

A SUMMARY OF ORDERED STATE RESONANCE¹

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I. Abstract

A review of the history of ferromagnetic resonance is presented with the shortcomings of previous theories illustrated. Recent developments in ferromagnetic resonance are presented and the generalization to multisublattice systems is reviewed and discussed.

II. Ordered State Resonance

A. Prologue

The original motivation for the work outlined in this paper started with an attempt to find a method for measuring the anisotropy energy as well as the interplane antiferromagnetic (AF) exchange in the common layered structures of *n*-ethylammonium CuCl_4 (*n*EA) and *n*-ethanediammonium CuCl_4

(*n*DA). Most of these order ferromagnetically within the plane of CuCl_4 's but the weak ferromagnetic (F) exchange between planes yields a 3D AF compound below the ordering temperature. In F resonance the strong exchange between ions is not explicit in the free energy or Hamiltonian since in the ordered F state all spins are aligned. That is, the exchange energy has already been *used* to provide the magnetization vector *M*. AF resonance, on the other hand, is typically treated as a two sublattice system where all the spins pointing in one direction are considered to be one sublattice and all those in the other direction are from the other sublattice. In this case the strong exchange in a standard 3D AF is still the exchange between neighboring spins and, therefore, becomes the exchange between sublattices and as such becomes part of the explicit free energy of the system. The fact that all the spins on one sublattice

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tice point in one direction and form the sublattice magnetization vector M_1 is implicit in the ordering but not because there is an exchange term between spins on one sublattice.

The situation is quite different for the layered structures mentioned above. They are unique in that each alternate ferromagnetic layer forms one of the sublattices, the other layers form the other sublattice. This is illustrated in Figure 1. Now the weak interlayer AF exchange becomes the exchange which shows up in the free energy and, therefore, becomes a measurable parameter along with the anisotropy energy or whatever else might be included in the free energy model.

The standard theories for ferro- and antiferromagnetic resonance soon proved inadequate to the task since some of the layered systems showed resonances that could only be described by systems with more than two sublattices. Furthermore, no single theory seemed to adequately describe the angle dependences that experiment revealed.

In what we present here, we will use the term *ordered state resonance (OSR)* to describe magnetic resonance in systems comprised of one or more distinct sublattices, where a sublattice is considered to be a group of spins moving coherently. F and AF systems are plentiful, but little work has been done on magnetic systems of more than two sublattices (1,2,3). In studying such systems, the need for a simple theory for magnetic resonance in systems comprised of multiple sublattices became apparent and we have successfully extended the theory of F and AF resonance to cover all such systems, in a straightforward manner (4). Indeed, we have been able to show that OSR is completely described by a simple equation involving a "Hessian matrix" of the system under consideration.

B. Background

F resonance, which is OSR for a one sublattice system, is well established as a method for the determination of anisotropy constants in a parametric expansion of free energy (5). Since the first article by Kittel (6) in 1947, several authors (7,8) have extended the theory for specific solutions while weakening their conditions for validity. In 1955, Smit and Beljers (9) published a simple equation for the resonance frequency, ω , in terms of second derivatives of magnetization polar θ and ϕ which appears

as

$$(\omega/\gamma)^2 = (F_{\theta\theta}F_{\phi\phi} - F_{\theta\phi}^2)/M^2 \sin^2 \phi. \quad (1)$$

In this equation $F_{\theta\theta} = \partial^2 F/\partial\theta^2$, for example, where the polar angles are shown in Fig. 2. The same expression has been published independently by Suhl (10) by Tannewald and Lax (11) and also by Gilbert (12). This form of the resonance equation has since become the standard in analyzing ferromagnetic resonance data (13,14,15). Recently it was shown by Basalgia, *et al.* (16) that this equation, while correct for most angles, cannot be used to find the correct high field limits when $\theta \neq \pi/2$ through a simple substitution of field angles for magnetization angles. Instead, it was shown in that work that a third form of the ferromagnetic resonance equation, which combines the advantage of Smit and Beljers method with that of orthogonal coordinate systems, allows direct substitution of field angles for magnetization angles in order to obtain high-field limits. It is this latter theory of ferromagnetic resonance which we have generalized to cover all magnetic systems describable by the sublattice concept. We outline this theory below.

