

Temperature variation of ferromagnetic relaxation in the 3d transition metals*

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We report measurements on the ferromagnetic resonance in single-crystal specimens of hcp Co, Ni, and Fe. The linewidth data have been analyzed to obtain the temperature dependence of the ferromagnetic relaxation frequency λ . These temperature dependences can be qualitatively understood in terms of recent theories. On the other hand, it turns out that in some cases λ is essentially constant over wide ranges of temperature. This behavior is unexplained at present.

I. INTRODUCTION

During the last decade, it has become well established¹ that the ferromagnetic resonance (fmr) in the 3d transition metals and their alloys can be adequately described in terms of the Landau-Lifshitz-Gilbert equation

$$\frac{d\vec{M}}{dt} = \gamma(\vec{M} \times \vec{H}) + \frac{2A\gamma}{M_s^2}(\vec{M} \times \nabla^2 \vec{M}) - \frac{\lambda}{\gamma M_s^2} \left(\vec{M} \times \frac{d\vec{M}}{dt} \right), \quad (1)$$

where $\gamma = ge/2mc$, M_s is the saturation magnetization, A is the exchange stiffness, and λ is a microscopic relaxation parameter. For comparison with experiments, Eq. (1) has to be solved along with Maxwell's equations, the constitutive relations between currents and fields, and boundary conditions $\partial \vec{m} / \partial \vec{n} = (K_s/A)\vec{m}$, where \vec{m} is the dynamic magnetization, K_s is the so-called surface anisotropy, and the gradient is to be taken along the outward normal to the sample surface. It turns out that in the linewidth, there is a significant contribution from the so-called exchange-conductivity mechanism, in addition to the contribution from the λ term. The prescriptions for the calculations were worked out, among others, by Ament and Rado^{2,3} (for the normal conductivity regime) and by Hirst and Prange⁴ (for the anomalous skin-effect regime). It is generally agreed that near 300 K, $\lambda \approx 10^8 \text{ sec}^{-1}$ for Fe, Ni, hcp Co, permalloy, and Ni-Cu alloys. On the other hand, a quantitative calculation of this parameter from a microscopic theory is not available at present.

In 1966, Bhagat and Hirst⁵ reported that in order to account for the observed linewidths in pure Ni for $T \leq 100$ K, it was necessary to make λ increase rapidly as the temperature was lowered. For $4 < T \leq 77$ K they had very few data, but the apparent λ was essentially temperature independent between 4 K and about 50 K. In addition, for $100 \leq T \leq 600$ K it has been found that λ is essentially temperature independent.⁶⁻⁹ Furthermore, it is interesting to note that for a Ni-5-wt%-Cu alloy there is no observable variation in λ for $4 < T < 300$ K.¹⁰

In Fe it has been found that the observed linewidths are consistent with a λ which is essentially

independent of T for $4 < T < 300$ K.^{11,12} However, it should be noted that all the measurements were made at frequencies (< 35 GHz) for which the exchange contribution to the linewidth was so large that no truly definitive temperature dependence for λ could have been deduced. This is especially true at low temperatures because the exchange term increases rapidly with increasing dc conductivity. On the other hand, for $300 < T < 900$ K there appears to be some evidence for a very slow increase in λ .^{13,14}

In this paper we present fmr measurements on: (i) single-crystal whiskers of hcp Co at several frequencies near 300 K and as a function of temperature between 4 K and about 700 K at 60 GHz (brief descriptions of this work have been presented recently^{15,16}); (ii) Single-crystal whiskers of Fe at 70 GHz for $4 < T < 300$ K (at 70 GHz the λ contribution to the linewidth amounts to roughly 30% at 300 K; thus one can put a somewhat more definitive upper limit on the temperature variation of λ); (iii) Single-crystal bulk samples of highly annealed Ni at 22 GHz for $4 < T < 300$ K (the main aim of these investigations was to determine more precisely the temperature dependence between 4 and 100 K).

