

Magnetic excitations in Dy₂Co₁₇

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1989 J. Phys.: Condens. Matter 1 2047

(<http://iopscience.iop.org/0953-8984/1/11/013>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.90

The article was downloaded on 10/05/2010 at 17:59

Please note that [terms and conditions apply](#).

Magnetic excitations in Dy₂Co₁₇

J H P Colpa†, S Sinnema†, P H Frings‡, J J M Franse† and
R J Radwański†

† Natuurkundig Laboratorium, University of Amsterdam, Valckenierstraat 65, 1018 XE
Amsterdam, The Netherlands

‡ Institut Laue–Langevin, 156X, 38042 Grenoble–Cédex, France

Received 3 May 1988, in final form 22 September 1988

Abstract. Inelastic neutron scattering measurements have been performed on the ferrimagnet Dy₂Co₁₇ in order to investigate the low-energy part (between 2 and 15 meV) of the magnetic excitations. The results are interpreted in terms of Heisenberg exchange and anisotropy parameters in a model Hamiltonian using linear spin-wave theory. The exchange and anisotropy parameters resulting from our analysis are compared with the parameters derived from high-field magnetisation data measured earlier.

1. Introduction

The compound Dy₂Co₁₇ is a member of the inter-metallic series R₂T₁₇ of rare-earth elements R and 3d transition metals T with a hexagonal crystal structure characterised by the unit cell (equal to 2 formula units) of figure 1. Dy₂Co₁₇ is a uniaxial ferrimagnet in which the magnetic moments prefer to lie in the *a*–*b* plane and in this plane directed along the *a* axis indicated in figure 1. High-field magnetisation studies on hexagonal easy-plane intermetallic compounds R₂T₁₇ have recently been reported (Sinnema *et al* 1986, 1987a). The results were described in terms of a number of microscopic parameters accounting for the inter-ionic exchange interactions and the crystal anisotropy of the sublattices. To have additional and independent data for the same parameters, we report here on measurements of the low-lying magnon dispersion relations in Dy₂Co₁₇ using the technique of inelastic neutron scattering.

Earlier, Clausen (1981) (see also Clausen and Lebech 1982) analysed similar data for Ho₂Co₁₇ and Ho₂Fe₁₇ using a model Hamiltonian and linear spin-wave theory. The crystal and magnetic structures of these compounds are very similar to those of Dy₂Co₁₇ except that in zero magnetic field the magnetic moments in Ho₂Co₁₇ and Ho₂Fe₁₇ prefer the *b* direction rather than the *a* direction (figure 1). The aim of the present paper is to use the same theory in analysing our data. In deriving formulae for the magnon dispersion relations, we follow Clausen as much as possible and for many details refer to his publication, trying to confine ourselves in the present paper to the points where our analysis differs from that of Clausen.

The paper is organised as follows. After a description of experimental details in § 2, we present the experimental results in § 3, where also the main differences from the results of Clausen (and Lebech) are indicated. In § 4 the model Hamiltonian of Clausen

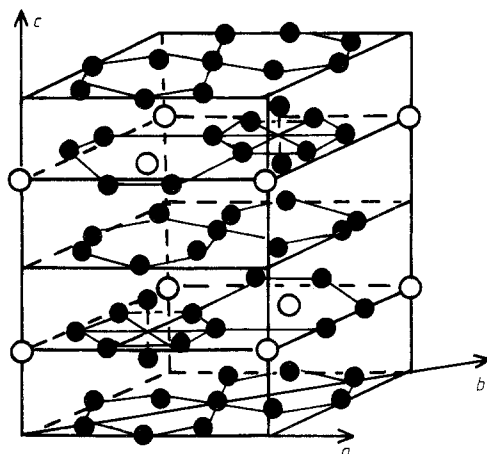


Figure 1. The hexagonal crystal structure of $\text{Dy}_2\text{Co}_{17}$ (○, dysprosium; ●, cobalt), showing the unit cell (=2 formula units) and the definition of the a , b and c directions ($a = 8.36 \text{ \AA}$; $c = 8.12 \text{ \AA}$).

is introduced; in the Appendix the spin-wave theory connected with this Hamiltonian, as described in detail by Clausen (1981) and Clausen and Lebeck (1982), is somewhat adapted and generalised in order to study the implications of the differences between our results and those of Clausen. In § 5, we give the parameters determined from a least-squares fit of the experimental points to the results following from the theory. We conclude with a discussion on the accuracy and other features of Clausen's calculations (and ours) in view of the simplifications used to derive closed expressions for the dispersion relations.