C. Ordered State Resonance

The generalized torque equation which describes OSR is given by

$$d\vec{M}^\sigma/dt = \gamma \vec{M}^\sigma \times \vec{H}^\sigma \quad (2)$$

where σ denotes the particular sublattice under consideration and \vec{H}^σ is the effective field acting upon sublattice σ , with \vec{M}^σ and \vec{H}^σ referenced to the local system of coordinates defined by $\vec{M}^\sigma = (0,0,M^\sigma)$ in equilibrium, rather than by the lab coordinate system. Note, the unusual use of the greek letters for sublattices is to emphasize that these are sublattices numbers even though the summations for an ℓ -sublattice system is now $\sigma = 1$ to ℓ . The numbers 1, 2, and 3 refer to the Cartesian components in the frame of the magnetization vectors with M_3^σ in the M^σ direction. We expand the 1 and 2 components in a Taylor series to first order about $M_1^\sigma = 0$, and $M_2^\sigma = 0$ and $M_3^\sigma = M^\sigma$ to obtain

$$\dot{M}_1^\sigma/\gamma \sim M^\sigma \{M_2^\sigma (H_3^\sigma/M^\sigma) + \sum_{\rho=1}^{\ell} [M_1^\rho A_{21}^{\sigma\rho} + M_2^\rho A_{22}^{\sigma\rho}]\}$$

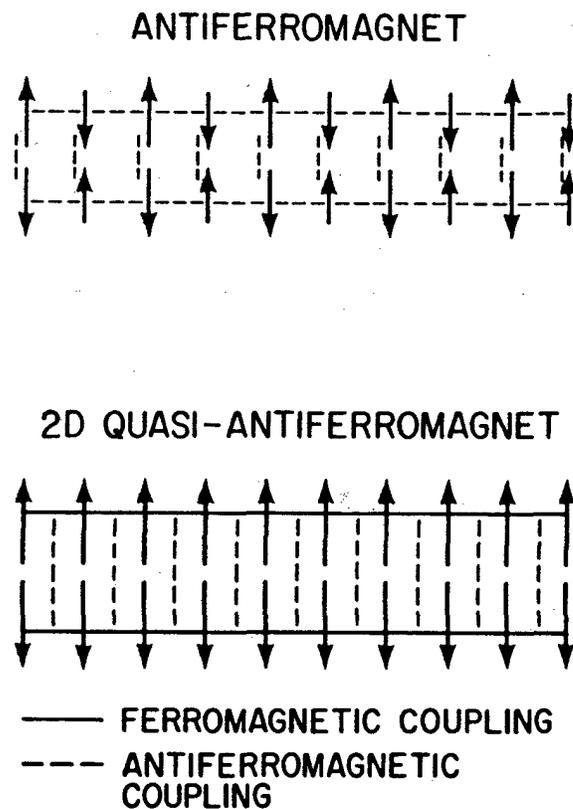


Figure 1: For traditional antiferromagnets, antiferromagnetic exchange between nearest neighbors leads to an overall antiferromagnetic system where as for the 2-dimensional layered compounds, we find an antiferromagnetic exchange between planes but strong ferromagnetic interactions within the planes.

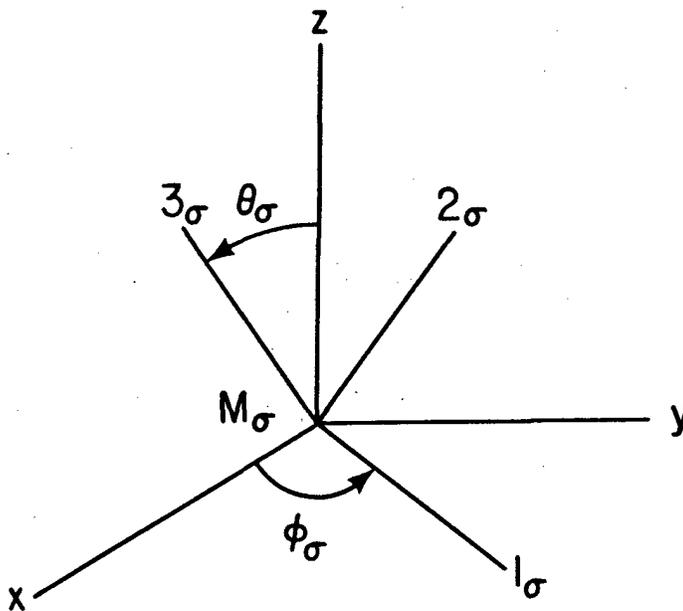


Figure 2: Relationship between the Euler angles ($\theta_\sigma, \phi_\sigma$) and the lab coordinate system.

