

Nonlinear infrared response of antiferromagnets

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We consider the nonlinear response of a uniaxial antiferromagnet, exposed to an elliptically polarized magnetic field of frequency Ω , with the field in the basal plane. Explicit expressions are obtained for the contribution to the magnetic susceptibility cubic in the Cartesian components of the applied field, which describe the response at frequency Ω . We use these to calculate, as a function of applied power, the transmissivity of a thin film. We show that such samples should exhibit bistability under conditions outlined.

I. INTRODUCTION

When a system of spins is exposed to intense radiation with frequency at or near its resonance frequency, the response of the system may be nonlinear. The best-known and most easily described mechanism is simple saturation of the resonance.¹ If we consider a two-level system, application of a sufficiently intense resonant field depletes the ground state, leading to a decrease of the effective absorption coefficient as the power increases.

In concentrated spin systems, such as ferromagnets, nonlinear behavior in the resonant response is found for power levels far below that required to saturate the resonance in the above sense. Many years ago, a theory of the microscopic origin of these low-power nonlinearities was put forward by Suhl,² in the recent literature, there is renewed interest in the area, because subharmonics and ultimately a chaotic response of the spin system are found, well beyond threshold.³ Suhl's instabilities have their physical origin in the buildup to nonthermal values of the population of short-wavelength spin waves, as power is applied to the spatially uniform main-resonance mode of the sample. The main resonance is coupled to such short-wavelength modes via nonlinear terms in the spin-wave Hamiltonian, and at elevated power levels pumps energy into them more rapidly than it can be removed by relaxation processes, to produce the nonthermal populations which then react back on the uniform mode.

The discussion of saturation effects and Suhl's pumping instabilities has been focused on the microwave-frequency regime. Here, in a typical experiment, the wavelength of the radiation is very long compared to the sample size. To excellent approximation, the exciting field may be regarded as spatially uniform, and throughout the discussion, retardation or propagation effects can be ignored in the description of the interaction of the electromagnetic field with the sample.

Antiferromagnets have resonance frequencies which lie in the infrared, and as noted some years ago,⁴ interesting propagation effects can influence the electromagnetic response of these materials, within which the wavelength of the resonant electromagnetic radiation can be small compared to the sample size. The electromagnetic normal modes are in fact polaritons, where strong dispersion is

present by virtue of the resonance in the magnetic-susceptibility tensor produced by coupling of the electromagnetic field to the antiferromagnetic resonance. Toussaint *et al.* have reported clear evidence for magnetic polaritons in their experimental study of Mn-doped FeF₂.⁵

In this paper, we discuss the nonlinear response of antiferromagnetically ordered spin systems. We suppose the material is exposed to an oscillatory magnetic field with Cartesian components in the basal plane, perpendicular to the anisotropy axis. If the field has frequency Ω , we obtain expressions for the contributions to the magnetization induced in the sample that are cubic in the field amplitudes, also with frequency Ω .

With these results in hand we may calculate the transmissivity of a thin film as a function of incident power. For a film of uniaxial material such as FeF₂ in zero external field, with c axis normal to the surface, this may be done through application of an approach developed recently for the analysis of dielectric films and multilayer structures.^{6,7}

If the film thickness is chosen suitably, as discussed below, we find bistability in the infrared transmissivity. Thus, antiferromagnetic films may serve as nonlinear circuit elements in optical structures designed to operate in the far infrared. In this regard, note that films of the thickness required are now available which are of very high quality. The material used in Ref. 5 had an antiferromagnetic-resonance width of less than 20 G; this figure is an upper limit and not the measured value of the line width.

Viewed from the perspective of the literature on nonlinear instabilities in magnetic media,^{2,3} the bistability examined here may be viewed as a new form of instability whose existence depends on the role of retardation in the description of the electromagnetic response of the film. As remarked earlier, such effects are unimportant in the microwave response but, as we will see, they may play a substantial role at infrared frequencies.

This paper is organized as follows. Section II discusses the calculation of the nonlinear response of the antiferromagnetic spins. Section III examines the transmissivity of the thin film as a function of power, and Sec. IV is a summary and general discussion.

II. THE NONLINEAR MAGNETIC SUSCEPTIBILITY OF UNIAXIAL ANTIFERROMAGNETS

In this section, we derive expressions for the transverse elements of the nonlinear susceptibility tensor of a uniaxial antiferromagnet that can be described by the two-sublattice model.⁸ The materials MnF_2 , FeF_2 , and CoF_2 are examples of crystals which can be described by this model. These compounds form crystals of the rutile structure, where the body-centered metal ions form sublattice A , say, and the corner metal ions form sublattice B .

We begin with the assumption that the crystal is at a low temperature, and then the magnetization on each sublattice can be regarded as saturated. We define the $+z$ ($-z$) direction as the direction of the spontaneous magnetization of sublattice A (B). We let \mathbf{m}_A and \mathbf{m}_B be the magnetization of each sublattice, and m_s is the absolute value of the saturation magnetization associated with each sublattice.

To calculate the nonlinear contributions to the magnetic susceptibility, we place the spins in a spatially uniform magnetic field

$$\mathbf{h}(\hat{\mathbf{t}}) = \hat{\mathbf{x}}h_x \cos(\Omega t + \phi) + \hat{\mathbf{y}}h_y \sin(\Omega t + \phi),$$

polarized elliptically in the xy plane. The response of the spin system to this field may be calculated from the torque equations

$$\frac{d\mathbf{a}}{dt} = \gamma(\mathbf{a} \times \mathbf{H}_{\text{eff}}^{(A)}) \quad (2.1a)$$

and

$$\frac{d\mathbf{b}}{dt} = \gamma(\mathbf{b} \times \mathbf{H}_{\text{eff}}^{(B)}), \quad (2.1b)$$

where γ is the gyromagnetic ratio, $\mathbf{a} = \mathbf{m}_A/m_s$, $\mathbf{b} = \mathbf{m}_B/m_s$, with $\mathbf{H}_{\text{eff}}^{(A)}$ and $\mathbf{H}_{\text{eff}}^{(B)}$ the effective fields acting on sublattice A and B , respectively. Supposing the spins are placed in a dc external magnetic field of strength H_0 directed along the z axis, $\mathbf{H}_{\text{eff}}^{(A)}$ and $\mathbf{H}_{\text{eff}}^{(B)}$ are given by

$$\mathbf{H}_{\text{eff}}^{(A)} = \hat{\mathbf{z}}(H_0 + H_A a_z) - H_E \mathbf{b} + \mathbf{h}(t) \quad (2.2a)$$

and

$$\mathbf{H}_{\text{eff}}^{(B)} = \hat{\mathbf{z}}(H_0 + H_A b_z) - H_E \mathbf{a} + \mathbf{h}(t). \quad (2.2b)$$

Following the prescription used earlier,⁴ we assume the internal field operating on each sublattice consists of an anisotropy field of strength $H_A(m_z/m_s)$ for each sublattice, and the exchange field of strength H_E generated by spins on the opposite sublattice.

