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Vibrational modes of silicon metalattices from atomistic and finite-element calculations

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CENTER FOR nanoscale science

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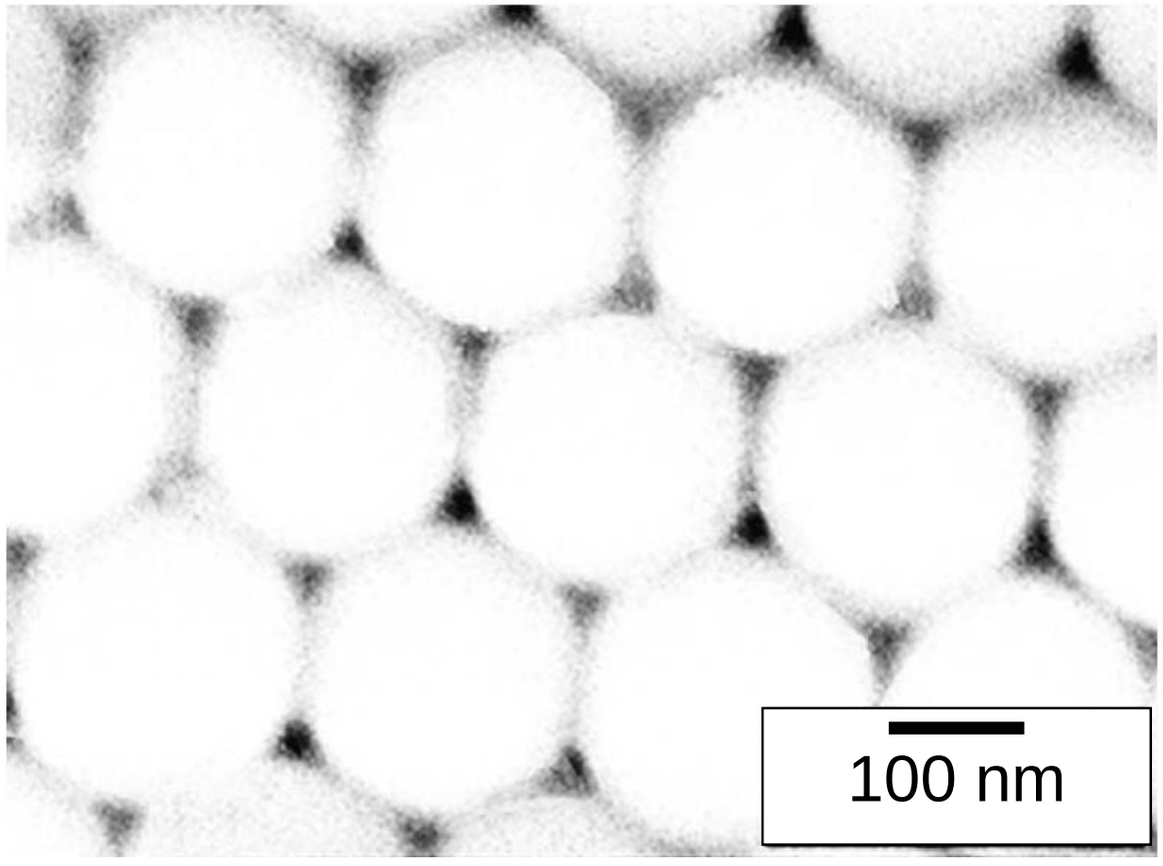
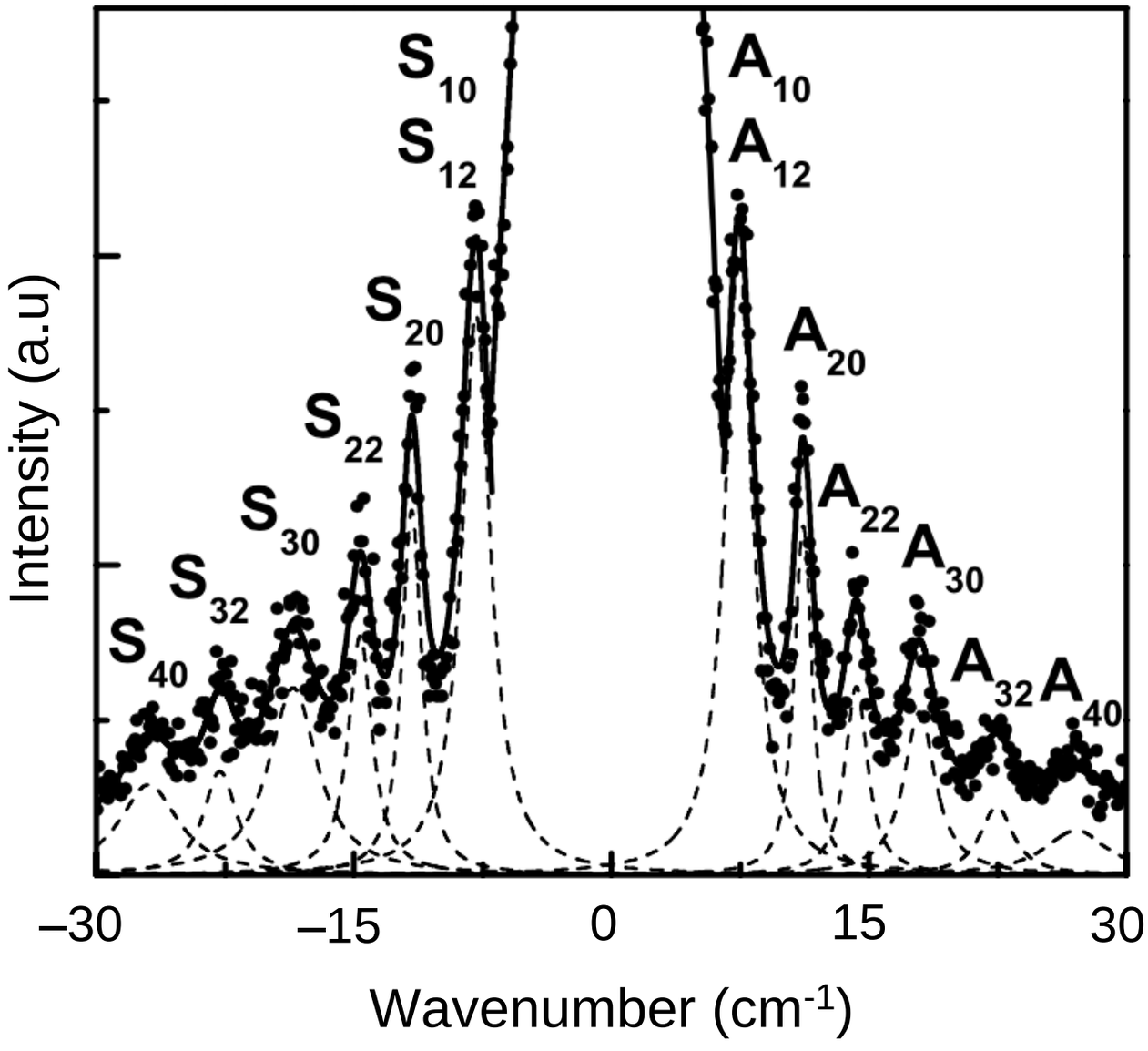
COLLEGE OF EARTH AND MINERAL SCIENCES

Raman spectroscopy at the nanoscale

Vibrational spectroscopy provides a highly sensitive probe of structural properties at the nanoscale.

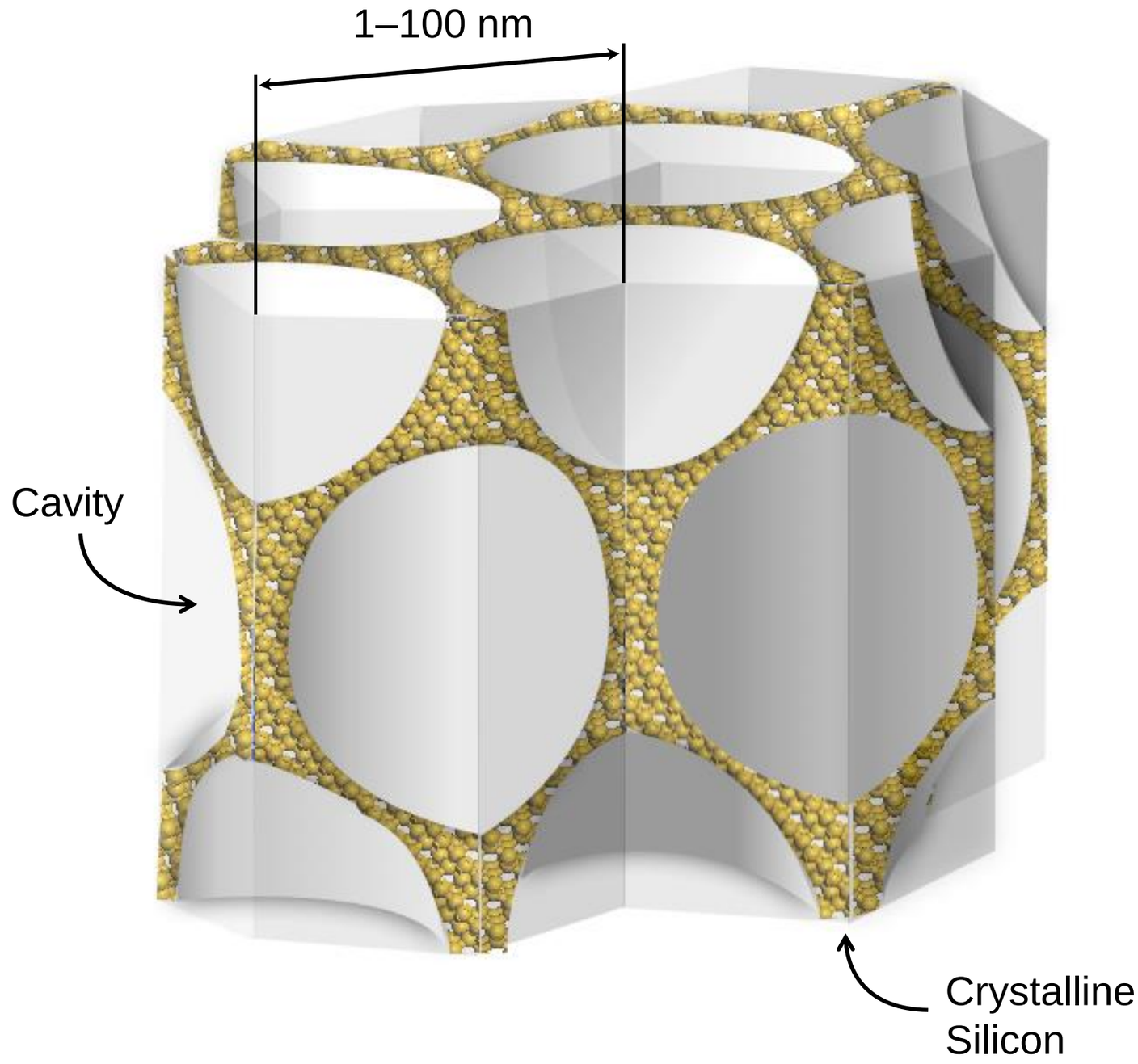




Kuok *et al., Phys. Rev. Lett.* 90, 25 (2003)

What is a metalattice?

