Hybridization and magnetism in small FePt alloy clusters

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Abstract. Magnetic properties of small, mass selected, Fe and FePt alloy clusters deposited on a remanently magnetized Ni/Cu(001) substrate have been investigated using x-ray magnetic circular dichroism. Strong size- and stoichiometry-dependent magnetic properties of Fe and FePt clusters are found, together with enhanced orbital moments compared to bulk and nanoparticles. Compared to pure Fe clusters, FePt alloy clusters have lower orbital and higher spin moments. In addition, they show a higher branching ratio of $B_r = L_3/(L_2 + L_3)$ due to stronger spin–orbital coupling and hybridization.

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1. Introduction

Nanoparticles are promising new materials for ultra-small magnetic storage devices [1]. With lower coordination numbers, the low-dimensional systems exhibit enhanced magnetic moments and this opens up new fields of applications for technological and biomedical sciences. To understand these fundamental interactions and their variation with the particle size, small, mass selected clusters are excellent systems as they are large enough to show complex phenomena and small enough to be treated with sophisticated theoretical methods. In recent work, magnetic properties as a function of the number of atoms per cluster have been studied for 3d and rare earth elements [2–6]. However, by changing also the composition of the clusters a much larger functional space is available. For example, already in current magnetic storage devices a larger magnetic anisotropy and, hence, a strongly increased bit density is realized by using a 3d–5d alloy system, such as CoPt or FePt. In 3d–5d alloys the large magnetic moments of the 3d electrons are combined with the rather large spin–orbit interaction $\xi$ of the 5d electrons. The spin–orbit interaction is about one order of magnitude higher for the 5d element Pt ($\xi_{\text{Pt}} = 0.6 \text{ eV}$), compared to Fe ($\xi_{\text{Fe}} = 0.08 \text{ eV}$) [7]. This can result in an increase of the orbital moment of the 3d electrons also inducing a larger magnetic anisotropy. This effect has been observed by Gambardella et al [8], where a huge orbital moment of $1.1\mu_B$ per atom together with a magnetic anisotropy of 9 meV per atom have been found for Co atoms deposited on a Pt(111) surface. Similar findings exist for clusters where a huge enhancement of 100% for the orbit to spin moment ratio has been found in the Co$_2$ cluster by adding a single Pt atom [9]. Tailoring the size and composition of very small, mass selected clusters will enhance the fundamental understanding of the influence of the alloying due to hybridization of the valence electrons on the magnetic properties.

In this paper we present experiments on small, mass selected alloy clusters with up to four atoms per cluster deposited on a magnetized surface taking Fe$_n$Pt$_m$ ($n, m \leq 2$) as a case study. We focus on the magnetic properties, including magnetic orbital ($m_\ell$), spin ($m_s$) moments and the orbital to spin ratio ($m_\ell/m_s$), by adding one or two Pt atoms to the Fe atoms and dimers deposited on remanently, out-of-plane magnetized thin Ni films on the Cu(100) surface [10, 11]. Exchange coupling to the substrate aligns the magnetic moments of the Fe and FePt clusters.

2. Experiments

To probe the magnetic properties of the clusters the x-ray magnetic circular dichroism (XMCD) method is applied using synchrotron radiation. Its ability to determine spin and orbital magnetic moments in an element-specific way makes it a favorable method to study the magnetic properties of deposited clusters in detail. The Fe and FePt alloy clusters are produced by high-energy ion sputtering using 30 keV Xe$^+$ ions to erode the cluster material and thereby produce the clusters from a high-purity Fe$_{50}$Pt$_{50}$ target. The production and mass separation of the clusters by a magnetic dipole field are carried out at a base pressure of $1 \times 10^{-8}$ mbar. Figure 1 shows the mass spectra of the clusters up to Pt$_3$, produced from the FePt target. A more detailed description of the cluster source and its capabilities are given in [12].

To prevent fragmentation and implantation into the substrate surface during deposition, the clusters are landed with a kinetic energy below 1 eV per atom and a soft landing scheme [13, 14] is applied using krypton as a buffer system. This scheme effectively suppresses fragmentation during cluster deposition [15, 16]. Krypton layers are desorbed by flash heating to $\approx 120$ K
Figure 1. Mass spectrum of Fe and FePt alloy clusters produced from a FePt (50%:50%) target sputtered by Xe$^+$ ions. As an example the four possible geometries for a FePt$_2$ cluster on an Ni/Cu(100) surface are shown. The arrangement of the clusters is chosen for convenience and would correspond to a much larger coverage (12%) than used in the experiment.

before the measurements and the randomly distributed clusters are thereby directly supported by the substrate surface. Low sample temperatures ($\approx$30–40 K) are used to inhibit cluster agglomeration and therefore, in combination with a low cluster coverage of about 0.03 monolayer, cluster–cluster interaction can be neglected.

