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Ideal adhesive and shear strengths of solid interfaces: A high throughput *ab initio* approach

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ABSTRACT

We release a computational protocol to calculate two intrinsic tribological properties of solid interfaces from first principles, namely the *adhesion energy*, γ, and the *ideal interfacial shear strength*, τ. These properties, which correspond to the energy required to separate two surfaces from contact and to the static friction force per unit area, respectively, are ruled by physical/chemical interactions between the surfaces in contact. First principles calculations based on Density Functional Theory (DFT) can accurately describe surface-surface interactions, offering the possibility to characterize the adhesive and shear strengths of materials *in silico*. We implemented the computational protocol as an AiiDA workflow (WF) that allows to obtain the γ and τ figures of merits in a high throughput manner. The software we produced uses a simple input file and most computational parameters determined automatically. To our best knowledge, this is the first time a high throughput approach has been used in tribology.

1. Introduction

In the late 20th century the invention of scanning probe techniques like the atomic force microscope [1], and the friction force microscope [2], boosted the field of nanotribology tremendously by providing means to measure single asperity friction [3]. Since classical continuum contact models may break down in nanoscale contacts [4], atomistic computer simulations where increasingly used to analyze the experimental data and make new predictions [5]. Ever increasing computer power also opened the way for parameter-free Density Functional Theory (DFT) simulations of chemical processes occurring at sliding interfaces [6–8], and to compute the adhesion and static friction properties of solid surfaces [9–13].

Even if most of the DFT-based works consider crystalline, infinite, flat surfaces, they do provide useful insights into the effects of the surface chemistry on the adhesive contribution to friction. This fundamental understanding is not only important for basic interface science, but also for practical application such as the design of lubricant materials. Results obtained by studying the interaction of idealized surfaces are, in fact, consistent with experiments and can explain experimental observations e.g. on the effects of passivating species on the tribological properties of diamond [14,15], of oxidation in MoS_2 [16], of iron passivation by graphene [17] and elements such as sulfur and phosphorus [18,19], which are contained in friction modifying lubricant additives.

While high throughput studies using DFT have become more common in recent years [20], a high throughput study of tribological properties of materials has not been attempted yet. In this paper, we present a computational protocol to calculate important tribological figures of merit connected to dry adhesive friction of solid interfaces with DFT methods. The protocol is implemented within the *Automated Interactive Infrastructure and Database for Computational Science* framework (AiiDA [21]) and is designed to be reliable and accurate while requiring only minimal user input. The philosophy behind AiiDA also ensures that the results are easily accessible in a queryable database which can be effortlessly shared. Furthermore the provenance of all calculations is preserved at all times and thus possible influences of computational parameters or pseudopotential choices are easily traceable even after considerable time has passed or after the person originally submitting the calculations is no longer associated with the project.

The workflow (WF) that we implemented in AiiDA calculates the

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following main quantities for the selected interface:

- The adhesion energy γ, which is the energy gained by mating two surfaces to form an interface. In the context of tribology this is a very important quantity, since high adhesion can lead to cold welding and high friction and wear.
- The ideal shear strength τ is the strength a material has before failing to shear stress. It is equivalent to the static friction force of a defect-free interface per unit area. The calculated τ should, then, be understood as an upper limit for the resistance to sliding along a certain direction. Therefore we refer to it as *ideal* shear strength.

In our WF we simulate shear as originally proposed in [10], where the ideal shear strength is obtained by the potential energy surface (PES) that represents the variation of the adhesion energy as a function of the relative lateral position of the two surfaces in contact. The adhesion energy corresponds to the minimum of the PES, while the shear strength is calculated from its derivative. This means that τ is calculated directly as an interfacial property, and not from the deformation of a bulk cell as previously done [22]. This has the advantage that the shear strength of heterogeneous interfaces and/or interfaces containing impurities can be calculated. The exact relation between this interfacial method of calculating the ideal shear strength, and the more common calculation via bulk properties, will be discussed in a forthcoming publication.

The obtained data for the adhesion and the ideal shear strength can be used as upper limits for real materials, which contain roughness, impurities, structure defects, and other imperfections. Nevertheless the knowledge of upper bounds for the capabilities of two contacting surfaces to resist detachment and shear forces is useful, e.g. for comparing different materials and choosing the most suitable for specific applications. The data can also be useful as physical input parameters for continuum mechanics models.

In Section 2 we will present our computational scheme in detail. Some illustrative results are then given in Section 3, followed by some concluding remarks in Section 4.

