FERROMAGNETIC RESONANCE IN BULKY γ-Fe—NI ALLOYS

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Results are presented of a ferromagnetic resonance investigation of γ-ferronickel alloys containing from 29 to 85 at.% Ni. The investigation was carried out at room temperature at frequencies of 2210 and 6375 MHz in tempered and annealed alloys. The resonant field strengths are minimum at 40—45 at.% Ni and rapidly increase in Invars on decrease of the amount of nickel. Thermal treatment strongly affects the resonant fields. The resonance line width as a function of nickel concentration and thermal treatment was also studied. An anomalous growth of the line width in the Invar region of the composition is observed. Various relaxation contributions to the experimental line width are estimated. Resonance line broadening due to Landau-Lifshitz damping is separated and the relaxation frequency λ is estimated. The dependence of λ on concentration and thermal treatment which is observed is ascribed to the effect of magnetic ordering on λ. The Bloch-Bloembergen relaxation frequencies in the alloys are presented.

INTRODUCTION

THE ferromagnetic-resonance (FMR) method for the investigation of 3-dimensional metallic ferromagnets came into use relatively recently, owing to both experimental and theoretical difficulties. In the former case, the difficulty was that the absorption signal in ferromagnetic metals is exceedingly small. The development of spectrometers or of installations with large signal/noise ratios has made it possible to start such investigations. The second difficulty, which persists to this day, is due to the uncertainty in the microphysical nature of the line width in bulky metallic ferromagnets.

The connection between the frequency of the experiment and the magnetic-resonance field H_r, at which FMR is observed, and also the possible broadening of the resonance line in ferromagnetic metals, is obtained by solving the equation of motion of the magnetization vector M in the form

$$\frac{1}{\gamma}\frac{d\mathbf{M}}{dt}=[\mathbf{MH}]-\frac{\lambda}{\gamma^2 M_s^2}[\mathbf{M}[\mathbf{MH}]]+\frac{2A}{M^2}[\mathbf{M}\nabla^2\mathbf{M}] \quad (1)^*$$

simultaneously with the corresponding Maxwell equations[1]; here $\gamma = g|e|/2mc$, A is the exchange parameter, and λ is the Lifshitz relaxation frequency. In the case when the effective anisotropic fields can be neglected, the connection between the frequency λ and the resonance field H_r takes the form

$$\omega=\gamma[H_r(H_r+4\pi M_s)]^{1/2}. \quad (2)$$

The possible relaxation processes that act in ferromagnetic metals in FMR and govern the resonance line width are represented in (1) by the second and third terms on the right. The second term describes the microphysical relaxation in a ferromagnetic metal, phenomenologically represented by the Landau-Lifshitz relaxation frequency. This type of relaxation broadens the FMR line by ΔH_λ. The third term describes the so called "exchange-induced" broadening of the resonance line $\Delta H_{A-\sigma}$, which results from the skin effect in a metallic ferromagnet. The total FMR line width in single crystals is the sum

$$\Delta H=\Delta H_\lambda+\Delta H_{A-\sigma}, \quad (3)$$

where $\Delta H_\lambda = 1.45\lambda\omega/\gamma^2 M_s$[2], $\Delta H_{A-\sigma} = 16\pi/3c(A$ $= 16\pi/3c(A\sigma\omega)^{1/2}$[3], σ is the conductivity, M_s the saturation magnetization, and c the speed of light.

In polycrystals, an additional FMR line broadening, ΔH_{anis}, can occur as a result of the scatter in the values of the resonance fields over the individual crystallites, and $\Delta H_{anis} \leq 2K/M_s$, where K is the anisotropy constant.

