Effect of the Magnetic Structures of Holmium Metal on the Absorption Spectrum of 3-mm Microwave Radiation*

JOHN L. STANFORD
Institute for Atomic Research and Department of Physics, Iowa State University, Ames, Iowa

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The absorption of 3-mm (100-GHz) microwave radiation has been studied in single-crystal holmium metal from 4–80°K in applied magnetic fields up to 26 kOe. A variety of absorption spectra (as a function of applied field) were observed and have been correlated with the complicated magnetic structures of metallic holmium. Sharp absorption peaks of strong intensity were observed at helical-fan and fan-fan field-induced phase transitions. Anisotropy studies in the 50–60°K range revealed only small anisotropy between "easy" ([10$\bar{1}$0]) and "hard" ([11$\bar{2}$0]) directions of magnetization, in contrast with published results based on neutron diffraction studies, but in agreement with magnetization and magnetoresistance data for holmium. The absorption was found to be independent of the relative polarization of H_{rf} with respect to H_{dc}, suggesting that it is characteristic purely of magnetic domain and phase effects. The zero–wave-number spin-wave resonances predicted for the heavy rare-earth metals by Cooper and Elliott were not observed, and it is shown that this is in agreement with the two- and sixfold anisotropy-constant calculations by Kasuya and a consideration of the sequence of field-induced magnetic phases of holmium. No evidence of a resonance in the helical antiferromagnetic phase was observed, in agreement with the prediction of Cooper and Elliott. The absorption of microwave power was found to be a sensitive method for detection of magnetic phase transitions, in some cases being much more sensitive than magnetization or magnetoresistance techniques.

I. INTRODUCTION

THE magnetic structure of holmium metal is a very complex function of temperature and applied magnetic field. The magnetization studies on single crystals of holmium by Strandburg *et al.*[1] revealed an intriguing series of steplike increases in magnetization with increasing applied field. The recent single-crystal neutron diffraction work by Koehler *et al.*[2] has shown that the field-induced magnetic structures of holmium are quite complicated, some phases containing as many as six wave-number components characterizing the rotation of the static magnetization from layer to layer in the crystal. Pronounced changes in the longitudinal magnetoresistance with applied field have also been observed in single crystals of holmium.[3,4] Here we report a study of the effect of the field-induced magnetic structures of single-crystal holmium on the absorption of 3 mm microwave power ($\omega/2\pi=100$ GHz).

Microwave power absorption in single crystals of holmium has been reported by Bagguley and Liesegang.[5] Their data, taken at 35 GHz, were not entirely reproducible and consisted of broad absorption lines observed in the 55-70°K temperature range. No absorption was observed below 50°K. Our experimental results, at 100 GHz, show a profuse variety of absorption spectra at temperatures ranging from 80° down to 4.2°K. Accounting for hysteresis effects, the data are sharp and reproducible and are found to be consistent with the complex magnetization phases previously reported for holmium.

II. MAGNETIC STRUCTURE OF HOLMIUM

In the absence of an applied magnetic field, holmium undergoes a transformation from the paramagnetic to a helical antiferromagnetic state at a Néel temperature of about 133°K. The axis of the helix is along the c axis (holmium is hexagonal close packed). At about 20°K, the helix transforms to a conical structure, with the cone pointed along the c axis, producing a net ferromagnetic component along that axis. The conical projections of the moments in the basal plane are found to be bunched around "easy" b axis directions[6] by the strong hexagonal anisotropy found at low temperatures.

With application of an external magnetic field, the structures become exceedingly complex and data interpretation can no longer be based on the zero-field structures. At 78°K, the helical state is forced into a series of "fan" structures by a field H applied in the basal plane. At about 18 kOe, a fan-I phase appears, with a second (fan II) phase appearing around 20 kOe.[2] Higher fields would presumably force the system into a pure ferromagnetic structure. Between 33 and 40°K, an applied field along a "hard" a-axis direction of magnetization changes the helix into two intermediate phases but with a small component of the magnetization normal to H. With H still along an a axis, two other temperature ranges with distinct magnetic phase characteristics, 25–33°K and 20–25°K, are reached before the low-temperature ferromagnetic region. Due to the large hexagonal anisotropy, the latter has ferromagnetic alignment of the moments along b axes for

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[1] D. L. Strandburg, S. Legvold, and F. H. Spedding, Phys. Rev. **127**, 2046 (1962).

