Devil's Stairs and the Commensurate-Commensurate Transitions in CeSb

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We have analyzed a simple model of a periodically modulated magnetic system. The phase diagram includes multiple phase transitions between commensurate phases, similar to those observed in CeSb. We argue that our model, and CeSb, are possible candidates for the "devil's stairs" behavior.

Many interesting physical systems undergo phase transformations to periodically ordered phases. The distinction is made between commensurate phases where the wave vector describing the modulation can be formed by (simple) rational fractions of the basic vectors of the reciprocal lattice and incommensurate phases where this is not the case. Usually, the periodicity depends upon temperature or pressure. Because of the coupling of the modulated structure to the lattice there is a tendency for the wave vector to lock in at commensurate values. The nature of the resulting wave-vector–versus–temperature curve has been the subject of many considerations. Dzyaloshinsky[1] used a Landau-type argument to suggest that the modulated phase is always commensurate. On the other hand, Pokrovsky[2] has argued that an incommensurate phase might exist under certain circumstances. McMillan[3] and Bak and Emery[4,5] have analyzed the transition from an incommensurate phase to a specific commensurate phase. The transition was found to be continuous.

The most interesting and complicated behavior for such systems has been found by Aubry.[6] He analyzed a simple one-dimensional structural model in an external pressure and found that the wave vector locks into an infinity of rational values. At each value the function remains constant in a finite pressure interval. The resultant wave-vector–versus–pressure curve is an example of the "devil's staircase" as described by Mandelbrot.[7] In this paper we shall see that a similar phase diagram may arise from a completely different model of a *real magnetic system,* namely cerium antimonide (CeSb). Our model explains in a simple way the main features of the complicated phase diagram for this system.

From the experimental side, it has recently been observed that the rare-earth magnet CeSb undergoes several phase transitions between various commensurate phases.[8] The crystal structure of CeSb is fcc. Below $T_N = 16.1$ K the spins are ferromagnetically alligned within $\{001\}$ planes, but periodically modulated perpendicular to these planes. The directions of the spins are also perpendicular to the ferromagnetic planes. The periodicity just below T_N is $\frac{3}{2}$ lattice constants, i.e., it extends over $N=3$ ferromagnetic planes. When the temperature is lowered the system passes through phases with periodicities $N=13$, 7, 18, and 11 ferromagnetic planes until at the lowest temperatures it ends up in a phase with $N=4$.

In order to shed some light at the mechanisms driving this type of behavior, we have undertaken a numerical study of a simple mean-field model. Despite its simplicity it displays a surprising richness of phase transitions when the temperature is varied. When constructing the model we had the very interesting experimental behavior of CeSb in mind, and the Hamiltonian is very similar to those usually applied to analyze similar rare-earth systems. However, no attempt was made to reproduce the correct details of the picture for this particular system at the cost of simplicity, since the main purpose of this work is to investigate the *general* features of modulated systems. Our model includes three temperature-*independent* parameters (one of which is just a scale factor), and reproduces the periodicities $N=4$, 11, 18, 7, and 13 in the same sequence as found in CeSb. However, by refining our analysis, we find more and more stable phases which become stable for smaller and smaller temperature intervals. This is consistent with the devil's-stairs behavior.

Our model is defined in the following way: Spins $S=\frac{5}{2}$ (which is the relevant value in the case of the Ce ions in CeSb) are situated in an fcc lattice and interact with near neighbor spins. The Hamiltonian describing the system may be taken to be of the form

$$\mathcal{H} = -\sum_{ii'} J_{ii'} \vec{S}_i \cdot \vec{S}_{i'}, \tag{1a}$$

where the summation is over pairs of spins. Within the mean-field approximation, which we shall adopt here, this Hamiltonian is replaced by the simpler one

$$\mathcal{H}_{\rm MF} = -\sum_j H_j S_{zj} + \tfrac{1}{2}\sum_j H_j \langle S_{zj}\rangle. \tag{1b}$$

Here S_{zj} is the perpendicular spin component for a magnetic ion in the jth ferromagnetic plane, and H_j is the effective field at each spin in this plane. This field is given by the mean-field equation

$$H_j = \sum_{j'} J(j-j')\langle S_{zj'}\rangle, \quad j'-j = 0, \pm 1, \pm 2, \ldots, \tag{2}$$

where the summation is over the near-neighbor planes, and the effective interplane interactions $J(n)$ are the sums of the interactions between a spin in a given plane and the spins in the nth nearest-neighbor plane. These interplane interactions are the parameters entering into our theory. In principle, the mean-field Hamiltonian may represent a much larger class of original Hamiltonians than that given by (1). For example, the inclusion of two-ion anisotropy which is needed to stabilize the directions of the spins within the planes does not alter the form (1b). Also we believe that the choice $S=\frac{5}{2}$ does not influence essentially the resulting physical picture. Similar models have been applied successfully to similar rare-earth systems with simple ferromagnetic or antiferromagnetic behavior.

