Imaging magnetic atoms by Photoelectron Diffraction

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Abstract. We study possible methods how to obtain imaging of magnetic atoms by use of photoelectron diffraction (PD). We have proposed a method to apply Daimon effect where forward focusing PD peaks are rotated; we use the difference of the PD intensities for different circular polarization but $-\phi$ is used for the $-\circ$ circular polarization only in the data handling. This technique allows us to obtain atomic image only of spin polarized atoms. In this work we further apply this technique to ferromagnetic and antiferromagnetic surface layers with in-plane magnetization adsorbed on a nonmagnetic layer.

1. Introduction

Bansmann et al. have observed circular dichroism in angular distribution (CDAD) of photoelectrons excited from spherically symmetric initial states of nonmagnetic species such as CO/Pd(111) [1]. Daimon et al. have observed strong CDAD excited from Si 2p core level on Si(001) surface, although this system is not chiral [2]. They proposed an interesting formula for the azimuthal shift $\Delta \phi$ from the forward focusing peak. In order to study these effects, some theoretical approaches have successfully been proposed [3–6] based on short-range-order multiple scattering theory. By using CDAD effects, Daimon has proposed a new technique to observe stereo photographs of surface atomic arrangements [7–9].

In order to extract only information on magnetic atoms, we apply the cancellation of rotated forward focusing peaks because of the Daimon effect for nonmagnetic atoms. Conventional CDAD is defined by the difference $\Delta I(\theta, \phi) = I^+(\theta, \phi) - I^-\,(\theta, \phi)$ for different circular polarization $\pm$. This CDAD is influenced by surrounding magnetic and nonmagnetic atoms. We thus rather use the modified CDAD $\Delta I(\theta, \phi) = I^+(\theta, \phi) - I^-\,(\theta, -\phi)$. This technique allows us to obtain atomic image only of spin polarized atoms, and to distinguish magnetic atoms with up spins from those with down spins; magnetization is assumed to be parallel to the X-ray incidence [10]. For this purpose the scattering atoms should be arranged with mirror symmetry in regard to the xz-plane. The atom at $\phi_a$ is transformed into the atom at $-\phi_a$ under the mirror symmetry operation. We thus expect the cancellation in the modified CDAD $\Delta I(\theta, \phi)$ when they are nonmagnetic equivalent atoms. In this we have specified the x-axis.

In this paper we furthermore apply this technique to ferromagnetic and antiferromagnetic surface layers with in-plane magnetization adsorbed on a nonmagnetic layer. The result
demonstrates potential use of this technique.

2. Theory

X-ray photoelectron diffraction (XPD) intensity \( I^\pm_\pm(k, \sigma) \) excited from the splitted subshell \( j_e (=3/2) \) measuring photoelectrons with momentum \( k \) and spin \( \sigma \) is written for the \( \pm \) X-ray circular polarization [11]

\[
I^\pm_\pm(k, \sigma) = | \sum_\alpha \sum_{\mu_e} e^{-i\mu_e} \sum_j e^{-k_e j} R_{\alpha\Lambda} \sum_{LL'} Y_{L'L'}(\hat{k})(1 - X)^{-1}|_{L'\Lambda} M^\pm_{L'L\alpha}(k; \sigma)|^2
\]

\[X^{\alpha\beta} = t^{\alpha\sigma}_l(k_r) G_{LL'}(k_r R_{\alpha\beta}) e^{-\kappa R_{\alpha\beta}(1 - \delta^{\alpha\beta})}
\]

where \( X \) is a spin-dependent square matrix, where matrix elements are labeled by atomic sites \((A, \alpha, \beta, \ldots)\) and angular momenta \( L \). \( d_{de}(k) \) is the distance from site \( \alpha \) to the surface of the solid along the direction of a photoelectron propagation \( k \), \( R_{\alpha\Lambda} \) is the position vector of the scatterer \( \alpha \) measured from photoelectron emitter \( A \).

