Trends in the development and application of the nanoindentation method

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Abstract

One of the most important trends now is the application of nanoindentation for fundamental investigations of elementary mechanisms of mechanical deformation under very high pressure in the local contacted region. Representative examples are given.
We have studied in detail the homogeneous generation of dislocations at room-temperature by nanoindentation in locally dislocations-free monocrystals. The discontinuity of load-penetration depth-curve referred to as Pop-in-effect is the result of the nucleation of the first dislocation loops and subsequent drastic plasticity response of the material by indentation due to multiplication processes.
The mechanical stresses responsible for this process were calculated in the framework of elastic contact theory (Hertz, Sneddon). The measured critical stresses for loop nucleation are in good agreement with theory of dislocations within the isotropic approach. Corresponding dislocation loops were proved by means of microscopy imaging techniques (transmission electron microscopy (TEM), cathodoluminescence imaging (CL), and imaging of dislocation-etched surfaces.

1. Trends of High Pressure Physics by Nanoindentation

Advanced depth-sensing hardness measurement equipment makes it possible to investigate mechanical properties of solids under high local stress at nanometer scale. The contact stresses induced in this manner are only limited by the local strength of the material investigated.

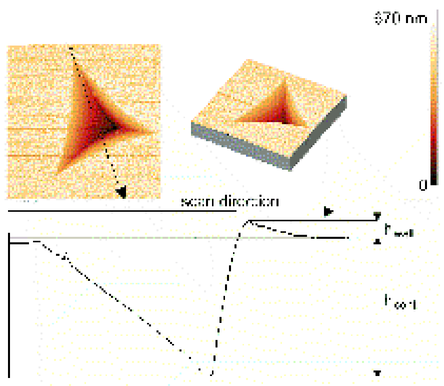


Fig. 1

Representative examples of the efficiency of high contact stresses will be given for amorphous and crystalline materials:

1.) It is well accepted that glass is plastic–viscous deformable only above the glass temperature T_G and glass is ideal brittle below T_G. We could analyze the velocity dependence of nanoindentation experiments at room temperature taking into account the viscosity reduction induced by the hydrostatic part of the contact stress [1,2,3]. The detection of wall formation, so called pile-ups, at nanoindents in glassy materials is a demonstrative evidence of high-pressure induced viscosity reduction. Enders [4] could show in Fig. 1 the wall formation at the border of crack–free indents for Na–Silicate glass (T_G = 535 °C) by AFM, integrated to the Nanoindenter II equipment. Also scratches, affected in glass at RT by nanoindentation, show pile-ups by AFM imaging [4].

2.) The wall formation in glass is connected with the occurrence of viscous shear bands [5]. The cutting of the shear bands in the viscosity reduced contact region leads at higher loads (nearly 1N) to formation of virgin cracks, the so called radial cracks. The frequency of the radial crack formation in dependence on the indenter load is a sensitive indicator for surface stresses [6].

3.) High-pressure induced phase transformation for example in monocrystalline Si could be detected by Kailer, Gogotsi et. al. [7] preferred as pop-outs in the unloading part of nanoindentation (Fig. 2 in [8]). The refinement of phase transition in Si monocrystals could be analysed in detail in combination with Raman scattering. Ultimately 7 modifications by Raman - Spectroscopy of Nanoindents could be detected [6].

