COMPOSITION DEPENDENCES OF FMR PARAMETERS IN GRANULAR $(Fe_{20}Ni_{80})_x Ag_{1-x}$ FILMS

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UDC 536.42;538.22;539.216
‰ 2001

FMR study was carried out for the system $(Fe_{20}Ni_{80})_x Ag_{1-x}$ in a wide range of the magnetic component concentration $0.079 < x < 0.85$. The experimental data for low x values are described on the basis of a simple model of the grain system. In this model, it is assumed that separate magnetic grains have some uniaxial anisotropy (or the shape of uniaxial ellipsoids), and they do not touch each other but are connected through the dipole-dipole interaction.

Introduction

In recent years, there was a great interest in granular magnets, in which ferromagnetic particles are distributed in a diamagnetic matrix. This interest is primarily due to the interesting technical applications of these materials as magnetosensitive elements. In [1] and [2 – 6], granular systems permalloy-silver and Co_xCu_{1-x} were analyzed by the FMR technique. The authors of [3, 4] observed well-defined FMR lines of a quite low width and made the conclusion about a strong interaction between grains. As a result, such a sample can be described as a one-layer magnetic thin film. The main attention of the above work was paid to the samples with the magnetic component concentration of about 10 – 20%. On the other hand, magneto-resistance measurements [7] showed the maximum of magneto-resistance at $x \approx 0.25$. To understand the properties of these materials, we carried out the FMR study for the system permalloy-silver in a wide range of the magnetic component concentration. We choose this system because the dissolubility of Fe-based alloys in silver is extremely low. At the same time, the magnetic anisotropy of $Fe_{20}Ni_{80}$ is quity small and, therefore, it is possible to observe the pure effect of space inhomogeneity (inhomogeneity in shapes and sizes of magnetic grains and in their distrubution).

Experimental Details

$(Fe_{20}Ni_{80})$ – Ag grain films (with the continuous row of composition in the range $0.07 < x < 0.9$) were prepared by co-evaporation of the previously prepared permalloy and silver, by means of highly stabilized electron beam guns in the oilless vacuum of 10^{-4} Pa. Films of approximately 200 nm in thickness were deposited at room temperature on the line of mica substrates (of the size of 48×60 mm each). The total length of the line was 450 mm. Films were deposited in such a manner that the entire set of compositions with a gradual change of concentration was prepared in the same run. The rate of deposition of each component in the central area of the substrate row was controlled by means of calibrated quartz sensors. The control films made of pure metals were prepared during deposition. The composition of grain films was determined by measuring the thickness of control films with an optical microinterferometer. The main advantage of this technique over the other is that the whole composition line of the films was prepared under the same conditions, excluding the undesirable influence of other factors on the process of their formation. For measurements of the structure and magnetoresistance, the mica substrates were cut into small stripes. Within one stripe, the film composition was practically constant. We used 3×4 mm samples for FMR measurments. FMR spectra were measured by means of a standard X-band radiospectrometer using a frequency of 9.46 GHz. The modulation of the magnetic field was done with a frequency of 100 kHz. Measurements were carried out at room temperature with two orientations of the magnetic field: in parallel and perpendicularly to the film plane.

Experimental Results

FMR spectra show well-defined lines for all the concentrations of FeNi in the range $0.008 < x < 0.48$. The intensity, shape, and position of the FMR lines

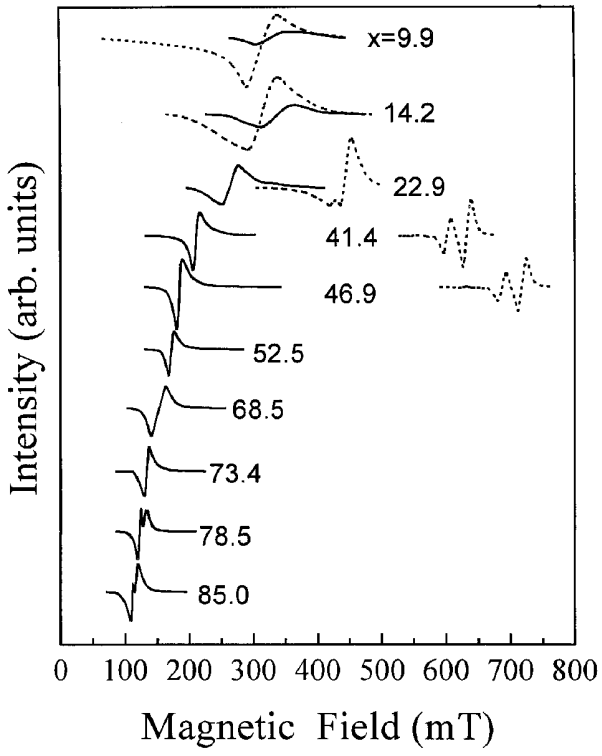

Fig. 1. FMR spectra for $(FeNi)_x Ag_{1-x}$ films in the entire x range for the parallel (continuous line) and perpendicular (dotted line) orientation of a sample in the external magnetic field

