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LOW LOAD VICKERS HARDNESS MEASUREMENTS OF NONCONDUCTING MATERIALS IN A SCANNING ELECTRON MICROSCOPE

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Abstract

Vickers hardness tests on microscopic small bodies, e.g., fibers, powder particles, thin layers, etc., require imprint dimensions near or even below the resolution limits of light microscopes. Hence, to detect and evaluate these miniature imprints a scanning electron microscope (SEM) has to be used. For such observations in a SEM, nonconducting samples have to be coated with a thin conductive layer. The influence of these films on the imprint size and thus on the hardness value can be rather significant. On the basis of a systematic investigation in the case of a layer much softer than the sample to be tested, methods for an elimination of the layer's contribution to the hardness result are presented.

+ This work is dedicated to Professor F. LIHL on the occasion of his 80th birthday.

Key Words: Vickers micro hardness, scanning electron microscope, coated system.

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Introduction

The extension of the Vickers hardness test down to loads of some 10^{-5}N opens a wide field of new applications not only in the vast developing branches of thin film-, fiber- and powder-technology but also in biology, mineralogy,etc. Evidently the desired miniature Vickers imprints necessary for the required spatial resolution can hardly be detected in a microscope if the imprint experiment was performed out of view. Hence, in-situ testing and evaluating inside a scanning electron microscope (SEM) is more or less a necessity for ultralowload testing on the Vickers principle. A suitable device was described a few years ago [2,3,4] and is now used in several research labs. Those users investigating nonconducting materials are faced with possible errors caused by conductive layers necessarily being deposited on their samples. Although such layers are at the most only some 10 nm thick their influence can be significant and will increase with decreasing imprint size.

In general Au or Au-Pd alloy films are used as conductive deposit which can conveniently be sputtered with air and thus in a rather simple DC-sputter system. Such deposits exhibit a Vickers hardness between 400 to 600 MPa (see Fig.2) and are therefore usually softer than most of the materials to be tested. Only polymers and other organic substances may be softer than Au or Au-Pd and would, in addition,behave differently due to a pronounced elastic redeformation [7].

In the present paper we deal with Vickers tests made on a hard body through a soft layer.

There are two approaches for an elimination of the layer's contribution. In the first one the layer is assumed to be ultra soft, i.e., it does not bear any part of the indentation load. A more elaborate correction procedure will account for a finite hardness - and thus for a substantial load bearing capacity - of the layer.

Before we deal with both aspects experimentally and theoretically in the system Au-layer/Si-substrate we shall briefly describe the apparatus used and the testing procedure in the SEM.

Principle of Vickers ultra micro hardness testing

The application of a bending cantilever arrangement to generate and transmit the load to the indentor is very adequate for an in-situ operating miniaturized testing device. The load is generated electromagnetically and measured by means of strain gauges attached to the indentor carrying spring (Fig.1). Loads ranging down to 10^{-5}N can be achieved this way*. A feedback between the strain gauges and the load generating coil forms a closed loop. A µ-processor produces a fully automated testing cycle which consists of the descent to the sample with various speeds, the indentation with a preselected load rate and the dwell time under test load for a given period. In a former paper the influence of these experimental parameters on the hardness results was demonstrated [5].

Experimental

Several considerations let silicon appear to be a suitable testing material: as a semiconductor it is sufficiently electrical conductive so that SEM investigations can be performed also without surface charging - i.e., without conducting layer. Secondly, the hardness of Si is at least one order of magnitude above the hardness of Au (although Si is known to form cracks at room temperature for loads above approximately 200 - 500 mN, its microplastic behaviour permits imprints without fracture below that value [9]).

To study the influence of a soft overlayer on the hardness result,a highly polished Si wafer was used. It was etched in diluted HF to remove the oxide overlayer and was then covered with gold films produced in a DC-sputtering system with air of 100 mbar. The distance between target and sample was 40 mm; sample and target diameters were 10 mm and 50 mm, respectively, so that a good uniformity across the entire sample surface was obtained. The deposition rate was kept constant to 10 nm/min. Seven different film thicknesses ranging from 5 nm to 180 nm were chosen. Samples either coated or uncoated were tested with loads covering a range between 0.5 mN and 20 mN; rate of load increase and dwell time were kept constant throughout all experiments to 100 µN/s and 10 s, respectively. All hardness data given had been evaluated from an average of 5 separate imprints. From the scatter of the individual data an average statistical error in hardness around 10% was estimated.

