This shows that $\text{Im}\Gamma_q^{zz}(t)$ is negligible compared to $\text{Re}\Gamma_q^{zz}(t)$ in the Weiss limit, as was already shown in RDL III Sec. V using a slightly different argument. If, instead of (C1), one starts from another well-known exact result

$$\frac{dR_q^{zz}(t)}{dt} = -2\,\text{Im}\Gamma_q^{zz}(t)\,, \qquad \text{(C7)}$$

which can be demonstrated by taking the time derivative of (5.2), one is, of course, not allowed to neglect the right-hand side! One writes instead, using (C3), (C4), and (C6),

$$\omega R_q^{zz}(\omega) = 2\Gamma_q^-(\omega) \simeq \beta\omega\Gamma_q^+(\omega)\,, \qquad \text{(C8)}$$

which is merely a restatement of (C4).

Let us finally point out that the neglect of $\text{Im}\Gamma_q^{zz}$ implies that the spectral function is symmetrical around $\omega = 0$; this result is a rigorous consequence of the Weiss limit although it is only approximately valid in realistic systems.

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Ferromagnetic Resonance in Nickel at High Temperatures*

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Ferromagnetic resonance measurements on high-purity nickel metal are reported at 23 and 32 kMc/sec and at temperatures varying between 20 and 380°C. Up to 300°C a temperature-independent relaxation parameter ($\lambda = 2.3\times10^8$ sec^{-1}) and $g = 2.21$ describe the data adequately. At higher temperatures, the lines become much narrower than expected from such a value of λ, and the g value becomes somewhat dependent on temperature as well as frequency.

INTRODUCTION

ROOM-TEMPERATURE measurements[1,2] on the ferromagnetic resonance (fmr) in nickel (bulk samples or whiskers) have shown that the resonance can be adequately described by a Landau-Lifshitz type of equation of motion for the dynamic magnetization. In addition, for frequencies above 1 kMc/sec, the exchange conductivity contribution to the linewidth is small and this allows one to claim with some confidence that the value of the relaxation parameter λ is 2.3×10^8 sec^{-1}, independent of frequency. Recently, Rodbell[3] has reported some measurements on single crystal whiskers at temperatures varying from room temperature to about 350°C. The experiments were done at 9 kMc/sec and his main results were that both the g value and λ were essentially constant over this range of temperatures. In this paper, we report observations of fmr on several bulk single crystals of high-purity (nominally 99.997%) nickel. The measurements have been made at 23.3 and 31.8 kMc/sec with temperatures ranging from 20 to ~380°C, the highest temperature at which a resonance signal was still discernible. Our results essentially confirm Rodbell's measurements up to about 300°C. At higher temperatures, however, (i) the lines become considerably *narrower* than would be expected from a temperature-independent λ, (ii) the linewidths are *not* proportional to frequency, and (iii) the g value is *not* independent of temperature and frequency.

EXPERIMENTAL METHOD

Sample preparation and microwave techniques were essentially similar to those described in earlier papers from this laboratory.[4] Cylindrical samples of diam 0.1 cm and length 1 cm were used with the cylinder axis either along ⟨100⟩ or ⟨111⟩. The magnetic field was applied along the cylinder axis and modulated at a low frequency. The sample cavity was maintained in a vacuum of better than 10^{-4} mm Hg throughout the heating cycles. This was found to be absolutely essential to prevent irreversible changes in the sample when it had been heated. The data reported below are taken from several heating cycles. All data for which the room-temperature linewidth at the conclusion of a heating cycle differed from that at the beginning were rejected. The sample temperature was monitored with a copper constantan thermocouple attached to the outside of the microwave cavity. The thermocouples were calibrated against an NBS standard platinum resistnace

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† Submitted by one of us (E.P.C.) in partial fulfillment of the requirements of a Master's degree at the University of Maryland.
[1] D. S. Rodbell, Phys. Rev. Letters, **13**, 471 (1964).
[2] S. M. Bhagat, L. L. Hirst, and J. R. Anderson, J. Appl. Phys. **37**, 194 (1966).
[3] D. S. Rodbell, Physics **1**, 279 (1965).
[4] S. M. Bhagat and H. O. Stevens, Jr., J. Appl. Phys. **39**, 1067 (1968); also, E. P. Chicklis and S. M. Bhagat, University of Maryland, Technical Report No. 859, 1968 (unpublished).

