

\*Work supported by the U. S. Army Research Office (Durham).

†Present address: Sandia Laboratories, Albuquerque, N. M. 87115.

‡Present address: Department of Physics, New York University, New York, N. Y. 10003.

§Alfred P. Sloan Fellow.

<sup>1</sup>N. J. Trappeniers and Th. J. van der Molen, *Physica* (Utrecht) **32**, 1161 (1966); N. J. Trappeniers and W. Mandema, *Physica* (Utrecht) **32**, 1170 (1966).

<sup>2</sup>C. W. Garland and B. B. Weiner, *Phys. Rev. B* **3**, 1634 (1971); B. B. Weiner and C. Garland, to be published.

lished.

<sup>3</sup>L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Addison-Wesley, Reading, Mass., 1969), Chap. XIV.

<sup>4</sup>V. L. Ginzburg and A. P. Levanyuk, *J. Phys. Chem. Solids* **6**, 51 (1958); V. L. Ginzburg, *Usp. Fiz. Nauk* **77**, 621 (1962) [*Sov. Phys. Uspekhi* **5**, 649 (1963)].

<sup>5</sup>C. W. Garland and R. Renard, *J. Chem. Phys.* **44**, 1130 (1966); P. D. Lazay, Ph.D. thesis, Massachusetts Institute of Technology, 1968 (unpublished).

<sup>6</sup>G. A. Samara, in *Advances in High Pressure Research*, edited by R. S. Bradley (Academic, New York, 1969), Vol. 3, Chap. 3.

## Spin-Wave Resonance Fields, Linewidths, and Intensities of a Permalloy Film: Theory and Experiment

G. C. Bailey and C. Vittoria

*Naval Research Laboratory, Washington, D. C. 20390*

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Using only the surface-anisotropy constants and the saturation magnetization as adjustable parameters, we have found good agreement between an observed and a calculated spin-wave resonance spectrum. A uniform magnetization, effects of exchange, conductivity, and Landau-Lifshitz damping are included in the theory.

The purpose of this paper is to show that the observed spin-wave resonance spectrum of a thin metal film can be fitted quite well by an extension of a theory<sup>1</sup> which takes into account four factors: exchange, electrical conductivity, an asymmetric anisotropy at the two surfaces of the film, and an isotropic Landau-Lifshitz damping constant  $\lambda$ . We present, for the first time, *quantitative* theoretical and experimental comparisons of all three important parameters observed in a standing spin-wave resonance spectrum: resonance field positions, line intensities, and linewidths. The results of this study, along with others,<sup>2</sup> suggest that, if care is taken in preparing the film, many of the contributions to the resonance spectrum as a result of inhomogeneities in the sample may be eliminated. Previous papers on this subject were concerned with studying the intensities of the resonance lines either alone or in combination with the positions.<sup>3-6</sup> No reports to date have been given of a consideration of all of the resonance parameters (including linewidths) simultaneously.

Most of the details of the film deposition and microwave techniques have been described previously.<sup>7</sup> In preparing the film, substrates were polished optically smooth and thoroughly cleaned, and the deposition was carried out only after the melt had come to thermal equilibrium with

its vapor. A rather fast evaporation rate of 38 Å/sec was used assuring that the grains of the film were randomly spaced and not columnar across the film thickness.<sup>8</sup> During evaporation, the pressure ranged from  $3 \times 10^{-8}$  Torr to a maximum of  $8 \times 10^{-7}$  Torr. The oven, which maintained a substrate temperature of 250°C during deposition, was turned off 5 min after the deposition was completed.

It was necessary to extend previous magnetic-resonance calculations<sup>1</sup> in order to account for realistic boundary conditions applicable to evaporated Permalloy films. The theory permits an asymmetrical rf excitation of the film for the following reason. Although the film is placed at the center of a rectangular cavity, any error in placement from the cavity center would result in an asymmetric rf excitation. In addition, the calculation permits asymmetrical pinning conditions, between the extreme limits of complete pinning to no pinning at all, at the two surfaces of the film. There may be several reasons why asymmetrical pinning occurs in actual films. One side of the film is rigidly attached to a substrate while the other side is free. In addition, the first few layers of the film structure, deposited at the beginning of the evaporation, may be quite different from the last few layers, deposited at the end of the evaporation. In both

cases, a difference in pinning conditions at the two surfaces would exist.