2. Experimental details

The inelastic neutron-scattering experiments were performed using the neutron spectrometer IN8 (with a graphite analyser and monochromator) at the Institut Laue-Langevin in Grenoble. The experiments were performed on two different $\text{Dy}_2\text{Co}_{17}$ single crystals, separately. Special sample shapes—and, for the construction of these, large crystals—were needed in order to minimise the very large neutron absorption in dysprosium (compare $\sigma_{\text{A}}^{\text{Dy}} = 940 \text{ b}$ and $\sigma_{\text{A}}^{\text{Co}} = 37 \text{ b}$).

The crystals were grown using the technique described by Sinnema *et al* (1987b). The first crystal (length, 30 mm; diameter, 5 mm) had a growth direction 15° from the c axis. For the first sample, four plates of 4 mm thickness were cut out of this crystal, each with a surface area of about 20 mm^2 ; we cut these plates in such a way that the a axis was perpendicular to the flat surface of the plates which is the most convenient orientation for the experiment. The plates were glued with epoxy resin (Stycast) onto an aluminium sample holder which was (apart from the sample) covered with cadmium. The second crystal (length, 30 mm; diameter, 7 mm) was grown onto a piece of the first crystal which served as a seed. This seed was sparked and mounted in such a way that its c axis was in the growth direction. In this way, it was possible to let the second crystal grow along the c axis. Because this geometry was much better than that of the first crystal, it was possible

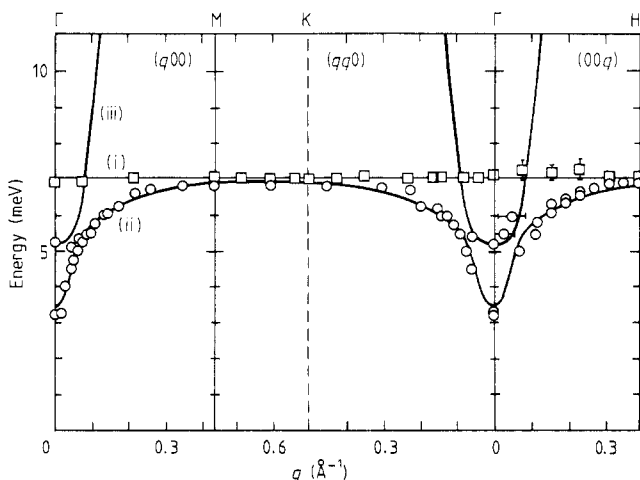


Figure 2. The magnon dispersion relations in Ho_2Co_{17} at 4.2 K (after Clausen (1981)).

to spark two large plates (each surface area was 100 mm^2) with the desired orientation from the second crystal. Unfortunately, the second sample showed some mosaic structure (2°) which caused a peak broadening in the experiments.

3. Experimental results

The inelastic neutron scattering studies on Dy_2Co_{17} were performed at 4.2 K and covered energy transfers between 2 and 15 meV. The experiments were performed with wavevectors q along the a , b and c directions (figure 1). The different dispersion relations (modes) have been observed around the (220) and the $(30\bar{1})$ reciprocal lattice points.

Clausen (1981) and Clausen and Lebech (1982) performed similar measurements on the isostructural compounds Ho_2Co_{17} and Ho_2Fe_{17} and reported (figure 2)

- (i) a non-dispersive mode,
- (ii) a weakly dispersive mode and
- (iii) a highly dispersive mode.

However, in spite of all our efforts to see mode (iii), which for the larger part is associated with the cobalt–cobalt exchange, in Dy_2Co_{17} we only observed the first two dispersion relations (figure 3). (A closer analysis showed, however, that we could obtain a better fit by assigning the points in the immediate neighbourhood of $q = 0$ to mode (iii) rather than to (ii); see § 5.) Our efforts to see mode (iii), which is expected to be very steep, implied the performance of q -scans rather than energy scans that we used for all other experiments. Indeed it is well known that, in R_nCo_m compounds, modes which do not involve precession of the rare-earth ions are rarely observed. For energy transfers larger than the energy transfer of the non-dispersive mode, Clausen did not observe the cobalt–cobalt exchange mode in Ho_2Co_{17} , either. The steep mode was measured, however, in $HoCo_2$ (Castets *et al* 1982) and this makes us think that the invisibility of the steep part of the mode (iii) is due to the mosaic structure of our samples, in combination with the counting rate which is low in view of the large neutron absorption by the samples.

