Curie temperature of PdFe films

J. R. Long and R. W. Mattozzi

Citation: Journal of Applied Physics **55**, 2359 (1984); doi: 10.1063/1.333664
View online: http://dx.doi.org/10.1063/1.333664
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/55/6?ver=pdfcov
Published by the AIP Publishing

Curie temperature of PdFe films

J. R. Long and R. W. Mattozzi
Virginia Polytechnic Institute and State University, Blacksburg, Virginia 24061

Ferromagnetic ordering of giant moments formed by dilute Fe in Pd host bulk alloys has been the subject of many investigations, but thin films have had little attention. Thin films exhibit evidence of a spin glass phase not observed in the bulk and a crossover from three-dimensional to two-dimensional ferromagnetic ordering. The range of ferromagnetic ordering for PdFe is long compared to that of pure Fe, so that the crossover to two-dimensional behavior should occur in relatively thick films. A study of the effect in pure Fe would require uniform films of a few tens of angstroms. The crossover is manifested by a depression of the Curie temperature, which is expected to vary as a critical exponent of the thickness. Using a mutual inductance technique with SQUID detection, the temperature dependences of the spontaneous magnetization and the ac susceptibility were measured for some sputtered films with approximately 8% Fe. As expected, the Curie temperature decreased as the film thickness decreased. Unlike Fe, the Curie temperature of PdFe is dependent upon microstructure, and the microstructure of a film may have a systematic thickness variation that could result in a false crossover effect. The extent to which a model sensitive shift exponent can be extracted from the results is discussed.

PACS numbers: 75.70.Dp, 75.50.Bb

INTRODUCTION

As the thickness of x of a ferromagnetic film approaches the correlation length of the ordering, a crossover from three- to two-dimensional ordering is expected. In the crossover regime, the Curie temperature T_C is expected to decrease as x decreases, and a critical parameter λ, the shift exponent, is defined.[1] The Curie temperature is then expected to be of the form

$$T_C(\infty) - T_C(x) \sim x^{-\lambda}. \quad (1)$$

To study this shift experimentally, samples must have a well defined x. For pure Fe, the correlation length is so short that films with a uniform thickness of a few tens of angstroms are required.[2] This difficult condition is made worse by the reactive nature of Fe.

The alloy PdFe has been studied extensively in bulk form.[3] Its ferromagnetism is understood to originate from the ordering of giant moments associated with Fe sites in the Pd host. The interaction is relatively long-ranged, and the crossover from bulk to two-dimensional behavior should occur at substantially greater x than in pure Fe. Thus, less difficulty is expected in producing uniform samples of PdFe in the crossover range, and the samples are not so reactive. Sputtered films of Fe^{57} enriched PdFe and their shift exponent were studied by McGrath who detected the ferromagnetic transition in the Fe^{57} Mössbauer spectra.[4] McGrath observed a shift, with a critical exponent of unity, suggestive of mean field ordering, which was unexpected and led to this study using direct magnetic measurements.

EXPERIMENT

Films were sputtered onto 6×2-mm rectangular substrates cut from glass cover slides. An rf cosputtering technique in a turbo pumped system was employed. The films were found to contain approximately 8% Fe as determined by Auger spectroscopy, which also measured the oxygen content of the films. One sample from the work of McGrath was available for our investigation. The films ranged in thickness from a few hundred to a few thousand angstroms. The thickness was measured by weight and by a Talysurf stylus device, which gave a greater thickness by several hundred angstoms than the weight measurements.

A study of the temperature dependence of the dc resistivity was made on the sample (JH) provided by McGrath. The balance of the measurements on this and our own samples were done in a SQUID susceptometer. In this susceptometer, the substrates were mounted in high vacuum on the end of a sapphire rod. The temperature of the rod was maintained by a feedback controller and measured with germanium and platinum resistance thermometers. The vertical position of the sample was controlled by a micrometer device above the cryostat, which located the sample in a superconducting astatic transformer, which sensed the flux in the film. The direct output of the (SHE) SQUID control was proportional to the dc magnetization of the film. In addition, the transformer was driven by a low frequency ac solenoid referenced to a lock-in detector. The output of the SQUID control was input to the lock-in with its output proportional to the rms ac susceptibility of the sample.

RESULTS

Figures 1 and 2 show results from four representative samples. Figure 1 is the SQUID signal proportional to the flux of the dc magnetization in the plane of the film in a longitudinal field$\simeq$0.1 Oe, trapped in the superconducting shield of the susceptometer. In Fig. 2, we show the lock-in output proportional to the ac susceptibility for an excitation of $\simeq$5 mOe peak to peak. The data shown are for cooling runs under these conditions, but negligible differences were observed when heating. No frequency dependence was observed in the 16–160-Hz frequency range of the apparatus.

