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A novel apparatus for *in situ* compression of submicron structures and particles in a high resolution SEM

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We report on the development and characterization of a novel *in situ* manipulation device to perform stressing experiments on the submicron scale inside a high resolution field emission scanning electron microscope. The instrument comprises two main assembly groups: an upper part for positioning and moving a mounted probe and a force sensor as well as a specimen support as lower part. The upper part consists of a closed loop tripod piezoelectric scanner mounted on a self-locking coarse positioning stage. Two interlocked steel springs and a linear variable differential transformer measuring the springs' deflections compose the lower part of the instrument. This arrangement acts as force-sensor and sample support. In comparison to already well-established concepts a wide measuring range is covered by adjusting the spring constant between 30 N/m and 50000 N/m. Moreover, the new device offers striking advantages with respect to force calibration and sample deformation measurements. Force calibration is performed using the eigenfrequency of the force detection system directly inside the SEM. Deformation data are obtained with high accuracy by simultaneously recording displacements above and below the specimen. The detrimental apparatus compliance is determined, and the influence on measured data subsequently minimized: an easy to validate two-springs-in-series model is used for data correction. A force resolution in normal direction of 100 nN accompanied by a sample deformation resolution of 5 nm can be achieved with the instrument using an appropriate load cell stiffness. The capabilities and versatility of this instrument are exemplified by compression experiments performed on submicron amorphous silica particles. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4749256>]

I. INTRODUCTION

Understanding mechanical properties on a nanometer scale is crucial for various applications, including microelectronics, sensors, and many applications in the field of particle technology and materials science. Although mechanical properties have to be known precisely to design suitable long living products, the mechanical responses of small structures to applied loads are poorly understood. This is especially true for nano-particulate systems, caused by the experimental difficulties to test these small structures individually. Studies on volumes confined by, e.g., applying rather small loads to a bulky material, are far more abundant. The probably most commonly employed technique is depth-sensing nanoindentation.¹ With respect to sample preparation and experimental procedures, indentation of bulky samples is comparatively simple. A correct interpretation of the obtained data is nevertheless far from being well established. Unambiguous correlation of features in recorded load displacement data and distinct processes within the material is not directly possible. Commonly accepted models and measuring procedures like the well-known Oliver and Pharr Method² do not account for systematic errors like pile-up around the indentation imprint. Additionally, effects stemming from the development of a three-dimensional and rather complex stress field under an

indenter generally overlays changes in deformation mechanisms at small scales: strain gradient plasticity theory,³ developed from torsion experiments on metal wires, is used to explain the indentation size effect observed for many metals and ceramics.⁴ Despite the above mentioned drawbacks, intriguing mechanical properties and deformation mechanisms have been reported using volume confinement by nanoindentation: single or multiple displacement-bursts in the range of the theoretical strength, serrated and highly localized flow phenomena, and phase transformations directly under the indenter are a few heavily cited examples.^{1,5}

To avoid the above mentioned problems a more intuitive way of testing small structures, ideally a simple geometry with uniaxial loading, can be used.⁶ Of course, these experiments are directly coupled to advances made in the field of electron microscopy and micromechanics: initially driven by the need of measuring hardness at very low loads, i.e., small residual imprints, several scanning electron microscope supported *in situ* indentation devices have been developed. Transmission electron microscopy (TEM) offers the possibility to study projections at the microstructural level directly, but suffers from the highly limited space in between the pole gap pieces and the need for electron transparent samples. Information obtained from scanning electron microscopy (SEM) is, in contrast, limited to regions near the surface. Nevertheless, the generally much larger space within the vacuum chamber of a SEM and the very high working distances allow for big samples. This renders laborious sample preparation and thinning procedures unnecessary. Moreover, the SEM imposes

