Ferromagnetic Resonance in Nickel at Low Temperatures*

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We have studied the ferromagnetic resonance linewidth in nickel at 9, 22, and 35 kMc/sec as a function of temperature from 300 down to 4°K. The 4°K linewidth is found to be independent of the resistivity of the sample but is much larger than the linewidth computed using any of the extant theories with a temperature-independent intrinsic relaxation. In order to fit the data it is necessary to assume that the intrinsic relaxation parameter is constant down to 130°K, but that on further lowering of temperature it increases rapidly (for no known reason) until it saturates at a value about 6 times its value at room temperature.

INTRODUCTION

UNTIL recently[1] ferromagnetic resonance (FMR) does not appear to have been observed in a pure high-conductivity metal at liquid-nitrogen temperature or below. This failure has sometimes been erroneously attributed to the action of the exchange-conductivity mechanism in conjunction with anomalous conductivity.[2] A correct analysis shows, however, that the anomalous form of conductivity should not have any upsetting effect on the resonance,[3] and should in fact effectively cut off the temperature-induced conductivity increase and thus limit the linewidth increase from the exchange-conductivity mechanism. Our recent measurements on FMR linewidths in iron single-crystal whiskers (Ref. 1) demonstrate very clearly the effects predicted by the nonlocal conductivity theory.

In this paper we report observations of FMR at liquid nitrogen temperatures and 4°K in several bulk single-crystal samples of nickel at 9, 22, and 34 kMc/sec. Two important results emerge. First, at 4.2°K the linewidth is very nearly independent of the resistivity of the sample for resistivity changes of a factor of 5. This result is certainly in accord with the nonlocal conductivity theory. However, the situation is somewhat complicated by our second result: As the temperature is lowered and/or the frequency increased a discrepancy between the observed and computed linewidths arises unless the relaxation damping is allowed to increase with decreasing temperature. In fact, at 4.2°K the measured linewidths at all frequencies are factors of 2 or more greater than the widths computed by using either the local or the nonlocal conductivity theory with the assumption that the intrinsic relaxation is represented by a temperature-independent Landau-Lifshitz parameter λ.

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[1] S. M. Bhagat, J. R. Anderson, and L. L. Hirst, Phys. Rev. Letters **16**, 1099 (1966).

[2] D. S. Rodbell [Physics **1**, 279 (1965)] has stated that some change in the character of the line took place as he cooled the sample down. We have seen no untoward change apart from the increased widths.

[3] L. L. Hirst and R. E. Prange, Phys. Rev. **139**, A892 (1965).

EXPERIMENTAL METHOD

Sample preparation and microwave techniques were for the most part similar to those in room-temperature work previously reported.[4] The samples had a nominal purity of 99.998%, and in order to check that the measurements were not being prejudiced by some peculiarity of the method used for growing the single crystals, we have used samples cut from single crystals obtained from several independent sources.[5] Both disk-shaped and cylindrical samples have been used. For low-temperature work the sample cavity was surrounded by an evacuated chamber and its temperature was monitored with a copper-constantan thermocouple. The temperature measurements are probably good to only 2 or 3°K. The magnetic field is either in the plane of the sample (disks) or along its axis (cylinder).

There are several experimental factors which may render observation of FMR more difficult at low temperatures. The most obvious of these is the weakened signal, due to the decreased skin depth which reduces the total line intensity, coupled with spreading of the intensity over an increased line width. However, FMR signals are strong enough that there need be no intensity problem, provided larger sample areas are used than would be appropriate at higher temperatures. In our experiments strong signals were detected both by the field-modulation technique (using modulation amplitudes of the order of 100 Oe) as well as by direct detection of the crystal output without field modulation, amplified by a Keithley 660 A guarded dc differential voltmeter.

A second experimental problem is a high sensitivity to the sample mounting which is not understood in detail but apparently has to do with the fairly strong magnetic force which will act on the sample if the field is nonuniform. The symptoms are distortions of the line shape, which, in the case of field-modulated curves, tends to alter the asymmetry more than the width. Whenever this occurs there is also an irregular absorp-

[4] S. M. Bhagat, L. L. Hirst, and J. R. Anderson, J. Appl. Phys. **37**, 194 (1966).

[5] Materials Research Corporation, Orangeburg, New York; Metals Research, Cambridge, England; single-crystal sphere kindly provided by Dr. E. R. Callen of Naval Ordnance Laboratory.

