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Observation of an interference effect for fluorescent x rays

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A peculiar angular pattern is observed from $K\alpha$-fluorescent x rays emitted from a Zn monoatomic layer in a Langmuir-Blodgett film onto a Au substrate. A pattern similar to Young's fringe is monitored with a non-energy-dispersive two-dimensional detector (imaging plate), and is quantitatively measured by scanning the slit with an energy-dispersive detector (pure Ge detector). The fringes are in close agreement with a theoretical estimate based on the interference among transmitted and reflected waves at interfaces in the sample.

The characteristic x-ray intensity emitted from surface atoms is maximized at the critical angle for total external reflection. This phenomenon has been reported by Hasegawa *et al.*[1,2] in the case of electron-impact excitation. The same phenomenon also has been reported in the case of the fluorescent x rays excited with an x-ray beam by Sasaki and Hirokawa.[3,4] These anomalous distributions were accounted for by the effects of refraction[1] and total external reflection[5] of fluorescent x rays.

Suzuki and Hasegawa[6] interpreted the angular distribution quantitatively by using the interference effect between the direct beam and the specularly reflected beam as shown in Fig. 1. The angular distribution for fluorescent x rays is then given by

$$I(\theta_t,z)=|E_t+E_r|^2 , \qquad (1)$$

where θ_t is the take-off angle for fluorescent x rays, and z is the distance between the source point (atom) of the fluorescent x rays and the reflecting surface, and E_t and E_r are the transmitted and reflected E-field plane waves for the fluorescent x rays. Although x-rays reflectivity is usually very small, about 10^{-5}, at normal incidence, it is nearly 100% below the critical angle (θ_c) for external total reflection. Thus, interference fringe (fluorescent x-ray interference: FXI) patterns can be clearly observed when

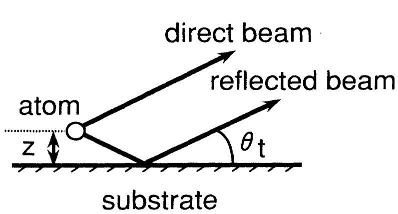

FIG. 1. Illustration of fluorescent x-ray interference (FXI) between direct beams and reflected beams at the substrate. Z is the distance between the substrate and the atom excited by the primary x-ray beam.

θ_t is smaller than θ_c.

The geometrical path difference Δ between E_t and E_r is described as

$$\Delta=2z\sin\theta_t . \qquad (2)$$

Therefore, positional information about the source atoms for the fluorescent x ray can be obtained through z by observing the FXI pattern.

To separate atoms from the substrate, a low-density layer which contains source atoms was deposited on a high-density substrate.[7,8] We used a Langmuir-Blodgett (LB) film for polydiacetylene[9] on a Au substrate, and a Zn arachidate is used as a source layer for the fluorescent x rays. A schematic cross-sectional view of the sample is shown in Fig. 2(a). Z was estimated at about 144 Å as follows: The thickness of the polydiacetylene monolayer was 29 Å measured by x-ray diffraction, and that of the Zn arachidate bilayer was 55 Å. The polydiacetylene was poly-(dibutyl 1-4,17-dioxo-5,16-dioxa-3,18-diaza-9,11-eicosadiynedioate), poly-3BCMU. This was prepared by uv photopolymerization of monomer crystals.

The experimental setup is shown in Fig. 2(b).[3,4] This experiment was carried out at the BL-8C2 experimental station in KEK Photon Factory 2.5 GeV storage ring. The stored current was between 300 and 350 mA. Synchrotron radiation was monochromated with a pair of Si(111) crystals and was tuned to an energy level where the Zn $K\alpha$ radiation became optimally excited. The output beam from the monochromator was monitored by an ionization chamber. The sample's incident x-ray beam was narrowed to 100 μm vertical×6 mm horizontal by slits. Incident radiation was linearly polarized, and the electrical vector was in a horizontal plane. Fluorescent radiation was measured at a 90° scattering angle in the horizontal plane to minimize background signals caused by elastic scattering.

The primary x-ray beam at a glancing angle of 3 mrad is totally reflected, and scattering background signals as

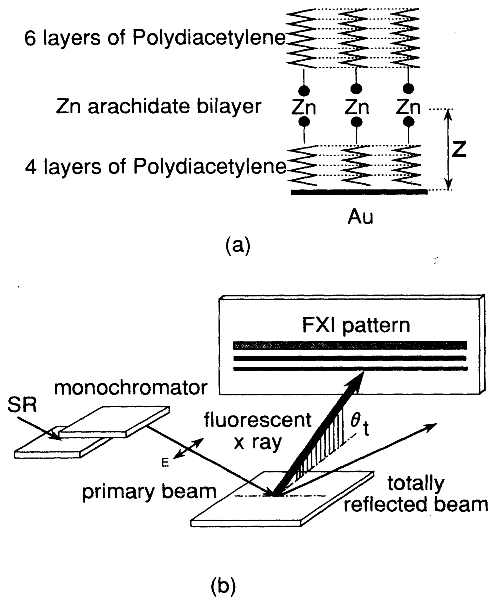

FIG. 2. (a) Structure of the sample: Cross section of the measured Zn containing LB film. (b) Schematic drawing of the experimental apparatus for FXI measurement. All experiments were carried out in air. The glancing angle of the primary x-ray beam is fixed at about 3 mrad in this experiment. E is direction of the electrical vector.

