

# SPIN POLARIZED PHOTOEMISSION AND MAGNETIC CIRCULAR DICHROISM STUDY OF FeAl THIN FILMS

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## ABSTRACT

It is well known that the equiatomic FeAl alloy crystallizes in a paramagnetic CsCl structure and is very stable in a wide temperature range owing to a significant charge transfer from Al to Fe. A presence of structural defects normally enhances the magnetic and magneto-optical properties of this alloy. In this study, spin-resolved photoemission and magnetic circular dichroism (MCD) were carried out on both ordered and disordered Fe<sub>0.52</sub>Al<sub>0.48</sub> alloy films. The disordered state in the alloy films was obtained by a vapor quenching deposition on cooled substrates. It is shown that the order-disorder transition in the Fe<sub>0.52</sub>Al<sub>0.48</sub> alloy films leads to a significant change in the spin polarization. From the MCD results the orbital and spin magnetic moments of the constituent atoms are obtained. According to the sum rule, the spin and orbital magnetic moments of Fe in the disordered FeAl film are  $\mu_{spin}^{SR} = 0.81\mu_B$  and  $\mu_{orb}^{SR} = 0.14\mu_B$ , respectively. The spin magnetic moment is also evaluated to be  $\mu_{spin}^{BR} = 0.77\mu_B$  by the branching ratio method employing a photon polarization of 90 %.

## I. INTRODUCTION

The study of Fe-Al intermetallic compound attracts a wide interest due to its high corrosion and oxidation resistance, and interesting magnetic properties. Equiatomic FeAl alloy crystallizes in a CsCl (B2) structure.<sup>(1)</sup> Because of the significant charge transfer from Al to Fe, this alloy is very stable in a wide temperature range. In the perfectly ordered stoichiometric FeAl alloy, Fe and Al atoms form interpenetrating primitive cubic lattices where Fe atoms have eight Al atoms as the nearest neighbor and vice versa. This alloy is not ferromagnetically ordered since the ferromagnetic nearest neighbor is absent. In contrast to the ordered state of FeAl alloy, the constituent atoms randomly occupy the sites of bcc lattice in the disordered one. This change in symmetry leads to the occurrence of local magnetic moments.

In this study, we investigate the influence of the structural order-disorder transition in the FeAl alloy film on the magnetic properties using spin-polarized photoemission spectroscopy (SPES) and magnetic circular dichroism (MCD). The magnetic moments of the constituent Fe atoms were also evaluated.

## II. EXPERIMENT

Fe<sub>0.52</sub>Al<sub>0.48</sub> alloy films of 150 nm thick were prepared by means of flash

evaporation onto Si substrate in a high vacuum of  $1 \times 10^{-5}$  Pa. The deposition rate was about 2 nm/s. An equilibrium ordered state in the FeAl alloy films was obtained by the deposition onto the heated substrates up to 680 K. In order to obtain a disordered state in the FeAl films, a vapor quenching deposition onto a cooled substrate was employed. The substrate temperature during the deposition was about 150 K.

The structural analysis of the films was performed by transmission electron microscopy (TEM). The field dependencies of magnetization for the ordered and disordered films were measured using a vibrating sample magnetometer (VSM) at room temperature in a magnetic field up to 1.8 T for in-plane geometry.

The MCD measurements were carried out at the 2B1 beam line, which is equipped with a spherical grating monochromator, of the Pohang Light Source (Pohang, Korea). The absorption process was monitored in the "photoelectron yield" mode at each photon energy, typically by measuring the sample drain current. The measured absorption spectra were normalized to the photon flux via dividing by the output of an upstream  $I_0$  detector. The FeAl films were magnetized using a permanent magnet set which enables to flip the magnetization direction.

The spin-polarized photoemission spectroscopy (SPPES) were performed at the U5UA undulator beam line of the National Synchrotron Light Source (Upton, N.Y.). A mini-Mott spin detector was employed for the spin analysis.<sup>(2)</sup> In order to eliminate an instrumental asymmetry, the sample was first magnetized in one direction and the intensities scattered left and right ( $I_L^+, I_R^+$ ) into the spin detector measured. The measurement was then repeated with the sample magnetized in the opposite direction ( $I_L^-, I_R^-$ ). From these four measurements, the polarization P is obtained via

$$P = \frac{1}{S} \frac{(I_L^+ I_R^-)^{1/2} - (I_L^- I_R^+)^{1/2}}{(I_L^+ I_R^-)^{1/2} + (I_L^- I_R^+)^{1/2}},$$

where S is the effective Sherman function which has a value of 0.07 in our case.<sup>(3)</sup>

### III. RESULTS AND DISCUSSION

According to the results of TEM study<sup>(4)</sup> the film deposition onto a substrate at 680 K leads to a formation of the ordered structure with a mean grain size of about 30 nm. A decrease in the substrate temperature down to 150 K leads to the formation of a considerably disordered polycrystalline alloy films with a mean grain size of 10 - 15 nm, which was identified by a few smeared diffraction rings.