Metalattices are 3D nanoscale structures consisting of crystalline aggregates, periodically repeated on the scale of 1-100nm.



Han and Crespi, *Phys. Rev. Lett.* **86**, 696 (2001)

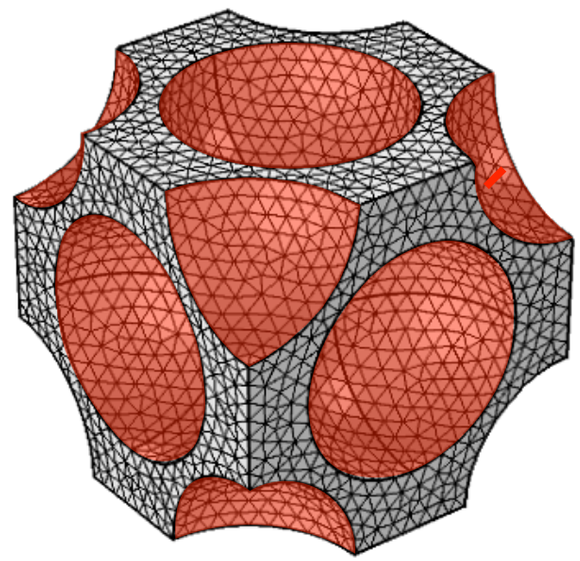
How to calculate acoustic modes

Acoustic wave equation:

$$\rho \frac{\partial^2 u_i}{\partial t^2} = \sum_{jkl} C_{ijkl} \frac{\partial^2 u_k}{\partial x_l \partial x_j}$$

Boundary conditions:

$$\boldsymbol{\sigma} \cdot \hat{\boldsymbol{n}} = 0$$



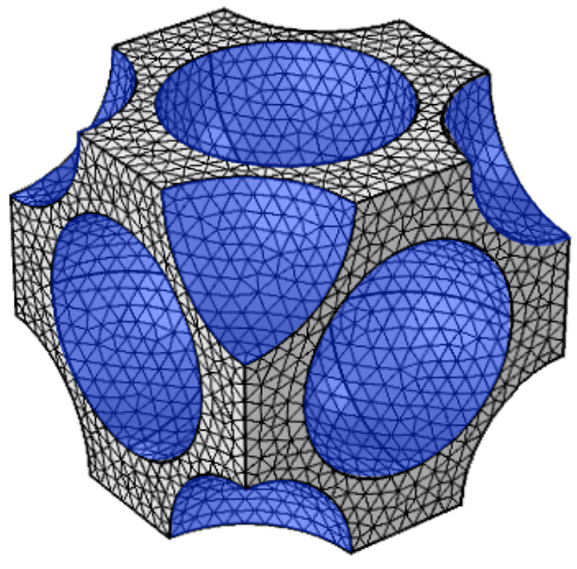
How to calculate optical modes

Optical wave equation:

$$\rho \frac{\partial^2 u_i}{\partial t^2} = -\rho \omega_0^2 u_i + \sum_{jkl} D_{ijkl} \frac{\partial^2 u_k}{\partial x_l \partial x_j}$$

Boundary conditions:

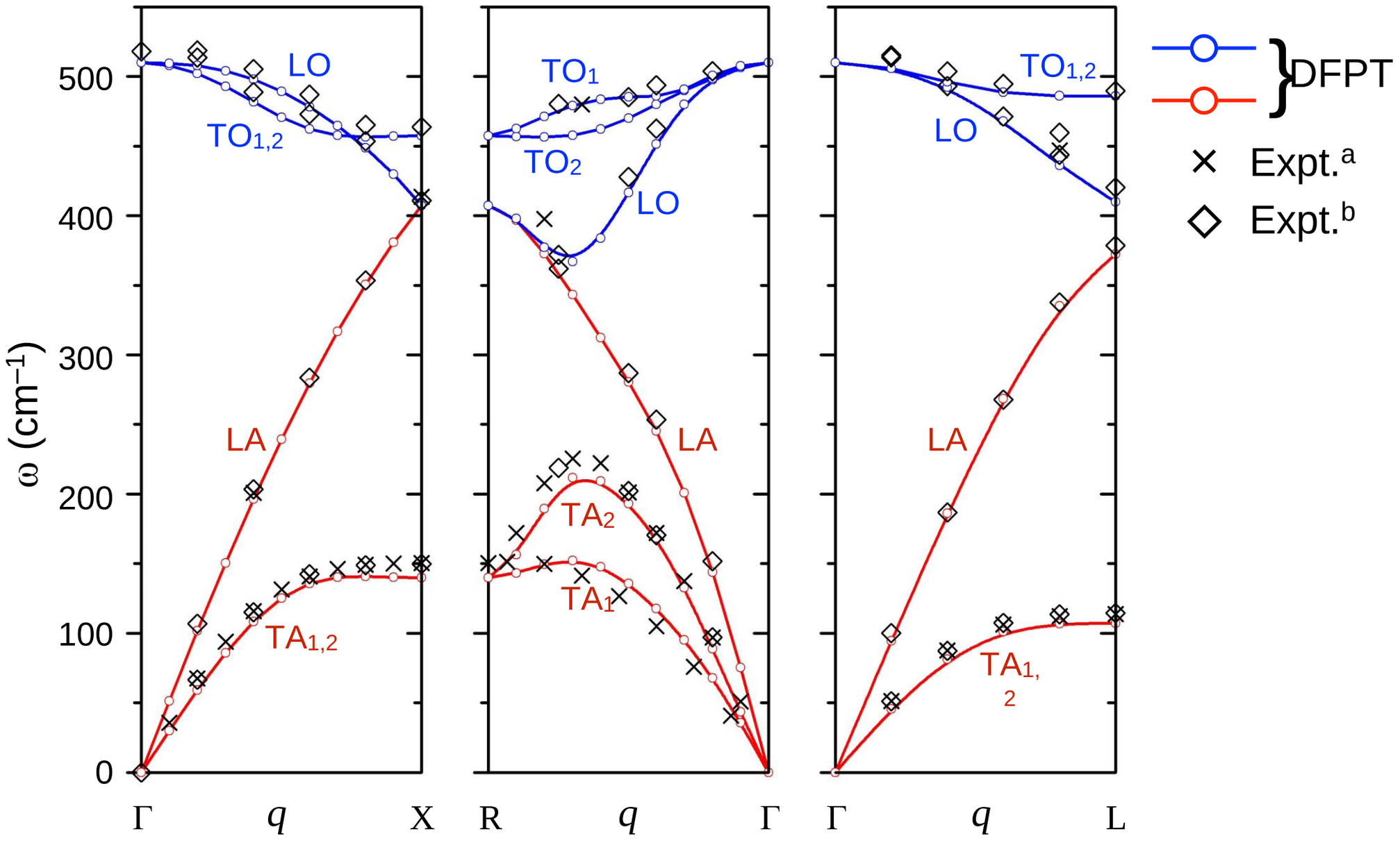
$$\boldsymbol{u} = 0$$



We need both acoustic and optical elastic coefficients C_{ijkl} and D_{ijkl}.

Thonhauser and Mahan, *Phys. Rev. B* **69**, 075212 (2004)

Phonon spectrum of silicon from density-functional perturbation theory



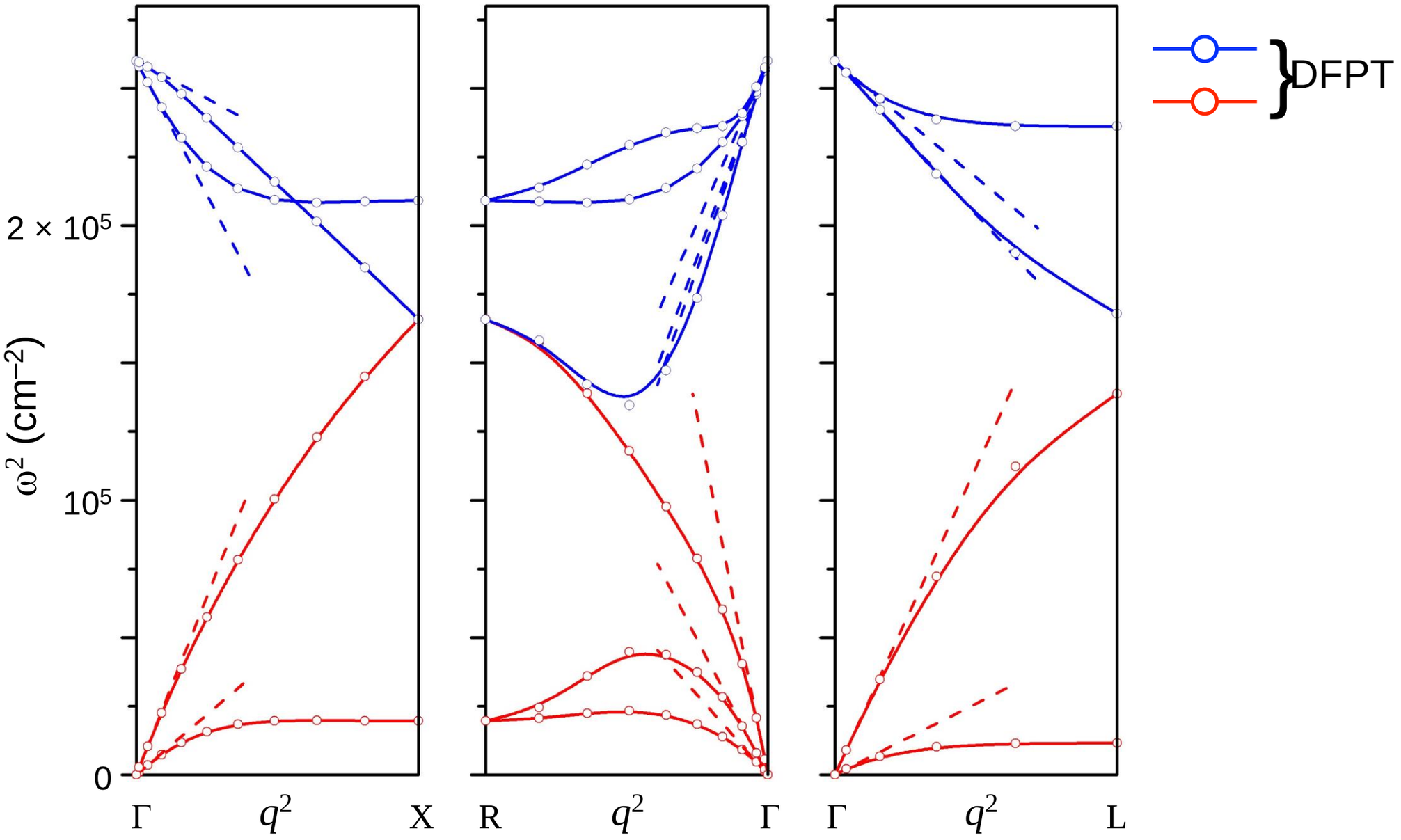
Calculation Details:

Functional = LDA (local density approx.)
Wavefunction Cutoff = 100 Ry
K-Point Sampling = 24 x 24 x 24

Experiment:

[a]Nilsson and Nelin, *Phys. Rev. B* 6, 3777 (1972)
[b]Dolling, *Proc. Symp. Inelastic Scattering Neutrons in Solids and Liquids 2,* 37 (1963)

Transformation of the phonon spectrum of silicon



By carrying out the transformation $q \rightarrow q^2$ and $\omega \rightarrow \omega^2$, all of the phonon branches become linear close to the Γ point.