The results of the experiments may be summarized as follows: (i) Both Ni and hcp Co exhibit a rapid increase in λ as T is lowered below about 100 K. However, in Ni the λ appears to become temperature independent below about 35 K. While no such saturation effect is obvious in Co, it has to be admitted that with the present precision it is not possible to ascertain if saturation occurs below ~ 20 K. (ii) The λ for Fe is essentially independent (to within a factor of 2) of T for $4 < T < 300$ K. (iii) The Co λ increases slowly as T is increased from 100 to 700 K. Our results will be compared, at least qualitatively, with some of the recent theoretical predictions.

II. EXPERIMENTAL

A. Measurement techniques

The fmr measurements were made using the well-known field-modulation technique using the

spectrometers described earlier.^{11,17} The temperatures were measured with a copper-constantan thermocouple attached to the outside of the microwave cavity in the vicinity of the sample. The thermocouple was calibrated against a Pt-resistance thermometer. For low-temperature work the samples were maintained in a helium atmosphere of about 10^{-2} Torr and the temperature was stabilized using a heater wound on the exterior of the cavity. We estimate that our temperature measurements are good to about 2–3 K for $T < 60$ K and about 1 K for higher T . For $T > 300$ K the sample was kept in a vacuum of about 10^{-5} Torr. The vacuum chamber was surrounded by a cylindrical heater, as described in our earlier papers.^{11,17}

B. Sample preparation

The hcp Co whiskers were grown by the hydrogen reduction of $\text{CoBr}_2 \cdot 6\text{H}_2\text{O}$,¹⁸ under conditions essentially similar to those described by Marker *et al.*¹⁹ A covered boat containing the salt was placed in a long quartz tube which, in turn, was inside a cylindrical oven. The following factors were found to be essential to the formation of hcp whiskers with their long axes along the [0001] direction: (i) temperatures of 680–700 °C, as measured by a thermocouple located in the center of the oven; (ii) hydrogen flow rates of ~ 2 cm³/min (for a 22-mm-i. d. reaction tube) mixed with He gas at a flow rate of 50–200 cm³/min; (iii) temperature gradients of about 50 °C/cm in the region of the reaction boat; and (iv) quartz or vycor chips scattered on top of the CoBr_2 , which had been thoroughly dried by first maintaining it in a moderate vacuum and then preheating it (in a He atmosphere) *in situ* to a temperature of 300 °C. Typically the whiskers were hexagonal in cross section, 2–20 μm thick and a few mm long. The resistivity was measured as a function of temperature for use in subsequent calculations. We found $\rho_{300}/\rho_{4.2}$ values in the range of 80–150. These values are somewhat lower than those reported by Marker *et al.*¹⁹

The [100] Fe-whisker specimens (~ 40 μm thick, ~ 5 mm long) were grown by the hydrogen reduction of $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ using the methods described earlier.¹² The Ni cylindrical (0.5-mm diam \times 8 mm long) samples were spark-cut from a single-crystal boule (nominal purity 99.997%), and electropolished and annealed in a high vacuum ($\sim 10^{-6}$ Torr) at temperatures of more than 1400 °C for several hours. After annealing, the samples were given a light electropolish and were then electroplated with a thin layer of Cu, leaving only the central $\frac{1}{3}$ exposed.¹¹

C. Sample mounting

The hcp Co samples were mounted on small pieces of glass, fused quartz or single-crys-

tal quartz. The single-crystal quartz was chosen so as to match the coefficients of thermal expansion of the whisker and the substrate. This was found to be particularly important for the high- T work. The sample was held on the substrate using very slight amounts of silicone oil, vacuum grease, or RTV-102 adhesive. In any case, only those data were retained for subsequent analysis where the linewidth was unaltered for several thermal cycles. The reproducibility of the linewidth for a variety of adhesives and/or substrates is taken to be a strong indication that our linewidth data are not significantly affected by induced inhomogeneous strains. On the other hand, it has to be admitted that with change of mounting, the line shape is not as repeatable as the linewidth. It should be noted that the fmr signals in hcp Co are often so strong that they give rise to spurious and pathological line shapes. To avoid the problems arising from this, first, the cavities were made very large (e. g., at 60 GHz the microwave cavities were effectively 50 wavelengths long). Second, we took observations at several frequencies lying close by to establish that the linewidth was not being affected. Taking into account measurements on several samples as well as several measurements on the same sample, we note that our hcp Co linewidths are good to about 10%, while the so-called asymmetry¹⁵ $\alpha = (S_l - 1)/S_h$ varies from nearly 0 to 0.3.