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much less design constraints to the *in situ* apparatus: Actuators and sensors can be completely integrated, limiting the required ports from outside the vacuum chamber to signal- and controlling-wires. Developments in the field of focused ion beam machining, the controlled metal deposition, and the integration of manipulators for moving, holding, and stressing objects combined with the ability to study organic and volatile samples by environmental techniques make *in situ* SEM experiments highly attractive.^{7,8} The first *in situ* stressing experiments in a scanning electron microscope were performed by Gane and Bowden,^{9,10} although the apparatus lacked depth-sensing capabilities, significant results on the yielding of low defective volumes and an observed displacement burst were obtained. At the same time several straining stages, operated from outside the vacuum chamber, were developed.^{11,12} Another major step was the development of the SEM hardness tester by Bangert *et al.*^{13–16} In this case the imaging capabilities of the SEM were solely used to measure the contact area precisely, overcoming limitations imposed by light microscopy. Rabe *et al.*¹⁷ developed the first real depth sensing *in situ* indenter, allowing for a direct measurement of tip displacement. In this case an inherently compliant load cell is used for force measurements. Since only indenter displacements are measured comparatively high errors in the calculated strains result.¹⁸ Today several sophisticated device concepts and improvements of already existing devices have been developed.^{19–21} Further details on the history and recent developments of electron microscopy based *in situ* testing devices are given by Legros *et al.*⁸ In summary, the whole span of applications from tensile testing to *in situ* tribology for both organic and inorganic mater in amorphous and crystalline state is now being covered.^{22–37} Even the first commercial solutions emerge for both SEM and TEM, mainly employing a patented three plate capacitor design for applying forces and measuring displacements at the same time.^{7,19,38,39} MEMS technology integrated into electron microscopes currently offers the best force and displacement resolution. However, the routine application is not yet possible.^{40–43} In parallel to the electron microscopy based *in situ* devices, scanning probe based concepts – often called triboscanners – have been developed. They allow for stressing and subsequent imaging, observing the stressing event itself is not possible.⁴⁴

Preparation of submicron samples used in the stressing experiments is possible by bottom-up or top-down methods. Although top down sample preparation, as performed by focused ion beam milling and lithographic processes, allows for shape control, the influence of a residual damage close to the surface in the received specimens is currently not understood;^{45,46} a remarkable difference is observed when the classical whisker tests by Brenner^{47–49} and other groups^{28,50–52} with the compression of FIB made pillar structures, both *in situ*⁵³ and *ex situ*.^{6,54–58} Bottom-up processes, whisker growth being just one example, offer a suitable and less time consuming approach. Especially the narrow control over particle forming processes in terms of particle-size, -shape and -phase composition imposed over typically gas- and liquid-phase synthesis routes offer a highly attractive alternative. Acquisition of data can be performed on a statistically relevant number of particles, i.e., up to 30 particles can

be tested within about 3 working hours with the presented instrument. Moreover, there is a strong need to experimentally characterize particle systems directly, since no analytical solutions for the complex interplay of stress fields in small particles and their mechanical properties are available. Despite the long history of *in situ* SEM and TEM based manipulators as well as triboscanners, and some recent work on aggregates of nanoparticles,^{63–65} to the best of our knowledge rather little work has been performed directly dealing with compression of submicron sized particles.^{10,59–62}

Here we report on the development of a novel *in situ* manipulator for mechanical characterization of free standing structures situated in a high resolution field emission scanning electron microscope. In contrast to already existing devices and well-established concepts, the device covers a wide measuring range by a simple adjustment of the spring based force sensor. Force calibration is performed using the eigenfrequency of the force sensor inside the SEM, right before experiments are carried out. Measured deformation data are of very high accuracy, since displacements are recorded above as well as below the specimen. Based on the unique design of the apparatus an easy to validate two-springs-in-series model can be used to remove the detrimental influence of apparatus compliance on measured data. Based on the applied spring, a force resolution below 100 nN accompanied by sample deformation measurements with an accuracy of 5 nm can be achieved.

II. EXPERIMENTAL SETUP

A. Configuration

A schematic diagram and a photographic image of the newly developed *in situ* manipulation device for compression experiments on the submicron scale are displayed in Fig. 1. The apparatus consists of two main assembly groups, which can be easily dismantled: the upper part, termed nanomanipulator, carries out all movements of the employed probe; the lower part, called load cell, acts as normal force sensor with integrated sample support. To fulfill all constraints imposed by the scanning electron microscope only vacuum compatible materials of low magnetic susceptibility were used: the load

FIG. 1. Schematic diagram (a) and a photographic image (b) of the *in situ* manipulation device. Two main assembly groups termed nanomanipulator and load cell are displayed. The nanomanipulator performs all probe movements by a linear coarse positioning stage and a piezoelectric tripod scanner. The load cell detects applied normal forces and supports the sample.