Figure 1 shows the in-plane magnetization loops,  $M(H)$ , of the ordered and disordered alloy films. For the ordered film, the magnetic moment increases with an applied magnetic field up to 1500 Oe, followed by a gradual decrease, which can be produced by a mixture of ferromagnetic and nonmagnetic phases.

The transition of the  $Fe_{0.52}Al_{0.48}$  alloy films from the ordered state into the disordered one causes a significant growth of the magnetic moment and the field dependence of M also changes. It is seen that the  $M(H)$  curve for the disordered state exhibits two different parts: a rapid increase of the magnetization in a comparatively low magnetic field (below 200 Oe), and a gradual increase with a

field up to 8000 Oe. It indicates that there are both soft magnetic phases and phases in which an external magnetic field induces magnetic moments.

The MCD spectra for Fe L<sub>2,3</sub> edge are presented in Fig. 2 and Fig. 3 for the ordered and disordered alloy films, respectively. After removing white line peaks,  $I^\pm(2p^{3/2})$ ,  $I^\pm(2p^{1/2})$ , and  $C^\pm$  were obtained, where  $I^\pm$  and  $C^\pm$  denote the integrated intensities and backgrounds, respectively, for the parallel (+) and antiparallel (-) configurations between the photon polarization and the sample magnetization.

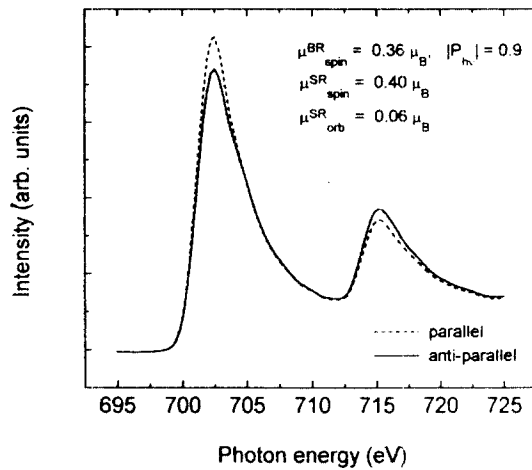


Fig. 2. MCD spectra from the ordered Fe<sub>0.52</sub>Al<sub>0.48</sub> alloy films.

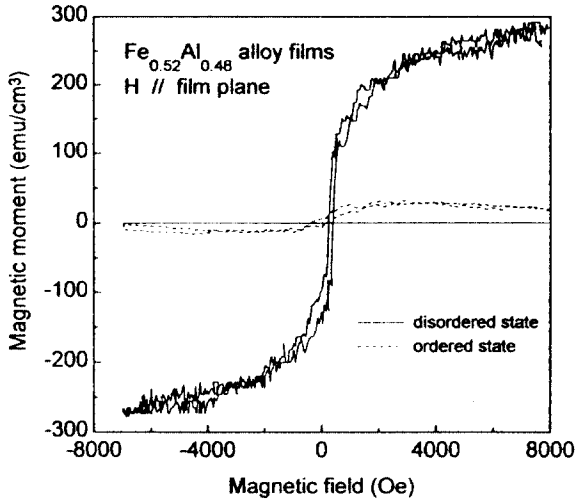


Fig. 1. Hysteresis loops for the ordered and disordered Fe<sub>0.52</sub>Al<sub>0.48</sub> alloy films

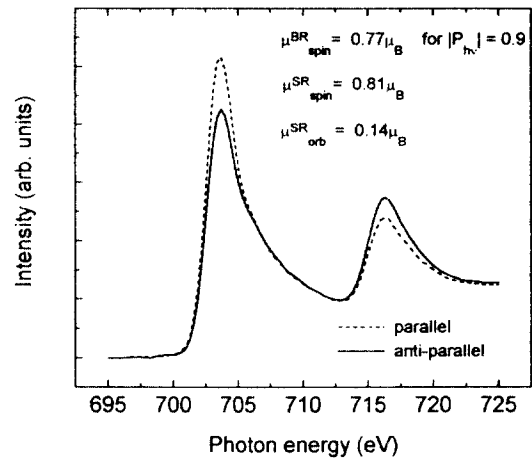


Fig. 3. MCD spectra from the disordered Fe<sub>0.52</sub>Al<sub>0.48</sub> alloy films

The branching ratio (BR) and the spin magnetic moments of 3d elements are related through the following equations<sup>(4)</sup>:

$$BR^\pm = \frac{I^\pm(2p^{3/2})}{I^\pm(2p^{3/2}) + I^\pm(2p^{1/2})} \quad \text{and} \quad \mu_{spin}^{BR} = \frac{4N}{|P_{hv}|} \left[ \frac{BR^+ - BR^-}{BR^+ + BR^-} \right],$$

where  $N$  and  $|P_{hv}|$  denote the number of 3d valence holes and the circular polarization of photons, respectively.

