Resonance Raman scattering provides new ways for high-sensitivity temperature probing

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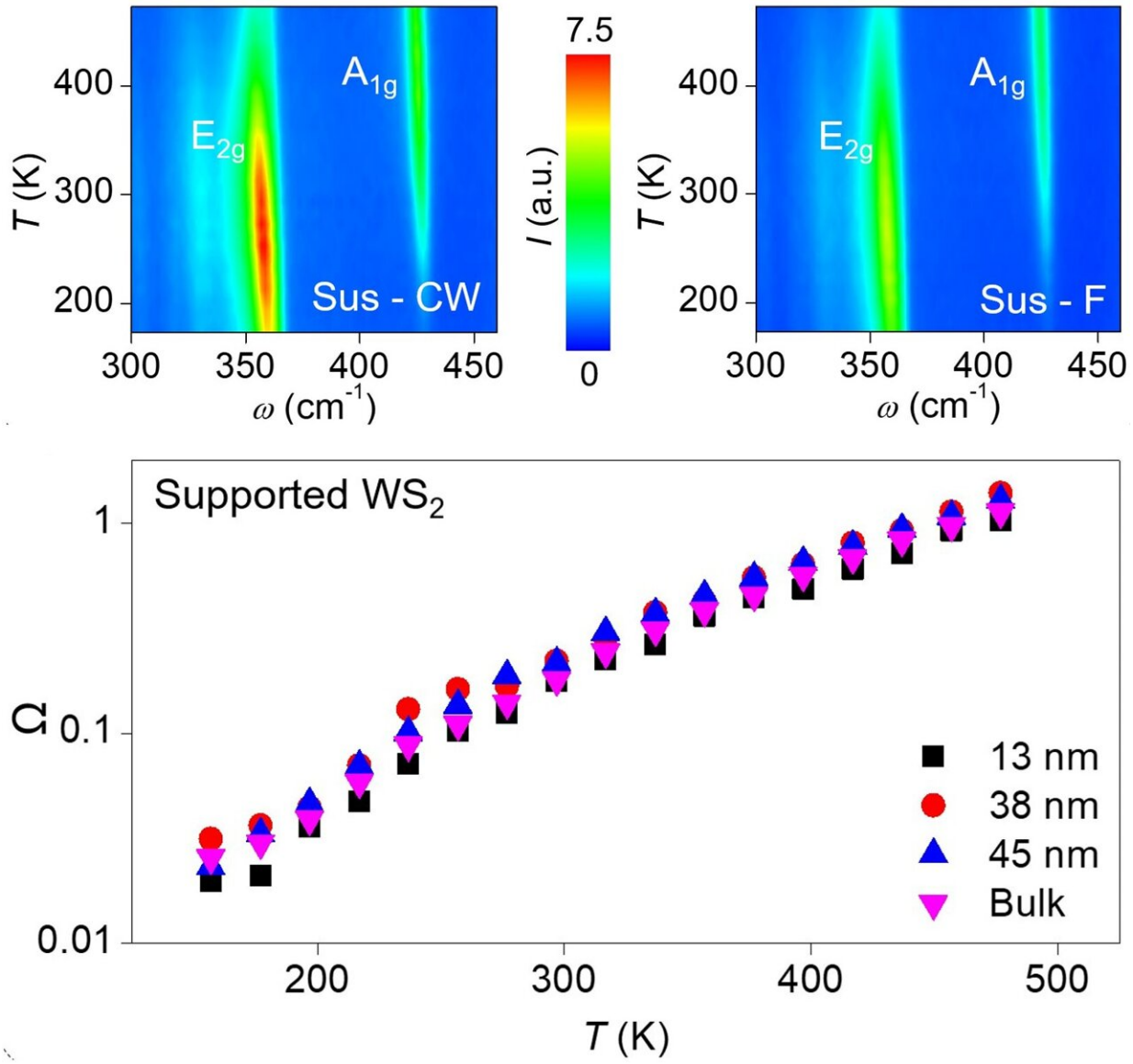

When a nm-thick WS_2 is experiencing resonance Raman scattering under 532 nm laser excitation, its two Raman peaks (A_{1g} and E_{2g}) have different variation behaviors against temperature, while their ratio ($\Omega = I_{A1g} / I_{E2g}$) shows universal behavior regardless of the sample structure (thickness, suspended or supported). This ratio changes by more than 100-fold from 177 K to 477 K, demonstrating its robustness in high-sensitivity temperature probing. Credit: Hamidreza Zobeiri et al

Thermal scientists from the Iowa State University, Shenzhen University, and Shanghai University of Engineering Science, have developed a new thermal probing technique based on the ratio of two resonance Raman scattering peak intensities.