If we define the characteristic frequencies $\omega_0 = \gamma H_0$, $\omega_A = \gamma H_A$, and $\omega_E = \gamma H_E$, then the equations of motion for the various transverse-spin components become

$$\dot{a}_x = \omega_0 a_y + \omega_A a_y a_z + \omega_E (b_y a_z - a_y b_z) - \gamma h_y(t) a_z, \quad (2.3a)$$

$$\dot{a}_y = -\omega_0 a_x - \omega_A a_x a_z + \omega_E (a_x b_z - b_x a_z) + \gamma h_x(t) a_z, \quad (2.3b)$$

$$\dot{b}_x = \omega_0 b_y + \omega_A b_y b_z + \omega_E (a_y b_z - b_y a_z) - \gamma h_y(t) b_z, \quad (2.3c)$$

and

$$\dot{b}_y = -\omega_0 b_x - \omega_A b_x b_z + \omega_E (b_x a_z - a_x b_z) + \gamma h_x(t) b_z. \quad (2.3d)$$

Assuming a_x , a_y , b_x , and b_y are always small compared to unity, we can write

$$a_z = (1 - a_x^2 - a_y^2)^{1/2} \cong 1 - \frac{1}{2}(a_x^2 + a_y^2) + \dots \quad (2.4a)$$

and

$$b_z = -(1 - b_x^2 - b_y^2)^{1/2} \cong -1 + \frac{1}{2}(b_x^2 + b_y^2) + \dots \quad (2.4b)$$

Upon employing Eqs. (2.4), we may rewrite Eqs. (2.3) to read, with $a_1^2 = a_x^2 + a_y^2$ and $b_1^2 = b_x^2 + b_y^2$,

$$\dot{a}_x = (\omega_0 + \omega_A + \omega_E) a_y + \omega_E b_y - \gamma h_y - \frac{1}{2} \omega_E (a_y b_1^2 + b_y^2 a_1) - \frac{1}{2} \omega_A a_1^2 a_y + \frac{1}{2} \gamma a_1^2 h_y, \quad (2.5a)$$

$$\dot{a}_y = -(\omega_0 + \omega_A + \omega_E) a_x - \omega_E b_x + \gamma h_x + \frac{1}{2} \omega_E (a_x b_1^2 + b_x a_1^2) + \frac{1}{2} \omega_A a_1^2 a_x - \frac{1}{2} \gamma a_1^2 h_x, \quad (2.5b)$$

$$\dot{b}_x = (\omega_0 - \omega_A - \omega_E) b_y - \omega_E a_y + \gamma h_y + \frac{1}{2} \omega_E (a_y^2 b_1 + b_y a_1^2) + \frac{1}{2} \omega_A b_1^2 b_y - \frac{1}{2} \gamma b_1^2 h_y, \quad (2.5c)$$

and

$$\dot{b}_y = -(\omega_0 - \omega_A - \omega_E) b_x + \omega_E a_x - \gamma h_x - \frac{1}{2} \omega_E (a_x b_1^2 + b_x a_1^2) - \frac{1}{2} \omega_A b_1^2 b_x + \frac{1}{2} \gamma b_1^2 h_x. \quad (2.5d)$$

From Eqs. (2.5), we shall extract the components of the response at frequency Ω , which are cubic in the amplitudes of the applied field amplitudes h_x and h_y . These are the components of the response that will prove of interest in Sec. III. We seek solutions in the form

$$a_x(t) = a_x \cos(\Omega t + \phi) + a'_x \cos(3\Omega t + 3\phi) + \dots \quad (2.6a)$$

and

$$a_y(t) = a_y \sin(\Omega t + \phi) + a'_y \sin(3\Omega t + 3\phi) + \dots, \quad (2.6b)$$

and use identities such as

$$\cos(3x) = \frac{3}{4} \cos x + \frac{1}{4} \cos(3x)$$

to isolate the coefficients of the terms which describe the response of the system at the frequency Ω . This leads to the following set of algebraic equations:

$$\Omega a_x + (\omega_0 + \omega_A + \omega_E) a_y + \omega_E b_y = \gamma h_y [1 - \frac{1}{8}(a_x^2 + 3a_y^2)] + \frac{1}{8}\omega_A a_y (a_x^2 + 3a_y^2) + \frac{1}{8}\omega_E [a_y (b_x^2 + 3b_y^2) + b_y (a_x^2 + 3a_y^2)] , \quad (2.7a)$$

$$\Omega a_y + (\omega_0 + \omega_A + \omega_E) a_x + \omega_E b_x = \gamma h_x [1 - \frac{1}{8}(3a_x^2 + a_y^2)] + \frac{1}{8}\omega_A a_x (3a_x^2 + a_y^2) + \frac{1}{8}\omega_E [a_x (3b_x^2 + b_y^2) + b_x (3a_x^2 + a_y^2)] , \quad (2.7b)$$

$$\Omega b_x + (\omega_0 - \omega_A - \omega_E) b_y - \omega_E a_y = -\gamma h_y [1 - \frac{1}{8}(b_x^2 + 3b_y^2)] - \frac{1}{8}\omega_A b_y (b_x^2 + 3b_y^2) - \frac{1}{8}\omega_E [a_y (b_x^2 + 3b_y^2) + b_y (a_x^2 + 3a_y^2)] , \quad (2.7c)$$

and

$$\Omega b_y + (\omega_0 - \omega_A - \omega_E) b_x - \omega_E a_x = -\gamma h_x [1 - \frac{1}{8}(3b_x^2 + b_y^2)] - \frac{1}{8}\omega_A b_x (3b_x^2 + b_y^2) - \frac{1}{8}\omega_E [a_x (3b_x^2 + b_y^2) + b_x (3a_x^2 + a_y^2)] . \quad (2.7d)$$

