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Target element dependent spin–orbit coupling in polarized $^4He^+$ ion scattering

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ABSTRACT

We studied low-energy (1.57 keV) electron-spin polarized $^4He^+$ ion scattering on various 5*d* transition metal targets. The scattered ion intensity generally differed between the incident He^+ ions with up and down spins. This spin dependent ion scattering is attributed to the spin–orbit coupling (SOC) that acts transiently on the He^+ 1*s* electron spin in the He^+-target binary collision. We observed that the amplitude of the spin dependence in ion scattering, i.e., the spin asymmetry, differs between 5*d* transition metal targets. This target element dependence of the spin asymmetry is discussed in terms of re-ionization of He^0, which originates from the neutralization of the He^+ ion during the He^+-target collision. Since the re-ionization is spin independent process, it degrades the effective spin polarization of the He^+ ion beam. This explains smaller spin asymmetry with the target on which He^0 is re-ionized with higher rate.

1. Introduction

Low-energy ion beams, typically He^+ ion beams of a few keV, have been widely utilized in solid surface analysis [1,2]. This includes ion scattering spectroscopy (ISS), secondary ion mass spectroscopy, and elastic recoil detection analysis [3]. In these analytical methods, the knowledge of the ion-surface interaction is essential for interpreting the data. For example, the scattered ion intensity *I*, which is a measured quantity in ISS, depends on the scattering cross section σ given by the scattering potential and the survival probability of the projectile ions from the neutralization P_S. It is noted that the re-ionization also contributes to the scattered ion intensity, in which ions are once neutralized and then re-ionized along their trajectories. Expressing explicitly the re-ionization with the probability $P_R, I \propto \sigma[P_S + (1 - P_S)P_R]$. Thus, the scattered ion intensity depends on both the neutralization and the re-ionization.

We hereafter discuss the relationship between He^+ ISS and an electron-spin. In He^+ ISS, the scattering potential has been traditionally expressed by the screened Coulomb function, which is an electron-spin independent central force potential [2]. Thus, the scattering cross section σ should be independent of the spin. The re-ionization of He^0 is also spin independent process. On the other hand, the survival probability P_S is primarily dependent on the transition rate of a surface electron into a He^+ 1*s* hole (Auger neutralization (AN)) [4]. Due to the Pauli exclusion principle, the AN rate depends on electron spin because the spin of a surface electron filling the He^+ 1*s* hole should be opposite to that of the He^+ 1*s* electron [5]. Actually, it is the principle of surface magnetism analysis by spin-polarized He^+ ion scattering spectroscopy (SP-ISS) [6]. In SP-ISS, the scattered He^+ ion intensity is compared between the incident He^+ ions with spins which are parallel and anti-parallel to the magnetization of the target. Due to the electron-spin polarization of the target which is responsible for magnetism, the scattered intensity differs between He^+ ions with parallel and anti-parallel spins. The scattered He^+ ions originate from the outermost surface of the target, because the ion penetrating into the subsurface are neutralized with high probability [2]. Therefore, SP-ISS is adequate for the analysis of surface magnetism.

In the conventional framework of low-energy ISS mentioned above, the scattered ion intensity should be independent of electron spin on non-magnetic surfaces since the AN rate is equal between spins. However, we recently observed spin-dependent He^+ ion scattering on non-magnetic targets, such as Au [7]. The observation was qualitatively interpreted in terms of the spin–orbit coupling (SOC) that acts transiently on the He^+ 1*s* electron spin in the He^+-target binary collision. 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

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collision. Thus, it is the effect of a Mott-like scattering, however, for He^+ ions rather than for electrons [8]. It was the first observation for electron-spin induced SOC in ISS. We believe that it is an important observation because it suggests that the non-central force potential arising from SOC should be taken into account in addition to the central force potential typically described by the screened Coulomb function to analyze the electron-spin effect in ion scattering even in low-energy range.

In the present study, we systematically investigated the target element dependence of SOC on non-magnetic 5*d* transition metal targets. We found that the spin dependent scattering remarkably differs between target elements. It is briefly discussed in terms of the re-ionization effect in SOC.