$$\dot{M}_2^\sigma/\gamma \sim -M^\sigma \{M_1^\sigma (H_3^\sigma/M^\sigma) + \sum_{\rho=1}^{\ell} [M_1^\rho A_{11}^{\sigma\rho} + M_2^\rho A_{12}^{\sigma\rho}]\} \quad (3)$$

where $A_{ij}^{\sigma\rho} = -\partial H_i^\sigma/\partial M_j^\rho$. One now seeks modes in which M_1^σ and M_2^σ vary harmonically in time, that is

$$M_1^\sigma(t) = M_1^\sigma e^{i\omega t} \text{ and } M_2^\sigma(t) = M_2^\sigma e^{i\omega t}. \quad (4)$$

The resultant system of equations for the σ th sublattices is

$$0 = M_1^\sigma (A_{11}^{\sigma\sigma} + H_3^\sigma/M^\sigma) + M_2^\sigma (A_{12}^{\sigma\sigma} + i\omega/\gamma M^\sigma) + \sum_{\rho \neq \sigma}^{\ell} [M_1^\rho A_{11}^{\sigma\rho} + M_2^\rho A_{12}^{\sigma\rho}]$$

and

$$0 = M_1^\sigma (A_{21}^{\sigma\sigma} - i\omega/\gamma M^\sigma) + M_2^\sigma (A_{22}^{\sigma\sigma} + H_3^\sigma/M^\sigma) + \sum_{\rho \neq \sigma}^{\ell} [M_1^\rho A_{21}^{\sigma\rho} + M_2^\rho A_{22}^{\sigma\rho}]. \quad (5)$$

From the form of Eq. (5) the condition for harmonic time dependence will be given by the determinant of the difference of two matrices,

$$\begin{bmatrix} A_{11}^{11} & A_{12}^{11} & A_{11}^{12} & A_{12}^{12} & \dots & A_{11}^{1\ell} & A_{12}^{1\ell} \\ A_{21}^{11} & A_{22}^{11} & A_{21}^{12} & A_{22}^{12} & \dots & A_{21}^{1\ell} & A_{22}^{1\ell} \\ A_{11}^{21} & A_{12}^{21} & A_{11}^{22} & A_{12}^{22} & \dots & A_{11}^{2\ell} & A_{12}^{2\ell} \\ A_{21}^{21} & A_{22}^{21} & A_{21}^{22} & A_{22}^{22} & \dots & A_{21}^{2\ell} & A_{22}^{2\ell} \end{bmatrix} \quad (6)$$

$$\begin{bmatrix} -H_3^1/M^1 & -i\omega/\gamma M^1 & & & & & 0 \\ i\omega/\gamma M^1 & -H_3^1/M^1 & & & & & \\ & & \dots & & & & \\ & & & -H_3^\ell/M^\ell & -i\omega/\gamma M^\ell & & \\ 0 & & & i\omega/\gamma M^\ell & -H_3^\ell/M^\ell & & \end{bmatrix} = 0$$

To complete the identification of the first matrix as the hessian matrix of the system with respect to coordinates M_1^σ and M_2^σ , we use the definition of the effective fields from ref. 16 and 17:

$$H_i^{eff} = -\partial F/\partial M_i. \quad (7)$$

The multiple sublattice generalized of this effective field is given by

$$H_i^\sigma = -\partial F/\partial M_i^\sigma \quad (8)$$

where F describes the free energy expansion of the system. Thus, using the definition of $A_{ij}^{\sigma\rho}$ from Eq. (3), we have the first matrix to be the Hessian matrix, H (1,2, ..., ℓ). If we then denote the second matrix as $\Omega(1,2, \dots, \ell)$, the generalized ℓ -sublattice resonance condition is seen to be

