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Theoretical studies on magnetic atom imaging by use of photoelectron diffraction

Masahiko Wada, Kouta Ito, Takehisa Konishi, Takashi Fujikawa *

Graduate School of Advanced Integration Science, Chiba University, 1-33 Yayoi-cho, Inage-ku, Chiba 263-8522, Japan

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A B S T R A C T

We theoretically study possible methods how to obtain imaging of magnetic atoms by use of photoelectron diffraction (PD). We propose a novel method to apply Daimon effect where PD peaks are rotated around forward focusing peaks. In usual circular dichroism, we simply use the difference of the PD intensities for different X-ray circular polarization. In contrast to this dichroism, we rather use the difference of the PD intensities for different circular polarizations but $-\phi$ is used for the – circular polarization only in the data handling. This technique allows us to obtain clear atomic image only of spin polarized atoms, and to distinguish magnetic atoms with up-spins from those with down spins. Some illustrative calculations demonstrate the potential use and also the limitation of this technique.

1. Introduction

Photoelectron diffraction (PD) from a deep core level of surface atom is useful to study local geometric structures of solid surfaces [1]. For those purposes PD spectra excited by linearly polarized and unpolarized X-ray photons have been used by many authors. Later 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) [2]. Daimon et al. have also observed strong CDAD excited from Si 2p core level on Si(001) surface, although this system is not chiral [3]. 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 [4–7] 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 [8–10].

The study of magnetic phenomena at solid surface has been greatly stimulated by the availability of several techniques which are capable to probe the first layer of a sample. Among them spin polarized low energy diffraction (SPLEED) [11] and spin polarized photoemission (SPPES) [11,12] are useful to study long-range magnetic order of surface layers. In contrast to these methods some useful methods using the PD techniques have been developed. For example the exchange coupling occurring in the final state after Mn 3s emission from Mn^{2+} in $KMnF_3$ splits apart the spin-up and spin-down photoelectron peaks in energy, so that a simple measurement of the spectrum yields peaks with different spin character, $S \pm 1/2$. There two peaks might then be expected to exhibit different spin-dependent scattering in a magnetically ordered environment [13,14]. This technique had been termed spin-polarized photoelectron diffraction (SPPD), and had been applied to other systems as MnO [15]. Timmermans et al. theoretically point out that spin–orbit correlations in the photolectron initial state are responsible for holographic spin-dependent contributions to the hologram intensity [16].

So far SPPD theory has been developed on the basis of one-electron nonrelativistic theory [13,17]. Further refinement has been found to include relativistic effects [18,19], and furthermore many-body effects based on the quantum electrodynamics (QED) [20,21]. We, however, use rather simple one-electron relativistic theory to provide simple physical interpretation.

In this paper we theoretically study a possible way to visualize magnetic atoms by use of circularly polarized light and excited photoelectrons from nonmagnetic atoms. This technique can distinguish magnetic atoms with up-spin polarization from those with down-spin polarization where the quantum axis is parallel to the X-ray incidence. We use the modified circular dichroism instead of the conventional circular dichroism: We use the difference of the PD intensities for the different X-ray circular polarization but $-\phi$ is used for the – circular polarization only in the data handling. So far no experimental and no theoretical approach has been found for that purpose.

2. Theory

Photoemission intensity measuring photoelectrons with momentum $\boldsymbol{k}$, spin σ is given for the $\pm$ X-ray circular polarization

* Corresponding author.
E-mail address: tfujikawa@faculty.chiba-u.jp (T. Fujikawa).

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$$I_{\pm}^{j_c}(\boldsymbol{k},\sigma) \propto \sum_{A} \sum_{\mu_c=-j_c}^{j_c} |Z_1^{\mu_c}(\boldsymbol{k};\sigma)_{\pm} + Z_2^{\mu_c}(\boldsymbol{k};\sigma)_{\pm} + \cdots| \tag{2.1}$$

where $Z_1^{\mu_c}(\boldsymbol{k};\sigma)_{\pm}$ is the direct term which suffers no elastic scatterings from surrounding atoms, $Z_2^{\mu_c}(\boldsymbol{k};\sigma)_{\pm}, Z_3^{\mu_c}(\boldsymbol{k};\sigma)_{\pm}, \ldots$ are the single, double, ... scattering amplitudes excited from a core function $\phi_c = R_{j_c l_c}(r) y_{j_c \mu_c}^{l_c}(r)$ ($y_{j\mu}^{l}$ is the Pauli spinor) [4]. The formula (2.1) can be derived in semirelativistic theoretical framework, where 2p core is split into $2p_{1/2}$ and $2p_{3/2}$ subshells. We apply the formula (2.1) to the photoemission from nonmagnetic atoms like Cu to get rid of multiplet splittings like Mn 3s photoemission from MnO crystals [15]. In this case effective one-electron formula (2.1) is a good approximate theoretical tool to analyze the SPPD pattern. For the later discussion we only show explicit formulas excited from a deep core $2p_{3/2}$ orbital ($j_c = 3/2$),