In the end, we seek the net dynamic magnetic moment induced by the field, whose components are proportional to $a_x + b_x$, and $a_y + b_y$. Thus we introduce $\mathbf{t} = \mathbf{a} + \mathbf{b}$, and the staggered magnetization $\mathbf{d} = \mathbf{a} - \mathbf{b}$, and rewrite Eqs. (2.7) in terms of these two variables. The results are instructive. One finds that

$$\Omega t_x + \omega_0 t_y + \omega_A d_y = -\frac{\gamma}{8} h_y (t_x d_x + 3t_y d_y) + \frac{\omega_A}{32} (3d_y^3 + t_x^2 d_y + 2t_x t_y d_x + 9t_y^2 d_y + d_x^2 d_y) , \quad (2.8a)$$

$$\Omega t_y + \omega_0 t_x + \omega_A d_x = -\frac{\gamma h_x}{8} (3t_x d_x + t_y d_y) + \frac{\omega_A}{32} (3d_x^3 + t_y^2 d_x + 2t_x t_y d_y + 9t_x^2 d_x + d_y^2 d_x) , \quad (2.8b)$$

$$\begin{aligned} \Omega d_x + \omega_0 d_y + (\omega_A + 2\omega_E) t_y &= \gamma h_y [2 - \frac{1}{16}(t_x^2 + 3t_y^2 + d_x^2 + 3d_y^2)] \\ &+ \frac{1}{32}(\omega_A + 2\omega_E) t_y (t_x^2 + 3t_y^2 + d_x^2 + 3d_y^2) + \frac{1}{16}(\omega_A - 2\omega_E) d_y (t_x d_x + 3t_y d_y) , \end{aligned} \quad (2.8c)$$

and

$$\begin{aligned} \Omega d_y + \omega_0 d_x + (\omega_A + 2\omega_E) t_x &= \gamma h_x [2 - \frac{1}{16}(3t_x^2 + t_y^2 + 3d_x^2 + d_y^2)] \\ &+ \frac{1}{32}(\omega_A + 2\omega_E) t_x (3t_x^2 + t_y^2 + 3d_x^2 + d_y^2) + \frac{1}{16}(\omega_A - 2\omega_E) d_x (3t_x d_x + t_y d_y) . \end{aligned} \quad (2.8d)$$

From the general structure of these equations, one may appreciate the crucial role played by the anisotropy energy, here represented through the parameter ω_A . If ω_A is set to zero, one sees that in the linear response of the system, both h_x and h_y couple only to the staggered magnetization \mathbf{d} , and the net magnetization \mathbf{t} vanishes. The two exchange-coupled sublattices each precess around the external Zeeman field H_0 , always remaining strictly anti-parallel when $\omega_A = 0$. In this limit, the only nonlinear terms in the equations for \mathbf{t} which survive are those explicitly proportional to the driving fields h_x and h_y . Through these, one obtains a net transverse magnetization \mathbf{t} cubic in the amplitude of the applied field. However, if one considers the case $\omega_A \neq 0$, and frequencies close to that of the antiferromagnetic resonances, one may establish that the dominant contributions to the nonlinear susceptibility have their origin in the terms in Eqs. (2.8) explicitly proportional to ω_A . The role of anisotropy is thus crucial.

From Eqs. (2.8), one easily obtains expressions for the linear response of the system to the applied field. If $\mathbf{t}^{(0)}$ and $\mathbf{d}^{(0)}$ denote the response calculated in the linear limit, then

$$t_x^{(0)} = (X + Y)h_x + (X - Y)h_y , \quad (2.9a)$$

$$t_y^{(0)} = (X - Y)h_x + (X + Y)h_y , \quad (2.9b)$$

$$d_x^{(0)} = (\omega_+ X + \omega_- Y)h_x + (\omega_+ X - \omega_- Y)h_y , \quad (2.9c)$$

and

$$d_y^{(0)} = (\omega_+ X - \omega_- Y)h_x + (\omega_+ X + \omega_- Y)h_y . \quad (2.9d)$$

In these expressions, we have

$$X = \frac{\gamma \omega_A}{\Omega_0^2 - (\Omega + \omega_0)^2} , \quad (2.10a)$$

$$Y = \frac{\gamma \omega_A}{\Omega_0^2 - (\Omega - \omega_0)^2} , \quad (2.10b)$$

where $\Omega_0^2 = \gamma^2 (2H_E H_A + H_A^2)$ is the zero-field antiferromagnetic resonance frequency, and finally

$$\omega_{\pm} = \frac{(\omega_0^{\mp} \Omega)}{\omega_A} . \quad (2.10c)$$

The nonlinear contributions to the response follow from a perturbation theoretic analysis of Eqs. (2.8). One proceeds by writing

$$\mathbf{t} = \mathbf{t}^{(0)} + \mathbf{t}^{(1)} + \dots \quad (2.11a)$$

and

$$\mathbf{d} = \mathbf{d}^{(0)} + \mathbf{d}^{(1)} + \dots , \quad (2.11b)$$

where $\mathbf{t}^{(0)}$ and $\mathbf{d}^{(0)}$ describe the linear response, and $\mathbf{t}^{(1)}$ and $\mathbf{d}^{(1)}$ are the first nonlinear corrections, given by terms of the form $h_x^n h_y^m$, where $n + m = 3$. The first corrections are found from

$$\Omega t_x^{(1)} + \omega_0 t_y^{(1)} + \omega_A d_y^{(1)} = -\frac{1}{8} \gamma h_y (t_x^{(0)} d_x^{(0)} + 3t_y^{(0)} d_y^{(0)}) + \frac{1}{32} \omega_A (3d_y^{(0)3} + t_x^{(0)2} d_y^{(0)} + 9t_y^{(0)2} d_y^{(0)} + d_x^{(0)2} d_y^{(0)} + 2t_x^{(0)} t_y^{(0)} d_x^{(0)}), \quad (2.12)$$

and similar relations deduced from Eqs. (2.8b)–(2.8d).

We may write

$$t_x^{(1)} = \sum_{n=0}^3 \alpha_n h_x^{3-n} h_y^n, \quad (2.13a)$$

$$t_y^{(1)} = \sum_{n=0}^3 \beta_n h_x^{3-n} h_y^n \equiv \sum_{n=0}^3 \alpha_{3-n} h_x^{3-n} h_y^n, \quad (2.13b)$$

and

$$d_y^{(1)} = \sum_{n=0}^3 \xi_n h_x^{3-n} h_y^n = \sum_{n=0}^3 \eta_{3-n} h_x^{3-n} h_y^n. \quad (2.13c)$$

The identification of β_n with α_{3-n} , and ξ_n with η_{3-n} is a direct consequence of the invariance of the form of the response under rotations of the reference axes about the z direction.

