Interatomic Resonant X-ray Raman Spectroscopy

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X-ray absorption fine structure (XAFS) is a structure appearing at higher energies than the X-ray absorption edge. It provides bond distances and coordination numbers around the X-ray absorbing atoms. When it is applied to low Z element, low energy X-ray and high vacuum environment are necessary because of the large absorption of air. X-ray Raman scattering uses hard X-ray to get the XAFS measurement of the low Z elements. The X-ray Raman, nevertheless, its sensitivity is very low and it is difficult to distinguish species of the same element. If the Raman scattering were enhanced by tuning the energy of the incoming photon to the absorption edge of the central atom to which the low z element is bound to, the problems of X-ray Raman would be solved. Such energy transfers from central atom to surrounding atoms would be less probable. However, to our knowledge no experimental nor theoretical estimations have been made yet for such probability. We dared to measure the interatomic resonant Raman signal in TaN using X-rays tuned to the excitation of the Ta L3-edge and Raman emission of the N K-edge to qualitatively assess of such an enhancement occurs. In this preliminary work, we demonstrated the possibility of this new type of interatomic resonant X-ray Raman spectroscopy.

X-ray Raman measurements were carried out in BL36XU of SPring-8. The X-ray emitted from tapered undulator was monochromatized with Si(220) double crystals. The sample was 10 wt.% TaN diluted with SiO₂. The emitted X-rays were analyzed by 4 Johan-type Ge(660) monochromator located at 820 mm from the sample. Incident X-ray were detected by an ion chamber and emitted X-ray from a sample were detected by two-dimensional pixel array detector. The obtained data were processed and analyzed by ImageJ.

The TaN sample was irradiated by X-ray energy at 9865 and 9886 eV. The emission intensities excited at 9886 eV were clearly changed at the N K-edge position in the same accumulation time as excited at 9865 eV. We found no peak in the Ta foil emission spectra in the same region excited by 9886 eV. This shows the possibility of enhancement effect on the N Raman signal by the neighboring Ta L_3 absorption.

In conclusion, we found a clear enhancement of the emission spectrum by interatomic resonance Raman. We coined phenomenon as Multi atom resonance X-ray Raman(MARX-Raman) which may lead to a new *in situ* and *bond-specific* XAFS spectroscopy for low Z elements.