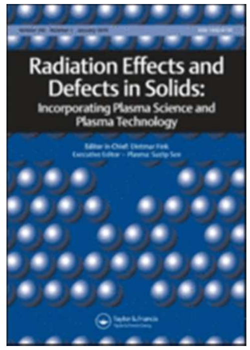


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|---|---|
| Journal: | *Radiation Effects and Defects in Solids* |
| Manuscript ID: | Draft |
| Manuscript Type: | Special Issue Article - REM-7-2014 |
| Date Submitted by the Author: | n/a |
| Complete List of Authors: | Ichinokura, Satoru; Univ. of Tokyo,<br>Hirahara, Toru; Tokyo Inst. of Tech.,<br>Hasegawa, Shuji; Univ. of Tokyo,<br>Sakai, Osamu; Natl. Inst. Mat. Sci.,<br>suzuki, taku; National Institute for Materials Science, |
| Keywords: | ions, metals, surfaces |
| | |

To appear in *Radiation Effects and Defects in Solids*
Vol. 00, No. 00, Month 20XX, 1–5

# Electron-spin dependent $^4He^+$ ion scattering on Bi surfaces

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(*v2.0 released January 2014*)



We studied low-energy (∼1.55 keV) electron-spin polarized $^4He^+$ ion scattering on Bi(111) ultrathin film epitaxially grown on a Si(111) substrate. We observed that the scattered ion intensity differed between the incident $He^+$ ions with up and down spins even though Bi is a non-magnetic element. To analyze the origin of this spin dependent ion scattering (the spin asymmetry) we investigated the detailed relationship between the spin asymmetry and the incident angle, the azimuthal angle, the scattering angle, and the incident energy. All the data indicates that the spin asymmetry originates from the scattering cross section owing to the non-central force in the $He^+$-Bi atom binary collision. The non-central force is mostlikely attributed to the spin-orbit coupling that acts transiently on the $He^+$ 1s electron spin in the binary collision.

**Keywords:** low energy ion scattering, electron-spin, spin-orbit coupling, atomic collision

## 1. Introduction

Particle-surface interaction has attracted attention since it is involved in a number of aspects related with both fundamental science and practical applications. It is particularly important in surface analysis by ion beams, such as low energy ion scattering spectroscopy (LEIS) (*1*), secondary ion mass spectroscopy (*2*), elastic recoiled detection analysis (*3*), and recent helium ion microscopy (*4*). Helium ion ($He^+$) is often utilized as a primary ion beam. This is because it is neutralized with quite high probability in the vicinity of surfaces owing to its large ionization energy, hence it is surface sensitive particularly with low energy of a few keV or less(*5*).
Ion scattering is also sensitive to surface electron-spin. One of the reason is that the ion neutralization obeys the Pauli exclusion principle. Because of the Pauli exclusion principle, a surface electron involved in the neutralization (Auger neutralization) has a spin whose direction is opposite to that of the $He^+$ 1s electron (*6, 7*). The other mechanism is spin-orbit coupling (SOC) which acts transiently on the $He^+$ 1$s$ electron spin in the $He^+$ ion-target atom binary collision. We recently observed spin dependent $He^+$ ion scattering (*8*). Briefly, in our previous study, we made electron-spin polarized $He^+$ ion scattering experiment (spin polarized $He^+$ ion scattering spectroscopy (SP-ISS)) on non-magnetic surfaces. Because the $He^+$ ion neutralization probability is independent of spin on non-magnetic surfaces, the spin dependent scattering is not caused by the Pauli exclusion principle. From the limited experimental data on Au and Pb in our previous study, we

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interpreted this spin dependent scattering in terms of SOC. It is the effect on the $He^+$ spin of the magnetic field induced by the $He^+$ ion angular motion around the target nucleus during the projectile-target binary collision. The $He^+$ ion-surface atom interaction has been conventionally discussed in terms of a screened Coulomb potential(*1, 9*) The (screened) Coulomb potential is so-called a central force potential, and thus, it is essentially not related with electron spin. However, so large spin asymmetry (∼10% for Au) was observed even with so low incident energy as about 1 keV.
In the present study, we extended our SP-ISS experiment to a bismuth (Bi) target which is also a non-magnetic element. We found spin dependent ion scattering appears also on Bi targets. The experimental data on Bi is consistent with the above-mentioned SOC model in ion scattering.