$$\det[H(1,2, \dots, \ell) - \Omega(1,2, \dots, \ell)] = 0. \quad (9)$$

The equilibrium conditions are obtained directly from the exact expression for the torque acting upon sublattice σ . We require that in equilibrium, all external torques on that sublattice vanish. If we thus look at the exact expression for torque,

$$\tau_1^\sigma = \gamma(M_2^\sigma H_3^\sigma - M_3^\sigma H_2^\sigma)$$

$$\tau_2^\sigma = \gamma(M_3^\sigma H_1^\sigma - M_1^\sigma H_3^\sigma), \quad (10)$$

we find that $H_2^\sigma = 0$ and $H_1^\sigma = 0$ in equilibrium. Then, using the definition of effective fields Eq. (8), we find that the generalized equilibrium condition for sublattice σ is given by

$$\partial F/\partial M_1^\sigma = 0 \text{ and } \partial F/\partial M_2^\sigma = 0 \quad (11)$$

where M_i^σ refer to local coordinates rather than laboratory coordinates. At this point one must decide how to write the free energy with regard to local coordinates ($M_1^\sigma, M_2^\sigma, M_3^\sigma$) when the external field, H, requires that F be written in lab coordinate. This conversion can conveniently be accomplished through the use of Euler matrix, B, defined by

$$\begin{bmatrix} x^\sigma \\ y^\sigma \\ z^\sigma \end{bmatrix} = \begin{bmatrix} \cos\theta_\sigma \cos\phi_\sigma & -\sin\phi_\sigma & \sin\theta_\sigma \cos\phi_\sigma \\ \cos\theta_\sigma \sin\phi_\sigma & \cos\phi_\sigma & \sin\theta_\sigma \sin\phi_\sigma \\ -\sin\theta_\sigma & 0 & \cos\theta_\sigma \end{bmatrix} \begin{bmatrix} 1^\sigma \\ 2^\sigma \\ 3^\sigma \end{bmatrix} \quad (12)$$

so that, for example,

$$\partial M_x^\sigma / \partial M_1^\sigma = b_{xl}^\sigma = \cos\theta_\sigma \cos\phi_\sigma. \quad (13)$$

Now to illustrate the procedure for obtaining resonant fields, H , we give a simple example.

D. Example: 1 Sublattice FMR

An important and well studied model is that of a ferromagnet (1-sublattice) with uniaxial anisotropy. Note that with $\ell = 1$, Eq. (9) and following is identical to the Basalgia, et al. formalism. Begin by writing the free energy of the system referenced to LAB coordinates as

$$F = (1/2)K(M_z^1)^2 - \vec{H} \cdot \vec{M}^1, \quad (14)$$

where \vec{H} is the external field and K is the uniaxial anisotropy constant and \vec{M}^1 is the sublattice magnetization. It is convenient to restrict H to the x - z plane so that the free energy becomes

$$F = (1/2)K(M_z^1)^2 - H_x M_x^1 - H_z M_z^1. \quad (15)$$

It is then necessary to find and solve the equilibrium angles from Eq. (11),

$$\begin{aligned} \partial F / \partial M_1^1 &= K M_z^1 b_{z1}^1 - H_x b_{x1}^1 - H_z b_{z1}^1. \\ &\text{and} \end{aligned} \quad (16)$$

$$\partial F / \partial M_2^1 = K M_z^1 b_{z2}^1 - H_x b_{x2}^1 - H_z b_{z2}^1.$$

Now, using the condition that the equilibrium coordinates will be that for which $\vec{M}^\sigma = (0, 0, M^\sigma)$ in the local coordinates, obtain

$$\begin{aligned} \partial F / \partial M_1^1 |_{eq} &= K M_z^1 b_{z1}^1 b_{z3}^1 - H_x b_{x1}^1 - H_z b_{z1}^1. \\ \partial F / \partial M_2^1 |_{eq} &= K M_z^1 b_{z2}^1 b_{z3}^1 - H_x b_{x2}^1 - H_z b_{z2}^1. \end{aligned} \quad (17)$$

From the definition of the Euler matrix, Eq. 12, we obtain

$$\begin{aligned} \partial F / \partial M_1^1 |_{eq} &= -K M^1 \sin\theta_1 \cos\theta_1 - H_x \cos\theta_1 \cos\phi_1 + \\ &H_z \sin\theta_1 = 0 \\ \partial F / \partial M_2^1 |_{eq} &= H_x \sin\phi_1 = 0 \end{aligned} \quad (18)$$

