New surface nonlinear spectroscopy capability: Picosecond-femtosecond broadband sum frequency generation system

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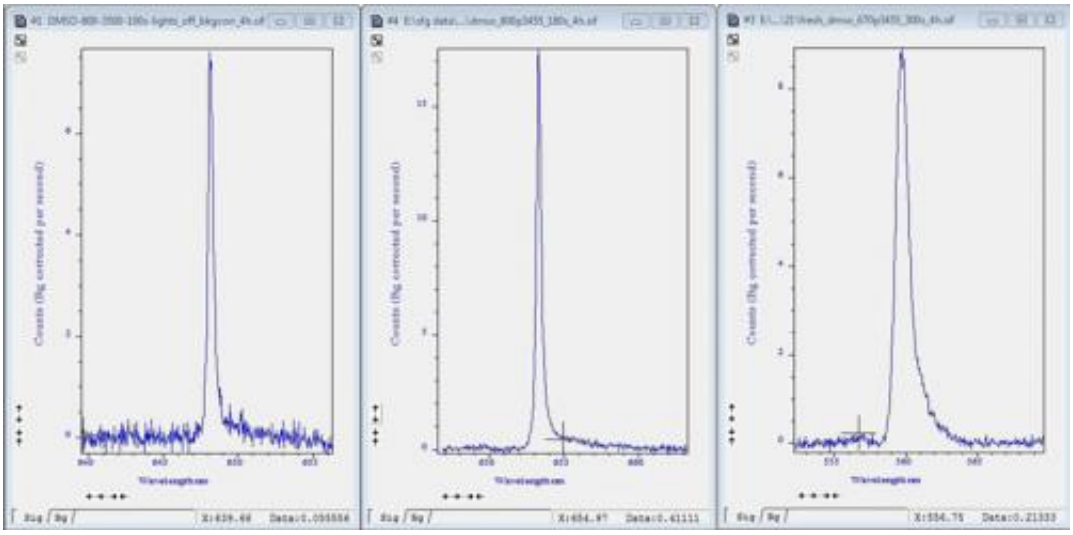

Steeper is better: A look at two spectra generated by the new system, as well as a comparison with previous capabilities. (Left) The first narrow band spectrum achieved in February 2011; (Middle) A subsequent spectrum with an improved signal-to-noise ratio taken a few days later; (Right) A broader spectrum for comparison, taken with a typical laser source.

In Dr. Hongfei Wang's spectroscopy laboratory at the end of EMSL's main hallway, the lights are always off. Because the instrumental capability his team has built uses lasers as its main weapon, light interference would hinder scientific results. So Wang, postdoctoral researcher Dr. Luis Velarde, and visiting scientist Dr. Xianyi Zhang

constantly wear headlamps in the lab, giving them the appearance of old-time coal miners. But instead of coal, they are using the new surface nonlinear spectroscopy capability to dig up never-before-seen data on molecular interactions at interfaces. Early in 2011, they struck a vein that could lead scientists in many fields to research gold.

When the first high resolution vibrational spectrum of the air/DMSO interface appeared on the screen (DMSO is Dimethyl sulfoxide, a very common and important solvent), Wang immediately began to celebrate: after shaking hands with the others and taking a few pictures with his phone, he burst into the hallway to show the evidence to whoever happened to be around. After six months of system design and configuration, and another six months of delivery, installation, and seemingly endless testing, it was finally up and running: the picosecond-femtosecond broadband sum frequency generation system was ready to provide a new generation of surface vibrational spectroscopy and imaging.

"I was very relieved," he said. "We expected it to happen, and it happened. Now we know we have something that is truly unique—the SFG community has been waiting for this, and many scientific fields will benefit."

Back in the lab, Velarde hadn't started celebrating yet. He wanted to make sure everything was just as they expected it would be, and that the spectrum was genuine proof of the system's capability. When he was satisfied, he finally let himself enjoy the moment as well.

For Wang, the milestone was more than a successful project at work; it confirmed his decision to leave his home nation and a job at the Chinese Academy of Sciences in 2009.

"EMSL is the perfect place to develop this capability. Not many places

offer an environment like this, and surface chemistry is crucial to all three of EMSL's science themes." He added, "After we generated the first spectrum, I called my wife to tell her: the decision to move here has been validated."

Sum frequency generation (SFG) is a highly specialized surface nonlinear spectroscopy technique scientists use to analyze molecular interactions at surfaces and interfaces of all kinds. It is an important, crosscutting technique that can unlock new discoveries in several energy, environmental, and health-related research areas. While the technique has been pioneered by Professor Ron Shen at Berkeley in the 1980s, there is a very small community worldwide that can perform these very difficult experiments to understand the one or few layers of molecules at various interfaces. As for resolution, strength, and efficiency of this capability, this recent "Moment" demonstrates that Wang and his team now stand alone. Previous techniques forced researchers to choose between signal strength and resolution, and carry out time-intensive examinations of each specific data point.

The new system in EMSL for the first time synchronizes two powerful lasers with completely different characteristics. Namely, one has very short laser pulses (35fs, 1fs=10^{-15} second) which provides the ultrafast time resolution, and another has very long pulses (100ps, 1ps=10^{-12} second) which provides the high spectral resolution. This offers the best of both worlds: reliable data is gathered in a few seconds or minutes, at more than ten times the best previously documented spectral resolution with the similar systems. Specifically, they achieved 0.7cm^{-1} spectral resolution, versus>15cm^{-1} resolution. With these gains, the tool is ready to reveal detailed molecular conformation and interactions at the molecular interface.

The capability is best shown by example: imagine a team of researchers who want to examine a sample with a liquid-liquid interface: oil and

water. This is an example of a "buried interface"—a difficult case for many experimental techniques. SFG specializes in this problem. Experimentalists like Wang interrogate the sample by shooting two pulsed lasers through the oil and water so they meet at the liquid-liquid interface at an exact time (on the picosecond scale). If done correctly, the signal the sample sends back is "surface sensitive"—which means it selects only information about the interface (the two-atom-wide molecular interaction the scientists care about), clearing away the background noise. The resulting spectra allow researchers to piece together what is truly happening on a molecular level—such as how these molecular groups are oriented. In the case of where oil and water meet, deeper fundamental information can provide new insights for environmental cleanup. In addition, the use of lasers causes far less damage to the sample (as opposed to bombarding it with ions) and allows scientists to perform in situ experiments that replicate true environmental conditions.

Wang is often invited to give talks around the world on surface nonlinear spectroscopy. On March 16, 2011, he gave a seminar of his latest findings at Oregon State University in Corvallis, Oregon, and he will do the same at Rice University in October 2011.

Like all of EMSL's experimental and computational tools, the new surface nonlinear spectroscopy capability is available at no cost to the global scientific community through EMSL's user proposal process.

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