Influence of the magnetic dipole T_z term on the XMCD spectra of clusters When T_z really messes things up

O. Šipr¹ J. Minár² J. Vackář¹ H. Ebert²

¹Institute of Physics, Academy of Sciences of the Czech Republic, Prague http://www.fzu.cz/~sipr

²Universität München, Department Chemie und Biochemie, München

30. July 2009 / XAFS-14 Camerino



Introduction: Clusters and magnetism

XMCD spin sum rule

Materials and computational method

Results: $\mu_{\rm spin}$ and $[\mu_{\rm spin} + 7T_z]/n_h$ for various systems

Conclusions: What we have to be prepared for



Introduction: Clusters and magnetism

XMCD spin sum rule

Materials and computational method

Results: $\mu_{\rm spin}$ and $[\mu_{\rm spin} + 7T_z]/n_h$ for various systems

Conclusions: What we have to be prepared for



Introduction: Clusters and magnetism

XMCD spin sum rule

Materials and computational method

Results: $\mu_{
m spin}$ and $[\mu_{
m spin}+7\,{\cal T}_z]/n_h$ for various systems

Conclusions: What we have to be prepared for



Introduction: Clusters and magnetism

XMCD spin sum rule

Materials and computational method

Results: $\mu_{\rm spin}$ and $[\mu_{\rm spin} + 7T_z]/n_h$ for various systems

Conclusions: What we have to be prepared for



Introduction: Clusters and magnetism

XMCD spin sum rule

Materials and computational method

Results: $\mu_{\rm spin}$ and $[\mu_{\rm spin} + 7T_z]/n_h$ for various systems

Conclusions: What we have to be prepared for



Clusters and magnetism: Why people like them ?

- ► Clusters have a high portion of surface atoms ⇒ average coordination number is smaller than in bulk
- Rule of thumb: The lower the coordination number, the larger the magnetic moment per atom
- Thin films and clusters have large magnetocrystalline anisotropy per atom (that's what you want for magnetic recording)



Clusters and magnetism: Why people like them ?

- ► Clusters have a high portion of surface atoms ⇒ average coordination number is smaller than in bulk
- Rule of thumb: The lower the coordination number, the larger the magnetic moment per atom
- Thin films and clusters have large magnetocrystalline anisotropy per atom (that's what you want for magnetic recording)



Clusters and magnetism: Why people like them ?

- ► Clusters have a high portion of surface atoms ⇒ average coordination number is smaller than in bulk
- Rule of thumb: The lower the coordination number, the larger the magnetic moment per atom
- Thin films and clusters have large magnetocrystalline anisotropy per atom (that's what you want for magnetic recording)



- Magnetic properties of *large assemblies* of clusters are macroscopic: no principal problems with measuring them
- To understand nanomagnetism, one needs to study individual small systems
- Magnetization of individual clusters (of just few atoms) cannot be measured by macroscopic methods
- Recent progress in studying magnetism of clusters is, to a large extent, associated with analyzing XMCD



(日) (四) (日) (日) (日) (日)

- Magnetic properties of *large assemblies* of clusters are macroscopic: no principal problems with measuring them
- To understand nanomagnetism, one needs to study individual small systems
- Magnetization of individual clusters (of just few atoms) cannot be measured by macroscopic methods
- Recent progress in studying magnetism of clusters is, to a large extent, associated with analyzing XMCD



- Magnetic properties of *large assemblies* of clusters are macroscopic: no principal problems with measuring them
- To understand nanomagnetism, one needs to study individual small systems
- Magnetization of individual clusters (of just few atoms) cannot be measured by macroscopic methods
- Recent progress in studying magnetism of clusters is, to a large extent, associated with analyzing XMCD



- Magnetic properties of *large assemblies* of clusters are macroscopic: no principal problems with measuring them
- To understand nanomagnetism, one needs to study individual small systems
- Magnetization of individual clusters (of just few atoms) cannot be measured by macroscopic methods
- Recent progress in studying magnetism of clusters is, to a large extent, associated with analyzing XMCD



XMCD appeals

XMCD (X-ray Magnetic Circular Dichroism):

sample is magnetized, measure the difference between absorption of left and right circularly polarized x-rays



