

leading to a structure change. Furthermore, as a resistivity anomaly has been observed in Nb_3Sn at 36°K , this could lead one to suppose that the martensitic transformation is characteristic of most superconducting A-15 type compounds.

In conclusion, it has been shown that a resistivity jump occurs at approximately the temperature where the martensitic transformation was observed by x rays. This anomaly was observed in several A-15 type superconducting compounds and a relationship seems to exist between the martensitic transformation and superconductivity. One must, however, keep in mind that cer-

tain factors could affect superconductivity without affecting the phase transformation.

I would like to thank E. S. Greiner for the V_3Si single crystals, W. H. Haemmerle for his help in obtaining the data, and D. Dorsi for preparing some of the compounds.

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²These films have been fully discussed in the following two papers: J. J. Hauser, Phys. Rev. Letters **9**, 423 (1962); J. J. Hauser and H. C. Theuerer, Phys. Rev. **129**, 103 (1963).

FERROMAGNETIC RESONANCE ABSORPTION LINEWIDTH OF NICKEL METAL. EVIDENCE FOR LANDAU-LIFSHITZ DAMPING

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This Letter describes the observation of the narrowest resonance absorption linewidth yet reported for metallic nickel single crystals at 9 kMc/sec. In addition to this observation, an analysis, based upon the temperature and frequency dependence of the resonance, shows that the "exchange conductivity" linewidth mechanism of Ament and Rado¹ is operative in the samples of this study although not dominant as in the case of iron "whiskers" previously reported.² In the present study it is found that a damping mechanism is required and although the physical origin of this linewidth contribution is unclear, it seems to be accurately characterized by the Landau-Lifshitz³ phenomenological relaxation frequency, λ , which enters the macroscopic torque equation as

$$d\vec{M}/dt = \gamma\vec{M} \times \vec{H} - (\lambda/M^2)\vec{M} \times (\vec{M} \times \vec{H}). \quad (1)$$

This is a remarkable result since the form of the term comes simply from the expansion of $d\vec{M}/dt$ into the three orthogonal coordinates: \vec{M} , $\vec{M} \times \vec{H}$, and $\vec{M} \times (\vec{M} \times \vec{H})$. The first term vanishes with the requirement that M be a constant of the motion, the second gives the resonance condition, and the third describes the breadth of the resonance line in the absence of exchange effects.

The samples used in these experiments were very small (e.g., a millimeter in extent and a micron thick) single crystals of nickel [both of "whisker" (filament) and platelet geometry]. These samples were grown by the hydrogen re-

duction of nickel bromide and very generously supplied by Dr. R. W. DeBlois of this Laboratory. The samples are first selected for optical qualities, i.e., specular surfaces with no obvious imperfections, and are then examined at 9.2 kMc/sec at 25°C and selected for sharpness of resonance absorption. From a few hundred considered visually, about 50 samples were chosen and of these about 25 had linewidths under 150 oersteds (i.e., the field separation between inflection points of the absorbed microwave power vs magnetic field; both rf and dc magnetic fields lie in the sample surface). Most of the selected group had widths of 130 oersteds but the narrowest of the group had a 114-oersted linewidth, this being roughly 25% of the width usually observed for bulk metallic nickel at this frequency and temperature. The resonance lines here reported have a characteristic asymmetry that serves as a "thumb print" to indicate the presence of the inhomogeneous broadening that results from the limited penetration of the rf fields by the metallic conductivity, coupled with the variation in the penetration depth as the resonance is traversed. The latter effect results from the field-dependent permeability, i.e., the resonance itself. If, for a moment, we neglect damping, this exchange-conductivity mechanism predicts for nickel at 9.2 kMc/sec, 25°C , and using an exchange stiffness, A , of 1×10^{-6} erg/cm a linewidth of 37 oersteds for unpinned surface spins. This value would be increased to 90 oersteds if