While the branching ratio approach explicitly retain a dependence upon polarization, a more sophisticated derivation lead to the sum rules (SR) for the orbital<sup>(6)</sup> and spin<sup>(7)</sup> magnetic moments. For 3d elements and 2p→3d transitions these are summarized as follows:

$$\mu_{orb}^{SR} = \langle L_z \rangle = \frac{4}{3} N \frac{\left[ \frac{\Gamma^+(2p^{3/2})}{C^+} + \frac{\Gamma^+(2p^{1/2})}{C^+} \right] - \left[ \frac{\Gamma^-(2p^{3/2})}{C^-} + \frac{\Gamma^-(2p^{1/2})}{C^-} \right]}{\frac{\Gamma^+(2p^{3/2})}{C^+} + \frac{\Gamma^+(2p^{1/2})}{C^+} + \frac{\Gamma^-(2p^{3/2})}{C^-} + \frac{\Gamma^-(2p^{1/2})}{C^-}}$$

and

$$\mu_{spin}^{SR} = 2 \langle S_z \rangle = 6N \frac{\frac{\Gamma^+(2p^{3/2})}{C^+} - \frac{\Gamma^-(2p^{3/2})}{C^-}}{\frac{\Gamma^+(2p^{3/2})}{C^+} + \frac{\Gamma^+(2p^{1/2})}{C^+} + \frac{\Gamma^-(2p^{3/2})}{C^-} + \frac{\Gamma^-(2p^{1/2})}{C^-}} - 3 \langle L_z \rangle - 7 \langle T_z \rangle.$$

If the magnetic dipole term  $\langle T_z \rangle$  remains very small as compared with  $\langle S_z \rangle$ , the spin and orbital magnetic moments are readily obtained.

The BR and the SR results are compared in Table 1.  $|P_{hv}|$  was 90%. The spin magnetic moment of Fe in the disordered film is about twice bigger than that in the ordered one in both BR and SR analyses. These results do not agree well with the aforementioned magnetization data which showed at least five times difference in magnetization between the ordered and disordered films. This discrepancy may be understood by considering that the surface magnetism contributes to the MCD results, and that the FeAl films are probable to be not magnetically homogeneous along their depth from the surface.

Another interesting feature of the MCD results in Table 1 is that the spin magnetic moments obtained by the BR approach are always smaller than those obtained by the SR approach. This is in a good agreement in the trend with the previous results of the Fe/Co(001) system and Fe/Pt multilayers reported by Tobin et al.<sup>(5)</sup> It seems that the magnetic dipole term which is usually neglected in the SR analysis also has a contribution to the spin magnetic moment, especially for noncentro-symmetric systems, e.g., with surfaces and interfaces as indicated in Ref. 9.

Table 1. Comparison of the spin and orbital amgnetic moments of Fe in the ordered and disordered Fe<sub>0.52</sub>Al<sub>0.48</sub> alloy films, obtained by using the BR and SR approaches.

	$\mu_{spin}^{BR}$	$\mu_{spin}^{SR}$	$\mu_{orb}^{SR}$
ordered Fe <sub>0.52</sub> Al <sub>0.48</sub>	0.36 $\mu_B$	0.40 $\mu_B$	0.06 $\mu_B$
disordered Fe <sub>0.52</sub> Al <sub>0.48</sub>	0.77 $\mu_B$	0.81 $\mu_B$	0.14 $\mu_B$

Spin-polarized photoemission spectra for the ordered and disordered Fe<sub>0.52</sub>Al<sub>0.48</sub> films are presented in Figs. 4 and 5 together with the spin polarizations. The order-disorder structural transformation enhances the spin polarization doubly in agreement with the MCD results. By comparing with the reported magnetic moment<sup>(8)</sup> and spin polarization<sup>(9)</sup> of the pure Fe, we found the spin magnetic moments of the constituent Fe atoms in the Fe<sub>0.52</sub>Al<sub>0.48</sub> alloy film to be  $\mu_{spin} = 0.25 \mu_B$  and  $0.55 \mu_B$  for the ordered and disordered states, respectively.

