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Controlled layer-by-layer thinning of oxygen-sensitive 2D materials

Owing to quantum confinement and interlayer coupling effects, the electronic and optical properties of 2D materials strongly depend on the number of layers. Hence, methods for controlling 2D materials' thickness with atomic layer precision are highly desirable. Yet, the very few methods reported so far have been demonstrated only for a specific material (i.e., MoS_2 or black phosphorous), make use of techniques that do not allow for scalability, and might even induce structural damage to it [1-4].
Here we present a simple and scalable method for thinning down oxygen-sensitive 2D materials. Notably, our strategy is based on an oxidation/reduction process, which can ultimately be controlled in order to achieve layer-by-layer thinning.
Through a combination of atomic force microscopy, X-ray photoemission spectroscopy, Raman spectroscopy and X-ray diffraction experiments, we investigate this method in details on a large number of 2D materials, comprising germanium arsenide (GeAs), germanium sulfide (GeS) and germanium disulfide (GeS_2) and black phosphorous (P). This suit of experimental techniques allows us to gather a holistic view of the thinning process, and to get insights into the fundamental chemical mechanism.
We also demonstrate the applicability of our method by using it to prepare and test electronic devices. Notably, we perform a systematic study of the carrier transport behavior of few-layers GeAs, the layer number of which is varied continuously by repeated oxidation/reduction cycles.
Our strategy, which we believe could be applied to other classes of 2D materials upon proper choice of the oxidation/reduction agent, could pave the way for the realization of 2D material-based devices, such as electronic or opto-electronic ones, where a precise control over the number of layers (hence over the material's physical properties) is needed.

References

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Figures

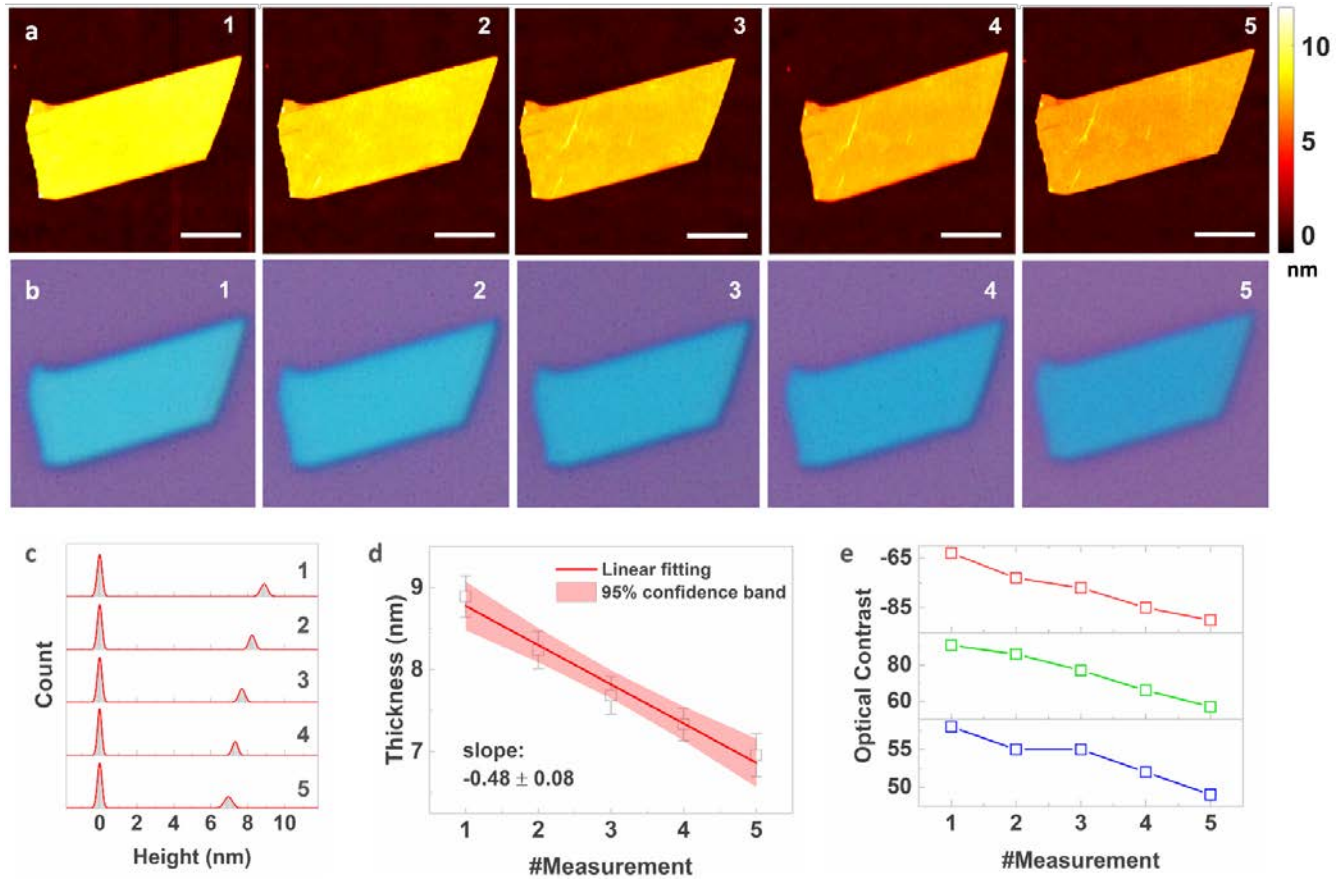

Figure 1: Controlled thinning of a GeAs flake. (a) Atomic force microscopy and (b) optical microscopy images of a GeAs flake collected after each oxidation/reduction cycle, for a total of 5 cycles. (c) Analysis of the atomic force microscopy images in (a). (d) Graph showing the direct proportionality between the thickness of the flake in (a) *vs* the number of oxidation/reduction cycles. (e) Graph showing the change in the R, G and B values in the optical images of the GeAs flake shown in (b) after each oxidation/reduction cycle.

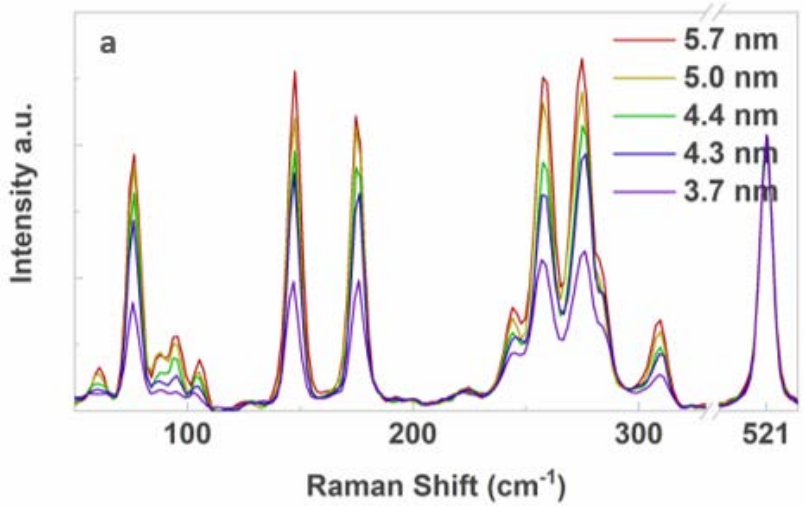

Figure 2: Raman spectroscopy of a thinned GeAs flake. Raman spectra taken on a GeAs flake gradually thinned down from 5.7 nm to 3.7 nm. The intensity of the characteristic peaks decreases as a function of the flake thickness, while the sample seems unaltered – i.e. neither peaks disappear nor new peaks emerge.