Dynamics of the Antiferromagnetic Spin-Flop Transition*

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(Received 11 September 1973)

The critical magnetic field producing spin flop in many antiferromagnets is too small to soften a three-dimensional magnon, i.e., to remove the energy barrier between the equal-energy phases. The barrier can be bypassed, however, via the softening of surface magnons in a smaller field, forming two-dimensional surface-spin–flop states, which broaden with increasing field and which catastrophically spread inward across the three-dimensional material as the critical field is approached.

It has been suggested that many first-order phase transitions are associated with normal modes which partially but not fully "soften," i.e., which almost go to zero frequency.[1] A nonzero energy barrier between two states of equal free energies provides restoring forces confining the normal-mode oscillations to one side of the barrier and to finite frequencies. In those first-order transitions without superheating or supercooling (i.e., without hysteresis) there arises the problem of how the system manages to hurdle the energy barrier. We exhibit here a possible mechanism for one such system.

As first shown theoretically by Néel[2] and experimentally by Poulis and Hardeman,[3] at a critical magnetic field H_3 (see Table I) applied along the preferred axis of an antiferromagnet (AF) there occurs a sudden, nearly 90° rotation of the sublattice vectors into the so-called "spin-flop" (SF) phase. The AF-SF phase transition is first order, there being a finite jump in the magnetic moment, and it can readily be made to take place near 0 K. One's first thought is that it occurs at that field H_4 at which the AF resonance frequency goes to zero, i.e., where the restoring torques vanish and a mode softens. Early work either assumed $H_3 = H_4$ or attempted to derive it.[4]

Anderson and Callen[5] identified the three possible critical fields H_2, H_3, and H_4 described in Table I. They argued that the AF-SF transition should occur at H_4 in increasing H but at H_2 in decreasing H; and that unless $H_2 = H_3 = H_4$, hysteresis should be observed.

Experimental evidence of the absence of hystereresis has been accumulating.[6] Furthermore, the AF→SF transition in at least three materials[7] takes place at a field measurably less than H_4 and theoretically evaluated as H_3, i.e., in the presence of a finite energy barrier.

Mills and Saslow[8] have shown that a field $H_1 < H_3$ fully softens a surface magnon; Mills[9] has demonstrated that above this same H_1 there can occur surface-spin–flop (SSF) regions, in which a few layers of spins near a surface have turned nearly 90°. According to Mills, the number of layers involved decreases with increasing H. We show that Mills's analysis omitted terms of the same order of magnitude as those included, and that in fact the number of layers *increases* with increasing H.

Our picture of the dynamics of the AF→SF transition is as follows: At a field H_1 surface magnons soften and SSF regions develop. Similar localized regions might also develop near impurity clusters, dislocation lines, and other imperfections. The SSF regions at first grow slowly with increasing H. Then as $H \to H_3$ the SSF regions catastrophically expand into three dimen-

TABLE I. Critical fields in antiferromagnets, in order of increasing size and at 0 K, for the energy given by Eq. (1). All fields are along the easy axis.

Field	Effect
$H_1^2 = H_E H_A + H_A^2$	Surface magnons soften in increasing H; SSF
$H_2 = H_3^2/H_4$	SF-phase magnons soften in decreasing H
$H_3^2 = 2H_E H_A - H_A^2$	Phase boundary, equal free energies, AF-SF
$H_4^2 = 2H_E H_A - H_A^2$	AF-phase magnons soften in increasing H
$H_5 = 2H_E - H_A$	Phase boundary, SF paramagnetism

sions and encompass the entire material. The leading edge of an SSF region moves much as does the edge of a Bloch wall. The system is continuously in stable equilibrium and the energy barrier is bypassed. The SF→AF transition should ideally simply reverse the above sequence.

