Ferromagnetic Metals in High Magnetic Fields

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High-field susceptibility results are presented for Mössbauer measurements on iron at low temperatures in fields up to 135 kOe and magnetization measurements on iron and nickel single crystals in fields up to 148 kOe with vibrating sample magnetometers. The Mössbauer measurements give the hyperfine field $H_n = H_{\text{int}} - (H_0 - H_{\text{DM}}) + \Delta H$, where H_{int} is the hyperfine field at zero applied field, H_0 is the applied field, H_{DM} is the demagnetizing field, and ΔH is a term which would reflect a change in the magnetization at high field, i.e., $\Delta H/H_n = \Delta M/M$. The Mössbauer results at 4.2°K are equivalent to $\chi = 2 \pm 7 \times 10^{-5}$ emu/cc. The magnetization experiments give $\chi \leq 4 \times 10^{-5}$ for iron and $\chi \leq 1.1 \times 10^{-5}$ for nickel at 4.2°K. We discuss the various contributions to the total susceptibility χ , give estimates of χ_d based on theoretical band calculations, and discuss various uncertainties in both the theoretical calculations and in values of the molecular field parameter which make detailed comparisons difficult. Finally, we compare with predictions based on Herring's phenomenological description of χ for ferromagnets.

I. INTRODUCTION

THE change of magnetization with applied magnetic field is an important property of ferromagnets because it allows a comparison with the predictions of band, or collective-electron, theory¹ and may have relevance in deciding between itinerant versus localized models of ferromagnetism.² Wohlfarth¹ has called attention to the fact that the collective-electron theory³ predicts a relative magnetization at absolute zero ζ_0 which may be less than one, depending on details of the density of states of the metal, $N(E)$, and the strength of the Weiss molecular field. The application of an intense external magnetic field should cause, at very low temperatures, an increase in ζ_0 if ζ_0 is indeed < 1 . Such a situation is, however, incompatible with theories based on localized atomic moments which require^{1,2} $\zeta_0 \approx 1$ at $T = 0^\circ\text{K}$.

In collective electron theory, the high-field band susceptibility χ_d at $T = 0^\circ\text{K}$ is given by

$$n\mu^2/\chi_d = \frac{1}{4}n\{[1/N(\epsilon_{F\uparrow})] + [1/N(\epsilon_{F\downarrow})]\} - k\theta', \quad (1)$$

if one ignores other contributions (discussed later). This result assumes that the total energy is the sum of single-particle energies plus an added exchange energy. In Eq. (1), n is the total number of electrons, $k\theta'$ is the molecular field represented by a characteristic temperature θ' , and $N(\epsilon_{F\uparrow})$ and $N(\epsilon_{F\downarrow})$ are the spin-up and spin-down densities of states at the Fermi energy, respectively. It is clear that if $\zeta_0 = 1$, $\chi_d = 0$, since $N(\epsilon_{F\uparrow})$ or $N(\epsilon_{F\downarrow}) = 0$.

II. MAGNETIZATION STUDIES

The smallest previous limits of the high-field susceptibility in Fe and Ni were established by Kapitza.⁴

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¹ E. P. Wohlfarth, *Phys. Letters* **3**, 17 (1962).

² C. Herring, in *Magnetism*, edited by H. Suhl and G. T. Rado (Academic Press Inc., New York, to be published), Vol. IV.

³ E. C. Stoner, *Phys. Soc. Progr. Phys.* **11**, 43 (1948); *Proc. Roy. Soc. (London)* **A165**, 372 (1938).

⁴ P. Kapitza, *Proc. Roy. Soc. (London)* **A131**, 243 (1931); see also W. E. Henry, *Phys. Rev.* **99**, A668 (1955).

For pulsed field changes of about 250 kOe, he found that the change in the magnetization was less than 1% (the limits of his error).

