ESTIMATION OF EXCHANGE CONSTANT *A* AND *g* FACTOR FOR Co_xNi_{1-x} MICROSPHERES FROM SIZE DEPENDENT FERROMAGNETIC RESONANCE MODES

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Abstract

The exchange constants A and the g factors for Co_xNi_{1-x} microspheres are estimated by comparing theoretical calulations of size dependent resonance modes with experimental data of spherical monodisperse Co-Ni particles. Only cylindrically symmetric modes are studied. Comparison with previously reported values is performed.

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I. INTRODUCTION

The internal structure, the properties and the response of nano and micro-scale ferromagnetic particles to: external magnetic fields, applied mechanical stresses or change in temperature, are of great importance due to the variety of applications in diverse fields. Chains of magnetic nanoparticles (called magnetosomes), help magnetotactic bacteria, birds, fish and bees to navigate in the earth's magnetic field [1,2]. The detection of ferromagnetic nanoparticles in the human brain and other human tissues raze questions of whether weak electromagnetic fields might have biological effects, including cancer [3]. Among the method,s used to destroy cancer tumors, is the use of vesicles that contain magnetic microspheres and can target drugs to specific location inside the human body via externally applied magnetic fields [4]. Moreover, colloidal suspensions of ferromagnetic nanoparticles in a liquid carrier, well known as ferrofluids [5], find many technological [6] and medical applications [7]. A phenomenological approach to describe such coupled phenomena in ferromagnetic solids is the continuum theory of micromagnetics [8,9].

For particles with diameters above $1\,mm$, magnetostatic forces dominate over exchange interactions. In this size range the resonance spectrum was studied for spheroids by Walker [10] and the corresponding modes are known as *magnetostatic or Walker modes* (MRM). For particles with diameters below 1μm, the exchange forces are the predominant ones. The corresponding modes of precession, *exchange resonance modes*(ERM), were studied by Aharoni for the sphere [16], but the analysis applies also to prolate and oblate spheroids. In this case the resonance frequency depends on the size of the particles as well as on the roots of the derivatives of spherical Bessel functions. These two originally theoretically predicted features of ERM could explain qualitatively [11–13] and in some cases quantitatively [14,15], related experiments. Surface anisotropy effects were discussed in [17] in order to account for the experimentally observed size dependence. Similar calculations have also been performed in [18]. In the intermediate size range, magnetostatic and exchange interactions are of the same order of magnitude and there is a mixing of the modes for particles above the size of about 1 μm. Such calculations have been performed previously [19,20], with the material parameters of magnetite, assuming cylindrical symmetry, and were completed in a recent communication [21], for the material parameters of Ni [20], in order to account for related experiments in this size range [11–15].

Ferromagnetic resonance has always been considered as the most accurate method of measuring the exchange constant A, and the g factor. It is the aim of the present work to complete the comparison of the theoretical analysis [21], with the experimental results for Co_xNi_{1-x} microspheres [11–15]. The estimated exchange constants A and g factors are compared with previous reported values [22].

II. THEORY

The magnetization dynamics in an external field is studied by the Landau-Lifshitz equation:

$$\frac{\mathrm{d}\boldsymbol{v}}{\mathrm{d}t} = \gamma_0 \boldsymbol{v} \times \left(\frac{C}{M_s} \nabla^2 \boldsymbol{v} - \frac{1}{M_s} \frac{\partial w_a}{\partial \boldsymbol{v}} + \boldsymbol{H} \right), \tag{1}$$

where $\boldsymbol{v}$ is a unit vector parallel to the magnetization, $C = 2A$ is the exchange constant, w_a is the anisotropy energy density, M_s is the saturation magnetization, t is time, γ_0 is the gyromagnetic ratio and $\boldsymbol{H} = \boldsymbol{H}_0 + \boldsymbol{H}'$ is the magnetic field which is composed of the applied field $\boldsymbol{H}_0$ and that, $\boldsymbol{H}'$, created by the volume and surface magnetic charges. For negligible surface anisotropy, the boundary conditions for the set of equations (1) are

$$\frac{\partial \boldsymbol{v}}{\partial n} = 0, \tag{2}$$

where $\partial/\partial n \equiv \boldsymbol{n} \cdot \nabla$ and $\boldsymbol{n}$ is the outward normal to the particle surface. The self-field $\boldsymbol{H}' = -\nabla V$, is determined from Maxwell' s equations of magnetostatics [8]

$$\begin{aligned} &\nabla^2 V_{\text{in}} = 4\pi \nabla \cdot \boldsymbol{M} && \text{inside the particle} \\ &\nabla^2 V_{\text{out}} = 0 && \text{outside the particle.} \end{aligned} \tag{3}$$

with the following boundary conditions on the particle surface

$$V_{\text{in}} = V_{\text{out}} \tag{4}$$
$$\frac{\partial V_{\text{in}}}{\partial n} = \frac{\partial V_{\text{out}}}{\partial n} + 4\pi M_s \boldsymbol{n} \cdot \boldsymbol{v}.$$