The Fe-whisker and Ni-sample mountings were essentially the same as reported in our earlier papers.^{11,12,17} The linewidths are determined to about the same precision as in hcp Co; however, the line shapes are more reproducible.

III. RESULTS AND INTERPRETATION: LINEWIDTHS

A. Hcp Co

As described in Refs. 15 and 16, we first made measurements near 300 K at frequencies of about 37, 60, and 135 GHz. In Table I we have listed the observed peak-to-peak widths Γ_{pp} . The 60- and 135-GHz linewidths correspond to room temperature; however, for an applied field along the c -axis, the anisotropy field is so large that the 37-GHz data had to be taken at $T \approx 350$ K. To our knowledge, none of the relevant parameters varies sufficiently rapidly with T for this difference to be significant.

TABLE I. fmr Linewidths in [0001] hcp Co whiskers near 300 K.

Frequency (GHz)	Linewidth (Oe)	
	Observed	Computed
37	155	157
60	205	205
135	340	365

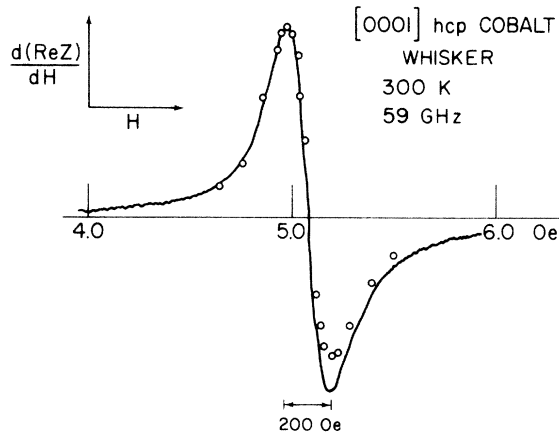


FIG. 1. Recorder trace of the field derivative of the microwave absorption in a [001] single-crystal whisker of hcp Co. The points were derived from the theory using the parameters given in the text in connection with Table I. The calculated $d(\text{Re}Z)/dH$ was normalized to the signal height at the low-field peak.

The calculated linewidths in Table I were obtained using the well-known methods^{2,3} outlined in Sec. I with the following values for the parameters:

$$g = 2.18, \quad \lambda = 1 \times 10^8 \text{ sec}^{-1},$$

$$K_s = 0, \quad A = 2.78 \times 10^{-6} \text{ erg/cm}^2,$$

$$4\pi M_s = 17900 \text{ Oe}, \quad \rho = 10.6 \mu\Omega \text{ cm}.$$

The following points should be noted: (i) For a uniaxial system the second term on the right-hand side of Eq. (1) must be replaced by

$$\frac{2\gamma A_{\perp}}{M_s^2} (\vec{M} \times \nabla_{\perp}^2 \vec{M}) + \frac{2\gamma A_{\parallel}}{M_s^2} (\vec{M} \times \nabla_{\parallel}^2 \vec{M}).$$

In the geometry appropriate to our experiment, only the first term in this expression will contribute. Our value for $A \equiv A_{\perp}$ has been taken from neutron-scattering data.^{20,21} (ii) The resistivity used here is appropriate to currents flowing along the symmetry axis. (iii) In principle, one could choose K_s values as large as 0.2 erg/cm^2 (with a concomitant reduction in λ to $0.7 \times 10^8 \text{ sec}^{-1}$) to fit the data in Table I within the limits of error stated above.

In Fig. 1 we have compared a typical observed line shape with the calculations. As mentioned earlier, the asymmetry is not very repeatable, and thus it is not surprising that the comparison between theory and experiment is far from satisfactory. In fact, the experimental asymmetries cannot be used to fix any of the parameters.

