Variable Moments and Changing Magnetic Behavior of Thin-Film FeNi Alloys

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INTRODUCTION

The electronic properties and magnetic behavior of FeNi alloys have been of special interest since 1897 when Guillaume [1] first reported an almost zero thermal expansion over a wide temperature range in face-centered cubic (fcc) crystals with a Ni concentration of around 35 atomic percent. This behavior was subsequently observed in various ordered and random binary alloy systems, and became known as the "Invar Effect" [2]. Despite much experimental [3] and theoretical [4] work, a full understanding of this important technological effect is lacking.

A general view, first advanced by Weiss [3], is that the Fe atoms first develop a large magnetic moment in the Ni-rich alloys, which expands their lattice as their number increases. At a critical Wigner-Seitz cell volume, the strain energy becomes too large and there is a phase transition from this "high-spin/high-volume" state into a 'low-spin/low-volume" state. In the bulk alloys, this instability begins around 60% Fe content, the Curie temperature falling precipitously, simultaneously with a 'martensitic' structural transformation to body-centerd cubic (bcc) symmetry [2]. Theoretical work predicts that the fcc phase can exist in two possible states: a ferromagnetic high volume state or an antiferromagnetic low volume state (2 γ state model) [3] with a volume change between the paramagnetic and the high spin state of ~7% [5], and 1% change between a non-collinear equilibrium state and the high spin state [4]. Experimental work shows a lattice expansion increasing linearly up to 3% at 65% Fe content followed by a sudden relaxation of 2% with increasing Fe content [6]. This work also shows that the martensitic structural transformation can be arrested in ultrathin alloy films epitaxially grown on a Cu(100) substrate. The nanometer-scale thickness effectively 'clamps' the crystal structure to that of the fcc substrate. Small changes in the Wigner-Seitz cell volume produce a small tetragonal distortion, which can be monitored by diffraction methods [6]. By growing ultrathin pseudomorphic fcc films, it is possible to focus on the effect of changing alloy composition on themagnetic and electronic behavior.

Here, we report changes in the magnitudes of both elemental magnetic moments with changing composition, measured with X-ray linear/circular dichroism as well as changes in the exchange splitting measured with spin- and angle-resolved photoemission.

RESULTS AND DISCUSSION

A plot of the change in the asymmetry amplitude, for both elements in the FeNi alloy measured with XMLDAD, being a measure of the expectation value of the atomic magnetic moment $\langle\mu\rangle$, is shown plotted as a function of composition, fig. 1 (left panel). We observe that both the Ni and Fe signals track a similar profile with changing composition. In the Ni-rich alloys, both signal increase linearly up to 65% that on the Fe showing the larger increase. Above 65% Fe content, both signals show a sharp decrease. The observed asymmetry amplitudes, suggest that a high-spin moment develops on the Fe with increasing Fe content that increases overall magnetization,

which then increases the polarization of the valence states surrounding the Ni atomic cores. The Ni thus develops a component that tracks the developing magnetization. Above 65% Fe-content, the high-spin moment on the Fe appears to collapse to a "low-spin value", causing the overall magnetization density to be lowered, which is sensed by the reduced polarization of the valence states on the Ni. A plot of the variation of a 'stoichiometric average moment' $xA(Fe_x)+(1-x)A(Ni_{1-x})$ is shown in fig. 1 (right panel).. The behavior is very similar to that reported for the variation of the saturated moment normalized to the volume of similar fcc films on Cu(100) & Cu(111) and measured with SQUID magnetometry [7]. The solid line is the behavior reported for FeNi alloys from neutron scattering measurements [2]. We note that the Ni-rich phase extrapolates to a value around μ=2.5-3.0μB, a value predicted theoretically for the "high-moment" metastable fcc phase [5]. The above 'mean magnetic moment' variation, normalizing the Ni asymmetry amplitude to be equivalent to the magnetic moment of metallic Ni is tracking closely the Slater-Pauling curve, the moment increasing linearly with increasing number of holes per atom in the valence electronic states. Above 65% Fe content, the average moment shows a sharp decline into a "low-spin" magnitude state, which could be the result of a collapse of the spin moment on the Fe atoms and/or a sudden decrease in magnetization due to a non collinear rearrangement of spins.

