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**COMMENTS AND REPLIES • <span style="color:red">OPEN ACCESS</span>**

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# 2D Materials

**COMMENTS AND REPLIES**

# Reply to comment on 'The Computational 2D Materials Database: high-throughput modeling and discovery of atomically thin crystals'

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## Abstract

In his comment Maździarz 2019 (*2D Mater.* **6** 048001) raises doubts concerning the reliability of our test for dynamical (in particular elastic) stability of monolayer materials, which neglects the shear components of the stiffness tensor and only considers the sign of the planar stiffness coefficients. We agree that our analysis has not been complete, but find that it suffices in practice except for very few cases (less than 1% of the materials). Nevertheless, for completeness we are currently calculating the shear components of the elastic tensor for all the materials in the C2DB.
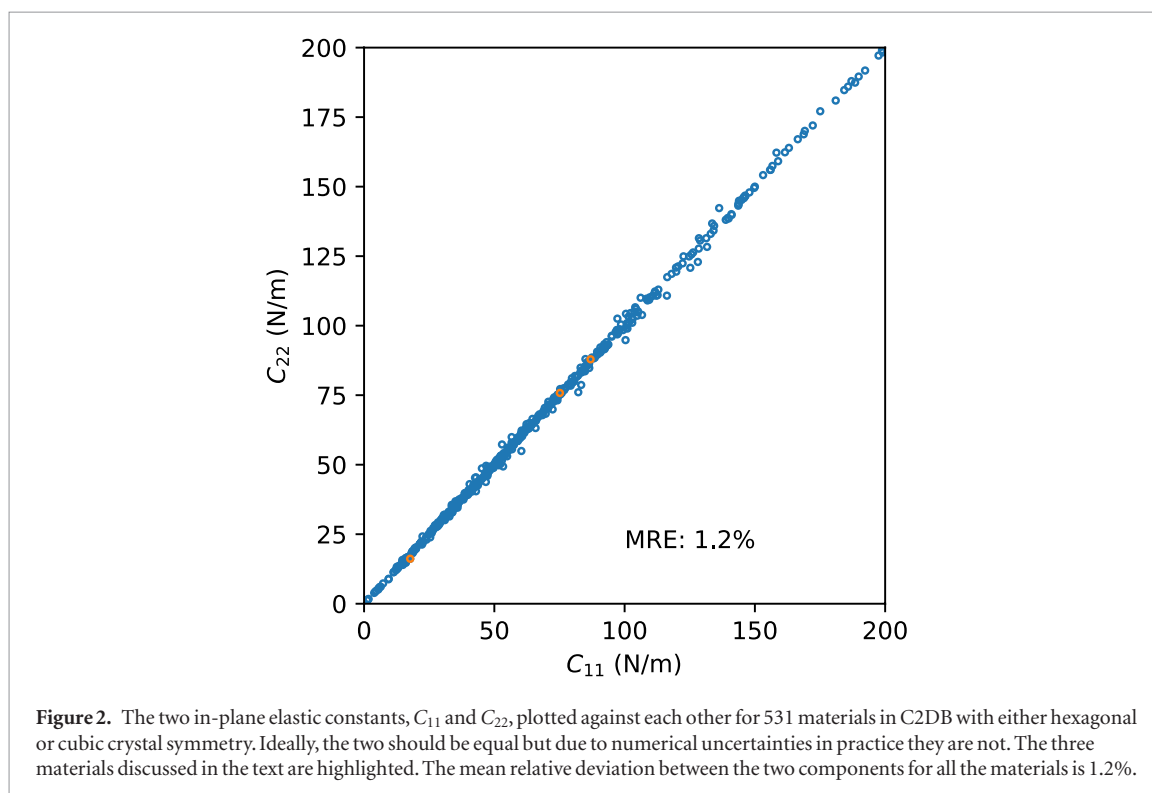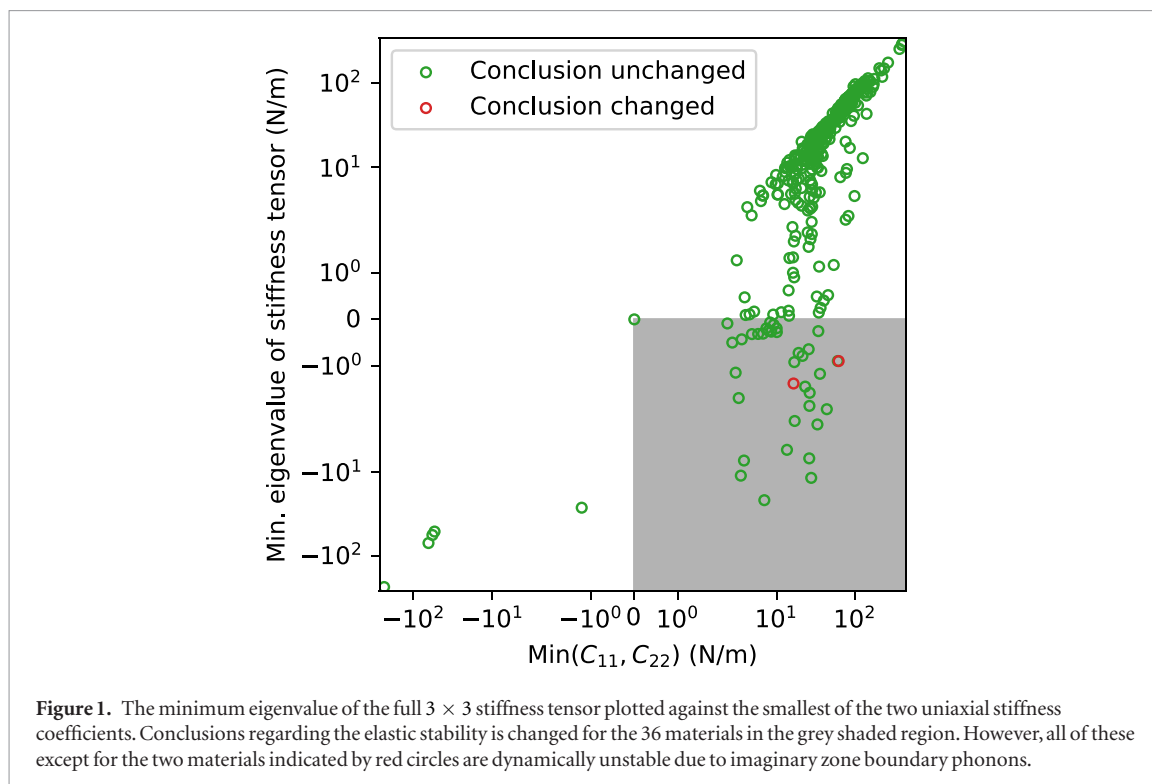
In our original paper [2], we described our strategy for testing whether a given hypothetized 2D material would be dynamically stable, i.e. whether it would spontaneously distort if all constraints imposed on the calculation (symmetries and unit cell size) were relaxed. In other words, the test for dynamical stability should assess whether the configuration of the given material represents a minimum or a saddle point of the potential energy surface. Regarding the atomic positions within the unit cell, we calculate the phonons at the corners of the Brillouin zone boundary (specifically the $\Gamma$-point phonons of the $2 \times 2$ repeated cell). The material is classified as dynamically unstable if at least one phonon with imaginary frequency is found. Concerning the shape of the unit cell, we calculate the components of the stiffness tensor corresponding to uniaxial deformations along the $x$, and $y$-axis, namely the $C_{11}$, $C_{22}$, and $C_{12}$ components in the Voigt notation. A material is classified as dynamically unstable if either $C_{11}$ or $C_{22}$ is negative.

As pointed out in the comment, the correct test for dynamical stability would involve, in addition to the phonon analysis, a diagonalization of the full stiffness tensor to check for negative eigenvalues. By considering only the sign of $C_{11}$ or $C_{22}$ there is a risk that a material is incorrectly classified as dynamically stable when in reality it would undergo a shear deformation.

