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Magnetic Modulation Vectors of CeSb - Model and Experimental Results

Janusz WOLNY, Bente LEBECH[1], Anna WNĘK and Hae Seop SHIM[2]

Faculty of Physics and Nuclear Techniques, University of Mining and Metallurgy, al. Mickiewicza 30, 30-059 Krakow, Poland
[1] *Risoe National Laboratory, Roskilde, Denmark*
[2] *Korean Atomic Energy Research Institute, Taejon, South Korea*

A model for commensurate transitions in centred tetragonal structure is developed and tested on magnetic structure of CeSb. The basis for the model is the relation between the symmetry direction of atomic structure and the direction of the propagation vector of magnetic modulated structure. The predicted values of modulation vector have been found in the neutron diffraction patterns measured for the single crystal of this compound.

KEYWORDS: CeSb, modulated structures, devil's staircase, neutron diffraction, rare earth, magnetic structures

§1. Introduction

The rare earth monopnictide CeSb (rock salt crystal structure, $Fm3m$) orders magnetically at the Néel temperature ($T_N = 16.5$ K) via a first order transition and the crystal structure transforms to the corresponding tetragonal structure ($I4/mmm$) at the same temperature.[1–5] The magnetic order is to a longitudinal commensurately modulated structure with modulation vector q along the unique tetragonal axis of length 2/3. Below T_N there are number of first order magnetic phase transitions between commensurately modulated structures with propagation vectors 2/3, 8/13, 3/5, 4/7 ..., reaching finally value of 1/2 at low temperatures. The first transition takes place just a few tenth of degree below T_N. The last transition is to the antiferromagnetic structure ($q = 1/2$) at about 9 K. Experiments show that some of these transitions are well behaved and take place at well defined temperatures on cooling and heating, respectively, and the thermal response is almost instantaneous. Other transitions are more chaotic and at these transitions there are even evidence of time dependence of the reordering of the magnetic structure. The unusual properties of CeSb were reviewed by Rossat-Mignod *et al.*[1] where the magnetic phase diagram was investigated under high magnetic fields as well as under high pressure. In our short communication we are not going to discuss the magnetic structure itself. Using only symmetry arguments we are trying to predict values of modulation vectors and compare them with our new results of neutron diffraction measurements.

§2. Model

A new model of commensurately modulated magnetic structures for the centred tetragonal structure is proposed. The main idea of this model is based on the following assumption: whenever the propagation vector of the modulation is parallel to the symmetry direction, the modulation should be commensurate with the basic structure, and vice versa. For the two components of modulation vector: one parallel to the symmetry direction and the other one perpendicular to this direction, disappearance of perpendicular component requires commensurate value for parallel component. If the length of the modulation vector changes continuously, passing through consecutive commensurate and incommensurate values, such behaviour has to be accompanied with the rotation of this vector in respect to the symmetry direction, as it is observed for modulated magnetic structures in Nd and Nd-Pr alloys .[6,7]
The light rare earth metals neodymium and praseodymium crystallise in a double hexagonal close packed (dhcp) structure with only minor differences in their lattice constants. It has been shown that the fundamental commensurate values may be given by the relations: $q_x = 1/n$ and $q_y = 0$, where n is an integer and q_x and q_y are two components of the modulation vector. The x-axis is along the direction of highest symmetry in the hexagonal plane of dhcp structure; y-direction is perpendicular to the highest symmetry direction. One-dimensional commensurate-incommensurate phase transitions for the modulation vectors associated with order, for instance at the hexagonal sites, is given by deviations from the equation, by $q_y \neq 0$. The modulation vectors change with the temperature and their y-component reaches zero for all commensurate values of the modulation length. It should be emphasized that if we consider the spin system as the adsorbate and the atomic structure as the substrate, the conditions given above are equivalent to the situation reported for monolayer films on solid surfaces, i.e. orientational epitaxy. Whenever the structure of the material adsorbed on a homogenous substrate becomes incommensurate with the substrate, the lattice formed by the adsorbed atoms is twisted in relation to the substrate lattice. The ground state for these systems is the incommensurate solid with static distortion waves, which has been treated theoretically in many papers.[8–10]

In neutron diffraction experiments for CeSb single crystal [1-5] no component of magnetic modulation vector perpendicular to the symmetry direction has been observed. Such experimental result requires for all possible modulation vectors to be commensurate. To find the commensurate values of modulations a very simple model is used as described below. The two limits for the length of modulation vector are 1/2, for lower temperatures, and 2/3, for higher temperatures, just below the temperature of the paramagnetic phase transition. There are infinitely many commensurate values between those two bounds, which finally lead to the well-known devil's staircase type of modulated structure.[11,12] However, some satellites for particular values of the modulation vector are more pronounced in the diffraction pattern (see the next paragraph for the experimental results) and the corresponding modulation vectors are more essential.

