considers the simplest case of an electron-electron interaction in a single spherical band. The predicted mass enhancement can be decreased significantly by the introduction of Hund's rule (intra-atomic exchange) [S. Doniach, Phys. Rev. Letters 18, 554 (1967)], and finite-range (inter-atomic) exchange interactions [Bucher et al., Ref. 3, and J. R. Schrieffer, Phys. Rev. Letters 19, 644 (1967)], but the relative strengths of these two latter interactions have not yet been determined.

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SURFACE SPIN-FLOP STATE IN A SIMPLE ANTIFERROMAGNET*

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It is well known that in antiferromagnetic materials, a magnetic field applied parallel to the direction of the sublattice magnetization may result in an instability of the ground state, if the strength H of the external field exceeds the value $H_c{}^{(B)} = (2H_E H_A)^{\frac{1}{2}}$, at $T = 0$. Within the framework of the molecular field approximation, it is not difficult to see that when $H > H_c{}^{(B)}$, the state sketched in Fig. 1(b) has an energy lower than that of the antiferromagnetic (AF) ground state illustrated in Fig. 1(a). The presence of the spin-flop instability may also be inferred by noting that as $H \to H_c{}^{(B)}$, the excitation energy of the $\vec{k} = 0$ bulk magnon with frequency $(2H_E H_A)^{1/2} - H$ sinks to zero. Foner[1] has provided a discussion of this "spin-flop" transition, along with a review of a number of experimental studies.

We have recently completed an investigation of the properties of surface magnons in a two-sublattice antiferromagnet of the CsCl structure, with a free (100) surface. A detailed discussion of the properties of these modes, along with their influence on the low-temperature properties of the system, will be given elsewhere.[2] For the present discussion, it is sufficient to note that at $k = 0$, the frequency of the (nondegenerate) surface mode is $(H_E H_A)^{1/2}$, provided surface pinning fields and changes in exchange constants near the surface are neglected and $H_E \gg H_A$. The effect on this result of changes in H_A and H_E near the surface is expected to be small, under conditions described in detail below. If a field H is applied antiparallel to the anisotropy field H_A seen by the surface spins, the excitation energy of the surface mode decreases to $(H_E H_A)^{1/2} - H$. For $H > H_c{}^{(s)} = (H_E H_A)^{\frac{1}{2}}$, the excitation energy of the surface mode becomes negative. By pursuing the arguments given above for
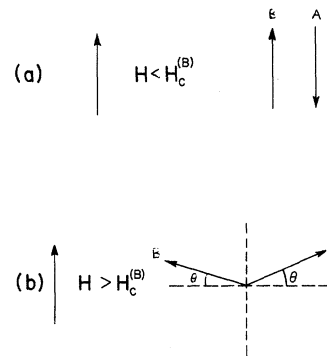

FIG. 1. (a) The bulk AF ground state when $H < H_c{}^{(B)}$. (b) The ground state for $H > H_c{}^{(B)}$; $e = H/2H_E$.

the bulk material, one concludes that for $H > H_c^{(s)}$, the AF ground state becomes unstable.

The purpose of this note is to discuss the nature of the new ground state when $H_c^{(s)} < H < H_c^{(B)}$. We find, in this region, that the spins within roughly H_E/H layers of the surface reorder. If the surface layer is constructed of A spins (assumed oriented antiparallel to the external field H in the bulk material), then when H exceeds $H_c^{(s)}$, the surface spins suddenly shift their orientation to become perpendicular to H. As one moves into the sample, the planes of A spins rotate until, far from the surface, they are aligned antiparallel to H. The B spins remain roughly antiparallel to the A spins; near the surface, the angle between a plane of A spins and an adjacent plane of B spins differs from $180°$ by an amount the order of (H/H_E), so that the layers near the surface in "the surface spin-flop state" (SSF) produce a magnetization in response to the external field. It is this induced magnetization which overcomes the increase in anisotropy and exchange energy to bring the SSF below the normal AF ground state in energy when $H > H_c^{(s)}$. It should be pointed out that in the discussion presented below, we employ a molecular-field approximation. The conclusions are thus not rigorous, but subject to the lim-

itations of the molecular-field theory. Also, we only examine the properties of the ground state so the theory describes the transition only at $T = 0$. We employ a Hamiltonian of the form