Films 34, 39, and 46 are $\simeq$8% Fe and were made at Virginia Tech. Film JH with 7.8% Fe was made by McGrath at Johns-Hopkins.[4] Each film was Auger analyzed and

 0021-8979/84/062359-03$02.40

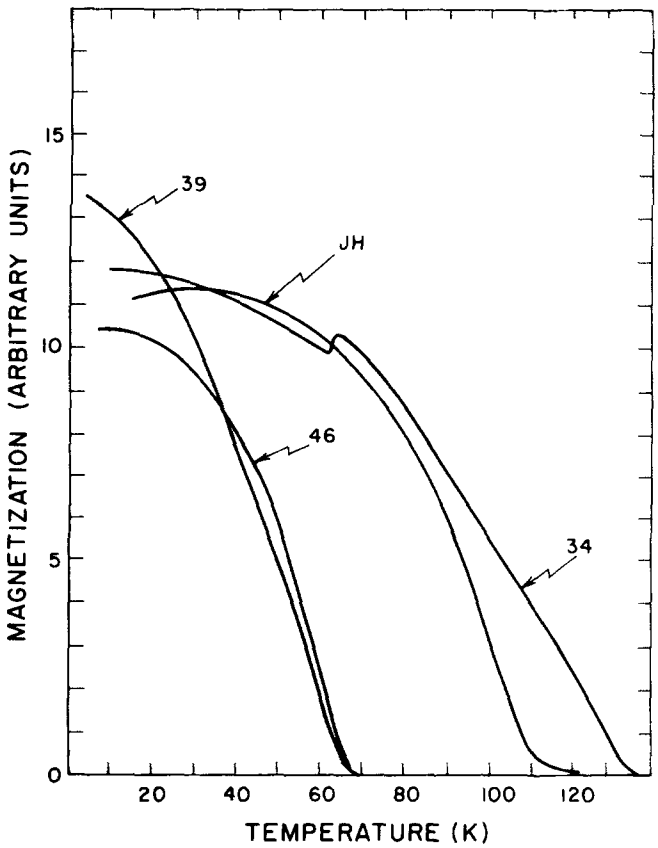

FIG. 1. The temperature dependence of the flux of the magnetization in the plane of four PdFe thin films with a 0.1-Oe longitudinal dc field.

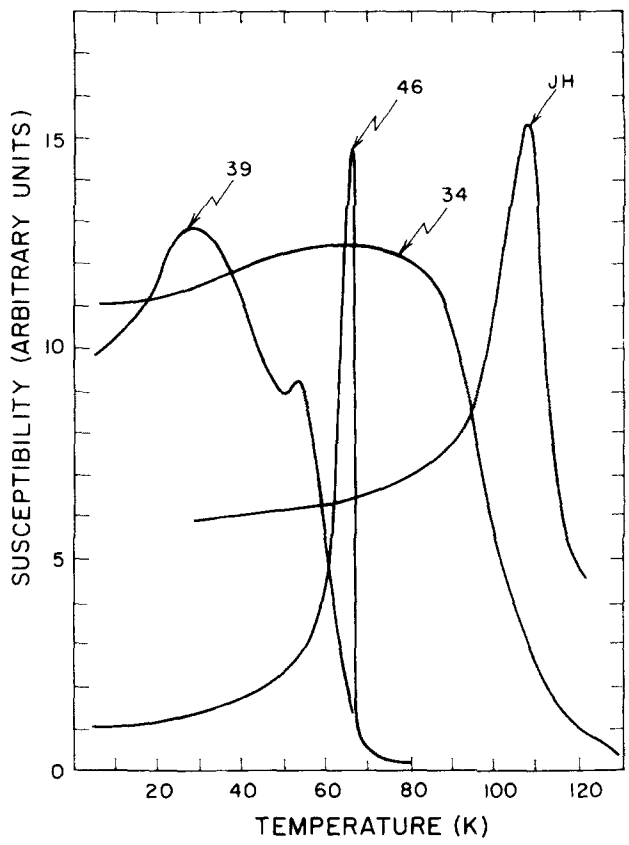

FIG. 2. The temperature dependence of the longitudinal ac magnetic susceptibility of four PdFe thin films with 5-mOe peak to peak excitation.

found free of contamination within the 1% resolution of the apparatus. Several films not shown were found to contain traces of oxygen. Those films had double transitions suggestive of spin glasses.[5]