It is a straightforward, but nonetheless tedious matter to obtain expressions for the coefficients α_n and η_n from the relations given above. We quote only the results, which are as follows:

$$\begin{aligned} \alpha_0 = \frac{XY}{\gamma^2 \omega_A^2} & \left[\frac{\gamma}{4} \{ [2\omega_0 \omega_+ (-\omega_0^2 + \Omega_0^2 + \Omega^2) + \omega_A (-\Omega_0^2 + \Omega^2 + \omega_0^2)(1 + \omega_+^2)] X^2 \right. \\ & + [\omega_0 (-\omega_0^2 + \Omega_0^2 + \Omega^2)(\omega_+ + \omega_-) + \omega_A (-\Omega_0^2 + \omega_0^2 + \Omega^2)(1 + \omega_+ \omega_-)] XY \\ & + [2\omega_0 \omega_- (-\omega_0^2 + \Omega_0^2 + \Omega^2) + \omega_A (-\Omega_0^2 + \omega_0^2 + \Omega^2)(1 + \omega_-^2)] Y^2 \} \\ & \left. + \frac{1}{8} \omega_A \{ [\Omega_0^2 - (\Omega - \omega_0)^2] (A_1 X^3 + A_3 XY^2) + [(\Omega + \omega_0)^2 - \Omega_0^2] (A_2 X^2 Y + A_4 Y^3) \} \right], \quad (2.14a) \end{aligned}$$

$$\begin{aligned} \alpha_1 = \frac{XY}{\gamma^2 \omega_A^2} & \left[\frac{\gamma}{4} \{ 2\omega_0 [2\omega_+ (\Omega^2 - \omega_0^2 + \Omega_0^2 + \Omega \omega_0) - \Omega \omega_A (1 + \omega_+^2)] + 2(\Omega_0^2 - \omega_0^2 - \Omega^2) [\Omega \omega_+ - \omega_A (1 + \omega_+^2)] \} X^2 \right. \\ & + \{ 2\Omega \omega_0 [\omega_A (1 + \omega_+ \omega_-) - \omega_0 (\omega_+ + \omega_-)] + \Omega (\Omega^2 + \omega_0^2 - \Omega_0^2) (\omega_+ + \omega_-) \} XY \\ & + \{ 2\omega_0 [2\omega_- (\omega_0^2 + \Omega \omega_0 - \Omega^2 - \Omega_0^2) - \Omega \omega_A (1 + \omega_-^2)] + 2(\Omega_0^2 - \omega_0^2 - \Omega^2) [\Omega \omega_- + \omega_A (1 + \omega_-^2)] \} Y^2 \\ & \left. + \frac{1}{8} \omega_A \{ [\Omega_0^2 - (\Omega - \omega_0)^2] (3A_1 X^3 - A_3 XY^2) + [(\Omega + \omega_0)^2 - \Omega_0^2] (A_2 X^2 Y - 3A_4 Y^3) \} \right], \quad (2.14b) \end{aligned}$$

$$\begin{aligned} \alpha_2 = \frac{XY}{\gamma^2 \omega^2} & \left[\frac{\gamma}{4} \{ 2\omega_0 [\omega_+ (\Omega^2 - \omega_0^2 + \Omega_0^2 + 4\Omega \omega_0) - 2\Omega \omega_A (1 + \omega_+^2)] + (\Omega_0^2 - \omega_0^2 - \Omega^2) [4\Omega \omega_+ - \omega_A (1 + \omega_+^2)] \} X^2 \right. \\ & + [\omega_0 (\omega_0^2 - \Omega^2 - \Omega_0^2) (\omega_+ + \omega_-) + \omega_A (\Omega_0^2 - \omega_0^2 - \Omega^2) (1 + \omega_+ \omega_-)] XY \\ & + \{ 2\omega_0 [\omega_- (\Omega^2 - \omega_0^2 + \Omega_0^2 + 4\Omega \omega_0) + 2\Omega \omega_A (1 + \omega_-^2)] + (\Omega^2 + \omega_0^2 - \Omega_0^2) [4\Omega \omega_- + \omega_A (1 + \omega_-^2)] \} Y^2 \\ & \left. + \frac{1}{8} \omega_A \{ [\Omega_0^2 - (\Omega - \omega_0)^2] (3A_1 X^3 - A_3 XY^2) + [\Omega_0^2 - (\Omega + \omega_0)^2] (A_2 X^2 Y - 3A_4 Y^3) \} \right], \quad (2.14c) \end{aligned}$$

$$\begin{aligned} \alpha_3 = \frac{XY}{\gamma^2 \omega_A^2} & \left[\frac{\gamma}{4} \{ [2\Omega \omega_+ (\Omega_0^2 + \omega_0^2 - \Omega^2) - 2\Omega \omega_0 \omega_A (1 + \omega_+^2)] X^2 \right. \\ & + [\Omega (\omega_+ + \omega_-) (\omega_0^2 + \Omega_0^2 - \Omega^2) - 2\Omega \omega_0 \omega_A (1 + \omega_+ \omega_-)] XY \\ & + [2\omega_- \Omega (\omega_0^2 + \Omega_0^2 - \Omega^2) - 2\Omega \omega_0 \omega_A^2 (1 + \omega_-^2)] Y^2 \} \\ & \left. + \frac{1}{8} \omega_A \{ [\Omega_0^2 - (\Omega - \omega_0)^2] (A_1 X^3 + A_3 XY^2) + [\Omega_0^2 - (\Omega + \omega_0)^2] (A_2 X^2 Y + A_4 Y^3) \} \right]. \quad (2.14d) \end{aligned}$$

In these equations, we have introduced the following quantities:

$$A_1 = \omega_+^2(3\omega_A - 2\omega_E) - \omega_+^2(\Omega + \omega_0) - 3\omega_+(\omega_0 + \Omega) + (2\omega_E + \omega_A), \quad (2.15a)$$

$$A_2 = 2\omega_+^2\omega_-(\omega_0 - \Omega) + 4\omega_+\omega_-(\omega_E - \omega_A) - 2\omega_A\omega_+^2 - 4\omega_+(\Omega - \omega_0) - 2\omega_-(\Omega - \omega_0) - 2(2\omega_E + \omega_A), \quad (2.15b)$$

$$A_3 = 2\omega_A\omega_-^2 - 2\omega_+\omega_-^2(\omega_0 + \Omega) - 4\omega_+\omega_-(\omega_E - \omega_A) - 4\omega_-(\Omega + \omega_0) - 2\omega_+(\omega_0 + \Omega) + 2(2\omega_E + \omega_A), \quad (2.15c)$$

and

$$A_4 = \omega_-^3(\omega_0 - \Omega) - \omega_-^2(3\omega_A - 2\omega_E) - 3\omega_-(\Omega - \omega_0) - (2\omega_E + \omega_A). \quad (2.15d)$$

While the above expressions give a complete description of the nonlinear response of the total moment to the applied field, quite clearly they are sufficiently cumbersome that they offer little insight into the nature of the response.

