The role of the spin-density wave and disorder in the density of states of sputtered Cr films

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Chromium plays a key role as a spacer layer in the well-studied landmark giant magnetoresistive (GMR) system, Fe/Cr. In these multilayered structures, most often produced through sputtering, the density of Cr states at the Fe/Cr interface is known to contribute strongly to the spin-dependent scattering that gives rise to the GMR behavior. Chromium itself holds a wealth of information about antiferromagnetism due to its unique spin-density wave (SDW) behavior. By varying the preparation conditions, we examine how stress and disorder alter the SDW and the density of states of Cr. We measured the specific heat of various Cr films from 2 to 300 K using our unique thin film microcalorimeters. Analysis of the low temperature specific heat allowed us to determine the Debye temperature and the electronic specific heat coefficient γ, which is proportional to the density of states at the Fermi surface. Fitting the low temperature heat capacity data to a Debye model shows a clear phonon softening in the more highly disordered sputtered films. From this, we are able to extract the band structure density of states and the strength of the electron-phonon coupling. Though it has been shown that the commensurate and incommensurate SDWs carve out varying parts of the Fermi surface, we find that the density of states at the Fermi energy is much more sensitive to disorder broadening than to the various SDW phases. © *2009 American Institute of Physics*.
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I. INTRODUCTION

Though chromium's role in the giant magnetoresistive (GMR) the Fe/Cr multilayer system is often glossed over as merely a nonmagnetic spacer layer, many recent studies have shown that understanding its magnetic behavior is crucial to understanding the complexities of the GMR structure (for a review, see Pierce *et al.*[1]). Chromium's antiferromagnetic spin-density wave (SDW) can be manipulated not just through the confinement of the Fe/Cr structure[2–4] but also through strain and alloying in both the bulk (for a review, see the work of Fawcett[5]) and in thin films.[3,6,7] The sensitive nature of the SDW has an impact on the total density of states (DOS) available at the Fermi surface due to the nesting responsible for the SDW and is therefore expected to play a role in GMR in the coupling mechanisms of the Fe/Cr multilayer system.[1,8,9]

The coupling of the Fe/Cr GMR system has been proven to be quite dependent upon disorder in the film.[1,10,11] The impact of disorder on chromium's magnetic characteristics has been well established also, particularly in sputter-deposited films where disorder was controlled by sputter gas pressure and substrate temperature.[7,12] This sensitivity allows one to affect the nature of the SDW through sputter deposition at various pressures.[7] With the well established qualitative links between disorder and chromium's SDW and the coupling in the Fe/Cr GMR structure and chromium's DOS, the question remains as to whether these effects are linked and/or if there is a relative significance in these effects in the oft-studied sputtered Fe/Cr GMR heterostructures.

II. EXPERIMENTAL DETAILS

In order to examine the links between SDW, disorder, and DOS, we have sputtered Cr films at various pressures onto our micromachined calorimeters.[13] We deposited a copper capping layer *in situ* to eliminate the effect of surface oxidation (especially important on the more disordered films[7]). Relaxation calorimetry was used to measure the specific heat of the Cr films from 2 to 300 K with the contributions of the copper layer and the device calculated from a separate addenda measurement. We then fit these data below ~20 K to C/T versus T^2 to obtain the Sommerfeld coefficient γ, proportional to $N(\varepsilon_F)$, and the Debye temperature Θ_D (Fig. 1 inset).

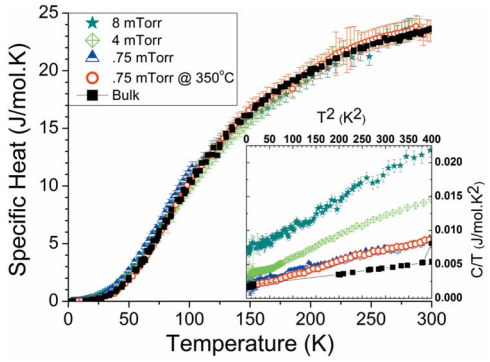

FIG. 1. (Color online) Specific heats of different sputtered Cr films compared to bulk data. Inset shows the C/T vs T^2 plot used to obtain the Debye temperature and Sommerfeld coefficient.