## 2. Experimental Method and Setup

We performed experiments in a high vacuum chamber (base pressure of $2 \times 10^{-9}$ Torr) equipped for SP-ISS(*10*). Electron-spin-polarized $^4He^+$ ions were generated by Penning ionization of spin-polarized metastable He $2^3S_1$ atoms (*11*). We employed an optical pumping technique to polarize metastable He $2^3S_1$ atoms(*12*). The polarization of the incident $He^+$ ion beam $P_{He^+}$ is defined as $(n_\uparrow - n_\downarrow)/(n_\uparrow + n_\downarrow)$, where $n_\uparrow$ and $n_\downarrow$ are numbers of projectile $He^+$ ions whose magnetic moment are parallel and antiparallel to the magnetic field, respectively. The spin polarization of the $He^+$ ion beam $P_{He^+}$ in the present experiment was about 0.2(*13*). The entire apparatus was surrounded by a three-axis coil to compensate the Earth magnetic field. An additional coil produced a weak guiding field (0.3 Oe), which was parallel to the vertical axis. Thus, the spin direction of the incident $He^+$ ion beam was defined by the guiding field, hence, it was polarized parallel or anti-parallel to the guiding field. To eliminate the effect due to the passage of time between the measurement for up and down spins, which is, for instance, the surface damage by the $He^+$ ion beam irradiation, surface contamination, and slight variation of $P_{He^+}$, we repeatedly changed the spin direction of the projectiles within 10 s. We set the scattering geometry as the spin of the incident $He^+$ ion to be perpendicular to both the scattering plane and the surface normal of the target. We hereafter define the incident angle $\alpha$ (exit angle $\beta$) as the angle between the surface normal and the incident (exit) ion beam. On the other hand, the scattering angle $\theta$ is defined as 180° - $\alpha$ - $\beta$ (See Figure 2 (a)). The scattered $He^+$ ions were measured using a rotatable hemispherical sector analyzer (Omicron SHA50). The measurements were conducted in a constant pass energy mode with a pass energy of 318 eV.

## 3. Sample preparation

Bi ultrathin films were grown by molecular beam epitaxy (MBE) in the same chanber of SP-ISS and characterized *in situ* using reflection high-energy electron diffraction (RHEED). Bi was deposited on the Si(111)-7x7 surface (n type,1 Ω cm) at room temperature and postannealed at 350 K, which makes the films atomically flat(*14*). In this paper, we use the rhombohedral indexing. One bilayer (BL) is defined as the atom density in the covalently bonded Bi(111) plane 1 (BL) $= 1.14 \times 10^{15}$ atoms/cm$^2$ and 0.39 nm thick. The thickness has been calibrated with RHEED by the completion of the Si(111)-$\sqrt{3} \times \sqrt{3}$-Bi phase which was also confirmed by the allotropic transformation from the Bi{012} phase into the Bi(111) phase(*14*). Figure 1 (a) shows a RHEED pattern of MBE-grown fresh Bi surface. The sharp spots correspond to diffraction from long-range ordered Bi(111) planes with a lattis constant of 0.45 nm(*15*). The Bi surface was relatively inert, and consequently, we successfully made (SP-)ISS measurements in the chamber with a base pressure of $2 \times 10^{-9}$ Torr without surface contamination.