In the dipole approximation, the photoexcitation matrix element \( M^\pm_{L\alpha\alpha}(k; \sigma) \) excited by circularly polarized light \((k // z)\) is given by [12,13]

\[
M^\pm_{L\alpha\alpha}(k; \sigma) = \sqrt{2 \pi} \int e^{i\mu_e k} \rho(l) G(L_e 1 \pm 1|L) \left( L_{1/2} | \sigma \mu_e \right),
\]

\[
\rho(l) = \int R_l(kr) R_{lc}(r) r^3 dr
\]

where \( \delta^{l\sigma} \) is the spin-dependent phase shift of \( l \) th partial wave at site \( \Lambda \), and \( R_l(kr) \) and \( R_{lc}(r) \) describe the radial parts of the \( l \) th partial wave and the core function. Gaunt integral \( G(L_e 1 \pm 1|L) = \int Y_{l1}(r) Y_{l1}(r) Y_{l1}(r) d\vec{r} \) is responsible for the angular momentum selection rule of the photoexcitation. The angular momentum representation of the site-\( l \) matrix \( t^{\alpha\sigma}_l(k) \) at site \( \alpha \) is given by

\[
t^{\alpha\sigma}_l(k) = -\frac{e^{2i\delta^{l\sigma}} - 1}{2ik}
\]

in terms of the phase shift \( \delta^{l\sigma} \) at site \( \alpha \) for spin \( \sigma \) and the photoelectron wave number \( k \).

We introduce modified CDAD which enables us to pick up only the information on magnetic atoms [10],

\[
\Delta I(\theta, \phi) = I^+(\theta, \phi) - I^-(\theta, -\phi).
\]

3. Results

Here we show some calculated results for perpendicular and in-plane magnetization models.

3.1. Cu-Gd\(_4\) model with perpendicular magnetization

First we consider perpendicularly magnetized Cu-Gd\(_4\) model, where circular polarized X-ray photons propagates parallel to \( z \)-axis. We consider photoemission from Cu 2p\(_{3/2}\) sublevel with kinetic energy of 100 eV \((\epsilon_k = 100 \text{ eV})\). We assume that Cu atoms are nonmagnetic whereas a Gd atom is strongly magnetized \( S = 7/2 \) which aligns parallel or antiparallel to the \( z \) direction. We consider the following three models: (a) ferro model, (b) antiferro model1 in which two Gd atoms have down spin at \( \alpha \) and \( \gamma \) sites, and (c) antiferro model2 in which two Gd atoms have down spin at \( \alpha \) and \( \beta \) sites.

Figure 1 shows the calculated modified circular dichroism \( \Delta I(\theta, \phi) \) for the different spin orders. Figure 1(a) shows four positive spots corresponding to forward focusing Cu→Gd (up spin) scatterings. Figure 1(b) shows two positive spots corresponding to the forward Cu→Gd
(up spin at $\beta$ and $\delta$) scatterings, and two negative spots to Cu→Gd (down spin at $\alpha$ and $\gamma$) scatterings. This approach can clearly distinguish ferromagnetic order from antiferromagnetic order [10]. We also consider the other antiferromagnetic model (c) where $\alpha$ and $\beta$ Gd atoms are magnetized with down spin and $\gamma$ and $\delta$ with up spin. The calculated modified circular dichroism is shown by (c) where $\alpha$ and $\gamma$ are in xz-plane, and is shown by (c)' where x axis is positively rotated around z-axis. In (c) the mirror symmetry in regard to xz-plane is lost. Only the spots for $\alpha$ and $\gamma$ which are in the xz-plane are found, and spots for $\beta$ and $\delta$ disappear. In (c)' the mirror symmetry in regard to the new xz-plane correctly gives rise to magnetic atom imaging. When we rotate the xy-plane with $-45^\circ$, we obtain perfectly green pattern for $\Delta I(\theta, \phi)$: We find no spot. We thus understand the importance of the mirror symmetry in regard to xz-plane.

In this paper we use the full multiple scattering calculations as given by eq.(1). In the previous paper the plane wave approximation was used at the same energy, $\epsilon_k = 100$ eV. The result is quite similar to each other, where four positive spots are found for the ferromagnetic order in the plane wave approximation [10].

