Singularities of radio-frequency absorption in nickel near the Curie temperature

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An experimental investigation of radio-wave absorption spectra in nickel near the Curie point (T_c) in a magnetic field, in the frequency range $\omega/2\pi = 1$–25 MHz, is reported. It is shown that resonant spectrum anomalies exist near T_c. Frequency vs field plots are drawn for the absorption peaks at various temperatures. The spectra obtained differ from the known frequency-field dependences for ferromagnetic resonance (FMR). The boundaries of the region in which anomalies exist for the directions [001], [1$\bar{1}$0], and [1$\bar{1}$1] are drawn on the frequency-temperature plane. It is observed that a change of the critical behavior (a crossover) occurs in the vicinity of $\tau \equiv (T - T_c)/T_c \approx -10^{-3}$.

1. INTRODUCTION

Critical dynamics of ferromagnets has been recently attracting much attention. The formulation of the dynamic-scaling hypothesis[1] has contributed to the understanding of the physics of dynamic phenomena near a second-order phase-transition point. Experiments on inelastic scattering of neutrons and on the measurement of high-frequency susceptibility have confirmed the conclusions of the dynamic scaling hypothesis and have shown that a magnetic field influences strongly the dynamic effects in ferromagnets. This holds primarily for experiments on polarized neutrons,[2] measurements of EPR spectra,[3,4] and investigations of the dependences of the high-frequency susceptibility on temperature in a magnetic field.[5–8] Using the dynamic-scaling hypothesis, Lazuta *et al.*[9] developed the most complete theory, which takes into account the influence of a magnetic field on the critical dynamics of a ferromagnet in the exchange ($4\pi\chi \ll 1$, where χ is the static susceptibility) and in the dipole ($4\pi\chi \gg 1$) temperature regions. This theory was further developed and generalized in Maleev's papers.[10–12] The influence of the magnetic field on the behavior of ferromagnets near the Curie point can be estimated from known inequalities. In statics, the field is regarded as weak if the condition[1]

$$g\mu H \ll T_c(\varkappa a)^{(5-\eta)/2} \qquad (1)$$

is met, where g is the Landé factor, μ the Bohr magneton, H the internal field, $\varkappa^{-1} \equiv R_c(\tau) = a|\tau|^{-\nu}$ the correlation radius a length barely shorter than interatomic, and η is the Fisher exponent. If H satisfies Eq. (1), the behavior of the system is determined by the temperature, and the field can be taken into account as a perturbation. In a strong field, i.e., if $g\mu H \gg T_c(\varkappa a)^{(5-\eta)/2}$, it is in fact the field rather than the temperature which determines the behavior of the system. In dynamics, the magnetic field must be compared not with the energy $T_c(\varkappa a)^{(5-\eta)/2}$, of the critical fluctuations, but with the damping Γ. If the Larmor precession $g\mu H$ is comparable with Γ, the field can no longer be regarded as weak.[9]

Among the first studies of the influence of a magnetic field on the critical dynamics were measurements of the temperature dependence of the high-frequency susceptibility in yttrium iron garnet.[7,8] It was shown by these studies that the magnetic field shifts the maximum of the imaginary part χ'' of the susceptibility as a function of temperature. Moreover, if an attempt is made to use the data of Refs. 7 and 8 to plot the dependence of χ'' on the magnetic field, it can be seen that χ'' is at certain temperatures a nonmonotonic function of the magnetic field. This nonmonotonicity can in no way be attributed to experimental error, but it is impossible to determine from the data of Refs. 7 and 8 the field dependence of χ'', since the field was varied discretely in both cited papers. Discrete variation of the magnetic field in experimental studies of the critical dynamics is a practice common to all the cited experiments.

