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Influence of modulus-to-hardness ratio and harmonic parameters on continuous stiffness measurement during nanoindentation

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ABSTRACT

Dynamic nanoindentation is a popular method for continuously probing the mechanical properties of a sample as a function of depth. It is shown here that special caution should be exercised when testing materials with high modulus-to-hardness ratios (E/H) at fast loading rates, as the choice of harmonic parameters can result in a significant underestimation of the contact stiffness and hence the elastic modulus. The origin of this behavior is traced back to a bias occurring during signal processing by the lock-in amplifier. The possible consequences of flawed measurements are highlighted and a practical method for detecting possible occurrence from the phase angle signal is presented.

1. Introduction

Dynamic nanoindentation measurements, first introduced by Pethica and Oliver in the late 1980s [1,2], have become very popular over the years, mostly because they allow extracting much more information from a single experiment than quasi-static nanoindentation. Dynamic measurement, also called continuous stiffness measurement (CSM), relies on imposing a small but fast oscillation onto the main loading signal. This is tantamount to performing a multitude of partial unloadings, which allows one to determine the contact stiffness quasi-continuously. Dynamic measurements have been widely used for investigating variations in hardness and Young's modulus through the sample thickness, which is of great interest in the case of graded and coated samples. In recent years, the continuous stiffness method has also been pivotal to the development of new methods aimed at investigating advanced mechanical properties such as the local strain-rate sensitivity [3,4] and creep stress exponent [5,6]. It has also been used for performing accurate measurements on submicrometer thin films [7–9]. An overview of its applications can be found in a recent publication by Durst et al. [10].

Despite its widespread application, the reliability of dynamic nanoindentation has sometimes been questioned. For example, Cordill et al. [11] suggested that the periodic oscillation could induce fatigue damage and therefore modify the mechanical properties of the sample being investigated. This hypothesis was confirmed by subsequent observations [12], albeit only for much larger oscillations than are usually used during nanoindentation. While investigating the indentation size effect, Durst et al. [13] observed discrepancies between dynamic measurements and quasi static tests at very small indentation depths. These were later accounted for by Pharr et al. [14] as flaws in property measurement caused by the use of continuous stiffness measurement techniques. Pharr et al. showed that for materials with a high modulus to hardness ratio (E/H), the assumption of a linear unloading does not hold true for oscillations larger than a few nanometers, resulting in a systematic bias in the evaluated stiffness. They consequently advocated reducing the amplitude of the oscillation to the minimum allowed by the noise of the measurement system.

In the present paper, we focus on a different type of bias also specific to materials with a high E/H ratio. Early observations by Vacchani et al. [15] pointed out that the outcome of spherical

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dynamic measurements on single crystalline aluminum could be dependent on the selected harmonic frequency. We show here that the more commonly used Berkovich indenter geometry is affected as well and that the issue is related to signal processing of elastic-plastic data by the lock-in amplifier.

2. Materials and methods

Nanoindentation experiments were performed with two similar Keysight (Chandler AZ, USA) Nanoindenter G200 systems with the continuous stiffness measurement (CSM) option. The XP head used for this study was equipped with a Berkovich diamond indenter fabricated by Synton MDP (Nissau, Switzerland). In one set of experiments, the Berkovich indenter was substituted with pyramidal punches of different opening angles, in order to investigate the effect of the indenter geometry on the stiffness evaluation. The centerline-to-face angles of these indenters were: 35.5° (cube-corner), 50°, 65.3° (Berkovich) and 80°, with further details to be found in Ref. [16]. All measurements were performed at a constant nanoindentation strain-rate $\dot{P}/P = 0.05\ s^{-1}$, which means that the load was increased exponentially over time. The harmonic frequency and amplitude (also called harmonic displacement) of the superimposed CSM oscillations were systematically varied between $f = 10,\ 20,\ 40$ Hz and $\Delta h_{RMS} = 1, 2, 4$ nm. Please note that the latter values are root mean square values, which means that the amplitude of the signal is actually $\Delta h = \sqrt{2}\cdot\Delta h_{RMS}$ and the peak-to-peak amplitude $\Delta h_{peak} = 2\sqrt{2}\cdot\Delta h_{RMS}$. The continuous stiffness evaluation by the Nanoindenter G200 is performed by a 7225NI (Ametek, USA) lock-in amplifier. An oscilloscope was connected to its “SIG MON” output in order to observe the pre-conditioned displacement signal fed to the lock-in amplification unit. For such measurements, a larger harmonic amplitude of $\Delta h_{RMS} = 8$ nm was used in order to achieve a sufficient signal-to-noise ratio.