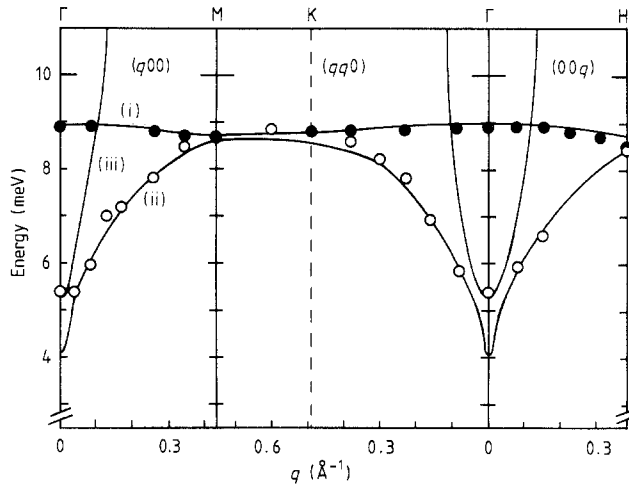


Figure 3. The magnon dispersion relations in $\text{Dy}_2\text{Co}_{17}$ at 4.2 K: \circ , points which are observed around the (220) reciprocal lattice point; \bullet , points which are observed around the (301) reciprocal lattice point; —, the results of the linear spin-wave model as approximated by equations (A1)–(A4) for the parameter values given in table 1.

Trying to use the linear spin-wave theoretical formulae derived by Clausen for the dispersion relations, we run up against the difficulty that mode (i) shows a weak dispersion whereas dispersion is absent in the corresponding mode measured by Clausen in $\text{Ho}_2\text{Co}_{17}$ (compare figures 2 and 3) and in $\text{Ho}_2\text{Fe}_{17}$. Accordingly, Clausen could assume a zero rare-earth–rare-earth exchange J_{RR} which essentially helped him to find closed expressions for the dispersion relations. In § 4 and the Appendix, we give the additional arguments that we used to derive closed expressions for the dispersion relations in which the J_{RR} are incorporated.

4. The linear spin-wave model

Following Clausen (1981) and Clausen and Lebech (1982), we interpret our experimental results in terms of a model Hamiltonian \mathcal{H} consisting of a Heisenberg exchange term and a single-ion crystal-field anisotropy term:

$$\mathcal{H} = \mathcal{H}_{\text{EX}} + \mathcal{H}_{\text{CF}} = \mathcal{H}_{\text{EX}} + \sum_i \mathcal{H}_{\text{CF}}(i) = -\sum_{ij} J_{ij} \mathbf{J}_i \cdot \mathbf{J}_j + \sum_i \sum_{l,m} B_l^m(i) O_l^m(i). \quad (1)$$

The summation indices i, j may correspond to both a rare-earth and a transition-metal ion. ($J_{ij} = J_{ji}$, and all pairs ij occur twice in the summation.) In this model the magnetic interactions are assumed to act between *spatially well localised* angular momenta \mathbf{J}_i . For the rare-earth ion, we use the spin number as given by Hund's rules for the trivalent positive ion ($J_{\text{Dy}} = 7.5$), for the transition-metal ion we use (Clausen and Lebech 1982) a so-called pseudo-spin $J_{\text{Co}} = 0.8$. As for the latter value, although J_{Co} is not an integer or half-integer and therefore forbidden, we follow Clausen and make our calculations for general allowed J_{Co} and substitute $J_{\text{Co}} = 0.8$ in the resulting formulae. In our analysis

the terms in \mathcal{H}_{CF} read in a coordinate system where the crystal c axis is directed along the z axis

$$\mathcal{H}_{CF}(\mathbf{R}) = B_2^0 O_2^0 + B_4^0 O_4^0 + B_6^0 O_6^0 + B_6^6 O_6^6 \quad \mathcal{H}_{CF}(\mathbf{T}) = B_2 O_2^0 \quad (2)$$

where O_l^m are Stevens' operators (Danielsen and Lindgård 1972), e.g.

