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Rapidly quenched $Fe_x Ta_{100-x}$ alloys^{a)}

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Binary alloys of Fe_x Ta_{100-x} were made by high-rate sputtering. A sharply defined boundary separates the composition ranges in which crystalline alloys $(93 \le x \le 100)$ and amorphous alloys $(15 \le x \le 90)$ are formed. The amorphous alloys with Fe concentrations up to the threshold of $x_c \simeq 65$ are nonmagnetic. For the samples with $x > x_c$, the magnetic ordering temperature (T_c) varies monotonically with Fe content to 200 K for a-Fe₉₀Ta₁₀. However, the crystalline alloys, with slightly higher Fe content, are strongly ferromagnetic. Their T_c 's are several hundred degrees higher, accompanied by large increases in magnetic moment and hyperfine fields. This discontinuity illustrates the sensitivity with which the magnetic properties of Fe-transition metal alloys depend on structure. In contrast, no discontinuity has been observed in Co-Ta alloys.

I. INTRODUCTION

The magnetic properties of binary alloys of Fe-TM and Co-TM, where TM is an early transition metal, have been a subject of interest in recent years. $^{1-6}$ The use of vaporquench methods, such as sputtering and evaporation, has greatly facilitated such studies. This is because equilibrium phase diagrams in most cases accommodate only very small solubility (<2%), which is too small to be useful for the studies of the evolution of the magnetic properties. Vaporquench methods allow alloys, either amorphous or metastable crystalline depending on the composition, to be fabricated over wide composition ranges. In this work, we report the studies of sputtered Fe-Ta alloys.

II. EXPERIMENT

Samples of Fe_xTa_{100-x} ($15 \le x \le 100$) were prepared using a magnetron sputtering device. Sputtering targets were made from homogeneous mixtures of pure Fe (99.9%) and pure Ta(99.9%). In all cases, thick films were obtained from liquid-nitrogen cooled substrates of various kinds. Samples used in the x-ray diffraction and Mössbauer measurements were 6-24 μ m thick, often with the substrates removed. Both conventional x-ray diffraction with Cu radiation and energy dispersive x-ray diffraction methods were used in the structure study. The hyperfine interactions were measured by a Mössbauer spectrometer using a ⁵⁷Co in Rh source. The magnetic ordering temperatures were determined by the zero velocity scan method.

III. RESULTS AND DISCUSSION

A. Structure and composition range of amorphous alloys

Binary alloys of $Fe_x Ta_{100-x}$ were prepared in the range of 15–100 at. % Fe. The amorphous state, as determined by x-ray diffraction, is realized over a wide composition range

of $15 \le x \le 90$. Typical x-ray diffraction patterns are shown in Fig. 1. For the amorphous alloys, the position of the first peak in the diffraction pattern $(q \text{ in units of } \mathring{A}^{-1})$ increases progressively with increasing x, as shown in the Fig. 2.

This monotonic increase of q with Fe concentration comes primarily from the fact that the atomic radius of Fe is smaller than that of Ta. It is also noteworthy that throughout the wide composition range, the value of q of a-Fe-Ta varies smoothly without a change of slopes. In contrast, in a few amorphous Fe-TM alloys, there is an apparent change of slope of the q vs x dependence suggesting a subtle structure difference between the Fe-rich and Fe-poor alloys.

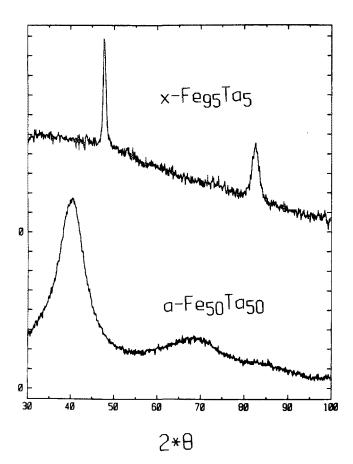


FIG. 1. X-ray diffraction patterns of crystalline and amorphous Fe-Ta alloys.

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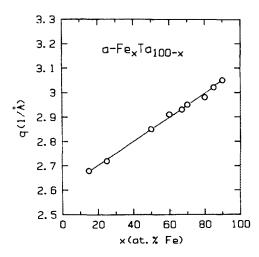


FIG. 2. Variation of the position of the first diffraction peak for Fe_xTa_{100-x} amorphous alloys as a function of x.

The composition range $(15 \le x \le 90)$ in which amorphous alloys of Fe-Ta are achieved agrees reasonably well with a recent model of Egami and Waseda.8 In this model, the composition range is dictated by the difference of the atomic volumes. Using the values of 2.097 and 3.30 Å³ for the atomic volumes of Fe and Ta, respectively, and the formula from Ref. 8, one obtains a composition range of $27 \le x \le 83$ for a-Fe_x Ta_{100-x}, in reasonable agreement with the experimental results.

While Fe₉₀Ta₁₀ is amorphous, the samples with slightly higher Fe content $(x \ge 93)$ are crystalline with bcc structure and very different magnetic properties.

