Spontaneous Resonances and Universal Behavior in Ferrimagnets: Effective-Medium Theory

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We reconsider the high-frequency susceptibility of partially magnetized uniaxial ferrimagnets. For a demagnetized sample, effective-medium theory reproduces the exact result. Spontaneous resonances appear in a band of frequencies, in which the susceptibility follows a universal behavior. A new effect is found; a weak magnetization m "digs a hole" in this resonant band, of width proportional to m. The formulas we propose agree with experimental data, from which microscopic parameters can be obtained. These also apply to the resistivity of a (2D) conductor in a random magnetic field.

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The response of a ferro(i)magnet to a small transverse oscillating magnetic field strongly depends on the overall magnetization of the sample. In the *fully magnetized* case, the susceptibility $\chi(\omega)$ has a typical resonant shape, corresponding to the damped precession of the spins around the external magnetic field H at the Larmor frequency $\omega = \gamma H$ (ferromagnetic resonance; γ is the gyromagnetic ratio, $\gamma/2\pi = 2.8$ MHz/Oe for a free electron). The line is quite narrow, and its shape satisfactorily accounted for by the Landau-Lifshitz-Gilbert equation. The situation is markedly different in *demagnetized samples* for which, surprisingly and despite several efforts,[1-4] no convincing quantitative theory is available. The imaginary part of the susceptibility of demagnetized $Ni_xZn_{1-x}Fe_2O_4$ (for example) exhibits two rather well resolved peaks, corresponding to the excitation of the internal modes of the system. The low-frequency peak (10 MHz) generally depends strongly on composition, porosity, and grain size, and is due to domain-wall motion, a mechanism that we shall not discuss further. The high-frequency peak ($\simeq 100$ MHz) is mostly insensitive to the grain size and is extremely broad (much broader than the ferrimagnetic resonance line): It extends up to the highest possible value of the demagnetizing field, $\omega_{max} = \gamma 4\pi M$, where M is the saturation magnetization. It is due to spin rotations, and very early was accounted for qualitatively by Polder and Smit,[1] who argued that the presence of domains of various sizes and opposite directions of the magnetization would induce a full distribution of local demagnetizing fields between 0 and $4\pi M$ and thus of resonances between $\omega_{min} = \gamma H_a$ (the anisotropy field) and $\omega_{max} = \gamma(H_a + 4\pi M)$ $(\simeq \gamma 4\pi M)$. A quantitative study of this effect amounts to solving Maxwell's equations for the magnetic field and induction in an inhomogeneous medium, where the value of the local susceptibility depends upon the orientation of the local magnetization (+ or − in a uniaxial ferrimagnet). This problem is thus of much the same nature as that of determining the average conductivity (or diffusivity) of a random mixture (except for the tensorial nature of the local magnetic susceptibility, see below) which has received enormous theoretical attention recently.[5,6] Apart from systematic weak-disorder expansions,[7] the most successful and versatile approach is the "effective-medium theory[8,9] (EMT) which, albeit approximate, is known to be exact in one dimension and for weak disorder (for all frequencies),[7,8] satisfies rigorous bounds,[10] and predicts qualitatively a nontrivial phenomenon: percolation.[8,11] The possible applications of EMT are so numerous (e.g., optical properties of fine metallic particles,[12] permeability of a mixture of porous media,[13] see also Refs. 5 and 6) that it is most important to exhibit dramatic predictions of EMT (such as percolation), and to validate them experimentally, at least qualitatively.