The conclusions of the dynamic-scaling hypothesis were verified for substances with different properties—metals and insulators. However, while the neutron-scattering experiments were performed on substances of both groups, the rf experiments were performed mainly on insulators (see the literature cited in Refs. 10–12). Exceptions are the FMR studies at the frequencies $\omega/2\pi = 10^{10}$–10^{11} Hz.[13,14] At these frequencies the FMR is observed in fields $H \gtrsim 3$ kOe, but in such fields $4\pi\chi \ll 1$ and the dipole temperature region is experimentally inaccessible.

In contrast to an insulator, in a metal it is easy to encounter a situation in which the spatial dispersion plays an important role. In fact, in an insulator the length λ of the electromagnetic wave is the largest parameter with the dimension of length, i.e., the sample is excited by the homogeneous magnetic field of the wave. In a metal, the electromagnetic field is concentrated in a skin layer whose depth $\delta = c/(2\pi\omega\sigma\mu)^{1/2}$, takes on the role of the wavelength (σ is the static conductivity and $\mu = 1 + 4\pi\chi$). Whereas in the paramagnetic phase the condition $\delta \gg R_c$ is well satisfied for frequencies $\omega \leqslant 10^7$–10^8 s^{-1} and for $\tau > 10^{-5}$ (these are estimates for nickel), in the ferromagnetic field there appear additional parameters with the dimension of length, such as the domain-wall thickness, which can be comparable with (or even larger than) δ, i.e., the situation is more varied in metals than in insulators.

2. CHOICE OF THE OBJECT FOR THE INVESTIGATION

The choice of nickel as the investigated object was not fortuitous. This metal is a well-investigated model isotropic (near T_c) ferromagnet with strongly developed fluctuations. It follows from measurements of the magnetization exponent β that the Ginzburg number for nickel is Gi > 0.1. It is precisely if $\tau \approx -0.1$ that the experimental points of the log-log plot of the magnetization vs temperature lie on a

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straight line having a slope 0.35 (Ref. 15). The equations of state of nickel were obtained from experiment and it was shown that the static scaling is quite well satisfied.[16] A check of the dynamic scaling in the exchange region, by means of neutron scattering, was carried out in Ref. 17. The high-frequency experiments, however, were practically all performed in the microwave band, and there are no radio-frequency measurement data analogous to those obtained for insulators. We report here the measured electromagnetic-wave absorption spectra of nickel near T_c under conditions of quasistatic magnetization reversal.

3. EXPERIMENT

The radiowave absorption spectra were measured with an autodyne oscillator and by a simple modulation procedure. This is the usual technique for measuring the spectra of continuous NMR.[18] The high-frequency coil had 15 turns of POZh-brand copper wire. The sample was secured to a copper stage and placed inside the hf coil. This made it possible to rotate the sample and align the crystallographic direction with that of the constant magnetic field. The device for rotating the sample inside the hf coil is shown in Fig. 1. To prevent oxidation of the sample and to decrease the temperature gradients, the heating was carried out in a quartz cell filled with helium. The oven in which the sample was heated was a copper cylinder 20 mm in diameter and ≈ 80 mm long, with wall thickness 1 mm and with a bifilarly wound heating coil.

The temperature was measured with a Pt–Pt 10% Rh thermocouple calibrated against the solidification temperatures of pure In, Cd, and Zn metals. The agreement with the international scale was within $\pm$ 1 K, and the temperature stability in the experiment was $\pm$ 0.01 K. The reversible-magnetization field was produced by an electromagnet whose power supply permitted the magnetic field to vary from $-$ 500 to $+$ 500 Oe for periods from several seconds to