the surface spins were completely immobilized.^{4,5} We reject this spin-pinning source of broadening as inconsistent with several experimental results (one of which is discussed below), but primarily rest its rejection on the temperature dependence of the linewidth at elevated temperatures. In Fig. 1 are displayed some linewidth data, normalized to room temperature, for several of the samples examined. The linewidth is seen to increase for decreasing temperature as expected on the exchange conductivity basis (No. 5 platelet is an exception discussed below). However, instead of decreasing for increased temperatures as this mechanism predicts, the linewidth remains essentially constant and then rises rapidly near the Curie temperature. Several likely mechanisms for linewidth are drawn on the same plot and the exchange-conductivity mechanism is seen to be a monotonic decreasing function of increasing temperature (proportional to magnetization/resistivity^{1/2}). Surface spin-pinning linewidth contribution would also follow this dependence. The mechanism of Kittel and Mitchell⁶ of the conduction electron spin interaction with the resonance-excited spin waves may be brought to the approximate form indicated by ρ/M (i.e., resistivity/magnetization) and is seen to be a more rapidly increasing function of temperature than the observed data. Lastly a Landau-Lifshitz λ term is plotted, its temperature dependence being M^{-1} for λ a constant. This func-

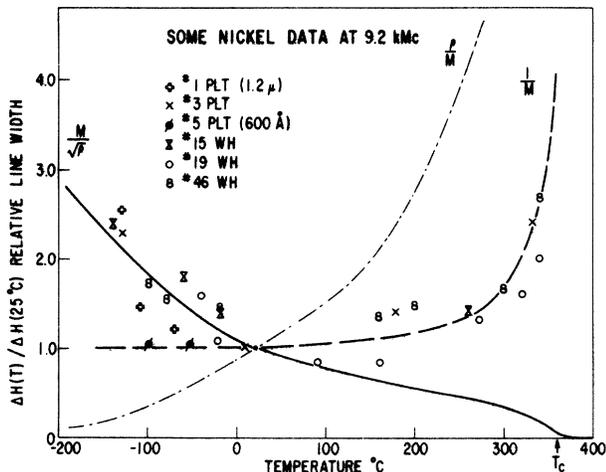


FIG. 1. The temperature dependence of the absorption linewidth of some of the samples studied. Both platelet and whisker samples are included with one platelet (No. 5) that is much thinner than the penetration depth of the rf exciting fields. All the data are normalized at 24°C.

tional behavior is true not only for insulators but also for metals as determined by calculation (below) and is seen to be a reasonable description of the observed high-temperature linewidth. By numerical solution of the Ament-Rado equation, including λ but excluding pinning, we find it possible to express $\Delta H_{\epsilon\lambda}$, the observed linewidth resulting from exchange-conductivity and damping, as the sum $\Delta H_{\epsilon\lambda} = \Delta H_{\epsilon 0} + 0.82\Delta H_{0\lambda}$, where $\Delta H_{\epsilon 0}$ is the linewidth for "exchange only" and $\Delta H_{0\lambda} = 3\lambda\omega/2\gamma^2M$ is the linewidth for "damping only." The value 0.82 is valid in the range $0 \leq \Omega L/\epsilon \leq 1$ and deviates to higher values beyond that range ($\Omega \equiv \omega/4\pi M\gamma$; $L \equiv \lambda/M\gamma$; $\epsilon^2 \equiv A/2\pi M^2\delta^2$; $\delta^2 \equiv \rho c^2/2\pi\omega$; the defining symbols have their usual meaning). The calculation of the actual line shape shows that as the damping increases the line broadens, shifts, and becomes less asymmetric, the asymmetry, the shift, and the width being unique functions of $\Omega L/\epsilon$; i.e., the effective damping-to-exchange ratio.

As noted earlier, No. 5 platelet shows a temperature-independent linewidth. This platelet is quite interesting since it is only 600 Å thick and should be essentially immune to the rf penetration problem although it should show marked effects from surface pinning. On the latter point it behaves exactly as much thicker samples with regard to the field necessary for resonance and also has approximately the same linewidth. If surface pinning were effective it should be easily noticed in the line position (at least 100 oersteds more shift than for the thicker samples). This platelet does show a symmetric resonance absorption whose linewidth is independent of temperature suggesting that this sample may be characterized by $\Delta H_{0\lambda}$ only, as would be expected for a situation of complete rf penetration. The calculation for such a situation is displayed in Fig. 2 in comparison to the observed line. The value of $\lambda = 2.5 \times 10^8$ cps was used. Subsequent resonance studies with this sample were precluded by its removal for x-ray orientation determinations.