thermometer. In order to minimize the effects of any thermal lag, data were taken only after the temperature had been stable for some 15 min. In addition, this was done both during the warming-up and the cooling-down portions of the cycle. No measurable differences were observed. The temperature measurements are probably good to 2 or 3°C. The linewidths are good to about 10%, except at high temperatures where the line shape is greatly distorted and the accuracy concomitantly poorer. The line centers were measured with a precision of about 50 Oe except at the highest temperatures, where the errors may be twice as large.

RESULTS AND DISCUSSION

A. *g* Value

Figures 1 and 2 show the observed variation of the resonance center as a function of temperature for 31.8 and 23.3 kMc/sec, respectively. The full lines in these figures are obtained as solutions of the Eqs.

$$\omega/\gamma=[(H+H_{\mathrm{aniso}})(H+H_{\mathrm{aniso}}+4\pi M_s)]^{\frac{1}{2}}, \qquad (1)$$

where γ is the gyromagnetic ratio $g|e|/2mc$, M_s is the saturation magnetization and H_{aniso} is the so-called anistropy field, $+2K_1/M$ for $H\|\langle 100\rangle$ and $-\frac{4}{3}(K_1/M)$ $-(4/9)(K_2/M)$ for $H\|\langle 111\rangle$. In our calculations we have taken $g=2.21$, and the values of K_1 and K_2 have been taken from the data of Rodbell.[3] The values of M were taken from the measurements of Weiss and Forrer[5] and require a word of explanation since at high temperature M is no longer independent of H. For the full lines in Figs. 1 and 2 we have used a constant $M_s=M_s(H_r)$, where H_r is the observed line center. It is clear that the data at 23.3 kMc/sec cannot be explained by a temperature-independent g.

Next, we made detailed calculations of the fmr line, using the computer program by Hirst[6] and taking explicit account of the variation of M with H. The data of Weiss and Forrer were represented by an expression of the form $M=cH^{1/\delta}$. For $g=2.21$ and $\lambda=2.3\times10^8$ sec^{-1}, the results of the computation are shown in Table I. For comparison we have also included in Table I the line center obtained using $M=M(H_r)$,[7] as well as the observed values. Since the effect of including the variation of M with H is to shift the computed line center to higher fields, the apparent agreement between the $g=2.21$ curve and the 31.8 kMc/sec data is somewhat spoiled for $T\gtrsim360$°C. Thus both at 31.8 and 23.3 kMc/sec, the observations would require a somewhat higher value for g in the high-temperature region.