For given effective fields the average spins $\langle S_{zj}\rangle$ are calculated by the formula

$$\langle S_{zj}\rangle = \left[\sum_{m=-5/2}^{m=5/2} m \exp(H_j m/T)\right] \times \left[\sum_{m=-5/2}^{m=5/2} \exp(H_j m/T)\right]^{-1}. \tag{3}$$

The problem is then to solve (3) self-consistently with (2), which (surprisingly) turns out to be a difficult task. For a ferromagnet or for a simple antiferromagnet there is only one pair of equations to solve. For a modulated system, however, there is in principle an infinity of coupled equations since the spins in different planes may always be different.

The calculation proceeds as follows. At each temperature the constraint is imposed that the spin configuration repeats itself after a certain number of planes, N. For each N the mean-field equations are solved numerically by means of an iteration procedure. As the starting configuration either a sinusoidal structure or the self-consistent configuration for the previous temperature is used. This setup is then used to calculate effective fields, which again are used to calculate a new spin configuration. Usually, self-consistency is obtained after only a few iterations. The average free energy per spin

$$F(N,T) = N^{-1}\sum_{j=0}^{N-1} -T \ln \mathrm{Tr}_j(\exp(-\mathcal{H}_{\rm MF}/T)) \tag{4}$$

is then calculated. Here Tr_j is the trace over the six spin states for a spin in the jth plane. This free energy is minimized when self-consistency is obtained. At each temperature the stable periodicity is the one which gives the lowest free energy. In principle, the calculation should be performed for N going to infinity to allow also an incommensurate phase. In practice we have carried out calculations for N up to 23. We believe that no additional significant insight can be achieved by extending the numerical calculation to higher values of N.

Near $T_{\rm N}$ the mean-field equations can be solved analytically. The stable periodicity here is given by the wave vector for which the Fourier transform $J(q)$ attains its maximum value, the corresponding spin configuration being purely sinusoidal. The parameters were chosen such that this wave vector is between $\frac{1}{3}$ and $\frac{1}{4}$. Figure 1 shows our result for the following choice of pa-

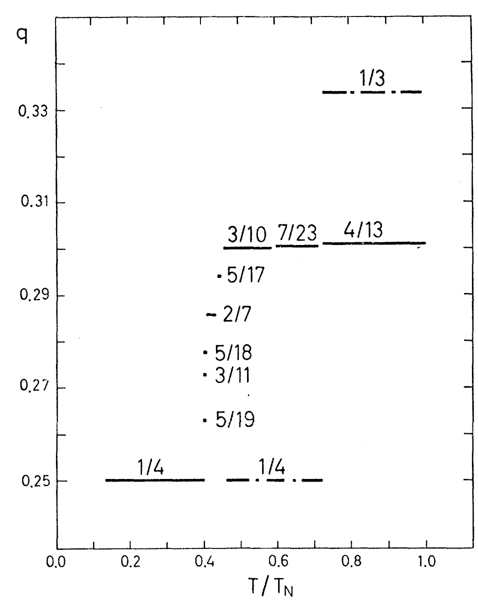

FIG. 1. Wave-vector–versus–temperature curves as calculated for our two models (full curve and dot-dashed curve, respectively). The rational numbers shown correspond to the main Fourier components.

rameters:

$$J(0) = -J(1) = 0.1240T_N, \quad J(2) = -0.0874T_N. \quad (5)$$

The values of $J(0)$ and $J(1)$ may arise from an antiferromagnetic nearest-neighbor- and a ferromagnetic next-nearest-neighbor interaction as found experimentally for other rare-earth antimonides.[9] There is little (or no) hope to calculate these parameters from first principles.

The wave vector varies stepwise in a way similar to that observed in CeSb.[8] The periodicity changes from $N = 4$ at $T = 0$ to $N = 13$ at T_N. In between, the periods 11, 18, and 7 occur in the same sequence as observed experimentally. Our model, with this particular set of parameters, does not reproduce the periodicity $N = 3$ near T_N, but the mean-field theory should break down in this regime anyhow. In fact, a renormalization-group argument may be applied to show that fluctuations will necessarily drive the transition first order at T_N in agreement with experiment.[10]

The stable configurations for the periods $N = 4$, 7, and 13 are shown in Fig. 2 together with their largest Fourier components. The spin arrangement for the periodicity $N = 13$ ($q = \frac{4}{13}$) which is stable near T_N is almost sinusoidal. The structure for $N = 7$ is rather distorted. This distortion gives rise to the higher harmonics which have been observed in the neutron scattering experiments.[8,11] The $N = 4$ configuration is again purely sinusoidal and the spins are almost fully developed ($|\langle S_{z\,j}\rangle| = \frac{5}{2}$).