The 20 monolayer thin Ni films have been prepared on the clean Cu(100) substrate and are remanently magnetized using short pulses from a small solenoid. The layer thickness has been calibrated and monitored by the intensity ratio of the Ni and Cu $L_{2,3}$ x-ray absorption spectra.

All measurements have been carried out at a base pressure below $2 \times 10^{-10}$ mbar. As a measure of the x-ray absorption, the total electron yield (TEY), in our experiment the total sample current, is recorded. The experiments have been carried out at beamline UE52-SGM [17] with a constant degree of 90% circular polarization at the BESSY II storage ring. The Fe and FePt alloy cluster samples have been prepared in situ in UHV at the synchrotron. XMCD spectra of the deposited clusters and the ultrathin film have been taken at the Fe $L$-edges and Ni $L$-edges in the photon energy ranges of 690–740 and 840–890 eV, respectively. A counting time of 3 s per data point has been used for each photon helicity. The beamline resolution for the Fe 2p–3d spectra is 200 meV [17], and so was the step width we set for the spectra. To avoid an influence of contaminations due to residual gas adsorption the total measurement time for each cluster preparation was kept below 120 min.
3. Results and discussion

In figure 2(a) the L-edge x-ray absorption spectra for all Fe and FePt clusters under study are depicted, after subtracting the background and a step function. The absence of other peaks in XAS besides the elemental $L_3$ and $L_2$ peaks at 708.2 and 721.5 eV indicates an oxide-free environment of the clusters. Within the uncertainty of the experiment no shift in the resonance positions or the 2p spin–orbit splitting is found. The spin–orbit coupling in the 3d states is not determined directly from the experimental absorption spectra, but is related to the branching ratio $B_r = A_{L3}/(A_{L3} + A_{L2})$ of the integrated intensity of the $L_3$ edge to the total absorption of the $L_3$- and $L_2$-edges [18, 19]. If we compare the branching ratio of all clusters shown in figure 2(b), the branching ratios are about 0.78 and 0.80 for Fe and FePt, 0.77 and 0.79 for Fe and FePt, respectively. As expected, the higher branching ratio shows a stronger spin–orbital coupling in the FePt alloy clusters, while all clusters have a higher branching ratio compared to bulk Fe of 0.75 [20]. The error bars have been estimated by averaging the data from different sample preparations and varying the background and step function.

Like the pure Fe clusters [2], all measured FePt alloy clusters show a ferromagnetic coupling to the magnetized Ni/Cu(001) substrate. By applying the XMCD sum rules [21, 22], it is possible to determine the effective spin magnetic moment $m_{\text{eff}}$ and the orbital magnetic moment $m_\ell$. $m_{\text{eff}} = m_s + 7m_\ell$ consists of the spin magnetic moment $m_s$ and the magnetic dipole moment $m_\ell$, which accounts for the asphericity of the spin moment distribution. In the following, we will refer to $m_{\text{eff}}$ as an effective spin moment denoted for simplicity as $m_s$. All absolute magnetic moments for $m_\ell$ and $m_\ell$ are given per d-hole in units of $\mu_B$ per hole.

In figure 3(a) the XMCD spectra of the Fe$_2$Pt$_n$ clusters are shown. For a general overview of the changes, the dichroism spectra are normalized to the same XMCD intensity at the $L_2$-edge.
Figure 3. Normalized (a) and integrated (c) $L$-edge XMCD spectra of Fe$_2$Pt$_n$ clusters deposited on a magnetized Ni/Cu(100) surface. In (b) the normalized $L_3$ XMCD signal is enlarged. The XMCD spectra have been normalized to the integrated intensity of the $L_2$-edge. For Fe$_2$ and Fe$_2$Pt the $L_3$ XMCD signal has the same height; however, the line shape of Fe$_2$Pt is narrower, resulting in the observed decreasing integrated $L_3$ XMCD intensity in (c) by adding Pt to Fe$_2$.