Linear regression of the first-principles phonon spectrum

Path	Branch	Linear Regression	Perturbation Expression
$\Gamma \rightarrow X$ $[q\ 0\ 0]$ $q \in [0,1]$	TA_1, TA_2	$\omega^2 = 9.08\times10^4\, q^2$	$\omega^2 = C_{44}/(a^2c^2\rho)\, q^2$
	LA	$\omega^2 = 2.64\times10^5\, q^2$	$\omega^2 = C_{11}/(a^2c^2\rho)\, q^2$
	TO_1, TO_2	$\omega^2 = 2.60\times10^5 - 1.98\times10^5\, q^2$	$\omega^2 = \omega_0^2 + D_{44}/(a^2c^2\rho)\, q^2$
	LO	$\omega^2 = 2.60\times10^5 - 4.52\times10^4\, q^2$	$\omega^2 = \omega_0^2 + D_{11}/(a^2c^2\rho)\, q^2$
$\Gamma \rightarrow K$ $[q\ q\ 0]$ $q \in [0, 3/4]$	TA_1	$\omega^2 = 1.27\times10^5\, q^2$	$\omega^2 = (C_{11} - C_{12})/(a^2c^2\rho)\, q^2$
	TA_2	$\omega^2 = 1.88\times10^5\, q^2$	$\omega^2 = 2C_{44}/(a^2c^2\rho)\, q^2$
	LA	$\omega^2 = 5.68\times10^5\, q^2$	$\omega^2 = (C_{11} + C_{12} + 2C_{44})/(a^2c^2\rho)\, q^2$
	TO_1	$\omega^2 = 2.60\times10^5 - 2.52\times10^5\, q^2$	$\omega^2 = \omega_0^2 + (D_{11} - D_{12})/(a^2c^2\rho)\, q^2$
	TO_2	$\omega^2 = 2.60\times10^5 - 3.70\times10^5\, q^2$	$\omega^2 = \omega_0^2 + 2D_{44}/(a^2c^2\rho)\, q^2$
	LO	$\omega^2 = 2.60\times10^5 - 2.40\times10^5\, q^2$	$\omega^2 = \omega_0^2 + (D_{11} + D_{12} + 2D_{44})/(a^2c^2\rho)\, q^2$
$\Gamma \rightarrow L$ $[q\ q\ q]$ $q \in [0, 1/2]$	TA_1, TA_2	$\omega^2 = 2.08\times10^5\, q^2$	$\omega^2 = (C_{11} - C_{12} + C_{44})/(a^2c^2\rho)\, q^2$
	LA	$\omega^2 = 8.96\times10^5\, q^2$	$\omega^2 = (C_{11} + 2C_{12} + 4C_{44})/(a^2c^2\rho)\, q^2$
	TO_1, TO_2	$\omega^2 = 2.60\times10^5 - 4.24\times10^5\, q^2$	$\omega^2 = \omega_0^2 + (D_{11} - D_{12} + D_{44})/(a^2c^2\rho)\, q^2$
	LO	$\omega^2 = 2.60\times10^5 - 4.17\times10^5\, q^2$	$\omega^2 = \omega_0^2 + (D_{11} + 2D_{12} + 4D_{44})/(a^2c^2\rho)\, q^2$

q in unit of $2\pi/a$, ω in cm^{-1}, and all other quantities are in S.I. units. $a = 5.40\times10^{-10}$ m; $\rho = 2370$ kg/m^3; $c = 3\times10^8$ m/s.

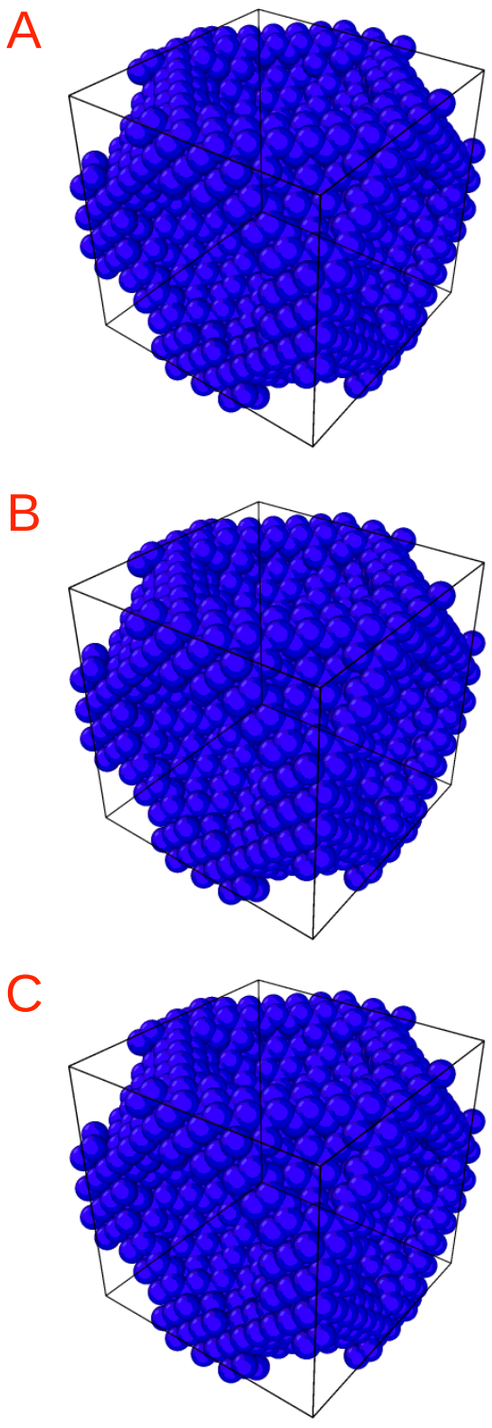
Elastic constants of the acoustic and optical modes of silicon from density-functional perturbation theory

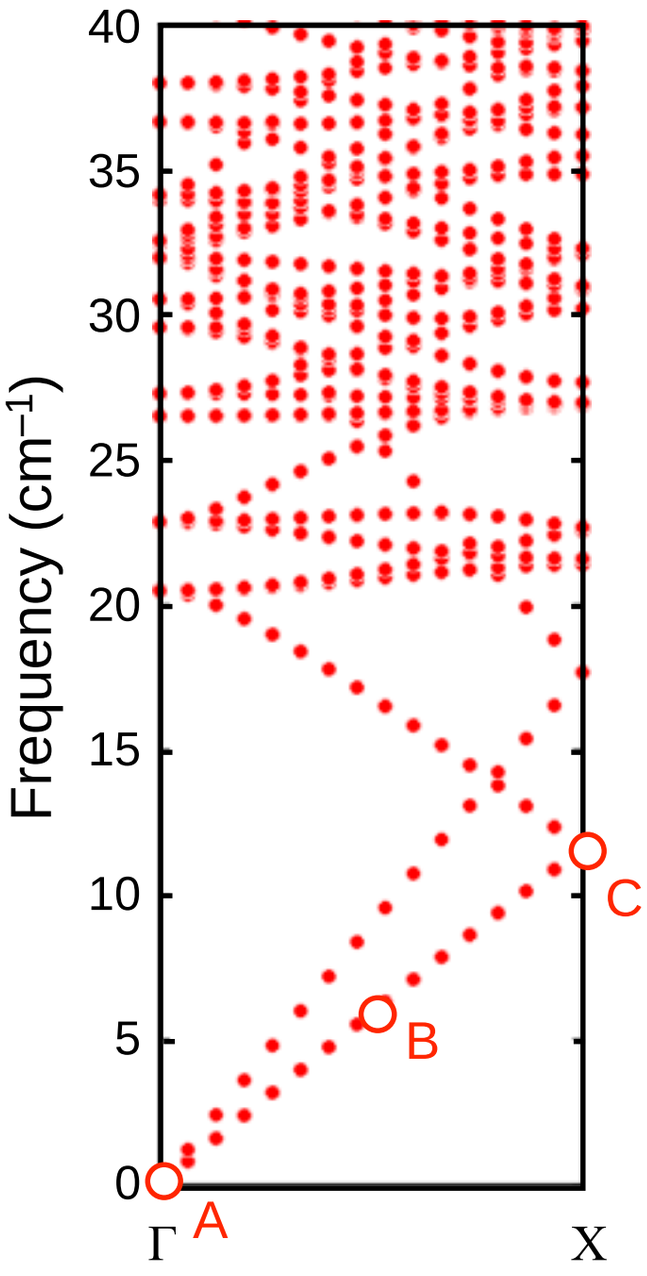
We exploit the linear regression and perturbation results to obtain:

	LDA	PBE	PBEsol	SW	SW[a]	Expt.
C_{11} (GPa)	160.2	156.9	161.5	152	151.4	168
C_{12} (GPa)	83.9	78.6	85.1	77.2	76.4	65
C_{44} (GPa)	56.9	56.5	54.9	56.7	56.4	80
D_{11} (GPa)	–28.1	–27.3	–28.4	–118.7	—	—
D_{12} (GPa)	121.2	127.7	127.6	8.1	—	—
D_{44} (GPa)	–122.8	–125.2	–120	–106.5	—	—