- Chemically selective one can probe "local" magnetism connected with particular chemical type of atoms
- \blacktriangleright XMCD sum rules gives access to $\mu_{\rm spin}$ and $\mu_{\rm orb}$ separately



A D F A B F A B F A B F

Two ways of moving an electron

(A quick and dirty introduction to magnetism)

► Spinning:





(日)



э

Two ways of moving an electron

(A quick and dirty introduction to magnetism)

Spinning:

```
\mu_{\rm spin} usually large, {\sim}2.2~\mu_B for Fe
```



Orbiting:

 $\mu_{
m orb}$ usually small, \sim 0.1 μ_B for Fe important for connection with MAE





э

$L_{2,3}$ edge of magnetic TM systems





$L_{2,3}$ edge of magnetic TM systems



< ロ > < 同 > < 回 > < 回 >

э

Problems with sum rules

Approximations involved in its derivation:

- Transition to a well-defined single band
- Core hole neglected

. . .

- Could be at least partly corrected for by renormalization, scaling and/or varying the L_{2.3} edge integration ranges
 - Justified when studying series of related systems:
 One can assume that the corrections will be similar for each member of the series



Problems with sum rules

Approximations involved in its derivation:

- Transition to a well-defined single band
- Core hole neglected

...

- Could be at least partly corrected for by renormalization, scaling and/or varying the L_{2,3} edge integration ranges
 - Justified when studying series of related systems:
 One can assume that the corrections will be similar for each member of the series



 T_z term: A feature, not a bug !

$$\frac{3}{I_A} \int \left(\Delta \mu_{L_3} - 2 \Delta \mu_{L_2} \right) \, \mathrm{d}E = \frac{\mu_{\mathrm{spin}} + 7 T_z}{n_h}$$

- I_A integrated isotropic absorption spectrum μ_{spin} d component of the local spin magnetic moment n_h number of holes in the d band
- T_z d component of magnetic dipole operator

$$T_z = \left\langle \hat{T}_z \right\rangle = \left\langle \frac{1}{2} \left[\sigma - 3\hat{\mathbf{r}} (\hat{\mathbf{r}} \cdot \sigma) \right]_z \right\rangle$$

T_z is a measure of the *asphericity of spin magnetization* [Stöhr & König PRL **75** 3748; Oguchi & Shishidou PRB **70** 024412]

 $\mu_{
m spin}$ can be obtained only in combination with $7T_z$!



 T_z term: A feature, not a bug !

$$\frac{3}{I_A} \int \left(\Delta \mu_{L_3} - 2 \Delta \mu_{L_2} \right) \, \mathrm{d}E = \frac{\mu_{\mathrm{spin}} + 7 T_z}{n_h}$$

- I_A integrated isotropic absorption spectrum μ_{spin} d component of the local spin magnetic moment n_h number of holes in the d band T d component of mean spectrum
- T_z d component of magnetic dipole operator

$$T_z = \left\langle \hat{T}_z \right\rangle = \left\langle \frac{1}{2} \left[\sigma - 3 \hat{\mathbf{r}} (\hat{\mathbf{r}} \cdot \sigma) \right]_z \right\rangle$$

T_z is a measure of the *asphericity of spin magnetization* [Stöhr & König PRL **75** 3748; Oguchi & Shishidou PRB **70** 024412]

 $\mu_{\rm spin}$ can be obtained only in combination with $7T_z$!



 T_z term: A feature, not a bug !

$$\frac{3}{I_A} \int \left(\Delta \mu_{L_3} - 2 \Delta \mu_{L_2} \right) \, \mathrm{d}E = \frac{\mu_{\mathrm{spin}} + 7 T_z}{n_h}$$

 I_A integrated isotropic absorption spectrum μ_{spin} d component of the local spin magnetic moment n_h number of holes in the d band T d component of momentia dipole encepter

$$T_{z} = \left\langle \hat{T}_{z} \right\rangle = \left\langle \frac{1}{2} \left[\sigma - 3 \hat{\mathbf{r}} (\hat{\mathbf{r}} \cdot \sigma) \right]_{z} \right\rangle$$

T_z is a measure of the *asphericity of spin magnetization* [Stöhr & König PRL **75** 3748; Oguchi & Shishidou PRB **70** 024412]

 $\mu_{\rm spin}$ can be obtained only in combination with $7T_z$!