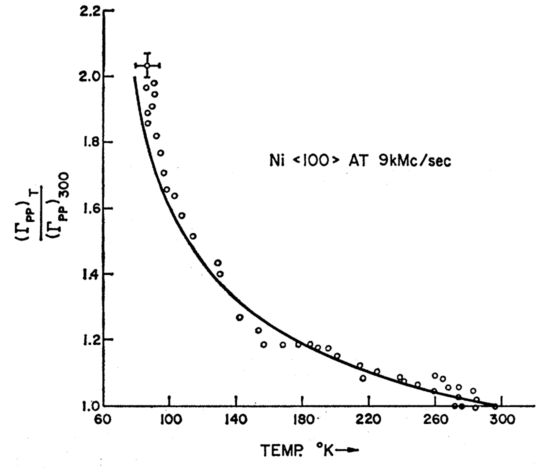

FIG. 1. Observed temperature variation of the linewidth in nickel at 9 kMc/sec. The full line is obtained using the normal conductivity theory, taking the conductivity to vary linearly with temperature and the other parameters as having the values given in Ref. 3.

tion in the vicinity of small applied field. For moderately bad mountings this signal can be of the same magnitude as the main resonance, and its absence gives a useful empirical test of the quality of the mounting. The best mountings were found to be those which were either viscous (as GE 7031 adhesive) or loose (as mounting in a block of Styrofoam). Worst mountings were those which held the sample elastically, such as rubber cement. One possible mechanism considered for the distortion was resonant mechanical vibrations of the sample at the field-modulation frequency; but this appears to be ruled out by the accurate correspondence which was found, even in the case of distorted lines, between signals using the modulation and direct techniques. In the case of satisfactory mountings the observed linewidths were essentially independent of the type of mounting.

A third possible experimental complication is worth mentioning. This can arise at 9 kMc/sec because of the large magnetocrystalline anisotropy of nickel at low temperatures. At this frequency it is necessary to observe the resonance with the steady field applied along the ⟨100⟩ "hard" axis, since the large anisotropy field along the ⟨111⟩ "easy" axis would shift the resonance to higher frequencies. In disk samples with the field applied along ⟨100⟩, at fields below the main resonance the magnetization becomes unstable and takes up a new orientation, with a resulting second low-field resonance.[6] On the other hand, as the applied field H is turned away from the ⟨100⟩ axis, the magnetization will tend to tilt away from H, with the tilt angle varying as the field is swept. A straightforward analysis shows a surprisingly strong angular dependence of the resonance behavior, which, in the example cited, causes the resonance to disappear if H deviates from the ⟨100⟩ axis by more than 5°. Such a misalignment would mean that upon cooling the sample and thus increasing the anisotropy, one would observe a fairly sudden disappearance of the resonance, which could easily be misinterpreted as some kind of anomaly.[7]

The large anisotropy field, or rather its variation with angle, also gives rise to problems in the case of ⟨100⟩ cylindrical samples. If the magnetic field is not accurately aligned along the cylinder axis, the observed width is considerably enhanced. A misalignment of about 10° at 80°K and about 5° at 4°K is sufficient to give a 50% larger width. Since we are unable to tip the magnetic field so as to test for misalignment in the horizontal plane, we may indeed be left with a slight residual broadening due to this effect. However, the repeatability of the data for several different mountings and samples is reassuring. Also, this effect is much smaller for the ⟨111⟩ samples; no variation of width is seen for deliberate misalignments of some 10° to 15°. Taking into account all the above effects we claim that the linewidths reported below are good to about 8%. As before, the linewidth is always defined as the field separation between the maxima of the derivative curve.

DISCUSSION AND RESULTS

First, consider the linewidths at temperatures between room temperature and 80°K. Figure 1 shows the variation of the linewidth (normalized to the room temperature value) at 9 kMc/sec in this temperature range. As the temperature is lowered the line broadens very slowly in the beginning, but at about 150°K the increase in width becomes much more rapid. This is as expected, because the linewidth at 297°K is dominated by the contribution due to λ (cf. Ref. 3) and therefore one should expect a rather slow increase when the temperature is first lowered due to the variation of the much smaller exchange contribution. In this region one uses essentially the computations described in Ref. 3 and the full curve in Fig. 3 shows the result of this calculation. The resistivity is taken as varying linearly with the temperature and the other parameters are taken from our room-temperature work. Although it is not evident

TABLE I. Observed and computed linewidths in nickel at 80°K ($\rho_{300}/\rho_{80}=14$).

Sample	Freq. (kMc/sec)	Observed	Calculated $K_s=0$	0.1	0.25	0.4	∞
Cylinders ⟨100⟩	9.0	380	220	280	310	320	410
Cylinders ⟨111⟩⟨100⟩	22.0	700	375	425	490	550	650
Disks [110]							
Cylinders ⟨111⟩	34.8	1100	550	550	600	750	850

[6] J. O. Artman, Phys. Rev. **105**, 74 (1957); L. L. Hirst and S. M. Bhagat, Bull. Am. Phys. Soc. **10**, 471 (1965).

[7] S. M. Bhagat and L. L. Hirst, University of Maryland Technical Report No. 506, 1965 (unpublished).

