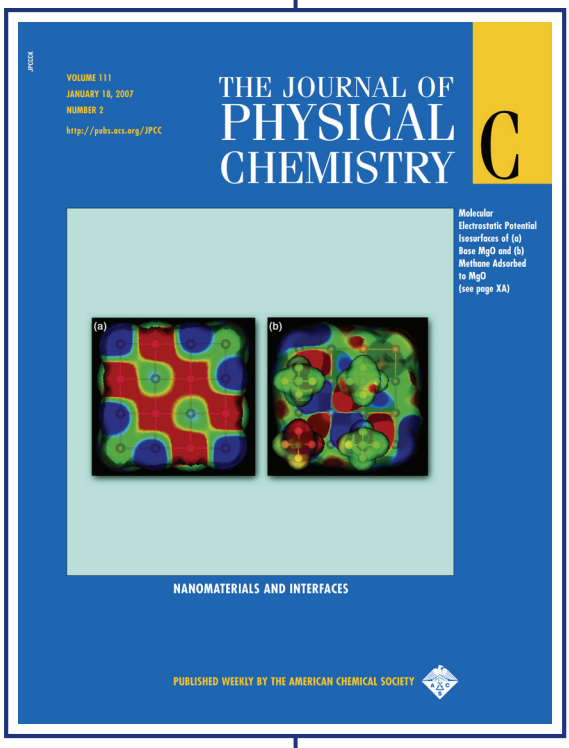
Oak Ridge Research Spotlighted in Chemistry Journal



Adsorption, the small-scale process by which one molecule attaches to another, plays a decidedly large-scale role in our lives. It is used to make thousands of the products that we take for granted—from margarine to gasoline to penicillin—and it is the process by which chemical sensors keep us safe at airports and fuel cells will likely store hydrogen in the high-tech future.

Given the importance of this process, it makes sense that we would want to understand it as well as possible. And given the microscopic scale at which adsorption takes place, it makes sense that computer simulation would greatly bolster that understanding.

Four Oak Ridge researchers using high-performance computers at the National Center for Computational Sciences (NCCS) have significantly enhanced our understanding of this process. Their achievement has landed them on the cover of a prestigious chemistry journal.

Michael Drummond, Bobby Sumpter, William Shelton, and John Larese are the authors of "An Electronic Structure Investigation of Surface-Adsorbate and Adsorbate-Adsorbate Interactions in Multilayers of CH_4 on MgO(100)." The paper is being featured in January as the cover article in the *Journal of Physical Chemistry C*, a publication of the American Chemical Society that focuses on nanomaterials and surface science.

Specifically, the Oak Ridge researchers resolved the molecular structure of successive layers of methane as they attach to magnesium oxide. Sumpter noted that the interaction of molecules with a surface is a fundamental problem facing scientists, but it relates to a variety of important applications, including catalysis and chemical sensors.

"It's very important to understand what drives the adsorption of gases, for example, on surfaces," he explained. "In catalysis, the first thing that happens is a molecule adsorbs on the surface. With sensors, if you're trying to sense some gas—which is a big deal in airports—the first thing that happens is you've got to get the molecule on the material that's part of the sensor. So it's very fundamental."

Indeed, catalysts are directly involved in the synthesis of an estimated one in five industrial products. The energy industry uses catalysis to produce gasoline and develop fuel cells. The food industry uses it to produce margarine and corn-based sweeteners such as fructose. The auto industry uses it in the catalytic converters that remove poisons from exhaust, turning carbon monoxide into carbon dioxide and nitrogen oxide into nitrogen and water.

The team's work builds on a 2006 paper they published in the journal *Physical Review B*. In that paper, they resolved the question of how a single layer of methane arranges on the magnesium oxide surface. In the more recent article, they tackle the question of how subsequent layers of methane arrange themselves.

According to Sumpter, the collaboration began with Larese, a neutron scientist with ORNL's Chemical Sciences Division. Because the Spallation Neutron Source was not yet operational, Larese used the ISIS facility in Great Britain for neutron scattering.

"He said, 'Is there any way one could do simulations on trying to understand what happens when you go from monolayer to bilayer to trilayer, because we see the neutron spectra showing a shift in the peaks.'"

Simulations to that point had not been able to resolve the question of how methane molecules were situated above the surface, Sumpter said. "Simple calculations will get it completely wrong."

"We've cleared it up, we think," he said, with the team using primarily the Cray X1E system known as Phoenix, but also the SGI Altix system known as Ram. "You have to be able to do a fairly high-level calculation, and that requires a reasonably good-sized computer to do these simulations."

Sumpter said the project is a good example of computer simulation complementing experimental results.

"The initial project was trying to understand what he (Larese) was seeing. That's typical: Theory and computation can do two things—we can try to understand an experiment, which happens a lot. Or we can actually come up with a material and tell experimentalists that they should look into it.

"It's a feedback. Without both, it's interesting but not complete, because if you don't have the experiment, it's just a piece of computational theory. With experiment, it's really there."

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