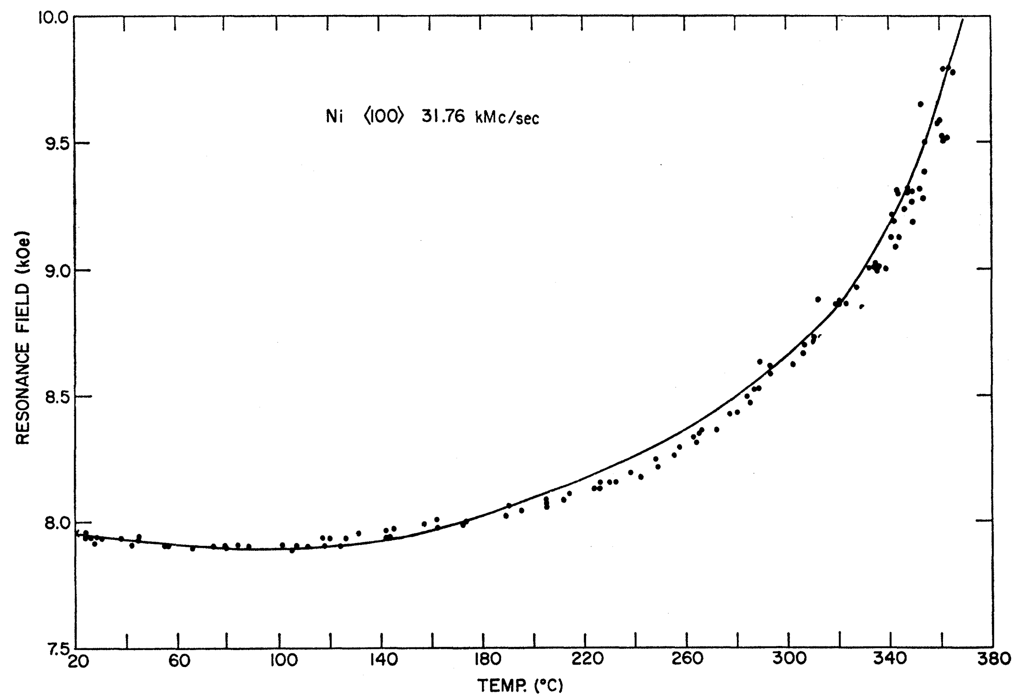

FIG. 1. Temperature variation of the observed resonance field at 31.8 kMc/sec. The full line was obtained as solution of Eq. (1) with $g=2.21$ and $M=M(H_r)$ as explained in the text. The different symbols stand for results of different heating cycles.

[5] P. Weiss and R. Forrer, Ann. Phys. (Paris) 5, 153 (1926).

[6] L. L. Hirst, Ph.D. thesis, University of Maryland, 1965 (unpublished).

[7] It must be understood that in the absence of a better theory of fmr at high temperatures the present calculations are done merely to indicate the trend in which the various corrections are likely to go. They are not at all intended to suggest that a precise value of g (or λ) can be obtained without considerable further effort.

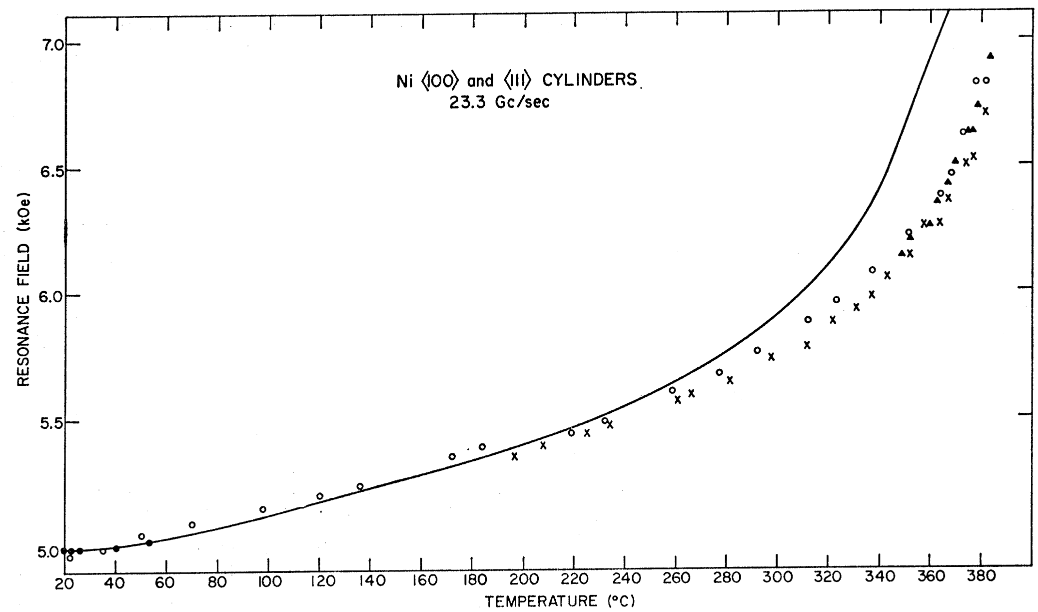

FIG. 2. Temperature variation of the observed resonance field at 23.3 kMc/sec. The full line was obtained as in Fig. 1.

In addition, the same value of g may not be adequate to account for the resonance field at both frequencies. However, for reasons outlined below it does not seem reasonable to try and derive specific values for g from the observations.

