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Surface stress: implications and measurements

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Abstract

Recent advances in the understanding of the role of surface stress for surface reconstruction, self-assembled pattern formation, alloying, shape and structural transitions, and adsorbate-induced stress are discussed. An understanding of the implications of surface stress requires a thorough theoretical investigation of the relevant processes on the atomic and mesoscale. Progress in the theoretical description of surface stress of clean and adsorbate-covered surfaces is described. A presentation of an optical deflection technique for stress measurements concludes the article.

Keywords: Surface stress; Adsorbate-induced stress; Surface reconstruction; Surface alloying; Self-organized pattern formation; Ab-initio stress calculations; Stress measurements

PAC Numbers: 62.20.-x; 83.85.St; 81.16.Rf; 68.35.Bs; 61.50.Ah; 71.15

1. Introduction and background

Surface stress has been identified as a decisive factor for the understanding of a wide variety of surface phenomena. As surface stress is nothing but a mechanical force acting in the surface region of any solid, its impact on the physical properties is essential where dynamical, structural and morphological issues are studied. However, its implications are not limited to these areas.

The role of surface stress has been discussed, e.g. for surface reconstruction, shape transitions in nanoscale particles, surface alloying, surface diffusion, epitaxial growth, and self-assembled domain patterns. It is expected that the inclusion of surface stress in the discussion of various physical phenomena will be of growing interest and relevance with the ongoing shrinkage of lateral and vertical dimensions of particles investigated in surface science, magnetism and materials science.

In contrast to the significant role which surface stress plays in the discussion of various aspects of surface science, experimental reports on its direct measurement are scarce. Surface stress measurements seem to originate from half a dozen labs worldwide.

This article offers first a brief account of the recent theoretical work devoted to surface stress, before a short overview of the correlation between surface stress and selected surface phenomena is given. A short description of a set-up for surface stress measurements concludes the article.

An excellent introduction to the role of surface stress in surface science is presented in the thorough reviews by Ibach [**1,**2], and a compilation of theoretical and experimental data as of 2001 is given in Ref. [3].

2. Ab-initio calculations of surface stress

Calculations of surface stress have been performed for rare gas crystals, ionic crystals, semiconductors, compounds, and metals, and are summarized in Ref. [3]. These calculations suggest an order of magnitude estimate of surface stress of 1 N/m, which corresponds roughly to an energy per surface atom of 0.6 eV. The large magnitude of this energy indicates that in view of the superior accuracy of present state-of-the art calculations, reliable values of surface stress should be obtainable by calculations. However, contact between calculated and experimental values of surface stress has not been established so far. The reason for this deficiency is the lack of reliable experimental data of the *absolute* value of surface stress, as discussed in Section 8. Presently, a comparison between theory and experiment is limited to the discussion of adsorbate-induced changes of surface stress. In cases where this comparison is possible, the agreement between theory and

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experiment is not always satisfactory. This is ascribed to many open questions, in both experiment and theory, which govern the atomistic and electronic origin of surface stress [**1,**4].

2.1. Clean surfaces

The reduced coordination of atoms in the surface layer vs. atoms within the bulk induces a corresponding redistribution of electronic charge. A well-known manifestation of the altered binding situation in the surface is the modified layer spacing (interlayer separation), which deviates in general from the bulk value (for most metal surfaces, the first layer spacing is slightly contracted). Within the surface plane, the atomic position remains at the respective bulk-like coordinates, as long as no surface reconstruction is observed. In the picture of a strain-dependent surface energy, this means that the bulk-determined intra-layer distance corresponds to zero strain, which however does not correspond to the minimum of the surface energy [5]. Fig. 1 presents a calculated surface energy vs. strain curve for Pt(111) [**4]. Such calculations can be extended to analyze the effect of adsorbate coverage, as shown in Fig. 1. At zero surface strain, the strain-dependent surface energy has a positive slope and this indicates a tendency of the surface to contract in-plane. This is the origin of the tensile surface stress which has been calculated for many systems [6].