For bulk systems it is usually negligible

- ► For surfaces, monolayers or wires, absolute value of 7T_z is about 20 % of µ_{spin} [Wu & Freeman PRL 73 1994 (1994); Komelj et al. PRB 66 140407(R) (2002)]
- For investigating trends of μ_{spin} within a series of systems, what matters is how T_z varies from one system to another
 - If variations in T_z are small, neglect of T_z causes just an overall shift of the deduced μ_{spin}
 - Could T_z vary in such a way that the overall trends of μ_{spin}+7T_z and μ_{spin} would be quite different ?
- Common experience: Variations in n_h do not really matter, their effect is more-or-less negligible



- For bulk systems it is usually negligible
- ► For surfaces, monolayers or wires, absolute value of 7T_z is about 20 % of µ_{spin} [Wu & Freeman PRL 73 1994 (1994); Komelj et al. PRB 66 140407(R) (2002)]
- For investigating trends of μ_{spin} within a series of systems, what matters is how T_z varies from one system to another
 - If variations in T_z are small, neglect of T_z causes just an overall shift of the deduced μ_{spin}
 - Could T_z vary in such a way that the overall trends of μ_{spin}+7T_z and μ_{spin} would be quite different ?
- Common experience: Variations in n_h do not really matter, their effect is more-or-less negligible



- ► For bulk systems it is usually negligible
- ► For surfaces, monolayers or wires, absolute value of 7T_z is about 20 % of µ_{spin} [Wu & Freeman PRL 73 1994 (1994); Komelj et al. PRB 66 140407(R) (2002)]
- ► For investigating trends of µ_{spin} within a series of systems, what matters is how T_z varies from one system to another
 - If variations in T_z are small, neglect of T_z causes just an overall shift of the deduced μ_{spin}
 - ▶ Could T_z vary in such a way that the overall trends of µ_{spin}+7T_z and µ_{spin} would be quite different ?
- Common experience: Variations in n_h do not really matter, their effect is more-or-less negligible



- For bulk systems it is usually negligible
- ► For surfaces, monolayers or wires, absolute value of 7T_z is about 20 % of µ_{spin} [Wu & Freeman PRL 73 1994 (1994); Komelj et al. PRB 66 140407(R) (2002)]
- ► For investigating trends of µ_{spin} within a series of systems, what matters is how T_z varies from one system to another
 - ► If variations in T_z are small, neglect of T_z causes just an overall shift of the deduced µ_{spin}
 - ▶ Could T_z vary in such a way that the overall trends of µ_{spin}+7T_z and µ_{spin} would be quite different ?
- Common experience: Variations in n_h do not really matter, their effect is more-or-less negligible



- ► For bulk systems it is usually negligible
- ► For surfaces, monolayers or wires, absolute value of 7T_z is about 20 % of µ_{spin} [Wu & Freeman PRL 73 1994 (1994); Komelj et al. PRB 66 140407(R) (2002)]
- ► For investigating trends of µ_{spin} within a series of systems, what matters is how T_z varies from one system to another
 - ▶ If variations in T_z are small, neglect of T_z causes just an overall shift of the deduced µ_{spin}
 - ► Could T_z vary in such a way that the overall trends of µ_{spin}+7T_z and µ_{spin} would be quite different ?

 Common experience: Variations in n_h do not really matter, their effect is more-or-less negligible



- ► For bulk systems it is usually negligible
- ► For surfaces, monolayers or wires, absolute value of 7T_z is about 20 % of µ_{spin} [Wu & Freeman PRL 73 1994 (1994); Komelj et al. PRB 66 140407(R) (2002)]
- ► For investigating trends of µ_{spin} within a series of systems, what matters is how T_z varies from one system to another
 - ▶ If variations in T_z are small, neglect of T_z causes just an overall shift of the deduced µ_{spin}
 - ► Could T_z vary in such a way that the overall trends of µ_{spin}+7T_z and µ_{spin} would be quite different ?
- Common experience: Variations in n_h do not really matter, their effect is more-or-less negligible



Our mission

- Take a series of supported clusters of different sizes
- \blacktriangleright For each cluster size, evaluate average of d components of $\mu_{\rm spin}$

$$\frac{1}{N} \sum_{j=1}^{N} \mu_{\rm spin}^{(j)}$$

and of $[\mu_{\rm spin} + 7T_z]/n_h$

$$\frac{1}{N} \sum_{j=1}^{N} \frac{\mu_{\rm spin}^{(j)} + 7 T_z^{(j)}}{n_h^{(j)}}$$

