

Electronic Supplementary Material

Fast and reliable identification of atomically thin layers of TaSe₂ crystals

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Supporting information to DOI 10.1007/s12274-013-0295-9

Characterization of atomically thin TaSe₂ crystals

Figures S1 and S2 shows the same information as Fig. 1 in the main text for other two TaSe₂ flakes. Both the optical and atomic force microscopy characterization of the flakes is shown.

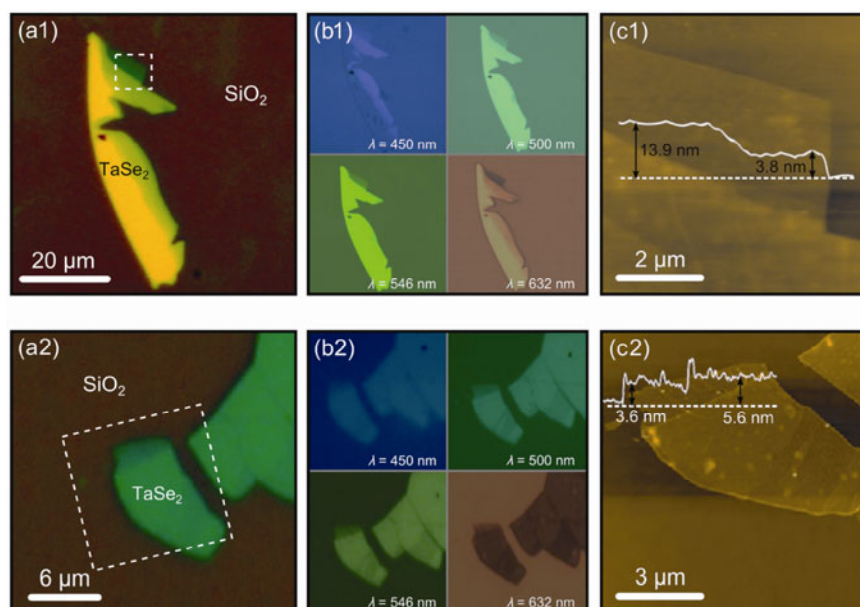


Figure S1 Optical and atomic force microscopy images of two different TaSe₂ nanosheets: (a) optical micrographs under white illumination of TaSe₂ flakes deposited on a 300 nm SiO₂/Si substrate; (b) different optical micrographs acquired at 450 nm, 500 nm, 546 nm and 632 nm by using narrow bandwidth filters; (c) AFM topography image of the region marked by a dashed rectangle in (a). A topographic line profile along the horizontal line is inserted to indicate the thickness of the flakes.

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Refractive index of TaSe₂

For the calculation of the optical contrast of atomically thin TaSe₂ crystals (see Figs. 2 and 3 of the main manuscript) we employed the complex refractive index reported for bulk 2H-TaSe₂ [S1]. In Ref. [S1], however, the two components of the complex dielectric permittivity are shown. In Fig. S3 we show the complex refractive index calculated from the dielectric permittivity values reported in Ref. [S1].

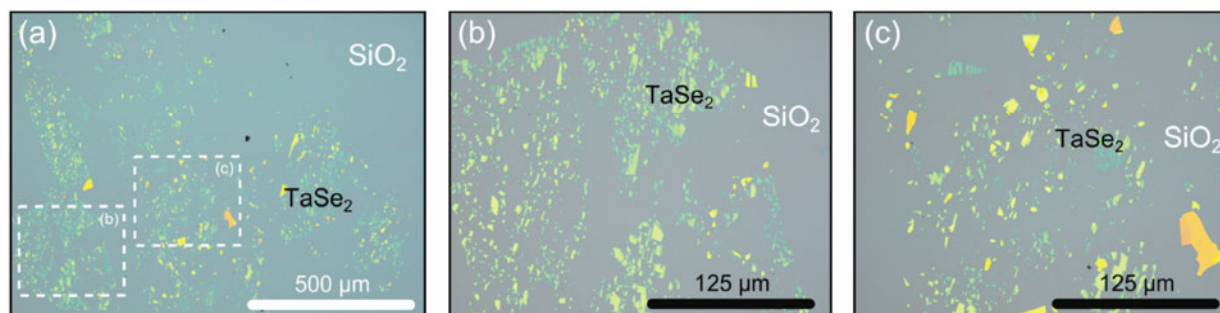


Figure S2 Low magnification optical images of few-layer TaSe₂ crystals exfoliated onto a 300 nm SiO₂/Si substrate. Greenish flakes are typically thinner than 10–15 layers. (b) and (c) Close up images of the areas marked by the dashed rectangles in (a).