The samples investigated in this study are listed in Table 1. They display a wide range of E/H ratios spanning between 8 and 231, which encompasses all common materials [17]. Special attention was paid to the three samples highlighted in bold. They include the single crystalline aluminum (sx-Al) and fused silica calibration standards that were provided with the nanoindenter system. In addition, an ultrafine grained aluminum sample was produced by severe plastic deformation of an AA1050 aluminum sheet through 6 cycles of the accumulated roll-bonding (ARB) process. All three materials display similar Young's moduli of around 70 GPa but very different hardnesses and thus E/H ratios. sx-Al and fused silica account for the extreme behaviors, while ufg-Al is more representative of most metals.

The simulated signals shown in the next section were generated by Python scripts written for evaluation purposes using the SciPy library for numerical equation solving.

3. Experimental observations of deviation in contact stiffness

3.1. General observations

As indicated in the previous section, the harmonic parameters in this study were systematically varied in order to quantify their effect on Berkovich indentation. Fig. 1 summarizes the contact stiffness measurements that were achieved on the three main materials. The most interesting findings concern sx-Al, as shown in Fig. 1 (top). In agreement with the previous observations from Vacchani et al. on spherical nanoindentation [15], the experiments show that the measured contact stiffness strongly depends on the harmonic frequency and amplitude used in the test, specifically, a low frequency or a small amplitude leads to smaller stiffness values than their larger counterparts. This behavior is not specific to sx-Al, but appears in less pronounced ways in other materials, too. The deviations visible in Fig. 1 appear to occur mostly at large penetration depth.

In order to understand this behavior, the sx-Al data was replotted as the Oliver-Pharr contact depth h_c against the Oliver-Pharr contact radius a_C calculated as:

$$h_c = h - \varepsilon\frac{P}{S} \tag{1}$$

$$a_c = \frac{S}{2\beta\cdot E_R} \tag{2}$$

taking the usual values of $\varepsilon = 0.75$ and $\beta = 1.0$ as well as the reduced moduli E_R values determined from indentations with $f = 40$ Hz and $\Delta h_{RMS} = 4$ nm. The important feature of a plot of h_c vs. a_c plot is that it should always, independent of the tested material, yield the same curve corresponding to the geometrical profile shape of the indenter (see Ref. [18]).

As shown in Fig. 2, this is not the case in the experiments on sx-Al. Specifically, there is good agreement between the measured and the actual indenter shapes up to a depth of approximately 1000 nm, which suggests that the continuous stiffness measurement is valid to these depths. However, for deeper indentations, the difference between the measured and actual values grows increasingly large. Interestingly, the expected value is recovered upon halting the indenter and holding for a period of time at fixed load (see holding segment in Fig. 2). These observations strongly suggest that the deviation in stiffness is a function of the loading rate, as this parameter grows exponentially during an indentation at constant strain-rate but falls to zero during the hold segment.

3.2. Effect of loading parameters

A thorough investigation of this effect was performed based on a quantitative evaluation of the difference between the measured

Table 1
Young's modulus to hardness E/H ratios calculated from measurements at $f = 60$ Hz and $\Delta h_{RMS} = 4$ nm RMS amplitude, averaging between 1500 and 2500 nm indentation depth.

Material	Abbrev.	Origin	E/H ratio
Fused silica	**FS**	Reference sample of Nanoindenter G200	**8**
Nanocrystalline nickel	nc-Ni	Pulsed electro-deposited Ni	41
Coarse grained nickel	cg-Ni	Recrystallized galvanic Ni	86
C45 steel	Steel	Pearlitic and ferritic recrystallized steel	96
Ultrafine grained aluminum	**ufg-Al**	Accumulated roll-bonded AA1050, 6 cycles	**125**
Coarse grained copper	cg-Cu	Accumulated roll-bonded p-Cu, recrystallized	172
Single crystalline copper	sx-Cu	Commercial purity, unspecified orientation	197
Single crystalline aluminum	**sx-Al**	Reference sample of Nanoindenter G200	**231**

Bold: reference materials.