$$Z_1^{\mu_c}(k;\sigma)_{\pm} = \sum_{L} Y_L(\hat{\boldsymbol{k}}) M_{L\ 1m_c}^{\pm}(k;\sigma), \tag{2.2}$$

$$Z_2^{\mu_c}(k;\sigma)_{\pm} = \sum_{\alpha L} \frac{e^{ikR_\alpha(1-\cos\hat{\theta}_\alpha)}}{R_\alpha} f_\alpha^\sigma(\hat{\theta}_\alpha) Y_L(\hat{\boldsymbol{R}}_\alpha) M_{L\ 1m_c}^{\pm}(k;\sigma), \tag{2.3}$$

$$Z_3^{\mu_c}(k;\sigma)_{\pm} = \sum_{\alpha\neq\beta} \frac{e^{ikR_{\beta\alpha}(1-\cos\theta_{\beta\alpha})+ikR_\alpha}}{R_{\beta\alpha}R_\alpha} f_\beta^\sigma(\theta_{\beta\alpha}) f_\alpha^\sigma(\theta_{\beta\alpha A}) \tag{2.4}$$
$$\times \sum_{L} Y_L(\hat{\boldsymbol{R}}_\alpha) M_{L\ 1m_c}^{\pm}(k;\sigma), \quad (m_c = \mu_c - \sigma/2)$$

where $M_{LL_c}^{\pm}(k;\sigma)$ is the atomic photoexcitation amplitude at the site A given in terms of the spin-dependent phase shifts $\delta_l^{A\sigma}$ on the site A and the radial integral $\rho_c(l)^\sigma$,

$$M_{LL_c}^{\pm}(k;\sigma) = \sqrt{\frac{2}{\pi}} i^{-l} \exp(i\delta_l^{A\sigma}) G(L_c\ 1 \pm 1|L) \left\langle L_c\ \frac{1}{2}\ \frac{\sigma}{2} | j_c \mu_c \right\rangle \rho_c(l)^\sigma. \tag{2.5}$$

The Gaunt integral $G(L_c 1 \pm 1|L) = \int Y_L^*(\hat{\boldsymbol{r}}) Y_{1\pm1}(\hat{\boldsymbol{r}}) Y_{L_c}(\hat{\boldsymbol{r}}) d\hat{\boldsymbol{r}}$ restricts $l = l_c \pm 1$, $m = m_c \pm 1$. The scattering angle $\hat{\theta}_\alpha$ is given by $\cos\hat{\theta}_\alpha = \hat{\boldsymbol{R}}_\alpha \cdot \hat{\boldsymbol{k}}$, and $\cos\theta_{\beta\alpha A} = \hat{\boldsymbol{R}}_{\beta\alpha} \cdot \hat{\boldsymbol{R}}_\alpha$, ($\boldsymbol{R}_{\beta\alpha} = \boldsymbol{R}_\beta - \boldsymbol{R}_\alpha$). The spin-dependent scattering amplitude $f_\alpha^\sigma(\hat{\theta}_\alpha)$ at the site α, $f_\beta^\sigma(\theta_{\beta\alpha})$ at the site β are key factors to give rise to imaging of magnetic atoms. They have a sharp peak in the forward direction, which is responsible for the forward focussing peak.

In the lowest approximation where the elastic scatterings from surrounding atoms are completely neglected, the photoemission intensity is simply given by

$$I_{\pm}^{3/2}(\boldsymbol{k},\sigma) \propto \rho_c(0)^2 - 4P_2(\cos\theta)\rho_c(0)\rho_c(2)\cos(\delta_0^{A\sigma} - \delta_2^{A\sigma}) + (3\cos^2\theta + 1)\rho_c(2)^2, \tag{2.6}$$

which are spin independent for nonmagnetic X-ray absorbing atoms: $\delta_l^{A+} = \delta_l^{A-}$. On the other hand when we take the scattering effects into account, the phase difference of $Y_{l,m_c\pm1}(\hat{\boldsymbol{R}}_\alpha)$ in $Z_2^{\mu_c}(\boldsymbol{k};\sigma)_{\pm}, Z_3^{\mu_c}(\boldsymbol{k};\sigma)_{\pm}, \ldots$ can give rise to the CDAD and also the peak rotation (Daimon effects) [3].