We can write the boundary conditions at the two surfaces of the film for the case of the applied field normal to the film. The first two equations which follow are simply statements of the fact that the tangential component of the rf magnetic field is continuous across the film surfaces; the last two equations describe the pinning conditions of the rf magnetization at the surfaces<sup>9</sup>:

$$\sum_{i=1}^2 [A_i \exp(-jk_i d) + B_i \exp(jk_i d)] = h_d, \quad (1)$$

$$\sum_{i=1}^2 (A_i + B_i) = h_0, \quad (2)$$

$$\sum_{i=1}^2 Q_i \left\{ \frac{K_s^{(d)}}{2A} [A_i \exp(-jk_i d) + B_i \exp(jk_i d)] + jk_i [A_i \exp(-jk_i d) - B_i \exp(jk_i d)] \right\} = 0, \quad (3)$$

$$\sum_{i=1}^2 Q_i \frac{K_s^{(0)}}{2A} (A_i + B_i) + jk_i (A_i - B_i) = 0. \quad (4)$$

The quantities  $k_1$  and  $k_2$  are the propagation constants which give rise to magnetic, circularly polarized field solutions. The propagation constants can be obtained from the secular equation of Ref. 1.  $A_i$  and  $B_i$  are the four unknowns ( $i=1, 2$ ) and represent the field strength of the resonant, circularly polarized waves;  $h_d$  and  $h_0$  are the surface rf magnetic fields at the two surfaces;  $K_s^{(d)}$  and  $K_s^{(0)}$  are the surface anisotropy constants at the two surfaces.  $A$  is the exchange-stiffness constant and  $Q_i = 1 - \frac{1}{2}j\delta^2 k_i^2$ , where  $\delta$  is the classical skin depth,  $\delta = c/(2\pi\sigma\omega)^{1/2}$ ;  $d$  is the film thickness. The surface impedances  $Z^{(d)}$  and  $Z^{(0)}$  at the two surfaces are given by

$$Z^{(0)} = \frac{c}{4\pi\sigma} j \sum_{i=1}^2 \frac{k_i}{h_0} (A_i - B_i),$$

$$Z^{(d)} = \frac{c}{4\pi\sigma} j \sum_{i=1}^2 \frac{k_i}{h_d} [A_i \exp(-jk_i d) - B_i \exp(jk_i d)].$$

If  $K_s^{(0)} = K_s^{(d)}$ , then we find that  $Z^{(0)} = Z^{(d)}$ . Analytical solutions for  $Z^{(0)}$  have been given previously<sup>10</sup> for a Gilbert form of damping.<sup>11</sup> However, we find that if  $K_s^{(0)} \neq K_s^{(d)}$ , then  $Z^{(0)} \neq Z^{(d)}$ . For either case the power absorbed<sup>12</sup> by the sample is given as

$$P = Z^{(d)} |h_d|^2 + Z^{(0)} |h_0|^2.$$

Analytical solutions of  $Z^{(0)}$  and  $Z^{(d)}$  are prohibitive to employ since the algebra becomes very involved. Our procedure was to calculate nu-

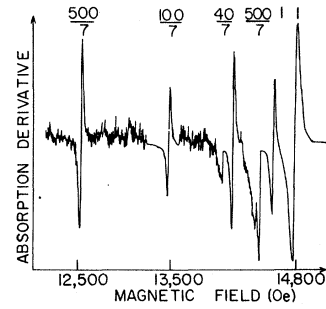


FIG. 1. Spin-wave resonance absorption derivative for the film with the dc magnetic field perpendicular to the plane of the film. The numbers above the lines are the gain settings used.

merically (by computer) the resonance spectrum for various values of  $h_d/h_0$ ,  $K_s^{(d)}$ , and  $K_s^{(0)}$ . The resonance field is defined in a conventional way as the field for which  $dP/dH_a = 0$ , where  $H_a$  is the applied field and the linewidth is obtained from the peak-to-peak width of the resonance line. The intensity of each mode is normalized with respect to the main line and corrected as described below.

