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# Temperature Dependence of the Ferromagnetic Resonance Linewidth in Thin Ni–Fe Films*

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Linewidths for ferromagnetic resonance with the static magnetic field in the film plane have been measured at frequencies from 1–9 Gc/sec and temperatures from 4.2°–300°K for Ni–Fe alloy films (77% Ni) 150 to 1500 Å thick. The linewidth exhibits a maximum in the vicinity of 80°K and the effect is generally larger in thinner films. The amplitude of the linewidth temperature dependence is independent of frequency and the maximum shifts to slightly higher temperatures with increasing frequency. The amplitude can be enhanced by heating the film at 150°C in air (oxidation) and removed by a similar treatment in a hydrogen atmosphere. This enhancement-removal cycle can be repeated reproducibly. The time required for saturation of the enhancement when the oxidation treatment was slowed down by annealing in a vacuum of $10^{-3}$ Torr instead of at atmospheric pressure was observed to be independent of film thickness, indicating that a volume-diffusion process is not involved. The apparent importance of surface oxidation suggests two possible mechanisms, valence exchange and exchange anisotropy, which may contribute to the linewidth temperature dependence. The discovery of a temperature-dependent linewidth in thin films which exhibits a maximum in the vicinity of 80°K and the identification of this dependence with a surface oxidation process are the primary results of this investigation.

Many investigations of the ferromagnetic resonance linewidth at room temperature have been reported which have attempted to characterize the losses associated with resonance. The present situation concerning resonance relaxation at room temperature has been summarized in a recent paper by Patton *et al.*[1] in conjunction with data on the thickness dependence of the room-temperature linewidth for resonance with the applied field in the plane of the film (parallel resonance). Very little work concerning the temperature dependence of the resonance linewidth in thin films has been reported. Such an investigation would be extremely useful in determining what relaxation mechanisms are important in thin films. The purpose of the present paper is to report the results of an investigation of the temperature dependence of the parallel-resonance linewidth $\Delta H$ for Ni–Fe alloy films. As a function of temperature, the linewidth exhibits a maximum in the vicinity of 80°K. The effect is generally larger in thinner films. The temperature dependence of $\Delta H$ resembles that predicted by the valence-exchange mechanism[2] and the slowly relaxing impurity mechanism.[3] An alternate explanation may be associated with exchange anisotropy.[4] Whatever the physical origin of this linewidth maximum, the discovery of a temperature-peak process associated with the resonance linewidth in thin Ni–Fe alloy films is surprising and interesting.

The nickel–iron alloy films used in this study were vacuum evaporated from a melt of 80% Ni and 20% Fe onto hot glass substrates at 300°C in a moderate vacuum of $10^{-6}$ Torr and in the presence of a uniform magnetic field in the plane of the substrate. Film composition was determined to be about 77% Ni using x-ray fluorescence techniques. Film thickness was determined magnetically, using a 20-cps hysteresis loop tracer, and optically, using the Tolansky multiple-beam interference technique. Linewidth measurements for parallel resonance were performed utilizing a nonresonant stripline section, field modulation, and phase-sensitive detection. The stripline was a conventional double-ground-plane system[5] designed to have an impedance of 50 Ω and provide a uniform microwave excitation field over the 1-cm-diam film sample mounted parallel to and against one ground plane. The modulation field amplitude was much less than the measured linewidths. High-power effects[6,7] were avoided by using very low microwave power levels. The static magnetic field for resonance was applied in the plane of the film and perpendicular to the microwave field using a Helmholtz coil pair. The phase-sensitive detector output was displayed as a function of applied static field using an $X$–$Y$ plotter and the linewidth $\Delta H$ was taken to be the field separation between the peaks of the absorption derivative.[8] The stripline assembly was contained in a conventional double-Dewar system and

* This work was done during the tenure of a National Aeronautics and Space Administration Graduate Fellowship.
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[1] C. E. Patton, C. H. Wilts, and F. B. Humphrey, J. Appl. Phys. **38**, 1358 (1967).
[2] W. A. Yager, J. K. Galt, and F. R. Merritt, Phys. Rev. **99**, 1203 (1955).
[3] R. W. Teale and K. Tweedale, Phys. Letters **1**, 298 (1962).
[4] W. H. Meiklejohn, J. Appl. Phys. **33**, 1328 (1962).
[5] Symposium on Microwave Strip Circuits, IRE Trans. Microwave Theory Tech. **MTT-3**, No. 2 (March 1955).
[6] J. B. Comly and T. Penney, Scientific Report No.8 (Series 2), Cruft Laboratory, Harvard Universiry, Cambridge, Massachusetts, December 1963.
[7] J. B. Comly, Scientific Report No. 5 (Series 3), Cruft Laboratory, Harvard University, Cambridge, Massachusetts, August 1965.
[8] Using the present experimental arrangement, the derivative of the absorption curve is obtained as a function of the applied static field. The values of the linewidth reported here can be converted to the usual half-power linewidth of the absorption curve by multiplying $\Delta H$ by $\sqrt{3}$ (assuming a Lorentzian line shape).