B. Linewidths

The fmr linewidth in a metal cannot, in general, be written in any simple form in terms of the parameters. However, for a situation like that of nickel, where the width is dominated by relaxation terms, one can write roughly

$$\Gamma_{pp}=1.45\lambda\omega/\gamma^2 M_s+(\Gamma_{pp})_{\mathrm{ex}}, \qquad (2)$$

where $(\Gamma_{pp})_{\mathrm{ex}}$ is the contribution due to the exchange conductivity including the surface anisotropy contributions, if any. The temperature variation of $(\Gamma_{pp})_{\mathrm{ex}}$ goes approximately as $M_s/\sqrt{\rho}$, where ρ is the dc resistivity of the metal. Detailed calculations show that at room temperature $(\Gamma_{pp})_{\mathrm{ex}}$ amounts to about 10% of the total linewidth for 23 kMc/sec (at 32 kMc/sec, the contribution is relatively smaller), while above about 300°C this contribution becomes entirely negligible. In Figs. 3 and 4 we have exhibited the observed linewidths at 23.3 and 31.8 kMc/sec, respectively, as a function of temperature. The full curves are derived from Eq. (2), using $\lambda=2.3\times10^8$ sec^{-1}, $g=2.21$, and $M=M(H_r)$. Clearly, the agreement between the theory and experiment is excellent up to about 300°C. At higher temperatures the observed lines are considerably narrower, the discrepancy becoming more and more serious as the temperature is increased. As is clear from Table I above about 360°C, part of the discrepancy is removed when one computes the linewidth using the fact that M is a function of H. An increasing g value will also lead to smaller widths. Further, the variation of M with H and a possible dependence of g on frequency will combine to produce linewidths which are *not* proportional to frequency as is observed experimentally. In principle, it may be possible to generate a set of g and λ values to fit the observations. However, in the absence of a dependable theory of magnetic relaxation the wisdom of quoting any specific values is highly questionable.

TABLE I. Calculated and observed characteristics of the ferromagnetic resonance in nickel at high temperatures. All values are in Oe. The following parameters were used in the computations: $g=2.21$, $\lambda=2.3\times10^8$ sec^{-1}. The values of M (and its dependence on H) were taken from Ref. 5.

Temp. °C		Freq. =31.76 kMc/sec				Freq. =23.3 kMc/sec			
		$H_r[M=M(H_r)]$	$H_r(M=cH^{1/\delta})$	$\Gamma_{pp}[M=M(H_r)]$	$\Gamma_{pp}(M=cH^{1/\delta})$	$H_r[M=M(H_r)]$	$H_r(M=cH^{1/\delta})$	$\Gamma_{pp}[M=M(H_r)]$	$\Gamma_{pp}(M=cH^{1/\delta})$
350	Calc.	9400	9400	1327	1150	6800	6800	975	1000
	Obs.	9300		1050		6150		550	
356	Calc.	9575	9650	1642	1400	6950	6950	1150	1070
	Obs.	9400		1150		6250		600	
360	Calc.	9700	9900	1978	1600	7170	7270	1300	1150
	Obs.	9550		1250		6300		650	
369	Calc.	9950	10650	3465	2700	7400	7900	2600	2350
	Obs.	...		...		6500		725	