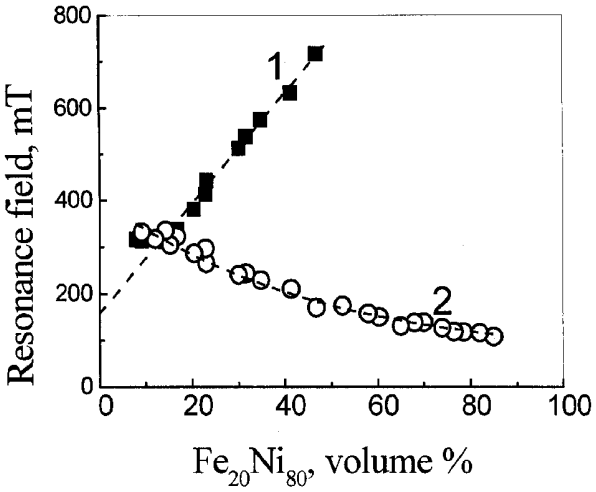

Fig. 2. Compositionalm dependence of resonance fields for parallel (open circles, line *2*) and perpendicular (solid squares, line *1*) orientations of a film with respect to the external magnetic field. Dotted lines correspond to Eqs. (6) and (7)

strongly depend on x (Fig. 1). One can schematically mark three ranges with different characteristics of FMR spectra.

At low concentrations of permalloy $x \approx 0.10 \div 0.15$, the line intensity for the perpendicular orientation of a film in the magnetic field is much lower than that for the parallel one. In this range, the resonance fields $H_{||}$ and $H_{\perp}$ practically do not depend on the concentration and coincide with each other within the limits of experimantal accuracy. The widths of lines are about 50 mT.

At high concentration ($x > 0.30$), one can observe a picture which is typical of FMR in continuous films. $H_{||}$ and $H_{\perp}$ lines are separated, their widths are small, not higher than 20 mT. The resonanse field $H_{\perp}$ has practically a linear dependence on the composition x (Fig. 2), and the dependence of $H_{||}$ on x weaker.

The FMR signal in the intermediate range $0.15 < x < 0.30$ is noticeably different from that at low and high concentrations. When the transition to this range occurs from the side of low concentrations, the line intensity grows rapidly and the dependence of resonance fields $H_{||}$, $H_{\perp}$ on x appears. Particularly, one can see the well-defined kink in the $H_{\perp}(x)$ dependence. The FMR lines are much wider than in case of high concentrations, their shape is different from the classical one.

All that indicated a substantial change in the properties of $(FeNi)_x Ag_{1-x}$ films at different concentrations of the magnetic component. The properties of this system change especially strong in the range of low and intermediate concentrations, where the signal outlook differs from the one typical of continuous films. It is known that a well-defined granular structure of material is realized in this range: the magnetic component forms small clasters which do not touch one another. On the other hand, we could not observe any considerable changes of the $H_{||}$ and $H_{\perp}$ dependences on x on the way from intermediate to high concentrations.

Theory

For the theoretical description of FMR spectra for the $(FeNi)_x Ag_{1-x}$ system in a wide range of concentrations, it is important to clarify two basic points: (i) to understand the behavior of the system at low concentrations where the granular structure is realized and (ii) to explain the transition from the granular system ($x < 0.30$) to a practically continuous film at high concentrations. Describing the film in the granular state, we assume that the magnetic phase forms separate clusters inside the nonmagnetic matrix, which do not touch each other. Since the grain size is much smaller than the single-domain size, we assume that atomic spins inside a grain are parallel to each other and form the total magnetic moment $\mathbf{m}_0$ of the grain. If the number of atoms in the grain n is large

enough (according to [8] $n \approx 10^3$), we can take that for the most part of atoms in the grain (except the surface layer), the character of interaction being the same as for the bulk sample. Then, at $T < T_{C,\infty}$, where $T_{C,\infty}$ is the Curi temperature of the bulk sample, we can assume that atomic spins in the grain are ferromagnetically ordered and the magnetic moment of the grain can be determined as $|\mathbf{m}_0| = M_s V$, where V is the grain volume and M_s is the saturation magnetization. We suppose that the energy of the i-the granule at an extermal magnetic field H_e can be written as:

