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Intrinsic magnetic resonance in nanoparticles: Landau damping in the collisionless regime

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Abstract

In a uniaxially anisotropic particle the frequency of intrinsic ferromagnetic resonance depends on the angle deviation of the magnetic moment from its equilibrium position. At low spin–lattice relaxation and in the presence of superparamagnetism, the regime of relaxation in an assembly of such particles closely resembles the Landau damping regime, which is well known in the kinetic theory of plasma. In the high-temperature limit in the case of polydisperse systems, particle volume and surface anisotropy affect the absorption lines differently. © 2002 Published by Elsevier Science B.V.

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Superparamagnetism is a well-known thermofluctuational effect responsible for observable loss of equilibrium magnetization in nanosize objects. Less known is the fact that the rotary diffusion of the magnetic moments of single-domain particles also strongly affects the precession mode. The essential rôle of thermal noise in the formation of the ferromagnetic resonance (FMR) spectra of fine-particles was reported in 1974 [1], and thus it has taken over two decades to fully understand its effect on the dynamic susceptibility. Let a magnetic moment $\boldsymbol{\mu}$ be deviated at zero temperature from its equilibrium position. Then, according to the Landau–Lipshitz equation

$$\mathrm{d}\boldsymbol{\mu}/\mathrm{d}t = -\gamma(\boldsymbol{\mu} \times \boldsymbol{H}_{\mathrm{eff}}) - \gamma\alpha(\boldsymbol{\mu} \times (\boldsymbol{\mu} \times \boldsymbol{H}_{\mathrm{eff}})) \qquad (1)$$

(with γ and α being the gyromagnetic ratio and the damping parameter, respectively) vector $\boldsymbol{\mu}$ begins to precess with an angular velocity $\sim\gamma|\boldsymbol{H}_{\mathrm{eff}}|$ circumscribing a conical surface with the symmetry axis along the effective field $\boldsymbol{H}_{\mathrm{eff}}$. With time the cone shrinks due to spin–lattice relaxation, and $\boldsymbol{\mu}_\perp$, the projection on the plane normal to $\boldsymbol{H}_{\mathrm{eff}}$, decays as a deterministic vector. Now let us introduce a heat bath, i.e., fluctuations that disorient the magnetic moment at random. In this way one "switches on" the orientational diffusion that accelerates the decay of the macroscopic (observable) quantity $\langle \boldsymbol{\mu}_\perp \rangle$, the ensemble average of $\boldsymbol{\mu}_\perp$. In Ref. [1], the appropriate theory was developed for the case of intrinsic FMR, where H_{eff} consists solely of the internal field of magnetic anisotropy $H_{\mathrm{a}} = 2K/I$, with K being the volume density of the magnetic anisotropy energy and I the particle magnetization. Assuming the perturbations $\boldsymbol{\mu}_\perp$ to be infinitesimal, the conclusion obtained in Ref. [1] was that with increasing temperature the character of the magnetic moment motion changes from a resonance to an overdamped one. In the high-temperature limit the decay rate (and thus, the linewidth $\Delta\omega$) is proportional to αkT, i.e., the product of the damping parameter and temperature. In other words, under heating, the absorption line of intrinsic FMR experiences unlimited *homogeneous* widening.

The predictions of Ref. [1] looked so reasonable that for a long period no revision seemed necessary. However, on closer inspection, the claimed proportionality $\Delta\omega \propto \alpha kT$ that yields $\Delta\omega = 0$ at $\alpha \to 0$ whatever the temperature, does not seem physically justified. A correct way to analyze the case of $\alpha \to 0$ was outlined in Ref. [2] and applied to intrinsic FMR in Ref. [3]. It turned out that at $\alpha = 0$ the linear-response (Kubo) theory gives an exact solution for the dynamic susceptibility. The result is not a zero-width line, as it follows from the approximation [1] in the limit $\alpha \to 0$. In the linear-response treatment, one explicitly takes into

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account that for intrinsic FMR the precession frequency is

$$\omega(\theta) = \omega_a \cos\theta, \quad \omega_a \equiv \gamma H_a, \tag{2}$$

and thus depends on the angle deviation θ of vector $\boldsymbol{\mu}$ from the easy axis.[1] In Ref. [1] this dependence was ignored, and the condition $\omega(\theta) = \omega_a$ was imposed assuming $\theta \ll 1$. However, a unique feature of nanoparticles is that even at sufficiently low temperatures the ratio $\sigma = \mu H_a/2kT$ can easily become less than unity. Writing down the Boltzmann law as $\exp(\sigma\cos\theta)$, one finds that at finite temperatures the magnetic moment orientations, θ, are scattered considerably over the interval $[0, \pi/2]$. Consequently, in a superparamagnetic particle a single response (precession) frequency ω_a is replaced by a response bandwidth $[0, \omega_a]$, i.e., a *non-homogeneous* widening of the absorption line takes place.

