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Spin-Wave Instability and Premature Saturation in Antiferromagnetic Resonance*

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A semiclassical theory of spin-wave instability in antiferromagnetic resonance is developed. The calculation is valid both for simple antiferromagnets and for canted systems when the canting is due either to single ion magnetocrystalline anisotropy or anisotropic exchange. The nonlinear terms leading to the instability are found to originate in the anisotropy and exchange energies. The critical radio frequency field for the onset of instability is calculated and in general is given by $h_c = 4\Delta H_0(\gamma\Delta H_k/\omega_0)^{1/2}$, where ΔH_0 and ΔH_k are, respectively, the uniform mode and spin-wave linewidths; and ω_0 is the zero-field antiferromagnetic resonance frequency. Experimental evidence for the existence of spin-wave instability in the canted antiferromagnet, $KMnF_3$, is presented and discussed. An anomalous low-level saturation of the rf susceptibility is attributed to the onset of spin-wave instability. A new technique for the measurement of spin-wave linewidths is described. This technique, which is based on the response of the resonant system to amplitude modulated microwave power, yields results in agreement with the instability theory. The spin-wave linewidth as determined by these experiments is approximately three orders of magnitude narrower than the linewidth of the uniform mode, indicating that the broadening mechanisms are grossly different in the two cases.

I.

THE premature saturation of the rf susceptibility in ferromagnetic resonance first observed by Damon[1] and Bloembergen and Wang[2] is well known. Bloembergen and Wang[2] demonstrated experimentally that this saturation occurred at an rf level far below that needed to reduce M_z by a significant amount. Suhl[3] attributed this anomalous saturation to instabilities in that portion of the spin-wave spectrum which is degenerate[4] in energy with the uniform mode. These instabilities result from a coupling of the uniform mode to the degenerate ($k\neq 0$) spectrum via nonlinear terms with the result that the uniform mode drives the degenerate spin waves in a choherent way. At a sufficiently high power level, the rate of growth of a degenerate spin-wave amplitude will exceed its relaxation rate, $(\gamma\Delta H_k)$. Under these conditions, the amplitude will grow exponentially with time, i.e., an instability will exist. In a previous paper,[5] it was shown that similar spin-wave instabilities are expected in simple antiferromagnetic systems. In this paper we present a more general theory of instability in antiferromagnetic systems valid for canted as well as simple antiferromagnets. In the more general antiferromagnetic case, the instability arises from nonlinear terms proportional to the exchange and anisotropy energies. Dipolar effects are relatively unimportant because the net moment, even on resonance, is small. The critical field for the onset of instability can generally be written

$$h_c = 4\Delta H_0(\gamma\Delta H_k/\omega_0)^{1/2},$$

where ΔH_0 and ΔH_k are, respectively, the uniform mode and spin-wave linewidths, and ω_0 is the antiferromagnetic resonance frequency. The effect of a canting interaction, if present, is simply to change the antiferromagnetic resonance frequency in the above expression.

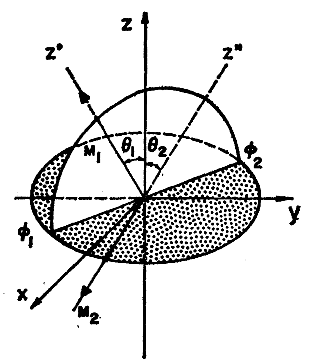

FIG. 1. Diagram showing the angles θ_1, ϕ_1, θ_2, ϕ_2 which define the new coordinate systems in terms of the x, y, z system. M_1 is along z'; and M_2 is along $-z''$ at equilibrium.

Dzyaloshinskii[6] used a thermodynamic approach to show that canted antiferromagnetism is an intrinsic property and may be understood as a direct consequence of crystal symmetry. The detailed physical interactions responsible for canting have been discussed by Moriya.[7,8] Moriya points out the existence of two mechanisms. The first is a single-spin magnetocrystalline anisotropy which differs for differing crystallographic sites,[7] and the second is anisotropic exchange.[8] The canting in a given crystal is due to one or the other (or perhaps both) of these two interactions. It is easily

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[1] R. W. Damon, Rev. Mod. Phys. **25**, 239 (1953).
[2] N. Bloembergen and S. Wang, Phys. Rev. **93**, 72 (1954).
[3] H. Suhl, Phys. Rev. **101**, 1937 (1956); Proc. IRE **44**, 1270 (1956); J. Phys. Chem. Solids **1**, 209 (1957).
[4] P. W. Anderson and H. Suhl, Phys. Rev. **100**, 1783 (1955).
[5] A. J. Heeger and P. Pincus, Phys. Rev. Letters, **10**, 53 (1963).
[6] I. E. Dzialoshinskii, J. Phys. Chem. Solids **4**, 241 (1958).
[7] Toru Moriya, Phys. Rev. **117**, 635 (1960).
[8] Toru Moriya, Phys. Rev. **120**, 91 (1960).

shown that whenever symmetry allows the existence of the anisotropic exchange of the form

$$D(S_1^iS_2^j-S_1^jS_2^i),$$

where i, j are x, y; x, z; or y, z and 1 and 2 denote sublattice; symmetry also allows a single-ion anisotropy of the form

$$K(S_1^iS_1^j-S_2^iS_2^j).$$

In Sec. II we give a detailed calculation of the instability criteria valid in all three cases; simple antiferromagnet, canting due to single-ion anisotropy, and canting due to anisotropic exchange. The results are, therefore, valid for a wide variety of magnetic materials.

Experimental evidence is presented to demonstrate the possible existence of spin-wave instabilities in the canted antiferromagnet, $KMnF_3$. An anomalous saturation of the rf susceptibility is observed at an rf power level considerably below that required to decrease the magnitude of the sublattice magnetization vectors, $|M^z|$, appreciably. Furthermore, the response of the system to amplitude modulated microwave power implies a spin-wave relaxation time of the order of that inferred from the "critical field" for the onset of saturation.