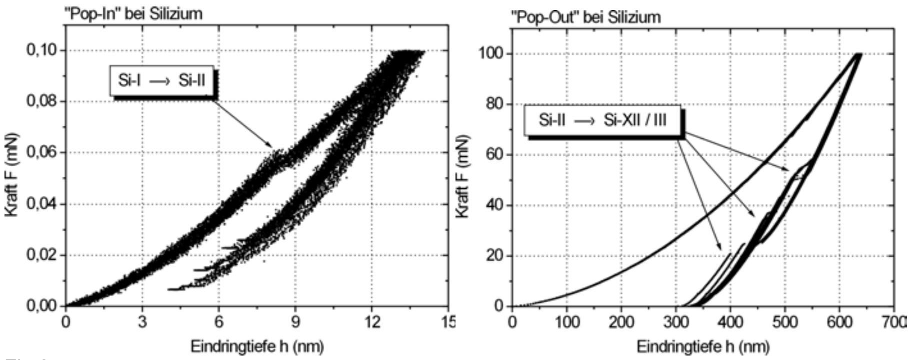


Fig.2

4.) Using the nanoindentation method for crystalline material where the critical load of phase conversion is higher than that of plastic strength one can observe the embryonic process of homogenous nucleation of dislocations, leading to one or more pop-ins in the loading curve. Firstly in 1997 Corcoran, Colton, Lilleodden and Gerberich [9] published a rapid communication "Anomalous plastic deformation at surfaces: Nanoindentation of gold single crystals", following by Michalske et. al. [10] and Bahr et. al. [11]. In the present paper we will concentrate on this specific effect and will look for the definite conditions of dislocation nucleation.

2. Theoretical Conditions limiting the High Pressure dislocation formation and their application on the nanoindentation method

- The HERTZ / SNEDDON calculation:

The deformation behavior before the pop-in is a pure elastic one. The load–penetration depth-curves recorded in loading and unloading direction are extremely congruent [8]. No permanent deformation could be detected by topographical inspection of the contact area after unloading. The manufacture restrictions which affect indenter rounding (at radiuses greater than 100 nm) leads to the growth of contact stress up to GPa values in the absence of plasticity carrying dislocations. Therefore this part of indentation experiment is specified for analyzing on the background of the well established elastic contact theories given by Hertz and Sneddon (summarized in [13]). Both theories lead to a power law between load F and penetration depth h. Following equations will be used:

$$\text{tip rounding}: z = c \cdot r^m \Rightarrow \quad \text{Sneddon}: F = B \cdot h^n \; ; h = b \cdot h_C \qquad (1)$$

$$n = \frac{m+1}{m}; \quad b = \frac{\sqrt{\pi} \cdot m \cdot \Gamma\left(\frac{m}{2}\right)}{2 \cdot \Gamma\left(\frac{m+1}{2}\right)}; B = 2E_{red} \frac{m}{m+1} \left(\frac{1}{c \cdot b}\right)^{1/m} \qquad (2)$$

r means the distance from the axis z of the body of revolution, h_C is the real contact depth between indenter and sample and E_{red} is defined by equation (5).

- The TRESCA – stresses:

The expression for the stress producing a stable dislocation loop τ_{crit} is obtained as a result of theoretical predictions for elastic energy of a dislocation loop in isotropic media [9]. For the case of dislocation nucleation it can be written as follows, omitting the thermal terms:

$$\tau_{crit} \approx \frac{G \cdot b}{2\pi \cdot r_C} \approx \frac{G}{10} \qquad (3).$$

Here G is the shearing modulus considered to be isotropic, b is the contribution of Burger's vector and r_c is the radius of the nucleated loop.
The maximum shearing stress (Tresca's stress) below the indenter τ_{Tresca} can be expressed through the indentation force and penetration depth at known tip radius of the indenter or Young`s modulus of the material, as it follows from Hertz contact theory:

$$\tau_{Tresca} = 0.465 \cdot p_m = 0{,}465 \cdot \frac{16}{9} \frac{E_{red}^2 \cdot h^2}{\pi \cdot F} = 0.465 \cdot \frac{4 \cdot E_{red} \cdot \sqrt{h}}{3 \cdot \pi \cdot \sqrt{R}} \qquad (4).$$

Here p_m is the mean contact pressure, h is the penetration depth and F is the corresponding indentation force. The reduced elastic modulus E_{red} consists of Young`s modulus and Poisson's ratio of the sample and indenter, i.e. E and ν, and E_{Ind} and ν_{Ind} (diamond: E_{Ind}=1141GPa, ν_{Ind}=0.07), respectively.

$$\frac{1}{E_{red}} = \frac{1-\nu^2}{E} + \frac{1-\nu_{Ind}^2}{E_{Ind}} \qquad (5)$$