In addition to these experimentally observed periods other periods become stable. The most interesting behavior takes place near $T = 0.4T_N$. By increasing the maximum N we find more and more phases, which show up on the borderlines between the "main" stable commensurate steps, thus increasing the number of steps. These new phases are stable for extremely small temperature intervals only. For example, the $N = 19$ phase is stable in an interval $\Delta T = 0.000\,003T_N$.

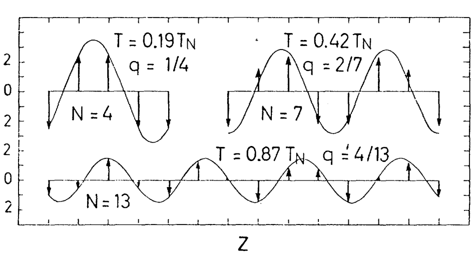

FIG. 2. Some stable spin arrangements. The curves show the largest Fourier component in each case.

The appearence of new steps thus by no means destroy the previous steps, and the curve does not approach an incommensurate one. The periods show up in a rather unpredictable way. For example, the period $N = 15$ never becomes stable.

This picture is consistent with the devil's staircase[7] but differs from Aubry's result[6] in the sense that certain periodicities do not become stable, in agreement with experiment. Of course, there is no hope to prove numerically (or experimentally) that the devil's staircase is complete, i.e., that the number of steps is infinite. We have performed calculations on several similar models defined in terms of different values of the parameters. The general features remain the same for a wide range of parameter values: We are able to increase the number of phases by refining the calculation. Because of the very general form of our Hamiltonian we believe that the devil's-stairs picture is quite general. The reason that it has not been more widely observed experimentally is probably associated with difficulties in resolving the fine steps. We suggest that an effort be made to investigate the details of temperature or pressure dependence of the wave vector for modulated systems, such as the rare-earth metals. More and more steps should appear when a more and more refined experiment is performed. In fact, the experiment by Fisher *et al.*[8] revealed *six* phases whereas the experiment by Rossat-Mignod *et al.*[11] (which was designed mainly to study the field dependence of the phase diagram) showed two or three phases only. An essential condition for the devil's staircase is the existence of metastable states with free energies which are (infinitely) close to the ground-state energy ("frustration"). These states appear here as the solutions of the MF equations with unstable values of N. The frustration arises because of a conflict in sign of the interactions. For Aubry's model, the devil's-stairs behavior is the result of interactions between incommensurate structures. An experimental consequence of the frustration is the "global hysteresis" where the periodicity jumps between metastable values. Experiments should be performed to look for such hysteresis.

For certain values of the parameters, the stable structures are, of course, purely ferromagnetic or antiferromagnetic, and the analysis is trivial. We have also studied a model defined by

$$J(0) = -J(1) = 0.1294T_N, \quad J(2) = -0.0970T_N,$$
$$J(5) = -0.0129T_N, \quad (6)$$

where we have forced the wave vector at T_N to be $q=\frac{1}{3}$. In this case (Fig. 1, dot-dashed line) there seems to be only *one* transition to a phase with $N=4$, and no new phases appear when N is increased, in contrast to the devil's-stairs behavior. However, by changing the parameters slightly, the devil's staircase immediately shows up again.

In conclusion, we have demonstrated that our model exhibits multiple phase transitions between commensurate phases when the temperature is varied, despite the fact that the model includes three temperature-independent parameters only, and we can increase the number of phases by refining the calculation. This is consistent with the devil's-stairs behavior. It would be interesting to investigate our model analytically to find out whether or not the devil's staircase is complete. We also suggest that accurate experiments be performed on periodic magnetic systems to search for the stepwise behavior.

We would like to thank Dr. S. Aubry for interesting discussions on the devil's stairs, and Dr. B. Lebech for explaining to us the experimental situation for CeSb.

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Textural Singularities and Frustration in Random-Anisotropy and Random-Field Models

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Spin models with random anisotropic fields and random magnetic fields are studied. It is shown that integral-index textural singularities of these fields act as disclination sources and participate in destroying long-range ferromagnetic order. Half-integral–index singularities give rise to frustration. The relation of these models to bond disordered spin-glasses is discussed.

Frustration is generally believed to play a fundamental role in determining the nature of spin-glass phases. The concept has, however, only been defined[1] and studied[2,3] for models where the frustrations are properties of the underlying bond structure. Recently spin models with random magnetic fields[3,4] and with random anisotropies[5,6] have been studied by several authors. The results show remarkable similarities to those found previously for bond-disordered spin-glasses. An interesting and related feature of these models is that long-range ferromagnetic order does not seem to exist below four dimensions.[4,6] Our purpose here is to show that one can indeed define frustration as an inherent property of the underlying (random) fields. Frustration defined in this way has properties very similar but not identical to those discussed by Tou-