At present there are very few published experimental papers on FMR in bulky metallic ferromagnets. Most investigations were performed on nickel, iron, and certain alloys, and the relaxation frequency was investigated sufficiently well only for pure nickel and iron. Thus, it is shown in[2,4,5] that the observed FMR line width in bulky metallic iron and nickel samples is satisfactorily described by Eq. (3). At high frequencies, the main contribution to the observed line width is made by the relaxation mechanism. The following estimates of the relaxation frequency at room temperature were obtained: $\lambda_{Ni} = 2.2 \times 10^8$ sec^{-1}[2,4] and $\lambda_{Fe} = 0.7 \times 10^8$ sec^{-1}[5]. The temperature dependence of the line width and the relaxation frequency in iron were investigated in[5]. It was found that the line width near the Curie temperature increases sharply, and the relaxation frequency changes from 0.7×10^8 sec^{-1} to 7×10^8 sec^{-1} with a maximum at ~ 550°C.

We report in this paper experimental results on FMR in γ-iron-nickel polycrystals, analyze the results on the basis of the phenomenological theory of ferromagnetic resonance in bulky ferromagnetic alloys, estimate the relaxation frequency in accordance with Landau-Lifshitz and Bloch-Bloemberger, and consider the influence of magnetic ordering on the relaxation frequency.

2. MEASUREMENT METHODS. SAMPLES

The measurements were performed with an FMR spectrometer with amplitude modulation. The high signal/noise ratio, larger than 20, was obtained by using narrow-band high-frequency amplification (with a

*[MH] ≡ M × H.

Comp., at. % Ni	g-factor (±0.01)		$4\pi M_s \pm 1.5\%$, G		Comp., at. % Ni	g-factor (±0.01)		$4\pi M_s \pm 1.5\%$, G	
	anneal	quench	anneal	quench		anneal	quench	anneal	quench
83.2	2.14	2.14	10000	9600	45.2	2.10	2.10	16200	16000
81.0	2.13	2.13	10150	9720	41.5	2.09	2.09	16200	15500
79.1	2.11	2.12	10650	10200	40.3	2.10	2.10	15950	15200
74.9	2.10	2.11	11850	11300	36.5	2.10	2.10	14950	14000
68.7	2.10	2.10	13900	13300	35.1	2.09	2.09	14000	12800
63.1	2.10	2.10	15300	14400	34.1	2.09	2.09	12900	11800
55.8	2.10	2.10	15900	15600	32.4	2.07	2.07	11100	9400
50.0	2.10	2.10	15950	15705	30.3	2.06	2.07	6000	5100
					29.0	2.06	2.06	1540	1160

pass band of several MHz), low-frequency amplitude modulation at 380 Hz with narrow-band low-frequency amplification (pass band 1–2 Hz), and synchronous detection. The output instrument was an automatic recorder that plotted the derivative of the absorption signal as a function of the external magnetic field at a fixed frequency. The magnetic field was calibrated with an MMR field meter (IMI-2). The resonant field was determined with accuracy not worse than 1%, and the accuracy of the line width, measured by determining the distance between the peaks of the absorption derivative, was not worse than 2%. A wire sample, electrically polished prior to the measurement, was placed either in a waveguide cavity (H_{012}) or in a strip cavity (depending on the frequency of the experiment), in such a way that the signal was detected from a section of 4 mm length at the center of a sample 40 mm long. This was necessary to exclude the influence of the inhomogeneities of the magnetization over the length of the sample on the FMR parameters. The magnetic field was applied along the sample axis.

The investigated alloys were smelted from electrolytically pure nickel and iron in a vacuum furnace. The samples were wires of 0.5 mm diameter (0.4 mm after polishing). The heat treatment was as follows: 1) heating in hydrogen to 1100°C, soaking for 5 hours, cooling at a rate of 250 deg/min in a hydrogen jet; 2) heating in hydrogen to 1100°C, soaking 5 hours, cooling at a rate of 250 deg/h to 600°C, cooling from 600°C to 500°C in three hours, and from 500°C to 400°C in 50 hours. There were five samples for each state and for each composition. The scatter and the values of the resonant fields for samples of one composition and one type of heat treatment did not exceed 1–1.5%. The analogous scatter in the values of the line widths was 6–8%.

The measurements were performed at two frequencies, 2210 and 6375 MHz at room temperature. The investigated compositions are given in the table.