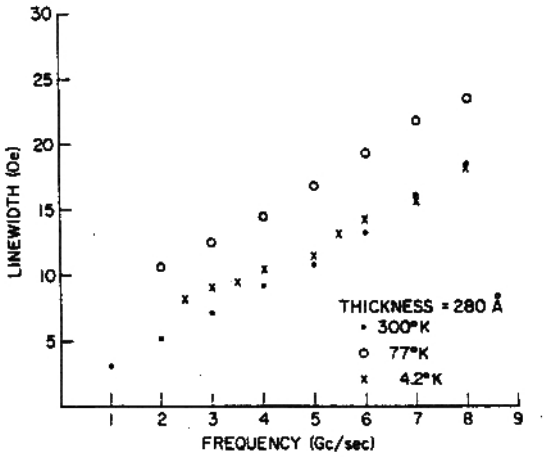


FIG. 1. Linewidth data as a function of frequency for a 280-Å-thick film at 300°, 77°, and 4.2°K.

measurements could be made down to about 2°K by pumping on the liquid helium. Temperature measurements were made using a calibrated carbon resistor and a pulsed-resistance bridge. For measurements between 300° and 77°K, the outer Dewar was filled with liquid nitrogen and measurements were made as the stripline assembly cooled down. The rate of cooling was about 1.5°K/min. For measurements between 4.2° and 77°K, linewidth data were taken as the system warmed up when the helium level fell below the stripline. The rate of temperature rise was about 1°K/min.

In Fig. 1, linewidth data for a 280-Å film as a function of frequency are shown for 300°, 77°, and 4.2°K. At all frequencies, the linewidth is larger at 77°K than at the two extremes. Similar effects were observed for films from 150 to 1500 Å thick. The magnitude of $(\Delta H_{77°K} - \Delta H_{300°K})$ appears to be larger for thinner films and is influenced by annealing treatments to be discussed shortly. The data in Fig. 1 indicate that the linewidth for parallel resonance in thin films does exhibit a significant temperature dependence. In order to obtain a more accurate picture of this dependence, the linewidth for a 150-Å film was obtained as a function of temperature from 4.2°K to room temperature, at 2, 4, and 6 Gc/sec. These data are shown in Fig. 2. Linewidth data at 2 Gc/sec could not be obtained below 70°K.[9] For the other two frequencies, the linewidth exhibits a maximum in the vicinity of 80°K.

One important conclusion which can be drawn from the data of Fig. 2 concerns the effect of eddy-current damping. The eddy currents induced in a metal film because of the magnetization motion have been calculated by Smith[10] for the situation where the film thickness is much less than the skin depth and the magnetization is constrained to remain in the plane of the film. From Smith's calculation, the resonance linewidth due to eddy-current losses is linearly proportional to the frequency and film conductivity, and is quadratic in film thickness. The shape of the curve is completely different than predicted by the temperature dependence of the conductivity. In addition, the observed temperature dependence is smaller for thicker films. Eddy-current losses cannot explain the present data concerning the temperature dependence of the linewidth for thin films.

Two important features can be seen from the data in Fig. 2. First, the amplitude of the linewidth peak is almost constant, independent of frequency. Second, the temperature at which the linewidth maximum occurs appears to shift to slightly higher temperature with increasing frequency. These two features are characteristic of the linewidth temperature dependence predicted by both valence-exchange[2] and impurity-relaxation[3] processes. Several difficulties occur in applying either of these two theories to the present data. If an impurity-relaxation mechanism is to be taken seriously, a quantitative understanding of the contribution of specific impurities to the peak in linewidth is required. Any explanation based on valence exchange must provide for the formation of divalent and trivalent ions for charge transfer. In addition, any modification of these two theories for metals must include the presence of conduction electrons.