Figure 1 shows the spin-wave spectrum of the 2700-Å-thick film (75% Ni-25% Fe) obtained at 9.44 GHz and room temperature. The implications of this spectrum concerning the quality of the film have been discussed before.<sup>7</sup> Table I shows the fit between theory and experiment for the resonance fields, linewidths, and intensities of the various lines. In obtaining the fit, we used the values of  $\lambda = 7.5 \times 10^7$  Hz and  $\sigma = 0.7 \times 10^5 \Omega^{-1} \text{cm}^{-1}$  obtained from previous studies of the linewidth-film-thickness dependence.<sup>2</sup> The values used for the exchange-stiffness constant,  $A = 1.14 \times 10^{-6}$  erg  $\text{cm}^{-1}$ , and for the  $g$  factor,  $g = 2.10$ , were obtained from the slopes of the  $H_n$ -vs- $n$  curves, which were nearly the same in both the perpendicular and parallel cases.<sup>7</sup> Thus there are only three remaining adjustable parameters

TABLE I. Resonance parameters, theory and experiment.

$H_n$ (Oe)		$\Delta H_n$ (Oe)		$I_n$	
Expt.	Theory	Expt.	Theory	Expt.	Theory
14780	14780	52	59	100	100
14569	14580	22	21	24	23
14390	14392	21	≈ 18	0.03	0.02
14151	14147	22	16	5.2	6.8
13481	13466	21	13	1.3	3.3
12536	12525	21	9	0.44	2.7

which we varied for the best fit to the spectrum. The values were  $4\pi M_s = 11\,580$  Oe,  $K_s^{(0)} = 0.87$  erg/cm<sup>2</sup>, and  $K_s^{(d)} = -1.1$  ergs/cm<sup>2</sup>. The signs of the two surface-anisotropy constants imply that a uniaxial rather than a unidirectional surface anisotropy was needed for the best fit. Finally, we have taken  $h_d/h_0 = 1$ . It is seen that the theoretical and experimental resonance fields agree to within 0.1% or better.

If we let  $n$  take the values 0, 2, 3, 4, 6, and 8 and calculate the resonance fields from a simple quadratic fit to the data,  $H_n' = a - bn^2$ , we obtain  $H_n' = 14\,690, 14\,555, 14\,387, 14\,150, 13\,473, 12\,535$  for  $n = 0$  through 8, respectively. It is clear that a good quadratic fit is obtained for the high values of  $n$  but not for the low values. By comparing  $H_n'$  and  $H_n$ , one sees that it is meaningless to talk about a departure of the spectrum field positions from an  $n^2$  law for small  $n$  since the field positions depend critically on the details of the boundary conditions at the two surfaces. For full pinning on both sides of the film, the  $n^2$  law would be obeyed but with  $n = 1, 3, 5, 7,$  and 9; for no pinning, only one line would be observed. In our case, the pinning must be at some intermediate value as predicted by the theory given above.

The corrected observed intensities<sup>13</sup> of the lines are in reasonable agreement with the theory although the experimental values decrease more rapidly with  $n$  than do the theoretical values. Better agreement for higher  $n$  could be obtained, of course, if one used smaller values of the surface anisotropies. However, using smaller values of  $K_s$  would also reduce the theoretical intensities of the second and third lines as well as reduce the good fit of the resonance fields. Thus the best overall fit is obtained with a compromise of the values  $K_s^{(0)}, K_s^{(d)}$ , and  $4\pi M_s$ .