▶ Compare how μ_{spin} and $[\mu_{spin} + 7T_z]/n_h$ depend on the cluster size



Our mission

- Take a series of supported clusters of different sizes
- \blacktriangleright For each cluster size, evaluate average of d components of $\mu_{\rm spin}$

$$\frac{1}{N} \sum_{j=1}^{N} \mu_{\rm spin}^{(j)}$$

and of $[\mu_{\rm spin} + 7T_z]/n_h$

$$\frac{1}{N} \sum_{j=1}^{N} \frac{\mu_{\rm spin}^{(j)} + 7T_z^{(j)}}{n_h^{(j)}}$$

▶ Compare how μ_{spin} and $[\mu_{spin} + 7T_z]/n_h$ depend on the cluster size



Our mission

- Take a series of supported clusters of different sizes
- \blacktriangleright For each cluster size, evaluate average of d components of $\mu_{\rm spin}$

$$\frac{1}{N} \sum_{j=1}^{N} \mu_{\rm spin}^{(j)}$$

and of $[\mu_{\rm spin} + 7T_z]/n_h$

$$\frac{1}{N} \sum_{j=1}^{N} \frac{\mu_{\rm spin}^{(j)} + 7 T_z^{(j)}}{n_h^{(j)}}$$

► Compare how μ_{spin} and $[\mu_{spin} + 7T_z]/n_h$ depend on the cluster size



Systems (1)







Distances between cluster atoms and the substrate taken so that structural relaxation is partially accounted for:

substrate	bulk interlayer	surface-to-cluster
	distance	distance
Ni(001) Au(111)	1.76 Å 2.35 Å	1.85 Å 1.78 Å

(Obtained from comparing atomic volumes and from other people's calculations [Martínez *et al.* PRB **71** 165425 (2002)])

Magnetization **M** perpendicular to the surface

Complete monolayers $N=\infty$ included as well (as end-points of the series...)





Distances between cluster atoms and the substrate taken so that structural relaxation is partially accounted for:

substrate	bulk interlayer	surface-to-cluster
	distance	distance
Ni(001) Au(111)	1.76 Å 2.35 Å	1.85 Å 1.78 Å

(Obtained from comparing atomic volumes and from other people's calculations [Martínez *et al.* PRB **71** 165425 (2002)])

Magnetization **M** perpendicular to the surface

Complete monolayers $N=\infty$ included as well (as end-points of the series...)





Distances between cluster atoms and the substrate taken so that structural relaxation is partially accounted for:

substrate	bulk interlayer	surface-to-cluster
	distance	distance
Ni(001)	1.76 Å	1.85 Å
Au(111)	2.35 A	1.78 A

(Obtained from comparing atomic volumes and from other people's calculations [Martínez *et al.* PRB **71** 165425 (2002)])

Magnetization ${\bf M}$ perpendicular to the surface

Complete monolayers $N=\infty$ included as well (as end-points of the series...)


- Ab-initio calculations, fully relativistic, LDA, SPR-KKR code http://olymp.phys.chemie.uni-muenchen.de/ak/ebert/SPRKKR
- Core hole neglected
- Calculate electronic structure of the "host" system (clean surface)
 - ▶ Tight-binding or screened KKR [Zeller et al. PRB 52 8807]
- Supported clusters are treated as a perturbation to the clean surface [Minár et al. Appl. Physics A 82 139 (2006)]
 - Green's function of the new system (cluster plus substrate) is obtained by solving the Dyson equation
- \blacktriangleright Atomic sphere approximation (ASA), $\ell_{max}{=}2$



- Ab-initio calculations, fully relativistic, LDA, SPR-KKR code http://olymp.phys.chemie.uni-muenchen.de/ak/ebert/SPRKKR
- Core hole neglected
- Calculate electronic structure of the "host" system (clean surface)
 - Tight-binding or screened KKR [Zeller et al. PRB 52 8807]
- Supported clusters are treated as a perturbation to the clean surface [Minár et al. Appl. Physics A 82 139 (2006)]
 - Green's function of the new system (cluster plus substrate) is obtained by solving the Dyson equation
- \blacktriangleright Atomic sphere approximation (ASA), $\ell_{max}{=}2$