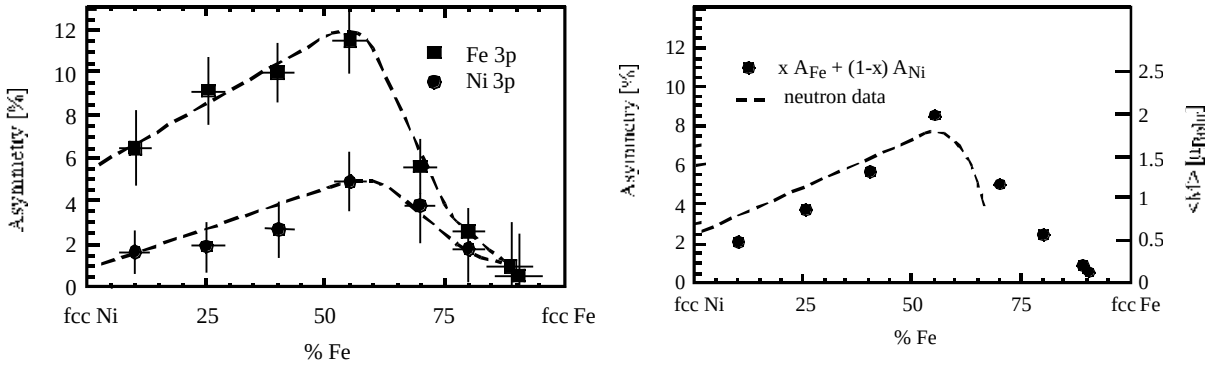

Figure 1. Left panel: Change in dichroism amplitude, A, as a function of FeNi alloy composition. Right panel: The variation of the 'stoichiometrically-weighted' dichroism signal amplitude (see text) with changing FeNi alloy composition. The dashed curve is the behavior observed in bulk FeNi alloys by neutron scattering. The right hand scale is determined from neutron and SQUID magnetometry data [2,7]

Spin polarized photoemission studies record a sudden decrease in the "mean-field" exchange splitting of the d-states with increasing Fe content through the critical "Invar transition". Angle-resolved photoemission imaging of states at the Fermi level [8] reveal a much smaller splitting of the sp-states, which also tracks the changing magnetization with changing composition. Spectral lineshapes reveal a decreased lifetime (i.e. decreased mean-free path for scattering) of the minority spin-polarized sp-states, in agreement with reported similar measurements on permalloy [9].Angle-resolved photoemission measurements of the sp-states, away from the regions of emerging minority d-states, along the <110> (Σ) symmetry direction, resolves the sp exchange splitting in reciprocal space. We observe that the spectral width of the minority-spin band of the sp-states is broader than that of the majority-spin sp-band.

This has been reported in similar measurements on permalloy, and is indicative of a shorter lifetime due to increased scattering and a shorter mean-free-path for the minority spin electrons. We also note that the lifetime broadening of the minority-spin sp-states increases significantly in the Fe rich alloys. The measured exchange splitting of the sp-states as well as the spin-resolved measured exchange splitting of the d-states track the behavior of the x-ray core-level photoemission dichroism.

This is to be expected on the basis of the overall magnetic energy being the sum of a 'local moment' energy on the 'atom(s) and a 'mean-field' exchange energy rising from the spin polarization of the itinerant valence states [10].
X-ray circular dichroism measurements allow to separate the orbital from the spin part of the local moments. Our measurements show the same concentration dependence of the local moments as t he linear dichroism measurements.

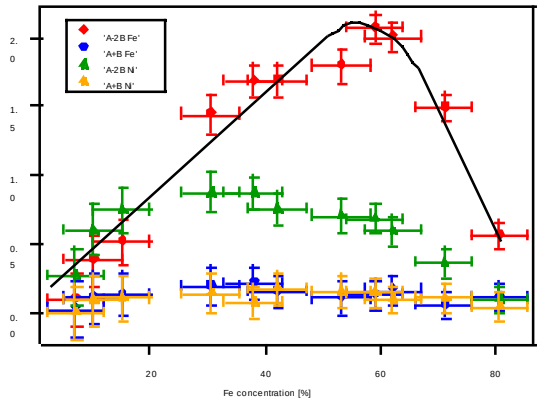

Figure 2. The variations of the spin, respectively orbital parts of the magnetic moments. [A-2B=-C/$\mu_B(\mu_s+\mu_D^{\alpha})$; A+B=-3C/2$\mu_B(\mu_O^{\alpha})$]. A, B are the areas underneath the difference peaks of the 2p spectra taken with magnetization up and down.

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