Consider an assembly of such particles (their easy axes being parallel) subjected to a circularly polarized probing field $\boldsymbol{h} \perp \boldsymbol{H}_a$. It would respond at any $\omega < \omega_a$, with the magnitude of the response depending on how densely the corresponding $\omega(\theta)$ state is populated. The absorption line χ'' (the imaginary part of the dynamic susceptibility) is calculated as follows. The Landau–Lipshitz equation for the intrinsic FMR is written as

$$\mathrm{d}\boldsymbol{e}/\mathrm{d}t = -\omega_a(\boldsymbol{en})[(\boldsymbol{e}\times\boldsymbol{n}) + \alpha(\boldsymbol{e}\times(\boldsymbol{e}\times\boldsymbol{n}))], \tag{3}$$

where $\boldsymbol{e} = \boldsymbol{\mu}/\mu$ and $\boldsymbol{n}$ are the unit vectors of the magnetic moment and easy axis, respectively. Then the friction term is replaced by $-\varepsilon$ that is an infinitesimal damping constant, and the circular components of $\boldsymbol{e}$ are expressed in terms of the polar angles as

$$e_x = \mathrm{Re}\Psi, \quad e_y = \mathrm{Im}\Psi, \quad \Psi = \sin\theta\exp(\mathrm{i}\varphi). \tag{4}$$

Substitution of Eq. (4) into Eq. (3) yields a solution of the form

$$\Psi(t) = \Psi(0)\exp[(\mathrm{i}\omega_a\cos\theta_0 - \varepsilon)t],$$
$$\Psi(0) = \sin\theta_0\exp(\mathrm{i}\varphi_0), \tag{5}$$

where index 0 denotes the initial values of $\boldsymbol{e}$. Clearly, Eq. (5) corresponds to the azimuthal motion and conserves the initial value θ_0. The dynamic susceptibility is evaluated with the aid of the Kubo theorem, which in terms of Eq. (5) may be written as

$$\chi = -\frac{c\mu^2}{kT}\int_0^\infty \mathrm{d}t\,\mathrm{e}^{-\mathrm{i}\omega t}\frac{\partial}{\partial t}\langle\Psi(t)\Psi^*(0)\rangle$$
$$= \frac{c\mu^2}{kT}\left\langle\frac{\omega_a\cos\theta_0\sin^2\theta_0}{\omega_a\cos\theta_0 - \omega + \mathrm{i}\varepsilon}\right\rangle, \tag{6}$$

with c being the particle number concentration; here the angular brackets mean the statistical averaging with respect to the equilibrium ($\boldsymbol{h} = 0$) state of the ensemble and the retained damping ε ensures a correct choice of the integration path.

Formula (6) describes the oscillatory response from a statistical assembly of magnetic moments. However, in the spirit of the Kubo theory the vertex angles θ_0 of the precession cones are taken as that distributed according to the Boltzmann (i.e., equilibrium) law $\exp(\sigma\cos\theta_0)$. Performing the configurational integration in Eq. (6) and then setting $\varepsilon = 0$, for the imaginary component of the dynamic susceptibility one gets

$$\chi''(\omega) = (\pi c\mu^2/2kTR)x(1-x^2)\exp(\sigma x^2),$$
$$x \equiv \omega/\omega_a, \quad R \equiv \int_0^1 \exp(\sigma y^2)\,\mathrm{d}y. \tag{7}$$

In the high-temperature limit, $\sigma \ll 1$, where $\exp(\sigma)$ approaches unity, Eq. (7) reduces to

$$\chi''(\omega) = (\pi c\mu^2/4kT)x(1-x^2). \tag{8}$$

Note that although the linewidths of individual oscillators are exactly zero, the lineshape (8), shown in Fig. 1 by a dashed line, has the finite width $\Delta x = \Delta\omega/\omega_a = 1$.

We remark that a striking similarity exists between the described effect in single-domain particles which is known as the *Landau damping* in the kinetic theory of plasma. Attenuation of electromagnetic waves in a fully ionized plasma is entirely due to this fundamental mechanism. Mathematically, in both cases a finite