## 4. Results and Discussion

Figure 2 (b) shows the SP-ISS spectra for up/down spin and their difference on Bi(111) surface. In the spectra, the scattering peak of Bi is observed at 1520 eV, which is consistent with the $He^+$-Bi atom binary collision energy. The scattered $He^+$ ion intensity is obviously different between up and down spins at the elastic peak position of Bi; it becomes maximum at the $He^+$-Bi binary collision energy. Considering that the spin asymmetry of the Fe(100) surface in the magnetic remanent state is about 5% (*7*), this spin asymmetry (about 5%) is obviously not due to the diamagnetism of Bi because the magnetic susceptibility of Bi ($-3.55\times10^{-9}$(*16*)) is too small to explain it. Thus, surface magnetism is not responsible for the spin asymmetry on the Bi target.
Figure 3 shows spin asymmetry $A$ at the elastic scattering peak position on a Bi obtained as a function of the 1.5 keV $He^+$ ion beam irradiation time. The ion beam flux at the Bi target position was about $2\times10^{11}$ $cm^{-2}s^{-1}$. The spin asymmetry $A$ is defined as $A = (I_\uparrow - I_\downarrow)/[P_{He^+}(I_\uparrow + I_\downarrow)]$ , where $I_\uparrow$ and $I_\downarrow$ are the scattered $He^+$ ion intensity using the incident $He^+$ ions with up and down spins, respectively. The distribution of the spin asymmetry is within the statistical error $1/\sqrt{P_{He^+}^2 I}$ in Figure 3. Thus, there is no effect on the spin asymmetry from the ion beam irradiation on Bi.
During ISS measurements, the sputtering effect occures, and so that the surface is seriously damaged after the long time measurement. Consequently, compositional mixing occurs at the interface of an ultrathin film/substrate system. Actually, the compositional mixing is observed in the inset of Figure 3 which shows ISS spectra before and after the ion beam irradiation for 4 hours. We can see a small peak around 1050 eV which corresponds to elastic scattering energy from Si. This indicates that the epitaxially grown Bi film is partly removed from the surface by the sputtering, hence, intermixed Bi-Si phase appears on the surface. Thus, the surrounding environment of the Bi atom involved in the $He^+$ ion scattering should be substantially modified. This was confirmed by RHEED observation after the measurements. Figure 1(b) shows a RHEED pattern after such a long-time measurement which took over 20 hours. The RHEED spots became streaks, which reflect lack of long-range order i.e. broken crystal periodicity on surface.
The fact that the spin asymmetry is constant through the ion beam irradiation measurements indicates that spin asymmetry is neither affected by the surface structure nor the surface elemental composition. We always observed the same asymmetry of Bi within the statistical error in the identical scattering geometry.
Figure 4 shows azimuthal angle dependence of the spin asymmetry. The azimuthal angle is defined as the target rotation angle around the surface normal from the Bi[$1\bar{1}0$]. The distribution of the spin asymmetry is within the statistical error, and there is no systematic change of the spin asymmetry.
The result in Figure 4 together with that in Figure 3 indicates that the spin asymmetry arises from the $He^+$-Bi binary collision. In other words, there is no effect that the Bi target is a solid. This consideration is supported by the experimental observation that the spin asymmetry of Bi was independent of both the incident and the exit angles (not shown).
The neutralization of the projectile ion is sensitive to its trajectory because of neighboring atoms of the collision partners(*17*). Therefore, the fact that the spin asymmetry is independent of the surrounding environment of the target atom manifests that ion neutralization has no effect on the spin asymmetry on a Bi target.

As mentioned in the introductory section, the spin asymmetry on non-magnetic surfaces observed on Au and Pb is suggested to be due to SOC in our previous study. If we assume that the similar mechanism is responsible for the spin asymmetry on Bi targets, the spin asymmetry should be related to the scattering angle $\theta$ as

$$A \propto \frac{\cos(\theta/2)}{1 + \frac{1}{\sin(\theta/2)}} \tag{1}$$

Equation (1) is obtained with the assumption that the $He^+$ spin is at the He nucleus and the effect of SOC is limited to the smallest $He^+$-target distance, i.e., the classical turning point(*8*).

Figure 5 shows the scattering angle $\theta$, $\theta$' dependence of the spin asymmetry $A$ measured on Bi. In this measurement, the scattering plane was parallel to Bi[$1\bar{1}0$]. In Figure 5, the spin asymmetry vanishes at 0° and 180°, and it has a single maximum in between these angles. These features are consistent with Eq. (1).