An important aspect of the stress calculations is that a relaxation of atomic positions needs to be considered, as such relaxations are known to influence the result considerably. Unfortunately, some calculations, e.g. Ref. [7], neglected these relaxations, and the presented values should be viewed with caution [8].

The decisive role of structural relaxation for the calculated surface stress is evident from the discussion by Marcus et al. [**9], who calculated a surface stress for Mo(100) of 2.4 N/m^2, when they allowed for relaxation of all layer spacings in their slab calculations. Their previous calculation with one homogeneous layer spacing throughout the slab produced surface stress which was a factor two larger [10].

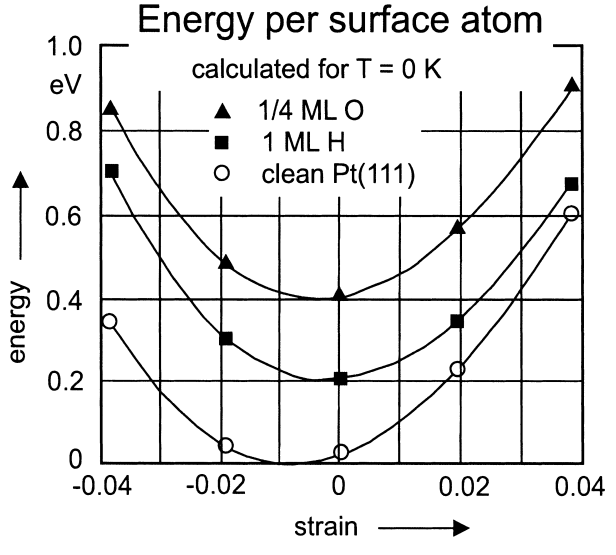

Fig. 1. Calculated energy change per Pt surface atom vs. surface strain. The data for H/Pt and O/Pt are displaced along *y* for clarity. Note that the slope of the curve is reduced due to adsorption at the position of zero strain, indicating an adsorbate-induced reduction of surface stress. Data from Ref. [**4].

3. Adsorbate-induced surface stress: experiment and theory

A compilation of the numerous measurements of adsorbate-induced surface stress changes and adsorbate–adsorbate interactions on metal and semiconductor surfaces is presented in Refs. [**1,3]. An illuminating discussion of adsorbate-induced changes of surface stress has been presented by Feibelman [**4]. Are the electronegativity, a work function change or the degree of filling of bonding and antibonding states the essential aspects which define adsorbate-induced surface stress changes? These and other aspects are discussed by Feibelman, and one has to realize that none of these effects alone can be identified as the sole driving force of surface stress.

Recently, Muller et al. have introduced another important aspect for consideration by pointing out that the spatial extension of electronic states may contribute to the stress via a direct orbital tensile interaction [11]. As a result, the role of charge transfer for the resulting surface stress change has to be reconsidered.

A proper discussion of adsorbate-induced surface stress requires, in addition to the stress data, reliable structural information on both the substrate and adsorbate. Recent calculations [8,12,**13] and experiments [**14,**15,16] suggest substantial adsorbate-induced structural distortions. Reliable models have to take these effects into account.

Up to now there is no complete understanding of the electronic origin of adsorbate-induced surface stress changes. This calls for further calculations and experiments to elaborate the relevant principles.

4. Surface stress and surface reconstruction

The existence of surface stress is correlated with the strain dependence of the surface energy as discussed above and shown in Fig. 1. A positive slope of the energy curve

reveals directly the tendency of the surface to increase the surface atom density, i.e. tensile surface stress should favor a surface which is more densely packed. This argument suggests surface stress as a driving force for surface reconstruction where the surface atom density is increased [5,6]. Indeed, the surface atom density is increased for several surface reconstructions like the herringbone reconstruction of Au(111) or the quasi-hexagonal 5×1 reconstruction of f.c.c. (100) surfaces.