$$O_2^0 = 3J_z^2 - J(J+1) \quad O_6^6 = \frac{1}{2}[(J^+)^6 + (J^-)^6]. \quad (3)$$

We apply the standard Holstein–Primakoff transformation from spin operators to creation and annihilation operators which is standard for an antiferromagnet or ferrimagnet (there are minor sign changes with respect to the calculations of Clausen in view of the moments which in zero magnetic field in our Dy_2Co_{17} point into an a rather than a b direction as in the holmium compounds studied by Clausen). After re-expressing the Hamiltonian obtained in terms of Fourier-transformed creation and annihilation operators, we are left in principle with the problem of diagonalising for each (allowed) value of the wavevector \mathbf{q} a 76×76 matrix ($76 = 2 \times$ the number of ions in a unit cell). Clausen simplified the calculation enormously by making several assumptions, roughly stated as follows.

(1) Clausen assumes a zero exchange interaction J_{ij} between ions i and j for inter-ionic distances larger than 3 \AA . For smaller distances the value of the exchange parameters depends only on the kind (R or T) of ions i and j , and not on the distance. In this way, only three exchange parameters occur in the problem: J_{RR} , J_{RT} and J_{TT} . (In the 1982 publication, in fact, two exchange parameters J_{TT} were introduced by Clausen and Lebeck: a nearest and a next-nearest exchange parameter. In the present paper, we only use the nearest exchange parameter.)

(2) In view of the appearance of a non-dispersive mode, Clausen assumes a zero rare-earth–rare-earth exchange: $J_{RR} = 0$.

(3) The highly dispersive mode corresponds to an in-phase precession of the T ions and can to a good approximation be derived from a calculation in which only one rather than 34 T ions per unit cell is assumed. The 34 positions of the T ions within the crystal structure are incorporated in Clausen's calculation by some averaging procedure. The coefficient B_2 (equation (2)) is the same for all T ions.

(4) The calculation of Clausen gives only one (triply degenerate) non-dispersive mode. This result was obtained because Clausen used an approximation in which the two crystallographically different rare-earth sites were treated as magnetically identical sites. This is realised by some redistribution of the crystallographic features of the R ions among themselves. The coefficients B_2^0 , B_4^0 , B_6^0 and B_6^6 (equation (2)) are the same for all R ions.

These assumptions enabled Clausen to derive closed expressions for the three modes (strictly speaking $3 + 2$ modes; the calculated non-dispersive mode (i) has threefold degeneracy). Since we found dispersion in mode (i) (figure 3) which in Clausen's case was dispersionless we cannot make assumption (2). However, the generalisation to non-zero J_{RR} of the closed expressions of Clausen for the three modes is not self-evident. The way in which we proceeded to obtain a generalisation is described in the Appendix.

5. Least-squares fit

We analysed our experimental data for Dy_2Co_{17} (figure 3) in terms of the linear spin-wave model described in § 4 and the Appendix. We determined the six free parameters

in this model, i.e. the three exchange constants J_{TT} , J_{RT} , J_{RR} , and the three single-ion anisotropy parameters A_R , B_R , $B_T = -0.816 A_T$ (equation (A3)) by a least-squares fitting procedure of the experimental data points to the results calculated from the model (equations (A1)–(A4)). In order to obtain a feeling for what we had to expect, we first applied the least-squares fitting procedure to the results of Clausen for $\text{Ho}_2\text{Co}_{17}$ as read from figure 2. We could reproduce the results of Clausen quite well but we found that the relatively small errors assigned by Clausen (1981) to each individual parameter only followed from a fit of all three dispersion relations simultaneously. In particular, if one does not include the highly dispersive mode (mode (iii)), the remaining experimental data can satisfactorily be fitted within a large range of the fitting parameter B_T ; the highly dispersive mode is much more sensitive to a variation of B_T . Since at first we thought that in $\text{Dy}_2\text{Co}_{17}$ we did not observe anything of the highly dispersive mode, we fitted our experimental data on $\text{Dy}_2\text{Co}_{17}$ with the value for the B_T parameter fixed on the value found by Clausen for $\text{Ho}_2\text{Co}_{17}$ using all three modes simultaneously ($B_T = -0.50$ meV). In fitting all points, there turned out to be a large difference between the experimental and calculated curves in the vicinity of $q = 0$ (of the order of 1 meV). Considerably better results were obtained in assigning experimental points in the immediate neighbourhood of $q = 0$ to the highly dispersive mode (see curve (iii) in figure 3). Because the region around $q = 0$ is the only part where mode (iii) is not very steep, this supports our conjecture, made in § 3, that the mosaic structure of our crystals is responsible for the non-visibility of (the steep parts of) mode (iii).