In Fig. 2 we present the observed linewidths as a function of temperature at 60 GHz. The data represent several samples. The full line was calculated as follows: (ii) For $T \geq 250 \text{ K}$ the normal conductivity theory^{2,3} was used. The temperature

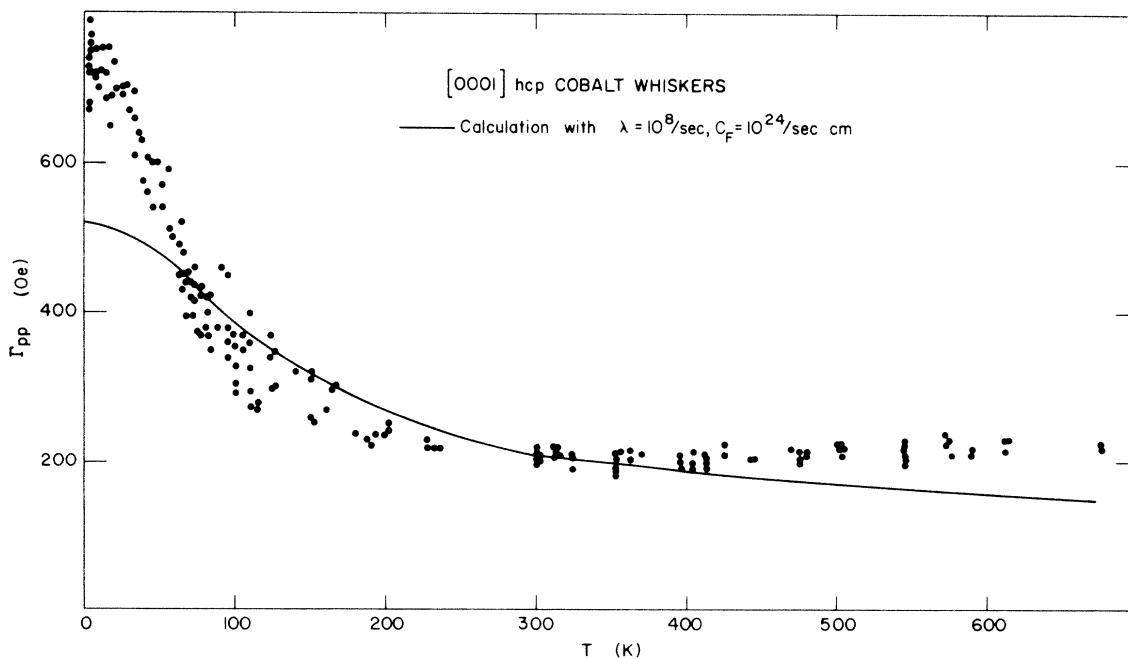


FIG. 2. Temperature variation of the peak-to-peak linewidth in [0001] whiskers of hcp Co at about 60 GHz. The full line was derived from the theory as described in the text.

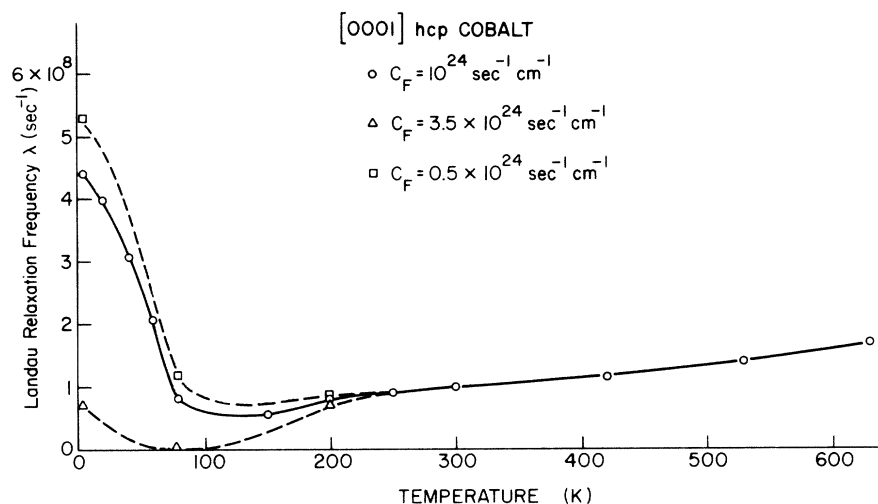


FIG. 3. Temperature variation of the ferromagnetic relaxation frequency required to account for the discrepancy noted in Fig. 2. The three values of C_F were chosen to show the sensitivity of λ to this parameter at low T . It should be noted that these curves give $3.5 \times 10^{24} \text{ cm}^{-1} \text{ sec}^{-1}$ as an upper limit for C_F .

