日本化学会 第103春季年会 (2023)

Experimental and Theoretical Development of Atomic Momentum Spectroscopy for Polyatomic Molecules

([1]*IMRAM, Tohoku University*, [2]*Grad. Sch. Sci., Tohoku University*) ○Satoru Kanaya,[1] Yuichi Tachibana,[1] Yuuki Onitsuka,[1] Noboru Watanabe,[1] Hirohiko Kono[2], Masahiko Takahashi[1]

Keywords: Electron Compton Scattering, Intramolecular Atomic Motion, Momentum Imaging Technique, Theoretical Quantum Chemistry

Intramolecular motions of the constituent atoms govern the functionality of a molecule and they have long been attracting great interest in a broad range of research fields. Such atomic motions have been studied mainly by laser vibrational spectroscopy, which measures frequencies of normal modes. Nevertheless, one may desire to have a technique to map the intramolecular motion of a specific single atom, because the origin of molecular functionalities is often motion of one or a few key atoms and not normal modes or the collective motion of many (or all) of the constituent atoms. This is the material reason why we have recently made a series of attempts towards development of electron-atom Compton scattering as atomic momentum spectroscopy (AMS).[1-3] We have already demonstrated for diatomic molecules that AMS enables one to observe momentum distributions of each atom, with different masses, due to intramolecular motion. In this study, we have aimed to extend the reach of AMS to polyatomic molecules both experimentally and theoretically.

For the purpose mentioned above, we have chosen CH_4 as the target molecule. Experimentally, AMS experiments were conducted at a scattering angle of 135° and at an incident electron energy of 2.0 keV, by using our multi-channel AMS apparatus.[1] As an example of experimental results, we present in Figure 1 an electron energy loss spectrum of CH_4. It is evident that two bands appear at around 0.3 and 3.7 eV, respectively. The former band is assigned to the C atom and the latter is to the H atoms. The difference in the width between the two bands is due to the difference in the Doppler broadening between the C and H atoms. Theoretically, we have developed a quantum chemistry-based theory to predict momentum distributions due to intramolecular atomic motions in polyatomic molecules. In the contribution, we will make a rigorous comparison between the experimental and theoretical atomic momentum distributions, in order to demonstrate that AMS has the ability not only to map the intramolecular motion of each atom with different masses but also to perform elemental composition analysis of a polyatomic molecule.

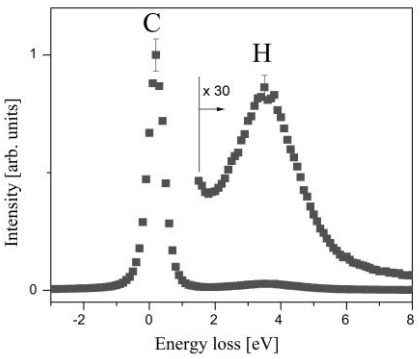

Fig. 1 An electron energy loss spectrum of CH_4.

1) M. Yamazaki, M. Hosono, Y. Tang, M. Takahashi, *Rev. Sci. Instrum.* **2017**, *88*, 063103. 2) Y. Tachibana, Y. Onitsuka, H. Kono, M. Takahashi, *Phys. Rev. A*. **2022**, *105*, 052813. 3) Y. Onitsuka, Y. Tachibana, M. Takahashi, *Phys. Chem. Chem. Phys.* **2022**, *24*, 19716.