If a large dc field H_0 is applied along a direction which is defined as the $z-$ axis, the magnetization is almost parallel to this direction z, so that v_x and v_y are small. This is usually the case in experimental studies of resonance. Then the differential equations (1) are linearized around the field direction and reduce to (the factor $( )e^{i\omega t}$, is omitted):

$$\left(\frac{C}{M_s}\nabla^2 - H_z\right) v_x - \frac{i\omega}{\gamma_o} v_y = \frac{\partial V_{\text{in}}}{\partial x} \tag{5a}$$

$$\left(\frac{C}{M_s}\nabla^2 - H_z\right) v_y + \frac{i\omega}{\gamma_o} v_x = \frac{\partial V_{\text{in}}}{\partial y}, \tag{5b}$$

where V_{in} is the potential due to the transverse magnetization v_x and v_y, and ω is the resonance frequency. The potential due to the z component is included in H_z, which for the case of the sphere studied here, has the form

$$H_z = H_0 - \frac{4\pi}{3} M_s + \frac{2K_1}{M_s}, \tag{6}$$

where K_1 is the anisotropy constant. MRM are solutions of the above set of equations with $C = 0$, while ERM are solutions, if we set $V_{\text{in}} = 0$ in Eqs (5) and omit Maxwell' s equations of magnetostatics (3-4).

In the case where magnetostatic and exchange forces are of the same order of magnitude, one has to solve the full set of equations. This has been attempted in the past and the modes of precession were calculated [19,20]. The recent experiments in this size range [11–15] forced us to reconsider the solution procedure. The original arbitrariness in the radial dependence of the solution was corrected and computations were performed for Ni microspheres [21]. In the present work the proposed solution procedure is extended in order to account for Co_xNi_{1-x} microspheres.

III. RESULTS AND DISCUSSION

The general solution and the frequency equation for the boundary value problem Eqs. (2-5) is given in [21]. We performed the calculations (like in related experiments) without any static magnetic field applied ($H_0 = 0$). All the computations were carried out with the same numerical algorithm but with material constants for Co_xNi_{1-x} microspheres given in Table I. Though the algorithm permits the computation of the size dependence of the resonance modes, we focused our attention in resonance frequencies, and thus on the estimation of the exchange constant A and the g factor, material parameters which are usually determined by ferromagnetic resonance. For this reason we kept the diameter, $2R$ of the sphere, fixed to the experimental value, for each particle with different chemical composition x (see Table I of Ref. [13]) and we repeated the solution procedure by varying A and g, until the eigenfrequency agreed with the experimental value. The eigenfrequencies $f^{(k,n)}$ are also included in Table I along with the experimental values f^{exp} ($f \equiv \omega/2\pi$) and the corresponding cylindrically symmetric resonance modes (k,n). Note that for higher concentration of Co higher modes are needed in order to satisfy the convergence criterion given in [21]. This is mainly due to the high magnetocrystalline anisotropy constant K_1 for these materials. The estimated g factors are in the expected range ($2.0 \leq g \leq 2.2$) typical for most ferromagnetic materials. The exchange constant A of Table I is plotted in Figure 1 as a function of the chemical composition x of Co_xNi_{1-x} microspheres. The vertical bars correspond to the fluctuation of A for each x, reported previously [22]. Apart from Ni and $Co_{50}Ni_{50}$ microspheres, all the rest exchange constants A are within the range of the previous tabulated values.

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REFERENCES

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[1] R. P. Blakemore and R. B. Frankel, Scientific American, December, 42 (1981).

[2] J. L. Kirshcvink and M. M. Walker, Science **269**, 1889 (1995).

[3] J. L. Kirschvink, Science **256**, May, 967 (1992).

[4] R. Langer, Science **249**, September, 1527 (1990).

[5] R. E. Rosensweig, *Ferrohydrodynamics* (Cambridge University Press, 1985).

[6] K.. Raj and R. Moskowitz, J. Mag. Mag. Mat. **85**, 233 (1990).

[7] W. Andrä and H. Nowak (eds.) *Magnetism in Medicine* (Wiley-VCH, 1998).