[2] W. C. Koehler, J. W. Cable, H. R. Child, M. K. Wilkinson, and E. O. Wollen, Phys. Rev. **158**, 450 (1967).

[3] L. E. Spanel, Ph.D. thesis, Iowa State University, 1964 (unpublished).

[4] A. R. Mackintosh and L. E. Spanel, Solid State Commun. **2**, 383 (1964).

[5] D. M. S. Bagguley and J. Liesegang, Phys. Letters **17**, 96 (1965); J. Appl. Phys. **37**, 1220 (1966); and to be published.

[6] W. C. Koehler, J. W. Cable, M. K. Wilkinson, and E. O. Wollan, Phys. Rev. **151**, 414 (1966).

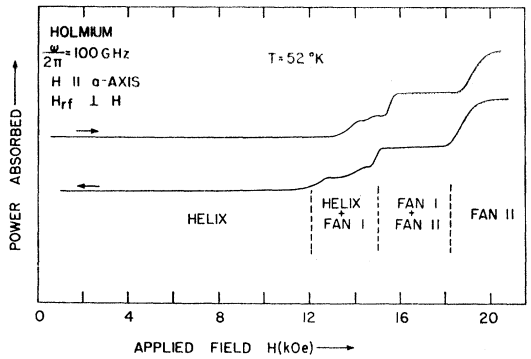

FIG. 1. Four distinct magnetic phases are observed in the 3 mm power absorption in single-crystal holmium metal as a function of applied field at 52°K. The sample is disc-shaped, with normal along [0001] and H applied in the plane of the disc along a magnetically hard [11$\bar{2}$0] direction. The increase in power absorption at the onset of the pure fan-II region is 25% of the zero-field absorption.

fields above 5 kOe, irrespective of whether H is applied along an a or b axis. A remanent state which has some of the characteristics of the fan-I phase is reached upon cycling of the field. A full account of the field-induced structures is given in detail by Koehler *et al.*[2]

III. EXPERIMENTAL APPARATUS AND SPECIMEN

A description of the experimental apparatus has been given elsewhere.[7] The specimen used in this investigation contained less than $\frac{1}{4}$% impurities, which were chiefly La, Tm, and Er. The specimen was disc-shaped, with a and b axes in the plane of the disc and the c axis normal to the disc. The diameter and thickness of the disc were approximately 9 mm and 1 mm, respectively. The microwave data were found to be very sensitive to the surface preparation of the specimen. The surface exposed to the microwave radiation was electropolished by a perchloric acid technique at −70°C.[8] The specimen

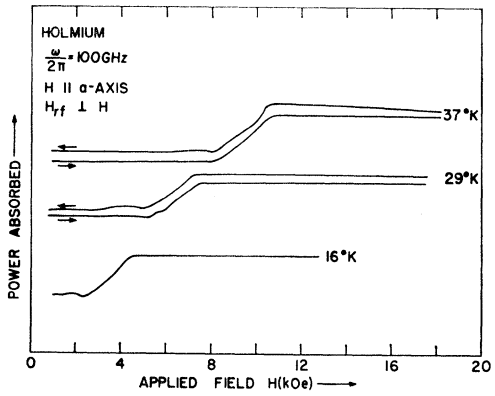

FIG. 2. Microwave power absorption in single-crystal holmium. The magnitude of the absorption change is ≈50% at each temperature.

[7] J. L. Stanford and R. C. Young, Phys. Rev. **157**, 245 (1967).
[8] E. N. Hopkins, D. T. Peterson, and H. H. Baker, U.S. Atomic Energy Report No. IS-1184 (unpublished).