It is important to note that absolute sensitivity or accuracy is not a major problem for these large moment measurements; rather, high relative differential sensitivity is required, and usually negligible systematic and instrumental corrections become dominant. We have combined the highest readily available fields with two magnetometers for these measurements. First, differential magnetic moments (DMM) were measured at 4.2°K up to 80 kOe in superconducting magnets with a vibrating sample magnetometer (VSM)⁵ modified for the low-temperature, axial-field configuration. Small single-crystal, spherical samples of Fe and Ni subjected to an applied field along the easy axes showed very small susceptibility over this field range. Second, DMM measurements were extended with a simplified VSM assembled for operation in the NML water-cooled, high-field solenoids. Briefly, this instrument is a low-frequency VSM of moderate sensitivity which combines flux integration, time averaging, and calibrated high-differential stability, and is specifically arranged to eliminate possible sample-positioning errors.⁶

The present measurements on Fe and Ni single crystals up to 148 kOe with the latter instrument show high-field susceptibilities for $\chi_{\text{Fe}} \leq 4 \times 10^{-5}$ and $\chi_{\text{Ni}} \leq 1.1 \times 10^{-5}$. The reproducibility of any data point over the entire field range had a maximum deviation of less than 0.05% to 0.1%. The available large field range permitted the detection and elimination of small systematic errors. Our results are in agreement with earlier measurements of Kapitza⁴ and Henry⁴ and with the Mössbauer measurements described below. We expect that any remaining systematic errors are small compared to the quoted upper limits and that the actual values are very close to these limits.

⁵ S. Foner, *Rev. Sci. Instr.* **30**, 548 (1959).

⁶ Further details about this instrument will be described elsewhere. S. Foner and E. J. McNiff, Jr. (to be published).

III. MÖSSBAUER STUDIES

The Mössbauer experiments were performed using a source of ^{57}Co in metallic iron together with an ^{57}Fe -enriched metallic iron foil absorber. Both source and absorber were at 4.2°K and in an external longitudinal magnetic field of about 135 kOe, resulting in a three-line absorption spectrum.⁷ The splitting between the outer lines is proportional to the hyperfine field H_n assuming H_n to be the same in both source and absorber. This configuration results in a much higher counting rate than if an unsplit single-line source at zero external field were used; consequently the statistical error is reduced for a given available counting time. Consider $H_n = H_{\text{int}} + H_{\text{DM}} + \Delta H - H_0$, with H_n the observed hyperfine field, H_{int} the hyperfine field at zero external field (338 kOe at 4.2°K), $-H_0$ the applied field, H_{DM} the demagnetizing field (21.8 kOe for a thin iron foil),⁸ and ΔH the discrepancy corresponding to a change in the magnetization. We assume strict proportionality between the hyperfine field and the magnetization. In terms of the volume susceptibility our result corresponds to $\chi = [\Delta H \times M] / [H_n \times (H_0 - H_{\text{DM}})]$, where M is the saturation magnetization in iron (~ 1.7 kG). Combining the results of three separate determinations of ΔH , we obtain the results, $\Delta H = 0.3 \pm 1.3$ kOe, and $\chi = (2 \pm 7) \times 10^{-6}$ emu/cc.

IV. DISCUSSION

The magnetic susceptibility of a metal, based on a simplified tight binding model of two types of bands (d and s), may be written as $\chi = \chi_s + \chi_d + \chi_{\text{v}} + \chi_{\text{diam}}$, where χ_s and χ_d are the spin paramagnetic contributions, χ_{v} is the paramagnetic contribution for partially filled degenerate bands (or Van Vleck temperature-independent paramagnetism for metals), and χ_{diam} is the diamagnetic contribution. We wish to deduce information about the band structure of ferromagnetic metals from measurements of χ at low temperatures. To do this we must first estimate the other contributions to χ . From measurements of χ in Cu, we estimate $\chi_{\text{diam}} = -1 \times 10^{-6}$ (units of χ are in emu/cc), which may be considered an upper limit because in both Fe and Ni the d band is not completely filled, and it is known that the $3d$ electrons are the major contributors to χ_{diam} . Similarly χ_s is very small in Ni ($\sim 0.9 \times 10^{-6}$ in the free-electron approximation for 0.6 "s" electrons) and in Fe ($\sim 1 \times 10^{-6}$ for one "s" electron).