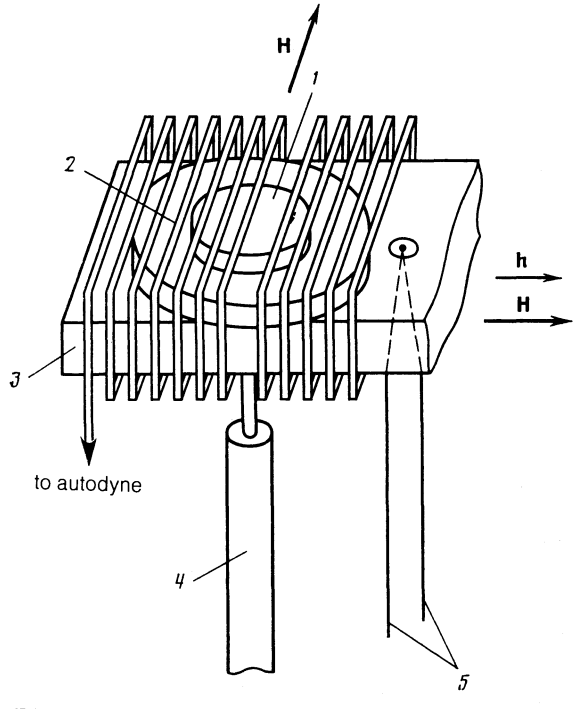

FIG. 1. Measuring cell of rf installation. The mechanism that rotates sample 1 consists of mounting stage 2, bearing plate 3, rod 4, and measuring thermocouple 5.

two hours. The field was measured with a Hall pickup calibrated against a NMR signal. The amplitude of the modulation field, also measured by NMR, was ≈ 0.5 Oe. The hf field amplitude h is estimated not to exceed 0.2 Oe. The last two values met the condition that the magnetic field be weak at $\tau \approx 10^{-4}$ (Ref. 1). The direction of the constant field **H** was always in the sample plane. The relative orientation of the fields **h** and **H** could range from **h**$\parallel$**H** to **h**$\perp$**H** (see Fig. 1).

Let us dwell briefly on the measurement method. It is known[18] that by using self-oscillator methods and a modulation technique it is possible to measure the change of the active losses in an oscillator circuit as the magnetic field is varied. The magnetization of the sample was reversed at constant temperature by a quasistatic magnetic field, and an x-y potentiometer was used to record the derivative of the absorption with respect to the field, dP/dH, as a function of the magnetic field. The magnetization reversal was carried out in both directions.

4. ORIENTING THE SAMPLE IN THE MAGNETIC FIELD

The sample, a nickel single crystal with resistance ratio $R_{300\ \mathrm{K}}/R_{4.2\ \mathrm{K}} \gtrsim 4000$, was a disk 5 mm in diameter and 0.3 mm thick. After mechanical finishing and bright-dipping the sample was annealed at $T = 1200$ K in a vacuum with residual pressure 10^{-5}–10^{-6} Torr for 3 hours. The sample surface was aligned, accurate to 0.5°, with the (110) plane, to which the crystallographic directions [001], [1$\bar{1}$0], and [1$\bar{1}$1] lead. The following simple considerations were used to align the field **H** with a given crystallographic direction. At room temperature the cubic anisotropy constant of nickel is $K_1 < 0$, while $K_2 > 0$. Two orientational transitions exist in a magnetic field.[19] The field strengths at which these transitions are observed depend on the direction of the field **H** at **H**$\parallel$[001] the difference between these fields is at a maximum. Corresponding to each orientational transition are two dP/dH dependences. To observe distinctly these transitions, the sample was first made single-domain by a magnetic field, after which the field was gradually decreased to zero. Figure 2 shows plots of dP/dH as the field is decreased, for different angles between **H** and the [001] direction. At **H**$\parallel$[001] the field difference between the peaks is a maximum (see Fig. 2). The accuracy of alignment with a given direction in the experiment was $\pm$ 2°.