- Ab-initio calculations, fully relativistic, LDA, SPR-KKR code http://olymp.phys.chemie.uni-muenchen.de/ak/ebert/SPRKKR
- Core hole neglected
- Calculate electronic structure of the "host" system (clean surface)
 - ► Tight-binding or screened KKR [Zeller et al. PRB 52 8807]
- Supported clusters are treated as a perturbation to the clean surface [Minár et al. Appl. Physics A 82 139 (2006)]
 - Green's function of the new system (cluster plus substrate) is obtained by solving the Dyson equation
- \blacktriangleright Atomic sphere approximation (ASA), $\ell_{max}{=}2$



- Ab-initio calculations, fully relativistic, LDA, SPR-KKR code http://olymp.phys.chemie.uni-muenchen.de/ak/ebert/SPRKKR
- Core hole neglected
- Calculate electronic structure of the "host" system (clean surface)
 - ► Tight-binding or screened KKR [Zeller et al. PRB 52 8807]
- Supported clusters are treated as a perturbation to the clean surface [Minár et al. Appl. Physics A 82 139 (2006)]
 - Green's function of the new system (cluster plus substrate) is obtained by solving the Dyson equation

 \blacktriangleright Atomic sphere approximation (ASA), $\ell_{max}{=}2$

- Ab-initio calculations, fully relativistic, LDA, SPR-KKR code http://olymp.phys.chemie.uni-muenchen.de/ak/ebert/SPRKKR
- Core hole neglected
- Calculate electronic structure of the "host" system (clean surface)
 - ► Tight-binding or screened KKR [Zeller et al. PRB 52 8807]
- Supported clusters are treated as a perturbation to the clean surface [Minár et al. Appl. Physics A 82 139 (2006)]
 - Green's function of the new system (cluster plus substrate) is obtained by solving the Dyson equation

• Atomic sphere approximation (ASA), $\ell_{\rm max}{=}2$

Results: $\mu_{\rm spin}$ and $[\mu_{\rm spin} + 7T_z]/n_h$



 $Fe_N/Ni(001)$



æ

ヘロト ヘ部ト ヘヨト ヘヨト

Results: $\mu_{\rm spin}$ and $[\mu_{\rm spin} + 7T_z]/n_h$





・ロト ・日下・ ・日下

Results: $\mu_{\rm spin}$ and $[\mu_{\rm spin} + 7T_z]/n_h$





Results: $\mu_{\rm spin}$ and $[\mu_{\rm spin} + 7T_z]/n_h$





<ロト <回 > < 回 > < 回 > < 回 > <

Results: $\mu_{\rm spin}$ and $[\mu_{\rm spin} + 7T_z]/n_h$



 FΖŰ

<ロト <部ト < 注入 < 注入 = 3</p>

$Co_N/Au(111)$: T_z changes the picture completely !



For Co_N on Au(111), trends of µ_{spin} and of [µ_{spin} + 7T_z]/n_h are exactly opposite.

Ignoring variations in T_z would lead to a false estimate of how µ_{spin} per atom depends on the cluster size

< ロ > < 同 > < 回 > < 回 >



 $Co_N/Au(111)$: T_z changes the picture completely !



For Co_N on Au(111), trends of µ_{spin} and of [µ_{spin} + 7T_z]/n_h are exactly opposite.

Ignoring variations in T_z would lead to a false estimate of how µ_{spin} per atom depends on the cluster size

< ロ > < 同 > < 回 > < 回 >



 $Co_N/Au(111)$: T_z changes the picture completely !



For Co_N on Au(111), trends of µ_{spin} and of [µ_{spin} + 7T_z]/n_h are exactly opposite.