$$w_i = \frac{1}{2}\frac{\beta m_{zi}^2}{V} - m_i H_e, \qquad (1)$$

where β is some phenomenological parameter connected with magnetic anisotropy of the granule z. This uniaxial anisotropy, which is normally directed to the film plane, can also appear as a result of stress in the system film – substrate, while preparing. In the first case, if one assumes that the shape of granules is a uniaxial ellipsoid with the main axis along z, then $\beta = 4\pi(N_z - N_\perp)$, where N_z and $N_\perp$ are the demagnetizing factors of the ellipsoid along z and perpendicular to it. Further, we assume that the parameter β is the same for all granules. If to combine this parameter with the shape anisotropy, it should mean that the level of deviation of the shape of granules from the spherical one is approximately the same, and the directions of ellipsoid axes are the same for all the granules.

The energy of the system of N particles consists of the sum of the separate energies (1) and their interaction energy. It is clear that the magnetic dipole interaction, which slowly decreases with distance [9], should be the most substantial (since the indirect RKKY exchange interaction [10] is a quickly oscillating function of distance).

The dynamical equation for the magnetic moment $\mathbf{m}_i$ is determined by the total energy of the system $W = W(m_1,...,m_N)$ and can be written as [9]

$$\frac{d\mathbf{m}_i}{dt} = \gamma\left[\mathbf{m}_i, \frac{dw}{dm_i}\right], \qquad \gamma = g\mu_B/\hbar, \qquad (2)$$

where g is the gyromagnetic ratio, μ_B is the Bohr magneton, and $\hbar$ is the Planck constant. Taking into account the formula for the energy of a separate grain (1) and the well-known expression for the energy of magnetic dipole interaction [9], Eq. (2) can be presented in the following form:

$$\frac{d\mathbf{m}_i}{dt} = \gamma[\mathbf{m}_i x (\mathbf{H}_e - \beta M_{iz}\mathbf{e}_z + \mathbf{H}_m)], \qquad (3)$$

where $\mathbf{M}_i = \mathbf{m}_i/\Delta V$ is the magnetization of the i-th grain and $|\mathbf{M}_i| = M_s$. The value of $\mathbf{H}_{mi}$ determines the field of magnetic interaction on the i-th grain,

$$\mathbf{H}_{mi} = \Sigma (r_{ij})^{-3}(3(\mathbf{n}_{ij}\mathbf{m}_j)\mathbf{n}_{ij} - \mathbf{m}_j), \qquad (4)$$

where $r_{ij} = |\mathbf{r}_{ij}|$, $\mathbf{n}_{ij} = \mathbf{r}_{ij}/r_{ij}$, $\mathbf{r}_{ij}$ is the vector connecting the i-th and j-th grains. It is easy to calculate the dipole sum (4) in the case where all the $\mathbf{m}_i$ are parallel, in analogy with the standard macroscopic approximation. Assuming that the grain sizes and average distances between them are small comparing to the film thickness, it is possible to express the value of $\mathbf{H}_{mi}$ through the magnetization of the sample $\tilde{\mathbf{M}}$, $\mathbf{H}_{mi} = -4\pi\tilde{M}_z\mathbf{e}_z$. In this case, $\tilde{\mathbf{M}} = x\mathbf{M}$, $\mathbf{M}$ is the magnetization inside the grain, therefore, we can write for the parallel orientation of magnetic moments:

$$\mathbf{H}_{mi} = -4\pi x M_z \mathbf{e}_z.$$

In view of the condition $\mathbf{m}_i \sim \mathbf{M}$, Eq.(3) can be reduced to the standard equation for the magnetic moment dynamics,

$$\frac{d\mathbf{M}}{dt} = \gamma[\mathbf{M} \times \mathbf{H}_{\text{eff}}(M)],$$

with the following simple expression for the effective field H_{eff}:

$$\mathbf{H}_{\text{eff}} = \mathbf{H}_e - (4\pi x + \beta) M_z \mathbf{e}_z. \qquad (5)$$