II. SEMICLASSICAL THEORY OF SPIN-WAVE INSTABILITY IN ANTIFERROMAGNETIC SYSTEMS

We shall use the coordinate systems x', y', z'; x'', y'', z'' indicated in Fig. 1, and assume a two-sublattice model. Thus, $M_1^{z'}\cong M_0$ and $M_2^{z''}\cong -M_0$. One can always represent the effective magnetic field experienced by sublattice 1 in the form

$$H_{\mathrm{eff}x'}^{(1)}=-[a_1m_{2x''}+b_1m_{2y''}+c_1m_{2z''}+d_1m_{1x'}+e_1m_{1y'}+f_1m_{1z'}],$$
$$H_{\mathrm{eff}y'}^{(1)}=-[g_1m_{2x''}+h_1m_{2y''}+j_1m_{2z''}+k_1m_{1x'}+p_1m_{1y'}+q_1m_{1z'}],$$
$$H_{\mathrm{eff}z'}^{(1)}=-[r_1m_{2x''}+s_1m_{2y''}+t_1m_{2z''}+u_1m_{1x'}+v_1m_{1y'}+w_1m_{1z'}],$$

with similar expressions for the components of the effective field acting on sublattice 2. The vectors $\mathbf{m}_1$ and $\mathbf{m}_2$ are unit vectors parallel to the magnetization vectors on the two sublattices. The coefficients appropriate to the crystals considered here are listed in Appendixes 2 and 3. The coefficients for a simple antiferromagnet may be obtained from Appendix 2 by setting $H_{A_2}=0$.

Suppose that the $k=0$ resonance mode (uniform precession) is excited in such a material, and that superposed on this mode a higher k spin wave is set up due, perhaps to a thermal excitation or to scattering from the $k=0$ mode. We wish to investigate the conditions under which such a spin wave will become unstable, and once set up, will grow rather than damp out. Proceeding in a manner similar to that of Suhl,[4] we write the equations of motion for such a system:

$$\frac{1}{\gamma}\frac{d}{dt}[\mathbf{m}_1+\delta\mathbf{m}_1(\mathbf{r})]=[\mathbf{m}_1+\delta\mathbf{m}_1(\mathbf{r})]\times[\mathbf{H}_{\mathrm{eff}}^{(1)}+\delta\mathbf{H}_{\mathrm{eff}}^{(1)}(\mathbf{r})],$$

with a similar expression for sublattice 2. $\delta\mathbf{m}(\mathbf{r})$ is assumed to be periodic and varies as $\cos(\mathbf{k}\cdot\mathbf{r})$. Note that the sum $\mathbf{m}_1+\delta\mathbf{m}_1$ is always constant in accord with the local conservation of the magnetization vector. We shall neglect terms which are second order in small quantites. We also neglect the k dependence of the exchange field and dipole-dipole effects. Just as in the ferromagnetic case, these two will simply provide a density of states for some value (or values) of the wave number k which are degenerate with the uniform mode,[9] and in this sense will cancel. On writing the equations of motion in this manner and equating spatially varying terms on each side (the equations for the constant terms determine the resonance frequency of the $k=0$ modes, and will not be considered further here) one finds, for zero applied field

$$\frac{1}{\gamma}\delta\dot{m}_{1x'}=(t_1-w_1)\delta m_{1y'}+h_1\delta m_{2y''}-t_1m_{1y'}\delta m_{2z''}+(h_1m_{2y''}-w_1m_{1z'})\delta m_{1z'},$$
$$\frac{1}{\gamma}\delta\dot{m}_{1y'}=(-t_1+w_1-d_1)\delta m_{1x'}-a_1\delta m_{2x''}+t_1m_{1x'}\delta m_{2z''}+[-a_1m_{2x''}+(w_1-d_1)m_{1x'}]\delta m_{1z'},$$
$$\frac{1}{\gamma}\delta\dot{m}_{2x''}=(-t_1+w_1)\delta m_{2y''}-h_1\delta m_{1y'}-t_1m_{2y''}\delta m_{1z'}+(h_1m_{1y'}-w_1m_{2y''})\delta m_{2z''},$$
$$\frac{1}{\gamma}\delta\dot{m}_{2y''}=(t_1-w_1+d_1)\delta m_{2x''}+a_1\delta m_{1x'}+t_1m_{2x''}\delta m_{1z'}+[-a_1m_{1x'}+(w_1-d_1)m_{2x''}]\delta m_{2z''}.$$

For simplicity we have dropped all terms containing products of the form $m_i\delta m_j$. These first-order nonlinear terms cannot contribute to the premature saturation of the uniform mode for they will not frequency modulate spin waves degenerate with $k=0$ at twice their natural frequency; and, therefore, will not cause the parametric pumping necessary for the instability. However, as shown by Suhl,[4] such first-order terms can cause spin waves at half the driving frequency to go unstable. This first-order Suhl instability shows itself as a subsidiary absorption at an applied field several hundred oersteds below that required for the $k=0$ resonance. The first-order terms are, therefore, treated separately in Appendix 1; and the conditions for the onset of a subsidiary absorption are discussed there. The above equations are easily rewritten in the more

[9] R. Loudon and P. Pincus (to be published).

convenient symmetric forms

$$\frac{1}{\gamma}\delta\dot{m}_1^+ = -iK\delta m_1^+ - i\frac{d}{2}\delta m_1^- - ih\delta m_2^+ - i\epsilon H_E\theta_c^2\delta m_2^- + itm_1^+\delta m_{2z''} + i\left(-hm_2^+ - \epsilon H_E\theta_c^2 m_2^- + wm_1^+ - \frac{d}{2}m_1^-\right)\delta m_{1z'},$$

$$\frac{1}{\gamma}\delta\dot{m}_2^+ = iK\delta m_2^+ + i\frac{d}{2}\delta m_2^- + ih\delta m_1^+ + i\epsilon H_E\theta_c^2\delta m_1^- + itm_2^+\delta m_{1z'} + i\left(-hm_1^+ - \epsilon H_E\theta_c^2 m_1^- + wm_2^+ - \frac{d}{2}m_2^-\right)\delta m_{2z''},$$

where

$$K = t_1 - w_1 + \tfrac{1}{2}d_1,$$
$$w = w_1 - \tfrac{1}{2}d_1,$$
$$d = d_1,$$
$$t = t_1,$$
$$h = h_1 + \epsilon H_E\theta_c^2.$$

The angle θ_c is the equilibrium canting angle as determined by setting the torque on each sublattice equal to zero. ϵ equals 1 for the single-ion anisotropy case, and equals (-1) when the canting is due to anisotropic exchange.