A much larger contribution arises from χ_{v} for which estimates have been made for a number of metals including paramagnetic Ni (although none of these estimates considered ferromagnetically occupied bands).

They include⁹ Ni (1.1×10^{-5} and 0.6×10^{-5}), if a molecular field term is or is not included, respectively, Cr (2.0×10^{-5} and 2.2×10^{-5}), V (2.3×10^{-5}), and Pt (0.3×10^{-5}). We estimate that $\chi_{\text{v}} \approx 1.1 \times 10^{-5}$ for Ni and 1.5×10^{-5} for Fe. By contrast, we note that the spin-wave contribution to the change in magnetization with field is completely negligible ($\chi \sim 10^{-8}$) at low temperatures in the range of fields used here.

Estimates of χ_d can be made from band calculations using Eq. (1), but these are of necessity crude because in ferromagnets both the density-of-states term and the exchange term have approximately the same value. Note that while we have separated out χ_s from χ_d in the above discussion the $N(\epsilon)$ terms from energy-band calculations contain combined s and d contributions. Since χ_s is very small, only a small error is made in using either procedure, but it does serve as a basis for making comparisons. For Fe, Cornwell and Wohlfarth,¹⁰ using Wood's¹¹ augmented plane-wave calculations, find $N(\epsilon_F) = 0.75$ and $N(\epsilon_F) = 0.35$ (states/atom·eV·spin) and a band splitting $\Delta E (= 2k\theta') = 1.35$ eV. This gives, by Eq. (1), $\chi_d = 1.1 \times 10^{-5}$. Thus, the sum of χ_d and χ_{v} is consistent with our measured upper limit and supports the accepted view that both bands in Fe contain holes.

For Ni, our present result $\chi < 1.1 \times 10^{-5}$ indicates that the entire susceptibility could arise from χ_{v} within the estimated uncertainties in these contributions. Such a result is consistent with one full spin-band in Ni, in agreement with previous expectations.¹²

Finally, Herring² has used a simplified phenomenological description to estimate χ_d . Using a difference in energy ΔE of 0.084 eV/atom for Fe, obtained from an analysis of magnetic specific-heat data, he finds $\chi_d = 2.7 \times 10^{-5}$. Applying this description to Ni, and using² $\Delta E = 0.02$ eV, one finds $\chi_d \approx 1 \times 10^{-5}$, which is larger than our estimated value and may be the result of a greater uncertainty in the estimate of ΔE from specific-heat data in this case.

Note added in proof. High-field susceptibility measurements for Fe and Ni have also been reported at this meeting by Herring *et al.* (p. 1340). Their values of χ are much larger than those reported here.

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⁹ See for example, M. Shimizu, T. Takahashi, and A. Katsuki, J. Phys. Soc. Japan **18**, 801 (1963). Because of space limitations, full references will be given in a forthcoming publication.

¹⁰ J. F. Cornwell and E. P. Wohlfarth, J. Phys. Soc. Japan **17**, Suppl. B-1, 32 (1962).

¹¹ J. H. Wood, Phys. Rev. **126**, 517 (1962).

¹² See for example, J. C. Phillips, Phys. Rev. **104**, 1263 (1956); *ibid.* **133**, A1020 (1964); and E. D. Thompson, E. P. Wohlfarth, and A. C. Bryan, Proc. Phys. Soc. (London) **83**, 59 (1964).

⁷ N. Blum and L. Grodzins, Phys. Rev. **136**, A133 (1964).

⁸ The source and absorber foils were 0.375 in. diam $\times$ 0.001 in. thick and 0.625 in. diam $\times$ 0.0002 in. thick, respectively.