

Excitations of a quantum solid

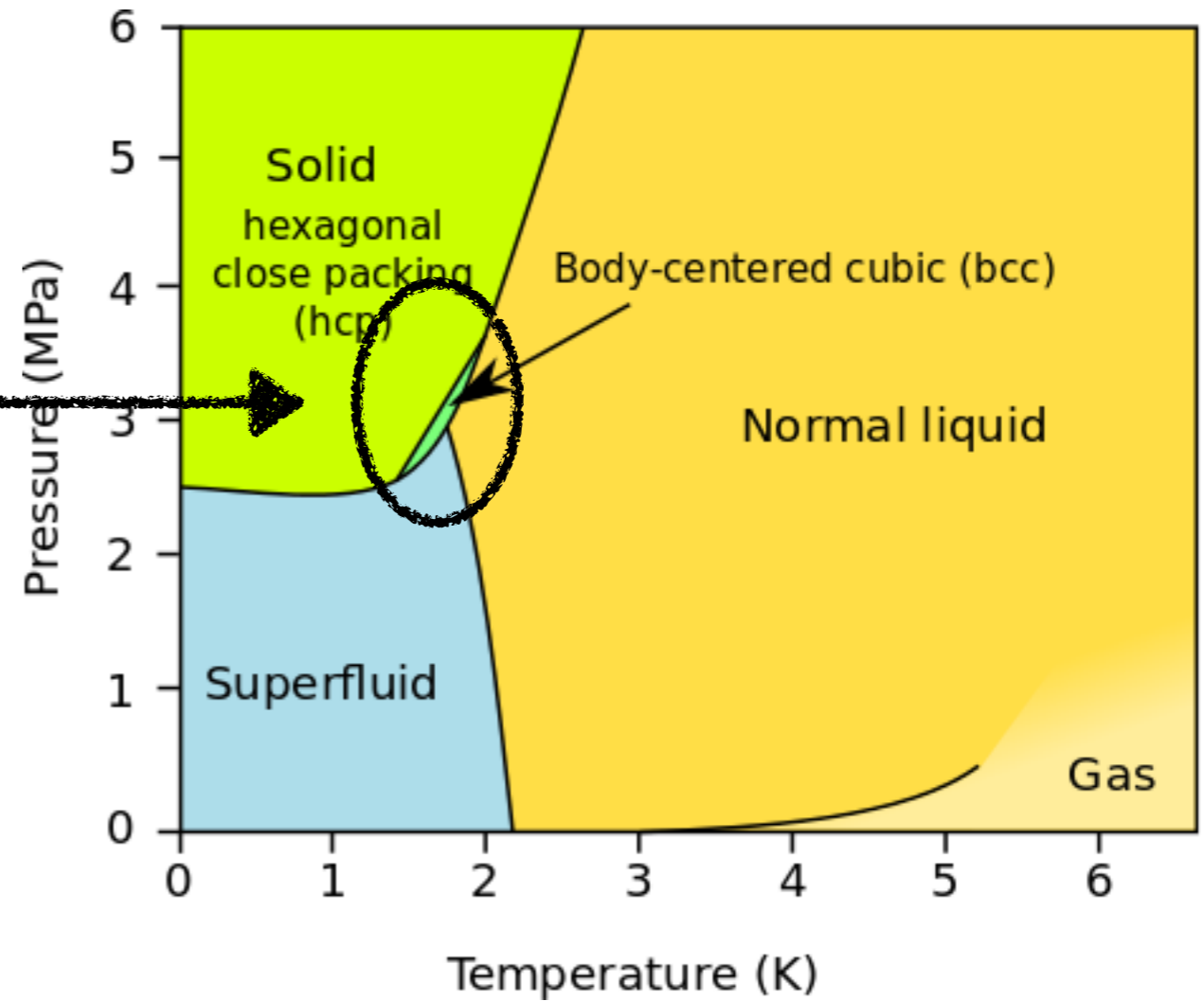
Daniel Podolsky

Periodic Table of the Elements

Atomic Number	Symbol	Name	Atomic Mass		
1	H	Hydrogen	1.008		
2	He	Helium	4.003		
3	Li	Lithium	6.941		
4	Be	Beryllium	9.012		
11	Na	Sodium	22.990		
12	Mg	Magnesium	24.305		
19	K	Potassium	39.098		
20	Ca	Calcium	40.078		
21	Sc	Scandium	44.956		
22	Ti	Titanium	47.88		
23	V	Vanadium	50.942		
24	Cr	Chromium	51.995		
25	Mn	Manganese	54.938		
26	Fe	Iron	55.933		
27	Co	Cobalt	58.933		
28	Ni	Nickel	58.693		
29	Cu	Copper	63.545		
30	Zn	Zinc	65.39		
31	Ga	Gallium	69.732		
32	Ge	Germanium	72.61		
33	As	Arsenic	74.922		
34	Se	Selenium	78.09		
35	Br	Bromine	79.904		
36	Kr	Krypton	84.90		
37	Rb	Rubidium	84.458		
38	Sr	Strontium	87.62		
39	Y	Yttrium	88.906		
40	Zr	Zirconium	91.224		
41	Nb	Niobium	92.906		
42	Mo	Molybdenum	95.94		
43	Tc	Technetium	98.907		
44	Ru	Ruthenium	101.07		
45	Rh	Rhodium	102.906		
46	Pd	Palladium	106.42		
47	Ag	Silver	107.868		
48	Cd	Cadmium	112.411		
49	In	Indium	114.818		
50	Sn	Tin	118.71		