The variables δm^+, δm^-, m^+, and m^- are not the normal mode variables for the problem even in the absence of the nonlinear terms. It is, therefore, convenient to first transform to the variables a_k^+, a_k^-, b_k^+, b_k^- which diagonalize the equations of motion in the absence of the nonlinear terms before considering the effect of these nonlinear terms on the resonance properties. To make this transformation we seek the matrix S such that

$$M' = SMS^{-1}$$

is diagonal, where M is the resonance matrix

$$M = \begin{bmatrix} -iK & -id/2 & -ih & -i\epsilon H_E\theta_c^2 \\ id/2 & iK & i\epsilon H_E\theta_c^2 & ih \\ iK & id/2 & ih & i\epsilon H_E\theta_c^2 \\ -id/2 & -iK & -i\epsilon H_E\theta_c^2 & -ih \end{bmatrix}.$$

The normal mode variables are then defined by the matrix equation

$$\begin{bmatrix} a_k^+ \\ a_k^- \\ b_k^+ \\ b_k^- \end{bmatrix} = S \begin{bmatrix} \delta m_1^+ \\ \delta m_2^+ \\ \delta m_1^- \\ \delta m_2^- \end{bmatrix}.$$

The transformation matrix S is given by

$$S = \tfrac{1}{2}\begin{bmatrix} 1-\eta_1 & 1+\eta_1 & 1+\eta_1 & 1-\eta_1 \\ 1+\eta_1 & 1-\eta_1 & 1-\eta_1 & 1+\eta_1 \\ 1-\eta_2 & 1+\eta_2 & -1-\eta_2 & -1+\eta_2 \\ 1+\eta_2 & 1-\eta_2 & -1+\eta_2 & -1-\eta_2 \end{bmatrix}$$

and

$$S^{-1} = \tfrac{1}{4}\begin{bmatrix} 1-\dfrac{1}{\eta_1} & 1+\dfrac{1}{\eta_1} & 1-\dfrac{1}{\eta_2} & 1+\dfrac{1}{\eta_2} \\ 1+\dfrac{1}{\eta_1} & 1-\dfrac{1}{\eta_1} & 1+\dfrac{1}{\eta_2} & 1-\dfrac{1}{\eta_2} \\ 1+\dfrac{1}{\eta_1} & 1-\dfrac{1}{\eta_1} & -1-\dfrac{1}{\eta_2} & -1+\dfrac{1}{\eta_2} \\ 1-\dfrac{1}{\eta_1} & 1+\dfrac{1}{\eta_1} & -1+\dfrac{1}{\eta_2} & -1-\dfrac{1}{\eta_2} \end{bmatrix},$$

where

$$\eta_1 = \left(\frac{A+B}{C+D}\right)^{1/2}, \quad \eta_2 = \left(\frac{A-B}{C-D}\right)^{1/2},$$

$$A = K-h, \quad B = \epsilon H_E\theta_c^2 - \tfrac{1}{2}d,$$
$$C = K+h, \quad D = \epsilon H_E\theta_c^2 + \tfrac{1}{2}d.$$

The subscripts k denote that a_k^+, a_k^-, b_k^+, b_k^- are related to the deviation variables δm_i^+, δm_j^-. We shall denote the uniform mode variables a_0^+, a_0^-, b_0^+, b_0^-. The a's and b's are the normal coordinates for the problem and are equivalent to the spin-wave variables which diagonalize the Hamiltonian. In terms of these variables, the equations of motion become (neglecting nonlinear terms)

$$\frac{1}{\gamma}\dot{a}_k^+ = i[(A+B)(C+D)]^{1/2}a_k^+ = i\frac{\omega_1}{\gamma}a_k^+,$$

$$\frac{1}{\gamma}\dot{b}_k^+ = i[(A-B)(C-D)]^{1/2}b_k^+ = i\frac{\omega_2}{\gamma}b_k^+.$$

For a simple antiferromagnet, the two frequencies are degenerate and equal to

$$\omega_1 = \omega_2 = \gamma(2H_EH_A)^{1/2},$$

as shown by Keffer and Kittel.[10] When the canting interaction is included, the degeneracy is removed and one finds

$$\omega_1 = \gamma(4H_{A_2}^2 + 2H_EH_{A_1})^{1/2},$$
$$\omega_2 = \gamma(H_{A_2}^2 + 2H_EH_{A_1})^{1/2}$$

in the case of single-ion anisotropy, and

$$\omega_1 = \gamma(2H_EH_{A_1})^{1/2},$$
$$\omega_2 = \gamma(D^2 + 2H_EH_{A_1})^{1/2}$$

[10] F. Keffer and C. Kittel, Phys. Rev. 85, 329 (1952).

for anisotropic exchange. The above frequencies are equal to the uniform mode frequencies for we have assumed degenerate spin waves. We now go back to the original equations of motion, including the nonlinear terms, and rewrite them in terms of the spin-wave variables. In performing this transformation, we note that to first order in $1/\eta \simeq (H_{A_1}/H_E)^{1/2}$

$$m_1^+ = m_2^+ = \tfrac{1}{4}[a_0^+ + a_0^- + b_0^+ + b_0^-],$$
$$m_1^- = m_2^- = \tfrac{1}{4}[a_0^+ + a_0^- - b_0^+ - b_0^-],$$

so that we may drop the subscript on m_i^+, m_j^+ in the nonlinear terms. The transformed equations have the form

$$\frac{1}{\gamma}\dot{a}_k^+ = i[(A+B)(C+D)]^{1/2} a_k^+ - \frac{i}{8}[U(m^+ - m^-)^2 \eta_1^{-1} + Y(m^+ + m^-)^2 \eta_1] a_k^- + \frac{i}{8}[U(m^+ - m^-)^2 \eta_1^{-1} - Y(m^+ + m^-)^2 \eta_1] a_k^+,$$

$$\frac{1}{\gamma}\dot{b}_k^+ = i[(A-B)(C-D)]^{1/2} b_k^+ + \frac{i}{8}[V(m^+ + m^-)^2 \eta_2^{-1} + X(m^+ - m^-)^2 \eta_2] b_k^- - \frac{i}{8}[V(m^+ + m^-)^2 \eta_2^{-1} - X(m^+ - m^-)^2 \eta_2] b_k^+,$$

with

$$U = t_1 - h_1 + w_1,$$
$$V = t_1 - h_1 - 2H_E\theta_c^2 + w_1 - d_1,$$
$$X = t_1 - w_1 + h_1,$$
$$Y = t_1 - w_1 + d_1 + h_1 + 2H_E\theta_c^2.$$

Since the a_k and b_k are the normal mode variables, they can be excited independently of one another. Therefore, we may set $b_k^+ = b_k^- = 0$ in the a_k^+ equation; and $a_k^+ = a_k^- = 0$ in the b_k^+ equation. Substituting for $(m^+ + m^-)$, and $(m^+ - m^-)$ one finds to first order in $1/\eta$.

$$\dot{a}_k^+ = i\omega_1 a_k^+ - \frac{i}{32}\omega_1 (a_0^+ + a_0^-)^2 (a_k^+ + a_k^-),$$

$$\dot{b}_k^+ = i\omega_2 b_k^+ + \frac{i}{32}\omega_2 (b_0^+ + b_0^-)^2 (b_k^+ + b_k^-).$$