Table 1 gives the parameters as derived by fitting all experimental points simultaneously. For J_{TT} and J_{RT} we found values of 17 meV and -0.21 meV, respectively. The value of J_{RT} found for $\text{Dy}_2\text{Co}_{17}$ (-0.21 meV) agrees well with the value derived from the high-field magnetisation experiments on $\text{Dy}_2\text{Co}_{17}$ (Sinnema 1988).

Using equation (A3), we derived a value of -12×10^{-6} meV for the coefficient B_6^0 , which corresponds to a value of -98 J kg^{-1} for K_4^{Dy} . This is acceptable; we could fit the magnetisation curves by the same value (Sinnema 1988). A value of 0.021 meV was obtained for $B^0 = B_2^0 - 390B_4^0 + 37537.5 B_6^0$ (equation (A3)) which agrees with the values $B_2^0 = 0.0307$ meV, $B_4^0 = 25.0 \times 10^{-6}$ meV and $B_6^0 = 0$ meV as derived from the values -1000 J kg^{-1} , 260 J kg^{-1} and 0 J kg^{-1} by which values for K_1^{Dy} , K_2^{Dy} and K_3^{Dy} we could fit the magnetisation measurements (Sinnema 1988).

The value of 2.2×10^{-3} meV found for J_{DyDy} is two orders of magnitude lower than J_{DyCo} and four orders of magnitude lower than J_{CoCo} . It turns out that introduction of the parameter J_{RR} into our fitting procedure has very little effect on the value of the other parameters as is to be expected from this small value of J_{DyDy} . We shall discuss another origin of the overall spread found for mode (i) in § 6.

As the linear spin-wave model is only an approximation, the results are in reasonable agreement with the data deduced from the high-field magnetisation measurements. In particular the agreement between the values for the inter-ionic dysprosium–cobalt exchange constant and between the values for the anisotropy parameter B^0 (equation (A3)) is very satisfactory.

6. Discussion

In the Appendix we described how we generalised to non-zero J_{RR} the expressions found by Clausen (1981) for the three (or five, in view of the threefold degeneracy of mode (i)) lowest-lying magnon dispersion relations. In § 5, we found that a relatively very small

Table 1. The parameters for Dy_2Co_{17} at 4.2 K as deduced from a least-squares fit of the neutron scattering results to the linear spin-wave model for Dy_2Co_{17} (equations (A1)–(A4)), and the magnetic parameters derived from these and from the high-field magnetisation measurements (Sinnema 1988). $J_R = 7.5$, $J_T = 0.8$, $B_0 = B_2^0 - 390B_4^0 + 37537.5B_6^0$ (equation (A3)); for the definition of the macroscopic parameters K_i^R , see equation (12a) of the paper by Clausen and Lebech (1982). The uncertainties in the parameters reflect the differences found when using the fitting procedure on different groups (different directions of \mathbf{q} , for example) of experimental results.