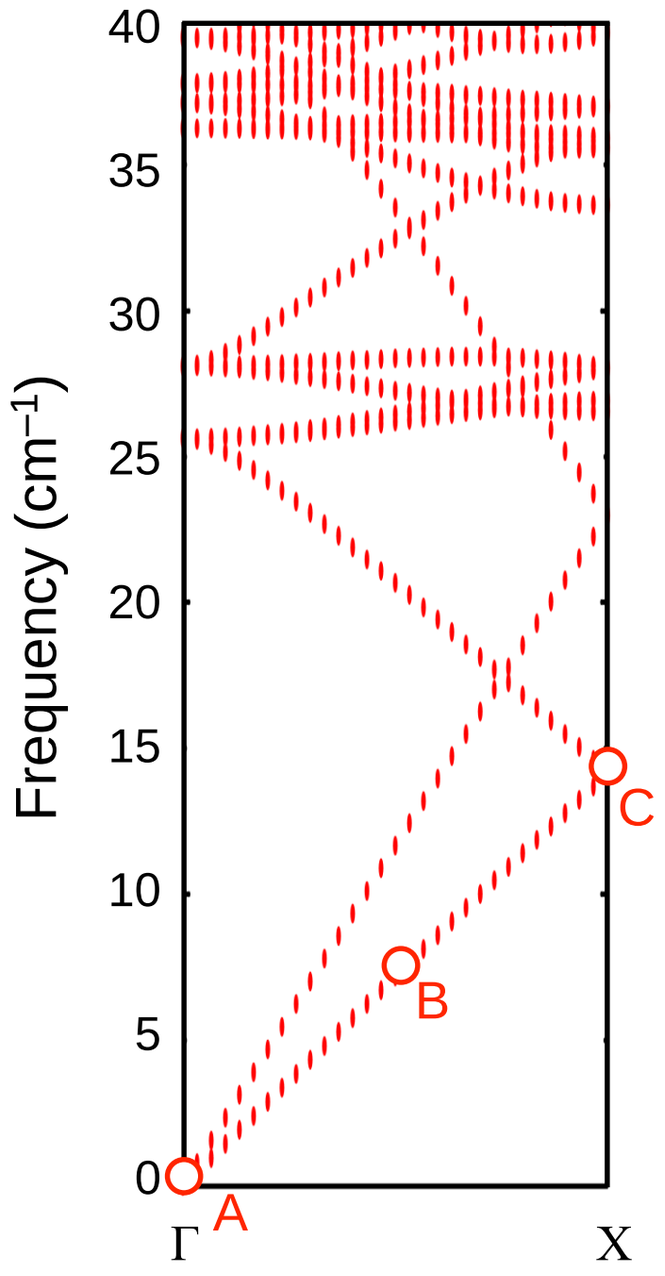
[a]Cowley, *Phys. Rev. Lett.* 60, 2379 (1988)

Acoustic vibrational modes

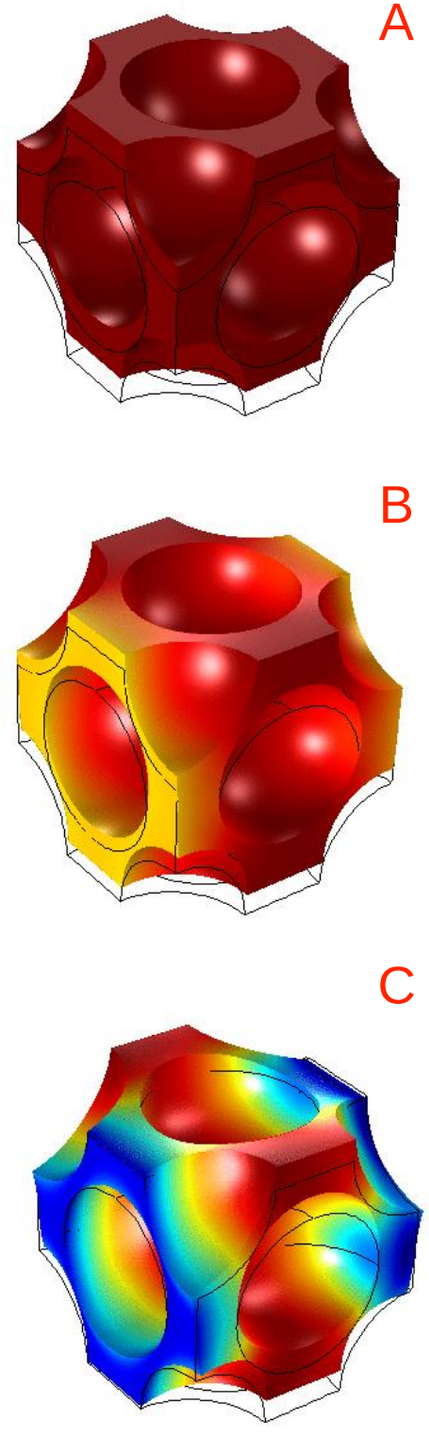




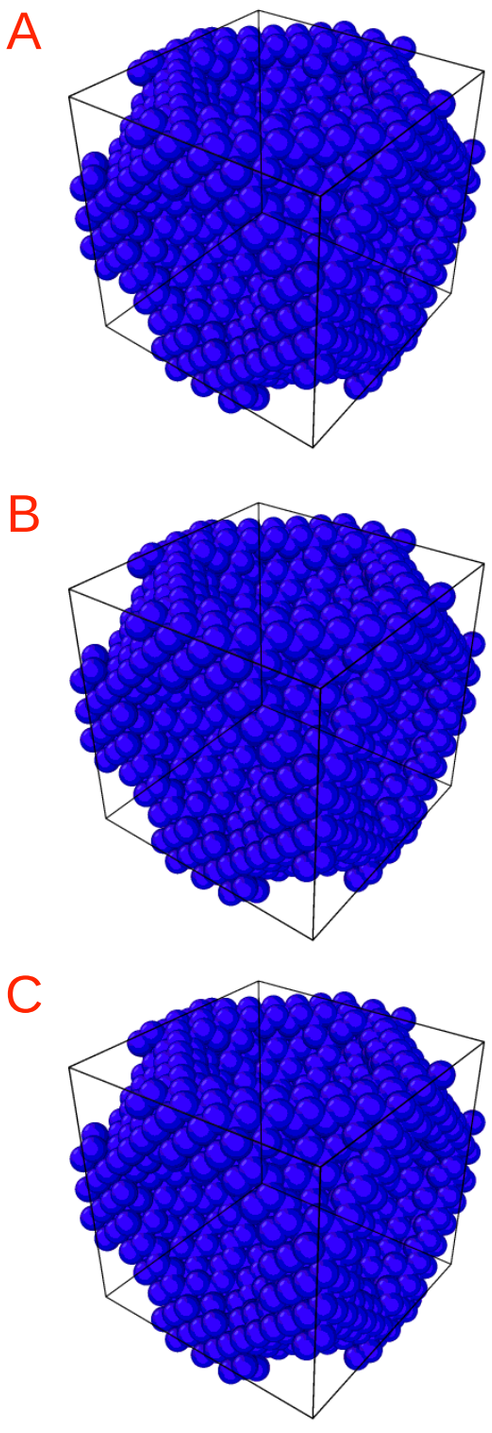
Method = DFT-based SW
Pore size = ~1.5 nm
Number of atoms = 2,140
Pore volume fraction = ~52%

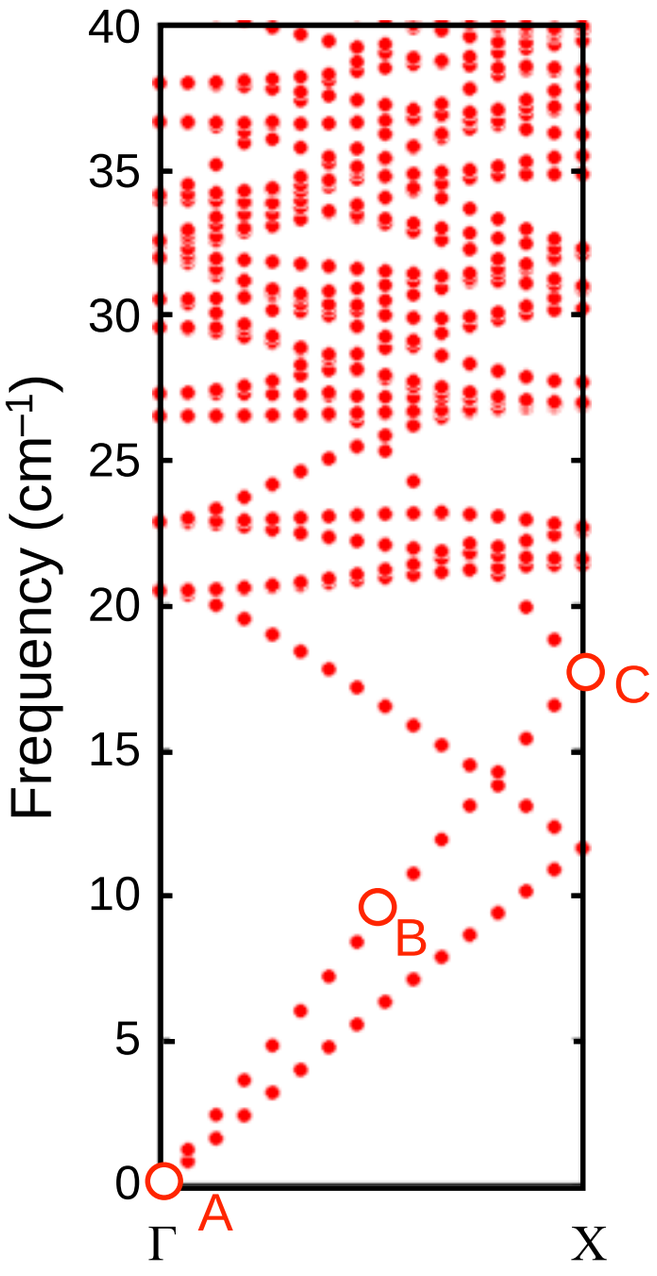


Method = DFT-based FEM
Pore size = 1.37 nm
Number of nodes = 63,918
Pore volume fraction = 52%



Acoustic vibrational modes (cont'd)