Following Mills[9] we write the energy as ES per surface spin, with

$$E=-\tfrac{1}{2}H_A\sum_{l=0}^{\infty}(\cos^2\alpha_{2l}+\cos^2\beta_{2l+1})+\tfrac{1}{2}H_E\sum_{l=0}^{\infty}[\cos(\alpha_{2l}-\beta_{2l+1})+(1-\delta_{l,0})\cos(\alpha_{2l}-\beta_{2l-1})] +H\sum_{l=0}^{\infty}(\cos\alpha_{2l}+\cos\beta_{2l+1}). \quad (1)$$

Here the A spins in the $2l$th layer from the ($l=0$) surface make angles α_{2l} with the preferred $+z$ direction, and the B spins in the $(2l+1)$th layer make angles β_{2l+1} with $+z$. The applied field H is directed along $-z$, and H_A and H_E are effective anisotropy and exchange fields, respectively, in the molecular field approximation.

The study is limited to 0 K. On minimizing (1) with respect to α_{2l} and β_{2l+1}, we obtain, for $l\geqslant 0$,

$$\sin(\alpha_{2l+2}-\beta_{2l+1})+\sin(\alpha_{2l}-\beta_{2l+1})=2\xi\sin\beta_{2l+1}-\zeta\sin 2\beta_{2l+1}, \quad (2a)$$

$$\sin(\alpha_{2l}-\beta_{2l+1})+(1-\delta_{l,0})\sin(\alpha_{2l}-\beta_{2l-1})=-2\xi\sin\alpha_{2l}+\zeta\sin 2\alpha_{2l}. \quad (2b)$$

Here $\xi\equiv H/H_E$ and $\zeta\equiv H_A/H_E$. We assume $H_A\ll H\ll H_E$, or $\zeta\ll\xi\ll 1$. Since $H_3^{\,2}\approx H_AH_E$ (as is well known and as will be shown), we must keep in mind that ζ is the same order as ξ^2. Let

$$\alpha_{2l}-\beta_{2l+1}=-\pi-\eta_{2l+1},\quad \alpha_{2l+2}-\beta_{2l+1}=-\pi+\epsilon_{2l+1}. \quad (3)$$

By our assumptions, neighboring spins are almost antiparallel and α and β vary slowly with l. Hence η and ϵ are small parameters (of order ξ, it develops). We insert (3) into (2), expand in powers of η and ϵ, and pass to the continuum limit. After much algebra there results, for $l>0$ and through order ξ^4,

$$2\,\partial^2\alpha/\partial l^2=(2\zeta-\xi^2)\sin 2\alpha+X; \quad (4)$$

$$X=\zeta\xi(\sin 2\alpha\cos\alpha-2\sin\alpha\cos 2\alpha)+\tfrac{1}{2}\zeta^2\sin 4\alpha+\tfrac{1}{2}\left(\epsilon^2\frac{\partial\epsilon}{\partial l}+\eta^2\frac{\partial\eta}{\partial l}\right) -\left(\eta^2+\epsilon^2+2\eta\frac{\partial\eta}{\partial l}-2\epsilon\frac{\partial\epsilon}{\partial l}\right)(\tfrac{1}{2}\xi\sin\alpha+\zeta\sin 2\alpha)-\frac{2}{3}\frac{\partial^4\alpha}{\partial l^4}.$$

We first solve (4) omitting X, which is of order ξ^3, and obtain

$$(\partial\alpha/\partial l)^2=(2\zeta-\xi^2)\sin^2\alpha+\text{const}. \quad (5)$$

The equation obtained by Mills[9] did not contain the 2ζ. The integration constant is evaluated by matching to the known infinite-medium solution. Thus, in the AF phase, in which $\xi^2<2\zeta$, with $\partial\alpha/\partial l=0$, $\sin\alpha=0$, we have const $=0$.

We next use the (lower energy) negative root of (5) to evaluate η and ϵ through order ξ^2, insert these into X, and thereby achieve an equation in α valid through order ξ^4, for $\xi^2<2\zeta$:

$$2\,\partial^2\alpha/\partial l^2=(a^2-2b^2\sin^2\alpha)\sin 2\alpha; \quad (6)$$

$$a^2\approx 2\zeta+\zeta^2-\xi^2,\quad b^2=2\zeta(\xi^2-\zeta).$$

This integrates to

$$(\partial\alpha/\partial l)^2=a^2\sin^2\alpha-b^2\sin^4\alpha+\text{const}, \quad (7)$$

and again we take const $=0$.