Neutron scattering fitting parameters	
J_{TT}	17 ± 5 meV
J_{RT}	0.21 ± 0.05 meV
J_{RR}	0.002 ± 0.003 meV
A_R	2.4 ± 0.2 meV
B_R	0.49 ± 0.1 meV
B_T	-0.50 meV (kept fixed in fitting procedure, see text)
Magnetic parameters derived from the neutron scattering fitting parameters	
B_2	0.68 meV
B^0	0.021 ± 0.005 meV
B_6^0	$(-12.0 \pm 0.5) \times 10^{-6}$ meV
K_4^R	-98 ± 5 J kg $^{-1}$
High-field magnetisation fitting parameters K_i^R and B -values derived from them	
K_1^R	-1000 J kg $^{-1}$
K_2^R	260 J kg $^{-1}$
K_3^R	0 J kg $^{-1}$
K_4^R	-200 J kg $^{-1}$
B_2^0	0.0307 meV
B_4^0	25.0×10^{-6} meV
B_6^0	0 meV
B^0	0.021 meV
B_6^0	-24.5×10^{-6} meV

value of J_{RR} suffices to explain the dispersion that we found in mode (i). In fact, we felt that the J_{RR} is so small that one, for example, would not be surprised if J_{RR} in reality has the other sign. Indeed, one can show that the approximations leading Clausen (and us) to formulae in closed form (equations (A1)–(A4)) are far too rough to determine J_{RR} of such a small order of magnitude as we found in § 5 with any (relative) accuracy. We give the following arguments. As stated in the Appendix, Clausen was able to find closed expressions for the dispersion relations of interest by a diagonalisation (for each allowed wavevector \mathbf{q}) of a 10×10 matrix, and we have taken Clausen's closed formulae as a starting point to generalise them for non-zero J_{RR} . This 10×10 matrix originates from a 76×76 matrix and the eigenvalues of the smaller matrix are at best only approximations for the lowest-lying excitation energies. We performed for some representative values of the wavevector \mathbf{q} the diagonalisation of the 76×76 matrices by brute-force techniques (the general method has been described by Colpa (1978)).

Table 2 gives the lowest-lying six (or more) excitation energies as calculated in this way for the same values of the parameters J_{TT} , J_{RT} , $J_{RR} = 0$, A_R , B_R and B_T as we found in § 5 (table 1). It is seen from table 2 that the agreement of the results as calculated from the closed formulae (A1)–(A4) (in parentheses in table 2) and those from the

Table 2. Lowest-lying magnon excitation energies for $\text{Dy}_2\text{Co}_{17}$ for some representative values of the wavevector \mathbf{q} as calculated from the diagonalisation of 76×76 matrices. (The parameters $J_{\text{RT}}, J_{\text{TT}}, A_{\text{R}}, B_{\text{R}}$ and B_{T} have the values in table 1 and $J_{\text{RR}} = 0$.) The digits given are significant. The values in parentheses are calculated by the approximating formulae (A1)–(A4) of Clausen. The last column contains the lowest-lying excitation energies (with their multiplicity) which cannot be brought into correspondence with the results of these formulae. The numbers 8.06, 8.87 and 47.2 each occur twice in the last row, i.e. the degeneracy is two.

		Lowest-lying magnon excitation energy (meV)					
\mathbf{q}	ξ	Mode (i)	Mode (i)	Mode (i)	Mode (ii)	Mode (iii)	Other modes
(000)	0.0	8.36	8.70 (8.73)	9.02	4.06 (4.07)	5.22 (5.23)	131
(ξ 00)	0.1	8.36	8.70 (8.73)	9.02	5.94 (5.99)	7.13 (7.32)	132
	0.2	8.36	8.71 (8.73)	9.00	6.97 (7.04)	15.0 (15.7)	128
	0.5	8.37	8.75 (8.73)	8.95	8.07 (8.40)	51.5 (64.6)	72.1, 129
($\xi\xi$ 0)	0.1	8.36	8.70 (8.73)	9.01	6.73 (6.79)	12.5 (13.0)	135
	0.2	8.37	8.73 (8.73)	8.97	7.82 (7.96)	33.0 (35.4)	104
	0.5	8.37	8.75 (8.73)	8.95	8.07 (8.70)	129 (157)	51.5, 72.1, 16
(00 ξ)	0.1	8.35	8.71 (8.73)	9.01	5.78 (5.81)	6.35 (6.41)	120, 166 (2 \times)
	0.2	8.33	8.73 (8.73)	9.00	6.70 (6.75)	12.3 (12.5)	101, 169 (2 \times)
	0.5	8.06	8.87 (8.73)	8.87	8.06 (8.24)	47.2 (48.3)	47.2, 190 (4 \times)