As further evidence for the support of the Landau-Lifshitz damping, let us consider the frequency dependence of the linewidth. The Ament-Rado linewidth depends on frequency as $\omega^{1/2}$; the Kittel-Mitchell mechanism as ω^{-1} ; and the Landau-Lifshitz contribution as ω . A sample (No. 10 whisker) was examined at 25°C both at 9.2 kMc/sec and at 34.9 kMc/sec,⁷ and found to have 135- and 410-oersted linewidths, respectively. The $\omega^{1/2}$ and the ω^{-1} predictions clearly fail while

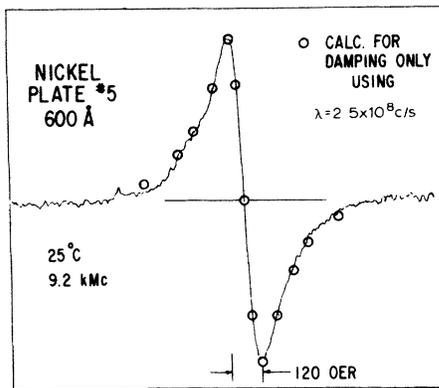


FIG. 2. The first derivative with respect to the steady magnetic field as a function of the steady magnetic field for a "thin" single-crystal platelet of nickel (No. 5, 600 Å thick, {100}). The circles are calculated for λ only using 2.5×10^8 cps for the relaxation frequency. The absorption line is seen to be symmetric as it should be in this case.

the use of Eq. (2) with $\lambda = 2.2 \times 10^8$ cps and $A = 1.0 \times 10^{-6}$ erg/cm predicts linewidths of 120 and 410 oersteds, respectively. The observed line asymmetry is also closely accounted for. Both observed and calculated lines at 9.2 kMc/sec and 34.9 kMc/sec are given in Fig. 3. The observations were both at 25°C. The agreement is quite striking.

It is also germane that the exchange shift associated with the exchange conductivity mechanism is, in magnitude, a large fraction of the concomitant linewidth (70% for $\lambda = 0$) but decreases with increased damping. By inclusion of the exchange shift appropriate to the "exchange conductivity" influenced resonance absorption and with the determination of the magnetocrystal-line anisotropy constants of nickel from our data, we find a temperature- and frequency-independent g value for nickel metal of 2.22 ± 0.03 from -140°C through 360°C , the Curie point.

Thus there seems to be strong evidence for the presence of a relaxation term of the Landau-Lifshitz form in nickel metal with $\lambda \sim 2.5 \times 10^8$ cps for the selected samples of this study. This value appears to depend upon the sample and its treatment since it increases when these samples are damaged (not an infrequent occurrence). The fact that such a term was not earlier identified in the case of iron whiskers² may be due to the higher magnetization and higher exchange stiffness of that material, both reducing the effectiveness of such a relaxation term relative to the pure exchange-conductivity broadened reso-

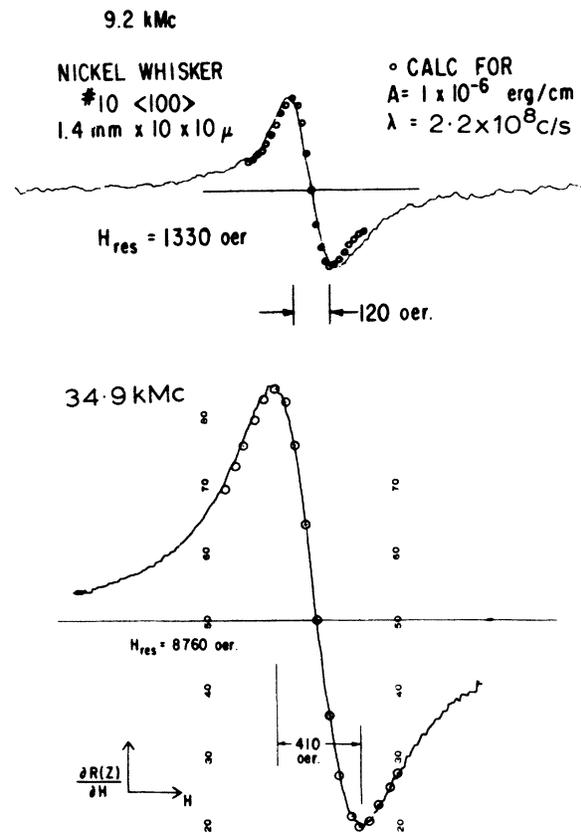


FIG. 3. The first derivative with respect to the steady magnetic field of the real part of the surface impedance as a function of the steady magnetic field of nickel "whisker" crystal No. 10 at 25°C for 9.2 and 34.9 kMc/sec. The line has an asymmetry and width due to exchange and conductivity effects with damping. Thick platelets of nickel behave in the same way. The calculated points are determined using an exchange stiffness, A , of 1×10^{-6} erg/cm and a Landau-Lifshitz "frequency" λ , of 2.2×10^8 cps.

