Supporting information

S1. Theory used for calculation of Young's modulus (*E*) and indentation hardness (*H*) from DCM-CSM nanoindentation data

According to the dynamic CSM mode, *E* and *H* are determined as a function of the surface penetration depth (*h*). The displacement response of the indenter at the excitation frequency (usually 75 Hz) and the phase angle between the two are measured continuously as a function of depth. Determination of the in-phase and out-phase portions of the response results in calculation of the contact stiffness, *S*, as a continuous function of displacement.

The calculation principle is based on a simple harmonic oscillator subject to a force oscillation:

$$K = \left(S^{-1} + K_f^{-1}\right) + K_s \tag{1}$$

where *K* is equivalent stiffness (it includes the stiffness of the contact *S*), K_f is load frame stiffness, and K_s is the stiffness of the support springs.

If the imposed driving force is $F = F_0 \exp(i\omega t)$ and the displacement response of the indenter is $h(\omega) = h_0 \exp(i\omega t + \varphi)$, the contact stiffness (*S*) of the sample and damping of the contact ($D_s\omega$) are given by:

$$S = \left(\frac{1}{\frac{F_0}{z_o}\cos\phi - (K_s - m\omega^2)} - \frac{1}{K_f}\right)^{-1} \tag{2}$$

$$D_s\omega = \frac{F_0}{z_0} sin\phi - D_i\omega \tag{3}$$

In a CSM mode, the excitation frequency (ω) is a set value. During experiments, we measure the displacement amplitude (z_0), phase angle (φ), and excitation amplitude (F_0). The damping factor of the indenter head (D_i), K_s, and parameter *m* are machine factors, determined by analyzing the system dynamic response when the indenter is hanging free. The elastic modulus of the test sample, E, is determined from the reduced modulus (E_r), given by:

$$E_r = \frac{\sqrt{\pi}}{2\beta}\frac{S}{\sqrt{A_c}} \tag{4}$$

where A_c is the projected contact area and β is a constant that depends on the indenter's geometry (β = 1.034 for a Berkovich tip).

Referring to Oliver and Pharr's theory [28, 29], the E can be extracted from the following equation:

$$\frac{1}{E_r} = \frac{1-v_s^2}{E_s} + \frac{1-v_i^2}{E_i} \tag{5}$$

where E_i and v_i represent the elastic modulus and Poisson's ratio of the indenter (for the diamond tip: E_i = 1141 GPa and v_i = 0.07), and E_s represents the elastic modulus of the sample.

The indentation hardness, H, is calculated as the ratio between the applied load (P) and projected contact area (A_c):

$$H = \frac{P}{A_c} \tag{6}$$

The projected contact area is calculated by evaluating an empirically determined area function of the contact depth, h_c:

$$A = f(h_c) \tag{7}$$

$$h_c = h_{max} - \varepsilon\left(\frac{P_{max}}{S}\right)$$

where h_{max} is the maximal penetration depth, ε is a geometric constant of the Berkovich indenter, and (P/S) expresses the extent of elastic recovery.