We now drop all nonlinear terms other than those which vary as $e^{i\omega t}$, for the other terms average to zero over a period and hence will have no net effect. Terms of the form $|a_0^+|^2 a_k^+$ and $|b_0^+|^2 b_k^+$ are also thrown away for these are secular and simply serve to shift the resonant frequencies slightly. Taking

$$a_k^+ = (a_k^0) e^{i\omega_1 t}, \quad b_k^+ = (b_k^0) e^{i\omega_2 t},$$

one finds

$$\dot{a}_k^0 = \frac{i}{32}(a_0^0)^2 \omega_1 (a_k^0)^*,$$

$$\dot{b}_k^0 = -\frac{i}{32}(b_0^0)^2 \omega_2 (b_k^0)^*.$$

Eliminating $(a_k^0)^*$ and $(b_k^0)^*$, respectively, by using the complex conjugate equations, one obtains

$$\ddot{a}_k^0 = (\tfrac{1}{32})^2 |a_0^0|^4 \omega_1^2 a_k^0,$$
$$\ddot{b}_k^0 = (\tfrac{1}{32})^2 |b_0^0|^4 \omega_2^2 b_k^0.$$

Just as in the ferromagnetic case, the solutions indicate that the spin-wave amplitudes are exponentially growing. However, we have neglected the spin-wave losses up to this point. We may formally put in these losses without reference to their microscopic origin by adding terms $-(\Delta\omega_k/\gamma)a_k^0$ and $-(\Delta\omega_k/\gamma)b_k^0$, respectively, to the above equations for a_k^0 and b_k^0, where

$$\Delta\omega_k = \gamma \Delta H_k,$$

with ΔH_k being the spin-wave "linewidth." Thus, the solution for the spin-wave amplitudes are exponentially growing only if

$$\tfrac{1}{32}|a_0^0|^2 \omega_1 > \gamma \Delta H_{k_1},$$
$$\tfrac{1}{32}|b_0^0|^2 \omega_2 > \gamma \Delta H_{k_2}.$$

The criteria for instability are

$$|a_0^0| = (32)^{1/2} [\Delta H_k / \omega_1 / \gamma]^{1/2},$$
$$|b_0^0| = (32)^{1/2} [\Delta H_k / \omega_2 / \gamma]^{1/2}.$$

Physically, the interpretation here is exactly analogous to that of the instability in ferromagnetic materials.[3] When the uniform mode reaches a critical amplitude, the effect of the nonlinear terms is to cause an exponential growth of the degenerate higher $-k$ spin waves. This exponential growth acts as a loss to the uniform mode and essentially holds its amplitude fixed at the critical value. Therefore, an increase in rf field does not yield a corresponding increase in transverse moment, since the higher k spin waves have no net transverse moment. This produces an apparent saturation of the uniform mode rf susceptibility in a magnetic resonance experiment.

III. CRITICAL RF FIELDS FOR INSTABILITY

In order to evaluate the critical fields for instability one must determine the uniform mode amplitudes as a function of rf field. Using the transformation matrix given in Sec. II,

$$a_0^+ = \tfrac{1}{2}[(m_1^+ + m_2^+ + m_1^- + m_2^-) - \eta_1 (m_1^+ - m_2^+ - m_1^- + m_2^-)],$$
$$b_0^+ = \tfrac{1}{2}[(m_1^+ + m_2^+ - m_1^- - m_2^-) - \eta_2 (m_1^+ - m_2^+ + m_1^- - m_2^-)],$$

or, in terms of the components along the coordinate axes

$$a_0^+ = (m_{1x'} + m_{2x''}) - i\eta_1(m_{1y'} - m_{2y''}),$$
$$b_0^+ = i(m_{1y'} + m_{2y''}) - \eta_2(m_{1x'} - m_{2x''}).$$

Noting that the two coordinate systems are defined by $\theta_1 = \theta_2 = \theta_c$; $\varphi_1 = 0$ $\varphi_2 = \pi$ at equilibrium, we have that

$$a_0^+ \simeq 2(\Delta\theta)[1-i],$$
$$b_0^+ \simeq 2(\Delta\theta)[1-i],$$

and

$$|a_0^0| = |b_0^0| = 2\sqrt{2}(\Delta\theta),$$

where $(\Delta\theta)$ is the angle through which each sublattice is tipped from its equilibrium axis by the resonant rf field, h_1. Note that we have used the fact that to first order in $1/\eta$, $m_1^+ = m_2^+$ so that the angles $\Delta\theta_1$ and $\Delta\theta_2$ are equal. To find $\Delta\theta$ as a function of rf field, one must solve the uniform mode equations of motion with the rf field included and calculate the rf susceptibility. Keffer and Kittel[10] have done this for a uniaxial antiferromagnet. Using their results, one finds

$$\Delta\theta \simeq h_1/2\Delta H_0,$$

where h_1 is the rf field and ΔH_0 is the linewidth of the uniform precession. The same result carries over to the canted antiferromagnetic case.[11] Thus, we have

$$|a_0^0| = |b_0^0| = \sqrt{2}(h_1/\Delta H_0),$$

and the critical rf field for instability is given by

$$h_{1\text{crit}} = 4\Delta H_0[\gamma \Delta H_{k_1}/\omega_1]^{1/2},$$
$$h_{2\text{crit}} = 4\Delta H_0[\gamma \Delta H_{k_2}/\omega_2]^{1/2},$$

with ω_1 and ω_2 being the resonance frequencies as calculated above. For the simplest case, the uniaxial antiferromagnet,

$$h_{\text{crit}} = 4\Delta H_0[\Delta H_k/(2H_E H_{A_1})^{1/2}]^{1/2}$$

indicating the possibility of unusually low critical fields in antiferromagnetic resonance.[12]

Since most resonance experiments are performed in a nonzero dc magnetic field, the effect of such a field on the instability must be considered. Clearly the dc field itself cannot give rise to nonlinear terms, consequently its effect is limited to changing the magnitudes of nonlinear terms by changing the equilibrium canting angles. Therefore, for antiferromagnets with large exchange fields, the instability criteria developed above will be relatively insensitive to applied fields.

IV. EXPERIMENTAL EVIDENCE OF SPIN-WAVE INSTABILITY IN $KMnF_3$

Static torsion measurements[13] on a single crystal of $KMnF_3$ have shown that this material is a canted antiferromagnet below 81.5°K. Detailed x-ray measurements[14] have determined the space group symmetry as $D_{2h}^{16}P_{bnm}$. This symmetry has been shown[13] to allow canting interactions of the type considered in Secs. II and III. Comparison of the experimental value of the weak moment with Pearson's theory[15] of single-ion anisotropy in $KMnF_3$ suggests that the canting is the result of such anisotropy rather than anisotropic exchange.