FIG. 2. Schematic drawing of the apparatus in side view as installed in the SEM (a). Infrared illuminated photographic image of the sample area in working position inside the SEM (b). Proper imaging is assured as demonstrated in (c) showing a probe in contact with a silica particle on a silicon substrate.

carrying frame of the device with total outer dimensions of $25\text{ cm} \times 12\text{ cm} \times 5\text{ cm}$ ($l \times w \times h$) is completely made from a standard aluminum alloy. Additionally used materials comprise a titanium alloy, spring steel, brass and vacuum compatible polymers for insulation. The apparatus is optimized to fit in the vacuum chamber of the scanning electron microscope used (Ultra FE-SEM Gemini, Carl Zeiss, Jena, Germany). It is fixed to the standard sample stage of the microscope by a dovetail joint at the flip side of the main frame. Tilting of the apparatus by 4° with respect to the electron beam, depicted in Fig. 2, and the low working distance of 7 mm, as a result of the unobstructed sample environment, enable proper imaging.

The nanomanipulator (upper part in Fig. 1) is composed of a closed loop piezoelectric tripod scanner (Anfatec Instruments AG, Oelsnitz, Germany) operated in open loop mode. The scanner is fixed to a custom-built linear coarse positioning stage. A probeholder is attached to the free end of the tripod scanner, fully compatible to the Agilent MTS XP system. The coarse positioning stage is used to bring probe and sample surface in close proximity. For final probe positioning and stressing only movements performed by the piezoelectric tripod scanner are used. The coarse positioning stage is designed as a sledge guided by a track in the housing of the manipulator. Forward movement is realized by a gear reduced stepper motor driving a fine thread screw; the screw pushes the sledge forward. A restoring force ensuring contact between sledge and fine thread screw is caused by two coil springs clutched asymmetrically inbetween sledge and housing. A resulting torque ensures self-locking, hence increases the position stability. Step sizes of 120 nm are realized by the stage while sample heights of up to 2 cm can be accommodated. The tripod scanner, transported by the sledge, spans a rectangular positioning area of $80\text{ }\mu\text{m} \times 80\text{ }\mu\text{m}$ in plane of the sample. The piezo range normal to the surface accounts for 5 μm . The tripod scanner can be positioned with sub-nanometer resolution. All scanner movements are monitored by strain gauges attached to the comprising piezostacks. Elongation normal to the sample surface is measured with a resolution of 5 nm using a strain gauge extending over the full length of the respective piezostack. The strain gauges are operated in temperature compensated Wheatstone half-bridges. To avoid a steplike movement normal to the sample surface,

as caused by the inevitable discretization of the computer controlled voltage signal applied to the piezostack, an additional resistance is introduced in series; a dampening RC circuit is formed using the intrinsic capacitance ($C = 350\text{ nF}$) of the piezostack. For smoothing, the time constant $\tau = RC$ of the dampening circuit is adjusted to about twice the step length in the discretized voltage control signal by adapting the additional resistance R in the circuit. Deformation rates in the range of several $\mu\text{m/s}$ can be conveniently achieved.

