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Concept for support and cleavage of brittle crystals

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We report on sample holders for crystals to be cleaved for the preparation of surfaces with large atomically flat terraces. The concept for mounting sample crystals is based on a vicelike clamping mechanism to securely hold the crystal in position while reducing the risk of fragmentation. Sample holders based on this concept and made of suitable materials allow preparation and cleavage of crystals in the ultrahigh vacuum at high or low temperatures. To cleave the crystal, we employ a scalpel blade mounted on a wobble stick to generate a highly localized stress field initiating the cleavage process. The sample holders are used for experiments of highest resolution scanning force microscopy, however, the concept can be transferred to any other system where cleavage faces of crystals are of interest. Exemplarily, scanning force microscopy results demonstrate that (111) cleavage faces of CaF_2 crystals can be prepared with steps only a few F–Ca–F triple-layers high and atomically flat terraces extending over areas of several μm^2. © *2009 American Institute of Physics.* [DOI: 10.1063/1.3152367]

I. INTRODUCTION

The unparalleled strength of scanning force microscopy (SFM) is the possibility of imaging insulating surfaces. In this context investigated samples are often brittle single crystals. For a wide range of materials, among them fluorides and other halides, high quality surfaces can be prepared by cleavage along low energy lattice planes. To accomplish this, it is necessary to mount the crystal on a sample holder that allows precise cleavage and avoids other damage to the crystal. The crucial part in this is the fixing of the sample crystal on the sample holder: the crystal needs to be well fixed but must not be squeezed to avoid uncontrolled cleavage or breakage. Furthermore, the crystals should have a large area contact to ground to drain off charges resulting from cleavage. Often, a further requirement is compatibility with low or high temperature treatment during preparation and measurement under ultrahigh vacuum conditions. Single crystal samples are most commonly cuboids with rectangular or quadratic profiles or rods that should all be well supported by the sample holder.

Often, brittle sample crystals are attached to the sample holder with a conductive glue on the basis of epoxy resin and the glue is smeared over the sides of the crystal to improve electrical contact. This type of attachment is easy to apply and provides satisfying results in certain cases. However, if the glue has been smeared over the breaking edge, cleavage will also break the glue. Thereby undefined constituents of the glue are inevitably deposited onto the freshly cleaved plane. Furthermore, there is no epoxy resin on the market that is specified for use at low temperatures and under ultrahigh vacuum conditions, nonetheless some resins are empirically used in this field of application. Likewise, the glue is a constraint for heating the sample to high temperatures.

To resolve this problem and prevent pollution of freshly cleaved surfaces, sample holders with a mechanical clamping mechanism have been developed.[1] The mechanical design of such a clamping mechanism needs to follow the requirements of the experimental setup. The sample crystal must be as firmly attached to the sample holder as possible, not only during measurements but also to allow high quality cleavage with devices inside an ultrahigh vacuum chamber. This must be achieved with a clamping pressure as low as possible to reduce strain inside the crystal which would lead to defects and breakage. Therefore, the clamping force must be applied as evenly distributed over the contact area as possible.

To achieve this, we devised a universally applicable vicelike mechanism to hold the sample crystal without any glue. As we operate several different types of UHV force microscopes operated in the noncontact mode (NC-AFM) we designed sample holders for use in a RHK VT-AFM 750 (system 1),[2,3] for use in a homebuilt LT-AFM (system 2) based on the design presented in Ref. 4, and for use in an Omicron VT-AFM 25 (system 3).[5] To obtain highest quality cleavage faces, we implemented a subtle cleaving procedure based on scratching the crystal and initiating cleavage by the tip of a sharp blade.

II. SAMPLE HOLDER FOR SYSTEM 1

The sample holder for the scan head of system 1 consists of a cylindrical copper base body with moldings for an *in vacuo* transfer system and slopes for the approach movement of a beetle type scan head (Fig. 1). Between these two components, the sample support is installed on a stainless steel plate.