The role of surface stress in reconstruction has been discussed by Ibach [**1,**2] and Cammarata [17]. A parameter in proportion to the difference between surface stress and the specific surface free energy, i.e. the strain dependence of the specific free surface energy, also called *excess* surface stress [**2], was introduced as an indicator for the driving force for the surface to reconstruct. This parameter indicates the surface reconstruction of Au(111) and Pt(111), but it fails to predict the 1×5 surface reconstruction of Ir(100). This last point deserves a comment, as the original publication [7] quoted a large value for the difference between surface stress and specific free energy, whereas a more recent publication by the same author gives a negligible difference between the two quantities [8,18]. This leads to the conclusion that the Ir(100) 5×1 reconstruction is *not* driven by surface stress [**2].

Surface stress changes which accompany surface reconstruction have been measured e.g. for Au(111) and Au(100) [*19]. The authors considered the energy gain due to surface stress relaxation and they concluded that the reconstruction of Au(100) is definitely not driven by surface stress relaxation, but the reconstruction of Au(111) might be [**2,*19].

The calculation of surface properties of the large $22\times\sqrt{3}$ unit cell of the Au(111) herringbone reconstruction involves too many atoms for present ab initio studies. It is expected that for larger unit cell systems, surface properties get more and more successfully tackled by ab initio-based molecular dynamics calculations [13,**20], which elucidate structure and stress on the atomic scale.

Surface stress has been proposed recently in a theoretical study as the driving force for the 1×4 reconstruction of TiO_2 [21]. This supports the notion that the concept of stress-driven reconstruction is of general validity, and it is not limited to metals and semiconductors.

It is worth pointing out that surface-stress driven surface reconstructions are not only of fundamental interest, but they also offer templates for self-organized growth. The preferred nucleation of Co in the elbows of the Au(111) herringbone reconstructions has been exploited to grow regular arrays of magnetic Co nano-columns [22]. The implications of the resulting self-organized pattern formation, to be discussed next, for the preparation of high density nanomagnetic structures have been reviewed by Oepen and Kirschner [*23].

5. Surface stress and self-assembled pattern formation

An aesthetically appealing [24,25] and technologically important [26] aspect of surface stress is its role in the self-organized ordering of nanostructures on mesoscopic length scales (several nm–100s of nm). Elastic interactions between stress domains mediate an ordering interaction which often results in the formation of regular self-assembled patterns on a length scale below standard lithographic techniques. A comprehensive review with emphasis on semiconductor technology is given by Shchukin and Bimberg [*27].

The essential aspect of the theoretical description of self-assembled pattern formation [*28,*29] is that the difference of surface stress between adsorbate-covered and free regions always favors the formation of stress domains, i.e. the ordered patterns represent areas of equal surface stress.

The discussed role of surface stress for pattern formation raises the question as to whether the surface stress or its variation over mesoscopic length scales could be measured directly. The cantilever-bending technique, to be described below, integrates over a macroscopic sample area (≈10 mm^2), and in general the stress variation on the mesoscale is expected to be averaged out in the measurements. However, in some cases the domain patterns are anisotropic, e.g. stripe domains, and proper sample orientation and two-axial curvature measurements could identify the stress anisotropy. An example is the surface stress anisotropy which has been measured successfully during growth of III–V compounds [30]. Stress measurements on systems with stress domains other than the Au(111) herringbone reconstruction have not been performed yet. Such measurements are of high relevance as they have the potential to deliver quantitative stress values.

An example of a chemisorbed self-organized pattern is the formation of checker-board patterns for the adsorption of N on Cu(100), where the surface is composed of patches of bare Cu and N-c(2×2) covered areas. A recent surface X-ray diffraction structural analysis supplemented by molecular dynamics and continuum elasticity calculations of structural relaxations suggested a surface stress difference between N-covered and free Cu(100) surface patches of 7 N/m [**15]. This is the first example which demonstrates that the correlation between a detailed atomic structure determination and atomic scale calculations offers a promising approach to *deduce* surface stress. Note however, that a model for the atomic interactions has to be invoked to deduce stress from a structure analysis. Recently we have combined in-situ surface X-ray diffraction measurements with curvature measurements for stress analysis. Thus, structure–stress relations are in principle directly accessible by experiments [31].