The homogeneous dislocation nucleation is possible only when there are no movable dislocations within the catchments area of the stress field.
The probability number N of the dislocations likely to be activated before the set up of the Pop-In-effect can be obtained in the course of approximation by multiplying the maximum surface area of the contact between indenter and material surface , reachable before the set up of the pop-in effect by the dislocation density of the material.

$$N = \rho_{Dislo} \cdot \pi \cdot R \cdot h_{Pop-In} \qquad (6)$$

Here R is the tip radius of the indenter and h_{Pop-In} is the penetration depth, by which the Pop-in-effect appears. For the numbers N<<1 the activation of the dislocation yet before the Pop-in-effect is most improbable, in other words, the occurrence of the pop-in effect is most probable.
Assuming the approximate ratio between shearing modulus G and Young's modulus E in the first order of approximation as $E \approx 3 \cdot G$ and substituting the maximum reachable shearing stress value obtained from the Equation (3) into the Equation (4), yields the penetration depth of the Pop-in effect:

$$h_{Pop-In} \approx 0.03 \cdot R \qquad (7).$$

As it can be seen, within the approximation used the penetration depth, by which the Pop-In effect appears does not depend on the material parameter.
As a result of this the number of the dislocation likely to be activated before the Pop-In effect can be written down as follows:

$$N \approx 0.09 \cdot \rho_{Dislo} \cdot R^2 \approx 110 \cdot \rho_{Dislo} \cdot h_{Pop-In}^2 \qquad (8)$$

Figure 3 provides the detailed insight into the essence of the equation (8). Both diagonals enclose the theoretically predicted threshold area between the appearance and non-appearance of the pop-in-effect in the depth-sensing hardness measurement. During the nanoindentation of the monocrystals, as the typical indenter roundings encounter R≈0,1µm, the pop-in-effect will appear with the high degree of certainty, assuming that the dislocation density does not exceed greatly the value of 10^9cm^{-2}. On the contrary, the Pop-in-effect is not to be expected for the indentation with the Rockwell indenter despite the low dislocation density of the exemplary Gallium Arsenide wafer. First measurements confirmed this statement. By the indenter rounding values of 0,1µm and 0,3µm, the pop-in-effect has been observed in all investigated samples. Using the 10µm-indenter, it appeared only in the comparatively

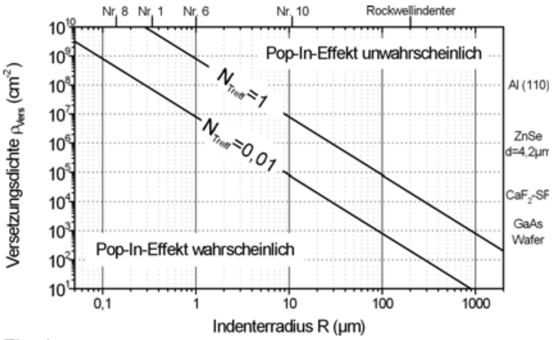


Fig. 3

dislocation-free samples (Tungsten and GaAs). The micro-hardness measurements with the Rockwell indenter showed even for GaAs no evidence of the Pop-in effect furthermore.

3. Experimental

Experimental results of the loop nucleation measurements show a good agreement with the theory of dislocations within the isotropic approach. The Pop-in-effect has been observed in metals (Al, Cu, Ni, W), ionic crystals (CaF_2 ,BaF_2) and semiconductors (CdTe, GaAs, GaP, InP, ZnSe). Corresponding dislocations were proven by means of microscopy imaging techniques (transmission electron microscopy TEM, cathodoluminescence imaging CL, and imaging of dislocation-etched surfaces).

For our depth-sensing hardness measurement the Nanoindenter^{TM} II by MTS Systems Corporation, (Nano Instruments Innovation Center, Oak Ridge, TN) has been used. The investigations have been performed at the constant loading and unloading rate of 1µN/s. Indenters with tip radiuses of 0.1µm, 0.3µm and 10µm, approximately, were used. For all materials the Pop-in-effect was observed exclusively in samples prepared so that the surface dislocation density keeps low, i.e. electrolytically polished metals, cleavage surfaces and wafers.