Our film 46 and film JH show clean and similar behavior. A sharp peak occurs in the ac response at the temperature where the dc magnetization vanishes. We interpret this to be the Curie temperature. A fit of the data above this temperature to a Curie-Weiss function gives a consistent value of T_C. In film JH, the resistivity was also measured, and a change in the slope of its temperature dependence was found at approximately the same temperature, $T_C \simeq 110$ K, as found in our magnetic data. The magnetic data for film 46 yields $T_C \simeq 67$ K. This lower value was unexpected. Film 46 was thicker than JH and was expected to have a higher transition. Visual inspection showed it to be thicker and we measured a thickness of $\simeq 1000$ Å to a certainty of $\simeq 300$ Å. Film JH was reported to be $\simeq 440$ Å thick by McGrath.[4] McGrath's Mössbauer measurements were not performed on film JH, but his data on similarly prepared samples of the same composition predicted a Curie temperature of $\simeq 40$ K for film JH. Thus, there are two problems: Why do our magnetic and transport measurements yield over twice the critical temperature expected from the Mössbauer data? Why does the thinnest film give the highest transition? We will return to these questions after considering the data on the other films.

Our films 34 and 39 do not exhibit such clean transitions and sharp peaks as 46 and JH. The ac susceptibilities, Fig. 2, of films 34 and 39 are rather like those of materials that have a double transition, from paramagnet, to ferromagnet, to spin glass.[5] The dc magnetization of film 34, the thickest film, is also double peaked. If, however, we rely upon the assumption that the dc signal represents a spontaneous magnetization, which disappears at T_C, we find the $\simeq 5000$ Å film 34 to have $T_C \simeq 132$ K, and the $\simeq 3000$ Å film 39 to have $T_C \simeq 70$ K. Films 34, 39, 46, and our other cosputtered films (not shown) exhibit the expected shift to lower Curie temperatures with decreasing thickness, but film JH does not fit the trend.

CONCLUSIONS

These data must be augmented and refined before a shift exponent can be determined. The Curie temperatures found are below reported bulk values,[3] tending to support the existence of a shift, but the correct bulk values are not certain. Reported bulk values for Fe concentrations of order 8% vary by over 100 K and tend to fall around 200 K. This uncertainty appears to be due to the dependence of the value of the spin enhanced moment of the iron upon microstructure. Thus, quite unlike pure Fe, the critical temperature for ferromagnetic ordering of the giant moments is strain dependent, and strain inhomogeneity leads to very broad transitions. Thus, we conclude that Curie temperature determinations in bulk or thin films are not very significant unless the Fe moments are also determined and one knows the dependence of the critical temperature upon the moment. Our data must be strongly influenced by this strain effect.

In addition to bulk strains, there is differential substrate strain. We note that the thinner films have more well defined transitions. In these films, the substrate may uniformly dominate the entire film. As the thickness increases, the effect of the substrate should be less homogeneous and result in a broadened transition, as observed for films 34 and 39. Films 34, 39, and 46 were sputtered under similar conditions and should have similar bulk strain. Thus, the progression of Curie temperatures they exhibit may be the dimensional shift. Film JH was sputtered in a different system. Though we cannot absolutely rule out some error in Fe content or thickness, it is probable that the higher Curie temperature of JH is consequence of less bulk strain grown into the film during sputtering. This does not resolve the discrepancy between our measurements on this film and the Mössbauer study of similarly grown films.[4] One possibility is that the substrate of this particular JH film had an inferior thermal contact with the substrate stage and self-annealed during deposition.

The magnetic properties of PdFe are dependent upon microstructure and trace oxygen. Films appear to manifest this dependency to a greater degree than bulk alloy. Thus, we observe much variety in the nature of the magnetic phenomena, including spin glass type behavior. Films of 8% Fe in Pd appear to exhibit the expected shift of their Curie temperatures due to the crossover from bulk to two-dimensional ordering, but definite and quantitative results must await further studies to resolve the dimensional shift from the spurious effects of strain and microstructure. Much of the previous research on this alloy appears tof have been biased by experience with pure iron, where the Curie temperature does not depend upon microstructure.

ACKNOWLEDGMENTS

We are indebted to R. D. McGrath for providing a sample and helpful information and to S. P. Bowen for valuable conversations. Comments by J. C. Walker and K. V. Rao concerning strain effects are also appreciated.

[1]M. E. Fisher, J. Vac. Sci. Technol. **10**, 665 (1973).
[2]J. Tyson, A. H. Owens, J. C. Walker, and G. Bayreuther, J. Appl. Phys. **52**, 2487 (1981).
[3]G. J. Nieuwenhuys, Adv. Phys. **24**, 515 (1975).
[4]R. D. McGrath, Magnetic Behavior in PdFe Thin Films, Ph.D. Dissertation, Johns-Hopkins University (1979).
[5]G. J. Nieuwenhuys, B. H. Verbeek, and J. A. Mydosh, J. Appl. Phys. **50**, 1685 (1979).