TABLE II. Observed and computed linewidths in nickel at 4°K.

Sample	Frequency (kMc/sec)	Observed	Anom.	Computed[a] ($\lambda=2.3\times10^8$ sec^{-1}) Normal	Computed [$\lambda=13.8\times10^8$ sec^{-1} anom.]
Cylinders	9.0	640	275	375, 470, 600	650
Cylinders ⟨100⟩ ⟨111⟩ disks [110]	22.0	1500	450	600, 700, 950	1450
Cylinders ⟨111⟩	34.8	2200	550	1150, 1300, 1600	2200

[a] The residual-resistivity ($\rho_{300}/\rho_{4.2}$) ratios of our samples are 30, 60, and 160. In the anomalous case the calculated widths are the same for all these; but for the normal case we got 3 values as given in column 3.

from Fig. 1, the discrepancy between the observed and computed linewidths begins to be significant as the temperature is lowered below about 130°K. In Table I we show the observed and computed linewidths at 80°K. The normal conductivity theory is applicable here, and computations have been made for several values of the surface anisotropy energy K_s. In Ref. 3 it was shown that we required a value of 0.25 erg/cm^2 to explain the room temperature linewidths. From Table I one can see that if we were willing to make K_s larger, we could probably explain the observed linewidth at 9 kMc/sec. However, even the assumption of complete pinning will be insufficient to explain the data at higher frequencies.

On cooling below nitrogen temperature the line broadens very rapidly. We have not made detailed measurements in this region, but it appears that most of the broadening occurs within 20 or 30°K of nitrogen temperatures. On cooling further down to 4°K the width remains essentially constant. Further, for the 3 sets of samples having residual resistivity ratios of 30, 60, and 160, there is no significant difference between the linewidths at 4°K.

At low temperatures the exchange-conductivity line-broadening mechanism is thought to dominate, and one uses a conductivity dependent on the wave number q. In the low-q (normal) region it approaches a constant σ_0, and in the high-q (extreme anomalous) region it approaches C_F/q; in the intermediate region an interpolation formula suggested by the Drude model can be used. [The interpolation formula is given in Eq. (17) of Ref. 2, but with a misprint; it should read

$$\tilde{\sigma}(q)=\sum\frac{C_F{}^{(s)}}{i\pi q}\left[\left(1-\left\{\frac{\chi^{(s)}}{q}\right\}^2\right)\ln\left\{\frac{\chi^{(s)}-q}{\chi^{(s)}+q}-\frac{2\chi^{(s)}}{q}\right\}\right].$$

In the present case there is no summation since only the conduction electrons contribute appreciably.] The extreme anomalous conductivity constant C_F depends on a Fermi-surface integral which cannot be calculated accurately, but can be roughly estimated from the total number of conduction electrons as $C_F=10^{24}$ cm^{-1} sec^{-1} for nickel. (The d electrons make a negligible contribution because of their large effective mass.)

In Table II, compared with the experimental widths, are given linewidths computed from the exchange-conductivity theory at 4°K. In the first column the value $C_F=1\times10^{24}$ is used, and in the second column fully normal conductivity is assumed, which for computation purposes is taken to be $C_F=10^{31}$. The exchange stiffness $A=0.75\times10^{-6}$ erg/cm is taken from thin-film spin-wave resonance experiments[8] (in approximate agreement with the value derived from the $T^{3/2}$ dependence of the magnetization). The surface anisotropy constant $K_s=0.25$ erg/cm^2 and the Landau-Lifshitz relaxation parameter $\lambda=2.3\times10^8$ sec^{-1} have been obtained from room-temperature FMR linewidth fits (Ref. 3). It is seen that the calculated widths are barely half of those found experimentally. However, it is worthwhile to note that the observed linewidths are independent of the residual-resistivity ratio and this is evidence that we are in the anomalous region.

Increase of surface anisotropy with decreasing temperature could give some extra broadening, but a strong temperature dependence of the surface anisotropy is not expected on such meager theoretical grounds as are available. In the case of iron, for instance, our measurements are consistent with a temperature-independent surface anisotropy. As already noted above in discussing the 80°K data, an increase of surface anisotropy even to the limit of complete pinning of the surface spins would not make up the discrepancy.

In interpreting the low-temperature data we have to proceed with extreme care, especially when the observed linewidths are too large in comparison with those given by any of the extant theories. One must make sure that this is not a purely experimental effect. Several sources of error can arise. First, one can question the purity of the samples. We have obtained spectroscopic analyses of our samples and found that they do not contain more than a few parts per million of any metallic impurity. However, since the residual resistivity ratios are not large we cannot exclude the possibility of absorbed gases. On the other hand, since the observed linewidths are the same for several shapes, sizes, and sources of samples, impurity scattering does not seem to be a very likely source of the extra linewidth. Next, one has to make sure that the sample was not strained during the cooling process. It is well known that an inhomogeneous strain will give rise to an extra width via magnetostric-

[8] H. Nose, J. Phys. Soc. Japan 16, 2475 (1961).