The elastic anisotropy, see e.g. [32,*33,34], has been

proposed as an important factor which determines the orientation of nanoscale objects or patterns on crystalline substrates. Lu and Suo suggested that, e.g. nano-stripes tend to align along compliant, i.e. elastically soft, directions [*35].

6. Surface stress and alloying

The role of elastic relaxation in surface layers has been discussed recently in view of its implications for surface alloy formation [36–39]. Burrowing of nanoparticles, i.e. the sinking of Co nm-sized particles in a Ag(100) substrate has been reported [40]. Both effects, surface alloy formation and burrowing of particles, are expected to change the surface stress of the substrate significantly. Respective stress measurements have not been performed so far, but they are expected to clarify the role of elastic relaxation and capillary forces as driving force of the intermixing.

7. Surface stress and its effect on shape and structural transitions

Shape fluctuations of 2-dimensional islands have been analyzed to derive step-free energies [41]. In recent studies it was suggested [42,43] that surface stress anisotropy should also be considered as a factor which influences island shape. Middel et al. [*44] have taken this approach to derive the anisotropy of surface stress on Ge(100) from an analysis of the shape of vacancy islands. The shape analysis of 2-dimensional nanostructures might be a promising way to deduce surface stress anisotropies in such cases, where a direct measurement of the surface stress anisotropy is not feasible.

Surface stress oscillations during epitaxial growth have been measured recently [16]. These measurements offer quantitative stress values which are important benchmarks for theoretical modeling of stress-induced structural and morphological changes. For Co on Cu(100) a monolayer oscillation of the elastic energy by 1 meV per surface atom was measured. Atomic scale stress calculations identified structural relaxation in islands as the driving force for the stress oscillations [16].

The in situ combination of scanning tunneling microscopy and stress measurements by the cantilever technique proves very promising in revealing the correlation between epitaxial growth mode and resulting stress. A recent example is the transition from Stranski–Krastanow to Vollmer–Weber growth with increasing Si-content of SiGe alloys, deposited on Si(100). The former induces a large compressive stress at the interface, the latter leads to an almost stress-free interface [**45].

With increasing film thickness one leaves the region of surface stress and enters the field of film stress [**1,32]. This is not the topic of this contribution, but it is nevertheless of current research interest, and further details on the correlation between film growth mode and stress in thin films are given in Refs. [*33,46–49].

8. Experimental techniques to measure surface stress

It is remarkable that there seems to be no experimental technique which measures surface stress of clean surfaces *directly*. The reason is that the mere existence of a force within a surface layer of a material has proved elusive for measurements up to now. The effect of surface stress on another measurable quantity, like surface phonons is clearly anticipated. A quantitative value of surface stress derived from phonon dispersion data requires model calculations, which render surface phonons as an indirect source for surface stress data.

The concept of surface stress implies that work has to be done against surface stress when straining a solid. In thin samples, the surface stress contributes significantly to the elastic response of the sample, and deviations between measurements on thicker vs. thinner samples can be exploited to derive the surface stress. Muller and Kern suggested an experimental approach in which the bending of a thin circular disc in the gravitational field was measured [50]. The resulting deflection depends on sign and magnitude of the surface stress. Such experiments have not been performed so far, however it is suggested that progress in both micro-machining and finite-element-modeling of bending makes these experiments feasible.

On nanoscale particles, surface and interface stress may lead to a measurable change of the atomic structure as compared to large specimens. Electron diffraction experiments have been performed on nm-size particles, and the results are briefly discussed in Ref. [3]. The shortcoming of this approach is that the elastic properties of the particles have to be described in a model, and the result of the calculated surface stress is expected to depend critically on the assumed 3-dimensional shape.