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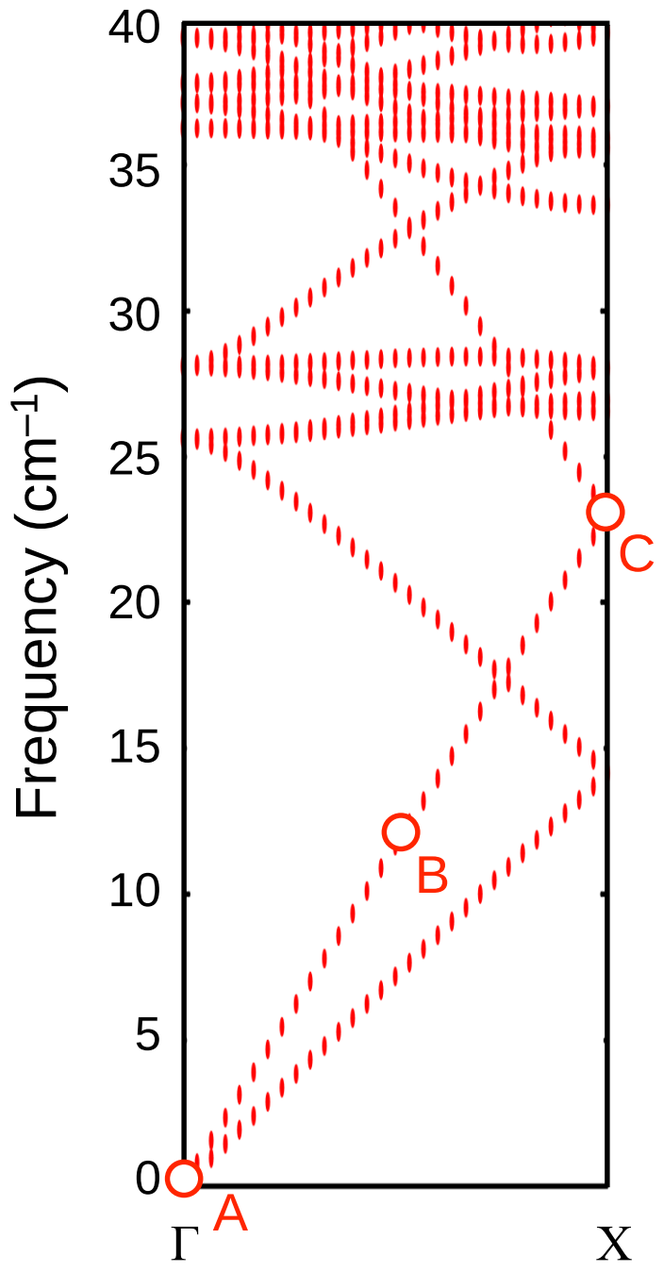

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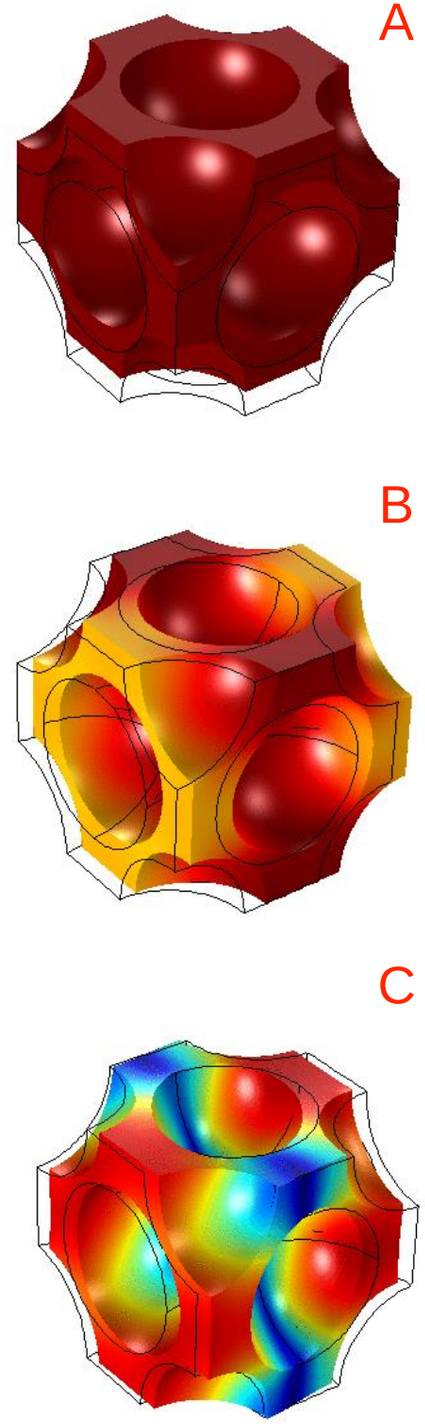
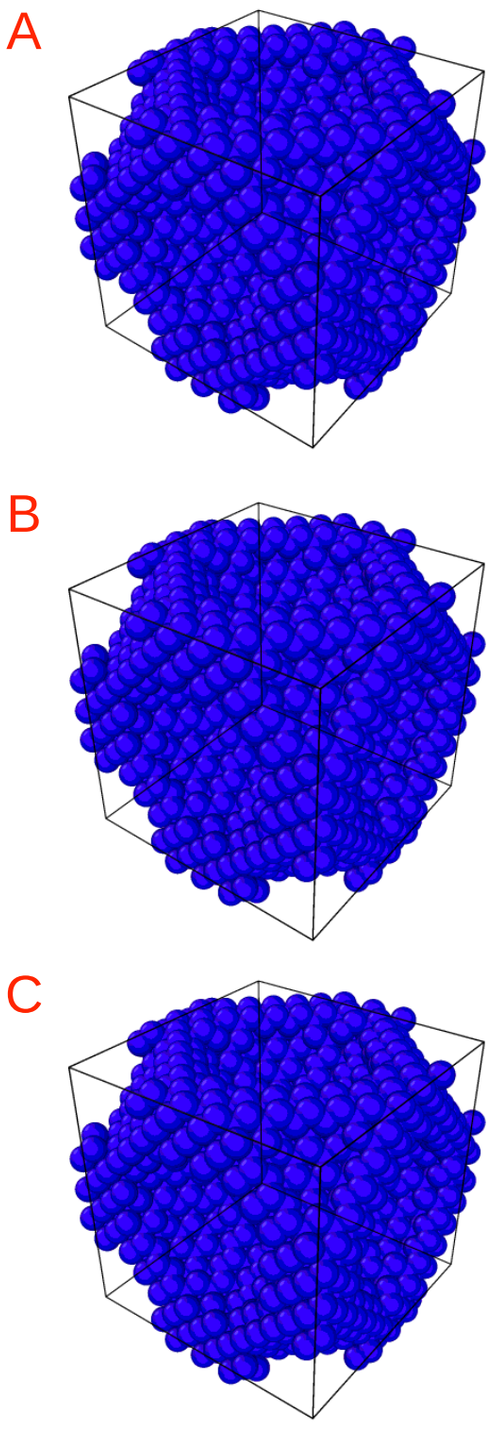
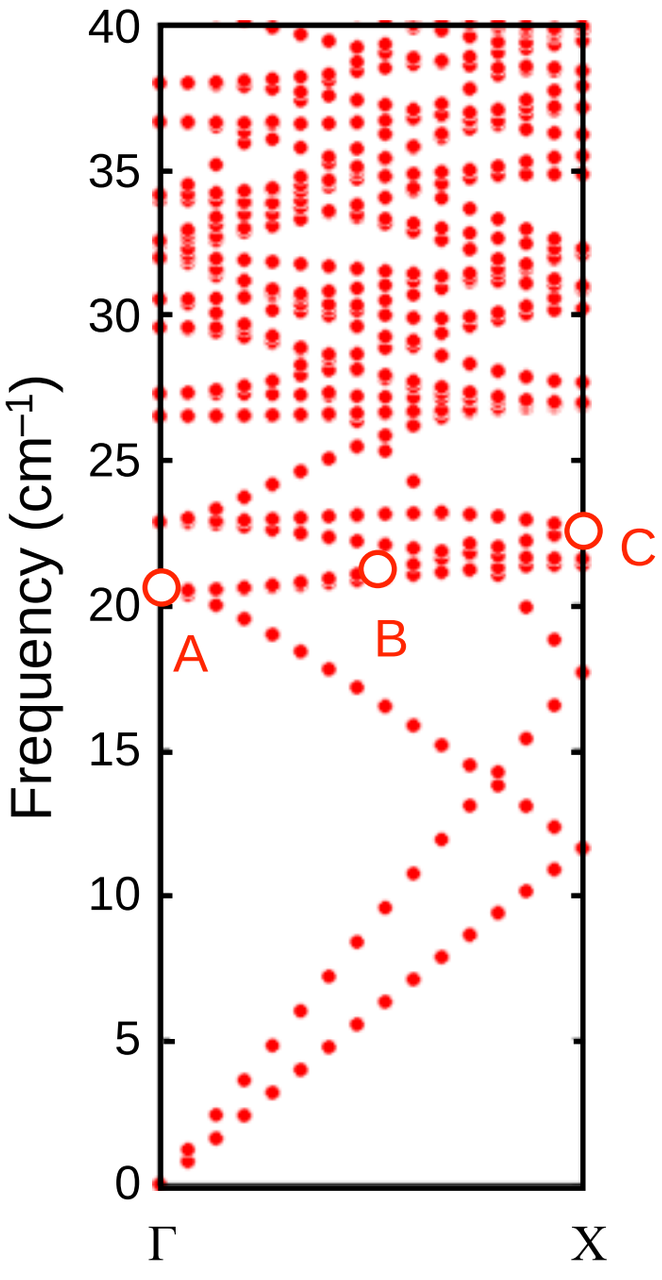
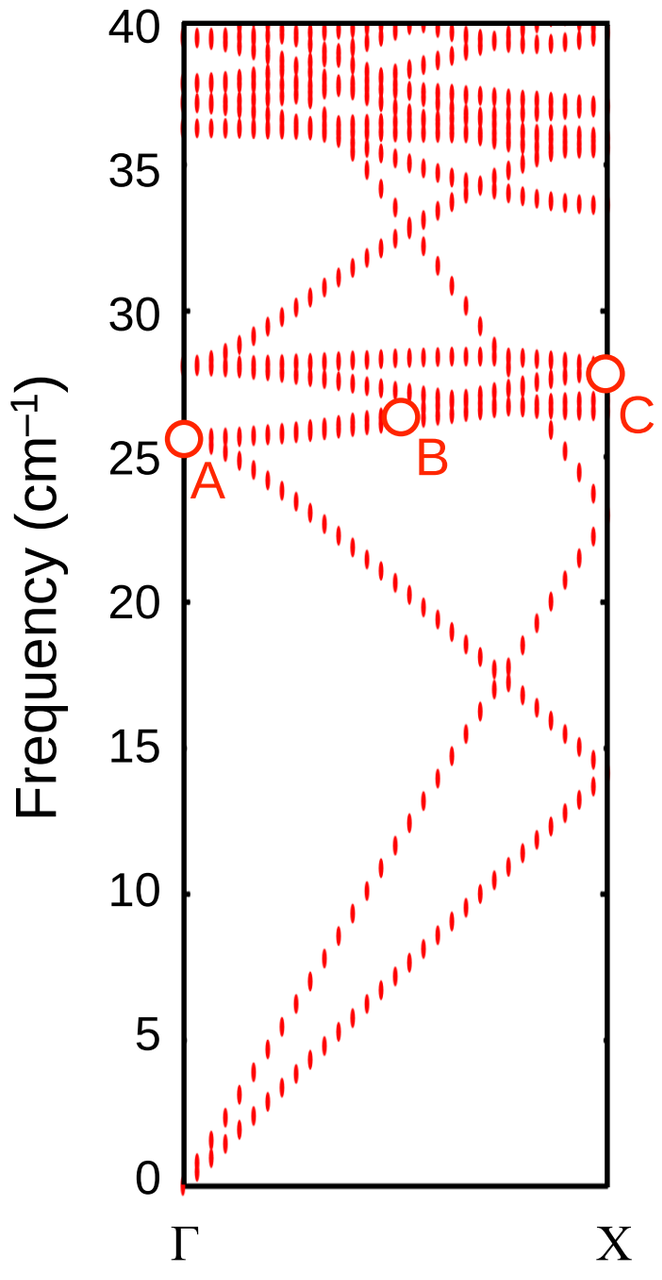
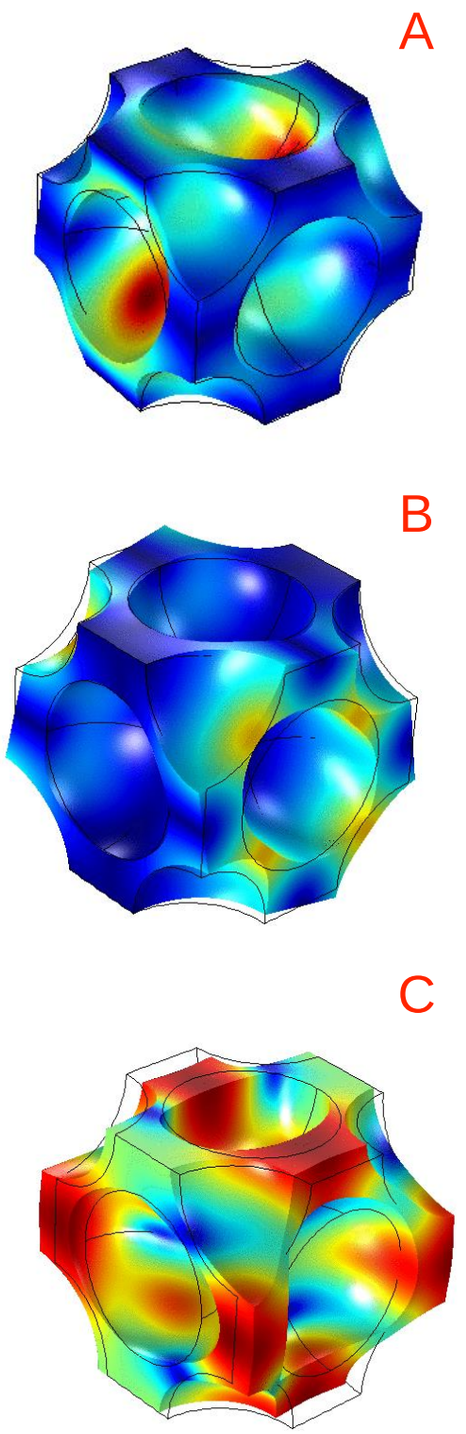


