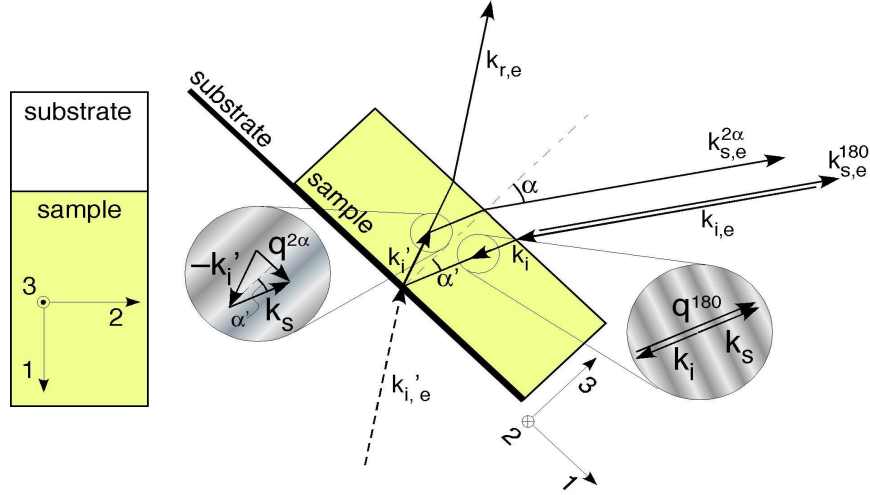


# Supporting Information



**Scheme S1.** Backscattering setup for Brillouin scattering.  $k_i$ ,  $k_s$ , and  $k_r$  are the incident, scattered, and reflected beams; 1 and 2 directions are in the growth plane of the sample, and direction 3 is normal to the growth plane;  $\alpha$  is the angle  $k_i$  makes with the 3 direction;  $q$  is the phonon propagation direction.

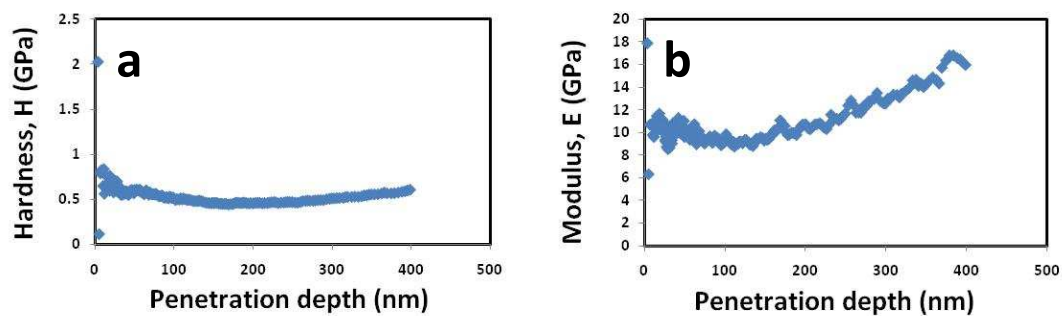
Scheme S1 shows this frequency shift measured at different locations on the film, here the inner and outer peaks are the  $2\alpha$  and  $180^\circ$  peaks, respectively. This frequency shift,  $\Delta f$ , in scattered light is proportional to the phonon velocity,  $v$ , using Equations 1 and 2

$$v_{180} = \frac{\lambda \Delta f_{180}}{2n}, \quad (1)$$

$$v_{2\alpha} = \frac{\lambda \Delta f_{2\alpha}}{2 \sin \alpha}, \quad (2)$$

where  $\lambda$  is the wavelength of the incident beam,  $n$  is the refractive index and  $\alpha$  is the angle between the incident beam and surface normal ( $45^\circ$ ). By combining Equations 1 – 2, along with the sample density, the longitudinal elastic modulus of the Kevlar film can be determined according to

$$c_{11} = \rho v^2 \quad (3)$$



**Figure S1.** Typical results from nanoindentation experiments for PDDA/ANF<sub>300</sub> LBL film with maximum depth set at 400 nm showing clear depth-sensing characteristic with depth bigger than 200 nm. a) Represents the hardness as a function of penetration depth and d) corresponding modulus. These results are from the loading part of the experiment.

Wave number (cm <sup>-1</sup> )	Assignment
1181	C-C ring stretching
1277	C-C ring stretching
1327	C-H in plane bending
1514	C-C ring stretching
1569	N-H bending, C-N stretching
1610	C-C ring stretching
1648	C=O stretching, C-N stretching, N-H bending

**Table S1.** The assignment of Kevlar active modes.