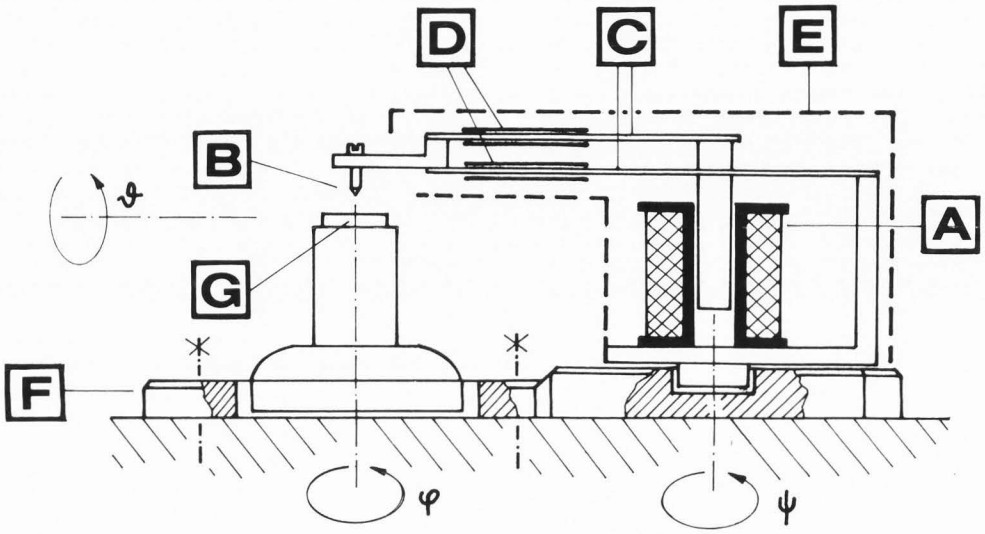

Fig.1 Principle of the mechanical part of the ultra micro hardness tester: (A) coil, (B) indentor, (C) double leaf spring, (D) strain gauges, (E) electro magnetic shield, (F) base plate, (G) sample, (φ) rotation of the sample, (ψ) sweep of the device, (ϑ) tilt of sample together with device.

For the communication between the control unit outside the SEM and the tester head only one additional electrical vacuum feedthrough has to be installed in the SEM. The tester head is directly mounted on the x-y stage and can be tilted with it. After centering the interesting sample detail on the screen the indentor tip is positioned right above it by independent electric drives on the head. The indentation procedure is then started and executed. After resetting the tip into the off-load position (out of view when the stage is tilted) the imprint appears on the screen and can be evaluated immediately.

Results

Fig.2 shows the results of hardness measurements on uncoated Si, on Si coated with different

* Lowering this limit would be possible to some extent, but cutting and handling of the required Vickers diamond tips (tip radii less than 0.2 µm) are extremely difficult.

Au-layers and on pure Au. The well known hardness increase with decreasing load for bulk materials can be observed for the coating and the substrate material [6,9]. A combined system, e.g. Au film on Si substrate, however, behaves differently: the graphs must approach the respective pure material's hardness values when the load either tends to zero or to infinity. In the first case the deformation zone around the indent is well confined within the coating, whereas at very high loads mainly the substrate will bear the corresponding large imprint. To illustrate this behavior different film thicknesses were chosen.

The hardness values obtained through the 5nm thick Au layer coincide with the corresponding ones of uncoated Si for loads above approximately 10 mN and represent the aforementioned case of high loads. Below that load a deviation towards lower hardness shows up gradually as expected. For the 10 nm and 20 nm thick films the coincidence with Si values shifts towards higher forces. The predicted approach at low loads - in our system a drop of hardness down to the values of gold cannot yet be observed with these films (imprints always penetrate into the Si substrate.

The graph for the 50 nm thick coating, however, already changes its slope at approximately 5 mN and bends in the direction of the graph of gold. This feature becomes more and more pronounced as the thickness of the gold coating increases. Simultaneously the hardness maximum tends to higher loads. With increasing film thickness the respective hardness values also decrease thus reflecting the growing influence of the soft cover.