The XMCD spectra for the different Fe$_2$Pt$_n$ clusters show only small variations. In particular, the Fe$_2$ and Fe$_2$Pt spectra are at first glance almost identical. However, a closer look reveals a different line shape of the Fe$_2$Pt XMCD $L_3$-signal compared to the Fe$_2$ signal. As a consequence of the line narrowing the integrated intensity at the $L_3$-edge is significantly smaller.

This narrowing of the XMCD signal can be attributed to a change, e.g. a narrowing of the unoccupied density of states, by adding Pt atoms to the Fe atom and dimer. Here looking at the shape of the XMCD $L_3$ spectra in figure 3(b) shows that the Fe$_2$Pt XMCD signal is smaller in particular on the high-energy side of the $L_3$-edge, indicating a shift of the DOS to lower binding energies. Adding a further Pt atom enhances this effect. Such a narrowing depending on the Fe content has been theoretically predicted for disordered FePt bulk alloys using the SPR-KKR method [23].

In table 1 and figure 4(a) the ratios of orbital to spin moments derived from the integrated spectra for all FePt clusters are summarized. The lower orbital to spin ratio with increasing the Pt content in the alloy clusters may be due to either a lower orbital moment or higher spin moment. Hence, also the absolute spin and orbital magnetic moments per 3d hole have been evaluated and are given in table 1 and depicted in figure 4(b). Within the error bars Fe atoms and dimers
Table 1. Magnetic spin and orbital moments for FePt clusters on Ni/Cu(001). By adding a Pt atom to Fe$^1$ or Fe$^2$ the orbital moments decrease while the spin moments increase. The error bar was estimated to be roughly 15% for the spin and orbital moment, including inaccuracies of the sum-rule-based analyses, and the error due to the small overlap in energy of $L_3$ and $L_2$ absorption edges of Fe. For each sample, four spectra have been averaged to obtain these results. The nanoparticle data are taken from $[24, 25]$.

| Cluster        | $m_s$ ($\mu_B$ per hole) | $m_l$ ($\mu_B$ per hole) | $m_l/m_s$ |
|----------------|--------------------------|--------------------------|-----------|
| Fe$^1$         | 0.55 ± 0.05              | 0.09 ± 0.01              | 0.17 ± 0.02 |
| FePt           | 0.68 ± 0.10              | 0.09 ± 0.01              | 0.14 ± 0.02 |
| FePt$^2$       | 0.58 ± 0.06              | 0.07 ± 0.01              | 0.13 ± 0.01 |
| Fe$^2$         | 0.57 ± 0.05              | 0.10 ± 0.01              | 0.18 ± 0.01 |
| Fe$^2$Pt       | 0.65 ± 0.06              | 0.08 ± 0.01              | 0.13 ± 0.01 |
| Fe$^2$Pt$^2$   | 0.64 ± 0.06              | 0.07 ± 0.01              | 0.11 ± 0.01 |

Nanoparticles

|                  | $m_s$ ($\mu_B$ per hole) | $m_l$ ($\mu_B$ per hole) | $m_l/m_s$ |
|------------------|--------------------------|--------------------------|-----------|
| Fe [24]          | 0.59                     | 0.026                    | 0.044     |
| FePt [25]        | 0.70                     | 0.065                    | 0.093     |

Adatoms

|                  | $m_s$ ($\mu_B$ per hole) | $m_l$ ($\mu_B$ per hole) | $m_l/m_s$ |
|------------------|--------------------------|--------------------------|-----------|
| Fe atom 3d$^6$ $^5$D | 1                        | 0.5                      | 0.5       |
| Fe atom 3d$^7$ $^4$F | 1                        | 1                        | 1         |
| Fe on Al/Ni$_3$Al [31] |                          |                          | 0.53 ± 0.09 |
| Fe on alkaline [32]  |                          |                          | 0.95 ± 0.05 |

show a high orbital moment of the order of 0.10$\mu_B$ and a spin moment around 0.56$\mu_B$, resulting in a huge orbit to spin ratio of 0.17.$^2$