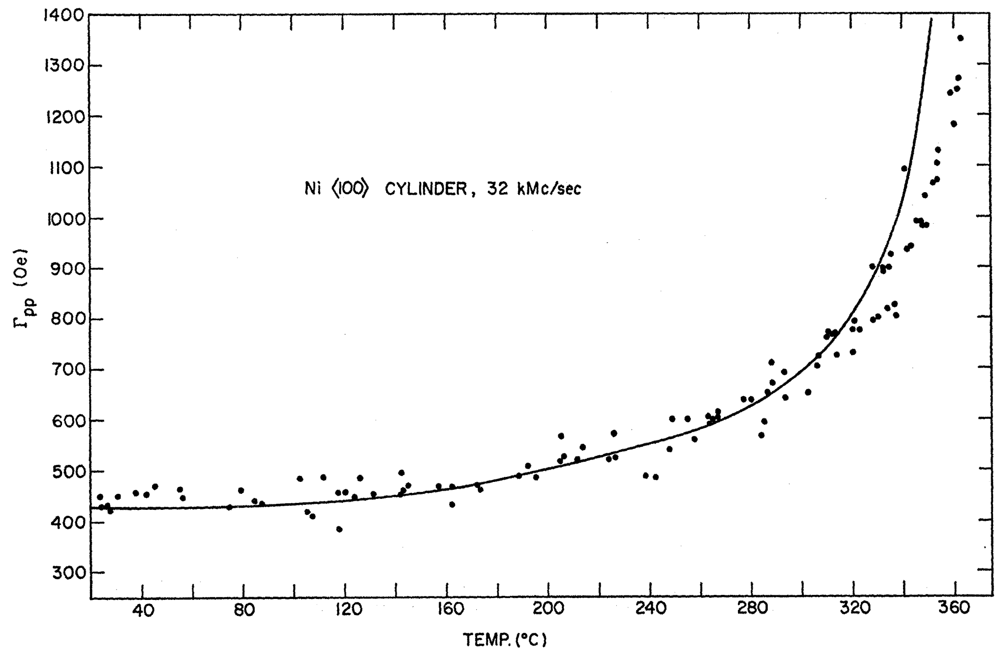

FIG. 3. Variation of observed linewidth with temperature at 31.8 kMc/sec. The full curve follows from Eq. (2), with $\lambda=2.3\times10^8$ sec^{-1} and $g=2.21$.

C. Asymmetry

As mentioned earlier, precise measurements on the resonance became very difficult at high temperatures because of considerable distortion in the line shape. Figure 5 shows a series of recorder traces of the observed signal at high temperatures. As can be seen, the lines become exceedingly asymmetric as the temperature goes above the Curie point (~357°C). An increasing asymmetry is, of course, to be expected because at high temperatures M_s is not large in comparison with H. It is well known that even at room temperature a relaxation-dominated line is not symmetric if one is working at very high frequencies such that H is much larger than M_s. Another way of understanding this phenomenon is to recall that for the parallel-field geometry, the Bloch-Bloembergen relaxation time τ_m is related to λ through the expression $1/\tau_m=\lambda(H/M_s+2\pi)$.

For H and Γ_{pp} much less than M, τ_m is almost constant over the resonance. However, when H is much larger than M_s, and the resonance is rather wide, τ_m will vary as the line is swept out and this will give rise to an asymmetric resonance.

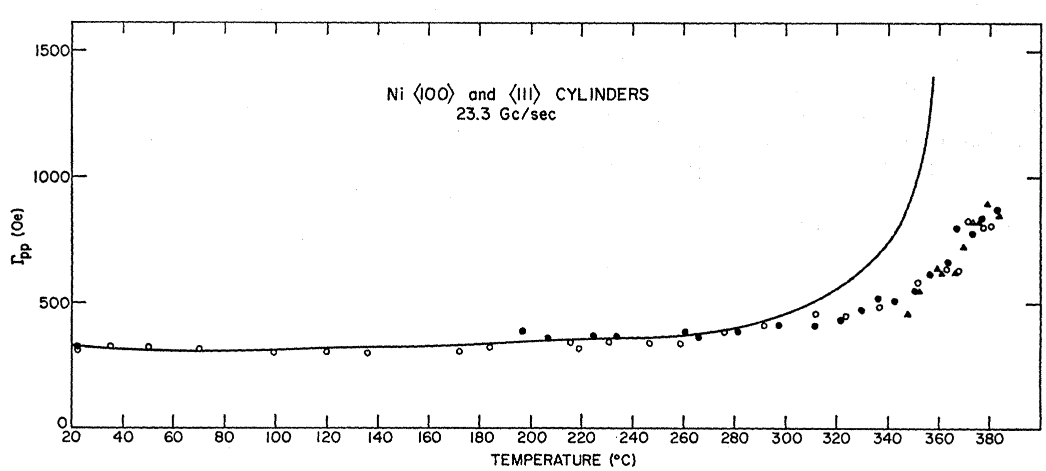

FIG. 4. Variation of observed linewidth with temperature at 23.3 kMc/sec. The curve represents Eq. (2), with $\lambda=2.3\times10^8$ sec^{-1} and $g=2.21$.