nance line. The effect of $\lambda = 2.5 \times 10^8$ cps on iron is to broaden the "exchange conductivity" linewidth, and that would be increased by 50% whereas for nickel this would result in a 300% increase. It is thus not inconsistent that such a damping term magnitude is also appropriate to iron whiskers and in fact such a term would help to explain the high-temperature linewidth behavior of iron whisker crystals previously reported.² The question as to the physical origin of this Landau-Lifshitz damping term must be answered, since there seems here to be the first clear evidence for its existence in the form originally prescribed by Landau and Lifshitz. The present observations suggest it to be structure sensitive and probably related to imperfections.

It is a pleasure to acknowledge here my gratitude to R. W. DeBlois for his very generous and skilled ability to supply the samples for this study. I thank G. Mahan and T. Moore for helping me to use a G. E. 225 Computer that performed some of the data analysis. I also thank Z. Frait for calling my attention to an error in the numerical evaluation of one of the parameters in an earlier draft.

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⁷I am grateful to T. W. Moore for providing his equipment for this measurement.

NATURE OF THE CRYSTALLINE SOLID AT ELEVATED TEMPERATURES

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We wish to report on some calculations performed in connection with the theory of melting developed by one of us (R.B.).¹ These calculations have not yet been completed to the point where they give the melting-point curve. We feel, however, that the results on the solid state are of sufficient interest to be made available immediately. Our findings indicate that considerable modification of the qualitative physical picture of a molecular solid is necessary when the ratio of the temperature to the Debye temperature satisfies $(T/\theta_D) \geq 0.4$.

A brief resumé of the theory given in I, as modified for this calculation, follows. The basic self-consistent equation of I is

$$\rho(r) = \frac{\exp[-\beta \int v(\vec{r}-\vec{r}') g_{\text{HC}}(\vec{r}-\vec{r}') \rho(\vec{r}') d^3r']}{\int \exp[-\beta \int v(\vec{r}-\vec{r}') g_{\text{HC}}(\vec{r}-\vec{r}') \rho(\vec{r}') d^3r'] d^3r}. \quad (1)$$

Here $\rho(r)$ is the single-particle periodic density,

$v(\vec{r}-\vec{r}')$ is the attractive part of the intermolecular potential, $\beta = (1/\kappa T)$, and $g_{\text{HC}}(\vec{r})$ is the hard-core exclusion factor given by

$$g_{\text{HC}}(r) = 1, \quad r > c, \\ = 0, \quad r < c, \quad (2)$$

where c is the hard-core diameter. Any periodic function may be written as

$$\rho(\vec{r}) = \sum_{\vec{R}i} \varphi(\vec{r}-\vec{R}i), \quad (3)$$

where $\vec{R}i$ are the lattice points. We assume that the solid is such that the particles are localized within their unit cells, and hence that in the i th cell only the i th member of the sum in Eq. (3) contributes significantly to $\rho(r)$. In that case, Eq. (1) reads

$$\varphi(\vec{r}) = \exp\left[-\sum_{R \neq 0} \beta \int r(\vec{r}-\vec{r}') \varphi(\vec{r}-\vec{R}) d^3r'\right] \left\{ \int_{\text{unit cell}} \exp\left[-\sum_{R \neq 0} \beta \int r(\vec{r}-\vec{r}') \varphi(\vec{r}'-\vec{R}) d^3r'\right] d^3r \right\}^{-1}. \quad (4)$$

The free energy (see I) is given by

$$-\frac{\beta F}{N} = -\int \varphi(\vec{r}) \ln \varphi(\vec{r}) d^3r - \left(\frac{\beta}{Z}\right) \sum_{R \neq 0} \int v(\vec{r}-\vec{r}') g_{\text{HC}}(\vec{r}-\vec{r}') \varphi(\vec{r}) \varphi(\vec{r}'-\vec{R}) d^3r d^3r'. \quad (5)$$

The equilibrium distance, R_0 , is determined as a function of T by minimizing F with respect to R_0 . The external pressure is assumed negligible. To facilitate calculation we have replaced the summation over the 12 near neighbors by an integration over a spherical shell.