2. Experimental method and setup

We performed experiments in an ultrahigh vacuum (UHV) chamber (base pressure of 5×10^{-11} Torr) equipped for SP-ISS. Electron-spin-polarized $^4He^+$ ions were generated by Penning ionization of spin-polarized metastable He 2^3S_1 atoms [9]. We employed an optical pumping technique to polarize metastable He 2^3S_1 atoms. 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.

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 gravity direction. 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. 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 as shown in Fig. 1.

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.

In the present SP-ISS experiment, we used targets which consist of a form of the pure elements Hf, Ta, Re, Ir, Pt, and Au. We cleaned the target surface by repeated cycles of annealing and sputtering in UHV. After the surface cleaning procedure, we confirmed that the surface contamination was successfully removed in the preliminary ISS measurement (Fig. 2).

3. Results and discussion

Fig. 2 shows typical ISS spectra after the surface cleaning of the target. All spectra exhibit the surface peak near the He^+-target

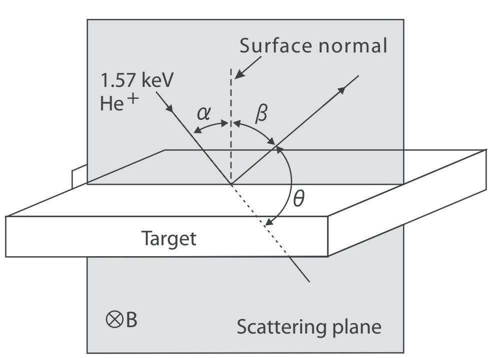

Fig. 1. Scattering geometry of the present study. The scattering plane is perpendicular to both the target surface and the magnetic field **B**.

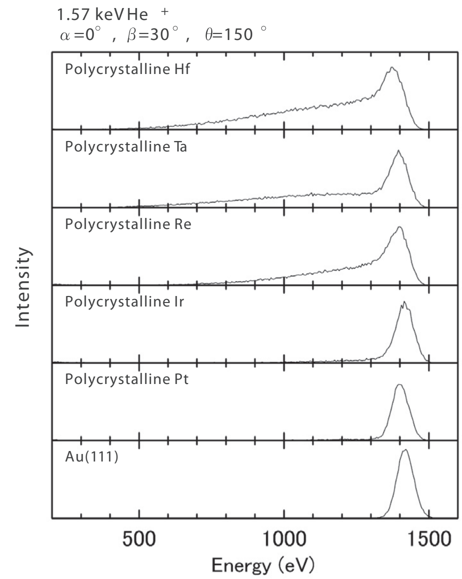

Fig. 2. ISS spectra on Hf, Ta, Re, Ir, Pt, and Au targets. The incident energy was 1.57 keV. The incident angle α, the exit angle β, and the scattering angle θ were 0°, 30°, and 150°, respectively.

binary collision energies. This shows that the surface cleaning of the target was successful. The shape of the peak is substantially different among the targets; the peak is symmetric in the energy scale for Pt and Au, while a pronounced peak tail appears in the low-energy side on Hf, Ta, and Re. The peak tail on Hf, Ta, and Re is due to stopping power accompanied with the neutralization followed by re-ionization ($He^+ + e^- \rightarrow He^0 \rightarrow He^+ + e^-$). The He^+ ions penetrating into the subsurface of the target lose their kinetic energy along their trajectory due to kinetic stopping and electronic stopping. The He^+ ions are neutralized with high probability along the penetrating trajectory. Thus, most of He^+ ions are neutralized at the subsurface. Subsequent backscattering followed by re-ionization is responsible for the peak tail on Hf, Ta, and Re. Since the peak tail appears even after the additional surface cleaning procedure, we interpret that it is not the consequence of the contamination on the target surface.

The re-ionization mechanism is electron promotion mediated by a collisional quasimolecule. It has been observed that the re-ionization probability is minimized for target elements with filled *d* orbitals located in a shallower energy position than the He 1*s* level. This indicates that re-ionization probability is in the following order on the 5*d* transition metal targets; Hf > Ta > Re > Ir > Pt > Au [10]. This target element dependent re-ionization of He^+ ions is consistent with the theoretical study [11]. The target element dependence of the re-ionization explains the symmetric peak shape on Pt, Au, and Ag and the peak tail at the low energy side on Hf, Ta, and Re in Fig. 2.