2. Description of the workflow

2.1. Summary of the workflow's capabilities

Fig. 1 offers a schematic representation of the WF, where the most important outputs, represented by green boxes, are summarized below.

1. The kinetic energy cutoff for the wavefunctions is converged to the selected accuracy using a very dense k-point grid.
2. The minimal k-point density able to satisfy the selected accuracy criteria is determined. This density is then used for all subsequent calculations to ensure minimal computational cost.
3. The lattice parameter is optimized using a simple bulk cell.
4. A surface slab is constructed according to the Miller indices provided by the user and the surface energy σ is calculated.
5. We construct the potential energy surface (PES) by displacing two slabs laterally (and relaxing them in the perpendicular direction) using high symmetry points for efficiency and then interpolating between them using radial basis functions (see Fig. 2). With the full knowledge of total energy as a function of lateral displacements, we are also able to identify the most likely sliding path, which connects the minima of the PES by traversing the lowest saddle points. This is known as the minimum energy path (MEP).
6. The perpendicular potential profile, $\gamma(z)$, is then obtained by displacing the two slabs normal to the interface plane (see Fig. 3). Thus

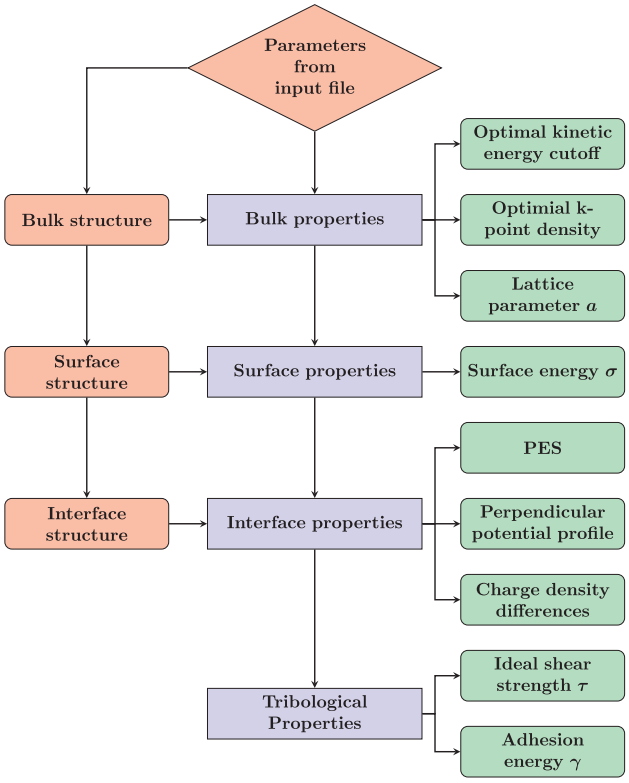

Fig. 1. Schematic representation of the WF. The input node is vizualized by the red diamond, structures are depicted as red rectangles, while blue and green rectangels represent the computations and output data, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

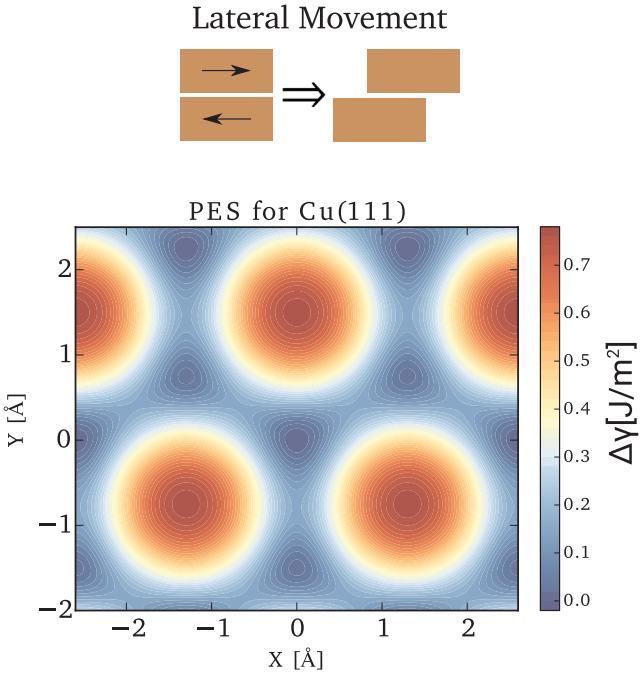

Fig. 2. Representation of the potential energy surface corrugation $\Delta\gamma = \gamma - \gamma_{min}$ and its connection to the lateral displacement of one surface relative to the other one.