Competing magnetic interactions in metastable Gd–Cr alloys

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Gadolinium and chromium are ferromagnetic and antiferromagnetic respectively. Thus we may expect that alloys of these elements will reveal the evolution between these types of magnetic order. We have used high rate magnetron sputtering to prepare metastable $\text{Gd}_x\text{Cr}_{1-x}$ alloys over the entire composition range. The alloys with $0.2 < x \leq 1$ are hexagonal close-packed, as is pure Gd. By using SQUID and vibrating sample magnetometry, we have investigated both a ferromagnetic and a spin-glass like state. A magnetic phase diagram has been determined.

Alloys of gadolinium and many of the transition metals (Fe, Co, Ni, Mn) have been investigated extensively, and shown to have interesting and in some cases applicable magnetic properties [1,2]. In contrast, little work on Gd–Cr alloys has been reported. Although Gd and Cr are immiscible under equilibrium conditions [3], we expect that metastable alloys may be formed by cosputtering elemental Gd and Cr under appropriate conditions [4].

The $\text{Gd}_x\text{Cr}_{1-x}$ system is also interesting from a fundamental point of view. Gd and Cr are respectively ferromagnetic and antiferromagnetic. Therefore, in a sequence of alloys with progressively lower Gd content, we expect ferromagnetic ordering to be gradually destroyed and replaced by antiferromagnetic ordering. In the intermediate composition region, we may observe more complex magnetic behavior; for instance, a spin-glass state may be observed at low temperature.

In this paper we report the fabrication and structural characterization of $\text{Gd}_x\text{Cr}_{1-x}$ alloys across the entire composition range. Further, we will discuss the magnetic behavior of the alloys with $x > 0.20$ which are shown to be single phase hcp.

$\text{Gd}_x\text{Cr}_{1-x}$ alloys ($0 \leq x \leq 100$) were prepared by dc magnetron sputtering from composite targets. The substrates were Corning 7059 glass plates, held in thermal contact with a copper plate cooled by a liquid nitrogen reservoir. Details of the sputtering process may be found elsewhere [5]. Throughout this work, we will refer to the $\text{Gd}_x\text{Cr}_{1-x}$ films by x , the relative atomic concentration of Gd. The structure of alloys was studied by using X-ray diffraction in the θ - 2θ geometry. The magnetic measurements were performed using SQUID and vibrating sample magnetometers.

X-ray diffraction measurements indicate that the samples with $x \geq 0.30$ are single phase hcp, while the $x = 0.20$ samples shows evidence of both bcc and hcp phases. We have found that in the hcp region the lattice parameters a and c vary slowly with Gd concen-

tration changing by only a few percent over the whole hcp range.

In fig. 1, we present the temperature dependence of the magnetization. For these measurements, the samples were cooled to 5 K at zero field, and the measurements were then made as the sample temperature was raised in an applied field of 1 kOe. All of the samples show a peak at low temperature followed by a gradual decline at higher temperatures. Furthermore, the samples with $x \geq 0.40$ reveal transitions suggestive of ferromagnetic ordering. We have estimated values of this transition temperature (T_c) by plotting M^2 vs T and extrapolating the linear portions of the curves to zero magnetization. We have also measured the temperature dependence of the magnetization under an external field of 10 kOe, up to temperatures of 450 K. We find that for $x \geq 0.40$ a Curie–Weiss law fits the data well in the high temperature region. We thus identify the alloys with $x \geq 0.40$ as ferromagnetic. Our results for T_c are summarized in fig. 2, in which we propose a magnetic phase diagram for the $\text{Gd}_x\text{Cr}_{1-x}$ alloy system.

In order to better characterize the peaks of magnetization at low temperatures shown in fig. 1, we have examined the zero-field-cooled and field-cooled temperature dependences of the magnetization under a

Fig. 1. The temperature dependence of the zero-field-cooled magnetization in a fixed applied field of 1 kOe.

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much smaller external field (25 Oe). The behavior of the samples with $x \leq 0.30$ is markedly different from the behavior of the samples of higher Gd concentration. For $x \leq 0.30$ we observe a cusp in the zero-field-cooled branch of the magnetization, which corresponds with the onset of irreversibility between the field-cooled and zero-field-cooled branches. The cusp and the irreversibility of $\chi(T)$ represents a typical spin glass to paramagnetic transition [6]. We thus suggest that for $x \leq 0.30$ a spin-glass state exists at low temperature.

The alloys with $x \geq 0.40$ show strikingly different behavior. Although the magnetization still shows a peak in the zero-field-cooled branch, this peak is broader than that seen in the alloys of lower Gd concentration. Furthermore, under the small applied field (25 Oe) the magnetization is seen to be irreversible for all temperatures up to the high temperature transition. The broad peak in the susceptibility persists, even up to $x = 0.90$. This suggests a transition to a mixed magnetic state, or to a reentrant state, as has been observed in many other alloy systems [7]. We denote this transition temperature T_f . Our results for T_f are summarized in fig. 2.

We have also studied the response of the magnetization to applied fields at temperatures of 5 and 75 K. Surprisingly, none of the alloys is completely saturated at either temperature, even at an applied field of 45 kG. Fig. 3 depicts the evolution of the field response

Fig. 3. The normalized magnetization vs. applied field at $T = 75$ K for $x = 0.50, 0.60$ and 0.90 .

with increasing Gd concentration. The absence of saturation above T_f further suggests the interpretation that the low temperature state of the gadolinium rich samples is magnetically inhomogeneous, rather than a reentrant state.

We have produced single phase hcp $\text{Gd}_x\text{Cr}_{1-x}$ alloys over a broad range of compositions. We have studied the magnetic ordering of these alloys, and have determined the magnetic phase diagram shown in fig. 2. The alloys with $x \leq 0.30$ exhibit only a spin glass phase. The alloys with $x \geq 0.40$ are ferromagnetic at high temperatures and also exhibit a spin glass-like peak in the susceptibility at low temperature. The width of these peaks, and the absence of saturation demonstrated in fig. 3 suggest that the low-temperature state is magnetically inhomogeneous, rather than a true reentrant spin glass.

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Fig. 2. Magnetic phase diagram for $\text{Gd}_x\text{Cr}_{1-x}$ alloys, showing paramagnetic (P), spin-glass (SG) and inhomogeneous regions (I, I').