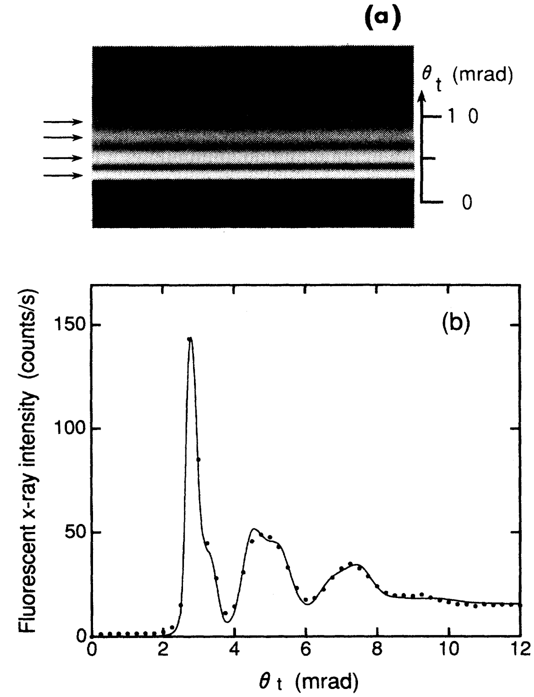

FIG. 3. (a) Zn $K\alpha$ FXI patterns were measured with an IP. The exposed time is 3 hours. High intensity areas are shown by the arrows. (b) The theoretical calculation (solid line) and the experimental result (closed circles) of Zn $K\alpha$-FXI pattern were measured by slit scanning method with a pure Ge detector. The amplitude of the calculated fringes was normalized based on the experimental FXI pattern. The signal integration time is 200 sec per point. Total thickness of the LB film is 566 Å. The complex refractive indices for air, LB film, and Au film are 1, $1-(3.26\times10^{-6})-(4.81\times10^{-9})i$, and $1-(3.67\times10^{-5})-(3.17\times10^{-6})i$ at 1.437 Å (the wavelength of the Zn $K\alpha$ line), respectively. These are fitting parameters in the χ^2 minimization.

well as fluorescent x rays from the substrate is well suppressed.[3] Furthermore, total intensity for the fluorescent x rays are strongly dependent upon the glancing angle.[7,8] Thus, the glancing angle of the primary beam is fixed at a glancing angle where the total intensity of the fluorescent x ray corresponds to the maximum value. The exposed area for the sample is 6×30 mm.

Angular distribution for fluorescent x rays is monitored with an imaging plate (IP, Fuji Film Co.)[10] as a non-energy-dispersive detector and a pure Ge detector as an energy-dispersive detector. We used the IP as a two-dimensional detector to observe the overall structure of the interference fringes. The distance from the sample to the IP was 900 mm. The IP was read out by a BAS-2000 system (Fuji Film Co.). However, the IP has no energy selectivity. Thus, a Ge detector was used as a fluorescent x-ray detector to compare the experimental result with the theoretical one. A slit 50 μm vertical$\times$12 mm horizontal was placed before the detector at a distance of 200 mm from the sample to determine the take-off angle. The angular distribution of the fluorescent x rays was measured by scanning the slit along the transversal direction. This arrangement enabled us to measure an angular resolution of less than 0.5 mrad below θ_t for 8 mrad.

Figure 3(a) shows the FXI pattern for Zn $K\alpha$ radiation emitted from a Zn monoatomic layer in the LB film with the IP. In the result of IP measurement, the take-off angle for the first-order fringe was determined by fitting to that measured by the Ge detector. The signal attained by the IP in our experiment almost completely resulted from Zn $K\alpha$ radiation. The background signal caused by elastic scattering and other fluorescent radiation were suppressed to very low levels by polarization and the total external reflection of the monochromatic primary beam. These effects were confirmed by using the Ge detector. The first-order fringe at $\theta_t\sim3$ mrad, the second order at $\theta_t\sim5$ mrad, and the third order at $\theta_t\sim7$ mrad are shown in Fig. 3(a). The fourth order at $\theta_t\sim9$ mrad can be seen when the signal is summed along the horizontal direction.