3. MEASUREMENT RESULTS AND THEIR DISCUSSION

1. Figure 1 shows the signals of the absorption derivative for samples with different contents of nickel and for different heat treatments. For samples with high nickel contents, the signal of the absorption derivative had a symmetrical shape. At 29–31 at.% nickel, the derivative signal becomes weak, and its shape asymmetrical. To observe the resonance signal in samples with 29 at.% Ni it was necessary to increase the gain appreciably.

A similar transformation of the absorption derivative signal was observed in single-crystal nickel wires near the Curie temperature[2]. An asymmetry of the derivative signal is observed also in the FMR signal at very high frequencies, $\gtrsim 50$ GHz[6]. It follows from these measurements that the resonance signal becomes asymmetrical at $H_r \geq 4\pi M_s$. A similar situation obtains also in our case for low-nickel alloys, since the Curie temperature of these alloys is close to room temperature[7], and the saturation magnetization is such (see the table) that $H_r \geq 4\pi M_s$. The possible cause of the asymmetry of the resonance line at $H_r \geq 4\pi M_s$ was considered in[2].

2. Figure 2 shows the experimental values of the resonance fields as functions of the nickel content in the samples at 6375 MHz. A similar curve was obtained for 2210 MHz. As seen from the figure, this dependence has a gently sloping minimum at nickel concentrations 40–50 at.%, whereas in the invar region one observes an anomalous growth of the resonance fields. A similar growth of the resonance field was observed near the Curie temperature in nickel[2]. Obviously, the rapid growth of the resonance fields in our case is of the same nature as in the case of nickel, since the change of the concentration, as well as of the temperature, brings the sample closer to the Curie

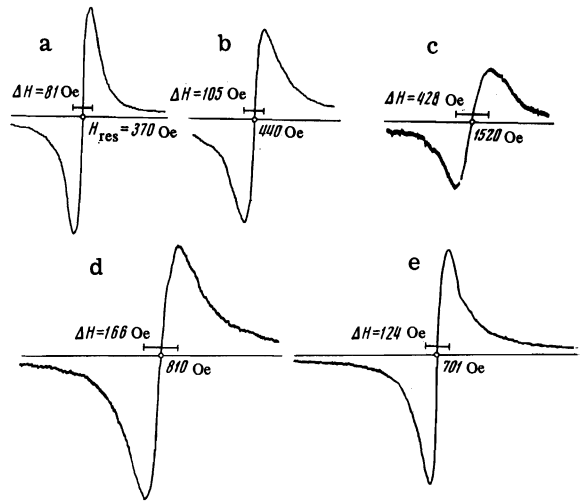

FIG. 1. Absorption derivative signals as functions of the nickel content in the samples and of the heat treatment (the times during which the signal was recorded differ): a–74.9 at.% Ni, annealing; b–32.4 at.% Ni, annealing; c–29.0 at.% Ni, annealing; d–30.3 at.% Ni, quenching; e–30.3 at.% Ni, annealing.

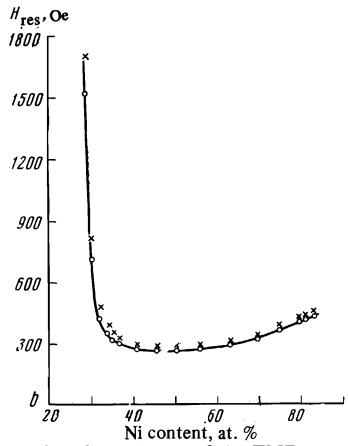

FIG. 2. Concentration dependence of the FMR resonance fields–X–quenching, O–prolonged annealing, ω = 6375 MHz.

point in the case of nickel. It is seen from Fig. 2 and from the table that the behavior of the resonance field correlates with the behavior of the saturation magnetization in accordance with formula (2). It is also seen from the figure that the values of the resonance fields for annealed alloys are less than for quenched alloys, and in the invar region of compositions the influence of the heat treatment on the resonance field is large. The influence of the heat treatment on the saturation magnetization, and consequently on the resonance fields for invar alloys, can be explained by means of the results of[8,9].