The load cell (lower part in Fig. 1) acts as force sensing sample support measuring the applied normal forces. It consists of two 20 μm thick spring steel foils clamped at the housing of the load cell. Both foils are interlocked by an all-thread titanium rod using two pairs of clamping jaws. In total this arrangement acts as a Hookean spring. An aluminum stub is attached to the upper free end of the rod as sample support. The female threaded magnetic core of the used linear variable differential transformer (LVDT - MHR 025, measurement SPECIALITIES, Virginia, USA) is screwed onto the titanium rod. The LVDT itself is fixed by three fine thread screws, arranged in an isosceles triangle, to the frame of the load cell. Using the LVDT deflection of the spring system is detected with an accuracy of about 2 nm using lock-in technology (Anfatec Instruments GmbH, Oelsnitz, Germany). The time constant for integration can be set in a range between 0.2 s and 5 s. Commonly a thousand data points are recorded for one compression experiment. The spring constant of the system can be set ranging from 30 N/m to 50000 N/m to aid a variety of different measuring purposes. To this end the tension of the interlocked steel foils is adjusted by changing the relative positions of the clamping jaws on the titanium rod. Changing the spring constant of the load cell leads to a changing force resolution: according to Hooke's law the deflection resolution of 2 nm in the load cell corresponds to resolution of the applied force ranging from 60 nN to 100 μN . A frictionless movement of the magnetic core inside the LVDT is ensured by aligning the coaxial positioned LVDT tube using the fine thread screws: dampening of the free oscillation is reduced iteratively.

Within the framework of the experiments presented here, a flat punch indenter made of boron doped diamond is used (Synton-MDP, Switzerland). Boron doping is employed to enhance conductivity of the indenter allowing for proper imaging of the indentation process. The plateau diameter of the indenter accounts for approximately 3 μm . Fig. 3 displays a SEM image (a) of the indenter and an atomic force microscopy image of the plateau (b) measured by a MultiMode SPM (Veeco Instruments, USA). The surface roughness rms is determined to 0.38 nm on a $2\text{ }\mu\text{m} \times 2\text{ }\mu\text{m}$ area.

B. Calibration and stiffness characterization of the instrument

Spatial calibration of the strain gauges attached to the tripod scanner is performed *ex situ* using an AFM and suitable calibration gratings. The calibration is verified *in situ* by SEM imaging of the probe movement. The spatial calibration of the LVDT in the load cell is directly traced back to the strain gauges: a proportionality factor (termed sensitivity)

FIG. 3. SEM image of the used diamond flat punch indenter (a) accompanied by an AFM image (b) characterizing the rms roughness of the plateau. The rms surface roughness accounts for only 0.38 nm (height data were processed using procedures for plane-fit and flatten).

transforming changes in the LVDT voltage output ΔU into the real deflection of the Hookean spring system Δd is experimentally determined. To this end, the sample support of the load cell and the tip of the tripod scanner are brought into a nonpositive contact and deflected together by elongating the tripod scanner for a measured distance Δz . A linear response characteristic, exemplified in Fig. 4, is obtained.

The slope, termed apparent sensitivity s , is defined according to

$$s = \frac{\Delta U}{\Delta z}. \quad (1)$$

Due to the finite stiffness of the components in the load train of the apparatus, e.g., adhering and counter supports of the piezoelectric stacks, the measured apparent sensitivity s is influenced by the apparatus compliance. Interpreting the elongation of the piezoelectric stack Δz as the sum of the spring system's deflection Δd and the compression of all components in the load train ζ , the true sensitivity σ is defined according to

$$\sigma = \frac{\Delta U}{\Delta d} = \frac{\Delta U}{\Delta z - \zeta} = s \frac{\Delta z}{\Delta z - \zeta}. \quad (2)$$

Assuming nonpositive contact with an applied normal force F and Hookean behavior of both, the load cell (spring constant $D = F/\Delta d$) and the apparatus compliance (spring

constant $\kappa = F/\zeta$), the true and the apparent sensitivity are linked by

$$s = \sigma \left(1 + \frac{D}{\kappa} \right)^{-1}. \quad (3)$$

This offers an elegant way to characterize and account for the instrumental compliance κ : measurements of the apparent sensitivity s for varying spring tensions, hence load cell constants D already suffices to determine κ by fitting the measured data.

