SAMPLE FABRICATION

The aluminum-coated soda lime glass substrates were cleaned by soaking in isopropanol and acetone for ten minutes each, and then rinsed with deionized (DI) water and dried under nitrogen flow. Following this, the substrates were coated with a 20-nm-thick silica layer via chemical vapor deposition. The colloidal suspensions for these samples were purified by centrifuging the suspension at 4000 rpm, discarding the supernatant and redispersing the particles in DI water or ethanol. This purification procedure was repeated three times for all samples, except in the case of Sample 2, where the colloidal suspension was centrifuged once. Details of the self-assembly parameters and the layer thicknesses characterized for the multilayer samples in this study are listed in Table S1. The samples with the microsphere suspension in water were left to dry in an oven at 75°C, while the samples immersed in the ethanol suspension were placed under a plastic container to avoid external airflows and left to dry under ambient laboratory conditions.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Concentration (% v/v)</th>
<th>Solvent</th>
<th>Drying Environment</th>
<th>Centrifuged</th>
<th>Layer Thicknesses Characterized</th>
<th>Monolayer Resonance Detected</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.2</td>
<td>Ethanol</td>
<td>Ambient</td>
<td>3x</td>
<td>1, 2, 3, 4, 5, 7, 8, 12</td>
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</tr>
<tr>
<td>2</td>
<td>0.01</td>
<td>DI Water</td>
<td>Oven, 75°C</td>
<td>1x</td>
<td>1, 2, 3, 4, 5, 9, 12</td>
<td>Yes</td>
</tr>
<tr>
<td>3</td>
<td>1</td>
<td>Ethanol</td>
<td>Ambient</td>
<td>3x</td>
<td>6, 7</td>
<td>No</td>
</tr>
<tr>
<td>4</td>
<td>0.5</td>
<td>Ethanol</td>
<td>Ambient</td>
<td>3x</td>
<td>2, 3, 4, 5, 6, 7, 8, 9</td>
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</tr>
<tr>
<td>5</td>
<td>0.3</td>
<td>Ethanol</td>
<td>Ambient</td>
<td>3x</td>
<td>2, 3, 4, 5, 6, 7, 9</td>
<td>No</td>
</tr>
<tr>
<td>6</td>
<td>0.01</td>
<td>DI Water</td>
<td>Oven, 75°C</td>
<td>3x</td>
<td>2, 3, 4</td>
<td>No</td>
</tr>
<tr>
<td>7</td>
<td>0.01</td>
<td>DI Water</td>
<td>Oven, 75°C</td>
<td>3x</td>
<td>2, 3, 4, 5, 6</td>
<td>No</td>
</tr>
</tbody>
</table>

LASER ULTRASONIC SETUP DETAILS

For the measurements on Sample 2, the pump beam was focused to an elliptical spot (436 µm x 76 µm at the 1/e² intensity level), whereas for all the other samples, the pump beam was focused to a 200 µm diameter circular spot. The interferometric signals were averaged over 10⁴ pump pulses for all samples, except in the case of the measurements on the monolayer, five-, seven-, eight- and twelve-layer regions of Sample 1, which were averaged over 10⁶ pump pulses to achieve an improved signal-to-noise ratio.

SIGNAL PROCESSING PROCEDURE

Time-resolved signals were recorded at a positive and negative phase setting by varying the optical path difference between the probe and the reference beams via a rotating fused silica window placed in the path of the probe beam. The net signal was obtained by subtracting the averaged waveforms collected at positive and negative phase settings. A segment of 0.75 µs of the signal starting from the sharp initial rise (corresponding to the arrival of the pump pulse) was used for further signal processing. This segment of the signal was zero-padded after the oscillations completely decayed below the noise floor, differentiated with respect to time to remove the thermal decay component from the signal, and normalized with respect to its maximum amplitude. A Fast Fourier Transform (FFT) was then applied to the normalized signal.