variation of M_s was taken from Honda and Masumoto.²² A was assumed to have the temperature dependence $[M_s(T)/M_s(300)]^2$, and the dc resistivity values due to Schulze²³ are used, confirmed, to some extent, by our observations. (ii) For $T \lesssim 250$ K the nonlocal conductivity theory⁴ was necessary. The dc resistivities were taken from our measurements on a few representative samples. In addition to the parameters mentioned above, the nonlocality brings into play a parameter C_F which is used to describe the wave-number-dependent conductivity. Unfortunately, in the absence of detailed information on the band structure of hcp Co, there is no *a priori* way to arrive at a definitive value for C_F . In the later discussion we will show how fmr can be used to put an upper limit on C_F . By comparison with our earlier determination from fmr measurements in Fe,^{11,12} we chose for C_F the value $10^{24} \text{ cm}^{-1} \text{ sec}^{-1}$ to motivate the discussion. Incidentally, this value is not inconsistent with C_F values that one would obtain by looking at the coefficient of the linear term in the specific heat of Co at low T .¹²

In order to account for the discrepancies evident in Fig. 2, it is essential to allow either C_F , λ , or K_s to be a function of temperature. Since C_F is a lumped parameter representing the band structure, it is difficult to see how C_F could vary with T . If we make K_s vary with T , holding λ constant, we have to let K_s become exceedingly large at $T \sim 600$ K to account for the discrepancy. Presumably, K_s is determined by the state of the sample surface and there is good reason to believe that severe oxidation could lead to higher K_s values. However, there is no way to make this process reversible in our measurements. In this connection, it may be useful to recall that in our earlier measurements of fmr in Fe and Ni at high T , any deterioration in the vacuum led to irreversible increases in the linewidth.

Figure 3 shows the temperature variation of λ required to account for the discrepancy observed in Fig. 2. Here we have used three different values of C_F . Clearly, C_F cannot be greater than $3.5 \times 10^{24} \text{ cm}^{-1} \text{ sec}^{-1}$. In any case, it is evident that to account for the observed linewidths, we need λ to reduce rapidly between 4 K and about 100 K, to go through a shallow minimum, and subsequently to rise slowly to a value of about $1.7 \times 10^8 \text{ sec}^{-1}$ at 650 K.

B. Fe

Figure 4 shows the observed linewidths in Fe [100] whiskers at 70 GHz as a function of temperature. The full curve is obtained from the theory in the same way as for hcp Co. The values of the parameters used were $C_F = 1.5 \times 10^{24} \text{ cm}^{-1} \text{ sec}^{-1}$,

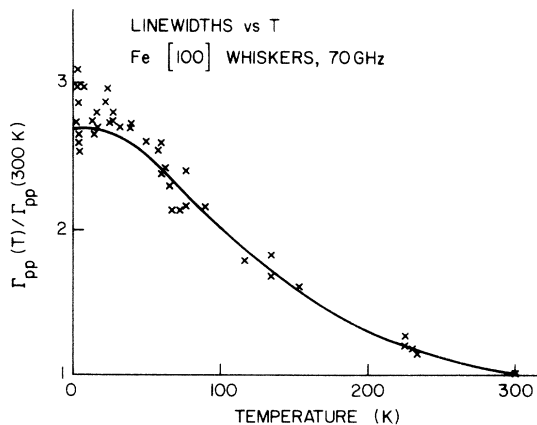


FIG. 4. Temperature variation of fmr linewidth in Fe [100] whiskers at 70 GHz. The full line was derived from theory as explained in the text.

