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FMR IN NICKEL NEAR THE CURIE TEMPERATURE (*)

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Résumé. — On a mesuré la résonance ferromagnétique à 9,4 GHz sur des échantillons massifs monocristallins de Ni pour 20° < T < 380° C. Jusqu’à 340 °C les mesures expérimentales sont consistantes avec des paramètres qui sont indépendants de la température et de la fréquence. Aux températures plus élevées l’analyse des résultats actuels et des résultats anciens aux fréquences plus élevées montre qu’aucune des équations du mouvement est satisfaisante. On suggère que les effets venant des fluctuations de la magnetisation sont responsables de ce désaccord.

Abstract. — We have measured FMR at 9.4 GHz in bulk single crystal Ni samples for 20° < T < 380° C. Up to 340° C the data are consistent with temperature and frequency independent parameters. At higher temperatures analysis of the present results along with our previous measurements at higher frequencies, shows that none of the equations of motion is adequate. It is suggested that effects arising from fluctuations of the magnetization may be responsible for this failure.

1. Introduction. — Several measurements [1, 2, 3] on FMR in metals show that at room temperature the lineshape can be adequately described by either the Landau-Lifshitz eq. [4]

\[ \dot{M} = \gamma [M \times H] + \frac{\lambda}{M_s^2} [M \times (M \times H)] + \frac{2 \lambda}{M_s^2} [M \times V^2 M], \]  

(1)

or the Gilbert eq. [5]

\[ \dot{M} = \gamma [M \times H] - \frac{\lambda}{\gamma M_s^2} [M \times \dot{M}] + \frac{2 \lambda}{M_s^2} [M \times V^2 M]. \]  

(2)

The most interesting conclusion is that the relaxation parameter \( \lambda \) is independent of frequency over a fairly wide range of frequencies. In pure Ni, measurements on whiskers [1] and bulk samples [6] for

\[ 20° < T < 300° C, \]

show that \( \lambda = 2.3 \times 10^8 \text{ s}^{-1} \) and \( g = 2.21 \), independent of \( T \), the exchange-conductivity contribution being small, especially at high \( T \). At higher temperatures the whisker data at 9 GHz are still consistent with the above values of \( \lambda \) and \( g \). However, bulk sample measurements at 23 GHz and 32 GHz indicated that to explain the results, both \( \lambda \) and \( g \) have to be functions of temperature and frequency. For instance, at 23 GHz the measured linewidth at 360 °C is about one half as large as that calculated using \( \lambda = 2.3 \times 10^8 \text{ s}^{-1} \). On the other hand, Salamon [7] found that the Wangness [8] equation

\[ \dot{M} = \gamma [M \times H] - \frac{1}{\tau} [T_{MO} H - M], \]  

(3)

was appropriate to describe his 23 GHz measurements in Ni at \( T > T_C \), the Curie temperature. However, anticipating the results of the sequel, the values of \( \tau \)

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obtained by him are nearly an order of magnitude shorter than those calculated by us using the linewidths of ref. [6].

In this note we present a new set of FMR measurements on bulk single crystal cylindrical (1 mm diameter \times 1 cm long) samples of pure Ni. The data were taken at 9.4 GHz in the temperature range

\[ 20° < T < 380° C, \]

the highest temperature at which the signal was still observable. The results confirm the conclusions of ref. [6] and show that none of the equations of motion is adequate unless somewhat unphysical assumptions are introduced.

2. Results and Discussion. — Techniques of sample preparation and mounting were similar to those described earlier [3, 6, 9]. For high temperature measurements it is essential to maintain a good vacuum (5 \times 10^{-5} \text{ mm.Hg}) throughout the heating cycle. Otherwise, the linewidths are not repeatable. During a measurement the temperature was stable to about 0.3°. From repeated measurements we conclude that the linewidths (\( T_{MO} \)) are good to about 10 % or \pm 150 Oe. The resonance fields (\( H_0 \)) are repeatable to \pm 50 Oe, the errors being somewhat larger at higher temperatures.