In common sample preparation for the SEM layers as thick as necessary to ensure conductability, i.e., around a few 10 nm, are deposited. Consequently the deviation in hardness from the real value will be significant mainly in the low load regime where it will exceed the statistical scatter. Two corrections are suggested in the following section to compensate for that.

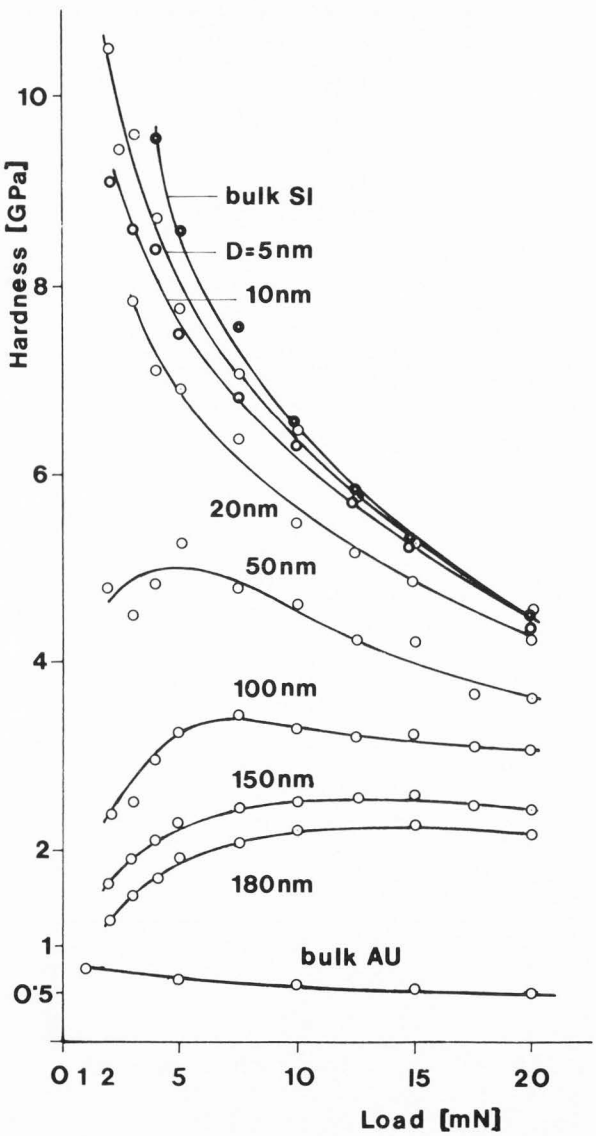

Fig.2 Hardness measurements on uncoated Si, on Si coated with Au-layers of different thicknesses D and on bulk Au.

Geometric correction

As already stated, the first approximation assumes an ultra-soft overlayer. Thus the film does not bear any portion of the force and only a pure geometric magnification of the imprint in the substrate will arise. In Fig.3a a more or

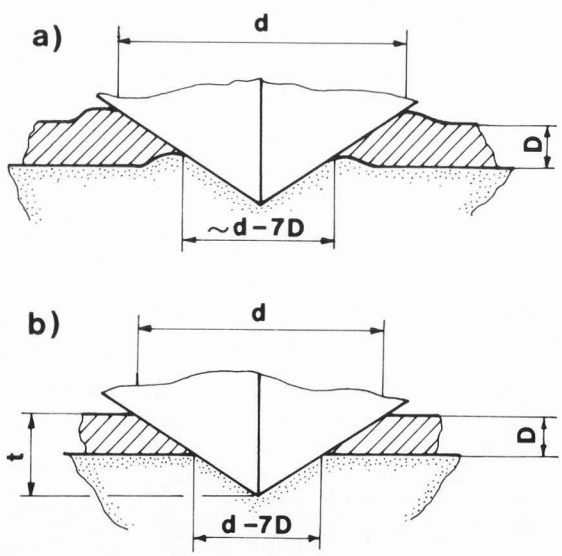

Fig.3a Model of the cross-section of an imprint; realistic shape.
b Simplified geometry of the cross-section of an imprint.
Film thickness D, imprint diagonal d, indentation depth t.