The sample crystal is placed inside a metal pouch bolted to the inlay of the sample holder. Three screws provide not only enough clamping force to secure the crystal but also enable its exact distribution so that a minimum strain is

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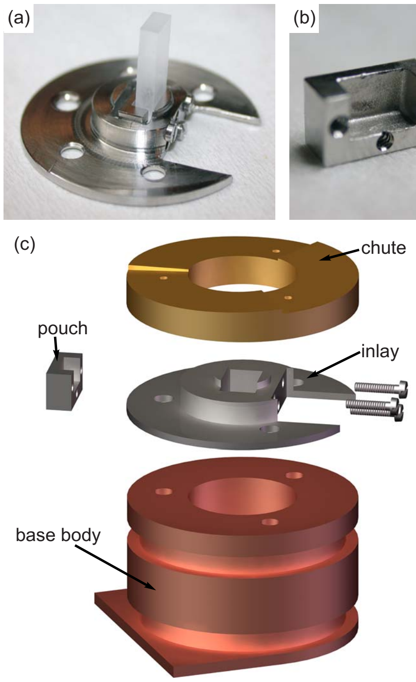

FIG. 1. (Color online) Sample holder for system 1. (a) Inlay plate with incorporated clamping mechanism. (b) Steel pouch for sample crystals. The three screws facilitate precise force adjustment. (c) The inlay with the clamping mechanism is assembled between the copper base body of the sample holder and the chute for the approach of the beetle type scan head.

generated inside the crystal. By the use of three screws and a metal pouch, any contortion of the sample during cleavage can be avoided.

As the space inside the slopes of the sample holder is limited, a more complex clamping mechanism cannot be installed without reducing the sample size unacceptably. The present design accommodates crystals with a profile of $2 \times 4\ \text{mm}^2$.

III. SAMPLE HOLDER FOR SYSTEM 2

The sample holder for system 2 is based on a simple base plate providing enough room for a more complex clamping mechanism consisting of a vicelike configuration of two chevrons holding the sample crystal (Fig. 2). One chevron is welded to the base plate, the other one is movable to allow for the clamping force to be applied diagonally through the crystal. The inner angles are routed so that the edges of the sample crystal are freestanding and the clamping force is transmitted through the side faces.

The clamping force is generated by a screw which is threaded through a metal plate that is welded upright onto the base plate. Bending of this plate allows to build up a spring force that ensures a firm clamping even if the sample holder is heated to high temperatures and thermal expansion will change dimensions of parts of the sample holder.

The clamping force is transmitted from the screw to the mobile chevron through a ruby ball. To keep this ball in position, dimples have been milled into the tip of the screw and the back plane of the mobile chevron. The transmission of the force through a hard sphere ensures that the mobile chevron can adapt to the position of the sample crystal without being contorted by turning the screw. The chevron welded to the base plate prevents contortion of the sample crystal during cleavage. The mobile chevron is dimensioned so that its lower plane solidly rests on the base plate to further increase lateral stability. Due to the symmetric configuration of the chevrons, sample crystals with a quadratic as well as circular profile can be used.

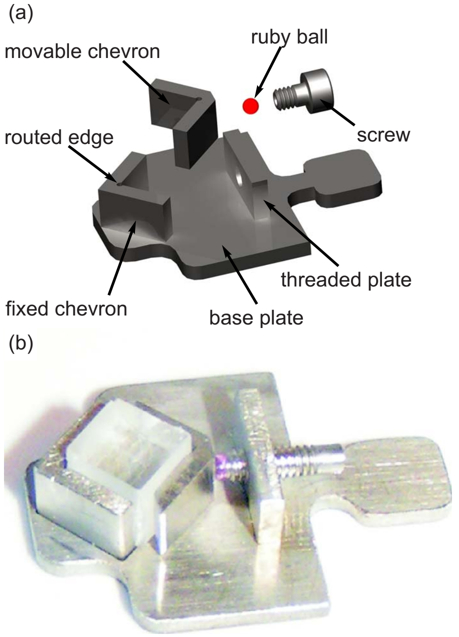

FIG. 2. (Color online) Sample holder for system 2. (a) Design drawing showing the clamping mechanism with a fixed and a movable chevron with routed inner edges. The clamping force is generated by a screw threaded through a spring plate. (b) Photograph of a crystal mounted in the sample holder.

