理学部セミナー

題目:
Nuclear Resonant Vibrational Spectroscopy for Observation of Extremely Weak Fe-H/D Features in Hydrogenases and Nitrogenases

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日時：平成29年6月14日(水) 16:00-17:00
場所：兵庫県立大学 播磨理学キャンパス
研究棟 739談話室
※Language: English

Wang氏は、放射光を用いた核共鳴非弾性散乱分光法を生体分子試料に適用している研究者です。その手法の解説と、最近の研究成果についてお話をしていただけます。手法にご興味のある方も歓迎です。ふるってご参加ください。

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Abstract

Hydrogenases (H₂ases) catalyze the reversible reaction of $2H^+ + 2e^- \rightarrow H_2$, while nitrogenases (N₂ases) catalyze the fixation of molecular nitrogen (N₂) in the atmosphere into bio-available NH₃. Since today’s world faces multiple pressures from the demands for sustainable energy and food resources, H₂ases and N₂ases have both attracted a lot of attention and have been intensively studied for decades. Although crystal structures are available for all of these enzymes, many key enzyme intermediates cannot be crystallized. We are therefore using spectroscopy as an alternative probe of these key intermediates.

Nuclear resonant vibrational spectroscopy (NRVS) measures vibrational transitions that occur together with nuclear transitions that are typically associated with the Mossbauer effect. For the study of Fe in biology, $^{57}$Fe NRVS has key features that complement traditional techniques such as infrared (IR) and Raman spectroscopies. For example, despite the complexity of these samples, $^{57}$Fe NRVS only sees normal modes that involve motion of the $^{57}$Fe nucleus.

Using $^{57}$Fe nuclear resonant vibrational spectroscopy (NRVS), we have characterized several important $^{57}$Fe-labeled proteins such as H₂ase and N₂ase. Following the successful observation of the Ni-H-Fe wag mode in Desulfovibrio vulgaris Miyazaki F [NiFe] H₂ase (DvMF for abbreviation) and the full Fe-H/D feature for several model complexes, we extended these studies to other enzymes, such as Chlamydomonas reinhardtii [FeFe] H₂ase (Cr-HydA1) and Desulfovibrio desulfuricans [FeFe] H₂ase (Dd-HydAB). Fe-hydride and Fe-deuteride vibrational modes in [FeFe] H₂ases were observed and interpreted by DFT calculations. With the advancement we have made for studying H₂ase, we have also better characterized the catalytic intermediates in N₂ase, such as the E₄ state.