To determine the spring constant of the load cell D , the eigenfrequency f of the load cell is used. Using the oscillating mass m (comprising the titanium rod with the attached magnetic core, the sample supporting stub, both pairs of clamping jaws and if applicable the sample itself) and the springs' mass m_s , eigenfrequency and spring constant are linked by

$$D = \left(m + \frac{1}{3} m_s \right) (2\pi f)^2. \quad (4)$$

This equation is valid for ideal harmonic oscillators taking mass of the springs into account. The oscillating mass m , and the springs' mass m_s are determined by weighing to 6.44 g (including specimen) and 0.52 g respectively. Fig. 5 shows an example of a hardly damped harmonic oscillation of the spring system inside the load cell. The method can be easily applied both in the SEM and outside: to measure eigenfrequency in the inherently low vibrational SEM

FIG. 4. Linear output signal U of the LVDT as a function of the piezo driven displacement z of the sample holder. The slope is the termed apparent sensitivity s of the LVDT in units of V/m.

FIG. 5. Free oscillation of spring system inside the SEM. The measured amplitude corresponds to a total deflection of 100 nm. The measured eigenfrequency is used to determine the spring constant with unrivaled accuracy.

FIG. 6. Apparent sensitivity s of LVDT as a function of spring constant D . Due to finite stiffness κ of the instrument a small compression, most likely due to the adhering of the piezostacks is observed.

environment, the oscillation of the spring system is deliberately excited by a slight movement of the SEM sample stage. Since masses and the eigenfrequency can be determined with deviations as small as per mill, the spring constant can be determined with unrivaled accuracy prior to each experiment.

As discussed above, the instrumental stiffness κ and true LVDT sensitivity σ are obtained by fitting Eq. (3) to several combinations of D and s . Fig. 6 shows the resulting fit with the free parameters $\kappa = 337.7 \pm 7.7$ kN/m and $\sigma = 184.5 \pm 0.2$ V/m. Both values are assumed to be constant instrument parameters and are used for further data evaluation. For quite typically loads of up to 1 mN the compression of the instrument accounts for 3 nm. Compared to the observed spring deflection of about 100 nm (spring constant of the load cell $D = 8765 \pm 18$ N/m for the subsequently presented experiments) this effect is rather small. Nevertheless it is accounted for this effect since correction of measured data is straightforward and easily applied during raw data processing.

C. Data processing

Force-deformation data are extracted from the probe movement normal to the sample surface Δz_m , measured by the strain gauge, and the spring deflection related to the change in LVDT output voltage ΔU_m . Using the spring constant D and true sensitivity σ , the acting normal loading force F can be calculated according to

$$F = D \frac{\Delta U_m}{\sigma}. \quad (5)$$

The sample deformation δ_t is given by the difference of the probe movement Δz_m , corrected for the unintended compression ζ , and spring deflection $\Delta U_m/\sigma$, as given by

$$\delta_t = (\Delta z_m - \zeta) - \frac{\Delta U_m}{\sigma} = \Delta z_m - \frac{\Delta U_m}{\sigma} \left(1 + \frac{D}{\kappa}\right). \quad (6)$$

The compression ζ is a function of the applied normal load and is estimated using the determined stiffness of the apparatus.

Free oscillations with amplitudes below 10 nm of the spring system occur typically before establishing the first contact between probe and sample. They are removed from the

recorded data using a low-pass filter: a cutoff-frequency right below the eigenfrequency of the spring system is chosen.

III. MEASUREMENTS AND RESULTS

To demonstrate the versatility and abilities in terms of probe positioning, force and displacement resolution, imaging, and stability of the newly developed *in situ* manipulation device, amorphous silica particles were compressed by a diamond flat punch indenter. Spherical silica particles were chosen as model system due the narrow particle size distribution and their good availability in different sizes over several orders of magnitude. The amorphous structure of the particles eliminates any influences of lattice alignment during the measurements. Hence, the system behaves isotropically.