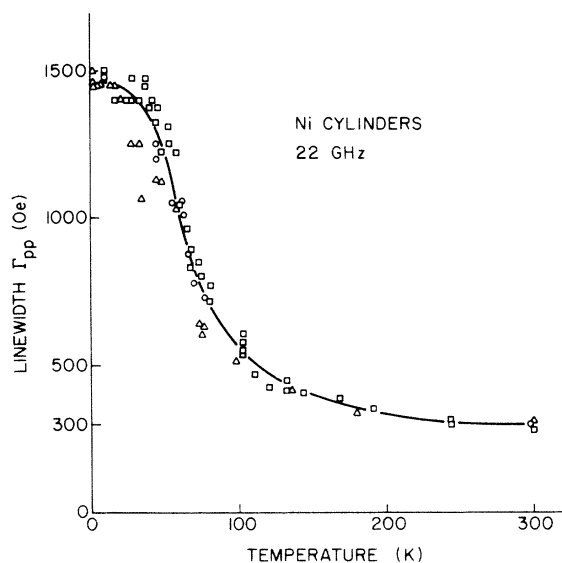


FIG. 5. Temperature variation of fmr linewidth in single-crystal cylinder samples of Ni at 22 GHz.

$A = 1.9 \times 10^{-6}$ erg/cm, $\lambda = 0.7 \times 10^8$ sec $^{-1}$, and $K_s = 0.1$ erg/cm 2 , the variation of the dc conductivity being taken from our earlier data¹² on such whiskers. It is clear that the data do not require any significant temperature variation in λ from 4 to 300 K. Considering that the λ contribution to Γ_{pp} is roughly 30% at 300 K, one can conclude that λ varies by less than a factor of 2 over this temperature range.

C. Ni

At room temperature we first measured the linewidths at 9 GHz to establish that our samples were, indeed, well annealed. Typically, we found $\Gamma_{pp} = 130$ Oe, which is very comparable to the whisker widths of Rodbell,¹⁶ although somewhat larger than the narrowest widths reported by Anders *et al.*⁹

Figure 5 shows the linewidths observed at 22 GHz as a function of T for samples whose resistivity ratio $\rho_{300}/\rho_{4.2}$ varied between 60 and 170. These data essentially confirm our earlier work on pure Ni; they cannot be described by a temperature-independent λ for $T \lesssim 150$ K. The detailed temperature dependence of λ deduced from the present data, following the analysis given in Ref. 4, is shown in Fig. 6. The other parameters relevant to this calculation are $A = 0.75 \times 10^{-6}$ erg/cm, $K_s = 0.1$ erg/cm 2 , and $g = 2.2$, with $4\pi M_s$ values taken from Pugh and Argyle.²⁴ The dc resistivity required for the computation was measured on samples which had been heat treated in a similar fashion.

We have used a value of $C_F = 1 \times 10^{24}$ cm $^{-1}$ sec $^{-1}$. In the case of Ni, most of the linewidth comes from the λ contribution, and the deduced temperature

variation in λ is very insensitive to the choice of C_F . It should also be noted that the observed linewidth at 4 K cannot be explained even if $K_s \rightarrow \infty$.

IV. DISCUSSION

In Fig. 7 we have collected all the existing data on the temperature dependence of the relaxation parameter λ . In addition to our data, we have included the results of measurements on (i) Ni by Rodbell ($130 < T < 600$ K)⁶ and by Anders *et al.* ($77 < T < 600$ K)⁹; (ii) Ni-Cu alloys by Lloyd and Bhagat ($4 < T < 300$ K)¹⁰; and (iii) Fe by Frait *et al.* ($300 < T < 900$ K)¹³ and Bhagat *et al.* ($4 < T < 300$ K).¹⁴

We suggest, with Lloyd and Bhagat, that the low- T behavior in Ni, Ni-0.17-wt%-Cu, and hcp Co is related to the rapid variation in the electronic mean free path. It is then very easy to see why there is no temperature dependence observable in the Ni-5-wt%-Cu alloy ($\rho_{300}/\rho_{4.2} = 3$).