![Diagram](image_url)

**Figure 1.** Left: Cu-Gd$_4$ models magnetized perpendicular to the surface: (a) ferro, (b) antiferro1 ($\alpha$ and $\gamma$ are magnetized with down spin), (c) antiferro2 ($\alpha$ and $\beta$ are magnetized with down spin). Right: Calculated modified CDAD $\Delta I(\theta, \phi)$. The calculated $\Delta I(\theta, \phi)$ are shown for the model (c) by (c) and (c)', where $\alpha$ and $\gamma$ are in the xz-plane, and xy-plane is rotated with $45^\circ$ around z-axis. In (c) we have no mirror symmetry with respect to xz-plane, whereas in (c)' we have the symmetry.

### 3.2. Cu-Gd$_4$ model with in plane magnetization

Next we consider in-plane magnetized Cu-Gd$_4$ model where Gd has $S= 7/2$ aligned parallel or antiparallel to the x direction with same photoelectron kinetic energy of 100 eV ($\epsilon_k = 100$ eV). Figure 2 shows $\Delta I(\theta, \phi)$ for the different spin orders (a) ferro model in which all Gd atoms are ordered parallel to x direction, (b) antiferro model1 which has two Gd with spin parallel to x direction at $\alpha$ and $\gamma$ sites, and (c) antiferro model2 which has two Gd with spin parallel to x direction at $\alpha$ and $\beta$ sites: Other two Gd atoms are magnetized in opposite direction.

Figure 2(a) shows the two negative and positive spots corresponding to Cu→Gd forward scatterings even for the ferromagnetic model. The inversion symmetry in regard to the center observed in Figs. 2(a) and (b) is explained by the symmetry consideration. The mirror symmetry
with respect to xz-plane gives rise to spin inversion of the spin x component $S_x$, $S_x \rightarrow -S_x$, and $Y_{11} \rightarrow Y_{1-1}$. The focusing spot near $\alpha$ site is approximately written

$$\Delta I(\alpha) = I_{x-}^+(\alpha) - I_{x-}^-(\alpha).$$

Rotation around z-axis with 180° gives by noting that $S_x \rightarrow -S_x$, and $Y_{1\pm 1} \rightarrow Y_{1\mp 1}$

$$\Delta I(\gamma) = I_{x-}^+(\gamma) - I_{x-}^-(\gamma) = I_{x+}^+(\alpha) - I_{x+}^-(\alpha).$$

Mirror symmetry operation in regard to xz-plane yields a relation

$$\Delta I(\gamma) = I_{x-}^+(\alpha) - I_{x-}^-(\alpha).$$

(7)

Comparing eqs.(7) and (8), we obtain a relation between $\Delta I(\alpha)$ and $\Delta I(\gamma)$

$$\Delta I(\alpha) = -\Delta I(\gamma).$$

(8)

In the same way, we can show that $\Delta I(\beta) = -\Delta I(\delta)$ in the model (a). That is also the case for the antiferromagnetic model (b). When the lack of the mirror symmetry as in (c), two spots corresponding to $\beta$ and $\delta$ are missing in the present imaging technique. In the imaging (c)' we find four spots because of the mirror symmetry. In this case we find that $\Delta I(\alpha) = \Delta I(\gamma)$, and $\Delta I(\beta) = \Delta I(\delta)$. This is easily explained by rotating the xy-plane with 180°

$$\Delta I(\gamma) = I_{x-}^+(\gamma) - I_{x-}^-(\delta) = I_{x+}^+(\alpha) - I_{x+}^-(\beta) = \Delta I(\alpha).$$

(10)

Figure 2. Left: Cu-Gd$_4$ model magnetized parallel to the surface: (a) ferro, (b) antiferro1 ($\alpha$ and $\gamma$ are down spin), (c) antiferro2 ($\alpha$ and $\beta$ are magnetized with down spin). Right: Calculated modified CDAD $\Delta I(\theta, \phi)$. The calculated $\Delta I(\theta, \phi)$ are shown for the model (c) by (c) and (c)', where $\alpha$ and $\gamma$ are in the xz-plane, and xy-plane is rotated with 45° around z-axis. In (c) we have no mirror symmetry with respect to xz-plane, whereas in (c)' we have the symmetry.
3.3. Fe/Cu ferro model with perpendicular magnetization

So far we have considered photoemission from one Cu atom. In real surface XPD spectra the photoemission from the second, the third, ... layers also contributes to the dichroism. As examples of more realistic systems, we consider iron monolayer and double layers on Cu(001) substrate. The model cluster is shown in Figure 3. Figure 3(a) and (b) show the calculated modified circular dichroism $\Delta I(\theta, \phi)$ for the ferromagnetic model with perpendicular magnetization. In (a) Fe monolayer and (b) Fe double layers on Cu substrate ($\epsilon_k = 90$ eV) are considered. The peak image from double-layer model is more complicated than from the monolayer model. We still have clear imaging of the magnetic surface layers.