In the present study, we discuss the spin dependent He^+ ion scattering in terms of the spin asymmetry *A*, which is defined as $(I_\uparrow - I_\downarrow)/[P_{He^+}(I_\uparrow + I_\downarrow)]$, where $I_\uparrow$ and $I_\downarrow$ are the scattered intensities with the incident He^+ ions whose magnetic moments are parallel and antiparallel to the guiding magnetic field, respectively.

The ISS spectrum and the spin asymmetry on the Au target are shown in Fig. 3. The scattering angle was 150°. The spin asymmetry of about −9% appears at the Au peak, while no spin asymmetry is observed for the secondary ions observed at around 30 eV (Fig. 3(a)). Moreover, no substantial difference in the spin asymmetry is detected between the single and the polycrystalline Au surface. These experimental facts are consistent with the previously-mentioned mechanism of SOC.

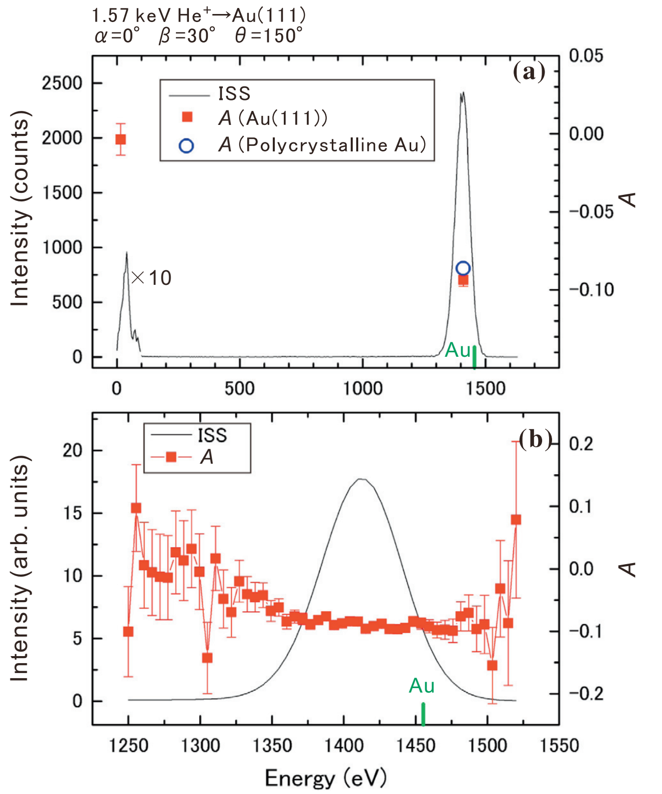

Fig. 3. ISS spectra (solid black curve) on the Au (111) surface, and the spin asymmetry on the Au (111) surface (red filled squares) and the polycrystalline Au surface (a blue open circle). The measurements were separately made for (a) a wide energy range and (b) a narrow energy range. The incident energy was 1.57 keV. The incident angle α, the exit angle β, and the scattering angle θ were 0°, 30°, and 150°, respectively. The He^+-Au binary collision energy is indicated at the bottom of the spectra. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

It is noted that the Au peak of ISS shifts to lower energy side by about 40 eV compared with the binary collision energy in Fig. 3(b). On the other hand, the energy for the maximum of the spin asymmetry agrees with the binary collision energy. Thus, the maximum of the spin asymmetry is located at higher energy than the ISS peak. This is explained as follows; in ISS, the He^+ ions lose their kinetic energy through the inelastic process which includes excitation in the target atoms. Thus, the surface peak of ISS generally appears at lower energy than the energy expected from the classical binary collision. The He^+ ions which survive the neutralization during the multiple scattering also reduce the peak energy. Actually, we observed that the Au peak position depends on the incident angle with the fixed scattering angle (not shown), suggesting that the contribution of the multiple scattering is not negligible. On the other hand, the multiple scattering may reduce the spin asymmetry, because the spin asymmetry is strongly dependent on the scattering angle θ as shown in Fig. 4. The absolute value of the spin asymmetry is the largest for θ of 150° on the Au target. Thus, if we observe the spin asymmetry on Au with θ of 150°, which is the condition of the result in Fig. 3, the spin asymmetry is the largest at the energy for the single scattering, that is the binary collision energy. In addition to the effect of the above-mentioned mechanism, spin relaxation of the He^+ ion in the inelastic collision may be also responsible for the peak energy difference between ISS and the spin asymmetry. The detailed mechanism of the spin relaxation in the inelastic collision is, however, not clear in the present time.