The assumption about the parallel orientation of all the $\mathbf{m}_i$ is a very strong simplification of the problem. It is known that granular magnetic systems exhibit superparamagnetic properties at low concentrations, which contradicts *a priori* the above supposition. On the other hand, all the $\mathbf{m}_i$ will be parallel in the presence of a sufficiently strong magnetic field $\mathbf{H}_e$. We shall discuss effects of nonparallelism of $\mathbf{m}_i$ later. Now let us return to the analysis of the case where all the $\mathbf{m}_i$ are parallel. It is natural that then one obtains formulas for the eigenmodes of the system which are similar to the standard Kittel's formulas for FMR modes. Particularly, for a parallel slab, we obtained the following expressions for the modes $\omega_{||}$ and $\omega_\perp$, when the magnetic field is parallel ($H = H_{||}$) and perpendicular ($H = H_\perp$) to the film plane, respectively:

$$\omega_{||} = \gamma\sqrt{H_{||}[H_{||} + (4\pi x + \beta) M_s]}, \qquad (6)$$

$$\omega_\perp = \gamma[H_\perp - (4\pi x + \beta) M_s]. \qquad (7)$$

Here, as previously, M_s is the magnetization of magnetic component of the material (inside a grain).

Formulas 96) and (7) are obtained for the diluted system, assuming the parallel orientation of grains. Now let us examine the 'ferromagnetic" limit, when the concentration of magnetic component is high, grains are touching each other, and the exchange interaction forms the total magnetization of a sample, $M = x M_s$. In this case, we can use formulas (6) and (7) again, considering the parameter β as the anisotropy constant. It is apparently expedient to process the experimental data on the basis of these formulas, considering the parameter β as a phenomenological constant.

The results of fitting the experimental data in the parameters g, M_s, β by formulas (6), (7) for the range $x > 0.20$ are shown in Fig. 2. The values of M_s, extracted from the curves for $H_{||}$ and $H_{\perp}$ ($4\pi M_s \approx 1.18$ T and $4\pi M_s \approx 1.10$ T, respectively) are in good agreement with each other and with the value $(4\pi M_s)_{\mathrm{FeNi}} = 0.97$T for the bulk sample of permalloy, which was used for the preparation of films. The value of $g = 2.1$ agrees with one known from the literature [5]. The value of the effective anisotropy field for a single grain $H_a = -\beta M_s$ appears to be equal to 140 mT for the $H_{||}(x)$ dependence and 160 mT for the $H_{\perp}(x)$ curve, or $\beta \approx -0.129$, $\beta \approx -0.136$, respectively. Therefore, values of the parameter β match each other within the experimental accuracy limits. It is difficult to connect the estimated values of β with the shape anisotropy of a sample as, in this case, the small quantity, $(N_z - N_{\perp}) \approx \approx -0.13/4\pi \approx -0.01$, should be the same for all the granules. It is difficult to imagine the reason for such a small, but same for all the granules, deviation of their shape from the spherical one. That is why we suppose that the β origin is a result of magnetic anisotropy, which is the same for all the granules. The shape of granules is supposed to be approximately spherical, that well correlates with data [12].

It follows from these data that, in the range of intermediate and high concentrations ($x > 0.20$), the resonance data can be described by the same formulas with regard of the effective uniaxial anisotropy with the easy axis perpendicular to the film surface.

As far as the range of low concentrations ($x < 0.15$) is concerned, the situation is principally different, see Fig. 2. It can be seen definitely, particularly from the curve $H_{\perp}(x)$, that the resonance field does not depend on concentration in this range. The resonance frequency with a good accuracy coincides in this range with its value for the isotropic case $\omega = g\gamma H$. So, both the shape anisotropy and effective anisotropy of grains do not appear in this range.

This behaviour is naturally connected to the fact that magnetic moments of grains are not saturated at low concentrations of permalloy and formulas (6), (7) cannot be used. To describe the resonance in a nonsaturated system, it is necessary to use the total system of coupled equations (3), (4) for variables $\mathbf{m}_i$. Moreover, the theory should include thermal fluctuations of granular magnetic moments, which determine the behaviour of the system in the superparamagnetic state. We are not able to achieve a consistent solution of this problem, and thus we restrict this discussion to the qualitative analysis.