[8] W. F. Brown Jr, *Micromagnetics* (John Wiley and Sons, 1963).

[9] A. Aharoni, *Introduction to the Theory of Ferromagnetism* (Oxford Univ. Press, 1996).

[10] L. R. Walker, Phys. Rev. **105**, 390 (1959).

[11] G. Viau, F. Ravel, O. Acher, F. Fiévet-Vincent, and F. Fiévet, J. Appl. Phys. **76**, 6570 (1994).

[12] G. Viau, F. Ravel, O. Acher, F. Fiévet-Vincent, F. Fiévet, J. Mag. Mag. Mat. **140-144**, 377 (1995).

[13] G. Viau, F. Fiévet-Vincent, F. Fiévet, T. Toneguzzo, F. Ravel and O. Acher, J. Appl. Phys. **81**, 2749 (1997).

[14] P. Toneguzzo, O. Acher, G. Viau, F. Fiévet-Vincent and F. Fiévet, J. Appl. Phys. **81**, 5546 (1997).

[15] P. Toneguzzo, Thèse, Université Paris 7-Denis Diderot, Paris (1998).

[16] A. Aharoni, J. Appl. Phys. **69**, 7762 (1991).

[17] A. Aharoni, J. Appl. Phys. **81**, 830 (1997).

[18] V. P. Shilov, J. C. Bacri, F. Gazeau, F. Gendron, R. Perzynski and Yu. L. Raikher, J. Appl. Phys. **85**, 6642 (1999).

[19] P. A. Voltairas, C. V. Massalas and I. E. Lagaris, Int. J. Engng. Sci. **31**, 1391 (1993).

[20] P. A. Voltairas and C. V. Massalas, J. Mag. Mag. Mat. **124**, 20 (1993).

[21] P. A. Voltairas, D. I. Fotiadis and C. V. Massalas, J. Appl. Phys. (submitted).

[22] *Landolt-Börnstein, Series III/19a, Magnetic Properties of Metals* (Springer, Berlin 1986).

FIGURES

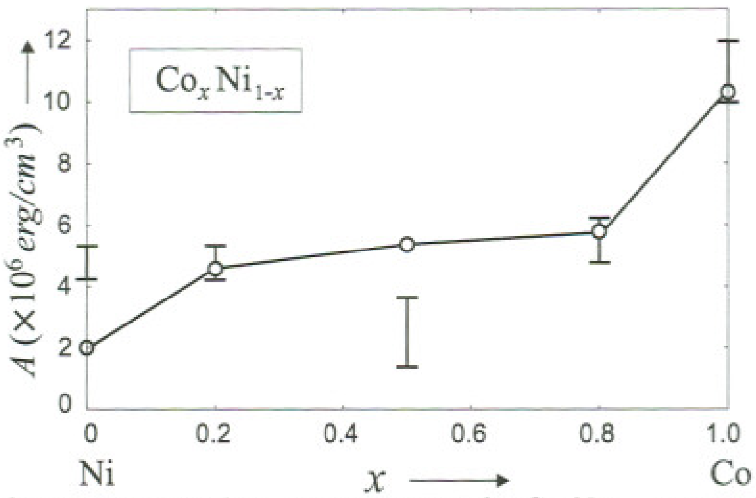

FIG. 1. The exchange constant A vs. composition x for Co_xNi_{1-x} microspheres. The vertical bars correspond to thin film measurements [22].

TABLES

TABLE I. Material parameters and experimental (f^{exp}) and computed ($f^{(k,n)}$) resonance frequencies for Co_xNi_{1-x} microspheres.

Materials	$4\pi M_s\,(G)$	$K_1\,(erg/cm^3)$	$A\,(\times 10^{-7} erg/cm)$	g	$2R\,(\mu m)$	Mode (k,n)	$f^{exp}\,(GHz)$	$f^{(k,n)}\,(GHz)$
Ni	508.8	-4.26×10^4	2.0	2.0	1.4	(24,48)	1.6000	1.6019
$Co_{20}Ni_{80}$	763.2	0	4.6	2.2	1.5	(28,56)	1.4000	1.3965
$Co_{50}Ni_{50}$	934.5	-1.5×10^5	5.4	2.2	2.0	(26,52)	3.0000	2.9944
$Co_{80}Ni_{20}$	1201.5	-4.8×10^6	5.8	2.1	2.0	(34,68)	4.0000	4.0258
Co	1445.0	5.2×10^6	10.3	2.2	2.0	(36,72)	6.5000	6.4293