

Order-(incommensurable disorder) phase transitions

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In first-order phase transitions in magnetic materials exhibiting a high-order spin, the short-range-order vector above the transition point may be incommensurable with the long-range-order vector below the transition point. This theoretical result explains some experiments on UAs. Some other materials which may exhibit this effect are pointed out.

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After the long-range order in a crystal has been disrupted, a short-range order of the same type as the long-range order is generally believed to remain. This assumption is correct for systems in which the long-range order and the short-range order are governed by the same interaction. If instead there are two (or more) competing interactions which are comparable in magnitude but which tend to establish different types of order and which depend on the temperature in different ways, then the long-range order before the transition and the short-range order after the transition may be governed by different interactions. This type of transition has been labeled¹ an “order-(foreign disorder) transition,” to make the distinction from ordinary “order-disorder” transitions, for which the short-range order corresponds in nature to the disrupted long-range order and which would naturally be called “order-(own disorder)” transitions.¹ We note that order-(foreign disorder) phase transitions are necessarily first-order transitions.

If a long-range order with a wave vector Q_{LO} exists above the transition temperature T_0 , then a short-range order with a wave vector Q_{SO} appears above T_0 . This new wave vector is determined from the condition of the maximum susceptibility $\chi(Q)$. In Ref. 1 we studied a particular case of an order-(foreign disorder) phase transition: a transition from an antiferromagnetic long-range order to a ferromagnetic short-range order corresponding to the commensurable wave vectors $Q_{LO} = (\pi/a)(1,1,1)$ and $Q_{SO} = (2\pi/a)(1,1,1)$. Since the paramagnetic Curie temperature of isotropic magnetic materials is $\Theta \sim \langle S_0 S_A \rangle$, the sign of Θ turns out to be positive, not negative, upon this transition.

In this letter we will prove the possibility of an order-(foreign disorder) phase transition of a more general type, in which the long-range-order wave vector Q_{LO} and the short-range wave vector Q_{SO} are incommensurable with each other. We will adopt a model which can be applied to the compound UAs, on which experimental data are available and will be discussed below. This model has a multispin interaction in addition to a two-spin interaction. The effect of the multispin interaction on the magnetic ordering is comparable to that of the two-spin interaction at $T = 0$, but it rapidly fades in importance with increasing temperature. The magnetic material is described as a set

of ferromagnetic planes within which there is an Ising exchange (P). Outside a given plane, the exchange interaction with neighbors from the nearest other plane consists of the ferromagnetic Ising exchange (J) and the antiferromagnetic four-spin anisotropic exchange (K). In addition, antiferromagnetic exchange with the next-nearest planes (V) is also taken into account. The Hamiltonian of this system can thus be written

$$\mathcal{H} = -1/2 J \sum (S_{gn}^z S_{g \pm 1, n}^z) - 1/2 P \sum (S_{gn}^z S_{g, n+\delta}^z) - 1/2 V \sum (S_{gn}^z S_{g \pm 2, n}^z) - \\ - 1/2 K \sum_{\delta \neq \pm \delta'} \left\{ (S_{gn}^z S_{g \pm 1, n}^z) (S_{g, n+\delta}^z S_{g, n+\delta'}^z) + (S_{g \pm 1, n}^z S_{gn}^z) (S_{g \pm 1, n+\delta}^z S_{g \pm 1, n+\delta'}^z) \right\}, \quad (1)$$

where n labels the atom in the plane, g is the index of the plane, and δ and δ' are the indices of the nearest neighbors in the plane. The calculations are carried out in the approximation of a self-consistent field. It is found, in agreement with Ref. 2, that when the four-spin interaction is taken into account the order-disorder phase transition may be a first-order transition. With $k = z(z-2)|K|S^2/J = 0.7$, $v = |V|/J = 0.51$, $p = z_e P/2J = 0.5$, for example, there is a first-order phase transition from an antiferromagnetic state to a paramagnetic state with $\tau_0 = T_0/2JS^2 = 0.07$. The paramagnetic susceptibility $\chi(Q)$ is expressed in terms of the spin correlators by the Kubo equation:

$$\chi(Q) = \frac{1}{T} \sum_{g \neq 0} \langle S_0^z S_g^z \rangle \exp(-iQg), \quad Q = (0, 0, Q). \quad (2)$$