From the resonance fields obtained at 6375 and 2210 MHz after quenching and annealing, we calculated the saturation induction $4\pi M_S$ and the g-factor (see the table). The difference between the saturation inductions of quenched and annealed states for one and the same composition increases with decreasing nickel concentration (below 36 at.% Ni). We note that the g factor is practically independent of the heat treatment and varies with composition in accordance with the data of[10,11].

3. The dependence of the line width in γ-iron-nickel alloys on the composition is shown in Fig. 3 for 6375 MHz. We see that the line width has a maximum at 40—45 at.% Ni and a steep rise in the region of invar compositions. The maximum corresponds to the maximum saturation magnetization of this series of alloys. The steep rise of the line width for invar compositions can be attributed to a decrease of the Curie temperature of these alloys with increasing iron content. A fast rise of the resonance line width was also observed in single crystals of nickel and iron in the region of the Curie temperature[2,5].

It is seen from Fig. 3 that the heat treatment influences the FMR line particularly strongly in the invar region of compositions. Thus, e.g., for the sample with 30.3 at.% Ni, the line width of the quenched alloy exceeds by almost 50% the line width of the annealed alloy, and the difference between the line widths for the quenched and annealed samples increases with decreasing nickel content, starting with 36 at.% Ni. It is seen from the table that this effect correlates with the behavior of the magnetization difference between the annealed and quenched states of the samples.

4. Let us examine the contributions made to the observed FMR line width by the different mechanisms considered in the introduction.

The upper limit of the possible resonance line broadening, due to the polycrystalline state, is shown for annealed samples in Fig. 3 in the form of a dashed line. The anisotropy constants were taken from[12]. The relative contribution of the line broadening ΔH_{anis} to the total line width at 6375 MHz does not exceed 10—15%, and there is a region of compositions where $\Delta H_{anis} \approx 0$.

The contribution of the "exchange-induced" broadening mechanism $\Delta H_{A-\sigma}$, due to the finite depth of penetration of the microwave energy into the samples, has been estimated from formula (3) and is shown in Fig. 3 by a dash-dot line. The exchange parameter A is taken from the data on spin-wave resonance[13]. In alloys containing less than 37 at.% Ni, the parameter A was not determined, and can be estimated from the formula[14]

$$A = DM_s / 2\gamma\hbar, \qquad (4)$$

where D is the parameter of the spin-wave dispersion relation ($E_{SW} = Dk^2$). From the data on the temperature dependence of the saturation magnetization (the 3/2 law)[15,16] and from neutron-diffraction data[17] it follows that D decreases in invars with decreasing amount of nickel. The contribution of the "exchange-induced" mechanism to the observed width is largest for the alloys with large nickel contents and amounts to about 35% at 6375 MHz (it is equal to 60—70% at 2210 MHz). With decreasing nickel contents, this form of resonance line broadening decreases, and amounts to several per cent for invars. Thus, in iron-nickel alloys at 6375 MHz, the broadening of the FMR line is due mainly to relaxation damping of the Landau-Lifshitz type.

5. We used formula (3) to calculate the relaxation frequency λ, using our results for 6375 MHz. The concentration dependence of the relaxation frequency is shown in Fig. 4. We see that the relaxation frequency varies considerably as a function of the nickel content, from $\sim 1.0 \times 10^8$ sec^{-1} to $\sim 7.5 \times 10^8$ sec^{-1}. The maximum value of the relaxation frequency occurs at a concentration 40—45 at.% Ni, and the minimum value at 75 at.% Ni. This result shows that the relaxation frequency is a function of the magnetic ordering. Indeed, the relaxation frequency has a lowest value in the region of 75 at.% Ni, where the alloys have atomic ordering. The ordered composition Ni_3Fe consists of four primitive cubic sublattices, of which 3 have neutral atoms with magnetic moment 0.6 μ_B at the sites and one has iron atoms with magnetic moments 2.8 μ_B. The atomic ordering is accompanied by formation of a unique magnetic ordering—magnetic sublattices (sublattice ordering). Quenching of these alloys has led to destruction of the Ni_3Fe superstructure and to almost doubling of the relaxation frequency. We note that the ordered Ni_3Fe structure has a magnetic saturation which is approximately 4% larger than the disordered structure. Consequently, the vanishing of the sublattice magnetic ordering leads to a growth of the relaxation frequency. It follows from neutron-diffraction data[18] that when the nickel content is decreased from 75 at.%, the degree of long-range order decreases linearly practically to zero at 45 at.% Ni, and a growth of the relaxation frequency is simultaneously observed. However, the sublattice ordering does not account for this entire growth. The existence of ordered regions in the disordered matrix can additionally increase the relaxation fre-

