

Detecting non-uniformities in 2D materials may lead to new medical sensors

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(Foreground) Doxorubicin molecule, detected using the van der Waals vertical heterostructure biosensor. (Background) Actual nanoscale optical image (sSNOM) of the heterostructure: large triangle is a single-layer MoS₂ island (ca. 3.7 micron wide); smaller triangle is a partially oxidized MoOS island; whole sample is covered with the monolayer graphene, with several wrinkles clearly seen in the map; darker graphene area corresponds to the region of extra charge doping. Credit: Jennifer M. McCann / Rotkin Group

A novel and better approach at detecting non-uniformities in the optical properties of two-dimensional (2D) materials could potentially open the door to new uses for these materials, such as the application of 2D materials for drug detection, according to a team of researchers.

"The Two-Dimensional Crystal Consortium (2DCC) is a world leader in 2D materials research and my lab often works with the 2DCC doing materials characterization for novel 2D materials," said Slava V. Rotkin, Frontier Professor of Engineering Science and Mechanics with an appointment in the Materials Research Institute at Penn State. "There is a big challenge in these studies: Frequently, optical properties of 2D materials are not uniform in space. Furthermore, they may vary at a very small spatial scale, down to a single atom."

Identifying and understanding such a variability of properties could be extremely important for certain applications of 2D materials, which are materials that are one to a few atoms thick. Such atomically thin materials, having an ultimate surface-to-volume ratio, may possess surface non-uniformities at the nanometer scale. This includes atomic impurities, adsorbates, defects, wrinkles, ruptures and so on. Such features can modulate the optical properties and result in variability of materials' properties.

"Despite this being critical for effectiveness in certain application of 2D materials, there is currently no truly effective approach to detect these variabilities," Rotkin said. "Due to their being so tiny, they are undetectable by optical tools and non-optical tools cannot resolve optical contrast."

Rotkin and other researchers were able to take one step toward a possible solution, which was outlined in a recent study in *ACS Nano*. This solution would potentially lead to better applications of 2D materials for medical sensing.

The researchers conducted experiments using a heterostructure material made of graphene, the 2D material version of graphite, and the inorganic compound molybdenum disulfide (MoS₂). The MoS₂ gives a [photoluminescence](#) signal that detects the amount of charge transfer between the graphene and the MoS₂ layers, and therefore can detect changes due to the bio analyte, in this case the cancer treatment drug doxorubicin (DOX), that can affect the charge. However, graphene itself can detect these changes via analysis by Raman spectroscopy, which detects unique vibrations in molecules. Raman microscope picks up shifts in the frequency of photons in the laser light beam caused by these vibrations.

"The two channels together allow a better calibration of two the signals against analyte concentration and the type of analyte," Rotkin said. "And additionally, graphene enhances the Raman signal of the analyte itself to the extent one can 'see' a signal from just a few molecules."

The researchers used DOX as their analyte because it is a common cancer drug, and there is an acute need for good medical devices for it, including sensors. Two types of [biosensors](#) are label-free biosensors, which can be used to detect a variety of drugs, and label-based biosensors, which can detect only a specific drug. The researchers used label-free biosensing.

"The label-based biosensor is like a lock that can be opened with only one key, but the label-free biosensor is like a lock with many different keys," Rotkin said. "We did not invent label-free multimodal biosensing, this approach has been in other studies. But an actual demonstration with a specific material is new and still important by itself."

This is significant because label-free biosensing is more challenging than label-based biosensing.

"We make it work by merging several sensors in one device, think about the lock and key analogy as three locks on one chain," Rotkin said "Specifically, we apply the DOX to our 2D material, which produces three different optical signals, constituting a multimodal sensing. By measuring three signals at once instead of just one like in a normal sensor, this allows us to detect DOX using label-free biosensing."

While Rotkin stresses they only gave a demonstration of the principle in the study, there are potential applications of this new mechanism of label-free biosensing. There potentially could be sensors that enable label-free sensing of bio-, chemical and/or medical analytes of interest with minimal sample preparation, in an abbreviated time frame, with low detection limits, and using samples containing substances other than the key analyte.

This could lead to steps for solving various health care challenges.

"Keeping in mind that there is a gap between fundamental research and its applications, I would say we contributed a brick to building a large set of nanotechnology/nanomaterials for biosensing and other applications," Rotkin said. "Label-free detection lays the groundwork for smart and integrated sensors, new bio-threat safety techniques and more individualized medicine and treatments, among others benefits."

In the meantime, there are also more immediate benefits to this research, according to Rotkin.

"This work gives us deeper knowledge of overall optical properties of 2D materials," Rotkin said. "We uncovered some of the mechanisms for one specific structure, graphene and MoS₂. But our nanoimaging method is applicable to many others, if not to all. Also, we hope to attract additional attention to the physics of 2D material heterostructures such as our composite material which combined the properties of

graphene and MoS2 single-layer materials."

The next steps for this research will include applying the materials component of their work to other projects at the 2DCC, including those involving quantum plasmonics and 2D non-linear optics. In addition, the research team will be looking for partners for researching practical applications.

"Since label-free detection is universal, we are not limited by a type of analyte, application nor problem," Rotkin said. "Still, there needs to be someone with a real problem to apply the approach. We are looking for collaborators from the world of medicine for some exciting new joint research."

More information: Tetyana Ignatova et al, Multidimensional Imaging Reveals Mechanisms Controlling Multimodal Label-Free Biosensing in Vertical 2DM-Heterostructures, *ACS Nano* (2022). [DOI: 10.1021/acsnano.1c09335](https://doi.org/10.1021/acsnano.1c09335)

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