The observed linewidths as shown in Table I drop off rapidly as the effects of exchange and conductivity diminish and then remain constant, whereas theoretical linewidths continue to decrease. This apparent inconsistency may be accounted for by postulating a small variation in the film thickness across the plane of the film. As shown previously,<sup>7</sup> a variation in the film thickness may broaden the line by an amount

$$\Delta H_n = \frac{2A n^2 \pi^2}{M} \frac{2\Delta d}{d^2} \frac{2\Delta d}{d},$$

where  $\Delta d$  is the change in thickness across the plane of the film. This additional broadening amounts to 12 Oe for a  $\Delta d$  as small as 8 Å. If

we plot the difference between the observed and calculated linewidths (see Table I) as a function of  $n^2$  (for  $n = 4, 6, 8$ ), we obtain a linear relationship. This suggests that the discrepancy may indeed be due to a small nonuniformity in the film thickness. The film could easily have a thickness variation of 8 Å (one or two atom layers).

The perpendicular resonance spectrum was obtained also on many other films of different thicknesses (350 to 2700 Å) and for different deposition rates (10 to 40 Å/sec). The spectra for all the films exhibited similar behavior—that is, the dependence of  $H_n$  on  $n^2$  was linear for all but the first one or two modes if the modes were labeled  $n = 0, 1,$  etc.; there was an alternation of intensity of the strong and weak modes, and the intensities of the higher-order modes were quite weak. Thus, it would be easy to fit the spectrum of another film of the same evaporation set from which the film discussed above came. However, to test the theory critically, we chose a film at random from another set which had been evaporated under conditions completely different. This film was 2010 Å thick and was deposited at a rate of 19 Å/sec; immediately following deposition, the film was annealed at 250°C in the presence of a planar magnetic field. After fitting the observed spectrum with the theory for this film, we found that  $K_s^{(0)} = 0.35$  erg/cm<sup>2</sup>,  $K_s^{(d)} = -0.5$  erg/cm<sup>2</sup>,  $4\pi M_s = 11\,150$  Oe, and  $g, \lambda, \sigma,$  and  $A$  were the same as for the thicker film. Again, a uniaxial rather than a unidirectional surface anisotropy must be used to characterize the film.

We have thus shown that the resonance parameters can be described quite well by a simple theory which assumes a uniform static magnetization across the film thickness. Any nonuniformity in the magnetization probably occurs at the film surfaces, and this has been accounted for by asymmetric surface anisotropies.

<sup>1</sup>C. Vittoria, R. C. Barker, and A. Yelon, *J. Appl. Phys.* **40**, 1561 (1969).

<sup>2</sup>G. C. Bailey and C. Vittoria, to be published.

<sup>3</sup>G. I. Lykken, and D. R. Schmitz, *J. Appl. Phys.* **41**, 1024 (1970).

<sup>4</sup>M. Ondris and H. Gartner, *Z. Angew. Phys.* **24**, 200 (1968).

<sup>5</sup>P. E. Wigen, C. F. Kooi, and M. R. Shanabarger, *J. Appl. Phys.* **35**, 3302 (1964).

<sup>6</sup>E. Schlömann, *J. Appl. Phys.* **36**, 1193 (1965).

<sup>7</sup>G. C. Bailey, *J. Appl. Phys.* **41**, 5232 (1970).

<sup>8</sup>R. H. Wade and J. Silcox, *Appl. Phys. Lett.* **8**, 7 (1966).

<sup>9</sup>G. T. Rado and J. R. Weertman, *J. Phys. Chem. Solids* **11**, 315 (1959).

<sup>10</sup>R. F. Soohoo, *J. Appl. Phys.* **32**, 1488 (1961).

<sup>11</sup>In the spectrum plot of Ref. 10, it is not clear what value of the damping constant was used.

<sup>12</sup>The surface impedance of only one side of the film is negative for some values of the applied field. Thus,

both terms in Eq. (7) must be used, or else the power absorbed by the film would have a physically meaningless negative value.