The spin asymmetry is independent of both the incident angle α and the exit angle β (not shown), while it is strongly dependent on

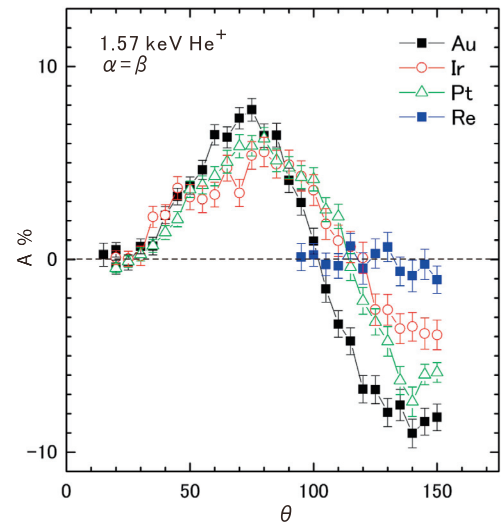

Fig. 4. Spin asymmetry A as a function of the scattering angle θ on 5d-transition metals (Au, Ir, Pt, and Re). A was measured for the He^+-target binary collision energy. The incident energy was 1.57 keV. The incident angle α was equal to the exit angle β, hence, $\alpha = \beta = (180^\circ - \theta)/2$.

the scattering angle θ as shown in Fig. 4. Actually, the distribution of the spin asymmetry was within the statistical error $1/\sqrt{P^2_{He^+} \cdot I}$ in both the α and β scan measurements. Furthermore, the spin asymmetry was also independent of the sample rotation angle around the surface normal, which is often called azimuthal angle. The detailed data on the relationship between the spin asymmetry and the scattering geometry is published elsewhere [12].

The above-mentioned angle dependence of the spin asymmetry manifests that the spin dependent He^+ ion scattering observed in Figs. 3 and 4 originates from the He^+-target binary collision. In other words, there is no effect that the target is a solid. Therefore, the spin asymmetry with similar angle dependence should be observed in gas-phase SP-ISS experiments.

Since the spin asymmetry arises from the He^+-target binary collision, either the scattering cross section σ or the survival probability of the projectile ion from neutralization P_S should be the origin of the spin asymmetry. The survival probability of the incident He^+ ion is sensitive to its trajectory because of neighboring atoms of the collision partners [13]. Thus, the fact that the spin asymmetry originates from the He^+-target atom binary collision, as derived from the angle dependence in Fig. 4, shows that ion neutralization has no effect on the spin asymmetry. Furthermore, we observed no change in the spin asymmetry of Bi in a Bi-Si composite system with the stoichiometric variation [12]. If the spin asymmetry is due to the survival probability, it should be affected by the stoichiometry at the surface, because the electronic state around the target atom is modified according to the composition. Therefore, the spin asymmetry should be due to the spin dependent scattering cross section.

The effect of SOC in the scattering cross section is intuitively interpreted as the effect on the projectile spin **S** of the magnetic field **H** induced by the projectile angular motion around the target nucleus during the projectile-target binary collision. The target nucleus can be considered to rotate around the projectile in the binary collision (Biot–Savart law); the SOC potential U_{SOC} in the collision between a projectile of mass M_1 and a target of atomic number Z_2 has the following form:

$$U_{SOC} = \mathbf{H} \cdot \mathbf{S} \propto (Z_2/|\mathbf{r}|^3)(\mathbf{r} \times M_1\mathbf{v}) \cdot \mathbf{S}, \quad (1)$$

where **v** is the velocity of the projectile and **r** is the position of the target nucleus as seen from the projectile. Eq. (1) shows that the effect of SOC should vanish when **r** is parallel or anti-parallel to **v**. This is the case for the scattering angle of 180°. Moreover, from the **r** dependence of SOC, it is obvious that the effect of SOC should also vanish at 0° by taking the relationship between the scattering angle and the impact parameter. These features are actually observed in Fig. 4. Moreover, Eq. (1) also indicates that the signs of the spin asymmetry for scattering to the left and right should be opposite as seen from the incident beams. This feature has been already confirmed in our previous study [7].

