第310回触媒化学研究センターコロキウム

共催　日本化学会北海道支部・応用物理学会北海道支部・触媒学会北海道支部・北大加速器科学研究会

Dynamic structure of supported Pt and Pt-Sn nanoclusters: Real-time, finite-temperature DFT/MD and X-ray Spectroscopy simulations

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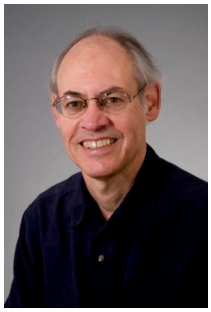

2012年7月9日（月）16：00―17：30

（創成科学研究棟4階セミナー室B・C）

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The nature of local atomic and electronic structure at the nanoscale is of both fundamental and technological importance. These local and generally dynamical properties are especially important for understanding the role of supported metalclusters, which are key ingredients in many catalytic processes. Understanding these properties thus requires real-time approaches, which are becoming increasingly important in studies based on x-ray absorption spectra (XAS). In this talk I will discuss a real-time approach for calculating dynamic structure based on ab initio, finite-temperature density functional theory/molecular dynamics and the real-space Green's function approach for XAS calculations based on the FEFF9 code [1]. This approach exploits recent theoretical advances that permit parameter-free calculations of the key many-body effects in the theory. This method is illustrated for nano-clusters of Pt [2] and Pt-Sn [3] supported on gamma-alumina. The simulations reveal a complex, non-equilibrium structure on multiple-time scales, including a hindered Brownian motion of the center of mass and fluctuating bonding. In particular their internal structure varies dynamically on a time scale of a few ps. Charge transfer between atoms in the cluster and between the cluster and the surface are particularly important. These results explain many unusual properties observed for supported Pt nanoclusters including large temperature dependence, anomalous disorder, and negative thermal expansion. For Pt-Sn, the simulations show that the Sn atoms are especially mobile, but the clusters still have well defined Pt-Pt and Pt-Sn coordination shells, and between 2 - 5 bonds between Pt or Sn atoms in the cluster and the O atoms at the surface. Simulations of the XANES are observed to be in reasonable agreement with experiment demonstrating the importance of taking into account the large, internal fluctuations in these structures. Implications for bonding and catalytic properties are also briefly discussed.

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[1] J. J. Rehr, J. J. Kas, M.P. Prange, A.P. Sorini, Y. Takimoto, and F. Vila, Comptes Rendus Physique, 10, 548 (2009).
[2] F. Vila, J. J. Rehr, J. Kas, R.G. Nuzzo and A.I Frenkel., Phys. Rev. B 78, 121404(R), (2008).
[3] F. Vila, J. J. Rehr, S. D. Kelly and S. R. Bare, Bull. Am. Phys. Soc. (2011).

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J.J.Rehr 先生は，理論 XAFS 解析手法として，広く使われる FEF を 1980 年代後半から開発され，XAFS 理論を先導されています．この 10 年近く触媒やナノ粒子の XAFS の理論解析に興味を持たれています．せっかくの機会ですので，多くの方の参加を期待しております．

310th Catalysis Research Center (CRC) Colloquium

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