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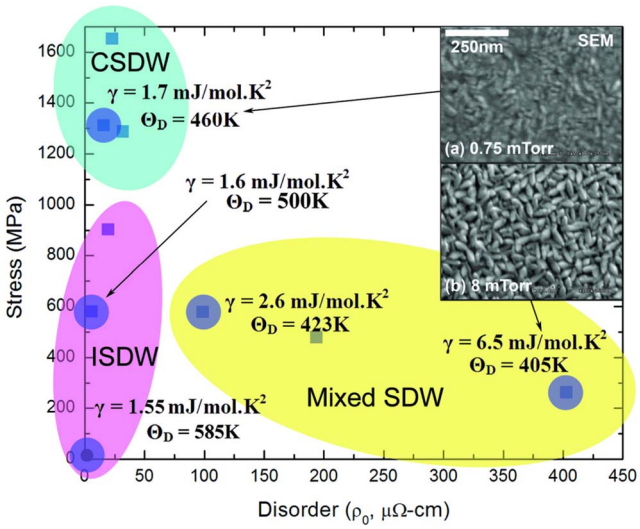

FIG. 2. (Color online) Magnetic phase diagram for sputtered Cr as a function of disorder and stress (Ref. 7). Overlayed onto the phase diagram are the Sommerfeld coefficient γ and the Debye temperature Θ_D obtained from specific heat measurements. The inset shows SEM data for samples grown at low (a) and high (b) pressure. Note that though (a) and (b) have roughly equivalent grain sizes, the high pressure sample has significantly larger grain boundaries.

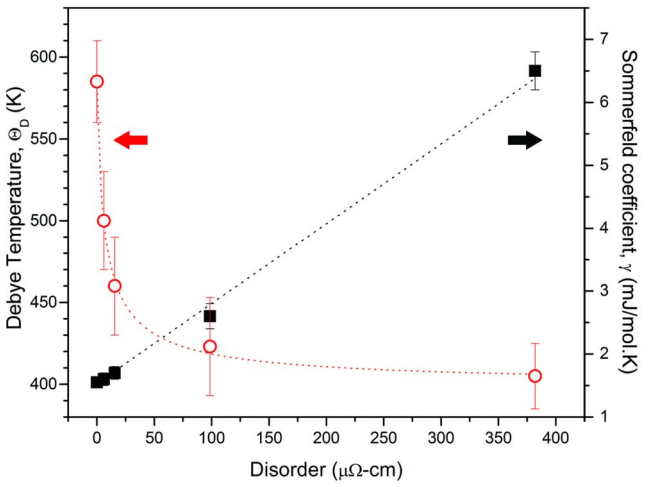

FIG. 3. (Color online) Measured γ (squares) and Θ_D (open circles) values as a function of disorder, where disorder is defined as the low temperature resistivity value ρ_0. Dotted lines are simply guides to the eye.

III. RESULTS

Figure 2 shows a phase diagram for Cr in the stress-disorder plane with the different types of SDW shown schematically.[7] We overlay the heat capacity results onto this phase diagram. Stress was determined through the use of a Tencor FLX-2320 to measure the wafer curvature before and after film deposition. Scanning electron microscopy (SEM) results in the inset of Fig. 2 illuminate the contrasting structure of the low pressure (a) and high pressure (b) films. These differing grain structures lead to a change in density from the bulk of as much as 23% in the most disordered films (Table I). This was taken into account when calculating the molar specific heat. Specific heat parameters γ and Θ_D are shown in Fig. 3 as a function of disorder, where disorder is characterized by the low temperature resistivity value ρ_0. Table I summarizes these results along with the calculated band structure DOS and electron-phonon coupling.

IV. DISCUSSION

Though there is no noticeable change in the Sommerfeld coefficient between the samples showing commensurate and incommensurate SDW behavior (CSDW and ISDW, respectively), there is a distinct correlation between γ and disorder, where increasing disorder shows a strong increase in γ (Fig. 3). With increasing disorder in these films, there is a corresponding increase in the size of the grain boundaries. TEM measurements show that these wide grain boundaries are often amorphous and, in general, the high-pressure films are exceptionally inhomogeneous.[7] As one would expect, this increasing disorder results in phonon softening, as evidenced in the decrease in Θ_D with increasing disorder (Fig. 3). Despite the differing C_P parameters for the various samples, they all approach the same high temperature limit (Fig. 1), which confirms that we are counting the modes for these disordered systems appropriately.[14]

Though the grains are on the order of nanometers (~25 nm), this leads to a surface to volume atomic fraction of 2% for even the film with the smallest grains, far less than the 30% seen in the 8.4 nm Pd nanocrystalline system exhibiting an enhanced heat capacity.[15] Therefore, confinement and surface effects are not a significant effect in these Cr films. Furthermore, extrinsic enhancements such as oxidation or water as seen in TiO_2 (Ref. 16) are not relevant because we have capped the sputtered Cr films *in situ*.