When the sample is completely demagnetized ($m = 0$, m is the reduced magnetization $|m| \leq 1$), EMT reproduces the exact result for two-dimensional domain structures (which is a very good approximation within one grain). It predicts that a resonance "band" appears between ω_{min} and ω_{max} and provides a formula [Eqs. (4) and (5) below] for $\chi(\omega)$ in the whole frequency range. The generalization to partly magnetized samples predicts a new effect: Resonances are *inhibited* in a frequency range of width $2\pi Mm$ centered on $\bar{\omega} = \gamma(H_a 4\pi M)^{1/2}$ which means that a "hole" in the $\chi''(\omega)$ curve appears around $\bar{\omega}$. Such an effect could be experimentally verified, and would thus test EMT in novel and rather "exotic" situations. The formula we obtain for $\chi(\omega)$ for $m = 0$ was in fact derived by Schlömann,[4] who explicitly solved the problem in the case of a very particular (and rather unrealistic) geometry of circular, concentric domains. We thus provide a sound foundation for this formula, which reproduces experimental results (Refs. 4 and 14, and below) quite successfully, and in fact allows one to extract from them useful microscopic information such as M, the relaxation time, and the anisotropy field.

We now present briefly the derivation of our results. The problem is to obtain the effective permeability $\hat{\mu}$ of a uniaxial magnetic sample broken into + or − domains of arbitrary cylindrical shape, with a local permeability tensor $\hat{\mu}_\pm$. A transverse time-dependent field $\mathbf{h}$ is applied to the sample. For sufficiently long wavelengths (larger than the domain size), one has $\nabla\times\mathbf{h}=0$ and $\nabla\cdot\hat{\mu}_\pm\mathbf{h}=0$, where the local Polder tensor in the plane (x,y) perpendicular to the magnetization reads[15]

$$\hat{\mu}_\pm=\begin{pmatrix}\mu & \mp i\kappa\\ \pm i\kappa & \mu\end{pmatrix} \tag{1}$$

with

$$\mu=1+\gamma^2\frac{4\pi MH_a}{\omega_{\min}^2-\omega^2} \tag{2a}$$

and

$$\kappa=\gamma 4\pi M\frac{\omega}{\omega_{\min}^2-\omega^2}. \tag{2b}$$

(We have neglected intrinsic relaxation, see below.) Equation (1) simply arises from the linearization of the equation of motion: $\partial_t\mathbf{M}=\gamma\mathbf{H}\times\mathbf{M}$. Note that off-diagonal terms change sign when $z\to -z$. Assuming that the domain structure is sufficiently random so that the equivalent medium is rotationaly invariant, the EMT averaging procedure states that the effective permeability tensor $\bar{\hat{\mu}}$ satisfies the self-consistent equation depicted in Fig. 1: A small + or − domain is immersed in a homogeneous medium characterized by $\bar{\hat{\mu}}$. This perturbs the overall permeability by a small amount $\delta\hat{\mu}_\pm$ which is calculated by solving Maxwell's equations in the geometry of Fig. 1 (in the quasistatic limit). $\bar{\hat{\mu}}$ is then obtained by requiring that $\sum_{+,-}\frac{1}{2}(1\pm m)\delta\hat{\mu}_\pm\equiv 0$.

This approximation scheme yields our central result: Writing

$$\bar{\hat{\mu}}=\begin{pmatrix}\bar{\mu} & -i\bar{\kappa}\\ i\bar{\kappa} & \bar{\mu}\end{pmatrix},$$

one has

$$\bar{\mu}^2-\bar{\kappa}^2=\mu^2-\kappa^2, \tag{3a}$$

$$\bar{\kappa}=m\kappa\bar{\mu}/\mu. \tag{3b}$$

Let us study the $m=0$ case first. From Eqs. (3) and (2) one gets

$$\bar{\mu}=(\mu^2-\kappa^2)^{1/2}=\left[\frac{(H_a+4\pi M)^2-(\omega/\gamma)^2}{H_a^2-(\omega/\gamma)^2}\right]^{1/2}, \tag{4a}$$