5. CHOICE OF CURIE TEMPERATURE

The choice of T_c plays an important role in the reduction of the experimental results. The published data cannot be used for this purpose. Thus, according to various sources, T_c of nickel fluctuates in a range 10 K, from 626 to 636 K. Naturally, these values of T_c cannot be used to describe the phenomena at $|\tau| < 10^{-2}$. The choice of T_c in a given experiment was based on a simple consideration supported by the most general properties of the high-temperature susceptibility. In weak magnetic fields that meet the condition $g\mu H \ll \Gamma$, the power absorbed is $P \propto H^2$ (Ref. 11). As $T \to T_c$, the region of fields for which $dP/dH \propto H$ contracts to zero, i.e., the width of the absorption line is a minimum at $t = T_c$ (see Fig. 3b and Fig. 5 below). It should be noted that the reasoning presented above is valid also when the scaling hypothesis is not valid. Neglecting hysteresis in the field H, the absorbed power P is an even function of the field, and in a

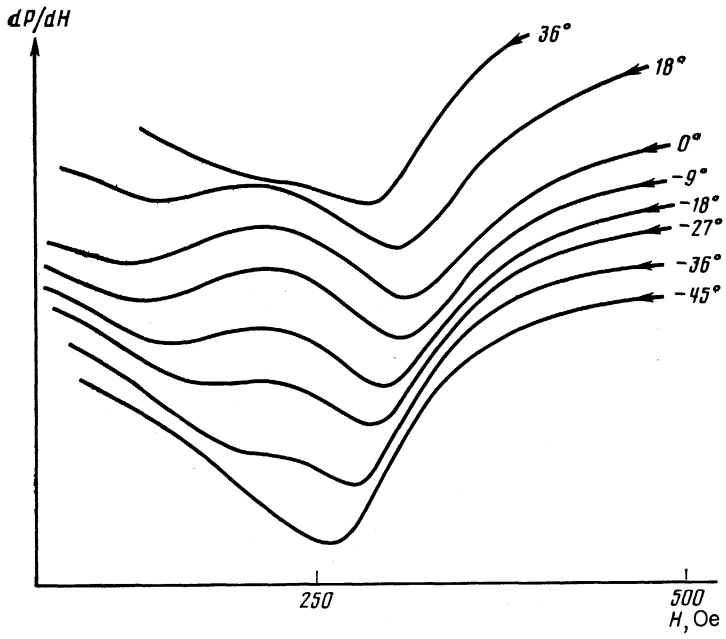

FIG. 2. Typical experimental curves used to orient the sample. The numbers on the plots are the values of the angle between H||h and the [001] direction.

weak field we have $P \propto H^2$. In our experiment the error in the determination of T_c from the minimum line width was ± 0.05 K ($T_c \approx 629$ K). The accuracy of T_c is limited by the instability of the temperature and by field hysteresis phenomena (see below).