A. Sample preparation

Silica spheres (sicastar[®] plain) with a mean diameter of 500 nm were obtained from micromod Partikeltechnologie GmbH (Rostock-Warnemuende, Germany). The received suspensions were diluted in deionized water (resistivity > 18 M Ω cm). No additional measures were taken to disperse the particles further after dilution. The diluted suspensions were cast onto pieces of a standard (100) silicon wafer with dimensions of 5 mm $\times$ 5 mm by a dip-coating procedure, followed by drying under ambient conditions. Cleaning of the wafer to remove organic compounds was carried out prior to silica deposition by rinsing with acetone and ethanol. To ease wettability by creating a hydrophobic surface a plasma treatment followed the cleaning. A silica concentration of about 250 μ g/ml turned out ideal to achieve a sufficient amount of well separated particles in the lateral scan range of the nanomanipulator on the substrate. Fixation of the silicon wafer on the sample holder of the load cell was performed using an electrically conductive adhesive. Special care was taken to ensure a coplanar alignment of sample, sample support, and the diamond probe to guarantee particle loading in normal direction only. The specimen and the probe were grounded to prevent electrical charging during imaging. As previously described the whole apparatus was tilted by 4 $^\circ$ and the working distance in the SEM was set to about 7 mm. The spring constant of the load cell was determined before the experiments were started inside the SEM.

B. Influence of electron irradiation on the behavior of silica particles

There are various literature reports on the influence of continuous electron irradiation on the mechanical properties of silica nanostructures.⁶⁶⁻⁶⁹ These experiments were performed in transmission electron microscopes with comparatively high acceleration voltages of up to 200 kV. An influence has not yet been reported for *in situ* SEM work and needs further clarification. To this end, force relaxation was studied for statically loaded single silica particles with and without parallel imaging. After an appropriate positioning of the flat punch indenter over a single particle and subsequently

FIG. 7. Influence of electron irradiation on a statically loaded silica particle (500 nm in diameter). (a) Deflection of the springs in the load cell, matching particle deformation. (b) Force obtained from the deflection and respective spring constant. (c) Extension of the tripod scanner normal to the sample surface. The beam is turned on at $t = 3$ s. Throughout the experiment probe position is maintained, the particle deformation increases suddenly, the applied load drops. Discrete steps with a period matching the scanning period of the electron beam ($T = 377$ ms) are observed in the deformation and force data.

blanking the 5 keV electron beam (intensity < 0.15 A/cm²), a load was applied by a movement of the diamond flat punch in the normal direction. After establishing mechanical equilibrium, the blanked beam was turned on again. An example of the obtained results is given in Fig. 7: it can be seen that a maximum static load of 0.48 mN was applied. Mechanical equilibrium was established within 1 s after the piezo came to rest. At $t = 3$ s the blanked beam is turned on again: although the position of the diamond probe remains constant (Fig. 7(c)), instant and sharp changes in spring deflection and force data (Figs. 7(a) and 7(b)) are visible. The sharp steps are followed by a stepwise compression of the particle matching the scanning period of the electron beam ($T = 377$ ms). As can be seen, load decreases by more than a factor of two, caused by the deformation of the stressed particle causing a relaxation of the load cell. The results indicate a decrease of yield strength by more than a factor of two during electron irradiation. The steps in the recorded data with typical falling times below 100 ms indicate that the beam influence decays instantly when radiation is turned off. As a consequence it seems unlikely that a temperature rise due to the impinging electrons causes the yield drop. Zheng *et al.*⁶⁹ suggested that electron beam induced bond switching events cause the softening of the silica particles. A similar experiment performed for the sole loading of the substrate did not result in a load drop. This indicates that the observed effect is based on the interaction of the impinging electrons and the silica particles. Since the conducted experiments show an influence of irradiation on the mechanical properties of silica, further loading experiments were always performed with blanked beam. Imaging

FIG. 8. Raw LVDT (a) and strain gauge data (b) obtained in a loading and unloading cycle of one silica particle with a diameter of 500 nm. LVDT signal shows a sharp step related to crack initialization and propagation, resulting in a decrease of load due to partial relaxation of the load cell (a).

was performed only during probe positioning prior to loading and after the loading experiment was finished.