• Ignoring variations in T_z would lead to a false estimate of how μ_{spin} per atom depends on the cluster size

< ロ > < 同 > < 回 > < 回 > < 回 >





 µ_{spin} can be obtained as an integral of spin polarization n_↑(E) - n_↓(E) = dµ_{spin}(E)/dE

$$\mu_{\rm spin}(E_F) = \int_{-\infty}^{E_F} \frac{\mathrm{d}\mu_{\rm spin}(E)}{\mathrm{d}E} \,\mathrm{d}E$$

<ロ> (日) (日) (日) (日) (日)



æ



$$n_{\uparrow}(E) - n_{\downarrow}(E) = \frac{\mathrm{d}\mu_{\mathrm{spin}}(E)}{\mathrm{d}E}$$

$$\mu_{\rm spin}(E_F) = \int_{-\infty}^{E_F} \frac{\mathrm{d}\mu_{\rm spin}(E)}{\mathrm{d}E} \,\mathrm{d}E$$





$$n_{\uparrow}(E) - n_{\downarrow}(E) = \frac{\mathrm{d}\mu_{\mathrm{spin}}(E)}{\mathrm{d}E}$$

$$\mu_{\rm spin}(E_F) = \int_{-\infty}^{E_F} \frac{\mathrm{d}\mu_{\rm spin}(E)}{\mathrm{d}E} \,\mathrm{d}E$$





$$n_{\uparrow}(E) - n_{\downarrow}(E) = \frac{\mathrm{d}\mu_{\mathrm{spin}}(E)}{\mathrm{d}E}$$

$$\mu_{\rm spin}(E_F) = \int_{-\infty}^{E_F} \frac{\mathrm{d}\mu_{\rm spin}(E)}{\mathrm{d}E} \,\mathrm{d}E$$





$$n_{\uparrow}(E) - n_{\downarrow}(E) = \frac{\mathrm{d}\mu_{\mathrm{spin}}(E)}{\mathrm{d}E}$$

$$\mu_{\rm spin}(E_F) = \int_{-\infty}^{E_F} \frac{\mathrm{d}\mu_{\rm spin}(E)}{\mathrm{d}E} \,\mathrm{d}E$$





• μ_{spin} can be obtained as an integral of spin polarization

$$n_{\uparrow}(E) - n_{\downarrow}(E) = \frac{\mathrm{d}\mu_{\mathrm{spin}}(E)}{\mathrm{d}E}$$

$$\mu_{\rm spin}(E_F) = \int_{-\infty}^{E_F} \frac{\mathrm{d}\mu_{\rm spin}(E)}{\mathrm{d}E} \,\mathrm{d}E$$





 µ_{spin} can be obtained as an integral of spin polarization
 du + (5)

$$n_{\uparrow}(E) - n_{\downarrow}(E) = \frac{\mathrm{d}\mu_{\mathrm{spin}}(E)}{\mathrm{d}E}$$

$$\mu_{\rm spin}(E_F) = \int_{-\infty}^{E_F} \frac{\mathrm{d}\mu_{\rm spin}(E)}{\mathrm{d}E} \,\mathrm{d}E$$





$$n_{\uparrow}(E) - n_{\downarrow}(E) = \frac{\mathrm{d}\mu_{\mathrm{spin}}(E)}{\mathrm{d}E}$$

$$\mu_{\rm spin}(E_F) = \int_{-\infty}^{E_F} \frac{\mathrm{d}\mu_{\rm spin}(E)}{\mathrm{d}E} \,\mathrm{d}E$$





▶ µ_{spin} can be obtained as an integral of spin polarization
(¬)

$$n_{\uparrow}(E) - n_{\downarrow}(E) = \frac{\mathrm{d}\mu_{\mathrm{spin}}(E)}{\mathrm{d}E}$$

$$\mu_{\rm spin}(E_F) = \int_{-\infty}^{E_F} \frac{\mathrm{d}\mu_{\rm spin}(E)}{\mathrm{d}E} \,\mathrm{d}E$$

T_z can be obtained as an integral of magnetic dipole polarization

$$T_z(E_F) = \int_{-\infty}^{E_F} \frac{\mathrm{d}T_z(E)}{\mathrm{d}E} \,\mathrm{d}E$$













T_z is very sensitive to position of *E_F*

<ロ> (日) (日) (日) (日) (日)



æ



- T_z is very sensitive to position of E_F
- Different slopes for ad-atoms and for monolayer



æ



- T_z is very sensitive to position of E_F
- Different slopes for ad-atoms and for monolayer
- Whether T_z will be larger for an ad-atom or for a monolayer depends both on cluster and on substrate

Sum rules can be checked



- Calculate the spectra theoretically
- Aply XMCD sum rules
- Compare \(\mu_{spin}\) and \(\mu_{orb}\) derived from XMCD spectra with moments calculated directly
- Upper integration boundary E_{cut} chosen so that there are exactly 10 electron states up to E_{cut} in the d band