Publishing in the *International Journal of Extreme Manufacturing*, the team led by Prof. Xinwei Wang at Iowa State University, systematically studied and proved that the ratio of two resonance Raman peak intensities of a 2D material can be used as an indicator for high-sensitivity temperature measurement. This new development will significantly broaden the traditional Raman-based temperature measurement (based on wavenumber shift) while significantly improving the measurement sensitivity and robustness.

Raman-based thermometry has been used for decades, mostly by tracking the wavenumber shift to measure temperature. This renders a very unique material-specific nature of Raman thermometry, making it possible to achieve very specific temperature measurement and probe a temperature drop across a sub-nm spacing.

However, Raman wavenumber is subjected to various experimental noises and uncertainties, such as optical focusing, optical interference within a material and across an interface. The ultimate measurement

sensitivity is documented low. Although Raman scattering intensity also changes with temperature, it is rarely used for temperature measurement since it is hard to control all the experimental conditions to well define the scattering intensity.

In resonance Raman scattering (e.g WS_2), due to the slight bandgap change against temperature, the scattered Raman intensity is very sensitive to temperature, and the intensity of a single Raman peak is still difficult to use for temperature measurement.

By using WS_2 nanofilms, either supported or suspended, the three teams at Iowa State University, Shenzhen University, and Shanghai University of Engineering Science discovered that the two Raman peaks of WS_2 (E_{2g} and A_{1g}), although each of them shows different variation trend against temperature, their intensity ratio surprisingly shows a very universal behavior, regardless of the material's physical size, suspended or supported, nm-level or macrosize.

Also this ratio shows dramatic change from 177 K to 477 K (>100-fold). This clearly demonstrates its capability for temperature measurement. Using this ratio as the indicator, the teams have characterized the thermal diffusivity and thermal conductivity of suspended WS_2 nanofilms with their energy transport state-resolved Raman (ET-Raman). The results agree very well with the measurement based on Raman wavenumber.

One of the team leaders, Prof. Xinwei Wang said, "This Resonance Raman Ratio (R3) method is superior to the classical wavenumber-based temperature measurement in three aspects."

First, since the intensity ratio is used, any optical focusing or optical interference-induced intensity shift will be automatically eliminated in the ratio. This will dramatically improve the measurement robustness.

Second, for many wavenumber-based methods, at low temperatures the Raman wavenumber becomes much less sensitive to temperature change, making the measurement less reliable.

However, the R3 method has an almost universal sensitivity from 177 K to 477 K. For even lower temperatures, measurement is possible by searching for appropriate materials whose bandgap change will cause larger intensity variation at lower temperatures. Third, the finding will make WS_2 a promising temperature sensor for measuring the temperatures of non-Raman active materials. The sensor's time response will be extremely fast (

This is very attractive for temperature monitoring in extreme manufacturing.

One of the team leaders, Prof. Yangsu Xie is leading her team to conduct active research to study thermal transport in nanoscale materials using Raman spectroscopy. She says that "the R3 method truly opens a new avenue to study a material's thermal response under either optical or other types of thermal loading. This will significantly improve our experimental capability of exploring nanoscale thermal transport physics that is hard to probe using other techniques."

"Also the R3 method still holds the material-specific feature, so it makes it possible to achieve temperature probing of very well defined physical domain. We are excited about the promising applications of this technique in high-resolution temperature monitoring in extreme manufacturing as well as in microelectronics."

Although the work only reported the R3 measurement using 532 nm laser-induced resonance Raman scattering, it is possible to choose other wavelength lasers (e.g. 633 nm, 488 nm, 785 nm) for resonance Raman scattering with materials of matched/close bandgap. This could extend

the temperature measurement range or shift the range to a designed level.

This high sensitivity makes it possible to employ the R3 method for monitoring materials' thermal response in extreme manufacturing for process physical understanding, control, and optimization with very high spatial resolution (~nm) and temporal response (

More information: Hamidreza Zobeiri et al, Robust and high-sensitivity thermal probing at the nanoscale based on resonance Raman ratio (R3), *International Journal of Extreme Manufacturing* (2022). DOI: 10.1088/2631-7990/ac6cb1

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