It follows from this equation that, for the parameter values adopted here, $\chi(Q)$ reaches a maximum at that value of Q_{SO} which satisfies the relation $\cos(Q_{SO}a) = -K_{01}/4K_{02}$, where $K_{0i} = \langle S_0^z S_i^z \rangle$; then $Q_{SO} \approx (\pi/a)(0;0;0.3)$. We thus have an order-(foreign disorder) phase transition with incommensurable Q_{LO} and Q_{SO} (an "incommensurable disorder").

This situation is observed experimentally in the diffuse critical scattering of neutrons by the UAs single crystal,³ which has a high magnetic anisotropy. A unique effect is observed: As T_N is approached from the high-temperature side, there is a sharp intensification of the scattering at $\mathbf{q} = (2\pi/a)(1,1,0.3)$ as if a magnetic ordering with $\mathbf{Q}_{SO} = (2\pi/a)(0,0,0.7)$ were beginning. Before a long-range order with this \mathbf{Q} can be established, however, a peak appears at $\mathbf{q} = (2\pi/a)(1,1,0)$, and there is a transition to an antiferromagnetic structure with $\mathbf{Q}_{LO} = (2\pi/a)(0,0,1)$. Sinha *et al.*³ interpreted these results as evidence of a virtual triple Lifshitz point, but Shapira *et al.*⁴ have shown that the necessary conditions for the existence of a Lifshitz point are not met in UAs and that the effect observed in Ref. 3 cannot be explained on this basis. The results of Ref. 3 essentially mean that an order-(incommensurable disorder) phase transition is being observed in UAs.

Several facts imply that a multispin interaction is responsible for the order-(foreign disorder) phase transition in UAs. First, at the Néel point no lattice distortion are observed⁵; i.e., this phase transition is caused by purely magnetic interactions. Second, as the temperature is lowered through the Néel point there is yet another first-order phase transition from a two-sublattice antiferromagnetic order (AFII) to a four-sublattice antiferromagnetic order (AFIV). At present, there is apparently no mechanism other than multispin exchange which can explain the low-temperature order-order

phase transitions.⁶ In principle, both of the structures AFII and AFIV, between which there is a phase transition and which are followed by an order-(foreign disorder) phase transition, can be found from model (1); we have not, however, been able to find exchange parameters such that the AFIV and AFII phases occur in the correct succession. This difficulty could be overcome, however, by sacrificing the simple structure of Hamiltonian (1), e.g., by introducing a four-spin exchange for second-nearest neighbors also.

We wish to emphasize that the order-(foreign disorder) phase transition is totally unrelated to the Ising nature of the interaction between atoms: This phase transition can also occur in isotropic magnetic materials with higher-order exchange (not necessarily four-spin exchange). An example is EuSe. The order-(foreign disorder) transition can be expected to be a typical property of materials exhibiting magnetic polymorphism (solid ³He, for example). Polymorphism is not, however, necessary for such phase transitions: Judging from the temperature dependence of the gap E_g in the optical absorption spectrum found in Ref. 7, a phase transition with $Q_{SO} \neq Q_{LO}$ occurs in MnO, in which the exchange is known to be quite different from Heisenberg exchange.⁶ This compound exhibits a first-order phase transition, below which dE_g/dT is negative, as it should be for an antiferromagnet; above the transition, this derivative is positive in a certain temperature interval, in the manner typical of a ferromagnetic short-range order.⁸ Neutron-diffraction studies will be required for an unambiguous identification of the type of short-range order in MnO and in materials exhibiting an order-disorder phase transition.

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