In our previous study, we observed that the spin asymmetry increases with the incident energy on a Au(111) surface. This may be understood by the enhanced SOC with the increase of the collisional energy as the case for Mott scattering(*18*). The simple SOC model suggests that the spin asymmetry is proportional to the third power of the incident velocity(*8*).

To validate this point, we measured the spin asymmetry on Bi as a function of the incident energy of the $He^+$ ion beam (Figure 6). The incident energy was controlled by sample biasing. In Figure 6, the spin asymmetry monotonically increases with the incident energy. Thus, the behavior is similar to that observed on Au in our previous study(*8, 19*). Compared with the result on Au, Bi exhibits much larger dependence on the incident velocity; it is estimated to be almost 10 times larger than that of Au.

## 5. Conclusion

We observed spin dependent $He^+$ ion scattering on a non-magnetic Bi(001) ultrathin film. From the relationship between the spin dependent scattering and the scattering geometry (incident, exit, scattering, and azimuthal angles), the structure / elemental composition of the target surface, and projectile incident velocity, we conclude that the origin of the spin dependent scattering is spin-orbit coupling (SOC) in the $He^+$ ion-Bi atom binary collision. Thus, the mechanism is identical to those on Au and Pb as demonstrated in our previous study(*8*). We have observed SOC in ion scattering on various targets with a relatively heavy-mass element including Bi. Therefore, the scattered ions should be generally spin polarized as a consequence of SOC if the ion has electron spin. This immediately implies the application of SOC in ion scattering to an electron-spin polarized ion source.

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## 6. Figure captions

Figure 1
(color online) (a) A RHEED pattern of Bi(111) fresh surface. (b) A RHEED pattern of Bi(111) surface after 20 hours of beam irradiation. The incident electron beam is parallel to Bi[11$\bar{2}$].

Figure 2
(color online) (a) Scattering geometry. The scattering plane was perpendicular to both the Bi surface and the magnetic field $B$. (b) SP-ISS spectra of the Bi(111) surface. The solid red and dashed blue curves are for up and down spins, respectively. The substracted spectrum of $I_{up} - I_{dn}$ shows a peak at $He^+$-Bi elastic scattering energy around 1520eV.

Figure 3
(color online) Spin asymmetry $A$ as a function of the 1.55 keV $He^+$ ion beam irradiation time. (Inset) ISS spectra before and after the ion beam irradiation for 4 hours. The elastic scattering peak from the Si substrate appears at around 1050 eV.

Figure 4
(color online) Spin asymmetry $A$ as a function of the azimuthal angle. The azimuthal angle is defined as the target rotational angle around the surface normal from Bi[1$\bar{1}$0].

Figure 5
(color online) Spin asymmetry $A$ as a function of the scattering angle $\theta$ and $\theta$ ', where $\theta$ and $\theta$' denote the left and right scattering as displayed in the inset.

Figure 6
(color online) Spin asymmetry $A$ for several kinetic energies. The kinetic energy was varied by sample biasing. Kinetic energy at zero bias (1550 eV) is determined by ISS elastic scattering peak position and the others are determined by the bias voltage applied to sample.

(a) (b)