![Figure 3. 2 layer Fe/Cu model (left). Calculated modified CDAD $\Delta I(\theta, \phi)$ for (a) 1 layer Fe/Cu ferromagnetic model and (b) 2 layer Fe/Cu ferromagnetic model.](image)

3.4. Fe/Cu ferro model with in plane magnetism

We next discuss the in-plane magnetized model of the same surface systems considered in Figure 3. Figures 4(a) and (b) show the calculated modified $\Delta I(\theta, \phi)$ for the ferromagnetic model magnetized in plane. In (a) Fe monolayer and (b) Fe double-layer on Cu substrate ($\epsilon_k = 90$ eV) are considered. The peak images in (a) and (b) become complicated and it is difficult to obtain useful information on surface magnetic order.

![Figure 4. 2 layer Fe/Cu model (left). Calculated modified CDAD $\Delta I(\theta, \phi)$ for (a) 1 layer Fe/Cu ferromagnetic model and (b) 2 layer Fe/Cu ferromagnetic model.](image)
4. Conclusion

We proposed a possible method to obtain only direct imaging of magnetic atoms by use of the photoelectron diffraction method that does not require any expensive spin detector. This approach applies the cancellation of the Daimon effect which rotates the forward focusing PD peaks with finite angles. The modified circular dichroism $\Delta I(\theta, \phi)$ picks up the information only of magnetic atoms. Illustrative numerical calculations clearly distinguish the magnetic atoms with different spin orientation in the case of the single magnetic layer on Cu substrate. Thicker magnetic layer, for example, double magnetic layers make the atomic image unclear because of increasing the number of emitter. The method is useful to study magnetic monolayer on nonmagnetic substrate. The images obtained for in-plane magnetized models show rather complicated features, however their symmetric patterns are well explained by use of symmetry consideration. We hope experimental studies will be performed to obtain the imaging magnetization as we proposed here.

Acknowledgement

The authors are grateful to have fruitful discussions with Dr. H. Shinotsuka.

References

[1] J. Bansmann, C. Ostertag, G. Schönhence, F. Fegel, C. Westphal, M. Getzlall, F. Schafers, H. Petersen, Phys. Rev. B 46 13496 (1992)
[2] H. Daimon, T. Nakatani, S. Imada, S. Suga, Y. Kagoshima, T. Miyahara, Jpn. J. Appl. Phys. 32 L1480 (1993)
[3] T. Fujikawa, M. Yimagawa, J. Phys. Soc. Jpn. 63 4220 (1994)
[4] T. Fujikawa, Physica B 93 208-209 (1995)
[5] P. Rennert, A. Chassé, T. Nakatani, K. Nakatsuji, H. Daimon, S. Suga, J. Phys. Soc. Jpn. 66 396 (1997)
[6] A. Chassé, P. Rennert, Phys. Rev. B 55 4120 (1997)
[7] H. Daimon, Phys. Rev. Lett. 86 2034 (2001)
[8] T. Nakatani, T. Matsushita, Y. Miyake, T. Nohno, A. Kobayashi, K. Fukumoto, S. Okamoto, A. Nakamoto, F. Matsui, K. Hattori, M. Kostugi, Y. Saitoh, S. Suga, H. Daimon, Prog. Surf. Sci. 71 217 (2003)
[9] F. Matsui, T. Matsushita, F.Z. Guo, H. Daimon, Surf. Rev. Lett. 14 1 (2007)
[10] M. Wada, K. Ito, T. Konishi, T. Fujikawa, Surface Science 602 2907-2914 (2008)
[11] H. Shinotsuka, H. Arai, T. Fujikawa, Phys. Rev. B. 77 085404 (2008)
[12] K. Ito, T. Konishi, T. Fujikawa, J. Surf. Anal. 14 328 (2008)
[13] K. Ito, T. Konishi, T. Fujikawa, e-J. Surf. Nanotech. 7 115 (2009)