These equations are exactly solvable. The second equation yields the solution $\phi_1 = 0$ so that the first of Eq. (18) becomes

$$-K M^1 \sin\theta_1 \cos\theta_1 - H_x \cos\theta_1 + H_z \sin\theta_1 = 0. \quad (19)$$

Eq. 19 can be solved exactly for the value of θ_1 , but its solution involves a fourth order polynomial. For more complicated free energy models, resort to iterative methods of solutions may be necessary.¹⁸

With the value of θ_1 , the next step is to obtain elements of the 1-sublattice Hessian matrix, $H(1)$,

$$H(1) = \begin{bmatrix} F_{M_1^1 M_1^1} & F_{M_1^1 M_2^1} \\ F_{M_2^1 M_1^1} & F_{M_2^1 M_2^1} \end{bmatrix} = \begin{bmatrix} K \sin^2 \theta_1 & 0 \\ 0 & 0 \end{bmatrix} \quad (20)$$

Then, the reduced Hessian matrix will be given by Eq. (6) with ω set to zero, that is

$$H_r(1) = \begin{bmatrix} -K \cos(2\theta_1) + H_x \sin\theta_1 / M^1 + H_z \cos\theta_1 / M^1 & 0 \\ 0 & -K \cos^2 \theta_1 + H_x \sin\theta_1 / M^1 + H_z \cos\theta_1 / M^1 \end{bmatrix} \quad (21)$$

Thus, the resonant frequency from Eq. (9), is given by

$$\begin{aligned} (\omega/\gamma)^2 &= (H_x \sin\theta_1 + H_z \cos\theta_1 - K M^1 \cos^2 \theta_1) \\ &(-K M^1 \cos(2\theta_1) + H_x \sin\theta_1 + H_z \cos\theta_1) \end{aligned} \quad (22)$$

where θ_1 is now the angle obtained by solving Eq. (19), the equilibrium conditions. Eq. (22), as mentioned earlier, is identical to that derived from the Basalgia, et al. formulas but differs from that obtained from the Smit and Belgers formalism. The latter appears as

$$\begin{aligned} (\omega/\gamma)^2 &= (H_x / \sin\theta_1)(H_x \sin\theta_1 + \\ &H_z \cos\theta_1 - K M^1 \cos(2\theta_1)) \end{aligned} \quad (23)$$

The difference between these two theories disappears when the equilibrium conditions are calculated. As an aside, one experiences great difficulties in inverting these resonance equations. However, in the high field limit, one may directly substitute field angles θ and ϕ for magnetization angles into the resonance equation obtained from Basalgia, et.

al. formalism.¹⁶ In this case, we obtain the simple expression for the resonance field,

$$H = KM^1(\cos(2\theta) + \cos^2\theta)/2 \pm [(KM^1)^2(\cos(2\theta) + \cos^2\theta)^2/4 + (\omega/\gamma)^2 - (KM^1)^2\cos^2\theta\cos(2\theta)]^{1/2} \quad (24)$$

where θ is now a field angle. For the parameter $KM = -1000$ and $\omega/\gamma = 3000$, we have sketched the behavior of the resonance field in Fig. 3 for the high field limit and also for the exact solution.

E. A 2-Sublattice Example

The model under consideration is that of an uniaxial system with an antiferromagnetic exchange between sublattices. An appropriate expression for such a system is

$$F = -(1/2)K[(M_z^1)^2 + (M_z^2)^2] - H_x[M_x^1 + M_x^2] - H_z[M_z^1 + M_z^2] + \epsilon[M_x^1M_x^2 + M_y^1M_y^2 + M_z^1M_z^2]. \quad (25)$$

According to Eq. (9), the resonance condition will be given by

$$\det[H(1, 2) - \Omega(1, 2)] = 0 \quad (26)$$

Evaluation of the Hessian matrix is straight forward and yields

$$H(1, 2) = \begin{bmatrix} -K\sin^2\theta_1 & 0 & \epsilon\cos(\theta_1 - \theta_2) & 0 \\ 0 & 0 & 0 & \epsilon \\ \epsilon\cos(\theta_1 - \theta_2) & 0 & -K\sin^2\theta_2 & 0 \\ 0 & \epsilon & 0 & 0 \end{bmatrix} \quad (27)$$

where, by symmetry, we have assumed the axial magnetization angles ϕ , ϕ_1 , and ϕ_2 are identical and thus, we are strictly working below the spin flop state.