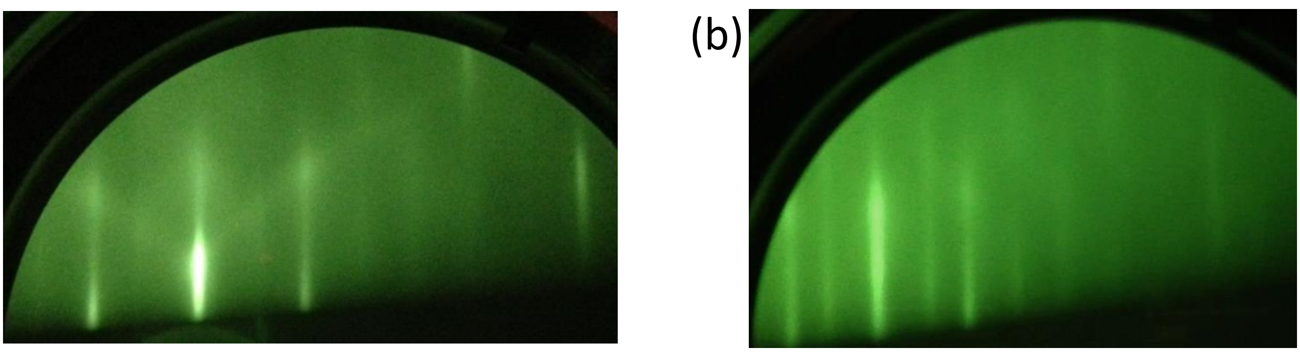



Figure 1. S.Ichinokura et al.

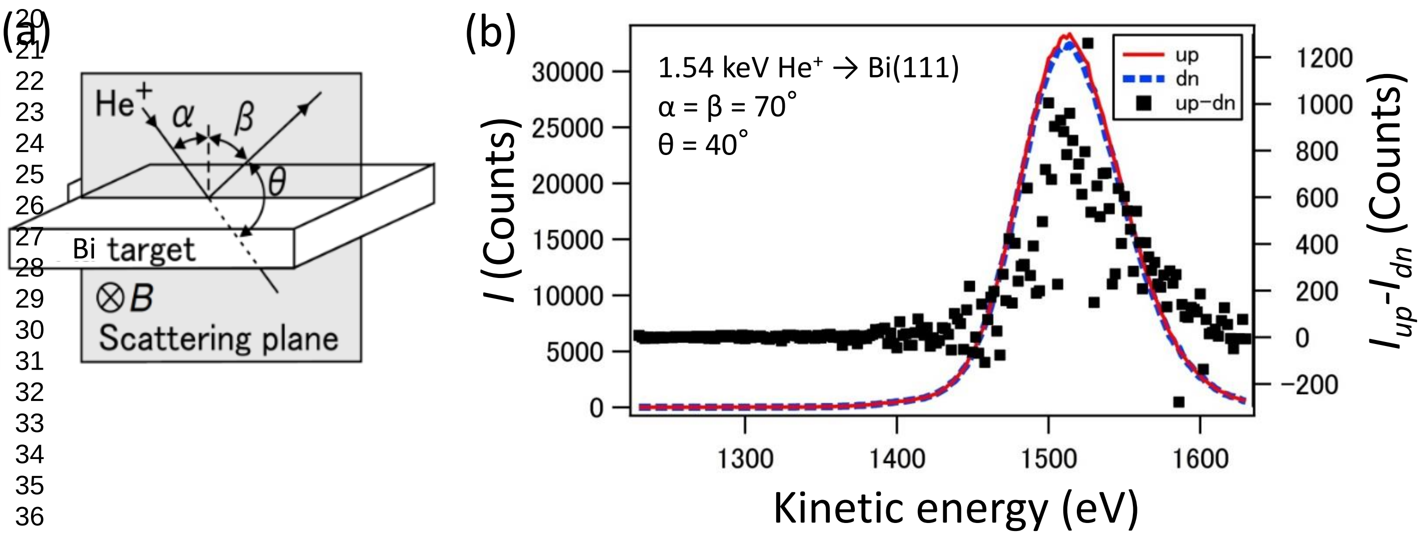


Figure 2. S.Ichinokura et al.