The only other parameter which varies significantly at low T is the magnetic anisotropy. However, (i) in hcp Co (see Fig. 8) most of the temperature variation of K_1/M_s occurs for $T \geq 150$ K, while most of the increase in λ occurs at $T < 100$ K; (ii) whereas λ is essentially constant, Ni-5-wt%-Cu alloys have a sizable variation in K_1/M_s at low T .

It could be argued that the sharp increase in Γ_{pp} at low T be attributed to effects of inhomogeneous strain. However, as noted above, several different types of mounting yield essentially the same linewidth. Furthermore, it is well known that in these metals most of the thermal contraction has

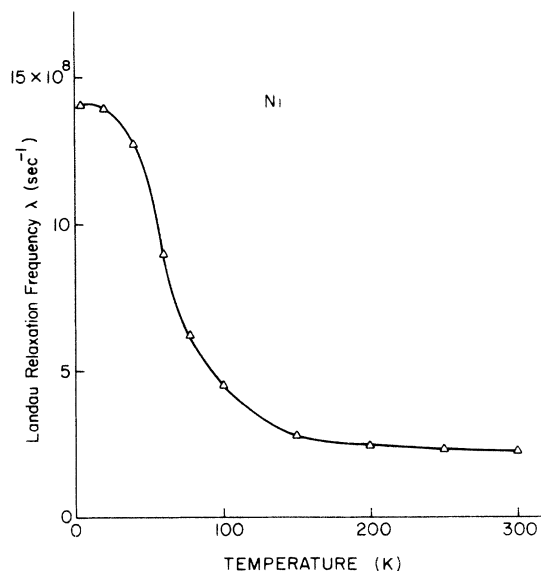


FIG. 6. Temperature variation of the ferromagnetic relaxation frequency λ required to account for the linewidths in pure Ni (Fig. 5). These values were obtained using the analysis given in Ref. 4.

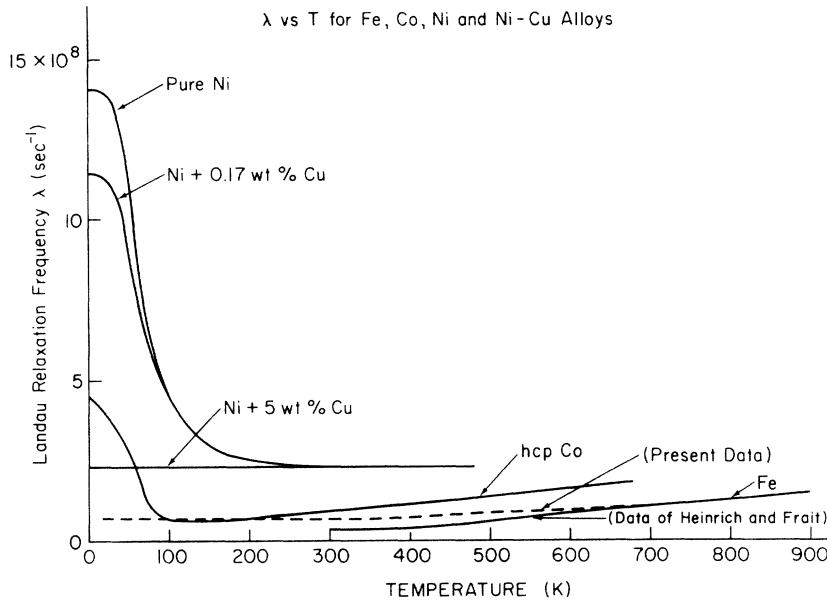


FIG. 7. Collected data on the observed temperature variations of the ferromagnetic relaxation frequency in $3d$ transition metals and alloys. In addition to our data, we have also included here the results of Refs. 6, 9, 10, 13 and 14.

taken place for $T > 77$ K.