$$\bar{\kappa}=0. \tag{4b}$$

These, in fact, coincide with the exact results: $\bar{\kappa}=0$ follows from the $z\to -z$ symmetry, while (3a) can be proven (for any m) due to a "duality symmetry" of two-dimensional Maxwell equations (along lines similar to Ref. 16). One can also invoke a general theorem of homogenization theory in 2D which states[17] that if the determinant of the local permeability tensor $\hat{\mu}$ is constant over the sample (which is the case here since $\hat{\mu}_+={}^t\hat{\mu}_-$), the determinant of the homogenized permeability is equal to this constant value. Therefore (3a) in fact relies and only relies on the very assumption of the existence of an isotropic effective medium. It does not apply, in particular, to a lamellar configuration of domains: In this (1D) case one may show that, for small intrinsic damping, the *only* resonant frequency is $\bar{\omega}=(\omega_{\min}\omega_{\max})^{1/2}$. EMT thus yields an exact result for Eq. (3a) and is probably only an approximation for Eq. (3b) (see Ref. 18).

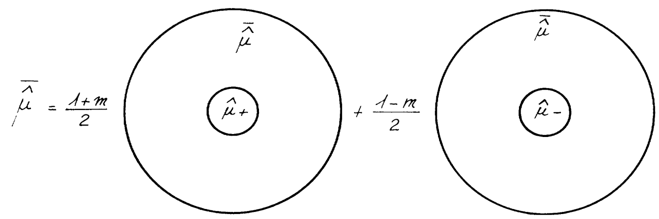

FIG. 1. Graphical representation of the effective-medium equation: A small "impurity" is included in the effective medium, and its contribution to the permeability must (to the lowest order) average to zero. Magnetostatic interactions between domains are thus in a way taken into account.

From (4a) one directly obtains that the susceptibility is purely imaginary for frequencies spanning the interval $[\omega_{\min},\omega_{\max}]$, while dissipation disappears outside this range. This is quite an extraordinary result: The medium is made of two components (which differ by $\kappa\to-\kappa$), each of which has only one resonance at $\omega_{\min}$, and one ends up with a dissipative medium in a whole band of frequencies. The physical origin of this effect is that a magnetic field along x excites a y component of the magnetization through the off-diagonal element of $\hat{\mu}$. This y component in − domains is out of phase with the neighboring + domains, and this creates uncompensated magnetic charges at the boundaries: + and − domains act in a sense as capacitances and inductances. The situation is analogous to the case of a (2D) mixture of an insulator and a metal[12] in equal proportion, for which the effective permittivity is given by[16] $\bar{\epsilon}=(\epsilon_{\rm met}\epsilon_{\rm ins})^{1/2}$. In the infrared range, the metal has a negative permittivity and the mixture is thus also characterized by "spontaneous resonances" (imaginary permittivity). We believe, however, that magnetic materials are far more convenient for the experimental investigation of this peculiar effect.

The analysis of Eqs. (3) in the partly magnetized case reveals that

$$\bar{\mu}^2=\mu^2\left[\frac{\mu^2-\kappa^2}{\mu^2-m^2\kappa^2}\right]$$

takes negative values only in a *restricted* interval of frequencies, which for $m\ll 1$, $\omega_{\max}\gg\omega_{\min}$ reads $[\omega_{\min},\bar{\omega}-2\pi m\gamma M]$, $[\bar{\omega}+2\pi m\gamma M,\omega_{\max}]$. Furthermore, the susceptibility is no longer diagonal, and the off-diagonal elements are linear in m for $m \ll 1$, a result first obtained by Rado[3] [(3b) in fact yields Rado's result in the large-frequency limit]. The above equation is correct only if one can neglect the external field and the induced demagnetizing field (of the order of NmM, where N is the demagnetizing factor along z).

Two modifications must be included in the previous theory to account quantitatively for experimental results on demagnetized samples.