less realistic cross-section (but not to scale) of an imprint is shown. The bulge formed by the expelled material is not uniform along the circumference of the imprint square and is smallest near the corners. We may ignore it in our considerations because according to Vickers standards the diagonal has to be measured. Fig.3b shows the simplified geometry. It is evident that the diagonal of the imprint relevant for the substrate's hardness is magnified. As can be calculated from the Vickers tip geometry (square pyramid, angle between opposite planes 136°) the additive term amounts to 7 times (exactly 7.0006 times) the film thickness D. For correction we simply have to subtract it from the measured diagonal d. The equation for Vickers hardness H calculated from the imprint surface area A and the test load L:

$$H = L/A = 1.85\ L/d^2 \qquad (1)$$

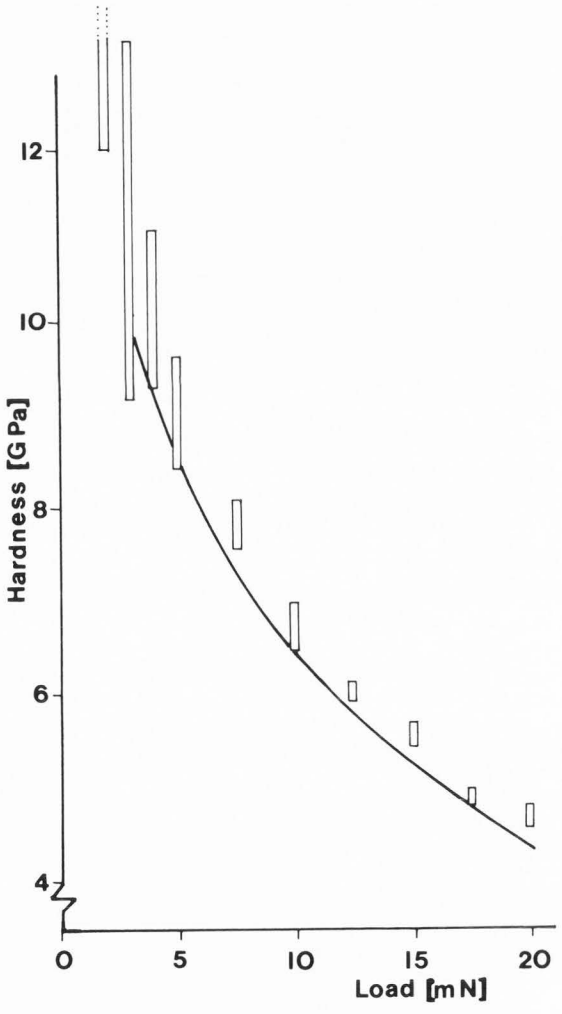

Fig.4 Results of the geometric correction. Hardness of uncoated Si (solid line) and corrected hardness values of Si coated with Au-films of different thicknesses D.

then yields a corrected value:

$$H^C = 1.85\ L/(d-7D)^2 \qquad (2)$$

The factor 1.85 results from expressing A by d^2 and by using the following units: GPa for hardness, mN for load, μm for all lengths. For convenience we abbreviate the relative extension of the imprint in the film:

$$7D/d = D/t = x \qquad (3)$$

(t...penetration depth) and obtain:

$$H^C = H/(1-x)^2 \qquad (4)$$

The results of such a correction for all film thicknesses are shown in Fig.4. In order to avoid a confusing plot of very close data only the range of the corrected values is presented. It can be seen from a comparison with Fig.2 that the correction is effective and leads to an excellent agreement with the graph for uncoated Si for all layer thicknesses. The remaining small systematic deviation towards higher values reflects the fact that the Au layer does have a certain hardness and thus actually reduces the imprint force into the substrate. The use of the total force in the calculation naturally must give such results.

Load correction

The second order correction ("load correction") is based on the geometric effect and on a distribution of forces among film and substrate. We assume (1) is valid also for the film and allows one to calculate the load carried by the film depending on the area in contact with the indentor. The total load L is distributed according to:

$$L = 2(H_f A_f + H_s A_s)/1.85 \qquad (5)$$

H_f, H_s...Hardness of film and substrate, respectively.