(日)



Sum rules can be checked



- Calculate the spectra theoretically
- Aply XMCD sum rules
- ► Compare µ_{spin} and µ_{orb} derived from XMCD spectra with moments calculated directly
- Upper integration boundary E_{cut} chosen so that there are exactly 10 electron states up to E_{cut} in the d band



Sum rules can be checked



- Calculate the spectra theoretically
- Aply XMCD sum rules
- ► Compare µ_{spin} and µ_{orb} derived from XMCD spectra with moments calculated directly
- Upper integration boundary E_{cut} chosen so that there are exactly 10 electron states up to E_{cut} in the d band



Verifying spin sum rule: $[\mu_{spin} + 7T_z]/n_h$ via two ways



Fe_N/Ni(001) Co_N/Au(111)

A (1) × A (1)

- ▶ Good agreement between evaluating $[\mu_{spin} + 7T_z]/n_h$ from electronic structure and from XAS
- Spin sum rule itself is valid (at least within the initial state approximation)



Can the results be trusted?

- Dealing not with isolated systems but with whole series of clusters, spanning from single ad-atoms to complete monolayers
- All the systems treated in the same manner
- Conclusions are not crucially sensitive to the accuracy of the calculations
- ► The fact that μ_{spin} and $[\mu_{spin} + 7T_z]/n_h$ may have opposite trends has thus been established quite reliably



Can the results be trusted?

- Dealing not with isolated systems but with whole series of clusters, spanning from single ad-atoms to complete monolayers
- All the systems treated in the same manner
- Conclusions are not crucially sensitive to the accuracy of the calculations
- ▶ The fact that μ_{spin} and $[\mu_{spin} + 7T_z]/n_h$ may have opposite trends has thus been established quite reliably



Can the results be trusted?

- Dealing not with isolated systems but with whole series of clusters, spanning from single ad-atoms to complete monolayers
- All the systems treated in the same manner
- Conclusions are not crucially sensitive to the accuracy of the calculations
- ► The fact that μ_{spin} and $[\mu_{spin} + 7T_z]/n_h$ may have opposite trends has thus been established quite reliably



Conclusions

- Knowing how T_z varies with cluster size is essential for applying XMCD spin sum rule in clusters
- ▶ Magnetic dipole term T_z is not just a minor factor that affects the analysis
- ▶ Whether the size-dependence of μ_{spin} and of $[\mu_{spin} + 7T_z]/n_h$ will be opposite or not depends both on the clusters and on the substrate
- At least when analyzing XMCD spectra of Co on noble metal surfaces (Pt, Au), the influence of T_z definitely should be taken into account



Conclusions

- Knowing how T_z varies with cluster size is essential for applying XMCD spin sum rule in clusters
- ► Magnetic dipole term T_z is not just a minor factor that affects the analysis
- ▶ Whether the size-dependence of μ_{spin} and of $[\mu_{spin} + 7T_z]/n_h$ will be opposite or not depends both on the clusters and on the substrate
- ► At least when analyzing XMCD spectra of Co on noble metal surfaces (Pt, Au), the influence of T_z definitely should be taken into account


Conclusions

- Knowing how T_z varies with cluster size is essential for applying XMCD spin sum rule in clusters
- ► Magnetic dipole term T_z is not just a minor factor that affects the analysis
- ▶ Whether the size-dependence of μ_{spin} and of $[\mu_{spin} + 7T_z]/n_h$ will be opposite or not depends both on the clusters and on the substrate
- At least when analyzing XMCD spectra of Co on noble metal surfaces (Pt, Au), the influence of T_z definitely should be taken into account



Conclusions

- Knowing how T_z varies with cluster size is essential for applying XMCD spin sum rule in clusters
- ► Magnetic dipole term T_z is not just a minor factor that affects the analysis
- Whether the size-dependence of μ_{spin} and of $[\mu_{spin} + 7T_z]/n_h$ will be opposite or not depends both on the clusters and on the substrate
- ► At least when analyzing XMCD spectra of Co on noble metal surfaces (Pt, Au), the influence of T_z definitely should be taken into account



Take-home message

Magnetic dipole term T_z is a bad guy...