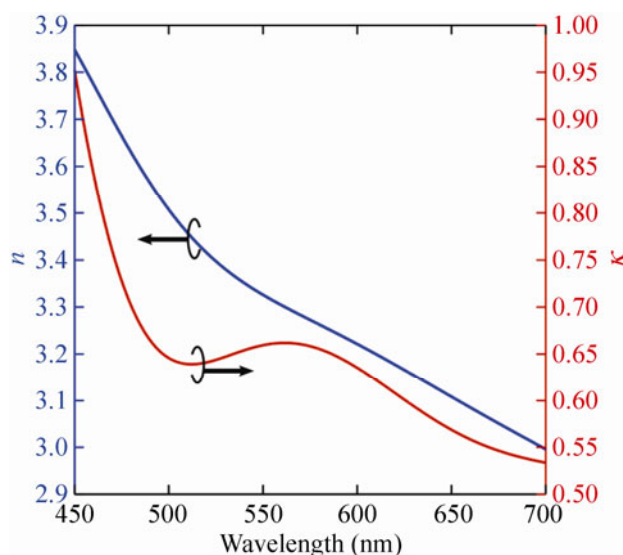


Figure S3 Complex refractive index (real part n and absorption κ) of 2H-TaSe₂ obtained from the dielectric permittivity values reported in Ref. [S1].

Optimal dielectric thickness to facilitate the optical identification

The optical contrast yielded by a single layer TaSe₂ sheet has been calculated using Eqs. (1) and (2) from the main text and the refractive index reported for bulk 2H-TaSe₂ [S1] (see Fig. S4). We have considered the case of different dielectric layers such as: SiO₂ (a), PMMA (b), Al₂O₃ (c), HfO₂ (d) and Si₃N₄ (e). The optimal dielectric thickness to identify ultrathin layers of TaSe₂ has been marked in Fig. S4 with white circles.

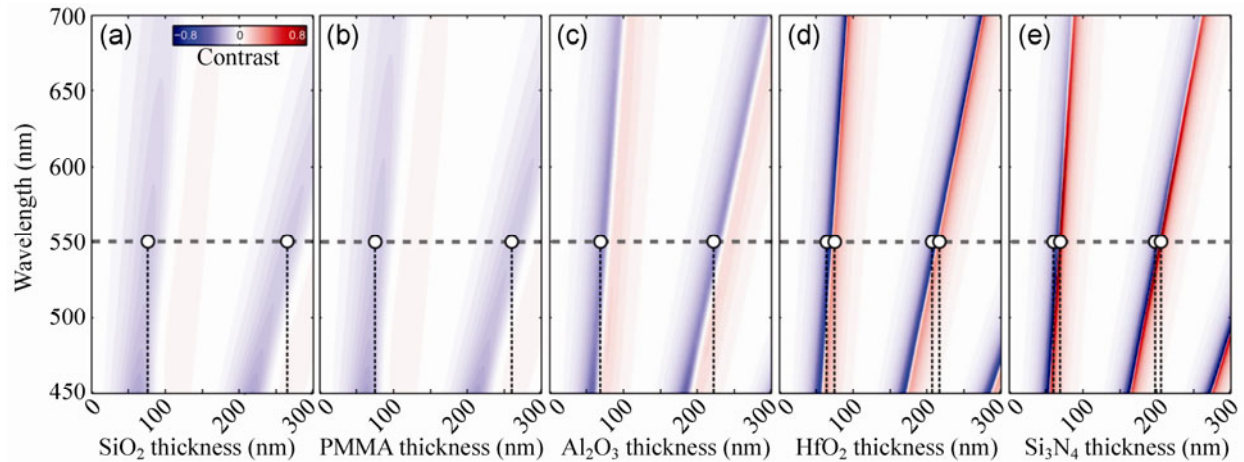


Figure S4 Optical contrast for a single layer TaSe₂, 0.64 nm thick, (colormap) as a function of the wavelength and dielectric thickness calculated with Eqs. (1) and (2) and the complex refractive index reported for bulk 2H-TaSe₂ [S1].

For SiO₂, for instance, the optimal thicknesses to facilitate the optical identification are 80 nm and 265 nm. In Fig. S5 we present the calculated optical contrast as a function of the illumination wavelength and the TaSe₂ thickness for these SiO₂/Si substrates.