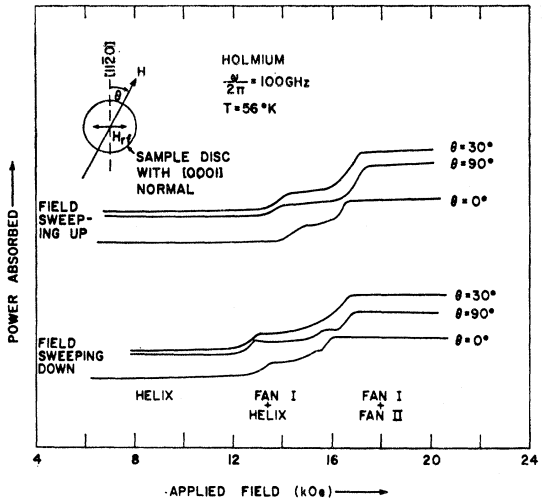

FIG. 3. Anisotropy studies at 56°K in single-crystal holmium. θ=0° is an a axis (magnetically hard direction) and θ=30°, 90° are b axes (easy directions of magnetization). H is applied in the plane of the disc-shaped sample, whose normal is along [0001]. This disc has a 9 to 1 diameter-to-thickness ratio, with a 9 mm diam.

was kept under vacuum to reduce oxide formation on the surface.

IV. DISCUSSION OF RESULTS

Typical experimental results are shown in Figs. 1–4, which are tracings of X-Y recorder graphs. Figure 1 is characteristic of the absorption spectra (as a function of field) observed above 40°K. Four distinct magnetic phases are evident. When demagnetizing effects are taken into account, the onsets of power absorption are found to be in good agreement with the onsets of the various field-induced magnetic phases in holmium.

Hysteresis

Figure 1 also shows significant hystersis associated with the field-induced fan-I phase. The two higher

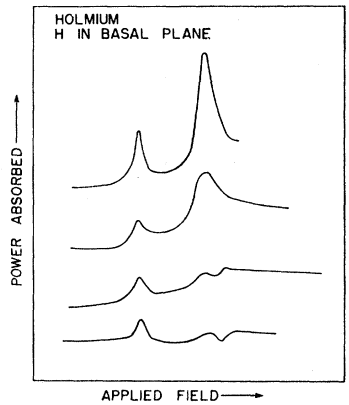

FIG. 4. Tracing of 3 mm power absorption observed at onsets of field-induced fan phases at liquid nitrogen temperature. The specimen is a disc of 9 mm diam and 1 mm thickness with normal along [0001]. The largest peaks occurred after temperature and field cycling and correspond to a 50% change in absorption.

field transformations are seen to occur with much smaller hysteresis. These observations are in agreement with the magnetization[1] and neutron-diffraction[2] studies on holmium.

Domain Orientation

The absorption curves are reminiscent of earlier observations of absorption onsets associated with domain-orientation/magnetostriction effects in terbium.[7] Indeed, as with terbium, the holmium power absorption onsets are correlated with the onsets of magnetostriction observed in single-crystal holmium.[9] In the pure ferromagnetic phase of terbium, the magnetostriction onset occured at fields around 5 kOe, depending on specimen shape and temperature. In holmium, the onset of magnetostriction is seen to occur immediately at the formation of a ferromagnetic phase since the fields necessary to induce the transitions ($\approx$10–20 kOe) are generally larger than those necessary to produce domain rotation.

The widths of the transition regions are found to be largely due to the effects of demagnetizing fields. The onset of a given phase appears to be at the onset of the microwave absorption rather than at the peak or knee of the absorption curve. The latter corresponds to the field value (plus demagnetizing field) at which the transition from one phase to another is essentially complete.

Absorption Peaks

Figure 4 shows the pronounced absorption peaks that were observed at the helical-fan and fan-fan phase transitions in the 60–80°K range. When the specimen was cemented down in the center as well as around the edges, the peaks were greatly diminished or disappeared. But with the disc held only by the edges, the peaks were observed, and as shown by the evolution in the tracings of Fig. 4, the peaks could be spectacularly enhanced by cycling the field and temperature. The applied field was at all times in the plane of the disc. Similar observations have been reported for dysprosium.[10,11] We have made polarization studies which reveal that the absorption peaks are independent of the polarization of H_{rf} with respect to the applied field H, both lying in the plane of the disc. We do not have a convincing explanation for these absorption peaks at present.