A much simpler picture emerges if we take the limit as the externally applied Zeeman field vanishes, $H_0 \rightarrow 0$. Then $X = Y = \gamma\omega_A / (\Omega_0^2 - \Omega^2)$, $\omega_- = -\omega_+ = \Omega / \omega_A$, while we also have $A_1 = -A_4$, $A_3 = -A_2$. Then one finds that

$$\lim_{H_0 \rightarrow 0} \alpha_1 = \lim_{H_0 \rightarrow 0} \alpha_4 = 0, \quad (2.16a)$$

while

$$\lim_{H_0 \rightarrow 0} \alpha_0 = \frac{\gamma^3 \omega_A \Omega^2 (\Omega^2 + \omega_A^2)}{(\Omega_0^2 - \Omega^2)^4}, \quad (2.16b)$$

and

$$\lim_{H_0 \rightarrow 0} \alpha_2 = -\frac{\gamma^3 \omega_A \Omega^2 (4\Omega_0^2 - 3\Omega^2 - 7\omega_A^2)}{(\Omega_0^2 - \Omega^2)^4}. \quad (2.16c)$$

In zero external Zeeman field, the relationship between the nonlinear component of the induced magnetization and the applied fields becomes

$$t_x^{(1)} = \alpha_0 h_x^3 + \alpha_2 h_x h_y^2 \quad (2.17a)$$

and

$$t_y^{(1)} = \alpha_0 h_y^3 + \alpha_2 h_y h_x^2. \quad (2.17b)$$

It is also possible to obtain simple expressions in finite field, where the frequency Ω is very close to $\Omega_0 \pm \omega_0$, the finite field antiferromagnetic resonance frequency. If we define $\Delta\Omega_{\pm}^2 = \Omega_0^2 - (\Omega \pm \omega_0)^2$, and assume $\Delta\Omega_{\pm}$ is very small, upon extracting the dominant terms in the response, we find

$$\alpha_0^{\pm} = \pm \alpha_3^{\pm} \cong \frac{\gamma^3 \omega^3 \Omega^2}{(\Delta\Omega_{\pm}^2)^4} \quad (2.18a)$$

and

$$\alpha_2^{\pm} = \pm \alpha_1^{\pm} \cong \frac{3\gamma^3 \omega_A^3 \Omega_0^2}{(\Delta\Omega_{\pm}^2)^4}. \quad (2.18b)$$

This completes our discussion of the nonlinear response of the uniaxial antiferromagnet, in the presence of an intense field. In Sec. III, we discuss the implications of the results obtained here.

III. POWER-DEPENDENT TRANSMISSION OF ELECTROMAGNETIC RADIATION THROUGH ANTIFERROMAGNETIC FILMS

In this section, we employ the results of Sec. II to explore the dependence on incident power of the transmission of electromagnetic radiation through thin antiferromagnetic films. We shall be led to the instabilities mentioned in Sec. I.

We consider a rutile structure antiferromagnet, with c axis oriented normal to the film. The source of the nonlinearity is coupling between the magnetic field in the infrared wave, and the spins. The results of the preceding section show that for frequencies near resonance, the nonlinear response coefficients may become substantial, so we focus on this regime.

We first recall the discussion of the transmissivity of such a thin film in the linear theory. We confine our attention to radiation normally incident on the film, so both its electric and magnetic field lie in the xy plane, perpendicular to the c axis. The dielectric tensor is diagonal in this case, and the electric field senses its value ϵ_{\perp} appropriate to the basal plane.

If a Zeeman field is applied parallel to the z axis, then the magnetic susceptibility tensor has nonzero diagonal elements $\chi_{xx}(\Omega) = \chi_{yy}(\Omega)$, and the off-diagonal elements are nonzero also, with $\chi_{xy}(\Omega) = -\chi_{yx}(\Omega)$, in a manner familiar from the theory of gyrotropic media. Explicit expressions for the nonzero elements of the magnetic-susceptibility tensor are given in Ref. 4, and also in Eqs. (2.9a) and (2.9b) of the present paper.

If plane-polarized radiation is incident on the film, then as the radiation propagates through the film, its plane of polarization rotates. This is the Faraday rotation; the transmitted radiation is also plane polarized, but the plane of polarization suffers a rotation through the angle $\Delta\theta$, controlled by the magnitude of the off-diagonal elements of the susceptibility tensor.

In zero applied external field, both $\chi_{xy}(\Omega)$ and $\chi_{yx}(\Omega)$ vanish identically. There is no Faraday rotation, so the problem of calculating the transmissivity is quite elementary. In the absence of an applied Zeeman field, it remains true also in the presence of the nonlinear response that a plane polarized field incident on the slab remains plane polarized always. We confine our attention to this simple case. The extension of the discussion to the case where the Zeeman field H_0 is nonzero would be most interesting to explore, but this is a nontrivial extension of the calculation presented below. It no longer remains

true that in the film the electromagnetic wave remains linearly polarized everywhere, with the plane of polarization rotating continuously as one moves through the film. At a general point in the film, the wave will be elliptically polarized, and its description requires solution of a non-trivial set of coupled, nonlinear differential equations for the two field components h_x and h_y .