In presenting the data we shall concentrate on the temperature regime above 300° C. Figure 1 shows the measured \( H_0 \) and \( T_{MO} \). First, consider \( H_0 \). Since the exchange terms are small eq. (1) and (2) can be solved easily to calculate the surface impedance. The results are exhibited in figure 1 for \( \lambda = 2.3 \times 10^8 \text{ s}^{-1} \) and \( g = 2.21 \). For the high \( T ( > 350° C) \) calculations it is important to note that \( M_s \) is not independent of \( H \). In our computations the field dependent values of \( M_s \) were taken from Weiss and Forrer [10]. Also included in the figure are the values of \( H_0 ( \text{designated « undamped ») which follow from solution of the Kittel equation (\( \omega_0/\gamma \))^2 = H_0 (H_0 + 4 \pi M_s). \) Here, the values of \( M_s \) appropriate to the observed \( H_0 \) were used. Several points are worthy of note :
FIG. 1. — Resonance field $H_0$ ($\pm$'s and upper curves) and linewidth $\Gamma_{pp}$ (O's and lower curves) as a function of temperature at 9.4 GHz. As described in the text, the curves were computed using eq. (1) and (2) and the Kittel relation $\langle v \rangle^2 = H_0(H_0 + A M_s)$. The data represent measurements on $<100>$ and $<111>$ cylinders.

(a) All the calculations agree with one another for $T < 350^\circ$C. Also, there is good agreement with the data for $T \lesssim 340^\circ$C.

(b) For $T > 350^\circ$C the increase in linewidth (or damping, represented by $\lambda/M$) pulls the resonance field and the overlap between (1) and (2) equations disappears. Since the observed $H_0$ also lie above the undamped values for $T > 370^\circ$C further discussion of equation (1) will be counterproductive.

(c) The apparent $g$-value increases as the temperature is raised.

The linewidths obtained from equation (2) using the above parameters are also shown in figure 1. Clearly above $350^\circ$C the observed lines are much narrower, and the results are not consistent with a temperature independent $\lambda$.

Next, the observed values of $H_0$ and $\Gamma_{pp}$, along with our higher frequency data [6], were used to calculate the values of $g$ and $\lambda$. It was found that both $g$ and $\lambda$ could be functions not only of temperature but also of frequency. In principle, there is no reason to believe that $\lambda$ should be frequency independent. However, a frequency dependent $g$ is clearly unphysical.

The failure of equation (2) prompted us to try equation (3). Once again the surface impedance was calculated to obtain values of $g$ and $\tau$ consistent with the measured values of $H_0$ and $\Gamma_{pp}$ at 9, 23 and 32 GHz. One consequence of this calculation was the discovery mentioned earlier that our values of $\tau$ at 23 GHz were much longer than those of ref. [7]. Unfortunately, again a frequency independent $g$ could not account for the observations.

We therefore conclude that neither the Gilbert nor the Wangsness equation is adequate to describe FMR over an extended range of frequencies for $T \gtrsim 350^\circ$C.

Recently, Leichner [11] calculated the temperature dependence of FMR linewidths near $T_c$ and claimed good agreement with the 32 GHz data of ref. [6]. However, his theory predicts the same temperature dependence for all frequencies in contradiction with experiments. In the analyses attempted so far only cursory attention has been given to the fact that the measurements cover a temperature regime around a phase transition. This has been done by making $M_s$ = a function of $H$. For a given $\lambda$ inclusion of this variation reduces the calculated widths but the observed widths are still much smaller (cf. Fig. 1). In this sense the dynamical equation appears to overestimate the effect of the transition. Perhaps a detailed consideration of spin fluctuations would explain the discrepancy. It is not easy to see how this could lead to narrowing. In principle, fluctuation narrowing may take place but we cannot demonstrate this in detail. However, fluctuations could lead to large shifts in $H_0$ because the terms in $V_2 M$ may become important, the space variation in $M$ being due to fluctuations rather than to the familiar skin effect. This might help to account for the awkward behavior of the apparent $g$-value.

References