8.1. Measurements of changes of surface stress

The examples above have shown that although the absolute value of surface stress is related with measurable quantities, its model-free determination remains a challenge.

By contrast, *changes* of surface stress due to adsorption can be measured quantitatively from the change of curvature of a thin substrate upon adsorption. The so-called bending beam technique as shown in Fig. 2 is used successfully to measure adsorbate-induced changes of surface stress of metal and semiconductor surfaces with high precision and sensitivity. The data analysis, the effect of substrate clamping and elastic anisotropy on the curvature are discussed in Ref. [3,51–53].

The two-beam technique of Fig. 2 offers the advantages

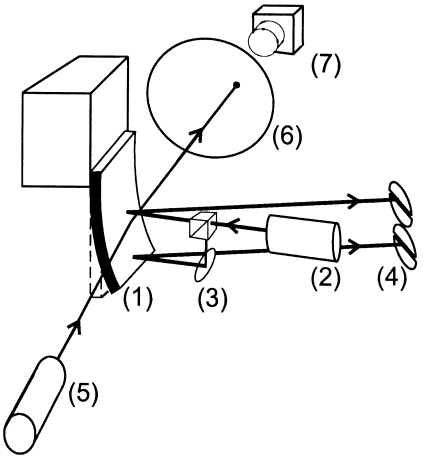

Fig. 2. Two-beam curvature measurement. An adsorbate-induced stress change on the surface leads to a curvature of the thin substrate, which is detected by laser deflection. The sample (1) is located inside a vacuum chamber, the curvature measurement tools ((2)–(4)) are outside under atmospheric pressure. (1) Sample, (2) laser, (3) beamsplitter, (4) two position-sensitive photo detectors, (5) electron gun, (6) low-energy-electron-diffraction (LEED) screen, (7) CCD camera. The inclusion of intensity measurements of diffracted electron beams ((5)–(7)) allows to correlate stress with layer filling during epitaxial growth [16].

of (i) a direct measurement of the curvature from the measured difference of the two position signals, and (ii) improvement of the signal-to-noise ratio, as vibrational noise, which affects both beams, is largely eliminated by the difference measurement [34,54].

8.2. Sensitivity of cantilever bending experiments

The magnitude of adsorbate-induced changes of surface stress depends on the adsorbate–substrate system under investigation. A typical order of magnitude is 1 N/m for a coverage of one adsorbate per surface atom. Under careful experimental conditions, stress changes as small as 0.01 N/m have been measured with the set-up shown in Fig. 2. This translates into a coverage sensitivity of the order of 1% of a monolayer. The corresponding variation of the energy per surface atom is of the order of meV. This estimate shows that even subtle stress variations which are described on a meV energy scale can be detected. Examples of such subtle effects are the H-induced stress in epitaxial Ni-films on Cu(100) [55], and the stress-relaxation in nm-size islands during epitaxial growth [16].

The signal-to-noise ratio of the stress measurement has been improved by three orders of magnitude if the stress-inducing effect can be modulated. This has been done for measurements of the magnetization-induced stress in monolayers, where the magnetoelastic coupling constants are derived from the curvature measurement [32,56]. Here, the film magnetization is flipped periodically between two well-defined states, and signal averaging or phase-sensitive detection schemes lead to an improved signal-to-noise ratio.

9. Conclusion and outlook

The role of surface stress for a large variety of different surface phenomena has been briefly described. The most striking deficiency in the experimental work is the lack of abundant data from direct stress measurements. It is hoped that this article might trigger more experiments devoted to this aspect. The exciting and promising possibilities of self-organized pattern formation on the nanoscale require theoretical modeling and experimental data which include both structure and stress information from the atomic to the mesoscale (hundreds of nm). Ab-initio based molecular dynamics simulations might be a promising tool for handling the large number of atoms which form the nanoscale patterns.

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