Focusing on the composition-dependent magnetic properties of the absolute moments, two trends are found in figure 4(b) by adding Pt atoms to the Fe clusters. On the one hand, an increase of the spin moment and, on the other hand, a reduction of the orbital moment are observed at the same time. Consequently, the orbital to spin ratio for Fe in the alloy clusters decreases with increasing Pt content. Compared with Fe$_2$, Fe$_2$Pt$_2$ has a 30% lower orbital moment and a roughly 10% higher spin moment and thus an almost 40% reduced orbital to spin ratio. The spin moments of the size selected clusters are of a comparable size with the corresponding Fe and FePt nanoparticles [24, 25]. Here the spin moment for the Fe atom and the dimer of $\cong 0.56 \pm 0.05\mu_B$ agrees fairly well with the value of $\mu_s = 0.59\mu_B$ found for Fe nanoparticles [24]. For Fe$_n$Pt$_m$ clusters the spin moment is in the range from 0.64 to 0.68$\mu_B$, which is slightly lower than the spin moment of 0.70$\mu_B$ for FePt nanoparticles found by Antoniak et al [25]. An exceptional case seems to be the FePt$_2$ cluster, which has a spin moment $\mu_s = 0.58\mu_B$ similar to the Fe atom and dimer.

$^2$ The absolute moments of deposited Fe$_n$ clusters presented in [2, 3] are too large by a factor of two due to wrong normalization.
Figure 4. Magnetic moments of Fe$_1$Pt$_{m}$ (●), Fe$_2$Pt$_{m}$ (■) clusters on Ni/Cu(100), nanoparticles (NP) and bulk or thin films. (a) The magnetic orbital to spin ($m_l/m_s$) ratio for the mass selected clusters. (b) Absolute magnetic spin and orbital moments per d-hole of Fe$_n$Pt$_m$ clusters. (c) Effective spin moment of mass selected clusters and nanoparticles (♦ [23–25]), bulk and of 50 nm thin films (▲ [23]) depending on the Fe content. For the FePt nanoparticles, the corrected (♦) and uncorrected (◇) data from [23] are given. For a better visibility, values with the same Fe content have been shifted slightly by ±1 at%. The dotted line is a guide to the eyes.

Whereas the spin moments are of comparable size, the orbital moments of the small Fe clusters are significantly larger than the nanoparticle values. For the Fe atom and dimer, the orbital moment is enhanced by a factor of ≈4 compared to Fe nanoparticles. The decrease of the orbital moment observed for the clusters with additional Pt and the opposite trend in the nanoparticles (here the orbital moment is increasing from 0.028$\mu_B$ to 0.065$\mu_B$ when Pt is added) lead to an almost vanishing difference between clusters and nanoparticles.

In figure 4(c) all spin moments of the clusters are given relative to their Fe content $x$ to compare them with Fe and FePt bulk, thin film and nanoparticle data [22, 23, 26]. Again, all magnetic moments are given per d-hole using the number of holes in the range of $n_d \approx 3.4$–3.5 given in [23–25]. For the Pt-rich, disordered bulk Fe$_x$Pt$_{1-x}$ ($x < 0.5$) large spin moments around $m_s \approx 0.85\mu_B$ are found, decreasing to 0.64$\mu_B$ with increasing the Fe content. For different FePt nanoparticles smaller magnetic spin moments have been found. However, from extended x-ray absorption fine structure measurements of the nanoparticles an Fe-rich environment of the local composition can be assumed. Correcting the Fe content accordingly, the magnetic spin moments of the nanoparticles agree with the disordered Fe$_x$Pt$_{1-x}$ bulk data [23].
Nevertheless, this inhomogeneous composition within the nanoparticles can hardly explain the rather low absolute magnetic moments in Fe\textsubscript{n}Pt\textsubscript{m} clusters, as the composition of the clusters is well defined. In a three-atom system on a (001) surface, there are, in principle, only four possible geometries, depicted in figure 1. If only the nearest-neighbor interaction is taken into account, two geometries are left and the magnetic moments of these different geometries should be at least similar.