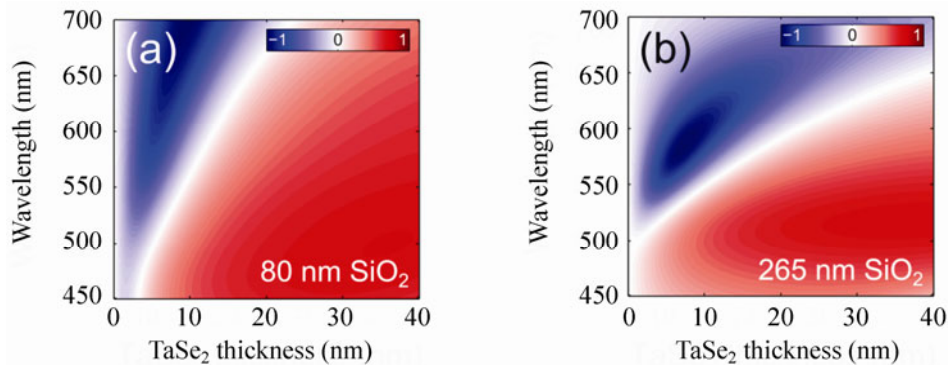


Figure S5 The calculated optical contrast as a function of the wavelength and the TaSe₂ thicknesses for substrates with (a) 80 nm and (b) 265 nm SiO₂ thickness (which are optimal to identify ultrathin TaSe₂ on SiO₂/Si wafers).

Characterization of the laser-induced defects

As pointed out in Fig. 5 of the main text, prolonged or high intensity Raman spectroscopy measurements on TaSe₂ flakes causes laser-induced defects. These defects can be easily identified because they drastically change the optical contrast of the TaSe₂ flakes. Figure S6(a) shows an optical image of TaSe₂ several flakes. The central flake has been subjected to a prolonged Raman spectroscopy image which damaged the flake as can be seen from the change in contrast inside the black frame in Fig. S6(a). Figure S6(b) shows the AFM topography of the damaged region.

As seen in Fig. S6, although the optical contrast drastically changed, the thickness of the flake was scarcely affected. This indicates that the laser-induced damage of the flake alters its optical contrast. This is in agreement with the occurrence of laser-induced oxidation, as tantalum oxide is expected to have a lower optical absorption coefficient than TaSe₂, thus explaining the reduced optical contrast. Therefore, optical contrast can be used to determine whether a Raman spectroscopy measurement has damaged the sample or not.

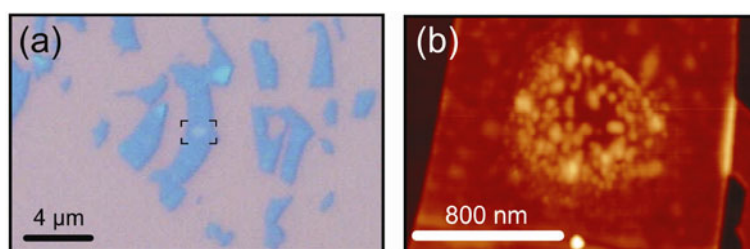


Figure S6 (a) Optical image of TaSe₂ after an intrusive Raman spectroscopy measurement. The laser-induced defects are clearly visible as a drastic change in optical contrast (see the region within the black frame). (b) AFM topography image of the region within the black frame in (a) showing that the region was modified during the Raman spectroscopy measurements.

Figure S7 shows another example where a high power laser was scanned over part of the surface of a TaSe₂ flake to intentionally damage it. The optical images before (a) and after (b) clearly show that one can identify the laser-modified part just by the difference in optical contrast. The AFM topography in Fig. S7(c) shows a slight decrease of the thickness of the region modified with the laser. Note that this small reduction in the thickness is not sufficient to explain the drastic reduction of the optical contrast in the scanned area.

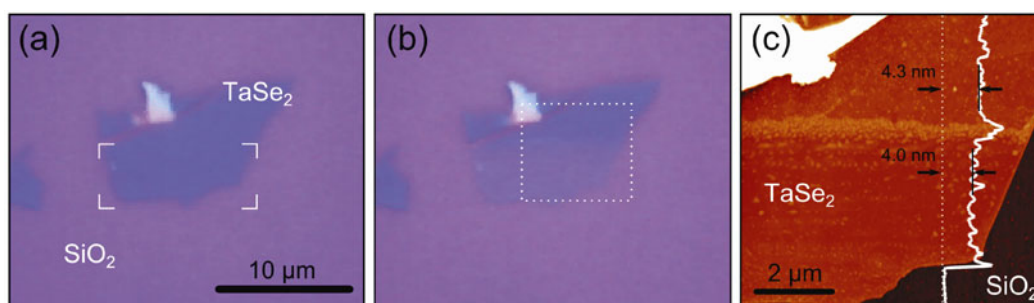


Figure S7 Optical images of TaSe₂ (a) before and (b) after scanning with a high-power laser (10 mW, 500 nm spot size, scanning steps of 500 nm with a waiting time of 0.1 s between steps). The laser scanning was carried out in the region denoted by the white frame in (a). (c) The AFM topography of the region marked by the dotted square in (b). A topographic line profile has been included in (c).