The area A_f and A_s are the corresponding contact areas between indentor and film or substrate, projected into the surface plane; their relations to the imprint diagonals are:

$$A_s \begin{cases} = (d-7D)^2/2 = d^2(1-x)^2/2 & x \leq 1 \\ = 0 & x > 1 \end{cases} \qquad (6)$$

and

$$A_f = d^2 x(2-x)/2 \qquad (7)$$

as can be seen from Fig.3.

After rearrangement of (5) to yield H_s and in combination with (1), (4), (6) and (7) we obtain:

$$H_s = (H - H_f x(2-x))/(1-x)^2 \qquad (8)$$

This equation includes the asymptotic case for large imprints ($x \to 0$) which gives:

$$H_s \to H \qquad (9)$$

i.e., the uncorrected value H approaches the true hardness of the uncoated substrate H_s. In the case x = 1 where the imprint extends only within the film, the trivial result $H = H_f$ is obtained.

The results which follow from the second order correction are plotted in Fig.5 together with the hardness of uncoated Si. For the gold cover a load-independent hardness of 500 MPa was assumed corresponding to the asymptotic value in Fig.2. The general systematic shift toward higher values as observed in the first order correction (Fig. 4) has now disappeared. For the sake of clearness only a selection of data is presented: all data omitted in the graph spread systematically between those of the extreme thickness values

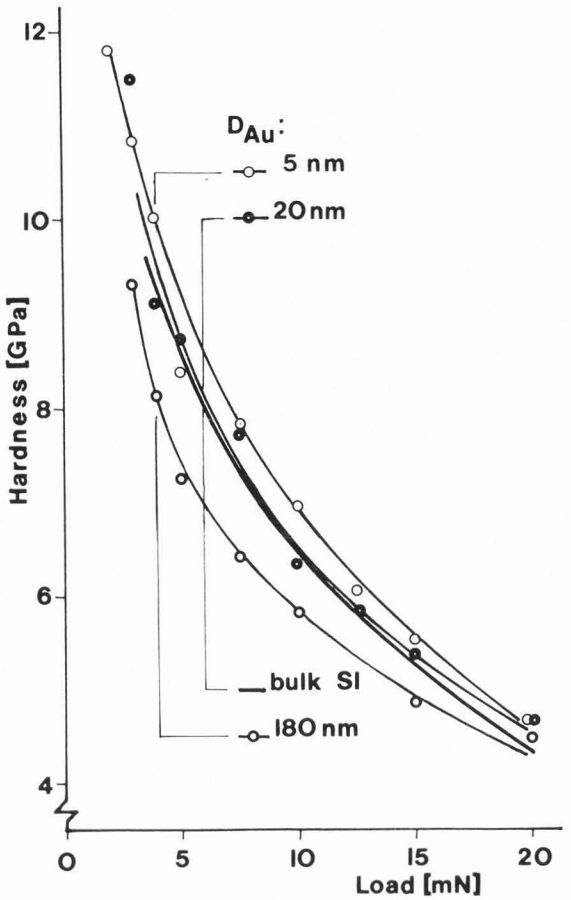

Fig.5 Result of the load correction. Hardness of uncoated Si (solid line) and corrected hardness values of Si coated with Au-films of different thicknesses D.

5 nm and 180 nm. For coverages up to 20 nm the correction leads to a hardness above the true value of Si. That means the Au layer's hardness was chosen too low. The opposite results are observed for films thicker than approximately 20 nm, i.e., Au was chosen too hard. We have to conclude that the Au films show a thickness varying hardness. In fact we can understand this behaviour if the internal stress in thin deposits is considered: coatings thin enough to consist of isolated islands are stressed compressively mainly by the surface and interface tension forces. When the deposit grows thicker the internal stress is inverted during coalescence and the tensile forces increase with thickness [1,8]. Considering the resistance such stressed films will offer a penetrating body we must obviously expect higher forces in the case of compression whereas a tensile bias will support indentation. This indeed resembles the observed behaviour because 20 nm corresponds roughly to that thickness where Au films, deposited at room temperature, coalesce and changes the sign of internal stress [1].