4. Results and Discussion

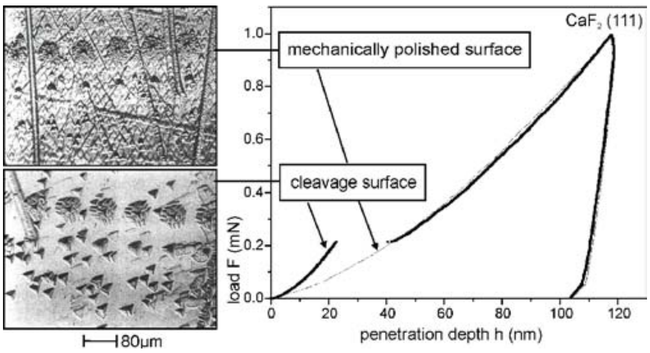


Fig. 4

Fig. 4 represents the comparison of measurement results for polished CaF_2 (111) surface and corresponding cleavage surface. The dependence for cleavage surface first shows the rapid increase with the subsequent so called Pop-in jump in the load curve. After the jump both surfaces show similar behaviour. The enclosed light microscopy images of subsequently dislocation-etched surfaces reflect the different dislocation densities and indicate dislocation rosettes around stress points induced by indenting (top to bottom: 100mN, 10 mN, 1mN maximum force). It should be taken into account that the diameter of 800nm size produced by the stress value of 1mN is considerably smaller than the corresponding etch pits.

Figure 5. presents the panchromatic cathodoluminiscence images of the indented ZnSe/GaAs layer. For given material bright and dark contrasts characterise α and β dislocations, respectively [14]. Marking indents produced by the force of 10mN are shown on the left side of figure 5, each followed by two rows for 1mN, 0.1 mN and

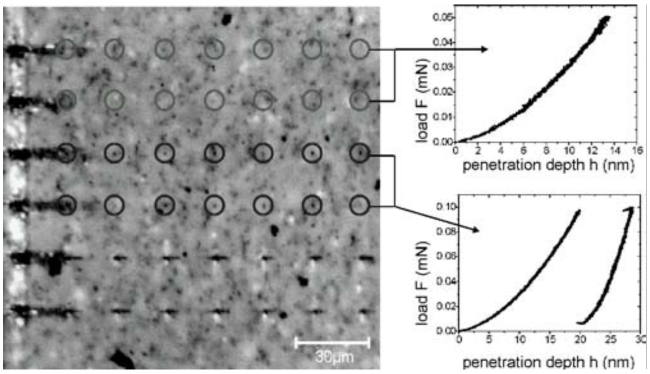
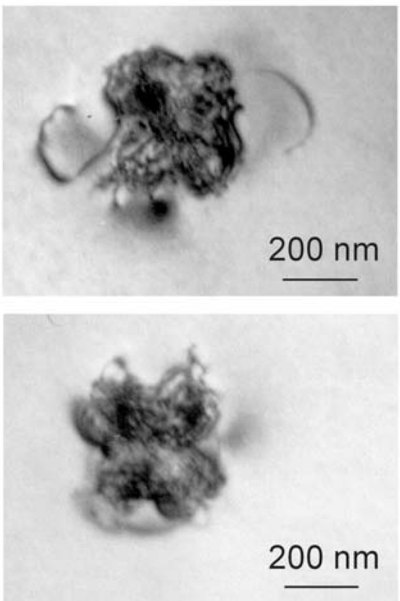


Fig. 5

0.05 mN force values, bottom to top. The corresponding measurement dependences obtained for indentation force values of 0.1 mN and 0.05 mN indicate that Pop-in effect does not appear for the force value of 0.05 mN (i.e. loading and unloading curves are reversible hence purely elastic) but is already set in by 0.1 mN. In the cathode luminiscence images the dark point contrast characterise the nucleating dislocations is observed for indentation force values from 0.1 mN onward. (The plotted circles are included to help locating indentations). It should be pointed out that though the CL-measurement resolution reaches 1 µm value, a much smaller dislocation loop (with diameter of 50 nm) still produces the contrast point.