Acoustic vibrational modes (cont'd)









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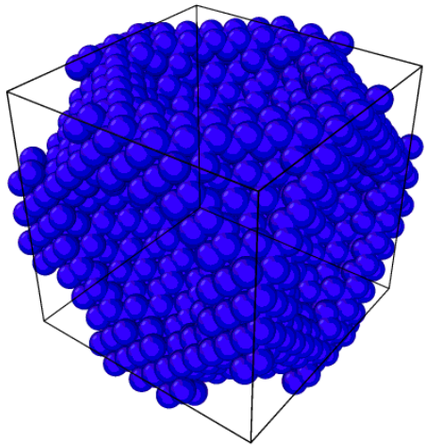
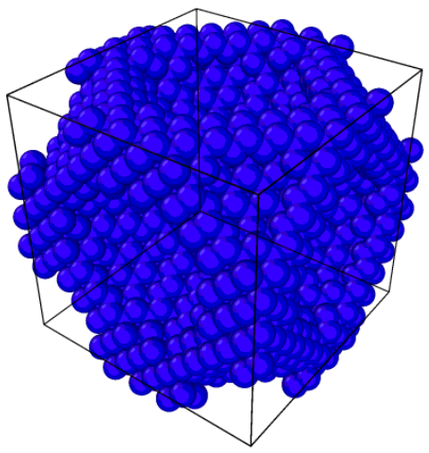
Optical vibrational modes

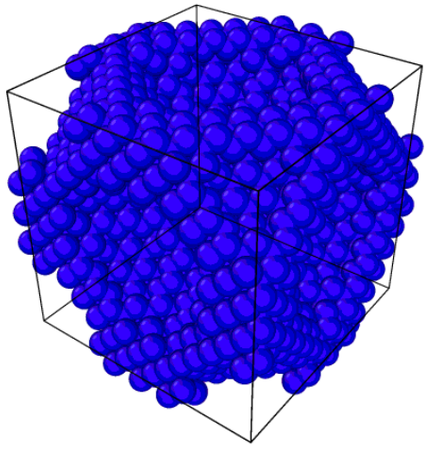
A

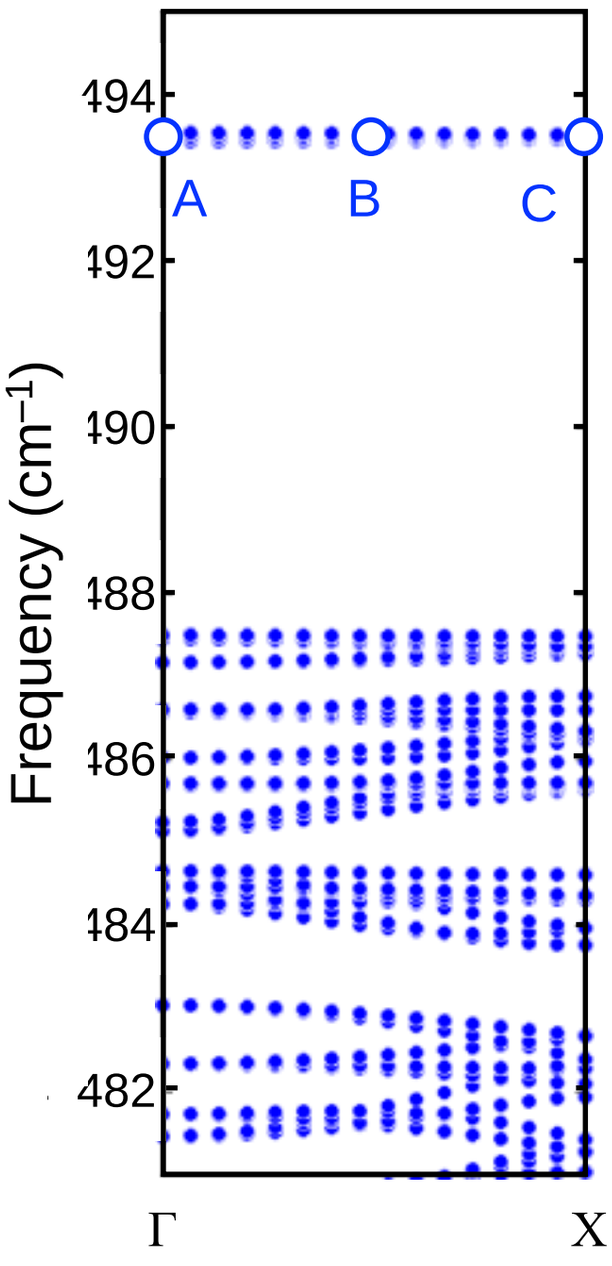


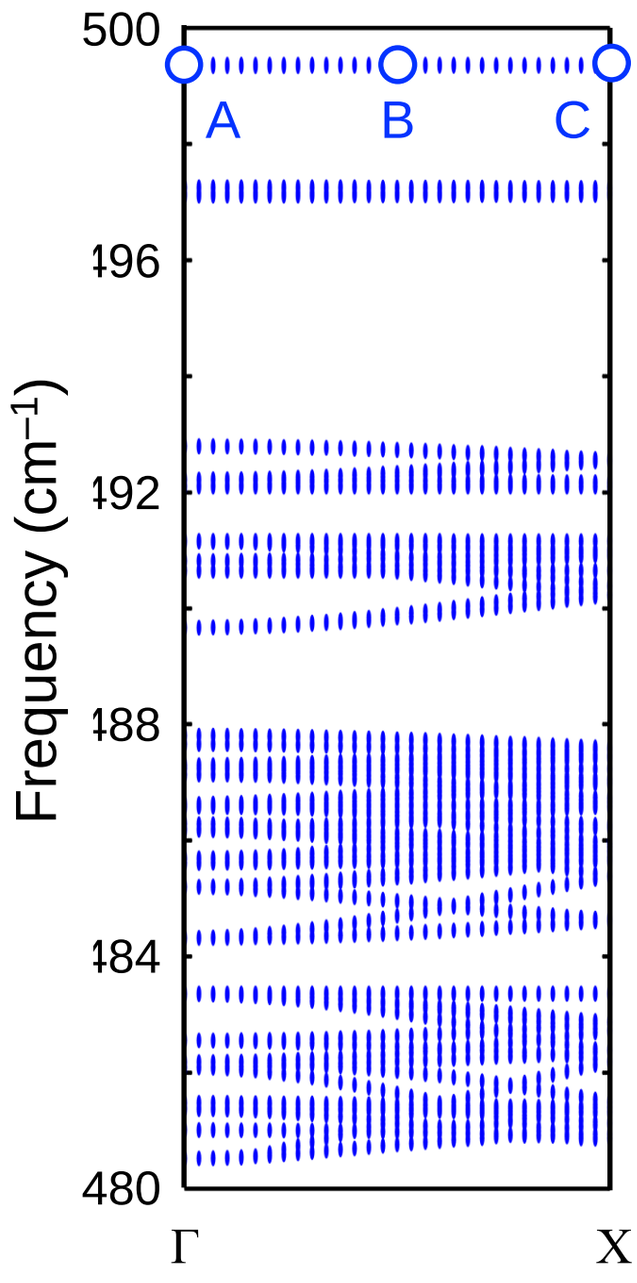
B



C







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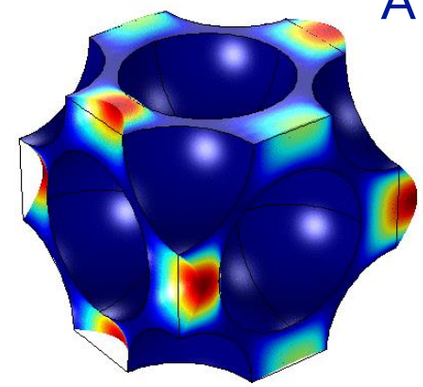

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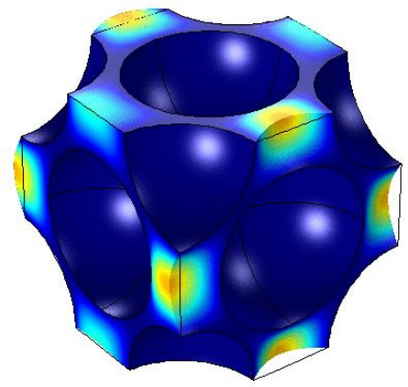