To describe the SSF state we take the minus root of (7), integrate, and set $\alpha_0=\pi/2$. The result is[10]

$$\sin^2\alpha_{2l}=\{1+[1-(b^2/a^2)]\sinh^2(2al)\}^{-1}. \quad (8)$$

This is now used, together with previous results, to evaluate the energy difference ΔE between the SSF and the AF state. The sums from $l=0$ to ∞ are replaced by integrations. We find, to order ξ,

$$\Delta E=\tfrac{1}{2}H_E(a-\xi). \quad (9)$$

This becomes negative at $\xi>\zeta^{1/2}$. Thus, when H exceeds H_1 and surface magnons soften, the SSF state is energetically favorable.

However, in a semi-infinite medium Eq. (2b) for $l=0$ allows only $\alpha_0=0$ or π. In an ideal finite crystal with A spins on the first ($l=0$) plane and B spins on the last plane there will occur, for $H>H_1$, two surface states, one converging inward from the first plane and one diverging inward from the last plane. The $l=0$ spins will turn through almost a full 180°, sending the region of 90° spin flop towards the middle of the crystal where it stops after meeting the diverging state coming from the last plane. The energy will approach twice that given by (9). In a real crystal the nature of the surfaces and of the interior will determine the precise location of the spin-flop region, and may also raise the critical field above H_1.

As $b \to a$, according to (8) all $\sin^2\alpha_{2l} \to 1$, and hence the SSF region expands across the entire crystal and a first-order phase transition takes place into the SF phase. The condition $a^2 = b^2$ is

$$\xi^2 = \xi_3{}^2 \equiv 2\zeta - \zeta^2, \qquad (10)$$

which marks the well-known AF-SF free energy boundary,[5] $H_3 \equiv \xi_3 H_E$. Above H_3, Eqs. (8) and (9) no longer apply, since the integration constant in (7) ceases to be zero.

Finally, we introduce a $\cos^4\alpha$ anisotropy, characterized by $\zeta' \equiv H_A'/H_E$, and also a Dzialoshinski-Moriya[11] interaction, characterized by $\mu \equiv H_D/H_E$. After relating the canting angle to α, it is found eventually that Eq. (6) and all the consequences still hold, with, to first order in ζ and ζ',

$$a^2 = 2\zeta + 4\zeta' - \xi^2 - \mu^2, \quad b^2 = 2\zeta'.$$

We note that the fields $H_3{}^2 = 2H_E(H_A + H_A') - H_D{}^2$ and $H_4{}^2 = 2H_E(H_A + 2H_A') - H_D{}^2$ are separated to first order in ζ', as has been pointed out by Jacobs *et al.*[7]

We wish to thank David Jasnow and Jack Semura for helpful comments.

*Research sponsored by the Air Force Office of Scientific Research, Air Force Systems Command, U.S. Air Force, under Grant No. AFOSR 71-2028.

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N-Body Forces in High-Density Nuclear Matter

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The contribution of n-body forces to nuclear matter energy is summed in closed form as a function of the nuclear density. It is seen that they provide an increasing amount of binding as the density increases beyond ordinary nuclear density. The presence of correlations suppresses this effect, but does not destroy it fully. The physics behind these phenomena is discussed.

We summarize here results obtained by us for the contribution of the *sum* of pionic many-body forces (i.e., intrinsic three-body forces + four-body forces + . . .) to the binding energy of nuclear matter in the presence of internucleon correlations. These higher many-body forces are certainly present in any real nuclear system, and have not yet been investigated, let alone summed. In addition, there are many interesting physical effects which may be expected to be produced by the sum of these forces, lending additional motivation for this work. The three-body force, on