The results of Sec. II may be summarized as follows. If, with Zeeman field set of zero, the film is excited by the time-varying magnetic field $h_y(t) = h_y \cos(\Omega t + \phi)$, then the magnetic moment m_y induced in the spin system may be written, with higher harmonics ignored,

$$m_y(t) = \chi_0(\Omega) [1 + \lambda(\Omega) h_y^2] h_y, \quad (3.1)$$

where the linear susceptibility is

$$\chi_0(\Omega) = \frac{2\gamma H_A m_s}{(\Omega_0^2 - \Omega^2)} \quad (3.2a)$$

and the nonlinear component of the response is controlled by

$$\lambda(\Omega) = \frac{\gamma^2}{2} \frac{\Omega^2(\Omega^2 + \omega_A^2)}{(\Omega_0^2 - \Omega^2)^3}. \quad (3.2b)$$

If we use complex notation, with $h_y(t) = h_y \exp(i\Omega t + \phi)$, then Eq. (3.1) is replaced by

$$m_y(t) = \chi_0(\Omega) [1 + \lambda(\Omega) |h_y|^2] h_y(t), \quad (3.3)$$

where now and in what follows, the physical response of the system at frequency Ω is given by $\text{Re}[m_y(t)]$.

Wave propagation in the medium is discussed by allowing h_y to vary with the coordinate z normal to the interface, with the relation between $h_y(z, t) = h_y(z) \exp(i\Omega t + \phi)$, and $m_y(z, t)$ controlled at each point by Eq. (3.2b). Within the film, for the simple case considered, Maxwell's equations give

$$\frac{\partial^2 h_y}{\partial z^2} + \frac{\Omega^2}{c^2} \epsilon_1 b_y = 0, \quad (3.4)$$

with ϵ_1 the dielectric constant in the basal plane, and we then have

$$\frac{\partial^2 h_y}{\partial z^2} + \frac{\Omega^2}{c^2} \epsilon_1 \mu_1(\Omega) h_y + 4\pi \frac{\Omega^2}{c^2} \epsilon_1 \chi_0(\Omega) \lambda(\Omega) |h_y|^2 h_y = 0. \quad (3.5)$$

We shall study the transmission of radiation through a film described by Eq. (3.5); we suppose the strength of the incident field which illuminates the film is h_0 . We then seek a solution of Eq. (3.5) with

$$h_y(z) = H_0 h(z) \exp[i\phi(z)], \quad (3.6)$$

with $h(z)$ and $\phi(z)$ real. We define $k = \Omega/c$, $n^2(\Omega) = \epsilon_1 \mu_1(\Omega)$, and we let $\gamma(\Omega) = 4\pi n(\Omega) \chi_0(\Omega) \lambda(\Omega) h_0^2$. In this paper, we shall confine attention to the spectral regime where both $n^2(\Omega)$ and $\gamma(\Omega)$ are positive. This is the case for frequencies below Ω_0 . In Ref. 6, an extension to the case $\gamma(\Omega) < 0$ is given. Equation (3.5) then breaks into two statements, once the real and imaginary parts are set equal to zero. These read as

$$\frac{d^2 h}{dz^2} - h \left[\frac{d\phi}{dz} \right]^2 + k^2 n^2(\Omega) h + k^2 \gamma(\Omega) h^3 = 0, \quad (3.7a)$$

and

$$2 \frac{dh}{dz} \frac{d\phi}{dz} + h \frac{d^2 \phi}{dz^2} = 0. \quad (3.7b)$$

Note that Eq. (3.7b) implies that the quantity

$$\xi = h^2 \frac{d\phi}{dz} \quad (3.8)$$

is independent of z . In the film, one may show that the time-averaged rate of energy flow along the z direction, $\langle S \rangle$, is proportional to ξ . In fact, one has $\langle S \rangle = (c^2 H_0^2 / 8\pi\omega\epsilon_1) (h^2 d\phi/dz)$ or $\langle S \rangle = (c^2 H_0^2 / 8\pi\omega\epsilon_1) \xi$. Since energy is conserved, in the appropriate units, the parameter ξ is the transmissivity of the film.

By combining Eq. (3.8) with Eq. (3.7a), one may obtain a second relation:

$$\left[\frac{dh}{dz} \right]^2 + \frac{\xi^2}{h^2} + k^2 n^2 h^2 + \frac{1}{2} k^2 \gamma h^4 = \tau, \quad (3.9)$$

where τ is a second constant of integration. If $h(z_0)$ is the value of the field at an arbitrary reference point z_0 , then we have

$$\int_{h(z_0)}^{h(z)} \frac{hdh}{(\tau h^2 - \xi^2 - k^2 n^2 k^4 - \frac{1}{2} k^2 \gamma h^6)^{1/2}} = \pm (z - z_0). \quad (3.10)$$

If $h(z)$ is found from Eq. (3.10), we then have information to find $\phi(z)$ everywhere:

$$\phi(z) = \phi(z_0) + \xi \int_{z_0}^z \frac{dz'}{h^2(z')}. \quad (3.11)$$

The relations above show that the solution within the medium is characterized by four constants, ξ , τ , $h(z_0)$, and $\phi(z_0)$. These are to be determined by submitting the solution of the nonlinear equation to the appropriate boundary conditions at each film surface. Note that in the description of the transmissivity of thin films provided by linear theory, four parameters are also required. In the linear theory, the solution within the film is described as a superposition of two plane waves, one propagating from right to left, and one from left to right. Each has wave vector of magnitude $\kappa = (\Omega/c)n(\Omega)$, where κ is pure imaginary in spectral regions where the product $\epsilon_1 \mu_1(\Omega)$ is negative. Each plane wave is described by a complex amplitude, which requires two parameters to specify.

The analysis of the response of the film to an incident electromagnetic wave requires the four parameters to be determined from the boundary conditions at each interface. A recent paper⁶ describes how this may be reduced to a search for a single parameter, ξ , which is bounded between zero and a finite value $\xi_M = \epsilon_1$ controlled by the requirement that the transmissivity of the film be less than unity. We summarize the approach used in Ref. 6, which applies as well to the present problem.