B. Magnetic ordering temperature

The magnetic ordering temperature (T_c) of amorphous $Fe_x Ta_{100-x}$ as a function of x is shown in Fig. 3. In all cases, the as-prepared samples were used for the measurements. The value of T_c , all of which are below 200 K, decreases monotonically with decreasing Fe concentration. The threshold for magnetic ordering was found to be near $x_c \simeq 65$ at. % Fe. Below the threshold $(x_c < 65)$, the samples are nonmagnetic. The magnetic hyperfine interaction and magnetization measurements, to be discussed later, show that in the

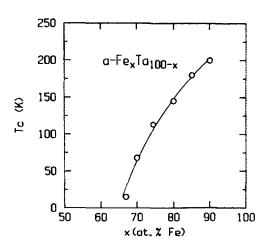


FIG. 3. The magnetic ordering temperatures of Fe_xTa_{100-x} amorphous alloys.

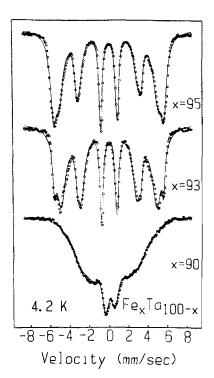


FIG. 4. Mössbauer spectra of Fe-Ta alloys at 4.2 K of crystalline Fe₉₅Ta₅, Fe₉₃Ta₇, and amorphous Fe₉₀Ta₁₀.

present system as in other Fe-early transition systems, the disappearance of the Fe moment is responsible for the loss of magnetic order.

For slightly higher Fe content, x > 93, the hyperfine interaction indicated the presence of two crystalline magnetic phases, both with T_c 's of more than 800 K, several hundred degrees higher than those of the amorphous samples. Structural transformation of these alloys at high temperatures prevents the determination of these T_c .

C. Magnetic hyperfine interaction

Mössbauer spectra of the $Fe_x Ta_{100-x}$ alloys with x = 90, 93, and 95 at 4.2 K are shown in Fig. 4. As mentioned above, the samples with x = 90 is amorphous, whereas the samples with x = 93 and 95 are crystalline. The difference in their spectra is obvious. For the crystalline samples, there are two bcc phases with magnetic hyperfine fields of

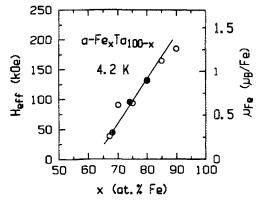


FIG. 5. Magnetic hyperfine fields at 4.2 K of amorphous Fe-Ta alloys. The solid circles represent magnetic moment data taken from Ref. 10.

340 and 308 kOe. These two phases were identified as α -Fe and saturated Fe-Ta crystalline alloys. For amorphous Fe₉₀Ta₁₀, despite very high Fe concentration, the value of H_{eff} is only about 185 kOe, which is 60% of that of crystalline Fe₉₃Ta₇. This large reduction is a consequence of structural difference. Naoe et al. have observed a similarly large change in saturation magnetization across the phase boundary. In contrast, the magnetization of Co_x Ta_{100-x} changes smoothly at the boundary separating the amorphous and the crystalline alloys without discontinuities. Data reported to date indicate that the properties of Fe-early transition metal alloys generally are very sensitive to local structure and coordination number whereas the properties of the Co-based alloys are not. 1-6 One notes also that pure Fe and pure Co exhibit similar characteristics. For example, both hcp Co and fcc Co are strongly ferromagnetic with similar magnetic moments whereas bcc α -Fe and fcc γ -Fe are entirely different. This might suggest that amorphous alloys resemble more of fcc structure than bcc structure.

Another plausible explanation comes from band structure considerations. In Co-TM alloys, the so-called "strong" magnetism is realized in which the majority subband is filled and holes exist in the minority subband only. In Fe-TM alloys, on the other hand, both subbands are partially filled. Since magnetism is determined by the difference of the states in the two subbands, it naturally explains the different behavior of Co-TM and Fe-TM alloys.

The values of $H_{\rm eff}$ (open circles) of amorphous Fe-Ta alloys decrease with decreasing Fe concentration as shown in Fig. 5. Also shown are the magnetic moments ($\mu_{\rm Fe}$) data (solid circles) reported by Fukamichi and Gambino. ¹⁰ The conversion factor between $H_{\rm eff}$ and $\mu_{\rm Fe}$ is 145 kOe/ $\mu_{\rm B}$, a number close to those found for many Fe-containing alloys.

The results clearly show that $H_{\rm eff}$ scales with $\mu_{\rm Fe}$. Equally important, both $H_{\rm eff}$ and $\mu_{\rm Fe}$ are extrapolated to zero at $x{\simeq}63$, which is near the magnetic threshold of $x_c{\simeq}65$. This establishes that the loss of magnetic order at $x{<}x_c$ is caused by the disappearance of the Fe moment.

IV. CONCLUSIONS

The wide compositional range of Fe-Ta alloys has been successfully made by sputtering. The magnetic phase diagram has been determined and the magnetic threshold found to occur at $x_c \approx 65$ at. % Fe. The disappearance of the magnetic moment is responsible for the loss of magnetic order. An abrupt change in magnetic properties has been observed at the boundary separating the amorphous and the crystalline states. This is caused by the difference in the atomic arrangement and the electronic structure between crystalline and amorphous solids.

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