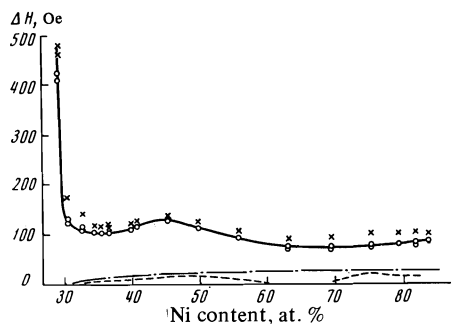

FIG. 3. Concentration dependence of the FMR line width. X—quencher, O—prolonged annealing, dashed—effective anisotropy field, dash-dot—"exchange-induced" mechanism, ω = 6375 MHz.

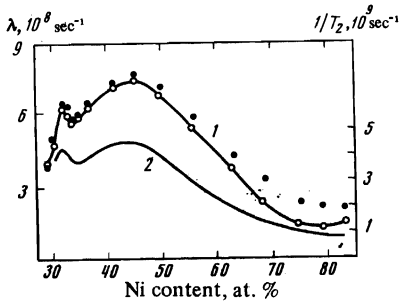

FIG. 4. Concentration dependences of the relaxation frequency after Landau-Lifshitz (curve 1) and relaxation frequency $1/T_2$ after Bloch-Bloembergen (curve 2).

quency. The agreement between the maxima of the saturation magnetization and of the relaxation frequency is not accidental and shows that they are interrelated.

In the invar region there occurs a destruction of another type of magnetic ordering, the orientation ordering that leads to a rapid decrease of the saturation magnetization with decreasing nickel content. Here, too, a rapid decrease of the Curie points is observed. This type of magnetic disorder causes the strongly pronounced maximum of the relaxation frequency at 32–33 at.% Ni. It is obvious that a relaxation-frequency maximum of the same type was observed in iron near the Curie temperature[5].

As shown by Mössbauer[19] and neutron-diffraction[8,20] data, invars are magnetically inhomogeneous. This circumstance is also responsible for the higher values of the relaxation frequency in this region of compositions, as compared with the region of high nickel contents. If we take an invar with the same magnetic saturation (~ 33 at.% Ni) as in Ni_3Fe, then the relaxation frequency in invar turns out to be larger by approximately 3 times. This confirms the foregoing.

6. Berk[21] obtained a formula for the Bloch-Bloembergen relaxation time T_2. Figure 4 shows the concentration dependence of the calculated value of $1/T_2$ for annealed alloys (curve 2). We see that a similar dependence of the relaxation frequency λ and of $1/T_2$ on the concentration is observed. It is interesting to note that, starting with ~ 30.5 at.% Ni, the value of $1/T_2$ increases sharply, and $1/T_2 \approx 1 \times 10^{10}$ sec^{-1} for the alloy with 29 at.% Ni (this is not shown in Fig. 4). This value of $1/T_2$ is comparable with the spin-spin relaxation in nickel at the Curie point, where $1/T_2 \approx 1.5 \times 10^{10}$ sec^{-1}[22]. This result favors the advanced assumption that the magnetic ordering exerts a strong influence on the relaxation frequency in iron-nickel alloys.

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