The film is illuminated from the left by an incident electromagnetic wave whose amplitude is H_0 ; to the left of

the film ($z < 0$) the magnetic field in the wave is thus $h(z) = H_0(e^{ikz} + R e^{-ikz})$, with R the reflection coefficient. To the right of the film, $z > d$, the transmitted wave is $h(z) = H_0 T \exp(ikz)$. The electromagnetic boundary conditions at the two surfaces then provide us with the following statements:

$$1 + R = h(0) \exp[i\phi(0)] , \quad (3.12a)$$

$$1 - R = \frac{1}{ik\epsilon_1} \left[\left. \frac{dh}{dz} \right|_0 + ih(0) \left. \frac{d\phi}{dz} \right|_0 \right] e^{i\phi(0)} , \quad (3.12b)$$

$$T e^{ikd} = h(d) e^{i\phi(d)} , \quad (3.12c)$$

and

$$T e^{ikd} = \frac{1}{ik\epsilon_1} \left[\left. \frac{dh}{dz} \right|_d + ih(d) \left. \frac{d\phi}{dz} \right|_d \right] e^{i\phi(d)} . \quad (3.12d)$$

We may obtain useful constraints from Eqs. (3.12a)–(3.12d). Quite clearly, Eq. (3.12c) gives us the relation

$$|T|^2 = h^2(d) \quad (3.13a)$$

while Eq. (3.12c) may be combined with Eq. (3.12d) to yield

$$\left. \frac{dh}{dz} \right|_{z=d} = 0 , \quad (3.13b)$$

and also

$$\xi = k\epsilon_1 h^2(d) . \quad (3.13c)$$

Equations (3.12a) and (3.12b) may be used to provide the relations

$$\int_{h^2(d)}^{h^2(z)} \frac{hdh}{\{[h^2(d) - h^2](h^2 - h_2^2)(h^2 + h_3^2)\}^{1/2}} = \pm k \left[\frac{\gamma}{2} \right]^{1/2} (d - z) , \quad (3.16)$$

where both h_2^2 and h_3^2 are positive numbers. We omit explicit expressions for these two quantities, in the interest of brevity. It is possible to express the integral in Eq. (3.16) in terms of Jacobi elliptic functions. One finds

$$\frac{1}{[h^2(d) + h_3^2]^{1/2}} \text{cn}^{-1} \left[\left. \frac{h^2(z) - h_2^2}{h^2(d) - h_2^2} \right|^{1/2} \left. \frac{h^2(d) - h_2^2}{h^2(d) + h_3^2} \right] = \pm k \left[\frac{\gamma}{2} \right]^{1/2} (d - z) , \quad (3.17a)$$

if $h^2(d) > h_2^2$, and

$$\frac{1}{(h_2^2 + h_3^2)^{1/2}} \text{dn}^{-1} \left[\left. \frac{h^2(d) + h_3^2}{h^2(z) + h_3^2} \right|^{1/2} \left. \frac{h_2^2 - h^2(d)}{h_3^2 + h^2(d)} \right] = \pm k \left[\frac{\gamma}{2} \right]^{1/2} (d - z) \quad (3.17b)$$

if $h^2(d) < h_2^2$. These relations may be inverted to give

$$h^2(z) = \begin{cases} h_2^2 + [h^2(d) - h_2^2] \text{cn}^2 \left[\left. \frac{\gamma(h^2(d) + h_3^2)}{2} \right]^{1/2} k(d - z) \left. \frac{h^2(d) - h_2^2}{h^2(d) + h_3^2} \right] & (h(d) > h_2) \\ -h_3^2 + [h^2(d) + h_3^2] \text{dn}^{-2} \left[\left. \frac{\gamma(h_2^2 + h_3^2)}{2} \right]^{1/2} k(d - z) \left. \frac{h_2^2 - h^2(d)}{h^2(d) + h_3^2} \right] & (h(d) < h_2) \end{cases} \quad (3.18)$$

$$\left. \frac{dh}{dz} \right|_0 = 2k\epsilon_1 \sin[\phi(0)] \quad (3.13d)$$

and

$$\xi + k\epsilon_1 h^2(0) = 2k\epsilon_1 h(0) \cos[\phi(0)] . \quad (3.13e)$$

Finally, Eqs. (3.13d) and (3.13e) may be squared and added to give

$$\left. \frac{dh}{dz} \right|_0^2 + \left[\frac{\xi}{h(0)} + k\epsilon_1 h(0) \right]^2 = 4k^2 \epsilon_1^2 . \quad (3.14)$$

Also note that Eq. (3.9), along with Eq. (3.13b), provides the statement

$$\tau = \frac{1}{2} k^2 \gamma h(d)^4 + k^2 n^2 h^2(d) + \frac{\xi^2}{h^2(d)} . \quad (3.15)$$

We may now proceed as follows. We choose a value of the parameter ξ in the acceptable range $0 \leq \xi \leq \xi_M$. Then from Eq. (3.13c) we may find $h(d)$, and the parameter τ from Eq. (3.15). The reference point z_0 is chosen to be $z = d$, and we now have enough information to find $h(z)$ everywhere from Eq. (3.10), and $\phi(z)$ from Eq. (3.11), if desired. We can inquire if the value of ξ chosen corresponds to a solution to the problem by checking if Eq. (3.14) is satisfied at $z = 0$; notice we need not calculate $(dh/dz)_0^2$ numerically, but we may make use of Eq. (3.9) applied at $z = 0$. Shortly we shall see that the ambiguity in sign on the right-hand side is of no concern. With this procedure, we may scan the whole range $0 \leq \xi \leq \xi_M$, to find the one or more values of ξ which yield a solution.

The integral in Eq. (3.10) may be arranged to read, after using relations given above,

It should be noted that the function $\text{cn}(y | a)$ is an even function of the argument y . As a consequence, the ambiguity in sign which occurs in earlier equations [Eq. (3.10), for example], leads to no difficulties. It is possible to take the limit as the nonlinear coefficient γ vanishes in the above relations, to recover the results of the conventional theory of the linear dielectric film. This is done explicitly in Ref. 6, where the case where $\gamma < 0$ is examined also, as remarked earlier. We discuss calculations of the transmissivity of FeF_2 films in Sec. IV.

IV. RESULTS AND DISCUSSION

In this section, we apply the results of Secs. II and III to the calculation of the power dependence of the transmissivity of thin antiferromagnetic films. We consider a film of FeF_2 , with c axis normal to the surface, and zero applied magnetic field H_0 . The radiation is assumed plane polarized, and strikes the film at normal incidence. For this material, the anisotropy field $H_A = 200$ kOe, the exchange field $H_E = 540$ kOe, and the magnetization m_s of one sublattice is 0.56 kOe. These parameters place the zero-field antiferromagnetic resonance at 47 cm^{-1} . The thickness of the film has been taken to be $105 \mu\text{m}$, equal to half the (vacuum) wavelength of a photon of frequency Ω_0 .

In Fig. 1, we show the transmissivity as a function of frequency Ω for frequencies near to and below the antiferromagnetic resonance frequency Ω_0 . These calculations assume the nonlinear coupling constant $\lambda(\Omega) = 0$. The transmissivity is seen to be an oscillatory function of frequency. This behavior has its origin in the frequency dependence of the linear susceptibility $\mu(\Omega)$. As one approaches the resonance frequency Ω_0 from below, $\mu(\Omega)$ increases and one finds a transmissivity of unity whenever a half-integral number of magnetic polariton wavelengths fit into the film.