Remanent State Effects

At low temperatures, remanent state effects were observed. In our experiment, a phase-sensitive detection system operating at 70 kHz applies necessary correction voltages to the klystron reflector to keep the klystron frequency locked to the resonance frequency of the cavity containing the sample. It was found that whenever the sample was cooled below about 20°K in zero field and then subjected to a slowly increasing applied field, the klystron suddenly became unlocked from the cavity frequency, probably due to large, rapid domain orientation effects. If the field were then reduced to zero and the klystron relocked onto the cavity frequency, a second upward field sweep would reveal a new absorption curve, indicating the existence of a remanent magnetization. As a result, reproducible microwave absorption data versus applied field could be obtained only for the remanent magnetic state. The neutron diffraction work of Koehler *et al.*[2] has shown that this remanent state has some of the properties of the fan-I phase and that cycling the applied field never reproduces the virgin state at low temperatures. Only warming the sample above 25°K eliminates the remanent state.

Anisotropy and Polarization Studies

Anisotropy studies were carried out for temperatures in the 50–60°K range and also at 78°K. Figure 3 shows data at 56°K which reveal that the anisotropy between a and b axes is not as great as that expected from the published phase diagrams of holmium in an applied magnetic field.[2] Spanel[3] has studied longitudinal magnetoresistance in single crystals of holmium. His data for **H** applied along $[10\bar{1}0]$ are given in Ref. 4, but the data for **H** parallel to a hard a axis have not been previously published. With the kind permission of Dr. Spanel, we reproduce here his $[2\bar{1}\bar{1}0]$ data in Fig. 5. Comparison of the $[10\bar{1}0]$ and $[2\bar{1}\bar{1}0]$ magnetoresistance data reveals only a small anisotropy in the basal plane above 50°K. The smaller anisotropy of our results near 56°K are in agreement with the work of Strandburg *et al.*[1] and of Spanel. At 78°K no

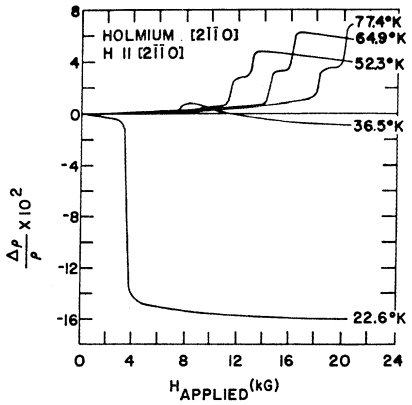

FIG. 5. The change in longitudinal magnetoresistance of single-crystal holmium versus applied field along $[2\bar{1}\bar{1}0]$ is plotted for various temperatures. These figures are given with the kind permission of Dr. L. E. Spanel.

[9] S. Legvold, J. Alstad, and J. Rhyne, Phys. Rev. Letters **10**, 509 (1963).

[10] F. C. Rossol, B. R. Cooper, and R. V. Jones, J. Appl. Phys. **36**, 1209 (1965); F. C. Rossol and R. V. Jones, *ibid.* **37**, 1227 (1966).

[11] F. C. Rossol, Ph.D. thesis, Harvard University, 1966 (unpublished).

anisotropy was observed between $[11\bar{2}0]$ and $[10\bar{1}0]$ field directions, again in agreement with the magnetization and magnetoresistance work on holmium.

Appreciable hysteresis was observed for the fan-I phase with applied field in both the $[10\bar{1}0]$ and $[11\bar{2}0]$ directions, as shown in Figs. 1 and 3. The onset of the fan-I plus fan-II structure is seen to have only slight hysteresis. The extra structure seen with decreasing field at the higher phase onset is associated with domain effects.

To minimize magnetic torque strain on the sample, field rotations during the anisotropy studies were performed with the field at its lowest value, about 0.5 kOe.

Finally, the rotation studies at 56°K (Fig. 3) show that the microwave power absorption is essentially independent of the polarization of the microwave H_{rf} with respect to the applied field H, which was swept slowly in time. Both H_{rf} and H lie in the basal plane. The microwave absorption data presented here may therefore be assumed to be characteristic purely of magnetic domain and phase effects since spin-wave absorption would be expected to be sensitive to the relative polarization of H_{rf} with respect to the magnetization direction, the latter being in general a function of the applied-field direction.

Sensitivity

Microwave absorption is found to be much more sensitive in some cases than magnetization or magnetoresistance in revealing different magnetic phases. For example, near 50°K, the onset of the pure fan-II phase is unnoticable in the $[2\bar{1}\bar{1}0]$ magnetoresistance results of Spanel (Fig. 5) and in the magnetization studies.[1] However, the same onset appears as a 25% increase in the microwave power absorption in Fig. 1. Thus, microwave absorption provides yet another sensitive probe for the investigation of magnetic systems.