As mentioned previously, the amplitude of the temperature effect was largest for the thinner films. In addition, the amplitude was observed to be larger for older films than for freshly evaporated samples. These two observations indicated that some contamination effect might be important, such as oxidation or diffusion through the film surface. For this reason, the influence of annealing treatments on $(\Delta H_{77°K} - \Delta H_{300°K})$ was investigated. The first annealing treatment consisted of simply heating the film to 150°C in a poor vacuum of $10^{-3}$ Torr, with an applied field of 50 Oe in the plane of the film along the easy axis. The vacuum-annealing

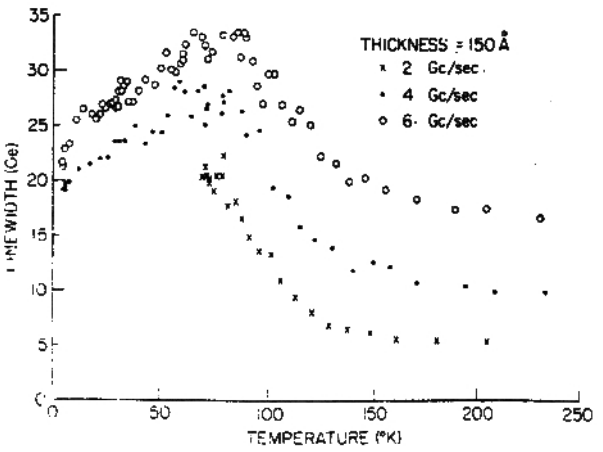


FIG. 2. Linewidth data as a function of temperature for 150-Å-thick film at 2, 4, and 6 Gc/sec.

[9] The disappearance of the 2-Gc/sec. resonance below 70°K may be associated with the anomalous increase in the anisotropy field at low temperatures reported by Mitchel and Briscoe [J. Appl. Phys. 37, 1489 (1966)]. However, the anisotropy increase should shift the hard-direction resonance to a higher field and it should still be observable. Both the easy- and hard-direction resonances disappeared off the low end of the field sweep at temperatures below 70°K.

[10] D. O. Smith, *Magnetism*, G. T. Rado and H. Suhl, Eds. (Academic Press Inc., New York, 1963), Vol. III, Chap. 10, pp. 506–7.

procedure should slow the contamination sufficiently, compared to annealing at atmospheric pressure, so that the increase of $(\Delta H_{77°K}-\Delta H_{300°K})$ could be observed at progressive stages of the process. If the linewidth enhancement were due simply to volume diffusion through the film surface, the time $t_s$ required for the enhancement to saturate should exhibit a marked thickness dependence. From a simple analysis of diffusion through a plane surface of fixed impurity concentration, one would expect $t_s$ to vary as the square of the film thickness. In Fig. 3, $\Delta H_{77°K}$ and $\Delta H_{300°K}$ are shown as a function of annealing time for films 190, 290, and 1350 Å thick. The enhancement in the linewidth at 77°K is greatest for the thinnest sample, but the saturation annealing time for all three films is about 80 h. Clearly $t_s$ is not quadratic in thickness and the enhancement in the linewidth maximum as a function of temperature is not due to volume diffusion.

The second annealing procedure which has been investigated could be better termed an oxidation–reduction treatment. The film was heated to about 150°C in a hydrogen atmosphere (reduction) or in air (oxidation) with an applied field of 50 Oe along the easy axis. In Fig. 4, $\Delta H_{77°K}$ and $\Delta H_{300°K}$ are shown after successive oxidation and reduction treatments. The first oxidation treatment for 12 h increased $\Delta H_{77°K}$ by over 2.0 Oe and had only a small influence on $\Delta H_{300°K}$. Next the film was reduced for 10 h. After this treatment, $\Delta H_{77°K}$ and $\Delta H_{300°K}$ were equal and almost 0.5 Oe lower than the initial room-temperature linewidth. By heating the sample in a hydrogen atmosphere at 150°C for 10 h, the linewidth peak as a function of temperature was completely eliminated. As shown in Fig. 4, repeated oxidation and reduction treatments had effects similar to those discussed above. Oxidation reproducibly enhanced the amplitude of the linewidth peak and reduction made the effect vanish. Whatever

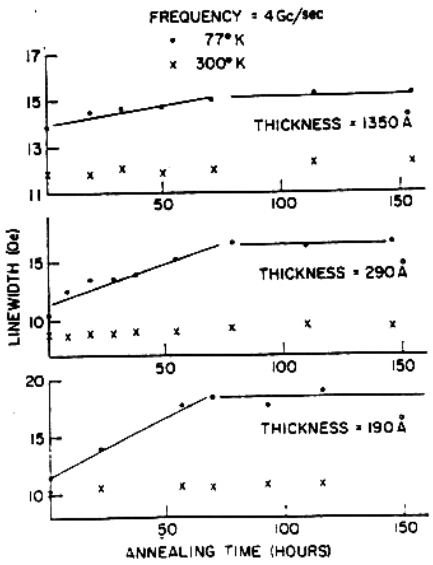


FIG. 3. Linewidth data (4Gc/sec) at 77° and 300°K for three films. 190, 290, and 1350 Å thick, as a function of annealing time at 150°C in a vacuum of $10^{-3}$ Torr with a 50-Oe field along the easy axis.