In Fig. 2, we show the frequency variation of the transmissivity, within the framework of a calculation that

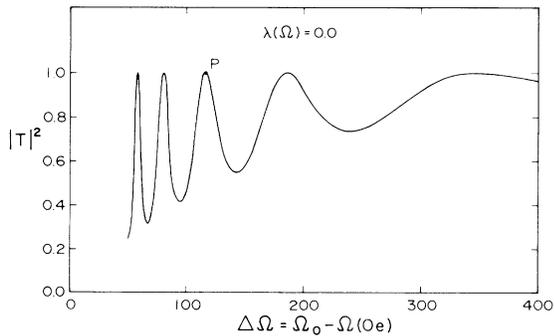


FIG. 1. The frequency variation of the transmissivity of a thin FeF_2 film as a function of frequency, for frequencies below the zero-field antiferromagnetic resonance frequency Ω_0 . The calculations assume zero external Zeeman field, and are calculated using linear-response theory [$\lambda(\Omega) = 0$]. The parameters are $H_A = 200$ kOe, $H_E = 540$ kOe, $m_s = 0.56$ kOe, and $\epsilon_1 = 4.0$. The film thickness has been chosen to be $105 \mu\text{m}$, equal to half the vacuum wavelength of a photon with frequency Ω_0 .

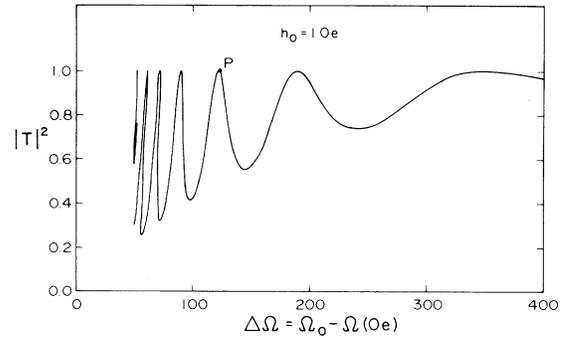


FIG. 2. The same as Fig. 1, but now the nonlinear response of the spin system has been incorporated. The strength of the magnetic field in the external wave has been chosen to be 1.0 G.

incorporates the nonlinear response of the spin system. Here the strength of the magnetic field in the incident electromagnetic wave is taken to be 1 G. The reader should note that the free-electron laser at Santa Barbara can generate pulses within which the magnetic field reaches 20 G.⁹ To the eye, the calculations in Fig. 2 look similar to those in Fig. 1. However, one sees power-dependent shifts of the various structures. For instance, at point P in Fig. 1 we see a resonance at $\Delta\Omega = 116$ G, and in Fig. 2 this feature has shifted down in frequency to 123 G. There is a region, with $\Delta\Omega$ below 70 G, where the transmissivity is a multivalued function of power. Here the film will exhibit hysteretic bistability.

In Fig. 3, we show the transmissivity for an incident wave within which the magnetic field is 10 G. The resonance at point P is now driven down to $\Delta\Omega = 220$ G. More importantly, we see the transmissivity becomes multivalued in nature in this spectral region, so we find bistability 200 G below resonance, at this power level. As remarked earlier, samples of FeF_2 have been fabricated with linewidths of less than 20 G,⁵ so point P in Fig. 3 lies 10 linewidths from resonance for such a sample. This suggests that it should be possible to carry out studies of bistability sufficiently far from resonance that absorption by the spin system is not a major problem.

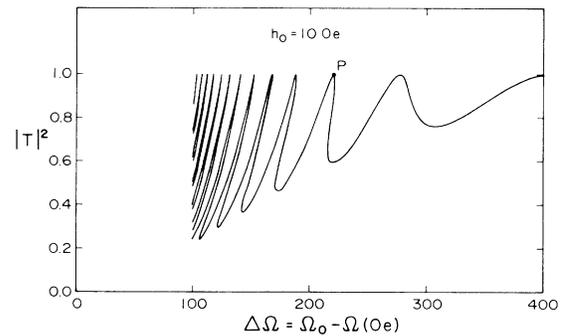


FIG. 3. The same as Fig. 2, but now the strength of the magnetic field in the incident wave is 10.0 G.

Of course, in any experiment at elevated power levels, sample heating simply from dielectric loss can be problematical. Sample heating can mimic the intrinsic effects studied here. If the sample is heated, the resonance frequency Ω_0 will decrease by an amount $\Delta\Omega$ proportional to $\langle h^2 \rangle$, the average of the square of the rf field in the wave over the film, if the film is heated uniformly. Such a shift in the resonance gives an effective nonlinear coupling constant $\lambda(\Omega)\langle h^2 \rangle = 2\Omega_0\Delta\Omega/(\Omega_0^2 - \Omega^2)$ which is positive below resonance, very much as the result derived in Sec. II. Note that the frequency variation of the contribution to $\lambda(\Omega)$ from heating differs from the expression in Eq. (3.2b) so at least in principle, detailed study of the dependence of the bistability on frequency and power can discriminate between the mechanisms described here. The use of pulses sufficiently short will also discriminate against heating effects.

Also, at elevated power levels, the analog of Suhl instabilities may be encountered in antiferromagnets, as well as ferromagnets.³ Rather little attention has been directed toward this question in the literature.¹⁰ Suhl discussed two distinctly different instabilities. The instability which sets in at lowest power is one in which a wave of frequency Ω_0 decays to a spin-wave pair, each member of which has frequency $\Omega_0/2$. For the example considered here, the antiferromagnetic resonance frequency is the lowest-

frequency spin-wave mode, so Suhl's subsidiary resonance absorption is absent. For experiments in a magnetic field, the same statement applies if one explores the response near the lower of the two resonance modes, at the frequency $\Omega_0 - H_0$ in our notation.

The second instability discussed by Suhl requires the existence of short-wavelength spin waves degenerate with the main resonance. There are no such waves either, at the level of description offered here. However, a full theory which incorporates dipolar fields generated by spin motions may allow such degenerate waves, in particular geometries. A detailed study is required to explore this point.

The study presented here suggests that antiferromagnetic films should exhibit bistability in their far infrared response, for power levels presently accessible in the laboratory. It will be intriguing to see experimental studies of this phenomenon.

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¹See the discussion which begins on page 44 of A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, London, 1961).

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⁹Allan R. King (private communication).

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