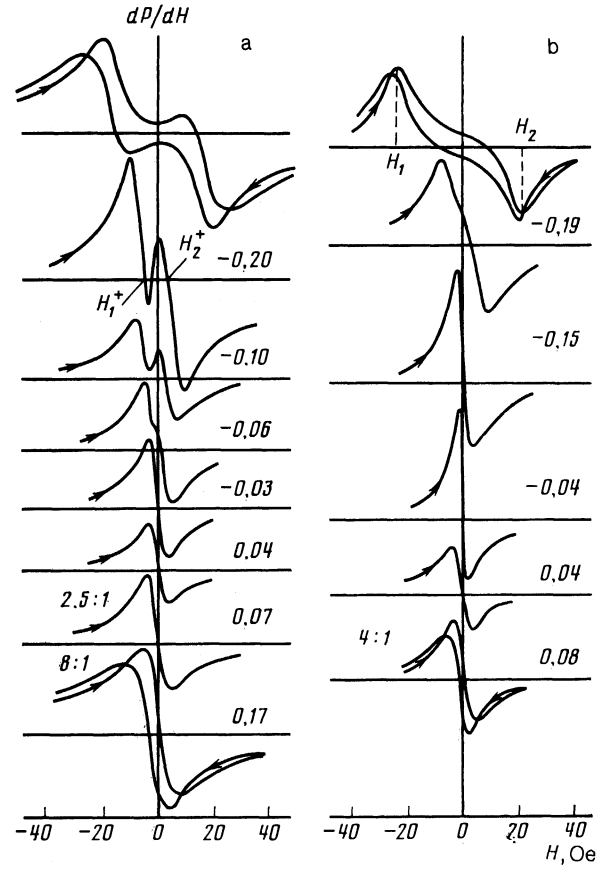

FIG. 3. Experimental plots of dP/dH for the $\mathbf{h}\perp\mathbf{H}\|[001]$ orientation at $T = T_c - 0.37$ K (a) and $\mathbf{h}\|\mathbf{H}\|[001]$ at $T = T_c - 0.54$ K (b); $\omega/2\pi = 5.4$ MHz.

6. RESULTS AND DISCUSSION

As already mentioned, we measured in the experiment the absorption derivative dP/dH on remagnetization of the sample by the field H. The experimental plots of the absorption line in a geometry $\mathbf{h}\perp\mathbf{H}\|[001]$ (a) and $\mathbf{h}\|\mathbf{H}[001]$ (b) at the frequency $\omega/2\pi = 5.4$ MHz for different temperatures is shown in Fig. 3. It must be noted first that the absorption line shows field hysteresis not only in the ferromagnetic phase but also in the paramagnetic one. In the paramagnetic region the hysteresis was observed up to $\tau \approx 2 \cdot 10^{-3}$. Further rise of temperature decreases the absorption-line intensity, and the temperature region $\tau > 2 \cdot 10^{-3}$ was not investigated in the present study. The causes of the hysteresis at $\tau > 0$ in a nickel sample of sufficiently high quality are not clear. Similar hysteresis phenomena were observed in the paramagnetic phases of $CdCr_2S_4$ and $CdCr_2Se_4$ (Ref. 20). No distinguishable temperature hysteresis was noted.

As seen from Fig. 3a, in the $\mathbf{h}\perp\mathbf{H}$ geometry the absorption line is strongly distorted, i.e., the $P(H)$ dependence is nonmonotonic. It was noted that distortions (singularities) of the line exist in a definite frequency and temperature region, and the boundaries of the singularity region were subsequently plotted with the frequency and temperature as the coordinates. To this end, we have determined at a fixed temperature the minimum frequency at which the absorption line is still distorted. The presence of singularities can be attributed to the existence of additional rf-power absorption from certain oscillations excited by the electromagnetic wave in the sample. The presence of the singularity boundary indicates that the spectrum of these oscillations has a gap. Figure 4a shows the boundaries of the singularity region for the directions [001], [1$\bar{1}$0], and [1$\bar{1}$1]. It is seen from Fig. 4a that the gap depends strongly on the crystallographic direction at $T < T_m$. The gap is a minimum for the [001] direction at $T < T_m$ (the easy axis in this temperature range and increases very rapidly at $T > T_m$.

It is possible to determine in experiment with sufficient accuracy (± 1 Oe) the fields at which of the absorption