In the course of studying the antiferromagnetic resonance in $KMnF_3$ in the low-temperature region from 1.8 to 4.2°K, an anomalous power dependence of the rf susceptibility was observed. This power dependence is shown for one of the resonance lines in Fig. 2. For very low powers the susceptibility is independent of power as one would expect for a linear system. Then the susceptibility is observed to increase, and finally a saturation effect is observed. The magnitude of the rf field at the sample was determined by measuring the power absorbed at 90°K, just above the antiferromagnetic transition temperature. Since the static susceptibility is known[16] and the linewidth directly measured, the rf field at the sample was obtained from the relation

$$P_s = \tfrac{1}{2}\omega\chi'' H_1^2.$$

The effect of increased Q at low temperature was taken into account. At 1.8°K, the onset of *saturation* corresponds to an rf field such that $h_1/\Delta H \simeq 5\times 10^{-3}$, where

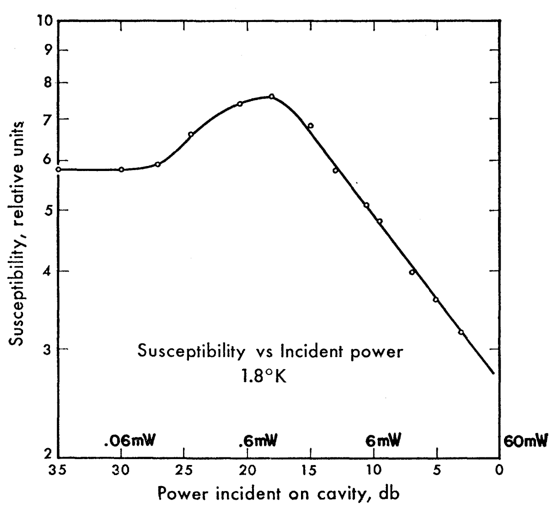

FIG. 2. Rf susceptibility versus incident power for $KMnF_3$ at 1.8°K. 0 dB corresponds to 60 mW of microwave power incident on the cavity. At this level the sample absorption is approximately 1 mW.

[11] A. J. Heeger, thesis, University of California, Berkeley, 1961 (unpublished).

[12] Equation (7) of Ref. 5 is in error. It should read

$$|a_0^0| = \frac{\sqrt{2}}{\epsilon}\left\{\frac{\Delta H_k}{(2H_E H_A)^{1/2}}\right\}^{1/2}.$$

The resulting critical field is then the same as that obtained here.

[13] A. J. Heeger, Olaf Beckman, and A. M. Portis, Phys. Rev. **123**, 1652 (1961).

[14] Olaf Beckman and Kerro Knox, Phys. Rev. **121**, 376 (1961).

[15] J. J. Pearson, Phys. Rev. **121**, 695 (1961).

[16] R. L. Martin, R. S. Nyholm, and N. C. Stephenson, Chem. Ind. (London) **1956**, 83 (1956); Shinji Ogawa, J. Phys. Soc. Japan, **14**, 1115 (1959); Kinishiro Hirawa, Kazuyoshi Hirakawa, and Takasu Hashimoto, *ibid.* **15**, 2063 (1960).

ΔH is the linewidth of the uniform mode. At higher temperatures in the liquid-helium region similar effects were observed. However, the three regions mentioned above were not so well defined, and the saturation did not set in until somewhat higher powers.

By using the field for resonance as an indicator of the spin temperature[17] of the electronic system, it is found that at maximum input power the spin temperature is raised above that of the helium bath by approximately 1°K. Since the Néel temperature for $KMnF_3$ is 88.3°K,[13] the magnitudes of the sublattice magnetization vectors are essentially unchanged by the rf power. Thus, the observed saturation occurs at a power level considerably below that required to change M_z by a significant amount.

If we now wish to interpret this saturation effect in terms of spin-wave instability, we may substitute experimental values for the above quantities and extract a value for the relaxation time of the higher k spin waves. The experiments were done at 10 k Mc/sec in an applied field. However, the applied field will not affect the instability criterion as discussed above. Using the results of Sec. II one obtains

$$\tau_k = 16(\Delta H/h_1)^2(1/\omega_1) \cong 10^{-5} \text{ sec},$$

which is equivalent to a spin-wave linewidth of

$$\Delta H_k = 5\times 10^{-3} \text{ Oe}.$$

This narrow spin-wave linewidth is to be contrasted with the relatively broad uniform mode width of 40 Oe. The fact that the observed linewidth of the uniform mode is roughly three orders of magnitude greater than the spin-wave linewidth suggests that the uniform mode is somehow statically broadened. This suggestion is consistent with the fact that the observed uniform mode width is independent of temperature.

Giordmaine[18] has noted that it is possible to measure the longitudinal relaxation time, T_1, in paramagnetic resonance by amplitude modulating the microwave power incident on the resonant cavity, and measuring the phase shift between the incident power and the resonance signal due to the sample. In a linear system, i.e., a system where the susceptibility is independent of incident power, the signal detected at the modulation frequency is proportional to $\chi''\Delta(H_1^2)$ and is clearly in phase with the modulation. However, in a nonlinear system the signal is proportional to $\chi''\Delta(H_1^2)+H_1^2\Delta\chi''$ and will not be in phase if the susceptibility, $\Delta\chi''$, cannot follow the modulation. Thus, the amplitude modulation technique is applicable to any resonant system in a region of nonlinear behavior; and measures the relaxation time of the saturation. In the case where the saturation is the result of spin-wave instability, τ_k, the spin-wave relaxation time, determines the response to amplitude modulated power. This may be seen in the following way. When the uniform mode is excited to the critical value, the magnetization vectors "stick." On increasing the rf field further, the excitation goes directly into the degenerate spin waves. Thus, a further increase in rf field does not increase the transverse moment so that an effective saturation of the rf susceptibility occurs. From this point of view, the $\Delta\chi''$ caused by the amplitude modulation results since there is no corresponding modulation of the transverse moment due to the instability. Only when the modulation frequency gets so high that the degenerate spin waves do not have time to react during the modulation cycle does the transverse moment begin to follow the modulation. Since the instability may be viewed as a driving of the degenerate spin waves by the uniform mode, the time needed for the spin waves to react is of the order of the relaxation time for these modes; i.e., $\tau_k = 1/\Delta H_k$. At low frequencies the spin-wave level will follow the modulation and $\Delta\chi''$ will be in phase. At high frequencies such that $\omega_M \gg 1/\tau_k$ the spin-wave level will remain constant during the modulation cycle and $\Delta\chi''$ will be zero; again the signal is in phase with the modulation. At frequencies $\omega_m \sim 1/\tau_k$, $\Delta\chi'' \neq 0$ and will not be in phase with the modulation. Thus, we expect a maximum phase shift when $\omega_m \sim 1/\tau_k$.