diagonalisation of 76×76 matrices is remarkably good. This justifies using the formulae (A1)–(A4) in fitting procedures as Clausen did, and as we did following him. On the contrary, table 2 shows that there is no question of a threefold degeneracy of mode (i), even for $J_{\text{RR}} = 0$. Rather the three modes of the model Hamiltonian (1) which correspond to the threefold-degenerate mode (i) of Clausen (equation (A1a)) lie close together and show an overall spread larger than 0.5 meV. We conclude that the degeneracy found by Clausen is not connected with the model Hamiltonian (1) but with the approximations leading to the closed formulae. We point out here that the formulae (A1)–(A4) of Clausen do not give for all \mathbf{q} -values the lowest-lying excitation energies. Fortunately this concerns only a part of mode (iii) far outside the range covered by our neutron scattering experiments.

In view of the results of the diagonalisation of 76×76 matrices, we do not understand why a spread of the order of 1 meV was not observed in $\text{Ho}_2\text{Co}_{17}$ and $\text{Ho}_2\text{Fe}_{17}$; with some effort, we may interpret the dispersion in mode (i) in our experiments on $\text{Dy}_2\text{Co}_{17}$ as such a spread. Obviously the reason why Clausen (and we) seemed to see only three dispersion relations instead of five is still an open question.

Appendix. Generalisation to non-zero J_{RR} of the closed expressions of Clausen and Lebech for the dispersion relations

We have proceeded in a somewhat unconventional way as follows. Studying the expression of Clausen (1981) and Clausen and Lebech (1982) for the dispersionless mode (threefold degenerate), we note that it has exactly the same form as one obtains in a system—we shall call it system I—of R spins J_R which are not coupled by any exchange J_{RR} but show single-ion crystal-field anisotropy, in an applied magnetic field equal to the exchange field due to the T ions. It turns out to be possible to obtain also the other two modes found by Clausen in a different way from that used by Clausen. Clausen obtains his closed expressions for the 3 + 2 modes by a diagonalisation of a 10×10 matrix. This 10×10 matrix can be considered to be obtained by some manipulations (i.e., summing up) of the rows and columns in the original 76×76 matrix in order to condense the features of the 34 T ions into just one T ion. We obtain a 4×4 matrix if we follow the same line of argument also for the R ions, leading to a theoretical treatment of a system II with one rather than four R ions in a unit cell (and, as in Clausen's treatment, with one rather than 34 T ions). Diagonalisation of this 4×4 matrix gives precisely the closed expressions found by Clausen for the two dispersive modes. Now it is a standard and straightforward procedure to find the (closed) formulae for the dispersion relations for systems I and II for non-zero J_{RR} and we obtain

$$N_R N_T [\hbar\omega(\mathbf{q})]^2 = a^2(\mathbf{q}) - d_R^2 \quad (\text{A1a})$$

$$N_R N_T [\hbar\omega(\mathbf{q})]^2 = \frac{1}{2}[a^2(\mathbf{q}) + b^2(\mathbf{q}) - d_R^2 - d_T^2] - c^2(\mathbf{q}) \pm \left\{ \frac{1}{4}[a^2(\mathbf{q}) - b^2(\mathbf{q}) - d_R^2 + d_T^2]^2 - c^2(\mathbf{q})[a(\mathbf{q}) - b(\mathbf{q})]^2 - (d_R + d_T)^2 \right\}^{1/2} \quad (\text{A1b, c})$$

where the number of R ions in the unit cell is

$$N_R = 4 \quad (\text{A2a})$$

the number of T ions in the unit cell is

$$N_T = 34 \quad (\text{A2b})$$

and

$$\sqrt{N_R/N_T} a(\mathbf{q}) = N_R A_R - 2J_R [J_{RR}(\mathbf{q}) - J_{RR}(\mathbf{0})] - 2J_T J_{RT}(\mathbf{0}) \quad (\text{A2c})$$

$$\sqrt{N_T/N_R} b(\mathbf{q}) = N_T A_T - 2J_T [J_{TT}(\mathbf{q}) - J_{TT}(\mathbf{0})] - 2J_R J_{RT}(\mathbf{0}) \quad (\text{A2d})$$