In the present calculations we use spin-dependent local optical potential developed by von Barth and Hedin [22]. More sophisticated optical potential are also available in literatures [23,24].

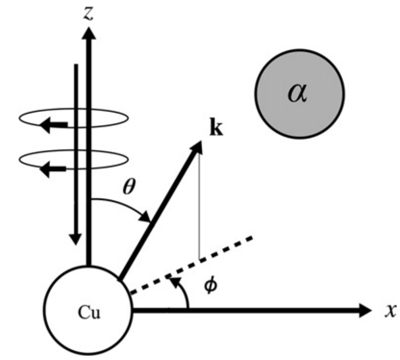

Fig. 1. Diatomic model where circularly polarized X-ray photons propagate in the z-direction. The scattering atom α is in the xz-plane with $\theta_\alpha = 45°$, $\phi_\alpha = 0°$.

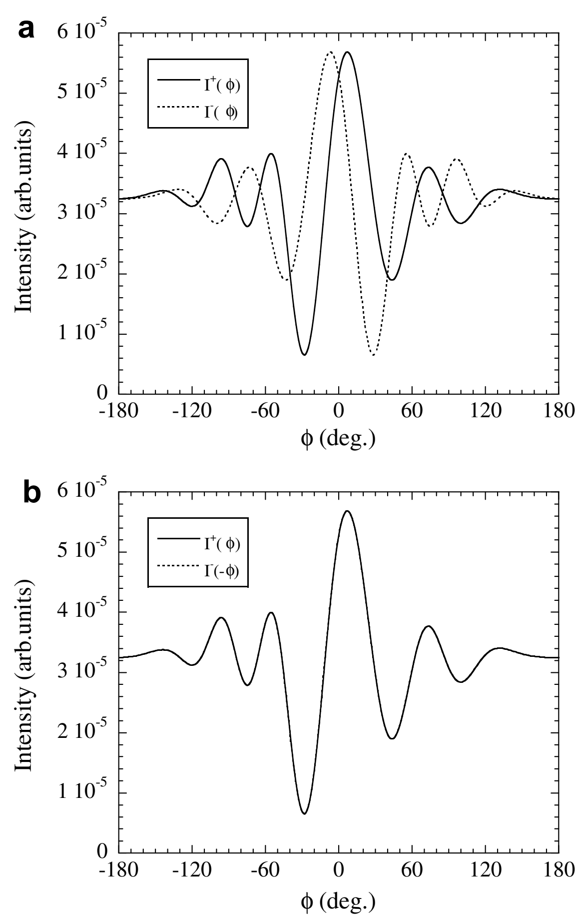

Fig. 2. Calculated XPD patterns for the photoemission from Cu $2p_{3/2}$ level with kinetic energy 100 eV ($\epsilon_k = 100$ eV) fixed at $\theta = \theta_\alpha = 45°$. The scatterer is a nonmagnetic atom (Cu). In (a) $I^{\pm}(\phi)$, and in (b) $I^{+}(\phi)$ and $I^{-}(-\phi)$ are shown. We observe that $I^{+}(\phi) - I^{-}(-\phi) = 0$.

3. Photoelectron diffraction pattern and circular dichroism

As described before, strong forward focusing peaks are expected along the direction from the emitter to the scatterers. At first let us consider simple diatomic systems, Gd–Cu and Cu–Cu as shown in Fig. 1, where circular polarized X-ray photons propagate parallel to z-axis. We consider photoemission from Cu $2p_{3/2}$ level with kinetic energy 100 eV ($\epsilon_k = 100$ eV): The scatterer Cu or Gd is in the xz-plane ($\theta_\alpha = 45°, \phi_\alpha = 0°$). We assume that Cu atoms are nonmagnetic whereas Gd have 7 up-spins parallel to z-axis. Fig. 2a shows the calculated XPD patterns $I^{\pm}(\phi)$ for ± circular polarization fixed at $\theta = \theta_\alpha = 45°$ for the Cu–Cu diatomic system where we assume that Cu–Cu distance is 2.56 Å. As expected the Daimon effect is clearly observed; the focusing peak around $\phi = 0°$ is rotated with 7°. We thus observe the spot at different position for different circular polarization. We also observe slowly oscillating tail due to the phase factor $\exp[ikR_\alpha(1 - \cos\hat{\theta}_\alpha)]$ in Eq. (2.3). Fig. 2b shows