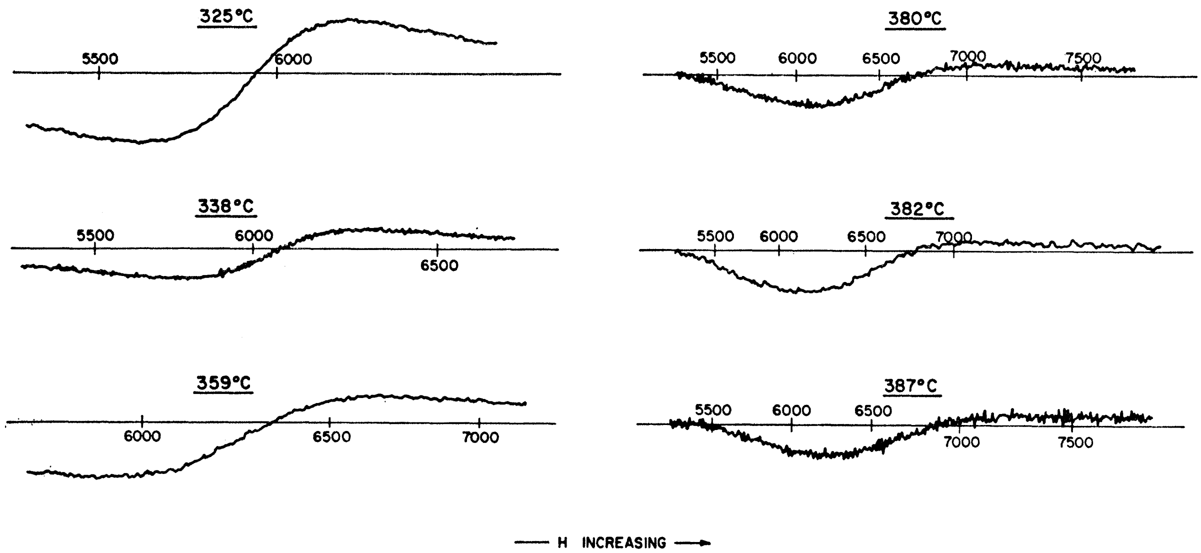

FIG. 5. Recorder traces of the 23.3 kMc/sec resonance signal at high temperatures. The gain of the system had to be continously increased during the rising-temperature sequence.

Recently, Salamon[8] has reported measurements on the magnetic relaxation in nickel at high temperatures (>350°C), using the Kerr rotation technique. He obtained the spin relaxation rate $1/\tau_m$ and the g value by making computer fits to the observed line shapes. His observations were made at 23.3 kMc/sec and the measurements on $1/\tau_m$ can be summarized by the expression

$$1/\tau_m = (4.6\times 10^{12}\ \text{sec}^{-1})(C/T)(H/M),$$

for temperatures between 350 and 380°C, where C is the Curie constant, H is the applied field, and M is the magnetization. In order to compare this result with the present data one has to recall that for his geometry (i.e., magnetic field perpendicular to the sample surface) the Landau-Lifshitz parameter λ is given by $\lambda = (1/\tau_m)(M/H) = (8.2\times 10^{10}/T)\ \text{sec}^{-1}$ (T in °K) when the value for C appropriate to nickel is substituted. Thus at the Curie point he would get a $\lambda = 1.3\times 10^8\ \text{sec}^{-1}$. This result is in qualitative agreement with the present data in that we also find the observed 21 kMc/sec linewidth at about 360°C to be about one-half of that calculated using $\lambda = 2.3\times 10^8\ \text{sec}^{-1}$. Also the reduction in λ with increasing temperature is clearly observed by us. Again, Salamon found a change in g value from 2.22 to 2.29 as he went from below T_c to above it. An increase in the g value at high temperatures is also observed by us but we seem to find no peculiarity near T_c; in fact, for the 21 kMc/sec data this reduction is already evident at temperatures well below T_c. The reason for this discrepancy is not at all clear.

In conclusion, we are led to suggest that (i) the fmr in bulk single crystals of nickel can be described between 20 and ~300°C in terms of parameters which are essentially independent of temperature and frequency, in agreement with the platelet and whisker results of Rodbell.[3] (ii) In order to fit the data at higher temperatures, it is necessary to assume that λ reduces and g increases with temperature and that both of them are somewhat dependent on frequency.

[8] Salamon, Phys. Rev. 155, 24 (1967).

ACKNOWLEDGMENTS

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