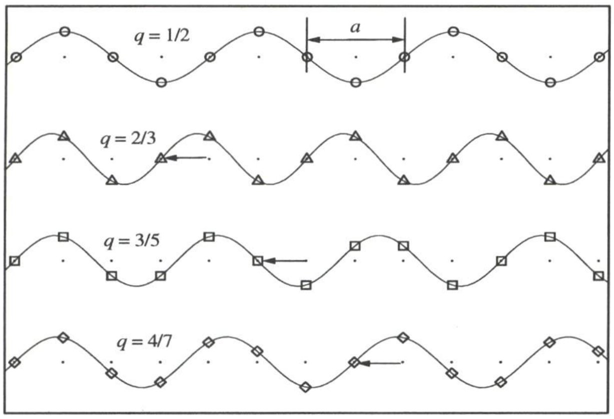

Fig. 1. Illustration of the model. A one-dimensional sinusoidal modulation is pinned on the atomic substrate of centred structure with a periodicity constant a. Starting from the modulation vector $q = 1/2$ after jumps of the nodes by 1/2 (indicated by arrows) new modulated commensurate structures are obtained: 2/3, 3/5, 4/7 respectively.

In the present model it is supposed that the magnetic commensurate structure is pinned on atomic layers, as it is shown schematically in Fig. 1. In this figure the modulation is shown as atomic displacements, rather then magnetic modulation. For this model type of modulation is not important and only the length and the orientation of modulation vector are considered. For the centred tetragonal structures the natural distance between the layers of atoms in symmetry direction is half of the basic period for atomic structure. The jumps between different nodes of modulation vector are accompanied with the change of the corresponding wavelengths by 1/2 (in relative units) and such behaviour can be observed for any integer multiplicity of the half wavelength (Fig. 1). This leads to the following recursive equation:

$$n\frac{\lambda_m}{2} = n\frac{\lambda_{m-1}}{2} \pm \frac{1}{2} \tag{2.1}$$

where λ_{m-1} and λ_m are the former and the next wavelengths, and n is the natural number (n = 1, 2, 3...). Minus sign describes the decreasing of wavelength (i.e. increasing of satellite index as it is observed in CeSb for heating) and plus sign corresponds to the decreasing of the satellite index, in CeSb observed for cooling. The above equation written for the satellite indices (q = $1/\lambda$) gives:

$$q_m = \frac{nq_{m-1}}{n \pm q_{m-1}} \tag{2.2}$$

Equation (2.2) gives broader class of modulation vectors than the one predicted previously,[1] when at low fields and high temperatures the so-called AFP-phases were described by square wave modulations of the wave vector $k = n/(2n-1)$. The above equation describes all possible modulations that can be obtained from particular wave vector in the frame of our model. Among these vectors one should expect the modulations with small value of n to be dominant. On the other side, however, small values of n lead to rather big reorientation of magnetic moments in each step of the transformation, which requires more energy to rearrange the structure. Competition between those two processes leads to some particular modulated structures, which are observed in the diffraction experiments as appropriate satellite reflections. For heating of CeSb from temperatures below 9 K the modulation starts with $q_1 = 1/2$. Some new modulation vectors, which can be obtain in our model from the vector $q_1 = 1/2$, are listed in the Table I. In this table there is a possibility of direct jump from 1/2 to 2/3 (for $n = 2$). However, such transition is hardly observed experimentally, due to the necessity of big rearrangement of the magnetic structure (only for some illustration see also Fig. 1). One of the next transitions, i.e. from 1/2 to 4/7, is dominating, and the corresponding satellite reflection at 4/7 is well developed in the observed diffraction pattern. The most probable transformation from 4/7 to 2/3 leads through 8/13 (n = 8 in Table I).