The strong dependence of γ on disorder suggests disorder broadening of the DOS. From a simple comparison of the

TABLE I. Table of experimental results on disordered sputtered Cr films compared to bulk Cr. Reference data is taken from Heiniger *et al.* (Ref. 22). Disorder is characterized by the low temperature resistivity ρ_0 (Ref. 25).

Growth conditions (mTorr)	Density (g/cm³)	ρ_0 ($\mu\Omega$ cm)	SDW state	γ (mJ/mol K)	Θ_D (K)	λ	$N(\varepsilon_F)$ (/eV-atom)
Bulk	7.14	···	ISDW	1.55	585	0.34[a]	0.49[b]
0.75 at 350 °C	7.14	6.1 ± 0.4	ISDW	1.6 ± 0.1	500 ± 30	0.35 ± .05	0.50 ± 0.04
0.75	7.14	15.6 ± 0.6	CSDW	1.7 ± 0.1	460 ± 30	0.37 ± .06	0.52 ± 0.04
4	6.15	98.6 ± 0.6	Mixed	2.6 ± 0.2	423 ± 30	0.51 ± 0.10	0.73 ± 0.10
8	5.53	382 ± 1	Mixed	6.5 ± 0.3	405 ± 20	0.97 ± 0.15	1.4 ± 0.2

[a]This value is obtained from the heat capacity results (Ref. 22) but there is a wide range of λ in the literature (Refs. 20, 23, and 24).
[b]See Laurent *et al.* (Ref. 19).

electronic lifetime in a disordered transition metal to that of the bulk, $\Delta E \Delta t = \hbar$ yields a broadening of 0.5–1 eV.[17,18] The Fermi energy of the bulk lies at a minimum,[19] so small changes due to disorder have large effects on the DOS.

The Sommerfeld coefficient $\gamma \propto N(\varepsilon_F)(1+\lambda)$, where $N(\varepsilon_F)$ is the band structure DOS and λ is the electron-phonon coupling constant. An increase in the band DOS also has an effect on screening and, thus, electron-phonon coupling. According to McMillan's theory on transition metal superconductivity in the weakly coupled case,

$$\lambda = \frac{N(\varepsilon_F)\langle \mathcal{I} \rangle^2}{M\langle \omega \rangle^2},$$

where $\mathcal{I}$ represents the electron-phonon matrix element.[20] It has been shown that the $\langle \mathcal{I} \rangle^2/(M\langle \omega \rangle^2)$ is roughly constant for transition metals,[21] meaning any change in the phonon spectrum results in a compensating change in the coupling element. Thus, changes in λ are related strictly to changes in $N(\varepsilon_F)$, and from this relationship we can calculate $N(\varepsilon_F)$ and λ given γ and Θ_D (Table I).

One sees that for the ISDW and CSDW states, the band structure DOS does not change (to within error). Any difference due to the AFM is expected to be small[26] given that only a subset of the Fermi surface participates in the AFM interaction. Because ε_F lies near a minima in the DOS,[19] small shifts and/or broadening are not going to change $N(\varepsilon_F)$ much. However, in the mixed SDW films where disorder is much greater, we see a large increase in $N(\varepsilon_F)$. This difference is approximately equivalent to the change in $N(\varepsilon_F)$ for crystalline and amorphous Mo,[27] which is isoelectronic to Cr. It should also be noted that there is an observed monotonic increase in the electron-phonon coupling with decreasing Θ_D. This agrees with the resistivity data on these samples,[25] where we see an increase in the Bloch-Gruneisen prefactor and thus λ_{tr} (the transport-derived electron-phonon coupling term[28]).

V. CONCLUSIONS

We have used our thin film calorimeters to measure the specific heat of the magnetron sputtered Cr films. From the low temperature data, we were able to obtain information about the phonon and electronic DOS as well as the electron-phonon coupling. We see no observable difference in the DOS between ISDW and CSDW Cr films. However, disorder plays a strong role in the films, not only in increasing the DOS through broadening but also in increasing the electron-phonon coupling through phonon softening and additional screening from the increased $N(\varepsilon_F)$. This increase in the DOS is important to consider when discussing the mechanism by which disorder may increase the GMR of the Fe/Cr multi-layer system.

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