Let us return to the general formula (4). if the magnetic moments grains are completely correlated, then, according to (5), the value of $\mathbf{H}_m$ is reduced to the z-component of the sample magnetization. It is clear that such a result can be obtained also in the other limit of small correlations. Indeed, if the deviations of granular magnetic moments from the average value $\langle \mathbf{m}_i \rangle$ (which is parallel to the average magnetization $\langle \tilde{\mathbf{M}} \rangle$) are totally chaotic, then magnetic poles emerging on the side surfaces of grains compensate each other at sufficiently small distances. In this case, we again obtain the expression like (5), which, however, includes the average value of the z-projection of the grain magnetization:

$$\mathbf{H}_{mi} = -4\pi \langle m_z \rangle (x/V)\, \mathbf{e}_z.$$

The value of $\langle m_z \rangle$ in an unsaturated field is determined by the grain susceptibility and can be significantly lower at not very strong fields than the nominal value $M_0 V$. Thus, the transition from the saturated to superparamagnetic state occurs while changing the system parameters, which should lead to 'switching off" the shape anisotropy and to a cusp in the dependence of the resonance frequency on the external field. Hence, one should change M_z to a much lower value $\langle m_z \rangle / V$ in formula (5) for the effective field. Then both $\omega_{||}$ and $\omega_{\perp}$ tend at $x << x_c$ to the value γH characteristic of an isotropic sample. This explains the transition from the range of intermediate concentrations to low ones in Fig. 2. According to this explanation, $x_c \approx 0.15$, which is much lower than the value characteristic of the percolation threshold.

The RKKY-interaction could be important for the ground state properties, but it affects no frequencies of a homogeneous magnetic resonance.

Note that the lines, that fit the dependences of $H_{||}$ and $H_{\perp}$ according to formulas (6) and (7) in Fig. 2, intersect near the value $x = x_c$. It means that $4\pi x_c + \beta$ is close to zero. This can be the coincidence of parameter values, as there is no interconnection of β and x_c in the presented qualitative ative model. Nevertheless, perhaps, a more strict and detailed

model could account the composition dependence of the parameter, which should be presented in (6) and (7) instead of M_s in the transition range from superparamagnetic granules to the state with saturated magnetization. The detailed description of this point is out of this paper.

One might think that the transition from the ordered to superparamagnetic state could be connected with a change in the external field, at least for the transverse field orientation. The analysis shows, however, that the internal field in a grain, $H_i = H_e - 4\pi x M_s$, does not depend in the saturated state region on the concentration and is close to the value of $\omega_0/\gamma \approx 320$ mT. Taking this fact into account, the behaviour of the resonance modes within the range of small x can be explained on the sole base of the assumption that the grain sizes depend on x. Such a behaviour of the particle size was experimentally established with the help of the electron microphotography technique for the granular system of the Cd/Ti and Tb/Ti type with nanometer scale particles [11]. With lowering x, the grain size decreases, and the superparamagnetic behaviour becomes dominant at certain value of $x = x_c$.

Note, that the model was built and calculations were carried out assuming that the granules of Py in Ag matrix have approximately the same size. It was shown experimentally for granular FeNi – Ag films with small x [1, 12].

We acknowledge to S.Ryabchenko for the fruitful discussion and important notes. This work was partly supported by project No. 1086 from the Science and Technology Center of Ukraine (STCU).

1. *Dieny B., Teixeira S.R., Rodmacq B. et a*l. // J. Magn. Magn. Mater. **130**, 197 (1994).
2. *Rubinstein M., Tejada J., Zhang X.X.* // J. Appl. Phys. **75,** 6557 (1994).
3. *Krishnan R., Lassri H., Seddat M. et al.* // Ibid. 6607 (1994).
4. *Hideki Watanabe, Iwao Ishida, Kouichi Hamada et al.* // J. Phys. Soc. Jap. **65,** 3680 (1996).
5. *Seshu Bai, Bhagat S.M., Krishnan R., Seddat M.* // J. Magn. Magn. Mater. **147,** 97 (1995).
6. *Khanana S.N., Linderoth S.* // Phys. Rev. Lett., **67,** 742 (1991).
7. *Pohorilyi A.N., Kravez A.F., Shipil E.V., Vovk A.Ya.* // Proc. 15th Intern. Colloquium on Magnetic Films and Surfaces, Queensland, Australia 1997, Pd-3.
8. *Peng Chubing, Chen Haiying, Li Guozhong, Dai Daosheng* // J. Appl. Phys. **76**, 7102 (1994).
9. *Akhiezer A.I., Bar'yakhtar V.G., Peletminskii S.V.* Spin waves. – Amsterdam: North-Holland, 1968.
10. *Vargas P., Altbir D.* // J. Magn. Magn. Mater. **167**, 161 (1997).
11. *O'Shea M.J.* // J. Phys. Cond. Matter (1998).
12. *Badia E., Battle X., Labarta A. et al.* // J. Appl. Phys. **82,** 677 (1997).

Received 24.03.99,
revised version – 07.06.01