V. MAGNETIC RESONANCE

No evidence of resonant spin-wave absorption of microwave power was seen in holmium. The types of absorption resonances expected generally for the heavy rare-earth metals will now be discussed, together with the probable reasons for their not being observed under the conditions of our investigation.

At least three types of resonant absorption of microwaves are to be expected in the heavy rare-earth metals. These have been treated in the theoretical work of Cooper and Elliott.[12] Briefly, for the regions of temperature and applied field where the magnetization lies in the basal plane, these possible resonances are as follows:

1. Microwave power may be absorbed in zero magnetic field if the microwave frequency coincides with the spin-wave energy gap at $q=0$ [$q=2\pi/$(wavelength of the spin wave)]. The frequency of the gap is given by

$$\omega/2\pi=(g\mu_B/h)\,(H_A 36H_h)^{1/2}, \tag{1}$$

where H_A and H_h are effective twofold and hexagonal anisotropy fields defined in terms of the twofold and hexagonal anisotropy constants K_2 and K_6^6 of the Hamiltonian of the system.[12]

2. In the presence of a magnetic field H applied along a hard direction of magnetization, the $q=0$ spin-wave energy gap is modified[13]:

$$\omega/2\pi=(g\mu_B/h)\,\{H_A(H-36H_h)\}^{1/2} \tag{2}$$

if $H>36H_h$, or

$$\omega/2\pi=(g\mu_B/h)\,\{H_A(H\cos(\tfrac{1}{6}\pi-\theta)+36H_h\cos 6\theta)\}^{1/2} \tag{3}$$

if $H<36H_h$. The value $36H_h$ is that field along a hard direction necessary to pull the magnetization M into the hard direction. In general, M makes an angle θ with respect to an easy direction. Equations (1)–(3) are true in the limit of $H_A\gg H$, M, H_h, which is valid for our experiment since, as will be discussed, $H_A\approx$ 275 kG at low temperatures in holmium. Since θ is a function of H, Eq. (3) must be solved in a self-consistent manner.

3. At high temperatures the hexagonal anisotropy decreases faster than the twofold anisotropy, so that H_A may still remain large while $H_h\approx 0$. In this regime, the resonance condition becomes

$$\omega/2\pi=(g\mu_B/h)\,\{H_A H\}^{1/2}. \tag{4}$$

Resonances of the second type have been reported at 9 and 38 GHz in dysprosium[5,10,14] and at 9 GHz in terbium.[5] Resonances in the third regime have been studied in terbium at 100 GHz.[7]

Application to Holmium

Equations (1)–(4) require that the magnetization lie in the basal plane. This is true for holmium for temperatures above about 20°K, even in the presence of an applied field in the basal plane. Kasuya[15] gives theoretical values for K_2 and K_6^6 for the heavy rare-earth metals. His values for Tb and Dy are in reasonable agreement with experiment.[16] Thus in the absence of direct experimental values, we use Kasuya's values of K_2 and K_6^6 for holmium to obtain a reasonable estimate of the fields necessary for the several types

[12] B. R. Cooper and R. J. Elliott, Phys. Rev. **131**, 1043 (1963); **153**, 654 (E) (1967).

[13] See Ref. 12 and B. R. Cooper, R. J. Elliott, S. J. Nettel, and H. Suhl, Phys. Rev. **127**, 57 (1962). The form of Eqs. (2) and (3) is that given in Ref. 11.

[14] H. A. Blackstead and P. L. Donoho, in Proceedings of the Sixth International Conference on Rare-Earth Research, Gatlinburg, Tennessee 1967 (unpublished).

[15] T. Kasuya, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York 1966), Vol. IIB Chap. 3.

[16] J. J. Rhyne and A. E. Clark, J. Appl. Phys. **38**, 1379 (1967).

of ferromagnetic resonance which might be observed at 100 GHz in holmium.

The value of the $q=0$ spin-wave energy gap at low temperatures (but $T>20°$K to keep M in the basal plane) is found, using Eq. (1) and Kasuya's values for K_2 and K_6[6], to be

$$\omega/2\pi \cong 400 \text{ GHz}.$$

This is higher than the frequency of our apparatus, 100 GHz, and thus should not be observed in our experiment.