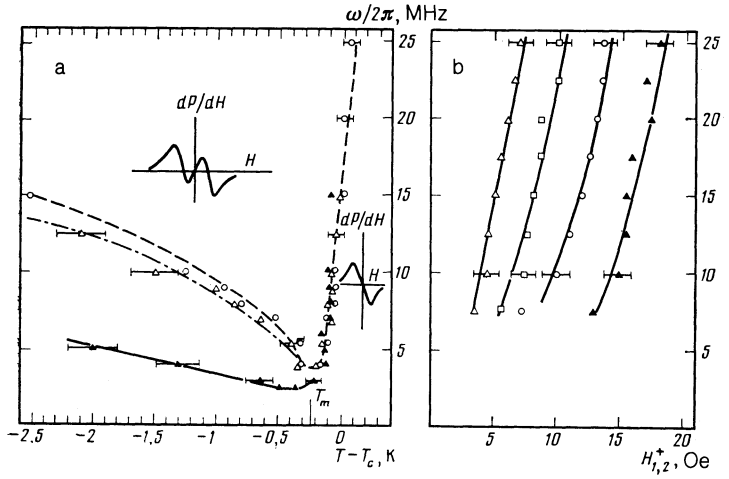

FIG. 4. a) Boundaries of the region of existence of resonant singularities for the directions [001] (▲), [110] (△) and [111] (○). b) Frequency-field dependences of the absorption extrema for different temperatures at $\mathbf{H}\|[001]$: ○,△—H_1^+, ▲,□—H_2^+; ○,▲—$T = T_c - 0.36$ K, △,□—$T = T_c - 0.23$ K.

lines and the zeroes of dP/dH have maxima. These fields are designated H_1^+ and H_2^+ and their values depend on the excitation frequency. Characteristic frequency vs field plots for the $\mathbf{h}\perp\mathbf{H}\|$ [001] orientation are shown in Fig. 4b. These plots are of similar form for the $[1\bar{1}0]$ and $[\bar{1}\bar{1}1]$ directions. It follows from the plots of Fig. 4b that the resonance frequency ω is an increasing function of the resonant field H at constant values of τ and ω in the limit as $H\to 0$ and $\tau\to 0$. This pair of facts contradicts the usual behavior of the FMR spectra.[13,14] It was shown in an experiment with polarized neutrons[21] that magnetic inhomogeneities exist in nickel near T_c and for $\tau<0$ and the singularities of the absorption spectrum can be apparently attributed to oscillations of these inhomogeneities.

There are no line distortions in this frequency band in the $\mathbf{h}\|\mathbf{H}$ geometry (see Fig. 3b). The most pronounced feature of the spectrum is the temperature dependence of the field positions of the absorption-line inflection points. They are labeled H_1 and H_2 in Fig. 3b. The form of this dependence is illustrated in Fig. 5. It can be seen from the figure that the fields H_1 and H_2 are unequal (hysteresis). After determining T_c by the method described above, the temperature dependences of H_1 and H_2 were plotted in log-log scale. They are shown for the $\mathbf{h}\|\mathbf{H}\|[110]$ orientation in Fig. 5. It can be seen that $H_{1,2}\propto|\tau|^{\beta'}$, and a change of the critical behavior (apparently a dipole crossover) takes place in the vicinity of $\tau\approx -1.2\cdot10^{-3}$. A theoretical estimate of the crossover temperature for nickel, $|\tau_d|\approx 3.9\cdot10^{-3}$, is given in Ref. 11. The measurement temperature in this experiment differed somewhat from the theoretical estimate, and to ascertain the nature of the interactions that cause this crossover it is necessary to carry out additional experiments at $|\tau|<10^{-3}$.

The critical exponents β' for H_1 and H_2, determined by least squares, are listed in Table I.