$$H = -K\sum_{\mathbb{1}a} S_z^{\;2}(\mathbb{1}_a) - K\sum_{\mathbb{1}b} S_z^{\;2}(\mathbb{1}_b) + H\sum_{\mathbb{1}a} S_z(\mathbb{1}_a)$$
$$+ H\sum_{\mathbb{1}b} S_z(\mathbb{1}_b) + \tfrac{1}{2}J \sum_{\mathbb{1}_a \delta} \vec{S}(\mathbb{1}_a + \vec{\delta}) \cdot S(\mathbb{1}_a),$$

where the spins are arranged on a semi-infinite lattice of the CsCl-like structure[3] with a (100) surface, and K, H, and J are positive constants. Only nearest-neighbor spins interact, and we ignore changes in J and K near the surface for the moment. The surface layer consists of A spins pointing in the $+z$ direction in situations where the normal AF ground state lies lowest in energy. We examine the energy of a state in which the A spins in the $2l$th layer from the surface make an angle α_{2l} with the z direction, and the B spins in the $(2l+1)$st layer make an angle β_{2l+1} with z, where $l = 0, 1, 2, \cdots$. The energy $\mathscr{E}$ of such a state may be written

$$\mathscr{E} = N_s SE,$$

where

$$E = -\frac{H_A}{2}\sum_{l=0}^{\infty}\{\cos^2\alpha_{2l} + \cos^2\beta_{2l+1}\} + \frac{H_E}{2}\sum_{l=0}^{\infty}\{\cos(\alpha_{2l}-\beta_{2l+1}) + (1-\delta_{l,0})\cos(\alpha_{2l}-\beta_{2l-1})\}. \tag{1}$$

The number of spins in a layer is N_S, and we have introduced the exchange field $H_E = 8JS$ and anisotropy field $H_A = 2KS$.

The spin configuration in the ground state must be such that the energy is minimized, i.e.,

$$\partial E/\partial\alpha_{2l} = \partial E/\partial\beta_{2l+1} = 0. \tag{2}$$

From Eqs. (2) and (1), one finds for $l \geq 0$

$$\sin(\alpha_{2l+2}-\beta_{2l+1}) + \sin(\alpha_{2l}-\beta_{2l+1}) = +2(H/H_E)\sin\beta_{2l+1} - (H_A/H_E)\sin 2\beta_{2l+1},$$

$$\sin(\alpha_{2l}-\beta_{2l+1}) + (1-\delta_{l,0})\sin(\alpha_{2l}-\beta_{2l-1}) = -2(H/H_E)\sin\alpha_{2l} + (H_A/H_E)\sin 2\alpha_{2l}. \tag{3}$$

Notice that the normal AF ground-state configuration, with $\alpha_{2l} = 0, \beta_{2l+1} = \pi$ for all l satisfies Eqs. (3). We will examine the case where $H_A \ll H \ll H_E$, so the term in (H_A/H_E) on the right-hand side of Eq. (3) will be ignored in what follows.

We first point out that Eqs. (3) reproduce the bulk spin-flop transition if applied to the

extended medium, with $-\infty < l < +\infty$. In this case, the factor $1-\delta_{l,0}$ is to be suppressed. Then if one asks for solutions with $\alpha_{2l}, \beta_{2l+1}$ independent of l, Eqs. (3) condense to

$$\sin(\alpha-\beta) = -\xi\sin\alpha,$$

$$\sin(\alpha-\beta) = +\xi\sin\beta, \tag{3'}$$

where $\xi = H/H_E \ll 1$ in the region of interest. Equations (3') admit a solution with $\alpha = \frac{1}{2}\pi + \epsilon$, $\beta = \frac{3}{2}\pi - \epsilon$, and $\epsilon = H/2H_E$. To order H^2, the energy of this spin-flop (sf) state is easily shown to be (within a multiplicative constant)

$$E_{sf} = -H_E - H^2/2H_E;$$

when this result is compared with the energy $E_N = -H_E - H_A$ of the normal state, one sees the spin-flop state is stable when $H > H_c^{(B)} = (2H_E H_A)^{\frac{1}{2}}$.