C. Deformation behavior of silica particles

To study the deformation behavior of single silica particles 32 loading experiments were conducted with applied maximum loads between 380 and 725 μ N and a loading rate of 700 μ N/s. Particle diameters were measured by SEM imaging with an accuracy of 10 nm. Prior to the stressing experiments, the rectangular working area of the tripod piezoelectric scanner was marked on the specimen by indenting one of the corners. To obtain variable maximum loads for a given loading rate, the diamond probe was positioned in different distances (50 – 150 nm) above the particles. Loading and unloading was performed by a continuous elongation of the tripod scanner, the probe traveled a total distance of 360 nm from its resting point to the front reversal point. Representative raw data for one loading experiment depicted in Fig. 8(a) show the recorded LVDT signal and its conversion to force before and after applying low-pass filtering. As can be seen filtering removes undesired oscillations especially present after the probe lost contact with the sample at the end of the experiments. In general the obtained data exhibits low noise and is of high quality. Fig. 8(b) shows the probe position as measured by the applied strain gauges resolved to 5 nm: deviations from the ideal triangular movement of the probe are caused by creep, non-linearity and hysteresis inherent to unregulated piezoelectric scanner movements. In addition, the analog low pass filtering used to smooth the control signal causes a slowed response to any signal changes. The real elongation of the piezostack is falling behind the computer controlled output signal: although retracing is not finished when data recording is stopped, particle indenter contact is already lost.

FIG. 9. Force – deformation plots of two silica particles (a, b). Reduced Young's moduli E^* and yield points were determined using a Hertzian model of two sphere – plane contacts. A displacement burst at a relative deformation of 0.40 ± 0.04 was observed at loads of 523 ± 46 μN. Side and top view images of the corresponding particles (c, d) reveal the connection of the burst to cracking in the meridian plane of the particles.

Side view images recorded immediately after finishing a loading experiment give valuable feedback: measured deformation can be checked and any erroneous probe sample alignment can be excluded. To obtain top view images of the stressed particles, the specimen was mounted on a standard SEM specimen holder after the stressing experiments were finished. Information on changes at the initially shadowed-sides of the particles are obtained. Locating the loaded particles is possible due to intentionally set indent at the beginning of the experiments, and recorded lateral coordinates of the piezoelectric scanner during stressing.

Fig. 9 shows the typically obtained force deformation behavior (obtained from the measured raw data as described in Sec. II C) for two representative measurements. In addition to a rather ductile behavior, indicated by the generally smooth curve progression, and the continuously increasing deviation from a purely Hertzian elastic behavior⁷⁰ indicated by the solid black lines, a displacement burst is visible in case of sufficient loading (Fig. 9(b)). During compression of 32 particles the displacement burst was observed for 21 particles at a load of 523 ± 46 μN, corresponding to a mean total relative particle deformation of $\delta_t/d = 0.40 \pm 0.04$. SEM imaging was used to analyze the reason for the displacement burst in the data: the observed displacement bursts are directly related to the formation of up to three cracks in a meridian plane of the particles, as exemplified in the SEM pictures of Fig. 9. Following smaller only sometimes observable bursts were attributed to the formation of additional minor cracks.

Reduced Young's Moduli were determined by applying Hertzian theory.⁷⁰ The theory delivers a mathematical relation between the deformation δ at each contact, modeled as a sphere contacting an infinite half space, and the applied external load F . Making the simplifying assumption that the deformation δ at both contacts equals exactly half of the total particle deformation δ_t load is given by

$$F = \frac{4}{3} E^* \sqrt{R} \left(\frac{\delta_t}{2} \right)^{3/2}. \quad (7)$$

Since the particle radius R was extracted from SEM images prior to stressing, the reduced Young's modulus E^* can be determined by fitting the model to the elastic range of the loading curves (Fig. 9). The obtained distribution of the reduced Young's modulus is shown in Fig. 10. The arithmetic average was calculated to 39.3 ± 5.7 GPa.