We have calculated the full $3 \times 3$ stiffness tensor, $C$, for 378 materials in the C2DB. We picked this set of materials because we already had calculated the shear deformations in connection with the calculation of their piezoelectric tensors. They cover representatives from all five types of 2D Bravais lattices. In figure 1 we show the minimum eigenvalue of $C$ plotted against $\min\{C_{11}, C_{22}\}$. There are 36 materials in the grey shaded area where our original assessment of dynamical stability based on the $C_{11}$ and $C_{22}$ components is wrong. Most of these are materials in the GeS$_2$ structure prototype. However, 34 of these have at least one imaginary zone boundary phonon and would therefore be classified as dynamically unstable in any case. Therefore, the stronger criterion based on the full stiffness tensor only leads to a different conclusion for two materials, namely GeSe$_2$ in the GeS$_2$ prototype and I$_2$Sb$_2$ in the CuI prototype, which are now classified as dynamically unstable.

Maździarz highlights three specific materials from C2DB , namely Au$_2$O$_2$–GaS, Ta$_2$Se$_2$–GaS, and Re$_2$O$_2$–FeSe, and criticises that (1) despite the hexagonal and cubic symmetries of the lattices $C_{11}$ and $C_{22}$ are not equal for these materials, and (2) the elastic stability of the crystals is not reflected by the signs of $C_{11}$ and $C_{22}$. Regarding (1), we acknowledge that $C_{11}$ and $C_{22}$ should be equal in these cases, but according to our calculations they deviate by 1.1%, 0.8%, and 9%, respectively. The average deviation for the 531 materials in C2DB with hexagonal or cubic symmetry is 1.2%, see figure 2. This is obviously due to numerics

**Figure 1.** The minimum eigenvalue of the full $3 \times 3$ stiffness tensor plotted against the smallest of the two uniaxial stiffness coefficients. Conclusions regarding the elastic stability is changed for the 36 materials in the grey shaded region. However, all of these except for the two materials indicated by red circles are dynamically unstable due to imaginary zone boundary phonons.



**Figure 2.** The two in-plane elastic constants, $C_{11}$ and $C_{22}$, plotted against each other for 531 materials in C2DB with either hexagonal or cubic crystal symmetry. Ideally, the two should be equal but due to numerical uncertainties in practice they are not. The three materials discussed in the text are highlighted. The mean relative deviation between the two components for all the materials is 1.2%.

as we also write in our original paper (page 9): 'for the isotropic materials $MoS_2$, $WSe_2$ and $WS_2$, $C_{11}$ and $C_{22}$ should be identical, and we see a variation of up to 0.6%. This provides a test of how well converged the values are with respect to numerical settings.' The deviation of 9% for $Re_2O_2$–FeSe is an outlier and we speculate that it arises due to the strong dynamical instability of this material (see below). We note that we could have decided to symmetrise the elastic tensors by hand such as to exactly reflect the symmetry of

the lattice. We have, however, refrained from such symmetrisation procedure because we believe it is relevant and more transparent to provide the raw rather than post-processed data. Similar considerations apply to many other quantities in C2DB. Regarding (2) we can essentially refer to the discussion in the first part of this paper. After calculating the full stiffness tensor for the three materials we obtain the same conclusions regarding the elastic stability of these materials as suggested in the Comment. However, as was the case for 99.5%

of the 378 test materials discussed above, irrespective of the stiffness tensor all three materials are correctly categorised as dynamically unstable in C2DB because they have zone boundary phonons with imaginary frequencies.

Despite the fact that only 0.5% out of the set of 378 materials are affected, we have decided to calculate the full stiffness tensor for all of the approximately 4000 materials currently in the C2DB. The full stiffness tensors for the 378 materials have already been made available in the C2DB, and data for the remaining materials will be available as soon as the calculations are done.

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## References

[1]  Maździarz M 2019 *2D Mater.* **6** 048001
[2]  Haastrup S *et al* 2018 *2D Mater.* **5** 042002