Electronic Supplementary Information For

Anisotropic Lattice Expansion of Threedimensional Colloidal Crystals and Its Impact on Hypersonic Phonon Band Gaps

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<u>Fitting the reflectance spectrum</u>



Figure S1. Experimental stop band peaks (o) measured at five different incident angles and the fitted curve using nonlinear least-squares algorithm. Note: the residual sum of squares (R²) of the fitting was close to one.

Derive vibration frequency from spring-hard-sphere model

To derive the vibration frequency of PS particle, we assume the colloid crystal as hard spheres connected by polymer springs. Let's first consider such a particle connected with two springs, whose spring constants are k_1 and k_2 , respectively (Fig. S2a).

It is easy to shown that with a small displacement dx, the force acting on the sphere is

$$d^{\vec{F}} = -\{k_1(\vec{l}_1 + d\vec{x}) - k_1\vec{l}_1\} + \{k_2(\vec{l}_2 + d\vec{x}) - k_2\vec{l}_2\} = -(k_1 + k_2)d\vec{x}$$
(S1)

Now consider a 3D colloid crystal as shown in Fig. S2b. Each sphere is connected with six inplane springs, and six cross-plane springs, so

$$d^{\vec{F}} = -\left(\sum k_i + \sum k_c\right)d\vec{x}$$
(S2)

The frequency becomes

$$v'/_{v^0} = \{ \left(\sum_{k_i}^{k_i} + \sum_{k_c}^{k_c} \right)' / \left(\sum_{k_i}^{k_i} + \sum_{k_c}^{k_c} \right)^0 \}^{0.5}$$
(S3)

If the PDMS chain acts as an ideal spring, the spring constant should be proportional to the inverse of its length. Without losing generality, we assume the spring constant is related to the sphere-to-sphere distance as

$$k \propto l^{-\alpha} \tag{S4}$$

Equation S3 and S4 give

$$v'_{v^{0}} = \{ \frac{\sum l_{i}^{-\alpha} + \sum l_{c}^{-\alpha}}{\sum l_{i}^{-\alpha} + \sum l_{c}^{-\alpha}} \right)^{\prime} / \left(\sum l_{i}^{-\alpha} + \sum l_{c}^{-\alpha} \right)^{0.5}$$
(S5)

Without any swelling, the colloid crystal is an fcc crystal with $l_i = l_c$. Because the swelling only happens along the cross-plane direction without any change on the in-plane directions, we get

$$v'/_{v^0} = \frac{1}{\sqrt{2}} [1 + (l'c'/_{l_0})^{-\alpha}]^{0.5}$$
 (S6)

From Fig. S2c, we get the cross-plane sphere-to-sphere distance after swelling is

$$l_{c}^{'} = (\frac{1}{3} + \frac{2}{3}\sigma^{2})^{0.5} l_{0}$$
(S7)

Equation S6 and S7 finally gives

$$v'/_{v^0} = \frac{1}{\sqrt{2}} [1 + (1/3 + 2/3\sigma^2)^{-0.5\alpha}]^{0.5\alpha}$$
 (S8)



Figure S2. (a) Force acting on a sphere connected with two springs. (b) Spring-hard-sphere model for fcc colloidal crystal. (c) Sphere-to-sphere distance after the swelling along cross-plane direction.