The reduced Young's modulus in turn is linked to the Young's modulus of the silica particles E_{SiO_2} for the given configuration “diamond-silica-silicon” by

$$E^* = \frac{E_1^* + E_2^*}{2}, \quad (8)$$

$$E_1^* = \left(\frac{1 - \nu_{\text{SiO}_2}^2}{E_{\text{SiO}_2}} + \frac{1 - \nu_{\text{dia}}^2}{E_{\text{dia}}} \right)^{-1}, \quad (9)$$

$$E_2^* = \left(\frac{1 - \nu_{\text{SiO}_2}^2}{E_{\text{SiO}_2}} + \frac{1 - \nu_{\text{Si}}^2}{E_{\text{Si}}} \right)^{-1}. \quad (10)$$

Using $\nu_{\text{SiO}_2} = 0.17$, $\nu_{\text{dia}} = 0.07$ and $\nu_{\text{Si}} = 0.22$ as Poisson's ratios and $E_{\text{dia}} = 1143$ GPa, $E_{\text{Si}} = 130$ GPa as

FIG. 10. Distribution of measured reduced Young's moduli of 32 tested silica particles with a mean diameter of 500 nm. Arithmetic average was determined to $E^* = 39.3 \pm 5.7$ GPa. A normal distribution with the given mean value and standard deviation is plotted as a guidance.

Young's moduli of the flat punch and the substrate, the Young's modulus of the silica particles can be calculated to $E_{\text{SiO}_2} = 44.7 \pm 7.6$ GPa, undermatching the bulk modulus of fused silica ($E_{\text{fused SiO}_2} = 76.4$ GPa).⁷¹ Similar values have been reported for silica microcapsules.^{72,73} The difference was attributed to production process of the silica particles. The particles studied here were produced by Stöber synthesis and differ from fused silica by a significantly reduced amount of siloxane bonds. The decreased crosslinking of the comprising network directly effects the elastic properties of the particles and results in a reduced Young's modulus.^{72,73}

Determination of stresses is challenging: there is no possibility to measure the contact area throughout the loading and unloading process with an appropriate accuracy. Models have to be used to estimate the radius of contact a and the contact area A . For the plastic regime of the loading process and the unloading process only rough estimations based on volume conservation exist. For the elastic regime of the loading process Hertzian theory⁷⁰ provides an accurate estimation of the contact radius

$$a = \sqrt{R \frac{\delta_t}{2}}, \quad (11)$$

$$A = a^2 \pi. \quad (12)$$

Thus yield strength p_Y can be determined by dividing the measured force F_Y at the yield point by the calculated contact area A_Y :

$$p_Y = \frac{F_Y}{A_Y}. \quad (13)$$

By assigning the yield point manually (datapoints in Fig. 9 start to deviate from the Hertzian fit) a yield strength of 6.1 ± 1.3 GPa is determined for the tested particles. This value significantly exceeds the compressive strength of bulk silica of 1.1 GPa. The value is in good agreement with other literature reports for submicron silica structures.^{69,74} The increase is caused by the significantly lowered amount of surface defects for submicron sized structures compared to bulk material.⁷⁴

IV. CONCLUSION

A novel concept for a SEM supported stressing apparatus for the mechanical study of structures in the micron and submicron range was introduced. The device consists of a custom build coarse linear positioning stage, moving a piezo-electric tripod scanner with an attached flat punch indenter. The load applied to a sample is supported by a spring based force sensing system: the so-called load cell consists of two thin steel foils interlocked by a rod. Deflection of the springs is measured by a linear variable differential transformer. This unique design offers striking advantages in terms of stiffness characterization, force calibration, and deformation measurement: compliance is determined by measuring sensitivity for varying load cell stiffness. Using a simple to apply and validate two-springs-in-series model allows for a subsequent correction of data for this detrimental influence. The spring constant is determined using eigenfrequency of the load cell

with unmatched accuracy; force resolution below 100 nN is easily possible. Deformation measurements are performed both above and below the sample with a resolution of 5 nm. The measuring range of the whole instrument can be adjusted in a wide range by changing the stiffness of the interlocked foils: spring constant can be set ranging from 30 N/m to 50000 N/m. To demonstrate the capabilities and versatility of the device, submicron silica particles with a mean diameter of 500 nm were characterized: a Young's modulus of $E_{\text{SiO}_2} = 44.7 \pm 7.6$ GPa and yield strength of $p_Y = 6.1 \pm 1.3$ GPa were experimentally determined using Hertzian contact theory. Electron irradiation caused a significant decrease in yield strength of the silica particles. For the future it is planned to investigate the influence of dehydration, polarity of the dispersing agent, the electron irradiation and heating in more detail.

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