<sup>13</sup>In order to compare the theoretical and experimental intensities, it was necessary to consider that, for the same mode, the experimental and theoretical linewidths were different and that the linewidth changed with order number. Thus, all intensities were normalized to the main line and multiplied by  $\Delta H_n/\Delta H_0$ .

## $\text{Na}_3[\text{Ce}(\text{C}_7\text{H}_3\text{NO}_4)_3] \cdot 15\text{H}_2\text{O}$ : A New Material for Millikelvin Thermometry\*

J. C. Doran,† U. Erich,‡ and W. P. Wolf

*Department of Physics and Department of Engineering and Applied Science, Yale University, New Haven, Connecticut 06520*

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Magnetic specific heat and susceptibility measurements have shown that  $\text{Na}_3[\text{Ce}(\text{C}_7\text{H}_3\text{NO}_4)_3] \cdot 15\text{H}_2\text{O}$  has extremely weak effective spin-spin interactions, suggesting that this material may be a useful alternative to cerium magnesium nitrate for very low temperature thermometry.

Since the original work on cerium magnesium nitrate (CMN), more than eighteen years ago,<sup>1,2</sup> the method used almost exclusively for determining temperatures below 100 mK has been to measure the susceptibility of a CMN thermometer. To a good first approximation this obeys Curie's law and the magnetic temperature  $T^* = \lambda/\chi$  is close to the true temperature  $T$ , where  $\chi$  is the susceptibility and  $\lambda$  is the appropriate Curie constant.<sup>3</sup> However at the lowest temperatures deviations from Curie's law become increasingly important and there has been a considerable amount of controversy over the proper  $T$ - $T^*$  relation under different conditions. In particular, there has been much discussion of the effects of using powdered CMN and nonellipsoidal sample shapes, the most common being a right circular cylinder with length equal to diameter, and even for the case of a single-crystal sphere there are significant discrepancies in the  $T$ - $T^*$  relation for  $T < 3$  mK.<sup>4-7</sup> It is clear that another salt which closely follows Curie's law at very low temperatures would be quite valuable, either for establishing a more reliable scale for CMN or as a thermometric substance useful in itself. In this Letter we report the first results for a substance we refer to as CDP, a promising new material of this kind.

CDP denotes "cerium dipicolinate," which is our abbreviation for trisodium tris-(pyridine-2,6-dicarboxylato) cerate (III) pentadecahydrate,  $\text{Na}_3[\text{Ce}(\text{C}_7\text{H}_3\text{NO}_4)_3] \cdot 15\text{H}_2\text{O}$ , one of a large class of

complex organic rare earth salts whose structures have recently been studied by Albertsson.<sup>8,9</sup> Many of these are magnetically very dilute with atomic volumes  $V_a$  close to  $1000 \text{ \AA}^3/\text{rare earth ion}$ , so that one would generally expect quite weak interactions between the magnetic ions. We chose CDP specifically for three main reasons: (i) All cerium isotopes have zero nuclear spin, so that there is no complication from the hyperfine structure; (ii)  $\text{Ce}^{3+}$  ions, with only one  $4f$  electron, have relatively weak magnetic moments which should further reduce the magnetic interactions; and (iii) CDP is readily prepared and is reasonably stable. These are the same considerations which first led to the work on CMN<sup>1</sup> but in CDP we have the additional advantage that the average spacing between the  $\text{Ce}^{3+}$  ions is some 16% larger while the mean square moment turns out to be some 31% smaller, with the result that the effective spin-spin interaction is only about one half of that found for CMN.

To obtain a quantitative estimate of the interactions in CDP we have carried out two types of experiments: (i) a measurement of the magnetic specific heat  $C_M$  in the region of 1 K using the Casimir-du Pré relaxation method,<sup>10</sup> and (ii) a preliminary study of the susceptibility down to 50 mK using a  $\text{He}^3$ - $\text{He}^4$  dilution refrigerator and a CMN thermometer. Some additional adiabatic demagnetization experiments to very low temperatures were also carried out.

For relaxation specific heat measurements one