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## Improvement of mapping quality by reflection of a laser beam in Resonance-SNMS

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Resonant ionization laser sputtered neutral mass spectrometry (R-SNMS) is a method of surface analysis. Since the sputtered neutral particles emitted having certain angular distribution, the wavelength (Doppler) shift will be occurred. Therefore, angularly spread particles cannot be ionized by the resonant laser due to the Doppler shift. In this study, we improved the mapping quality of R-SNMS and coped the Doppler shift by using a reflected laser beam.

### 1. Introduction

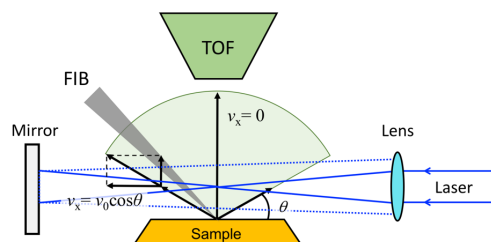
Mass spectrometry is an effective way to identify elements including isotopes. For example, the focused ion beam time-of-flight secondary ion mass spectrometry (FIB-TOF-SIMS) can draw the element distribution with a high lateral resolution [1]. On the other hand, element can be selected by ionization of sputtered resonance neutral particles using resonant laser sputtered neutral mass spectrometry (R-SNMS) [2]. It can effectively distinguish between the isobaric interfered elements. However, as compared with a flat sample, the particles with a bumpy surface are more difficult to analyze, because the distribution of sputtering space depends on the surface shape of the

sample. Therefore, in R-SNMS, some sputtered particles have a velocity component of the laser wave number vector. Hence, the resonance wavelength shifts due to the Doppler effect. Here, we propose a method for coping the Doppler effect in the mapping of the R-SNMS.

### 2. Principle and Method

As shown in Fig. 1, sputtered atoms from the sample are emitted after being irradiated by the FIB. It is assumed that the initial velocity of the sputtered neutral particles is  $v_0$ . Its horizontal velocity component should be  $v_0 \cos \theta$ . Therefore, resonant ionization space is limited to a small range if the spectral width of the laser is narrow. Here, we use a flat mirror to reflect the laser beam that passed through the sputter space. Thus, the resonant ionization space will become wider. In turn, the ionization efficiency will be increased.

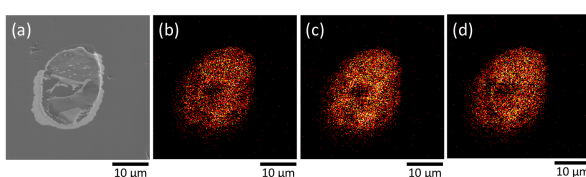
Here, we selected energy level between  $5s^25p$  (ground state) and  $5s^26s$  as a resonance absorption scheme for indium (In) sample. The sample was In powder on a Si substrate. Here, a wavelength tunable Ti:Sa laser developed by the authors was used [3]. The



**Fig. 1** Schematic of the R-SNMS setup. The solid line is the incident light of the laser beam, and the dotted line is the reflected light (when a flat mirror is used).

fundamental wavelength was fixed at 820.56 nm and was converted by a BBO crystal to a wavelength of 410.28 nm to match the resonance absorption scheme of In. The energy of the laser pulse was 3 mJ/pulse. The pulse width was 30 ns, and the wavelength line width was 3 pm. The laser was incorporated into the lab-made FIB-TOF-SIMS equipment. We mapped the same sample in case of the laser beam passes over sample for one time ("one way") and two times ("round trip"), respectively. The following are called "one way" and "round trip".

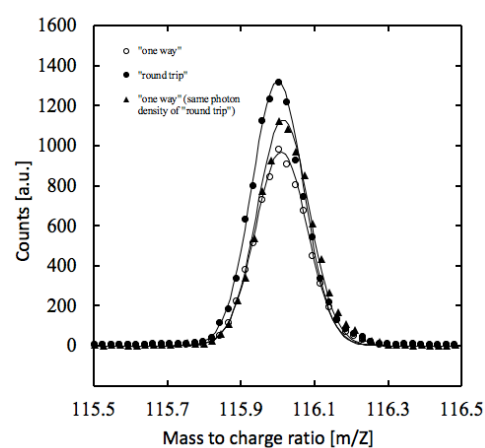
### 3. Result and Discussion



**Fig. 2** FIB-induced secondary electron image and R-SNMS mapping information of an indium particle. (a) Secondary electron image. (b) R-SNMS  $^{115}\text{In}^+$  image that laser beam passes over sample for one time ("one way"). (c) R-SNMS  $^{115}\text{In}^+$  that laser beam passes over sample for two times ("round trip"). (d) R-SNMS  $^{115}\text{In}^+$  image that laser beam passes over sample for one time with the same photon density of (c).

Figure 2 shows the R-SNMS mapping result of the information of an indium particle. The R-SNMS  $^{115}\text{In}^+$  mapping quality of "round trip" (Fig. 2(c)) has been improved than "one way" (Fig. 2(b)). Next, we set the "one way" photon density in the scattered space to be the same as "round trip". The R-SNMS  $^{115}\text{In}^+$  mapping quality of "round trip" (Fig. 2(c)) has been improved than "one way" (Fig. 2(d)) at the same photon density. In Fig.2 (b) and (c), although the maximum count is the same, in Fig.2 (c), the number of maximum count pixels is more than Fig.2 (b). The image becomes clearer due to the change in contrast. Figure 3 shows the peak intensity of the resonant mass spectrum. The intensity of the peak is "round trip", "one way (same photon density of "round trip")", and "one way" in descending order. And they are corresponding to Fig. 2(b), (c), and (d), respectively. The count enhancement of the resonant mass spectrum is due to the combination of the increase in photon density and the Doppler shift effect coping. The difference between Fig. 2 (d) and (c) and the difference in peak intensity indicate that the Doppler broadening was coped. On the other hand, as

shown in Fig. 1, the laser beam was collected by a lens before incidence, the diameter of the reflected beam was larger than incidence. Hence, mass spectrum widening is due to the reflected beam becomes thick, resulting in a time difference in the vertical direction of the resonant ionization of neutral particles. In case of "one way", only neutral particles in the direction perpendicular to the sample will be ionized by resonance. In the case of "round trip", the resonant ionization space will become wider.



**Fig. 3** Resonant mass spectrum of indium.

### 4. Conclusion

The mapping quality of the R-SNMS mapping was improved by a single reflected laser beam. This improvement is due to the increase in the count number of the resonant ionization. The increase in the number of count benefits both from the increase in photon density caused by reflection and from the expansion of the ionization space caused by the Doppler shift coping of the reflected laser beam. We will use more irregular particles to study the coping of the Doppler shift by reflected laser beam in future plan.

### 5. Acknowledgements

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### 6. References

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