Let us consider this in somewhat more detail. The equations of motion for the spin-wave variables are rewritten (we consider only the a_k branch for brevity).

$$\dot{a}_k{}^+ = i\omega_k a_k{}^+ - i\nu_{k0}a_0{}^+ - ip_k(a_0{}^+)^2 a_k{}^- - \frac{1}{\tau_k}a_k{}^+,$$

where ω_k is the spin-wave frequency, $p_k = \frac{1}{32}\omega_1$, and τ_k is the spin-wave relaxation time. The term $i\nu_{k0}a_0{}^+$ expresses the coupling between the uniform mode and the degenerate spectrum due to scattering from lattice imperfections; ν_{k0} is the scattering matrix element. This scattering term must be included if one is interested in the detailed effect of the instability on the uniform mode. We assume that the rf field is amplitude modulated and has the form $h_1 + \Delta h_1 e^{i\omega_m t}$. The resulting spin-wave amplitudes are, therefore, also modulated and take the form $a_k{}^+ + \Delta a_k{}^+ e^{i\omega_m t}$. Then, the time-varying terms give

$$\left(i\omega_m + \frac{1}{\tau_k}\right)\Delta a_k{}^+ = -i\nu_{k0}\Delta a_0{}^+ - ip_k[2a_0{}^+ a_k{}^- \Delta a_0{}^+ + (a_0{}^+)^2 \Delta a_k{}^-].$$

Eliminating $\Delta a_k{}^-$ by using the complex-conjugate equation, we obtain

$$\Delta a_k{}^+ \simeq -i\frac{[\nu_{k0} + 2p_k a_0{}^+ a_k{}^-]\tau_k}{1+i\omega_m\tau_k}\Delta a_0{}^+.$$

[17] A. J. Heeger, A. M. Portis, D. Teaney, and G. Witt, Phys. Rev. Letters 7, 307 (1961).
[18] J. A. Giordmaine, Bull. Am. Phys. Soc. 5, 418 (1960).

The uniform mode amplitude is given by (see Sec. III)

$$\dot{a}_0^+ = i\omega_0 a_0^+ - i\sum_k \nu_{0k} a_k^+ - \frac{1}{\tau_0} a_0^+ + \sqrt{2}\gamma h_1.$$

Again, equating time-varying terms

$$\left(i\omega_m + \frac{1}{\tau_0}\right)\Delta a_0^+ = -i\sum_k \nu_{0k}\Delta a_k^+ + \sqrt{2}\gamma\Delta h_1,$$

and substituting the above value for Δa_k^+, we obtain

$$(1+i\omega_m\tau_0)\Delta a_0^+ = \sqrt{2}\frac{\Delta h_1}{\Delta H_0} - \tau_0\sum_k \nu_{0k}\frac{[\nu_{k0}+2p_k a_0^+ a_k^-]\tau_k}{1+i\omega_m\tau_k}\Delta a_0^+.$$

We assume that for the degenerate spin waves τ_k is approximately independent of k. This assumption should be particularly good for antiferromagnetic systems where dipole-dipole effects are small so that degeneracy occurs only over a relatively narrow range of k values. Defining

$$A = \sum_k \nu_{0k}(\nu_{k0} + 2p_k a_0^+ a_k^-),$$

we obtain

$$\Delta a_0^+ = \sqrt{2}\frac{\Delta h_1}{\Delta H_0}\frac{1}{(1+i\omega_m\tau_0) + A\tau_0\tau_k/(1+i\omega_m\tau_k)},$$

and there is a phase shift between Δh_1 and the uniform mode response, Δa_0^+. When $\tau_0 \ll \tau_k$, as appears to be the case for $KMnF_3$, the phase shift angle is given by

$$\tan\phi = A\tau_0\tau_k \frac{\omega_m\tau_k}{1+A\tau_0\tau_k+\omega_m^2\tau_k^2}$$

indicating a maximum phase shift when

$$\omega_m\tau_k = (1+A\tau_0\tau_k)^{1/2}$$

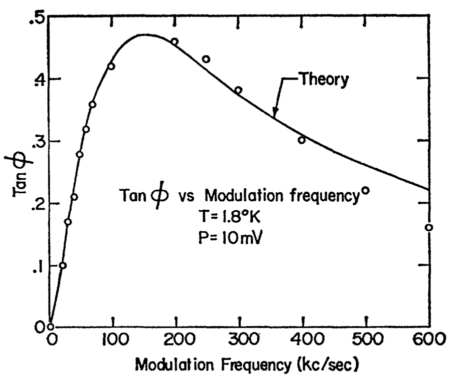

FIG. 3. The $\tan\phi$ versus modulation frequency, where ϕ is the phase shift of the signal relative to the amplitude modulated microwave power. The peak occurs at approximately 150 kc/sec indicating a spin-wave relaxation time of the order of 1.7×10^{-6} sec.

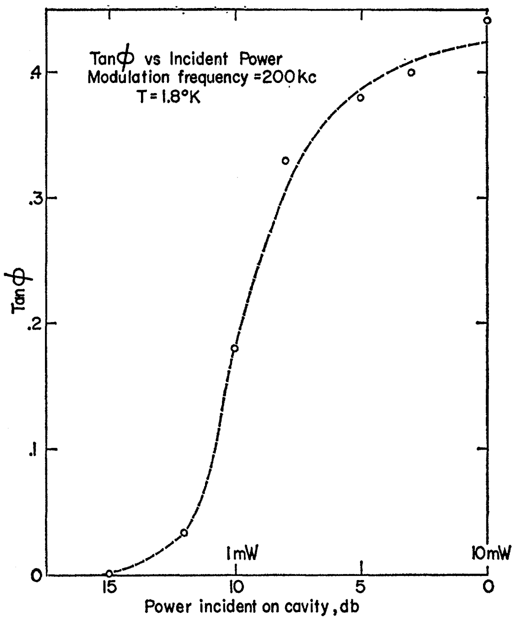

FIG. 4. The $\tan\phi$ versus microwave power at a fixed modulation frequency of 200 kc/sec. A rapid increase in the phase shift occurs at the instability threshold.

as expected from the physical arguments given above. The quantity A expresses the total coupling between the uniform mode and the degenerate spectrum and is expected to increase sharply above the instability threshold.