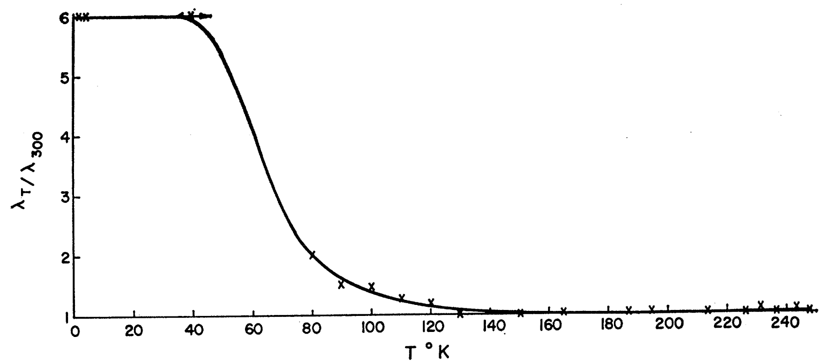

FIG. 2. Temperature variation of the effective relaxation parameter in nickel.

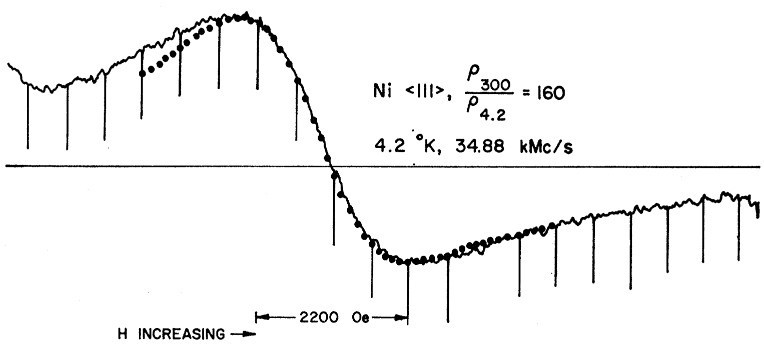

FIG. 3. Experimental resonance curve in nickel with theoretical points calculated using $\lambda=13.8\times10^8$ sec^{-1}, $C_F=10^{24}$ cm^{-1} sec^{-1}, and $A=0.75\times10^{-6}$ erg/cm^2.

tion. There are two reasons to believe that this is not the source of the extra widths observed by us. Firstly, the linewidths are independent of the methods of mounting discussed earlier. Again, it is well established that the broadening due to inhomogeneous strain will also lead to a shift in the line center, the amount of the shift being of the order of the extra width. [It may be worthwhile to note that[9] at low temperature the magnetostriction constant Λ_{111} is one-half as large as Λ_{100} and therefore one would expect to see a highly anisotropic linewidth if it was being mediated by magnetostriction.] In our measurements we have seen no evidence of such a shift because the value of the anisotropy constant K_1 computed from the position of the center of the line, including an adequate correction for the exchange shift,[10] agrees to within a few percent with the value obtained from the torque measurements.[11] The situation with respect to K_2 is not so clear. Our data are consistent with a very small value of K_2/M ($\sim$100 Oe).

The only parameter left at our disposal is the Landau-Lifshitz λ. So far we have treated it as being temperature-independent and used the room temperature value of 2.3×10^8 sec^{-1}. Unfortunately, the microscopic origin of λ is not known and therefore it is not possible to say what one should expect for its temperature variation. Rodbell's data[12] in the high-temperature region show that λ is sensibly constant. We completely agree with this for temperatures above about 130°K, but at lower temperatures it does not appear that a temperature-independent λ will explain the data. If we assume that all of the discrepancy can be attributed to the variation of λ with temperature, we find for λ the values shown in Fig. 2. The computed widths at 4°K with $\lambda=13.8\times10^8$ sec^{-1} are shown in the last column of Table II, and in Fig. 3 we show how well these values of the parameters describe the details of the line shape.

ACKNOWLEDGMENTS

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[9] W. D. Corner and G. H. Hunt, Proc. Phys. Soc. (London) A68, 133 (1955). See also W. D. Corner and F. Hutchinson, *ibid.* 72, 1049 (1958).

[10] L. L. Hirst and S. M. Bhagat, University of Maryland Technical Report No. 507, 1965 (unpublished).

[11] I. M. Puzei, Bull. Acad. Sci. USSR, Phys. Ser. 21, 1077 (1957).

[12] D. S. Rodbell, Phys. Rev. Letters 13, 471 (1964).