# Killer pulses help characterize special surfaces

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Detecting deadly fumes in subways, toxic gases in chemical spills, and hidden explosives in baggage is becoming easier and more efficient with a measurement technique called surface-enhanced Raman scattering. To further improve the technique's sensitivity, scientists must design better scattering surfaces, and more effective ways of evaluating them.

Researchers at the University of Illinois, led by chemistry professor Dana Dlott, have devised a method to evaluate substrate surfaces by using a series of killer laser pulses. They describe the method and report measurements for a commonly used substrate in the July 18 issue of the journal *Science.*

Surface-enhanced Raman scattering, which functions by adsorbing molecules of interest onto rough metal surfaces, typically enhances the Raman spectrum a million times. Hot spots can occur, however, where the electric field enhancement can be a billion or more.

Current surface characterization techniques cannot tell hot spots from cold spots, and create an average value across the entire substrate surface.

"Looking at a spectrum, you can't tell if it's the result of a small number of molecules in hot spots or a large number of molecules in cold spots," Dlott said. "Two materials could have the same average spectrum, but behave quite differently."

Dlott, graduate student Ying Fang and postdoctoral research associate Nak-Hyun Seong came up with a way to measure the distribution of site enhancements on the substrate surface. Using killer laser pulses, their technique can count how many molecules are sitting in the hottest spots, how many are sitting in the coldest spots, and how many are sitting between the two extremes.

The killer pulse is a short duration laser pulse with a variable electric field. When the electric field is strong enough, it rips a molecule apart, "killing" it.

"If a molecule is in a very hot spot on the substrate, where the electric field enhancement is really big, it takes only a weak pulse to kill it," Dlott said. "If the molecule is in a very cold spot, then it takes a really big laser pulse to kill it."

Dlott, Fang and Seong demonstrated their technique by measuring the distribution of local enhancements for benzenethiolate molecules on a substrate of silver-coated nanospheres 330 nanometers in diameter.

To characterize the surface, the researchers first measured the initial Raman intensity. Then they put in a weak killer pulse, which destroyed the molecules in the hottest spots. After measuring the new Raman intensity, they put in a bigger pulse and destroyed the molecules in slightly colder spots. The researchers continued with bigger and bigger pulses until all the benzenethiolate molecules were destroyed.

"We found the hottest spots comprised just 63 molecules per million, but contributed 24 percent of the overall Raman intensity," Dlott said. "We also found the coldest spots contained 61 percent of the molecules, but contributed only 4 percent of the overall intensity."

Measurements like these, of the distribution of local site enhancements, will help researchers design better scattering surfaces for sensor applications.

Prior to this work, no one knew if the Raman intensity was dominated by a small number of hot molecules or a large number of cold ones. Dlott, Fang and Seong have answered that important scientific question; not just with a yes or no, but with a full determination of exactly how many molecules there are in each level of hot or cold.

"Now, when evaluating a new surface-enhanced Raman material, instead of knowing just the average intensity, we know the highest, the lowest, and everything in between," Dlott said.

Source: University of Illinois at Urbana-Champaign

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