For various applications the sample holder can be manufactured from different materials, e.g., aluminum for low temperature or tantalum for high temperature operation.

The size of crystals that can be used in this sample holder must exceed the length of the side panels of the chevron and is limited by the distance between the fixed chevron and the upright threaded plate. Our current sample holders are either made of stainless steel or aluminum and can accommodate crystals with side lengths or diameters between 3 and 4.5 mm. The sample holders have been used to heat crystals up to 650 K to remove excess charges due to cleavage.

IV. SAMPLE HOLDER FOR SYSTEM 3

The principles of the above described design have also been employed to build a sample holder for system 3 (Fig. 3), which was additionally modified to accommodate sample crystals with a rectangular profile.

The standard sample holder for this system consists of two parallel plates. One is used to mount it in the *in vacuo* transport system, the other one serves as thermal insulation when placed inside the scan head. Of these two only the lower one can be used for mounting the clamping mechanism whereby the chevrons must be high enough to bridge the gap between the two plates. The top of the chevrons should not be lower than the upper plate of the sample holder

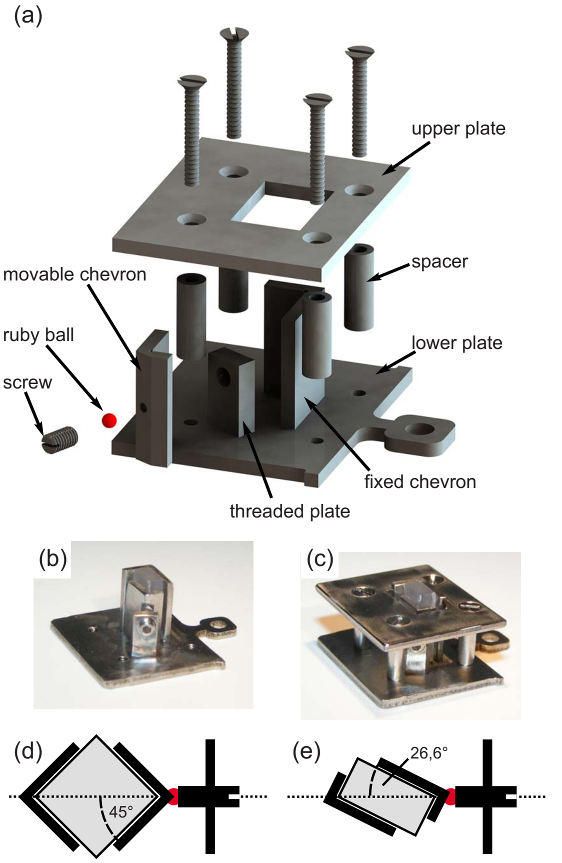

FIG. 3. (Color online) (a) Sample holder for system 3 consisting of two parallel plates. The clamping mechanism is mounted onto the lower plate using the height between the two plates for secure sample holding and large area electrical contact. (b) Clamping mechanism mounted on the lower plate. (c) Assembled sample holder. (d) Line of force in a sample holder for samples with a quadratic profile. The chevrons are tilted by 45°. (e) In order to apply the clamping force through two opposite edges of crystals with a rectangular profile the chevron must be tilted by an angle of $\alpha = \arctan(\text{shortside}/\text{longside})$ with respect to the line of force. This amounts to 26.6° for a crystal with 1:2 aspect ratio.

because the cleavage plane of the crystal will be most likely parallel to the top of the chevron and thus could be not reachable for the SFM tip.