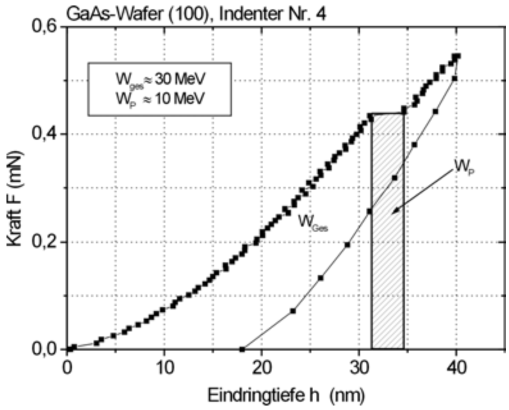


Fig 6

Figure 6 shows the TEM image of the indentation, whereby the loading has been withdrawn shortly after the Pop-in jump [15]. It can be seen that lots of dislocations appear during the jump. It means that at the starting point of the pop-in effect the homogeneous nucleation of the first dislocation loop takes place followed by its explosive growth and multiplication through interactions. At the end of the pop-in jump there is the whole dislocation network, that is to say, the crystal is plastified.

In figure 7 the maximum shearing stress estimated from the measurement curves according to equation (4) is compared with theoretical predictions for the shearing stress value at the

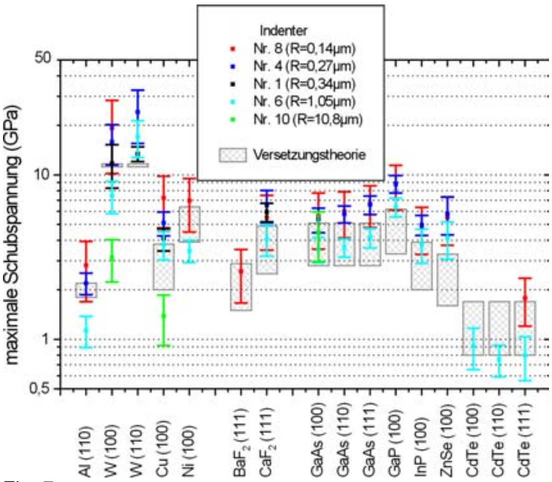


Fig. 7

homogenous nucleation of the dislocation loop (equation 1). Having in mind that these predictions are based on isotropic approach, one must assume that measurement results reveal a good agreement with theory for all investigated materials. The decrease of the maximum available shearing stress for indenters with greater rounding radiuses points to the corresponding increase of the loop radius (see equation 3).

5. Summary and Conclusions

We could show exemplarily that the trend of the development of nanoindentation methods is dominated by investigations of the fundamentals of mechanical deformation behavior of both, amorphous and crystalline materials. The nanoindentation methods have reached a new quality as a tool of high pressure physics without the use of very expensive pressure chambers. In this combination two facts are very important:

1. the realization of contact stresses in the GPa range needs small volumes as representative of a perfect sample, free from strength reducing defects, and indenter tips with small radii (nearly 100 nm) only operating by nanoindentation technique.
2. The controversial discussed indentation size effect is in this sense a necessary result of the mechanical deformation behaviour of solid or viscous materials by contact stresses. Here we agree with Nix [16,17], who has discussed this situation as Indentation Size Effect after pop-in.

Some details for these conclusions are investigated by nanoindentation in combination with AFM, TEM and CL – techniques. The development of Nanoindenters with integrated AFM is to recommend and offers the opportunity that the indented sample can be rest(?) at the sample holder for topographic investigations. The obtained experimental results confirm the suggestion that the Pop-in effect appears as a manifestation of homogenous nucleation of the first gliding dislocation loop. This loop induces further dislocations through multiplication (Frank-Read source), which then can be followed by the subsequent plastic deformation [16,17].

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