In order to obtain more information about the nature of the laser-induced damage on the TaSe₂ flakes we carried out Raman spectroscopy measurements on a TaSe₂ flake while increasing the laser power (Fig. S8). The flake was deposited onto a gold electrode to avoid the strong signal from the silicon underneath (with intense peaks around 950 cm⁻¹, 520 cm⁻¹ and 300 cm⁻¹), which hampers the identification of some of the laser-induced features in the Raman spectra. For a low power laser, the Raman spectra show the E_{2g}^1 and A_{1g} peaks (see Fig. 4 of the main text) and a background signal. With increasing power, however, a feature around 255 cm⁻¹ starts to develop (as seen in Fig. 5 of the main text) and becomes more and more pronounced as the exposure to the laser increases. This Raman peak matches the energy of the most intense Raman peak for crystalline Ta₂O₅ in that part of the spectrum [S2]. Furthermore, together with the appearance of the peak at 255 cm⁻¹, two broad peaks around 1,350 cm⁻¹ and 1,550 cm⁻¹ develop. These peaks correspond to a photoluminescence emission around 550 nm to 560 nm. Note that photoluminescence emission of similar characteristics has been observed for Ta₂O₅ of different forms (nanorods, nanoblocks, etc.) [S3, S4].

In summary, the observed change in the optical contrast, the presence of a Raman peak at 255 cm⁻¹ and the photoluminescence emission around 550–560 nm suggest that the laser-induced defects are probably due to the oxidation of tantalum to give Ta₂O₅. Nevertheless, more work would be necessary to study this oxidation process in more detail, which is out of the scope of the current work.

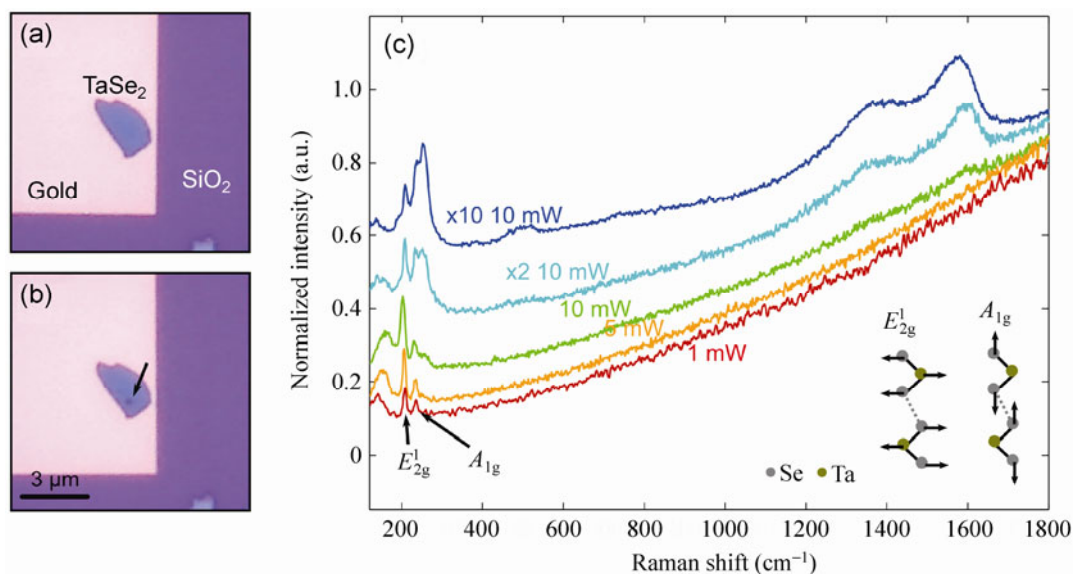


Figure S8 Optical images of TaSe₂ deposited on a gold electrode (a) before and (b) after performing several Raman spectroscopy measurements with different laser power. (c) Raman spectra measured on the TaSe₂ with 1 mW (1 s accumulation), 5 mW (1 s accumulation), 10 mW (1 s accumulation), 10 mW (2 s accumulation) and 10 mW (10 s accumulation) laser power.

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