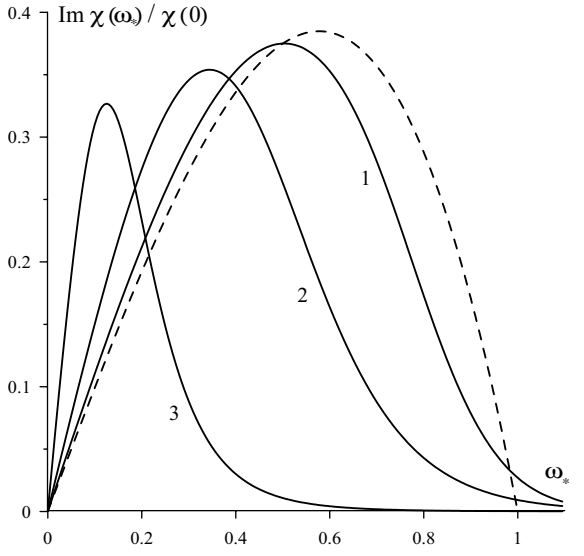

Fig. 1. Absorption line of intrinsic FMR in an assembly of nanoparticles with uniaxial surface anisotropy. The polydispersity is described by the gamma distribution; $\beta = 50$ (1), 10 (2), and 1 (3), the dashed line corresponds to a monodisperse system.

[1] In fact, Eq. (2) is a particular case of a more general problem considered in Ref. [4], where it is shown that a uniform precession is an exact solution for the finite-amplitude case as well.

absorption line results from the averaging of a Lorentzian line of an infinitesimal width over some distribution of resonance frequencies. Physically, this means that the characteristic frequencies of the process under consideration are far greater than the "collision rate" responsible for the energy diffusion in the system.

Another specific feature of intrinsic FMR in the $\alpha \to 0$ limit is that, unlike the majority of magnetic resonance phenomena, it is insensitive to the polydispersity of the particles. Indeed, since the resonance frequency, ω_a, by definition does not contain the particle size d, the size averaging applies only to the prefactor in formula (8). There μ^2 transforms to $\pi^2 I^2 \langle d^6 \rangle / 36$ and one observes that neither the frequency dependence nor the width, $\Delta\omega$, of the absorption line $\chi''(\omega)$ is affected. However, this conclusion holds only for the particles with volumetric magnetic anisotropy, i.e., $H_a = \text{const}(d)$. If this is the case [5,6] where the surface anisotropy is dominant, then situation changes. Then the basic frequency ω_a becomes inversely proportional to the particle size, and, accordingly, the absorption line of intrinsic FMR becomes sensitive to polydispersity. Introducing a uniaxial surface anisotropy of the Aharoni-type [7] with the energy density K_S, one finds that

$$\omega_a(d) = 12\gamma K_S / Id. \tag{9}$$

To demonstrate the effect, which the particle polydispersity has on $\chi''(\omega)$, we average Eq. (8) with the gamma distribution $f(d) = (d/d_0)^\beta \exp(-d/d_0)/\Gamma(\beta + 1)$; here Γ is the gamma function. In terms of the gamma distribution, the mean particle diameter is $\langle d \rangle = (\beta + 1)d_0$. In Fig. 1, the curves of χ'' for different β are plotted as a function of the dimensionless frequency $\omega_* = \omega/\omega_a(\langle d \rangle)$. Note that for all the curves the mean particle size, $\langle d \rangle$, is kept constant. For comparison, the absorption line of a monodisperse system ($d = \langle d \rangle$, $\beta = \infty$) is also presented. One sees the remarkable effect which the surface anisotropy has on χ'': the broader the distribution, the narrower is the absorption line of intrinsic FMR.

In conclusion, intrinsic FMR in superparamagnetic nanoparticles is a unique situation where the linear-response spectrum of the magnetic moment is directly affected by its equilibrium distribution function. The absorption lineshape has a striking resemblance with one describing the attenuation of electromagnetic waves in a fully ionized plasma (Landau damping).

In a polydisperse system, the absorption line of intrinsic FMR has a parabolic shape as long as the particle magnetic anisotropy is of a volumetric origin. A tendency of the line to deviate from the parabolic profile (8) (see Fig. 1) and to acquire an exponential shape, indicates the presence of surface magnetic anisotropy of the particles.

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References

[1] Yu.L. Raikher, M.I. Shliomis, Sov. Phys. JETP 40 (1974) 526.
[2] R.S. Gekht, Fiz. Met. Metalloved. (Metal Phys. & Metallograph) 55 (1983) 225.
[3] Yu.L. Raikher, V.I. Stepanov, Phys. Rev. B 51 (1995) 16428.
[4] G. Bertotti, C. Serpico, I.D. Mayergoyz, Phys. Rev. Lett. 86 (2001) 724.
[5] F. Gazeau, J.-C. Bacri, F. Gendron, R. Perzynski, Yu.L. Raikher, V.I. Stepanov, J. Magn. Magn. Mater. 186 (1998) 175.
[6] V.P. Shilov, J.-C. Bacri, F. Gazeau, F. Gendron, R. Perzynski, Yu.L. Raikher, J. Appl. Phys. 85 (1999) 6642.
[7] A. Aharoni, J. Appl. Phys. 61 (1987) 3302;
A. Aharoni, J. Appl. Phys. 64 (1988) 6434.