Figure 3 shows data, taken with a low Q ($Q\sim10^3$) cavity, of $\tan\phi$ versus frequency indicating a maximum phase shift at a frequency of approximately 150 kc/sec. These data were taken with an X-band microwave spectrometer of conventional design. The amplitude-modulated rf was obtained[19] by inserting into the line a hybrid junction with one port going to a matched load, the second to a crystal detector, the third to the klystron, and the fourth to the cavity containing the sample. By applying an external sine wave voltage to the crystal and thereby changing its impedance periodically, a modulated rf output was obtained. The solid curve in Fig. 3 is a fit of the above expression for $\tan\phi$ to the data. The best fit gives $A\tau_0\tau_k = 1.49$, $\tau_k = 1.7\times10^{-6}$ sec. The fit is quite good except at the highest frequencies where the accuracy is poorest and phase shift due to the Q of the cavity may be of importance. This value for τ_k is in reasonable agreement with that determined from the saturation curve considering the fact that the "critical field" is not well defined by the susceptibility curve (Fig. 2).

The above theory predicts a large increase in the phase shift observed at a given frequency when the spin system is taken above the threshold for instability. In Fig. 4 we show data of $\tan\phi$ versus power taken at

[19] A. M. Portis (prviate communication).

$f=200$ kc/sec. At low rf levels no phase shift is detected. However, above a critical power the phase shift becomes appreciable. This critical power level is well defined and appears to be a good indicator of the instability threshold.

Thus, we may conclude that the observed decrease in rf susceptibility in $KMnF_3$ is due to spin-wave instability. However, two comments should be made. Firstly, the instability mechanism would not appear capable of explaining the initial increase in rf susceptibility as shown in Fig. 2. Secondly, on the basis of the physical arguments made above suggesting that the uniform mode amplitude is held fixed at the critical value, one expects the rf susceptibility to fall off like $1/h_1$; whereas the experiments indicate a less rapid falloff. This discrepancy very likely arises from the lack of the inclusion of uniform mode scattering in a detailed calculation of the susceptibility above the threshold.[20]

CONCLUSION

We have shown that nonlinear terms in the equations of motion for antiferromagnetic resonance lead to instability of spin waves degenerate with the uniform mode when the rf driving field exceeds a critical value. The source of the nonlinear terms which cause this instability is found to be in the anisotropy and exchange energies. The instability criteria indicate that extremely low-level saturation is to be expected in antiferromagnets with high anisotropy.

Experimental evidence for the existence of spin-wave instability in the canted antiferromagnet, $KMnF_3$, is presented and discussed. The anomalous low-level saturation of the rf susceptibility is attributed to spin-wave instability. An independent measurement of the spin-wave relaxation time confirms this interpretation. The data are in general agreement with the instability theory developed in this paper. The spin-wave linewidth, ΔH_k, is found to be approximately three orders of magnitude narrower than the uniform mode linewidth; indicating that the broadening mechanisms are grossly different in the two cases.

ACKNOWLEDGMENTS

Many valuable discussions with Professor A. M. Portis and Professor P. Pincus are gratefully acknowledged.

APPENDIX 1.

First-Order Suhl Instability in Antiferromagnets and Canted Antiferromagnets

We consider here the effect of first-order nonlinear terms on the resonance properties. The appropriate equations of motion including first-order nonlinear terms are

$$\frac{1}{\gamma}\delta\dot{m}_{1x'}=(t_1-w_1)\delta m_{1y'}+h_1\delta m_{2y''}-u_1m_{1y'}\delta m_{1x'}-(u_1m_{1x'}+r_1m_{2x''})\delta m_{1y'}-r_1m_{1y'}\delta m_{2x''},$$

$$\frac{1}{\gamma}\delta m_{1y'}=(-t_1+w_1-d_1)\delta m_{1x'}-a_1\delta m_{2x''}+(r_1m_{2x''}+2u_1m_{1x'})\delta m_{1x'}+r_1m_{1x'}\delta m_{2x''}-c_1(\delta m_{1z'}+\delta m_{2z''}),$$

$$\frac{1}{\gamma}\delta m_{2x''}=(-t_1+w_1)\delta m_{2y''}-h_1\delta m_{1y'}-u_1m_{2x''}\delta m_{1x'}-(u_1m_{2x''}+r_1m_{1x'})\delta m_{2y'}+r_1m_{2y''}\delta m_{1x'},$$

$$\frac{1}{\gamma}\delta m_{2y''}=(t_1-w_1+d_1)\delta m_{2x''}+a_1\delta m_{1x'}+(r_1m_{1x'}+2u_1m_{2x''})\delta m_{2x''}+r_1m_{2x''}\delta m_{1x'}+c_1(\delta m_{1z'}+\delta m_{2z''}).$$

For a simple antiferromagnet, and for a canted system where the canting is due to anisotropic exchange, the coefficients c_1, r_1, and u_1 are all zero; and no first-order instability is expected. When single-ion anisotropy causes the canting

$$u_1=c_1=r_1=H_{A_2}.$$

Substituting this value into the above equations and transforming to the normal mode variables with the help of the transformation matrix in the text, one finds

$$\dot{a}_k{}^+=i\omega_{1k}a_k{}^++\frac{1}{4}\gamma H_{A_2}\frac{1}{\eta_1}(b_0{}^++b_0{}^-)(a_k{}^--a_k{}^+),$$

$$\dot{b}_k{}^+=i\omega_{2k}b_k{}^++\frac{i}{4}\gamma H_{A_2}\frac{1}{\eta_2}(b_0{}^++b_0{}^-)(b_k{}^--b_k{}^+),$$

taking $b_0{}^+=b_0{}^0e^{i\omega_0t}$, $a_k{}^+=a_k{}^0e^{i\omega_{1k}t}$, $b_k{}^+=b_k{}^0e^{i\omega_{2k}t}$. We now want to drop all terms other than those which vary as $e^{i\omega_{1k}t}$ in the first equation; and all except those which vary as $e^{i\omega_{2k}t}$ in the second equation. There are no such terms unless

$$\omega_0=2\omega_{1k}\quad\text{or}\quad\omega_0=2\omega_{2k}.$$

These conditions are possible since ω_k is a monotone increasing function of k for such a system. Thus, for some value of k, the above conditions can be fulfilled. However, it is clear that this will occur far off resonance for the uniform mode. The instability criterion, then, is

$$\frac{1}{4}\frac{\gamma H_{A_2}}{\eta_1}|b_0{}^0|>\gamma\Delta H_k,\quad |b_0{}^0|>4\frac{H_E}{H_{A_2}}\left(\frac{\Delta H_k}{H_{A_2}}\right).$$

This criterion will be quite difficult if not impossible to achieve, expecially since one must drive the $k=0$ mode far off resonance. Thus, we conclude that the first-order instability is not expected to be important even in the single-ion-anisotropy case.