For some very brittle samples it was found to be more convenient to use crystals with a rectangular basis with an aspect ratio best chosen to be 1:2. Furthermore, if rectangular crystals are cut with the same orientation with respect to the surface crystal lattice the orientation of the crystallographic directions will be the same for all measurements. To exert the clamping force diagonally through the crystal, the alignment of the chevrons on the base plate is tilted by an angle $\alpha = \arctan(\text{shortside}/\text{longside})$ with respect to the line of force [Fig. 3(e)]. This results in an angle of 26.6° for a crystal with an aspect ratio of 1:2.

Built out of stainless steel these sample holders are used in variable temperature experiments between room temperature and 35 K after heating samples up to 550 K for discharging.

V. *IN SITU* CLEAVAGE

Cleavage of single crystals in the ultrahigh vacuum is a simple method to produce clean and well ordered surfaces with a small number of steps with a limited step height.[7] For several years we have been interested in perfect cleavage faces of CaF_2 to explore their atomic structure[8] and morphology[9] as measured by NC-AFM, compare it to theoretical predictions,[10,11] and use them, for instance, for molecular manipulation experiments.[12] In a series of trials to obtain perfect surfaces, we could dramatically improve the quality of cleavage faces.

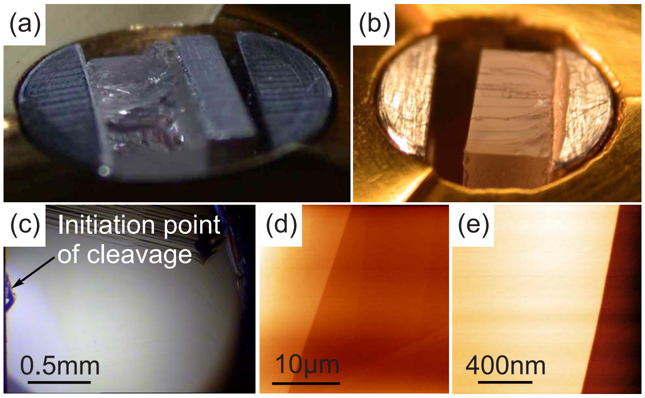

FIG. 4. (Color online) CaF_2 crystals cleaved along the (111) lattice plane. (a) Crystal cleaved by exertion of blunt force. The crystal was clamped by a mechanism not preventing torsion. (b) Crystal cleaved by exertion of blunt force after a predetermined breaking point was generated by scratching with a scalpel blade. (c) Optical microscopic view of a crystal cleaved with the tip of a scalpel blade. The tip was scratched over the left side plane while the contact pressure was slowly increased until the crystal broke. The cleavage was initiated on the left side. (d) Magnified view of the crystal surface shown in (c) taken with contact mode force microscopy. The largest step has a height of three F–Ca–F triple layers. On the lower terrace, triple-layer steps are barely visible. (e) NC-AFM image taken in UHV showing atomically flat terraces extending over 1 μm^2.

A major finding is that high cleavage quality can quite easily be obtained by cleaving small samples while cleavage of large samples often results in very rough surfaces with many high steps. Therefore, we reduced the sample size from initially 10×10 mm^2 to 2×4 mm^2. Furthermore, the way of clamping the crystal may strongly influence the cleavage result. In Fig. 4 we document the step-wise improvement yielded during the development of our sample holder design.

At first, brittle crystals were cleaved by applying blunt force using the crystal as a large lever arm. The resulting surfaces were mostly extremely rough, as shown in Fig. 4(a). The more precise cleaving is executed, the more rare are disturbances in the surface structure. Results could greatly be improved by specifying the predetermined breaking point by carving chamfers at the appropriate height into the sides of the crystal. However, the distribution of the breaking force over a relatively large area results in strain lines of macroscopic steps in the surface, as shown in Fig. 4(b).