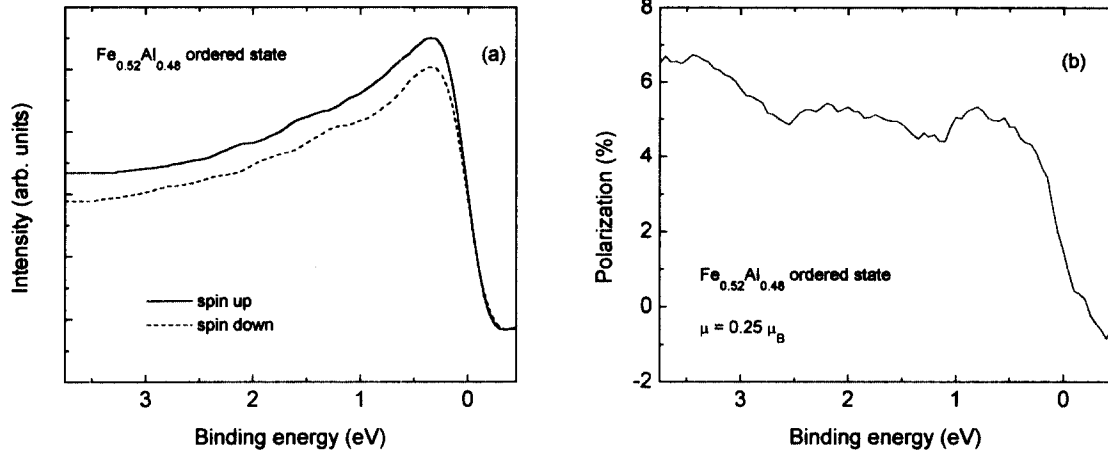


Fig. 4. SPPES spectra (a) and spin polarization (b) from the ordered  $\text{Fe}_{0.52}\text{Al}_{0.48}$  alloy film.

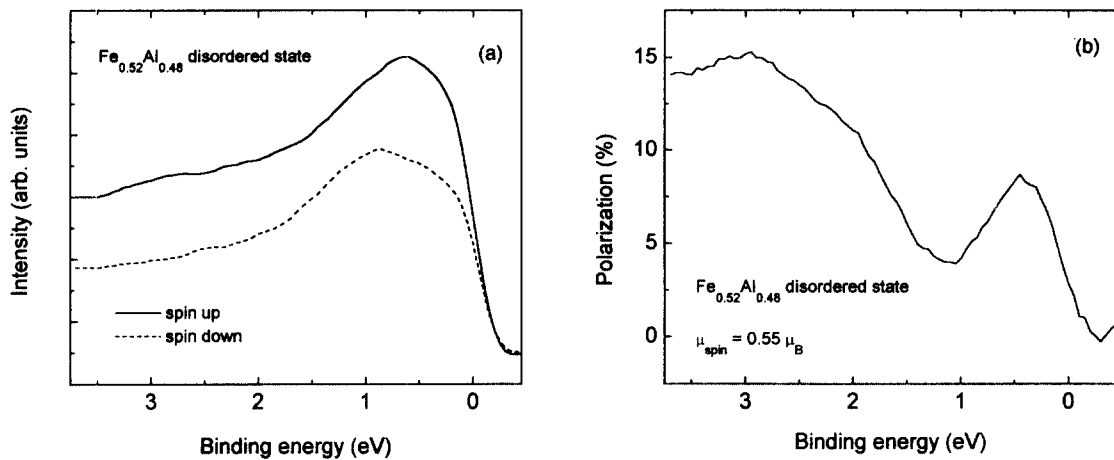


Fig. 5. SPPES spectra (a) and spin polarization (b) from the disordered  $\text{Fe}_{0.52}\text{Al}_{0.48}$  alloy film.

These lower values of the spin magnetic moment than those by the MCD analysis can be attributed to the probing-depth difference between MCD and SPPES. Owing to the lower kinetic-energy valence-band photoelectrons, SPPES are more surface-sensitive than MCD which probes photoelectrons from core-levels induced by x-ray. Hence it is thought that a magnetic structural difference between the top surface layers and the underlying ones might affect the results. Another possibility is a contamination during the SPPES measurements. Even though the surface had been thoroughly cleaned before each SPPES measurement, the sample surface was apt to be contaminated even in an ultrahigh vacuum during the measurement of a long data acquisition time. In this case, the SPPES results are more likely to be affected than the MCD ones.

#### IV. CONCLUSIONS

Ordered and disordered  $\text{Fe}_{0.52}\text{Al}_{0.48}$  alloy films were prepared by the flash evaporation technique. The field dependence of magnetization shows that the

order-disorder structural transition enhances the magnetic moment of the alloy films by about five times. The SPPES and MCD analysis indicate that the spin magnetic moment of the constituent Fe atoms and the spin polarization are also enhanced about twice. It is reasoned that this discrepancy between the magnetization data and SPPES/MCD results comes from an inhomogeneous magnetic structure along the depth from the sample surface. The spin magnetic moment obtained by the BR approach is always smaller than that by the SR approach partly because the neglected magnetic dipole term play a role for the spin magnetic moment. The difference in the analysed value of the spin magnetic moment by SPPES and MCD may indicate a magnetic structural difference between the top surface layers and the underlying ones.

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