There is an order in the absolute value of the spin asymmetry on 5*d*-transition metals as Au > Ir, Pt > Re ∼ 0 in Fig. 4. Moreover, we observed no substantial spin asymmetry on Hf and Ta regardless of the scattering angle as Re (not shown). The target element dependent spin asymmetry on 5*d*-transition metals is explained by the re-ionization effect on the effective beam polarization as described below.

The number of the He^+ ions which survive the neutralization with the probability P_N is written as $n_0(1-P_N)$, where n_0 is the number of the He^+ ion in the primary beam. On the other hand, the number of He^+ ions which originate from re-ionization with the probability P_R is written as $n_0 P_N P_R$. Because re-ionization is spin independent process, the number of He^+ ions originating from re-ionization should be equally divided between up and down spins. Thus, half of the re-ionized He^+ ion has the same spin with that of the primary ion. If the number of the primary He^+ ion with up (down) spin is $n_0^{\uparrow}(n_0^{\downarrow})$, the number of the scattered He^+ ions with up spin $n_1^{\uparrow}$ which experienced either the survival from the neutralization or a neutralization followed by a re-ionization is expressed as

$$n_1^{\uparrow} = n_0^{\uparrow}(1-P_N+0.5P_NP_R)+0.5n_0^{\downarrow}P_NP_R. \quad (2)$$

In the same manner,

$$n_1^{\downarrow} = n_0^{\downarrow}(1-P_N+0.5P_NP_R)+0.5n_0^{\uparrow}P_NP_R. \quad (3)$$

Thus, considering the neutralization and the re-ionization, both of which occur in prior to SOC along the He^+ trajectory, the He^+ beam polarization which effectively acts on SOC (the effective He^+ ion beam spin polarization P'_{He^+}) is expressed as

$$P'_{He^+} = P_{He^+}\frac{1-P_N}{1-P_N+P_RP_N}. \quad (4)$$

Eq. (4) shows that the spin asymmetry is affected by the factor $P'_{He^+}/P_{He}[=(1-P_N)/(1-P_N+P_RP_N)]$ if re-ionized He^+ ions are involved in the appearance of the spin asymmetry. Thus, smaller spin asymmetry should appear with the target on which larger P_R is expected. Actually, the target element dependence of re-ionization probability reported for He^+ ISS is consistent with the above-mentioned order of the spin asymmetry on 5*d* transition metals shown in Fig. 4 [10]. The He^+ ion loses the kinetic energy of about 20 eV due to the electron promotion in the re-ionization, but it is comparable to the energy width of the incident He^+ ion beam (∼25 eV) [9]. Thus, the effect of the energy loss due to the re-ionization was not observed in the spin asymmetry.

4. Conclusion

We studied SP-ISS on non-magnetic 5*d* transition metal targets. The spin dependent ion scattering is attributed to spin–orbit coupling (SOC) in the He^+ ion – target atom binary collision, which is intuitively understood as the effect of the transient angular motion of the He^+ 1*s* electron around the target nucleus during the collision. The spin asymmetry is strongly dependent on the target elements. It is interpreted in terms of re-ionization of He^0, which originates from the neutralization of the He^+ ion along its trajectory. Because re-ionization is spin independent process, it reduces the effective beam polarization. Thus, larger spin asymmetry appears on the transition metal target on which the re-ionization takes place with smaller rate. The present study indicates that He^+ ions are generally spin polarized after scattering even from non-magnetic surfaces. It is also indicated that the non-central force potential arising from SOC should be considered in addition to the central force potential typically described by the screened Coulomb function to analyze the electron-spin effect in ion scattering even in the low-energy range.

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