(a) Intrinsic dissipation can be phenomenologically described by substituting $i\omega$ by $(i\omega - 1/\tau)$ in the denominator of Eqs. (2). (See Ref. 19 for a more precise discussion.) This will smear out the divergence at $\omega_{\min}$ appearing in (4a) and will add a real part to χ in the resonance "band." Namely, for $\omega \ll \omega_{\max}$,

$$\bar{\mu}'' = \frac{\omega^2-\omega_0^2}{(\omega^2-\omega_0^2)^2+4\omega^2\tau^{-2}}\gamma 4\pi M\omega\,, \tag{5a}$$

$$\bar{\mu}' = \frac{2\omega^2\tau^{-1}}{(\omega^2-\omega_0^2)^2+4\omega^2\tau^{-2}}\gamma 4\pi M\,, \tag{5b}$$

with $\omega_0^2 = \omega_{\min}^2 + \tau^{-2}$.

In particular, in the regime $\omega_0 \ll \omega \ll \omega_{\max}$ (reasonable values are $\omega_0/2\pi \simeq 20$–50 MHz and $\gamma 4\pi M \simeq 12000$–$14000$ MHz), one has

$$\bar{\mu}' \simeq \gamma 8\pi M\tau^{-1}/\omega^2\,, \tag{6a}$$

$$\bar{\mu}'' \simeq \gamma 4\pi M/\omega\,. \tag{6b}$$

Both the real and the imaginary part of the permeability of insulating ferromagnets thus exhibit a *universal* power-law behavior in a rather wide frequency range. Note also that Eq. (5b) shows that μ' no longer takes negative values: Interactions between domains have converted a resonant response [Eqs. (2)] into a relaxation response [Eqs. (5)].

(b) The z axis may vary in direction from grain to grain. The simplest way to take this effect into account is to neglect grain-grain magnetostatic interactions; the angular average then simply multiplies the above results [Eqs. (6)] by a factor $\frac{2}{3}$.[4] More generally, one may leave this angular factor α as an adjustable parameter (restricted to the range [0,1]) to allow for angular and magnetic correlations. As an illustration, we have fitted the well-known susceptibility of $Ni_{0.36}Zn_{0.64}Fe_2O_4$ obtained by Verweel[20] in 1964 in the range 80–1000 MHz, where the domain-wall contribution is assumed to be small. The laws (6a) and (6b) are obviously followed on the log-log plot of the data (see Fig. 2, inset), from which one obtains just by inspection $\alpha\gamma 4\pi M \simeq 10000$ MHz and $2/\tau \simeq 40$ MHz.[21] From independent M measurements[22] (and taking into account the value of the gyromagnetic factor[2] $g \simeq 2.2$), one finds $\alpha \simeq 0.73$, which is indeed close to $\frac{2}{3}$. The full theoretical curves for χ' and χ'' are compared to experimental points in Fig. 2. Agreement is very good for both χ' and χ'' simultaneously, except in the low-frequency part where a precise determination of τ is important and where the domain walls certainly start to contribute significantly.[19] The same quality of fit has been achieved[19] on other NiZn compositions and $MnZnFe_2O_4$. The sudden change of behavior of χ for $\omega_{\max}$ [see Eq. (4a)] is also very clearly observed experimentally.[2]