We found that cleavage results can greatly be improved if the area of application of the force is reduced. We accomplish this by using extremely sharp and hard steel blades of scalpels normally used for sample preparation for biological experiments. To cleave the crystal, the tip of the blade is scratched over the side plane of the crystal along the edge of the pouch or chevron, ever increasing the pressing force until the breaking point is reached and the crystal is cleaved. Cleavage is induced by introducing a highly localized stress

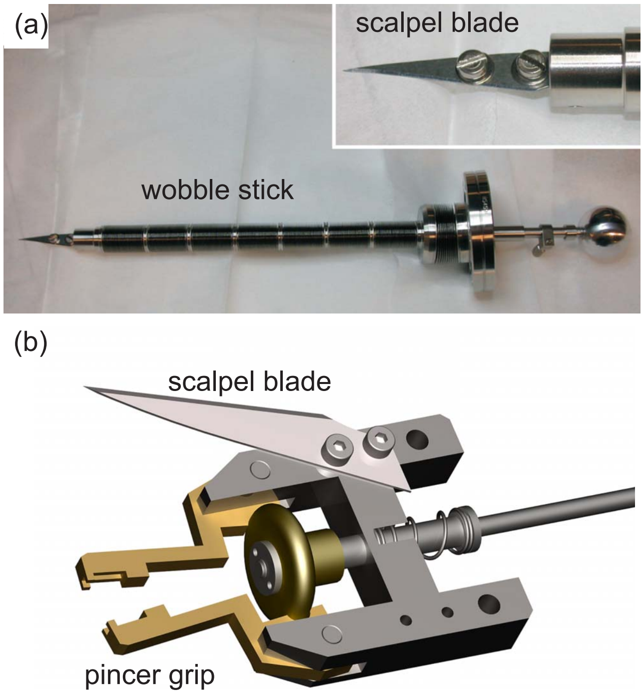

FIG. 5. (Color online) (a) Wobble stick with attached scalpel blade. (b) Drawing of a scalpel blade attached to the frame of the pincer grip of the wobble stick (Ref. 6) which is part of an *in vacuo* transfer system.

field acting at one point and breaking the crystal along a well defined lattice plane. Applying this procedure, it is straightforward to reproducibly generate surfaces with a small number of steps with a low height and very large atomically flat areas, as documented in the optical and scanning force microscopy images shown in Figs. 4(c)–4(e). Using one of the clamping chevrons as a guide rail is extremely useful for beetle type microscopes having a very small vertical positioning tolerance due to the limiting height of the chutes used for coarse approach.

We mount the scalpel blade on wobble sticks either installed solely for this purpose [Fig. 5(a)] or already installed as part of an *in vacuo* transfer mechanism. In the latter case, the scalpel blade can, for instance, be mounted on the frame for the pincer grip that is used for the transfer of tip and sample holders in system 2 [Fig. 5(b)]. As a wobble stick provides a high degree of maneuverability and assuming the given microscope provides enough travel range with the sample coarse approach, it is also possible to cleave a crystal several times if one does not use a guide rail.

After cleavage, the surface may be charged what hampers or even prevents measurements with the force microscope or electron emission based surface analysis techniques. Therefore, cleaved CaF_2 crystals are heated for at least 1 h to 120 °C to increase the mobility of charge carriers and speed up discharge.[13] Experience from many preparations shows that heating with insufficient power even for a prolonged period of time will not remove the charge while excessive heating may result in irreversible surface modification and destroy the crystal. The temperature should be slowly ramped up and down to avoid excessive thermal stress. We have transcribed the preparation procedure to KBr crystals, finding it necessary to increase the heating temperature up to 180 °C. When following this heating procedure, a clean KBr(100) surface can be obtained as an exception even by cleavage in air and rapid transfer into the UHV chamber. It was further found that during experiments performed on KBr, ion gauges and other electron emitting devices in the UHV chamber should be turned off, as electrons reaching the surface may cause radiation damage.[14]

VI. CONCLUSION

We developed sample holders for brittle sample crystals allowing to mount the crystal without using epoxy resin but by a mechanical clamping mechanism. Cleaving of the crystals is performed by scalpel blades mounted on wobble sticks and yields very large atomically flat terraces.

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