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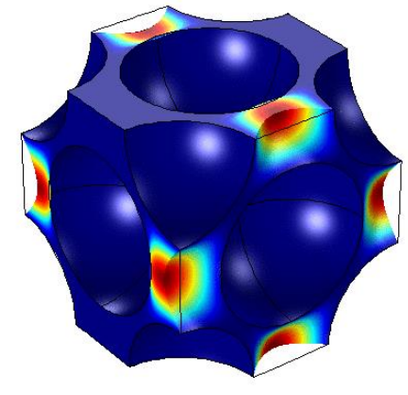

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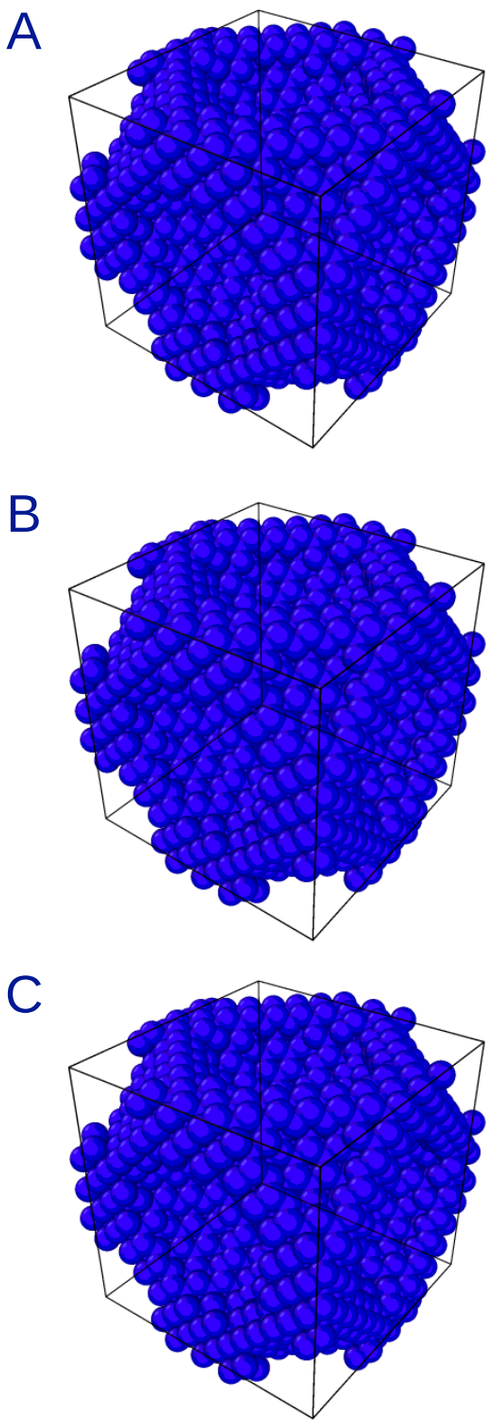
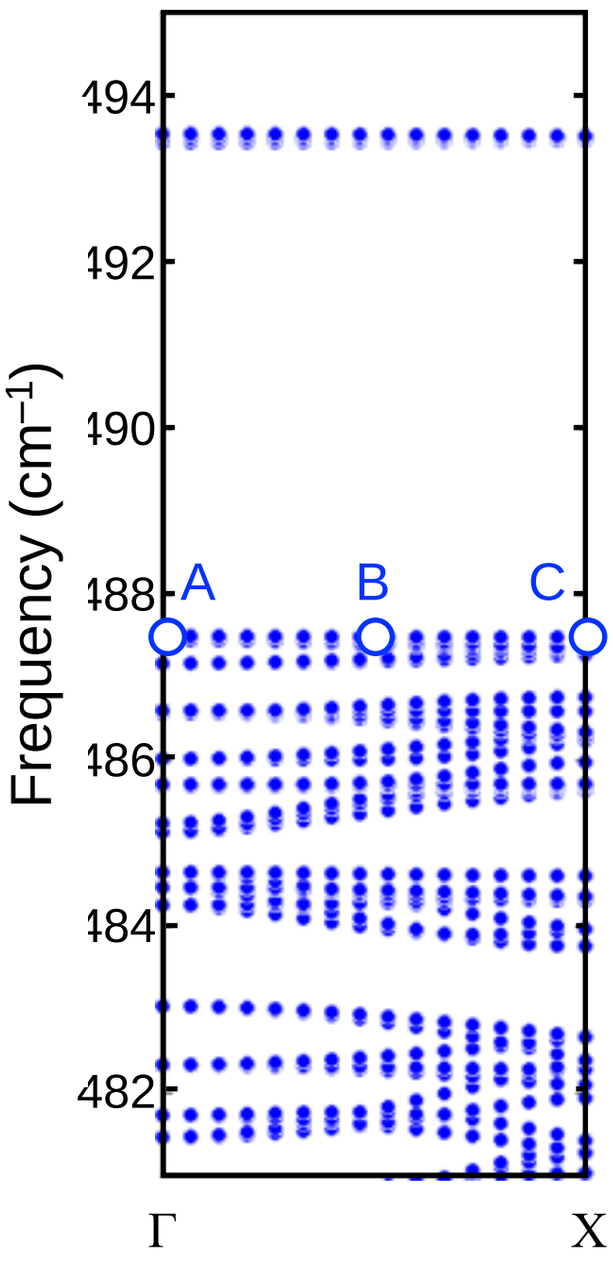
Optical vibrational modes (cont'd)





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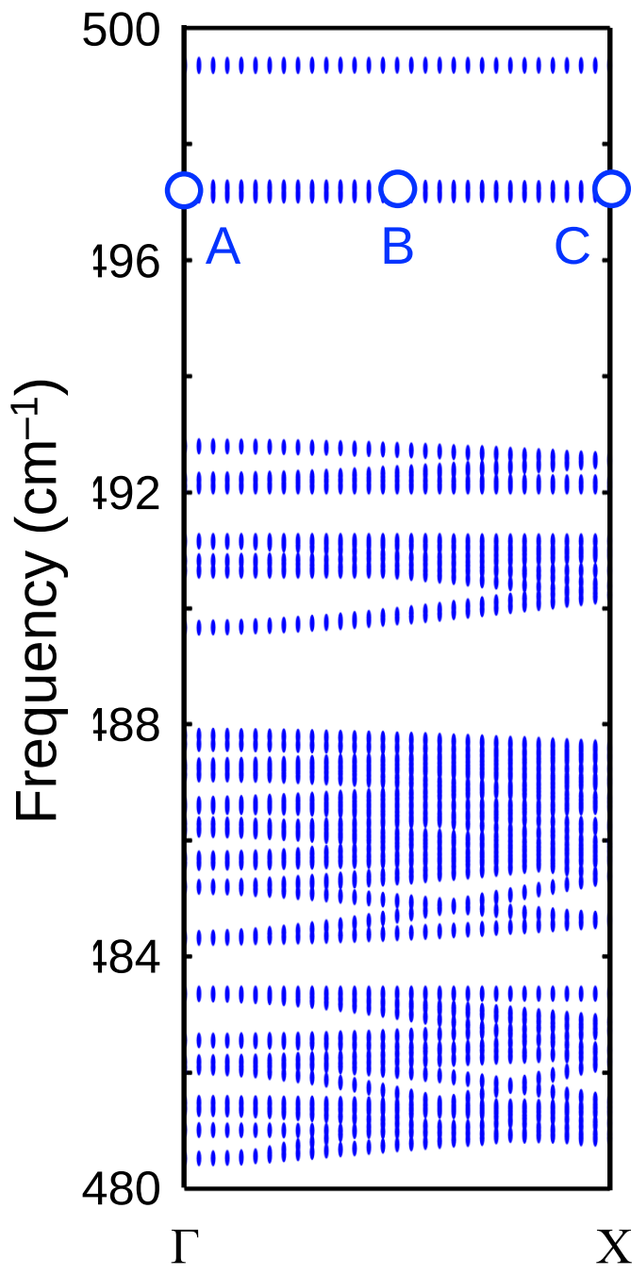