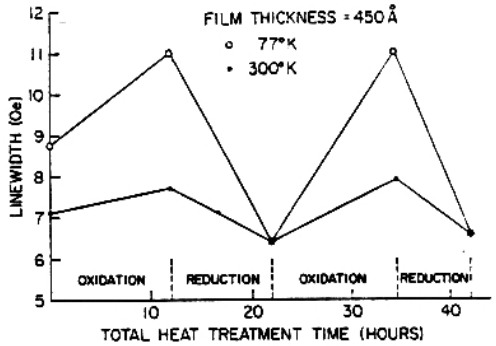


FIG. 4. Linewidth data (4 Gc/sec) at 77° and 300°K for a 450-Å film after successive oxidation and reduction treatments at 150°C and atmospheric pressure in a 50-Oe easy-axis field.

the physical mechanism causing the linewidth maximum as a function of temperature, the present annealing and oxidation–reduction data indicate that surface oxidation is involved.

Two possible mechanisms are suggested by the apparent importance of surface oxidation in producing the linewidth maximum as a function of temperature, valence exchange and exchange anisotropy. Valence exchange, or the $Fe^{2+}$–$Fe^{3+}$ charge transfer relaxation mechanism, which is important in high-conductivity spinel ferrites, is a possible source of linewidth in crystals containing both $Fe^{2+}$ and $Fe^{3+}$ ions on a single type of crystallite site.[2] Surface oxidation may well result in such a situation in which the conduction electrons have no effect on the surface oxide $Fe^{2+}$–$Fe^{3+}$ charge-transfer relaxation. The oxidation–reduction treatments discussed above are very similar to the treatments discussed by Hagedorn[11] in connection with his observation of anomalous hysteresis loops in Ni–Fe alloy films at low temperatures. Hagedorn proposed the formation of an antiferromagnetic oxide on the film surface and attributed the anomalous loop behavior to exchange anisotropy.[12] The rotational hysteresis loss for the Fe–FeO system does show a rapid increase below 120°K,[13] similar to the increase in linewidth in Fig. 2. However, complete data at low temperatures, to show whether there is a peak, are not available. It is entirely possible that exchange anisotrophy may also be connected with the linewidth data reported here.

## SUMMARY

Linewidths have been measured for films 150 to 1500 Å thick at temperatures from 4.2° to 300°K and frequencies from 1–9 Gc/sec. The linewidth exhibits a maximum in the vicinity of 80°K. The effect is generally larger in thinner films. The amplitude of the linewidth temperature-dependence $(\Delta H_{max}-\Delta H_{300°K})$ is independent of frequency and the maximum shifts to slightly higher temperatures for increasing frequency. These features are characteristic of the linewidth temperature-dependence predicted by valence-exchange and impurity

[11] F. B. Hagedorn, J. Appl. Phys. **38**, 1362 (1967).
[12] J. S. Kouvel and C. D. Graham, Jr., J. Phys. Chem. Solids **11**, 220 (1959).
[13] W. H. Meiklejohn, J. Appl. Phys. **29**, 454 (1958).

relaxation mechanisms. The linewidth maximum can be enhanced by heating the film at 150°C in air (oxidation) and removed by a similar treatment in a hydrogen atmosphere (reduction). This enhancement-removal cycle can be repeated reproducibly. The time required for saturation of the enhancement when the oxidation treatment was slowed down by annealing in a vacuum of $10^{-3}$ Torr instead of at atmospheric pressure was observed to be independent of film thickness, indicating that a volume diffusion process is not involved. The importance of surface oxidation in producing the linewidth maximum suggests two possible mechanisms, valence exchange and exchange anisotropy, which may contribute to the effect reported here. The pertinent points concerning these mechanisms have been discussed. The most important results of this investigation are: (1) the discovery of a temperature-dependent linewidth in thin films which exhibits a maximum in the vicinity of 80°K and (2) the identification of this dependence with a surface oxidation process.