan phase.

The various structures are associated with characteristic neutron-diffraction patterns illustrated in Fig. 3. An examination of the neutron-diffraction intensities which Koehler *et al.*[6] associate with the phase which they designate as "fan I" reveals a striking correspondence with the helifan ($\frac{3}{2}$) pattern, as shown in Table II, with a very weak fundamental at $\mathbf{Q}_0/3$, where $\mathbf{Q}_0$ is approximately the wave vector of the helix, but strong second and third harmonics. The basic periodicities of this structure are $2\mathbf{Q}_0/3$ for the component of the moments parallel to the field, and $\mathbf{Q}_0$ for the perpendicular component; the weak $\mathbf{Q}_0/3$ peak arises as the result of interference between them. Similar but more detailed neutron-diffraction results have recently been obtained by Axe, Bohr, and Gibbs.[13] As may be seen from Fig. 3, the changes in the basic wave vector are substantial, even though the underlying exchange function is constant, and they agree very well with those observed by neutron diffraction. For the helix, fan, and helifan ($\frac{3}{2}$) struc-

TABLE II. The reduced scattering intensities of the helifan ($\frac{3}{2}$) structure at 50 K, when a field of 11 kOe is applied along the b axis and the scattering vector is along the c axis. The experimental results are derived from the Fourier components of the ordered moments given in Table I of Ref. 6 (fan I, **H**‖**b**).

Harmonic (n)	$I(\pm n\mathbf{Q})$ Expt.	Calc.
0	0.063	0.105
1	0.007	0.003
2	0.115	0.122
3	0.260	0.230
4	0.077	0.085
5	0.010	0.006

tures the experimental (theoretical) values of **Q** are, respectively, 0.208 (0.211), 0.170 (0.168), and 0.063 (0.066), relative to the reciprocal-lattice vector $\mathbf{b}_3$. The period of the fan phase increases relative to that of the helix because of the resulting increase in the opening angle of the fan. This allows a decrease in the exchange energy which is greater than the concomitant increase of the Zeeman energy. The change in **Q** in the various helifan phases is therefore to a very good approximation proportional to their magnetization.

A detailed consideration[11] of the magnetization data[2,3] indicates that the metastable helifan (2) phase may replace or coexist with the stable ($\frac{3}{2}$) structure in such measurements. The occurrence of phases which are not thermodynamically stable may be explained by a closer examination of the structures of Table I. The helifan (n =integer) configurations may all be derived from the helix by transforming one in n (−) blocks into (+) blocks. Since there are only odd numbers of adjacent (+) blocks, the helicity of the helical regions is conserved. On the other hand, generating the helifan ($\frac{3}{2}$) structure from the helix requires the metamorphosis of regularly spaced (− + −) blocks into (+ − +), and the helicities of neighboring helical regions are reversed. We should expect that the energy barrier for such a complex process would be relatively high and therefore suggest that experiments such as neutron diffraction, which are performed at a fixed field over a long time scale, detect the pure thermodynamically stable ($\frac{3}{2}$) phase, while a rapidly increasing field may induce metastable (n) domains. The helifan (2) structure has apparently been observed in the magnetization measurements, and other stable or metastable helifans may be involved in the five phases observed by Akhavan and Blockstead.[5] In addition, the very pronounced hysteresis which they observed is consistent with the fact that, for instance, the helifan ($\frac{3}{2}$) phase is readily formed from the fan in decreasing fields.

Helifans may also occur in other rare-earth systems, for example, Dy and Er, where periodic ordering is observed, and the magnetic subharmonics recently observed in neutron-diffraction studies of Nd in an applied field[14] may presumably be associated with the analog of helifan structures for the longitudinal wave. It would clearly be of interest to explore the boundaries of the helifan region when the exchange is altered by temperature or alloying, and to examine the metastable helifan structures by neutron diffraction in a changing magnetic field. We are at present embarking on such a study in the Ho-Er system.

The stability of the helifan structures is determined by the form of the two-ion coupling, especially the long-range component. If the exchange is sufficiently short range, the helix, helifans, and fan are almost degenerate at the critical field; it is the interaction between the blocks which differentiates between these structures. Our calculations have been based on a realistic model of a particular physical system. The general properties of helifan structures, including their relative stability under different conditions, could, however, be elucidated by studying simple models, such as the $S=\frac{1}{2}$ Heisenberg system with long-range exchange in a magnetic field.

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[1]T. Nagamiya, K. Nagata, and Y. Kitano, Prog. Theor. Phys. **27**, 1253 (1962).

[2]D. L. Strandburg, S. Legvold, and F. H. Spedding, Phys. Rev. **127**, 2046 (1962).

[3]J. L. Feron, thesis, University of Grenoble, 1969 (unpublished); see also B. Coqblin, *The Electronic Structure of Rare-Earth Metals and Alloys: The Magnetic Heavy Rare-Earths* (Academic, London, 1977), Figs. 198 and 199.

[4]A. R. Mackintosh and L. E. Spanel, Solid State Commun. **2**, 383 (1964).

[5]M. Akhavan and H. A. Blackstead, Phys. Rev. B **13**, 1209 (1976).

[6]W. C. Koehler, J. W. Cable, H. R. Child, M. K. Wilkinson, and E. O. Wollan, Phys. Rev. **158**, 450 (1967).

[7]J. Jensen, J. Phys. F **6**, 1145 (1976).

[8]D. Gibbs, D. E. Moncton, K. L. D'Amico, J. Bohr, and B. H. Grier, Phys. Rev. Lett. **55**, 234 (1985).

[9]C. C. Larsen, J. Jensen, and A. R. Mackintosh, Phys. Rev. Lett. **59**, 712 (1987).

[10]J. Jensen, J. Phys. (Paris), Colloq. **49**, C8-351 (1988).

[11]A. R. Mackintosh and J. Jensen, in Proceedings of the Sir Roger Elliott Sixtieth Birthday Symposium, 1989 [Oxford Univ. Press, Oxford (to be published)].

[12]R. A. Cowley and S. B. Bates, J. Phys. C **21**, 4113 (1988).

[13]J. D. Axe, J. Bohr, and D. Gibbs (private communication).

[14]K. A. McEwen (private communication).