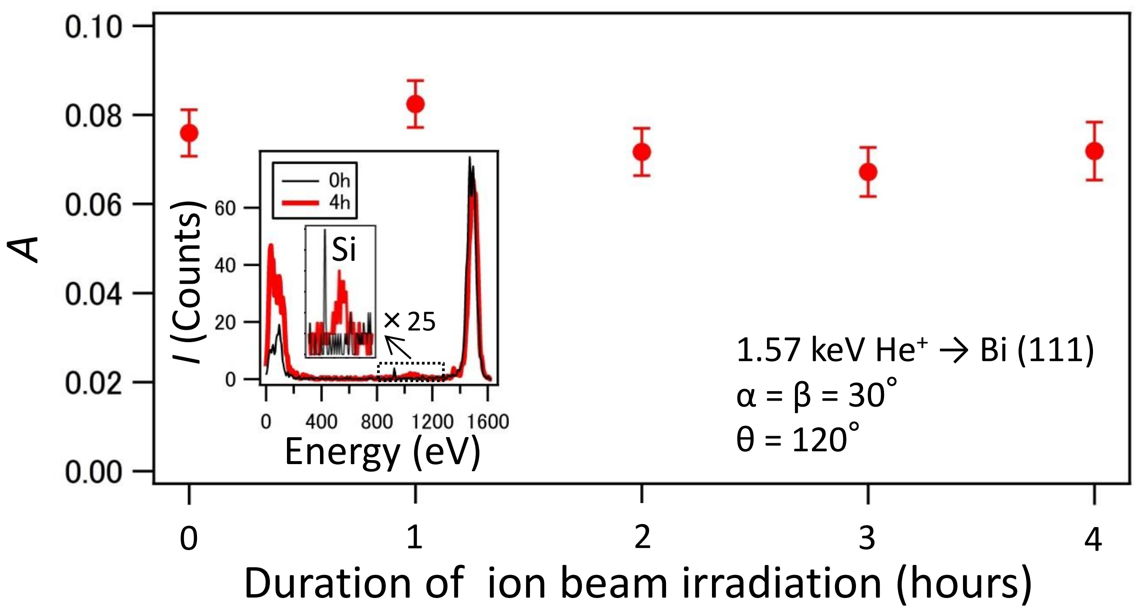




Figure 3. S.Ichinokura et al.

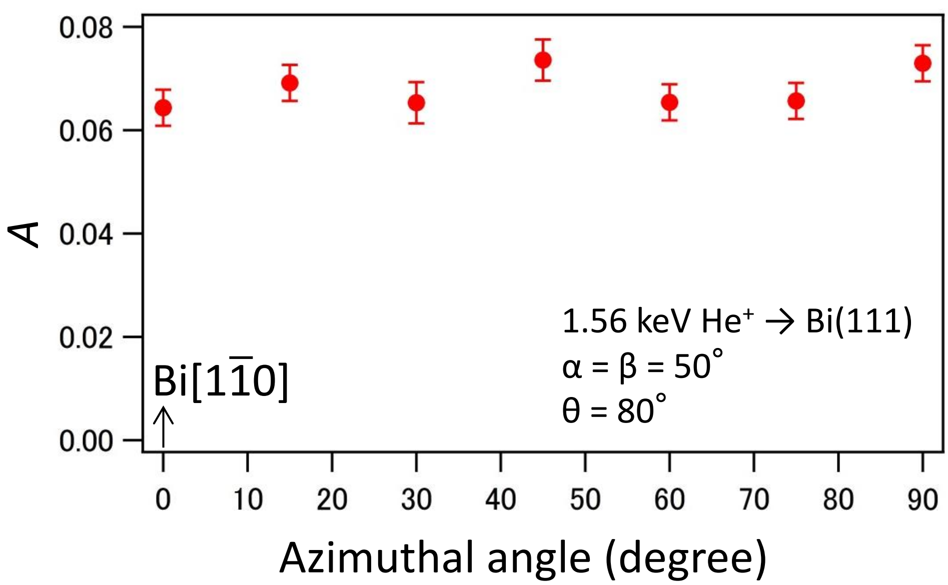




Figure 4. S.Ichinokura et al.