The other matrix, the frequency matrix, is given by

$$\Omega(1, 2) = \begin{bmatrix} \beta_1 & -i\omega\alpha_1 & 0 & 0 \\ i\omega\alpha_1 & \beta_1 & 0 & 0 \\ 0 & 0 & \beta_2 & -i\omega\alpha_2 \\ 0 & 0 & i\omega\alpha_2 & \beta_2 \end{bmatrix} \quad (28)$$

where

$$\beta_\sigma = (1/M^\sigma)\partial F/\partial M_3^\sigma|_{eq}. \quad (29)$$

$$= -K\cos^2\theta_\sigma - H_x/M\sin\theta_\sigma - H_z/M\cos\theta_\sigma + \epsilon\cos(\theta_1 - \theta_2)$$

and $\alpha_\sigma = 1/\gamma M^\sigma$. The expansion of Eq. (26) yields a polynomial in terms of $(\omega/\gamma M)^2$,

$$(\omega/\gamma M)^4 - (\omega/\gamma M)^2(H_r(1) + H_r(2) + 2X(1, 2)) + H_r(1, 2) = 0 \quad (30)$$

where the "reduced" Hessian matrices are given by

$$H_r(\sigma) = \begin{vmatrix} -K\sin^2\theta_\sigma - \beta_\sigma & 0 \\ 0 & -\beta_\sigma \end{vmatrix} = \beta_\sigma(\beta_\sigma + K\sin^2\theta_\sigma) \quad (31)$$

and

$$H_r(1, 2) = \begin{vmatrix} -K\sin^2\theta_1 - \beta_1 & 0 & \epsilon\cos(\theta_1 - \theta_2) & 0 \\ 0 & -\beta_1 & 0 & \epsilon \\ \epsilon\cos(\theta_1 - \theta_2) & 0 & -K\sin^2\theta_2 - \beta_2 & 0 \\ 0 & \epsilon & 0 & -\beta_2 \end{vmatrix} \quad (32)$$

so that

$$H_r(1, 2) = (\epsilon^2 - \beta_1\beta_2)(\epsilon^2\cos^2(\theta_1 - \theta_2) - [K\sin^2\theta_2 + B_1][K\sin^2\theta_2 + B_2]) \quad (33)$$

and

$$X(1, 2) = \begin{vmatrix} \epsilon\cos(\theta_1 - \theta_2) & 0 \\ 0 & \epsilon \end{vmatrix} = \epsilon^2\cos(\theta_1 - \theta_2). \quad (34)$$