[20] H. Suhl, J. Appl. Phys. **30**, 1961 (1959).

APPENDIX 2

Coefficients for the Single-Ion-Anisotropy Case

The Hamiltonian appropriate to this system has the form[9] (for no external field),

$$H=\lambda \mathbf{M}_1\cdot\mathbf{M}_2-\frac{K_1}{2M^2}[(M_1{}^z)^2+(M_2{}^z)^2]+\frac{K_2}{M^2}(M_1{}^xM_1{}^z-M_2{}^xM_2{}^z).$$

By changing the sublattice magnetization vectors by arbitrary small amounts, one finds the effective fields on the two sublattices

$$\mathbf{H}_{\text{eff}}{}^{(1)}=-H_E\mathbf{m}_2+H_{A_1}m_1{}^z\mathbf{z}-H_{A_2}(m_1{}^x\mathbf{z}+m_1{}^z\mathbf{x}),$$
$$\mathbf{H}_{\text{eff}}{}^{(2)}=-H_E\mathbf{m}_1+H_{A_1}m_2{}^z\mathbf{z}+H_{A_2}(m_2{}^x\mathbf{z}+m_2{}^z\mathbf{x}).$$

To obtain the coefficients one must now transform to the new coordinate systems x', y', z'; x'', y'', z''. The results are

$$a_1=H_E[\cos\theta_1\cos\theta_2\cos(\phi_2-\phi_1)+\sin\theta_2\sin\theta_1],$$
$$b_1=H_E\cos\theta_1\sin(\phi_1-\phi_2),$$
$$c_1=H_E[\cos\theta_1\sin\theta_2\cos(\phi_2-\phi_1)-\cos\theta_2\sin\theta_1],$$
$$d_1=-H_{A_2}\sin2\theta_1\cos\phi_1-H_{A_1}\sin^2\theta_1,$$
$$e_1=H_{A_2}\sin\theta_1\sin\phi_1,$$
$$f_1=H_{A_2}\cos\phi_1\cos2\theta_1+H_{A_1}\sin\theta_1\cos\theta_1,$$
$$g_1=H_E\cos\theta_2\sin(\phi_2-\phi_1)$$
$$h_1=H_E\cos(\phi_2-\phi_1),$$
$$j_1=H_E\sin\theta_2\sin(\phi_2-\phi_1),$$
$$k_1=H_{A_2}\sin\theta_1\sin\phi_1,$$
$$p_1=0,$$
$$q_1=-H_{A_2}\cos\theta_1\sin\phi_1,$$
$$r_1=H_E[\cos\theta_2\sin\theta_1\cos(\phi_2-\phi_1)-\sin\theta_2\cos\theta_1],$$
$$s_1=H_E\sin\theta_2\sin(\phi_1-\phi_2),$$
$$t_1=H_E[\sin\theta_2\sin\theta_1\cos(\phi_2-\phi_1)+\cos\theta_2\cos\theta_1],$$
$$u_1=H_{A_2}\cos\phi_1\cos2\theta_1+H_{A_1}\sin\theta_1\cos\theta_1,$$
$$v_1=-H_{A_2}\cos\theta_1\sin\phi_1,$$
$$w_1=H_{A_2}\cos\phi_1\sin2\theta_1-H_{A_1}\cos^2\theta_1.$$

The coefficients a_2, b_2, $\cdots w_2$ are similar except one must interchange subscripts 1 and 2 on all angles and furthermore the sign of H_{A_2} must be changed. $H_E=\lambda M$, $H_{A_1}=K_1/M$, and $H_{A_2}=K_2/M$. The equilibrium values of θ_1, ϕ_1, θ_2, ϕ_2 are found by setting the torque on each sublattice equal to zero. The resulting conditions are

$$-j_1+q_1=0,$$
$$-c_1+f_1=0.$$

APPENDIX 3

Coefficients for the Anisotropic Exchange Case

The appropriate Hamiltonian is

$$H=\lambda\mathbf{M}_1\cdot\mathbf{M}_2-\frac{K_1}{2M^2}(M_1{}^2+M_{2z}{}^2)+\frac{D}{M}(M_{1x}M_{2y}-M_{1y}M_{2x}).$$

This leads to the following coefficients:

$$a_1=H_E[\cos\theta_1,\cos\theta_2\cos(\phi_2-\phi_1)+\sin\theta_2\sin\theta_1]-D[\cos\theta_1\sin\theta_2\cos\phi_1-\sin\theta_1\cos\theta_2\cos\phi_2],$$
$$b_1=H_E\cos\theta_1\sin(\phi_1-\phi_2)-D\sin\theta_1\sin\phi_2,$$
$$c_1=H_E[\cos\theta_1\sin\theta_2\cos(\phi_2-\phi_1)-\cos\theta_2\sin\theta_1],+D[\cos\theta_2\cos\theta_1\cos\phi_1+\sin\theta_2\sin\theta_1\cos\phi_2],$$
$$d_1=-H_{A_1}\sin^2\theta_1,$$
$$e_1=0,$$
$$f_1=H_{A_1}\sin\theta_1\cos\theta_1,$$
$$g_1=H_E\cos\theta_2\sin(\phi_2-\phi_1)+D\sin\phi_1\sin\theta_2,$$
$$h_1=H_E\cos(\phi_1-\phi_2),$$
$$j_1=H_E\sin\theta_2\sin(\phi_2-\phi_1)-D\cos\theta_2\sin\phi_2,$$
$$k_1=p_1=q_1=0,$$
$$r_1=H_E[\cos\theta_2\sin\theta_1\cos(\phi_2-\phi_1)-\sin\theta_2\cos\theta_1]-D[\sin\theta_1\sin\theta_2\sin\phi_1+\cos\theta_1\cos\theta_2\cos\phi_2],$$
$$s_1=H_E\sin\theta_1\sin(\phi_1-\phi_2)+D\cos\theta_1\sin\phi_2,$$
$$t_1=H_E[\sin\theta_2\sin\theta_1\cos(\phi_2-\phi_1)+\cos\theta_2\cos\theta_1]+D[\sin\theta_1\cos\theta_2\cos\phi_1-\cos\theta_1\sin\theta_2\cos\phi_2],$$
$$u_1=H_{A_1}\sin\theta_1\cos\theta_1,$$
$$v_1=0,$$
$$w_1=-H_{A_1}\cos^2\theta_1.$$

The equilibrium conditions are, once again

$$-j_1+q_1=0,$$
$$-c_1+f_1=0.$$