Figure 3(b) shows the angular distribution for Zn $K\alpha$ radiation measured with a Ge detector, which was set for this radiation. The zero point of take-off angle was the angle at which the intensity for fluorescent x rays measured with the Ge detector disappeared. The statistical error rate for the experiment data, defined by the square root of the number of detected photons, corresponded to less than half the diameters for the circles. The positions of the four fringes shown in Fig. 3(a) are in close agreement with the values in Fig. 3(b).

We calculated the angular distrbution $I(\theta_t,z)$ for the fluorescent x rays with a characteristic matrix of a stratified medium.[11] The intensity of $I(\theta_t,z)$ can be determined by using the reciprocity theorem.

When a fluorescent x-ray source has a distribution expressed by $N(z)$, the partial yield can be written by this formula:

$$I(\theta_t)=\int N(z)I(\theta_t,z)dz \ . \quad (3)$$

The theoretical FXI pattern in Fig. 3(b) can be determined by χ^2 minimization, which was designated for the experimental data measured by the Ge detector. This fitting data was uniform and had roughness-free interfaces. As a result of the χ^2 minimization, we can obtain values for the refractive indices of the LB film and the Au substrate, the total thickness of the LB film, and the coordinate (z) of the Zn atomic layer. Except for the position of the Zn atomic layer, all information can be obtained by measuring x-ray reflectivity. The values for the refractive indices determined by reflectivity of the Au substrate and the LB film on the Au substrate at a Zn $K\alpha$ wavelength line, and the value for the total thickness of the LB film on the Au substrate were in close agreement with the values attained by the χ^2 minimization.

We assume that the Zn atoms have a Gaussian shape distribution curve in this FXI pattern calculation. As a result of applying the calculated FXI pattern to the experimental data, the Zn atomic layer is shown to be at a mean position $z=227\pm3$ Å, and has a width of $\sigma_z=26\pm2$ Å. These values (z and σ_z) were 63.8% data reliable. The measured z value is larger than what was expected. We expected the σ_z value to be smaller than that for the experimental results because the source layer is monoatomic. The difference between the expected values and the experimental results can be accounted for by misfitting at the LB film-Au substrate interface caused by the surface roughness and LB film stacking faults.[12]

According to calculations, the peak shape for the first-order fringe at 3.0 mrad is influenced by the presence of an air-LB film interface. This phenomenon is caused by the refractive effect[1] that occurs here. Without this effect, the peak height for the FXI pattern would be estimated to be about 4 times that for above θ_c for the air-Au interface. The peak height in the experiment is roughly 11 times higher.

The same information for x-ray standing waves[7,8] (XSW) generated by total external reflection can be obtained so that for measuring fluorescent x-ray intensity as a function of the glancing angle for the primary x-ray beam. Here must be considered the substrate reflection and the surface refraction.[13]

The signal intensity for our FXI measurements is lower than that of the XSW.[7] However, FXI measurements with energy sensitive detectors have some advantages. Fluorescent radiation occurs independently from the primary beam. Parallel and monochromatic x rays are unnecessary, and white x-ray beams, electrons and ions can be used as the primary beam.

In summary, we observed a peculiar angular distribution for fluorescent x rays emitted from a Zn monoatomic layer in a Langmuir-Blodgett film onto a Au substrate. The observed fluorescent x rays show patterns similar to these of Young's fringe. This pattern was in close agreement with the theoretical estimate for interference among transmitted and reflected waves at interfaces in the sample. In addition, the enhanced peak height was assigned to the refractive effect for the air-LB film interface.

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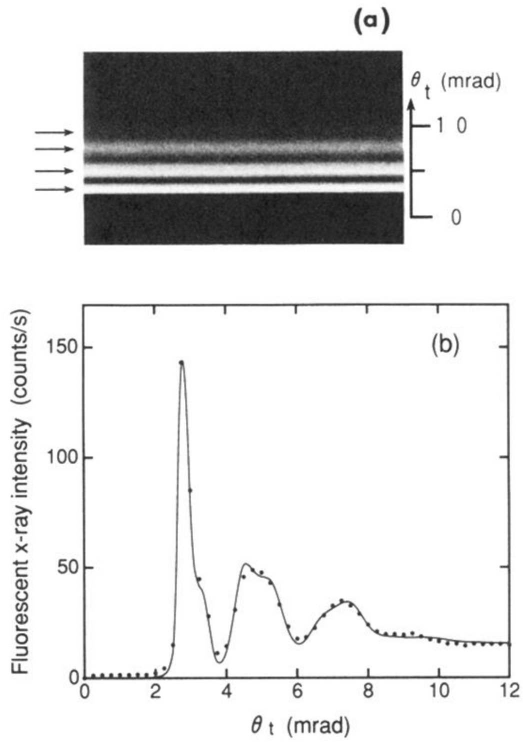

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