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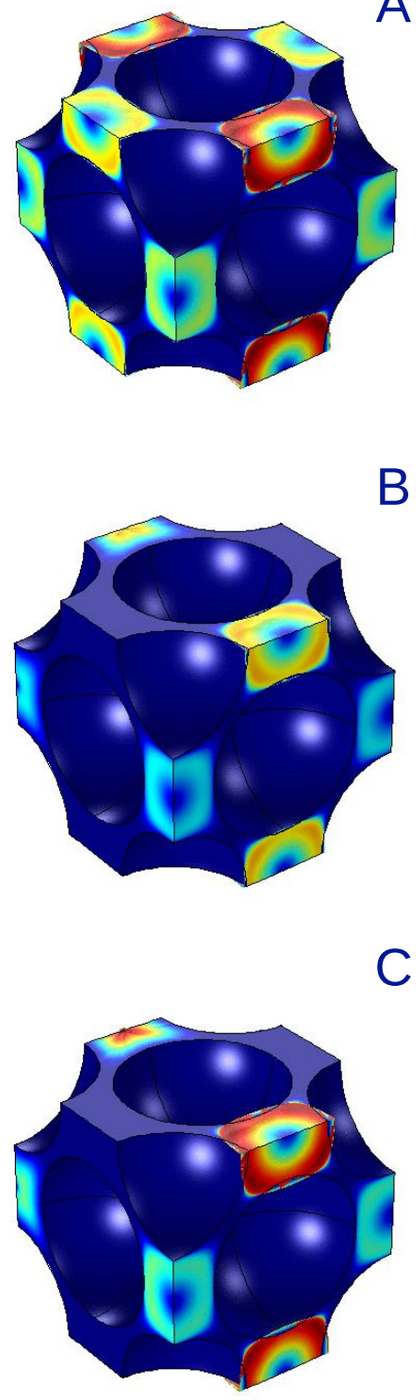
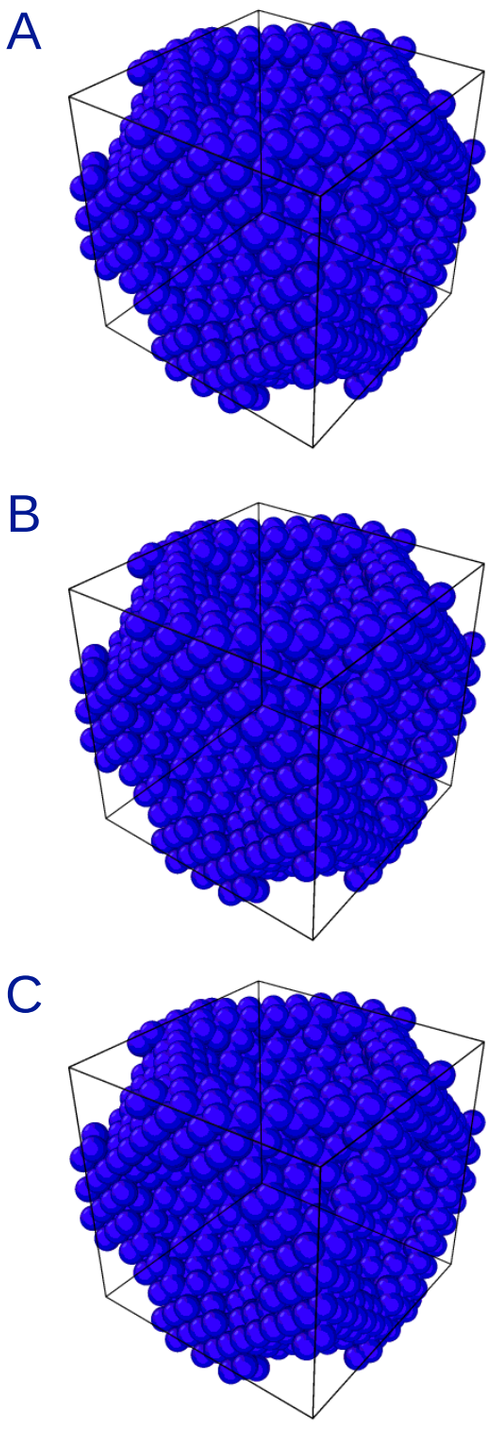
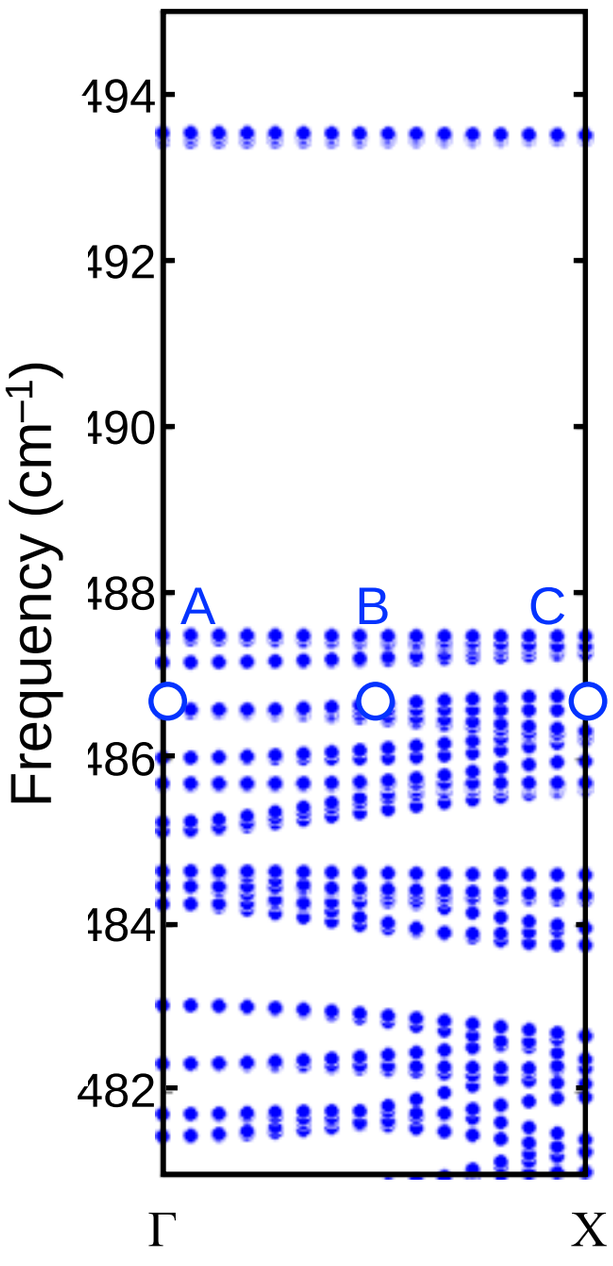
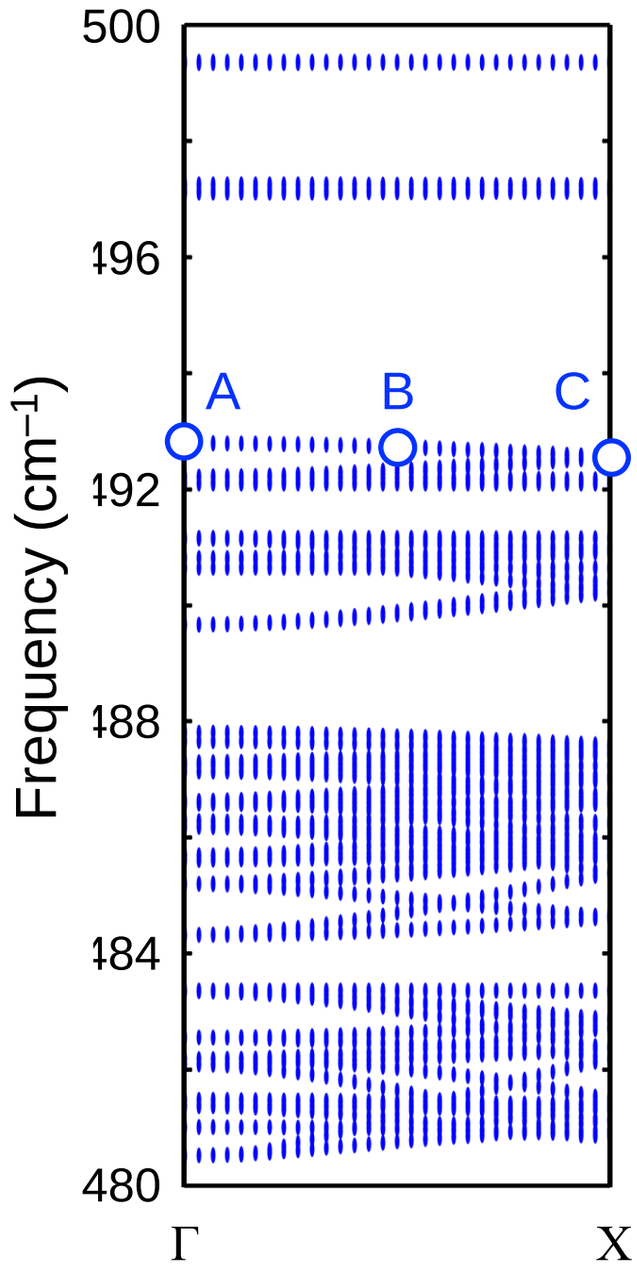
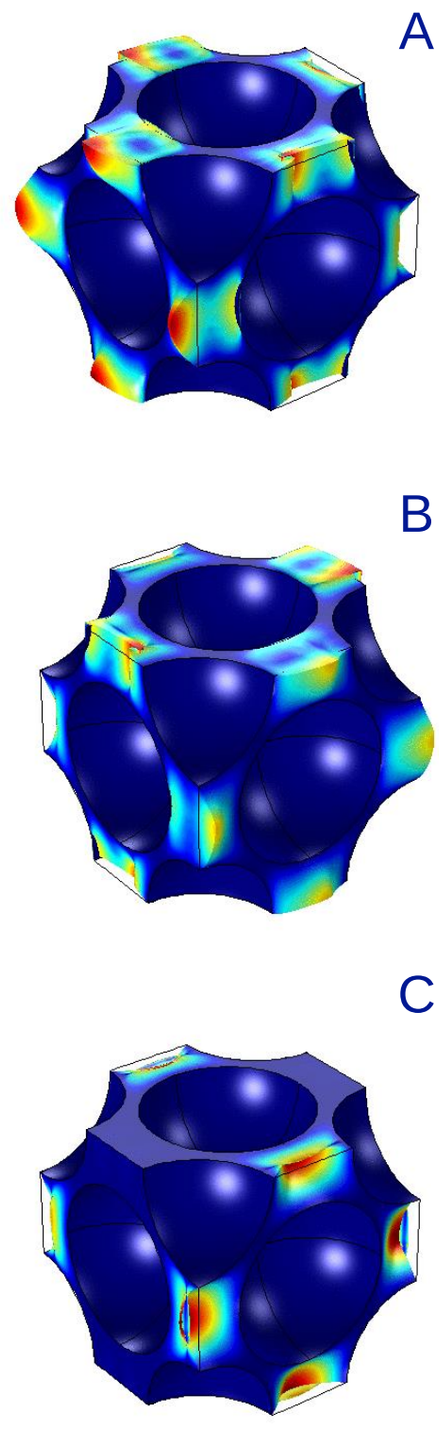


Optical vibrational modes (cont'd)









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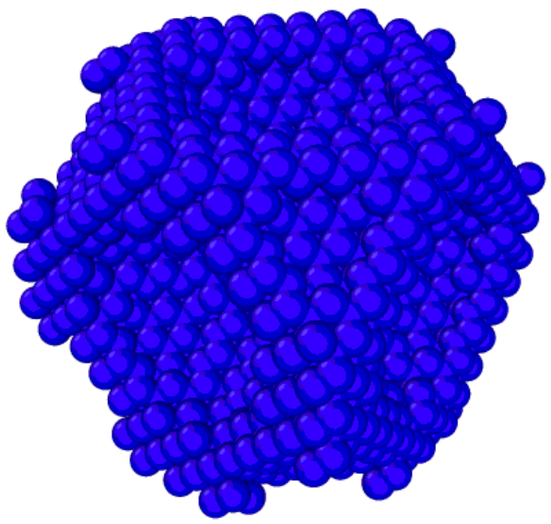
Summary

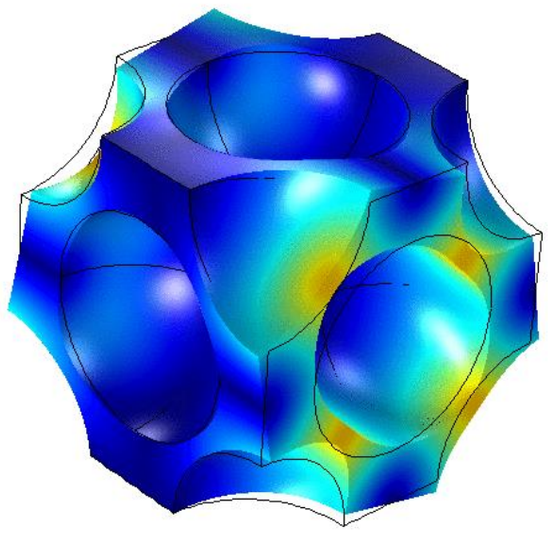
We have calculated the vibrational modes of silicon metalattices (silicon crystal with an array of nanopores) using COMSOL.

We have determined the acoustic and optical phonon dispersion spectrum of silicon metalattices in agreement with atomistic models.

Those results open up the possibilities to predict the influence of nanoscale geometry on the optical response of silicon metalattices.



atomistic



continuum

Funding: NSF-DMR 1420620