WORK OF ADHESION AT THE SPHERE-SUBSTRATE AND SPHERE-SPHERE INTERFACES

We estimate the work of adhesion between identical polystyrene microspheres \( w_{p-p} \) and that between the polystyrene microspheres and the silica-coated substrate \( w_{p-S} \) in terms of the Hamaker constant \( A \), using \( w = \frac{A}{12\pi D_0^3} \).
where \( T = 293 \text{ K} \) is room temperature, \( \nu_{e1} = 2.3 \cdot 10^{15} \text{ Hz} \) is the main electronic absorption frequency of polystyrene in the UV, \( k \) is Boltzmann’s constant, \( h \) is Planck’s constant, \( \epsilon_1 = 2.55 \text{ F/m} \) and \( \epsilon_3 = 1.0 \text{ F/m} \) are the permittivities of polystyrene and air, respectively, and \( n_1 = 1.56 \) and \( n_3 = 1.00 \) are the refractive indices of polystyrene and air, respectively. All of the preceding constants are tabulated in Ref. 1. Using Eq. 1, we calculate \( A_{131} = 6.53 \cdot 10^{-20} \text{ J} \). This gives a work of adhesion \( w_{p-P} = 0.06 \text{ J/m}^2 \) between the polystyrene microspheres.

Similarly, we use Eq. 1 to calculate the Hamaker constant for two silica surfaces (denoted as medium 2) interacting in air (medium 3), with the corresponding material properties of silica: permittivity \( \epsilon_2 = 3.8 \text{ F/m} \), refractive index \( n_2 = 1.45 \), and electronic absorption frequency in the UV \( \nu_{e2} = 3.2 \cdot 10^{15} \text{ Hz} \) [1]. We obtain \( A_{232} = 6.36 \cdot 10^{-20} \text{ J} \). We use a combining relation to obtain an approximate value for the Hamaker constant between polystyrene and silica, \( A_{132} \approx \sqrt{A_{131}A_{232}} \), such that \( A_{132} \approx 6.44 \cdot 10^{-20} \text{ J} \). The corresponding work of adhesion between polystyrene and silica is \( w_{p-S} = 0.06 \text{ J/m}^2 \).

**CALCULATION OF THE EFFECTIVE INTERLAYER CONTACT STIFFNESS USING THE DMT MODEL**

The normal contact stiffness between two colloidal particles \( G_{N, DMT} \) is derived from the linearized force-displacement curve prescribed by the DMT model [2], and is expressed as, 

\[
G_{N, DMT} = \frac{3}{2} (2\pi w_{p-P} R_e E_{P-P}^2)^{1/3},
\]

where \( w_{p-P} \) is the work of adhesion between the two polystyrene surfaces, \( R_e \) is the effective radius (equal to half the radius of the particle), and \( E_{P-P} \) is the effective modulus of the contact and is defined in terms of the Young’s modulus \( E \) and the Poisson’s ratio \( \nu \) of the particle, \( E_{P-P} = \frac{3}{4} \frac{E}{1-\nu^2} \). The effective interlayer normal contact stiffness \( G_{c, DMT} \) is then derived by accounting for the contributions of the three normal \( G_{N, DMT} \) and transverse \( G_S \) contact springs each in an HCP unit cell along the out-of-plane direction to be 

\[
G_{c, DMT} = G_{N, DMT}(2 + \nu^*),
\]

\( \nu^* = \frac{G_S}{G_{N, DMT}} = \frac{2^{1-\nu}}{2-\nu} \) is the ratio of the interparticle shear and normal contact stiffnesses [3, 4].

**ESTIMATION OF DISPLACEMENT AMPLITUDE IN THE ALUMINUM FILM**

We estimate the thermoelastic strain in the aluminum film induced by the pump laser pulse using the relation in reference [5], \( \eta_0 = 3k_0\alpha_L(1 - R)Q/(\alpha_p C_\rho_0 c_L^2) \), where \( k = 67.6 \text{ GPa} \) is the bulk modulus, \( \alpha_L = 23.1 \text{ mm/m-K} \) is the linear thermal expansion coefficient, \( \rho_0 = 2.7 \text{ g/cm}^3 \) is the density, \( C = 2.46 \text{ MJ/m}^3 \text{ K}^{-1} \) is the volumetric heat capacity, \( R = 0.92 \) is the optical intensity reflection coefficient, \( c_L = 6.8 \text{ nm} \) is the optical penetration depth, \( c_L = 6300 \text{ m/s} \) is the longitudinal wave speed in aluminum [6], \( \alpha_p \) is the area of the pump spot, and \( Q = 7 \mu \text{J/pulse} \) is the pump pulse energy at the sample. From the thermoelastic strain, we estimate the displacement in the aluminum film to be \( u_z = \eta_0 \xi_0 = 340 \text{ pm} \).