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Center for Novel Material Science under Multi-Extreme Conditions
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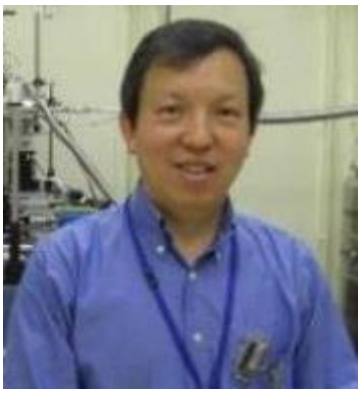
題目：

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



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Abstract

Hydrogenases (H_2ases) catalyze the reversible reaction of $2H^+ + 2e^- \rightarrow H_2$, while nitrogenases ($N_2$ases) catalyze the fixation of molecular nitrogen (N_2) in the atmosphere into bio-available NH_3. Since today's world faces multiple pressures from the demands for sustainable energy and food resources, H_2ases and N_2ases 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 H_2ase and N_2ase. Following the successful observation of the Ni-H-Fe wag mode in *Desulfovibrio vulgaris* Miyazaki F [NiFe] H_2ase (*Dv*MF 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] H_2ase (*Cr-HydA1)* and *Desulfovibrio desulfuricans* [FeFe] H_2ase (*Dd-HydAB*). Fe-hydride and Fe-deuteride vibrational modes in [FeFe] H_2ases were observed and interpreted by DFT calculations. With the advancement we have made for studying H_2ase, we have also better characterized the catalytic intermediates in N_2ase, such as the E_4 state.