$$c(\mathbf{q}) = 2\sqrt{J_R J_T} J_{RT}(\mathbf{q}) \quad (\text{A2e})$$

$$\sqrt{N_R/N_T} d_R = 2N_R B_R \quad (\text{A2f})$$

$$\sqrt{N_T/N_R} d_T = 2N_T B_T. \quad (\text{A2g})$$

The parameters A_R , B_R , A_T and B_T are related to the anisotropy parameters in the Hamiltonian (equation (1)) by

$$\begin{aligned} A_R &= 21B^0 - 157659B_6^0 & A_T &= 0.9B_2 \\ B_R &= -10.87B^0 - 58283B_6^0 & B_T &= -0.816A_T = -0.734B_2 \\ B^0 &= B_2^0 - 390B_4^0 + 37537.5B_6^0. \end{aligned} \quad (\text{A3})$$

The $J_{\alpha\beta}$ in equations (A2) are Fourier transforms of the exchange constants, and the

expressions depend on the direction of q (the ξ -values are measured in units of the length of the corresponding lattice vectors).

(i) For $q = (\xi, 0, 0)$ (q pointing in a b direction (figure 1)),

$$\begin{aligned} J_{TT}(q) &= J_{TT}[80 + 176 \cos(\pi\xi/3) + 96 \cos(2\pi\xi/3)] \\ J_{RT}(q) &= J_{RT}[12 + 32 \cos(\pi\xi/3) + 32 \cos(2\pi\xi/3)] \\ J_{RR}(q) &= J_{RR}[4 + 8 \cos(2\pi\xi/3) + 4 \cos(4\pi\xi/3)]. \end{aligned} \quad (\text{A4a})$$

(ii) For $q = (\xi, \xi, 0)$ (q pointing in an a direction),

$$\begin{aligned} J_{TT}(q) &= J_{TT}[44 + 96 \cos(\pi\xi/3) + 104 \cos(2\pi\xi/3) + 80 \cos(\pi\xi) + 28 \cos(4\pi\xi/3)] \\ J_{RT}(q) &= J_{RT}[20 + 16 \cos(2\pi\xi/3) + 32 \cos(\pi\xi) + 8 \cos(4\pi\xi/3)] \\ J_{RR}(q) &= J_{RR}[8 + 8 \cos(2\pi\xi)]. \end{aligned} \quad (\text{A4b})$$

(iii) For $q = (0, 0, \xi)$ (q pointing in the c direction),

$$\begin{aligned} J_{TT}(q) &= J_{TT}[108 + 144 \cos(\pi\xi/2) + 48 \cos(0.2\pi\xi) + 48 \cos(0.3\pi\xi) + 4 \cos(0.6\pi\xi)] \\ J_{RT}(q) &= J_{RT}[24 + 48 \cos(\pi\xi/2) + 4 \cos(0.7\pi\xi)] \\ J_{RR}(q) &= J_{RR}[12 + 4 \cos(\pi\xi)]. \end{aligned} \quad (\text{A4c})$$

Several coefficients in the expressions for J_{RT} and J_{TT} differ from those of Clausen because Clausen has taken into account the partly disordered crystal structure that was determined in his crystals (Christensen and Hazell 1980). As stated above, equations (A1)–(A4) are generalisations of the formulae derived by Clausen for non-zero J_{RR} (we think that B_R and B_T in the formulae for the dispersion relations of Clausen should be multiplied by 2).

References

- Castets A, Gignoux D and Hennion B 1982 *Phys. Rev. B* **25** 337
 Christensen A N and Hazell R G 1980 *Acta Chem. Scand.* A **34** 455
 Clausen K N 1981 *Risø National Laboratory Report* Risø-R-426
 Clausen K and Lebech B 1982 *J. Phys. C: Solid State Phys.* **15** 5095
 Colpa J H P 1978 *Physica A* **93** 327
 Danielsen O and Lindgård P-A 1972 *Risø National Laboratory Report* No 259
 Sinnema S 1988 *Thesis* University of Amsterdam
 Sinnema S, Franse J J M, Menovsky A and Radwanski R J 1986 *J. Magn. Magn. Mater.* **54–7** 1639
 Sinnema S, Franse J J M, Radwanski R J, Menovsky A and de Boer F R 1987a *J. Phys. F: Met. Phys.* **17** 233
 Sinnema S, Verhoef R, Franse J J M and Menovsky A A 1987b *J. Cryst. Growth* **85** 248