As mentioned above, the idea of alloying 3d and 5d metals is to increase the magnetic moments and, in particular, the orbital moments of magnetic materials, by increasing the spin–orbit interaction due to hybridization of 3d and 5d electrons. For nanoparticles this behavior is observed; however, in the few-particle limit studied here, the situation is different. From the branching ratio \( B_r \) the spin–orbit interaction enhancement for the Fe 3d electrons is found for the alloy clusters. Nevertheless, this does not necessarily result in much larger magnetic moments of the FePt clusters. For example, for Fe\textsubscript{n}Pt\textsubscript{2} the largest branching ratio \( B_r \) is found (see figure 2), indicating a larger spin–orbit interaction of the Fe d electrons with increasing Pt content. Here, the spin moments are slightly enhanced but much less compared to nanoparticles or Fe\textsubscript{n}Pt\textsubscript{1−x} bulk. For the orbital moments even a quenching due to the 3d–5d hybridization is found. The maximum in the spin moment depending on the Pt content might be explained by the narrowing of the Fe 3d states. Adding a further Pt atom, however, will increase the hybridization further. For Fe\textsubscript{n}Pt\textsubscript{2} this finally results in lower spin moments. This model is also consistent with the data for the ratio of orbital and spin moments measured for Fe adatoms on different, weakly interacting surfaces (Al/Ni\textsubscript{3}Al \cite{31} and alkaline metals \cite{32}) given in table 1. In these systems the hybridization of the Fe 3d electrons with the substrate is very weak, resulting in large ratios, similar to the free atom values, if a different number of 3d electrons is taken into account, e.g. 3d\textsuperscript{6} and 3d\textsuperscript{7} on the Al/Ni\textsubscript{3}Al and alkaline surface, respectively. For all Fe\textsubscript{n}Pt\textsubscript{m} the ratio is always clearly below the atomic value, decreasing with increasing Pt amount, e.g. strong 3d–5d hybridization.

Reduced magnetic moments in FePt alloys have been discussed also in terms of non-collinear spin structures \cite{27}. At least for our cluster data such structures can be almost ruled out. The lowest spin moment is found for FePt\textsubscript{2} with only one Fe atom, which will have a collinear alignment to the magnetized Ni substrate. For Fe\textsubscript{2}Pt\textsubscript{2} a similar moment as for Fe\textsubscript{1}Pt\textsubscript{1} is found, which makes a non-collinear alignment also unlikely. Nevertheless, such a non-collinear spin structure might become important for larger clusters, as already for thin Fe films with fcc- and bcc-like structures a ferromagnetic as well as an antiferromagnetic ordering is found \cite{28}. Very complex spin structures have recently been observed in FePt surface alloys \cite{7}. There might still be a size-dependent influence of the magnetic dipole term \( m_r \) on the effective spin moment. However, these effects, in general, are regarded as rather small and should not influence the observed trend found for FePt clusters.

Reduced magnetic moments might also result from unsaturated Fe moments on the magnetized Ni/Cu(100) surface. This should have at least a similar effect on all clusters and would not be able to explain the observed quantum size effects on adding single atoms. Nevertheless, one has to take into account that the magnetized Ni/Cu(100) substrate will influence the magnetic properties of FePt clusters. This becomes obvious when the completely different size dependences of free \cite{29} and deposited \cite{2,3} Fe\textsubscript{n} \((n \geqslant 3)\) clusters are compared. Thus, further studies on different magnetic substrates or in external magnetic fields are urgently required.

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A further interesting aspect is a comparison of the magnetic properties of FePt alloy clusters with that of CoPt [9], since they are both composed of 3d and 5d elements and have comparable properties in bulk materials. Although both alloy clusters have higher spin–orbit coupling, the Pt atom has a totally opposite effect changing the magnetic properties. Whereas an enhancement of 100% for the orbital to spin ratio has been found in the Co$_2$ cluster on adding a single Pt atom, a reduction of 30% is observed in the case of the Fe$_2$ cluster.

Here it would be highly interesting to study also the induced magnetic moments of the Pt atoms within the cluster. Unfortunately, within the present study these magnetic moments are not detectable. Nevertheless, at least a ferromagnetic alignment of the Pt moments can be expected, which has been observed e.g. in FePt nanoparticles [23]. Regarding the magnitude of the magnetic moments in Ni/Pt layer systems, induced magnetic moments in the Pt layers around $\approx 0.2\mu_B$ per atom have been found [30].

4. Summary

In conclusion, in this paper the influence of alloying on the magnetic properties of 3d metal atoms with 5d atoms has been studied in the few-particle limit taking FePt as a case study. The spin moments of Fe$_n$Pt$_m$ ($n, m \leq 2$) clusters are of comparable size to those of Fe$_x$Pt$_{1-x}$ nanoparticles; however, the orbital moments are larger by a factor of two. With increasing the Pt content the orbital moments are decreasing, whereas the spin moment shows a maximum for Fe-rich and Fe$_n$Pt$_m$ and the Pt-rich cluster FePt$_2$ shows a lower spin moment. This behavior is interpreted in terms of the hybridization of Fe 3d and Pt 5d electrons, which has been estimated from the narrowing of the $L_3$ XMCD lineshape as well as the $L_3/L_2$ branching ratio.

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