We now examine Eqs. (3) for the semi-infinite sample described above. Introduce a quantity η_{2l}, defined by

$$\beta_{2l+1} = \pi + \alpha_{2l} + \eta_{2l}. \tag{4}$$

Evidently $\eta_{2l} \ll 1$ for all l, since otherwise the increase in exchange energy compared with that in the normal AF state would become large. We shall assume $\eta_{2l} \ll 1$. This assumption will be consistent with the solutions we obtain. Assuming further that α_{2l}, η_{2l} vary slowly with l in a distance of one lattice constant allows us to write Eqs. (3) in the approximate form

$$2\eta_{2l} + (\alpha_{2l} - \alpha_{2l+2}) = -2\xi \sin\alpha_{2l}, \tag{5a}$$

$$\eta_{2l} + (1 - \delta_{l,0})\eta_{2l-2} + (1 - \delta_{l,0})(\alpha_{2l-2} - \alpha_{2l}) = -2\xi \sin\alpha_{2l}. \tag{5b}$$

Comparing the left-hand sides of Eqs. (5) for $l \neq 0$, we find

$$\eta_{2l-2} - \eta_{2l} = 2\alpha_{2l} - \alpha_{2l+2} - \alpha_{2l-2}.$$

In a continuum limit, this becomes

$$\frac{\partial \eta_{2l}}{\partial l} = 2\frac{\partial^2 \alpha_{2l}}{\partial l^2}.$$

Integrating this relation from $l = \infty$ (where it is assumed that α_{2l} and η_{2l} vanish) to some finite value of l yields

$$\eta_{2l} = 2\partial \alpha_{2l}/\partial l \quad (l \neq 0). \tag{6}$$

Equation (5a) may then be written

$$\partial \alpha_{2l}/\partial l + \xi \sin\alpha_{2l} = 0, \tag{7}$$

which may be integrated to yield

$$\tan\tfrac{1}{2}\alpha_{2l} = \tan\tfrac{1}{2}\alpha_0 \exp(-\xi l), \tag{8}$$

where α_0 describes the orientation of a spin in the surface layer. Notice that if the sign of H is changed, no solution of Eq. (7) is obtained that is well behaved as $l \to \infty$. The parameter α_0 is determined by requiring that Eqs. (5) be satisfied for $l = 0$. In the continuum limit, for $l = 0$, the equations require

$$\frac{\partial^2 \alpha_{2l}}{\partial l^2}\bigg|_{l=0} = 0.$$

Straightforward differentiation of Eq. (8) shows

$$\frac{\partial^2 \alpha_{2l}}{\partial l^2}\bigg|_{l=0} = \tfrac{1}{2}\xi^2 \sin 2\alpha_0.$$

Thus either $\alpha_0 = \pi$ or $\alpha_0 = \frac{1}{2}\pi$. If $\alpha_0 = \pi$, then Eq. (8) shows $\tan\alpha_{2l} = \infty$ everywhere, while $\eta_{2l} = 0$. This just corresponds to a normal AF ground state, but with the A and B spins interchanged. Therefore, we examine only the case $\alpha_0 = \frac{1}{2}\pi$, corresponding to a state in which the A spins in the surface layer are oriented perpendicular to the applied field.

Employing Eqs. (8) and (4), we now compute the difference in energy ΔE between the normal AF ground state and the "surface spin-flop" state. Notice the simple result

$$\cos\alpha_{2l} = \frac{1 - \exp(-2\xi l)}{1 + \exp(-2\xi l)}.$$

The change in energy may be evaluated by noting that η_{2l} is small, and $\alpha_{2l+2} - \alpha_{2l}$ is also small. Since all quantities vary slowly, sums over l may be replaced by integrations. We illustrate by computing the change in anisotropy energy:

$$\Delta E_A = +\frac{H_A}{2}\sum_{l=0}^{\infty}\{2 - \cos^2\alpha_{2l} - \cos^2\beta_{2l+1}\} \cong H_A\sum_{l=0}^{\infty}\{1 - \cos^2\alpha_{2l}\} \quad \text{(to lowest order in } H/H_E\text{)}$$

$$= 4H_A\sum_{l=0}^{\infty}\frac{\exp(-2\xi l)}{[1 + \exp(-2\xi l)]^2} \cong \frac{2H_A}{\xi}\int_0^{\infty} dl \frac{\partial}{\partial l}\frac{1}{1 + \exp(-2\xi l)}.$$

Finally $\Delta E_A = +H_A H_E / H$.