In conclusion we may recommend the geometrical correction of the imprint size as a quick and satisfactory procedure. Nevertheless, in situations where the geometric model is strongly insufficient, i.e., comparable hardness of film and substrate one ought to be cautious and thus use coatings only as thin as necessary in order to reduce their influence as far as possible. We found that the second order correction ("load correction") can suffer from the lack of appropriate hardness data of the coating but on the other hand it can provide information on a variation of mechanical properties with thickness in thin films.

Acknowledgement

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Discussion with Reviewers

B. E. Artz: The geometric correction factor $1/(1-x)^2$ appears to over correct the measured hardness as expected. The factor contains the term, x, which depends directly on the film thickness and inversely on the square root of the load. How accurately are the thickness and load

known? Could a small error in the film deposition rate have a significant effect on the amount of over correction and consequently your analysis of this effect?

D. L. Davidson: Nothing is said about the films, how they were deposited, and especially how their thickness was determined. There is almost always error in this process, and it is unlikely that the even values for thickness shown on the figures was, in fact, the actual value of the film-thickness.

Authors: Thickness was controlled via sputter time. Based on a careful calibration done by optical interference thickness measurements (Tolansky interferometry) on thick layers (0.3 µm thick) the error was confined to 5%. The load reproducibility is 5.10^{-5}N and thus the statistical load error is only 2% of the lowest load applied. An error in deposition rate will result in an error of film thickness and will enter the correction calculations. The effect of a small error, however, is insignificant for $x \ll 1$, i.e., for layer thicknesses small compared to the penetration depth. Even for x up to 0.5 (the most unfavourable case in our experiments) which is definitely far from the practical conditions in SEM investigations a thickness error of 5% will cause an error in hardness of only 10%.

B. E. Artz: You stated that the gold film should have a hardness of 400 - 600 MPa. Judging from your plot the film value could lie anywhere in this range. Using 500 MPa you get a transition thickness of 20 nm. What happens when you use 400 (or 600) MPa? Do you still get reasonable transition values?

Authors: The transition thickness value will only be slightly affected. If we assume either 400 or 600 MPa for the load correction the deviation from the value obtained with 500 MPa will be less than 2% for films up to 50 nm. With thicker films obviously the respective deviations are larger but are still less than 6%.

D. L. Davidson: Why does the hardness increase so dramatically as the load decreases for uncoated Si?

Authors: The hardness increase with decreasing load is a well known behaviour for homogeneous metals and semiconductors, but there is no general agreement on its interpretation. Possible causes for the effect are either "...local extreme work hardening..." [9] or "...an increase in the stress necessary to operate dislocation sources..." [6]. Violating the similarity condition of the imprint geometry at different loads due to the finite tip radius may also contribute.

D. L. Davidson: In discussing the effects seen, the stress internal to the film is thought to be a factor. This stress should be dependent upon how the film is deposited: has that been investigated? Why has friction between the indenter and specimen been ignored in this consideration?

D. L. Davidson: Even extremely small amounts of lubricants, or contamination on the indenter, should be important to the results. Has this been observed or investigated?

Authors: Such effects, indeed, occur, but it was not our intention to investigate them in detail. Regarding friction, lubricants, contaminations and film deposition parameters we may consider them as constant throughout the experiments.

D. L. Davidson: The island concept for very thin films has been evolved to explain results for very thin films. Is there any direct evidence from your work to support this concept?

Authors: In our work there is no direct evidence. Our conclusions were drawn from our previous experiments of Au films on glass and from the literature quoted [1,8].

M. J. Yacaman: What is the influence of the microstructure of the Au and Au-Pd layer?

M. J. Yacaman: It is well known that slight changes on deposition parameters can produce dramatic changes on this metal film structure. This is particularly true for Au and Au-Pd. How can one assure in the SEM (without using high resolution TEM) that the layer is introducing a reproducible effect?

Authors: Internal stress due to an island or a continuous structure of the film was discussed. Any other structural effects were not studied but constancy of deposition parameters was carefully observed.

M. J. Yacaman: Do you expect any influence of interphase effects? For instance dislocation structures in the interphase film-substrate? Also some oxide layer produced during deposition.

Authors: Both correction procedures are based on simplifying conditions; therefore interface or dislocation structure effects were not considered.