It can be seen from the table that the exponents β' differ

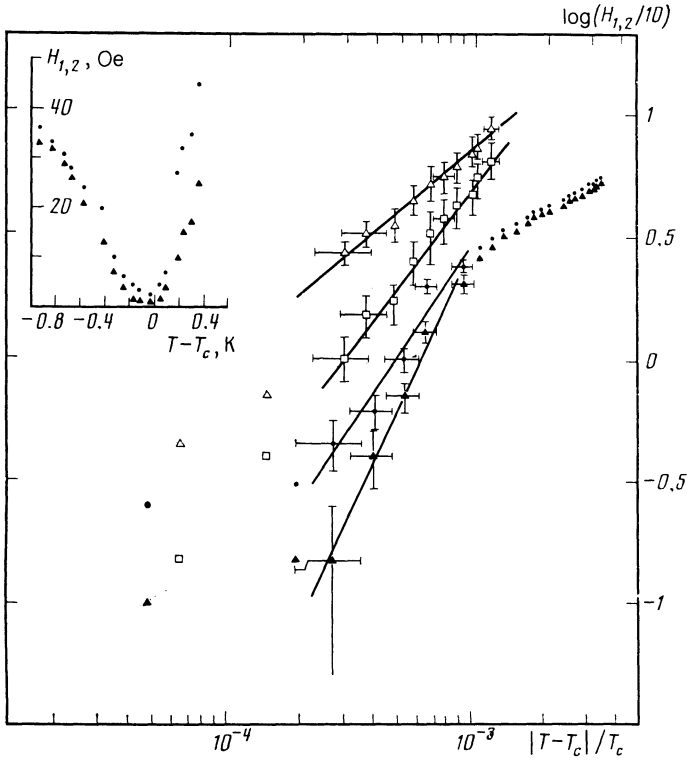

FIG. 5. Temperature dependences of H_1 and H_2 for $\mathbf{H}\|\mathbf{h}\|[110]$: ▲,□—$H_1$; ●,△—$H_2$; ▲,●—$T<T_c$; □,△—$T>T_c$. Inset: temperature dependences of H_1 and H_2 in linear scale.

TABLE I.

	β′					
	[100]		[1$\bar{1}$0]		[1$\bar{1}\bar{1}$]	
	H_1	H_2	H_1	H_2	H_1	H_2
$-4\cdot10^{-3}<\tau<-10^{-3}$	0.53±0.02	0.53±0.02	0.54±0.02	0.53±0.02	0.63±0.02	0.57±0.02
$-10^{-3}<\tau<-2\cdot10^{-4}$	$2.5^{+1.0}_{-0.5}$	1.9±0.3	$2.2^{+0.7}_{-0.2}$	$1.5^{+0.1}_{-0.4}$	$1.7^{+0.3}_{-0.4}$	$1.0^{+0.6}_{-0.1}$
$10^{-4}<\tau<10^{-3}$	$0.66^{+0.10}_{-0.15}$	0.52±0.15	$1.30^{+0.20}_{-0.30}$	0.85±0.15	$1.23^{+0.20}_{-0.30}$	$0.70^{+0.40}_{-0.25}$

for $\tau<0$ and $\tau>0$. This is not surprising, since nickel has at $\tau<0$ magnetic inhomogeneities[21] and a domain structure, and therefore behaves differently at $\tau<0$ and $\tau>0$.

The foregoing experimental results show that in nickel near T_c the $P(H)$ dependence for the $\mathbf{h}\|\mathbf{H}$ orientation is a monotonic function of H. $P(H)$ becomes nonmonotonic in the $\mathbf{h}\perp\mathbf{H}$ geometry, when the electromagnetic wave excites certain oscillations in the sample. These oscillations exist in the ferromagnetic phase in a definite frequency and temperature region. The form of the frequency versus field plots of these oscillations differs from the known FMR spectra. As for the temperature dependences of H_1 and H_2, it would be desirable to recalculate them in terms of the internal field. To this end it is necessary to know the dependence of the static susceptibility on the temperature and on the magnetic field. It is impossible to use for this purpose the equations of state,[16] since they do not describe phenomena with field hysteresis and have been determined from experiment at $|\tau|>4\cdot10^{-3}$. Their extrapolation to the region $|\tau|<10^{-3}$ is doubtful. It can be noted that measurement of χ at temperatures $|\tau|<10^{-3}$ is difficult, for in the case of a (nickel) sample of finite size χ is not proportional to $|\tau|^{-\gamma}$ in this temperature region, but is determined by the demagnetizing factor. The type of the observed crossover can be identified only from additional experiments performed at $\tau<10^{-3}$. No such experiments are known at present. In addition, actual experiments have shown that, at least in the ferromagnetic phase, nickel in anisotropic near T_c and caution must be exercised when using it as a model isotropic ferromagnet.

To describe the foregoing results theoretically it is necessary to obtain an expression for the high-frequency susceptibility of a ferromagnet of finite size not only in the limits of strong and weak magnetic fields, but also in intermediate magnetic fields. The author knows of no such theory.

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