Recently, Kambersky²⁵ proposed a model for spin relaxation in ferromagnetic conductors in which explicit account is taken of the spin-orbit coupling within a band picture. The main idea is that this coupling mixes the up and down spin bands, and relaxation can then occur either via electron scattering by phonons or via a "flow" of carriers from or to regions in which the Fermi surface is locally deformed due to the presence of a spin wave. Korenman and Prange²⁶ have worked out the details of the latter process, taking into account the influences of the wave-number-dependent conductivity at low T . They conclude that if there exist sizable regions in the band structure where the "predominantly up" and "predominantly down" spin bands are separated by the spin-orbit energy, one should expect the kind of low- T behavior observed in Ni. In other words, λ should increase with reducing T and exhibit a saturation effect when the mean free path becomes very long at sufficiently low T . The absence of an obvious saturation in our hcp Co data is somewhat surprising, because the hcp Co and the pure Ni samples have comparable resistivity ratios. Finally, while this model gives a reasonable qualitative description, a quantitative fit to the data must await further calculations, especially after more details of the band structure of these materials become available.

The absence of a large increase in λ in Fe at low T could be due to one of two reasons: (i) there are no favorable regions in the bands, or (ii) the spin-orbit coupling is intrinsically smaller. The latter is indicated by the fact that $g-2$ for Fe is smaller than that of Ni and hcp Co by nearly a factor of 2.

Also, it is well known that the magnetocrystalline anisotropy (which is another manifestation of the spin-orbit coupling) in Fe is much smaller than that in Ni or hcp Co. Furthermore, recent de Haas-Van Alphen data suggest that the spin-orbit parameter in Fe²⁷ is about 0.0015 Ry as compared to a value of 0.0075 Ry for Ni.²⁸

The slow increase in λ at higher T , i.e., $100 < T < 700$ K for hcp Co and $300 < T < 900$ K for Fe, is probably related to the phonon-assisted spin-orbit

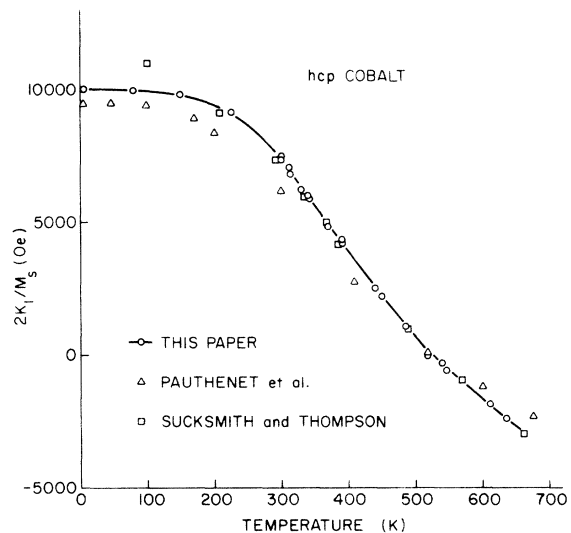


FIG. 8. Observed variation of the first anisotropy constant of hcp Co as a function of temperature. For comparison we have also included the data of Refs. 30 and 31.

effect discussed by Kambersky.²⁵ Again, a direct quantitative comparison with the theory is not possible at present.

Finally, it should be pointed out that at present there exists no theory to account even qualitatively for the temperature-independent part of λ shown in Fig. 7, although a preliminary attempt has been made recently by Kambersky.²⁹

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APPENDIX

We have used the resonant-field data for hcp Co

to determine the temperature dependence of the first-order anisotropy constant K_1 . In Fig. 8 we have plotted $2K_1/M_s$ as a function of temperature. These data were obtained using the parameters listed in the text in connection with Fig. 2. The low- T values given in Fig. 8 are somewhat dependent on the choice of C_F , since the latter controls the amount of "exchange shift" in the line center. With the range of C_F values given in Fig. 3, $2K_1/M_s$ will show a range of about 300 Oe at 4 K. It should be noted, though, that since the low- T linewidths are very large, the precision is not very good. However, in comparing our results with the earlier data of Sucksmith and Thompson,³⁰ we find excellent agreement for $T > 200$ K. On the other hand, the data of Pauthenet *et al.*³¹ (which, incidentally, were taken on Sucksmith's samples) differ significantly from our observations.

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