In a similar fashion, we find for the Zeeman and exchange contributions

$$\Delta E_z = -2H = -2\Delta E_{ex}.$$

Thus the energy difference ΔE between the normal AF ground state and the SSF state is

$$E_{SSF} - E_N = H^{-1}[H_A H_E - H^2].$$

When $H_c > H_c^{(s)} = (H_A H_E)^{\frac{1}{2}}$, the SSF state lies below the normal AF ground state in energy.

The transition has the character of a first-order phase transition. For the model considered here, the normal AF ground state is stable for all $H < H_c^{(s)}$. A discontinuous transition to the SSF state occurs as soon as H is raised above $H_c^{(s)}$. Notice that the disturbed region for H above $H_c^{(s)}$ occupies a three-dimensional volume of space of macroscopic extent; the transition to the SSF state is not similar to a phase transition in a two-dimensional system. When $H > H_c^{(B)}$, a discontinuous transition to the bulk spin-flop state occurs.

One means of observing the transition to the SSF state would be to measure the magnetization versus H of a thin film, with the field applied parallel to the sublattice magnetization. If the magnetic moment per site is μ_0, the transition to the SSF state would be signaled by the appearance of a moment of magnitude $4\mu_0 N_s$ independent of H,[4] for a thin film with $H > H_c^{(s)}$.

The model Hamiltonian employed in the discussion presents an oversimplified picture of the surface. When $H_E \gg H_A$, in the spin-wave theory[2] the fractional change f in the field $H_c^{(s)}$ that results from a change δH_A in the anisotropy field seen by a surface spin is f $\cong (H_A / H_E)^{1/2} (\delta H_A / H_A)$, assuming that δH_A is parallel to the z axis. For our model, if the z axis lies in the surface layer, then δH_A is parallel to $\hat{z}$, assuming that H_A is dipolar in origin. In this case we estimate $\delta H_A \cong -0.3 H_A$,

so $|f| \ll 1$ if $H_E \gg H_A$. If the exchange coupling between spins in the layers $l_a^z = 0$ and $l_b^z = 1$ is $J' \neq J$, and we define $H_E' = 8J'S$, then to first order in $H_E' - H_E$, one may show that $f = (H_E' - H_E)/2H_E$, when $H_E \gg H_A$. It is difficult to estimate changes in H_E near the surface in a reliable way. However, it is known[5] that in Ni the change Δa_0 in the lattice constant in the surface layer is a few percent of the value a_0 in the host. In a crystal in which the magnetic ion is an S-state ion, the value of J may not be greatly altered by the lowered symmetry of the crystal field in the surface layer. If the change in J comes solely from the change in lattice constant near the surface, and if $(\Delta a / a) \approx 0.05$ is a typical change in a_0, then one might expect $(H_E' - H_E)/H_E \approx (\Delta a / a) \approx 0.05$. Thus, changes in J near the surface may alter the estimate of $H_S^{(c)}$ by a few percent.

The reason for the insensitivity of $H_S^{(c)}$ to changes in H_E and H_A in the limit as $H_E \gg H_A$ has its origin in the fact that the energy decrease responsible for lowering the energy of the SSF state comes from a region $\approx (H_E / H_A)^{1/2} \gg 1$ layers thick. Small perturbations produced by altering H_E or H_A somewhat near the surface do not greatly affect the energy of the SSF state. When $H_E \approx H_A$, the field $H_S^{(c)}$ becomes very sensitive to these changes.

[1] S. Foner, in Magnetism, edited by G. Rado and H. Suhl (Academic Press, Inc., New York, 1963), Vol. I.

[2] D. L. Mills and W. Saslow, to be published.

[3] The B spins are assumed situated at the body center of a tetragonal cage of A spins.

[4] In the SSF state, the mean magnetization/layer affected by the transition increases linearly with H; however, the number of layers affected decreases as $1/H$. Thus for $H_c^{(s)} < H < H_c^{(B)}$ the moment is independent of H.

[5] A. U. MacRae, Science 139, 379 (1963).