Thus, the resonance frequency will be given by

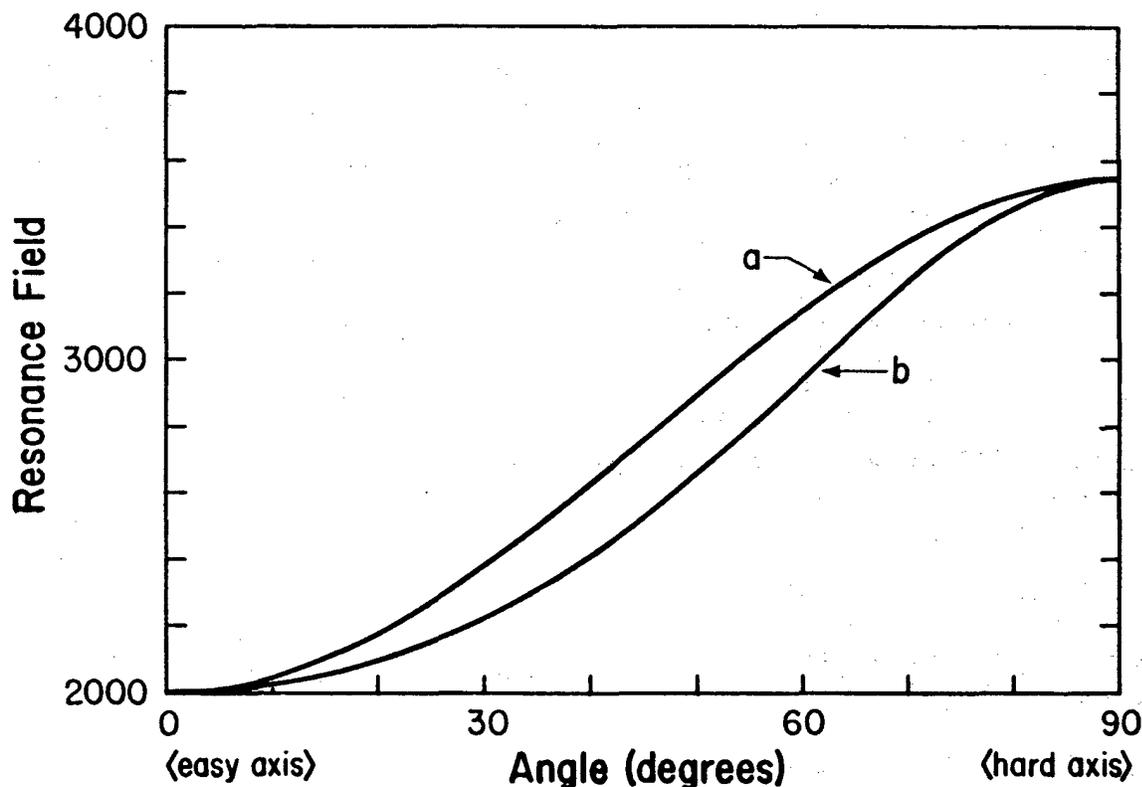


Figure 3: Curve a: Resonance field as a function of polar field angle with the assumption of a high field limit so that \vec{M}^1 and \vec{H} are parallel. Curve b: Resonance field as a function of polar field angle θ , including correction, due to \vec{M}^1 and \vec{H} not being parallel.

$$(\omega/\gamma M)^2 = (1/2)[H_r(1) + H_r(2)] + X(1, 2)$$

$$\pm [[(1/2)(H_r(1) + H_r(2)) + X(1, 2)]^2 - H_r(1, 2)]^{1/2} \quad (35)$$

One step remains to be done, however, namely the angles θ_1 and θ_2 must be obtained from the equilibrium conditions given by Eq. (11). In this case, the system to be solved is

$$KM\sin\theta_1\cos\theta_1 - H_x\cos\theta_1 + H_z\sin\theta_1 - \epsilon M\sin(\theta_1 - \theta_2) = 0$$

$$KM\sin\theta_2\cos\theta_2 - H_x\cos\theta_2 + H_z\sin\theta_2 + \epsilon M\sin(\theta_1 - \theta_2) = 0 \quad (36)$$

where we have implicitly assumed $M^1 = M^2 = M$.

The solution to this system may be obtained by expanding about $\theta_1 \approx 0$ and $\theta_2 \approx \pi$ (assuming the z-axis to have an attractive anisotropy field) or other numeric methods may be employed. In principle, however, one can obtain the angles θ_1 and θ_2 and thus obtain the 2 - sublattice resonance frequency.

F. A discussion of higher sublattice systems

In the above example, we considered only the simplest system where an exact solution was possible for the equilibrium coordinates. The solution involved a quartic equation. For higher sublattice systems, the problem becomes rapidly more complicated, however, the equilibrium conditions may be linearized and solutions obtained for small deviations from zero-field angles. In addition, several specific cases of higher sublattices systems may be exactly solvable for very specific cases but in general the solution of the equilibrium condition will require numerical analysis. In principle, however, one could obtain at least the high-field limiting cases for the higher sublattice system through the use of Eq. (9) without solving for equilibrium conditions. This is particularly important for our work with the lower dimensional compounds which do not necessarily have enormous antiferromagnetic exchange fields present.

III. Conclusion

We have reviewed the generalization of ferro- and antiferromagnetic resonance theory. We call this ordered state resonance since it includes systems of many sublattices and is valid for general free energy models. For the case of a 1-sublattice (ferromagnetic) system with uniaxial anisotropy, it was shown that an exact solution to the equilibrium equations is not only possible but actually valid in more experimental circumstances than earlier theories. The application of OSR to the two sublattice system is also reviewed.

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