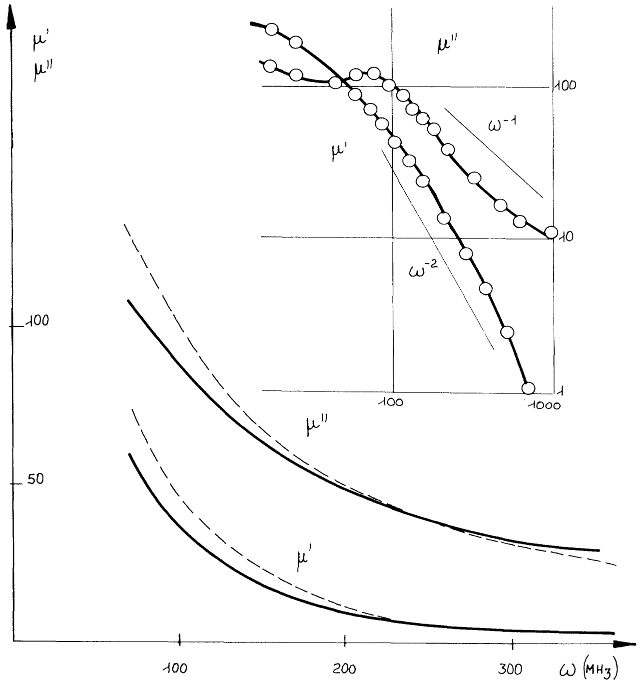

FIG. 2. Verweel's susceptibility data on NiZn (Ref. 20) (dashed lines) compared with expressions (5a) and (5b) (full lines). Inset: These data are on a log-log plot which makes apparent the laws ω^{-2} and ω^{-1} for χ' and χ''.

A very interesting analog of the magnetic problem presented here is the conductivity of a two-dimensional conductor in a random magnetic field $\pm B$ (or a mixture of two conductors of opposite ratio of the carrier charge to the carrier mass in a constant magnetic field). Denoting ω_c as the cyclotron frequency, the local resistivity tensor reads (in a Drude approximation)

$$\hat{\rho} = \rho_0 \begin{pmatrix} 1+i\omega\tau & \omega_c\tau \\ -\omega_c\tau & 1+i\omega\tau \end{pmatrix}.$$

In a *symmetrical* configuration ($\langle\omega_c\rangle = 0$), the average resistivity is scalar and equal to

$$\bar{\rho}(\omega) = \rho_0[1+(\omega_c\tau)^2-(\omega\tau)^2+2i\omega\tau]^{1/2}.$$

In particular, for $\omega_c\tau \gg 1$, the dc resistivity is much enhanced, $\bar{\rho}(\omega=0) = \rho_0\omega_c\tau$; due to spontaneous resonances, the relevant inelastic time is ω_c^{-1} rather than τ.

Summarizing, we have proposed that a partially magnetized ferromagnet realizes a particularly interesting and original "random" medium where "mixture" equations could be neatly studied and tested. We have adapted EMT to this problem and showed that it predicts new, nontrivial effects. It leads to the exact expression for $m=0$. As first recognized by Schlömann[4] in a particular case, a resonance band appears, terminating at $\gamma 4\pi M$, which explains a well-known experimental fact named "low-field losses" or "Polder-Smit resonances.[1,2] For small intrinsic relaxation, this resonance band would nearly entirely disappear for a regular lamellar arrangement of domains. The theory also predicts an "absorption trough" for $m \neq 0$, of width proportional to m: We claim that it would be a stringent test of EMT to evidence this "antiresonance" band. Conversely, comparison with experiments allows one to extract from susceptibility measurements on demagnetized samples values of M, H_a, and τ: This has recently been successfully tried in Refs. 14 and 19. Note finally that EMT may be used, along the same lines, to calculate the influence of inclusion of nonmagnetic phases. The theory for nonuniaxial magnets and ferromagnetic films will be presented elsewhere.[19]

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[18]Note that it does *not* coincide with the formula one would naively obtain by applying the standard EMT formula to the eigenvalues of the permeability tensor. The latter would lead to Eq. (3a) but not to (3b)—instead one would get $\bar{\kappa}=m\kappa$, and hence a *narrowing* of the band, instead of a hole for nonzero m.

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[21]This value of τ^{-1} is of the order of magnitude of γH_a, which is not unexpected in a demagnetized sample, where the typical fluctuations of magnetic field should be $\simeq H_a$ and no dipolar narrowing should occur; see C. W. Haas and H. B. Callen, in *Magnetism,* edited by G. T. Rado and H. Suhl (Academic, New York, 1963), Vol. 1. It would be interesting to compare this value with the one obtained in a ferrimagnetic resonance experiment.

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