Ferromagnetic resonance of the second type may be observed by utilizing an applied field along the magnetically hard a-axis direction in holmium. At low temperatures in holmium, we estimate from Kasyua's value for K_6[6],

$$36H_h \cong 200 \text{ kG} \gg (\hbar^2\omega^2/g^2\mu_B{}^2H_A) \qquad (5)$$

using $\omega/2\pi=10^{11}$ sec^{-1}. Thus, even at 100 GHz, the resonance fields at low temperatures will occur near $H=36H_h\cong 200$ kG, far outside the range of our apparatus. However, as discussed in Sec. IV, the hexagonal anisotropy is essentially zero ($H_h\approx 0$) at 78°K. At temperatures between 20 and 78°K, this type of ferromagnetic resonance might be expected to occur within the fields used in this investigation ($H\leq 26$ kOe) since H_h decreases rapidly with increasing temperature. In fact, no obvious resonance of this type was observed probably because either (i) the resonances were obscured by the various phase transitions, or (ii) the field values over which a given magnetic phase exists do not coincide with the fields necessary for resonance, (iii) the resonance intensities are much weaker in holmium than in terbium, or (iv) Eqs. (2) and (3) may not adequately describe the resonance conditions in the very complex fan phases of holmium. For a simple fan phase, the resonance conditions are not qualitatively different from Eqs. (2) and (3). However, the neutron-diffraction results show that the various field-induced fan phases in holmium are extremely complex, many different wave-number components being necessary to describe the magnetization.

Below 20°K, remanence effects do not allow a pure ferromagnetic state. Above 20°K, there is only a short temperature range from 25–33°K where our maximum available field along $[11\bar{2}0]$ could produce a pure ferromagnetic state. However, estimates based on the magnetization data[1] show that the hexagonal anisotropy field H_h is still so large at 30°K that a field of 18 kOe only pulls the magnetization about 2° away from the easy $[10\bar{1}0]$ direction. The resonance fields will be in the vicinity of $36H_h$, which is the field necessary to pull M to the field direction along a hard $[11\bar{2}0]$ direction, 30° away from the easy direction. Thus, the field necessary to pull the $q=0$ spin-wave gap down to 100 GHz at this temperature is much greater than the capability of our magnet (26 kOe), and the resonance could not be observed. In fact from the magnetization data, we estimate that $36H_h$ is still $\approx$200 kG at this temperature. For temperatures above 33°K and field along $[11\bar{2}0]$, the pure ferromagnetic state could not be reached with our maximum field.

Above 78°K, where the hexagonal anisotropy is essentially zero, ferromagnetic resonance in the third regime [Eq. (4)] may be observed. Scaling the experimental data on Tb[7] by Kasuya's K_2 values, one estimates that at 100 GHz this type of resonance should be observed in holmium for $H\sim$10–15 kOe. But at $T\geq 78°$K, this field is not sufficient to bring about a transition from the helix to a ferromagnetic state. Thus, this type of absorption should not be observed in holmium, and our data confirm this expectation.

Finally, at 100 GHz, we observe no resonance absorption in the pure helical phase for fields up to 26 kOe or up to the field-induced transition to the fan-I phase, whichever is lower. This is in agreement with the prediction of Cooper and Elliott that in the helical phase the only absorption at $q=0$ should be at zero frequency.[12]

VI. SUMMARY

The absorption of 100 GHz microwave radiation has been studied in single-crystal holmium metal for applied fields up to 26 kOe and in the temperature range of 4–80°K. A wide variety of absorption spectra, as a function of field and temperature, were found. Hysteresis, remanence, and magnetic domain effects were observed. Polarization studies showed that the absorption was independent of the relative orientation of H_{rf} and H_{dc} and that only small anisotropy exists in the basal plane for $T>50°$K. Large absorption peaks whose intensity and shape were functions of the magnetic history and mounting of the specimen were noted at several field-induced phase transitions. A discussion of types of $q=0$ spin-wave resonances expected for holmium is presented and it is shown that Kasuya's calculated values for the anisotropy constants of holmium yield resonance fields outside the range of our apparatus or in ranges inaccessible because of magnetic phase transitions. It was found that the microwave absorption technique is much more sensitive in resolving magnetic phases than magnetoresistance or magnetization methods.

ACKNOWLEDGMENTS

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