Table I. Values of modulation vectors obtained from the model for heating of CeSb. Starting values of modulation vectors are 1/2 and 4/7 respectively. The most expected values of modulation vector have been printed in bold.

n	q_1	q_2	q_3	q_4	q_5	q_1	q_2	q_3
1	1/2					4/7		
2	1/2	2/3				4/7		
3	1/2	3/5				4/7		
4	**1/2**	**4/7**	**2/3**			**4/7**	**2/3**	
5	1/2	5/9	5/8			4/7	20/31	
6	**1/2**	**6/11**	**3/5**	**2/3**		4/7	12/19	
7	1/2	7/13	7/12	7/11		4/7	28/45	
8	**1/2**	**8/15**	**4/7**	**8/13**	**2/3**	**4/7**	**8/13**	**2/3**
9	1/2	9/17	9/16	3/5	9/14	4/7	36/59	36/55

§3. Experimental Results

In order to develop a model for the variation of the magnetic modulation vector the antiferromagnet CeSb was reinvestigated by neutron diffraction on a single crystal at Risoe National Laboratory. Several scans around the (200) nuclear reflection were measured at different temperatures below 20 K. Selected diffraction patterns are presented in Fig. 2. In this figure one can easily noticed satellite reflections at related position 1/2, at low temperatures, reaching finally in many steps the position at 2/3 near the Néel temperate. The date obtained for heating in the temperature range 15.3-17.4 K have been fitted to 5 gaussians, with the same width and centred at positions marked in Fig. 3. The only fitted parameters are the satellite intensities and a common very small shift of the scattering vector scale. Results of these fits are shown in Fig. 3 and fully support the model described above. The most probable sequence of the modulation vector is the following: 1/2, 4/7, 8/13 and 2/3.

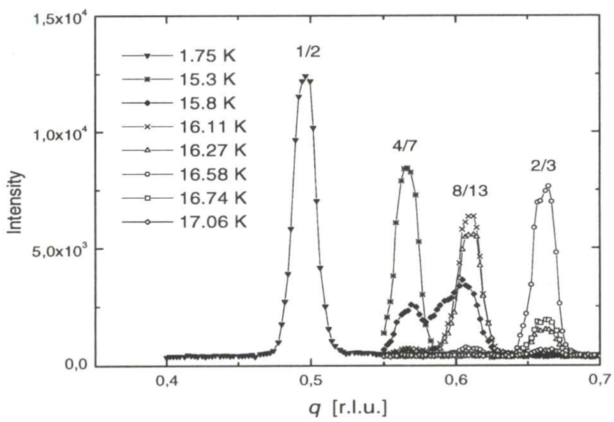

Fig. 2. Neutron diffraction scans through the satellite reflections $(2q0)$ of CeSb at different temperatures.

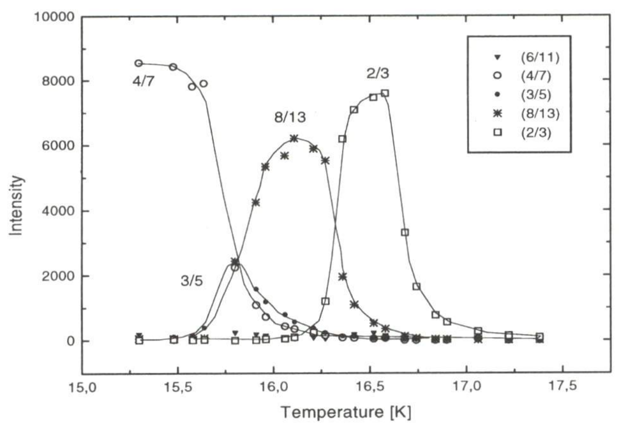

Fig. 3. Fitted intensities of five different satellites versus temperature when heating. The solid line is only a guide to the eye.

One can easily notice the very pronounced contribution of the satellite indexed as 8/13. This satellite overlaps with the two neighbours, 4/7 and 2/3, in a wide temperature range (Fig.3). Such behaviour fully supports the predicted transition from 4/7 to 2/3 via 8/13, as it is pointed out in Table I.

It should be emphasized that 8/13 does not belong to the class of scattering vectors described by $k = n/(2n-1)$.[1] Some contribution from satellite reflections in line 6 of Table I, i.e: 1/2, 6/11, 3/5, 2/3 is also visible. We can also expect some other satellites to appear in the diffraction pattern depending on the thermal history of the sample. Many of them could be observed in the diffraction pattern of the sample with particular heat-treatment. However, they should be mostly unstable and develope with time of annealing.

§4. Conclusions

The model related to the symmetries of modulated structure and the atomic structure has been developed and tested for CeSb magnetic structures. Applying this model to the centred tetragonal structure it was possible to predict possible values of modulation vector for commensurately modulated structures. The obtained values of modulations lead to the devil's staircase structure, however, some values seems to be more favourable. Analysis of neutron diffraction data for CeSb magnetic structure agrees with the results of the model. All the expected satellite reflections with strongest intensities have been found experimentally. Traces for some weak reflections are also evident in the measured diffraction patterns, but more data are still needed to support the model. It should be also emphasised here that for transitions of the devil's staircase type, the resulting structure is very sensitive to the history of the heat treatment and more accurate measurements are still required.

§5. Acknowledgements

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