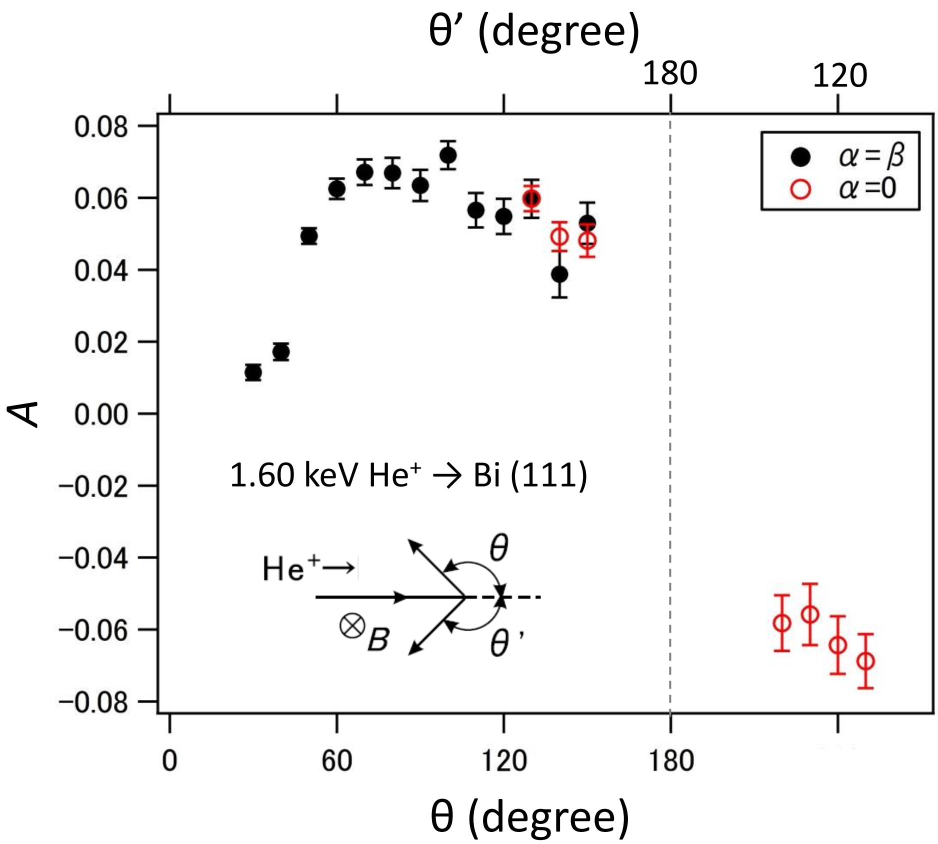




Figure 5. S.Ichinokura et al.

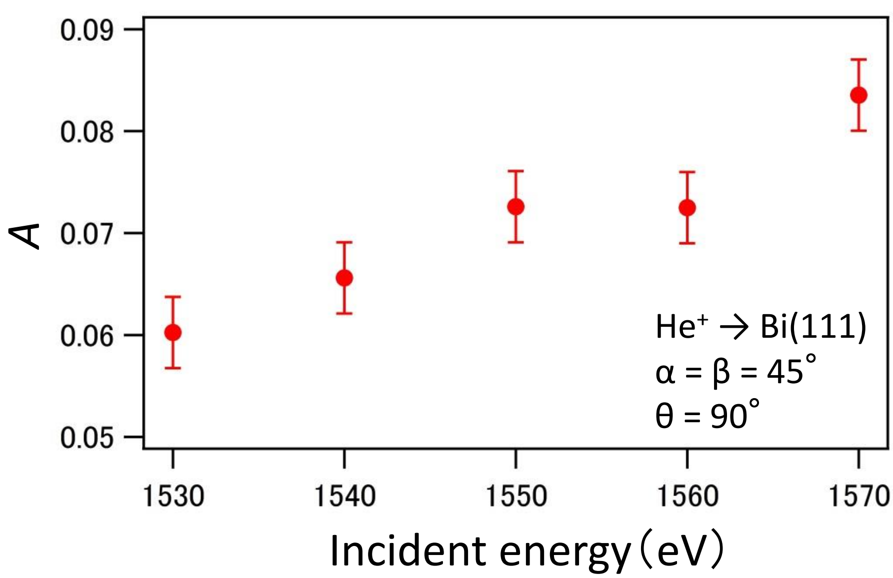




Figure 6. S.Ichinokura et al.