51	Sb	Antimony	121.760		
52	Te	Tellurium	127.6		
53	I	Iodine	126.904		
54	Xe	Xenon	131.29		
55	Cs	Cesium	132.905		
56	Ba	Barium	137.327		
57-71	Lanthanide Series				
72	Hf	Hafnium	178.49		
73	Ta	Tantalum	180.948		
74	W	Tungsten	183.85		
75	Re	Rhenium	186.207		
76	Os	Osmium	190.23		
77	Ir	Iridium	192.22		
78	Pt	Platinum	195.08		
79	Au	Gold	196.967		
80	Hg	Mercury	200.59		
81	Tl	Thallium	204.383		
82	Pb	Lead	207.2		
83	Bi	Bismuth	208.980		
84	Po	Polonium	[209]		
85	At	Astatine	209		
86	Rn	Radon	222.018		
87	Fr	Francium	223.029		
88	Ra	Radium	226.025		
89-103	Actinide Series				
104	Rf	Rutherfordium	[261]		
105	Db	Dubnium	[262]		
106	Sg	Seaborgium	[266]		
107	Bh	Bohrium	[264]		
108	Hs	Hassium	[268]		
109	Mt	Moscovium	[268]		
110	Ds	Darmstadtium	[268]		
111	Rg	Roentgenium	[272]		
112	Cn	Copernicium	[277]		
113	Uut	Ununtrium	unknown		
114	Fl	Flerovium	[289]		
115	Uup	Ununpentium	unknown		
116	Lv	Livermorium	[293]		
117	Uus	Ununseptium	unknown		
118	Uuo	Ununoctium	unknown		

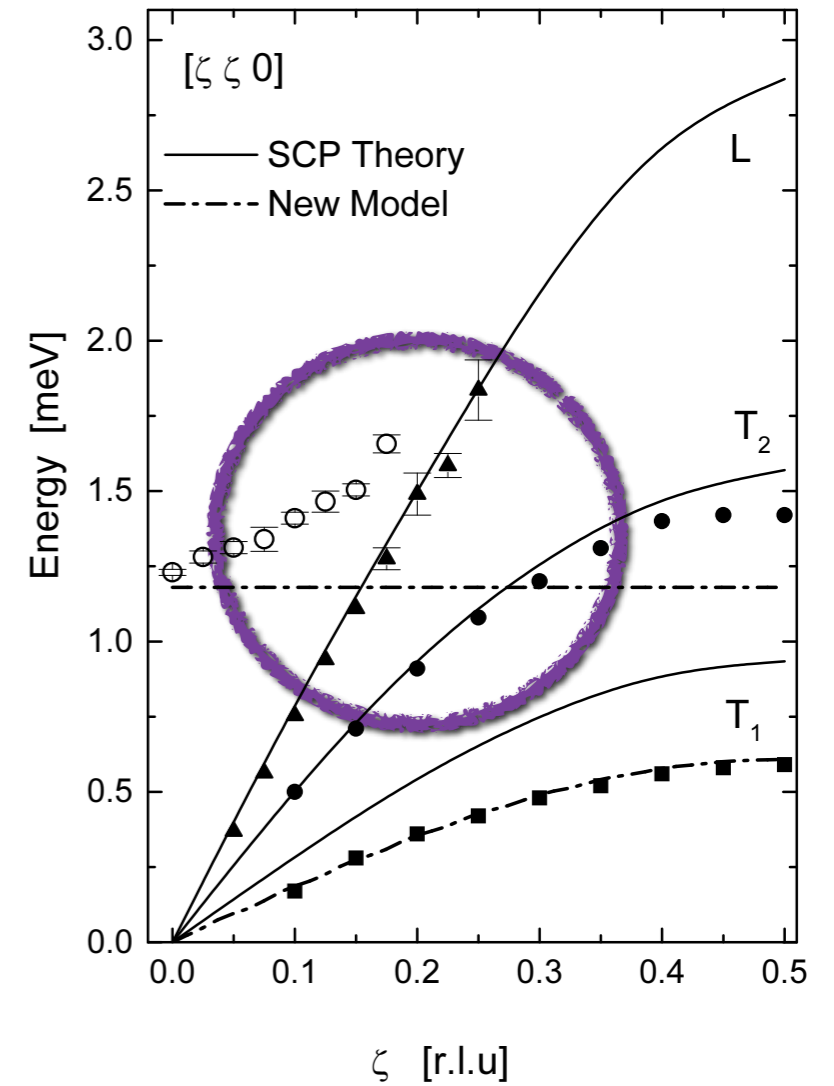
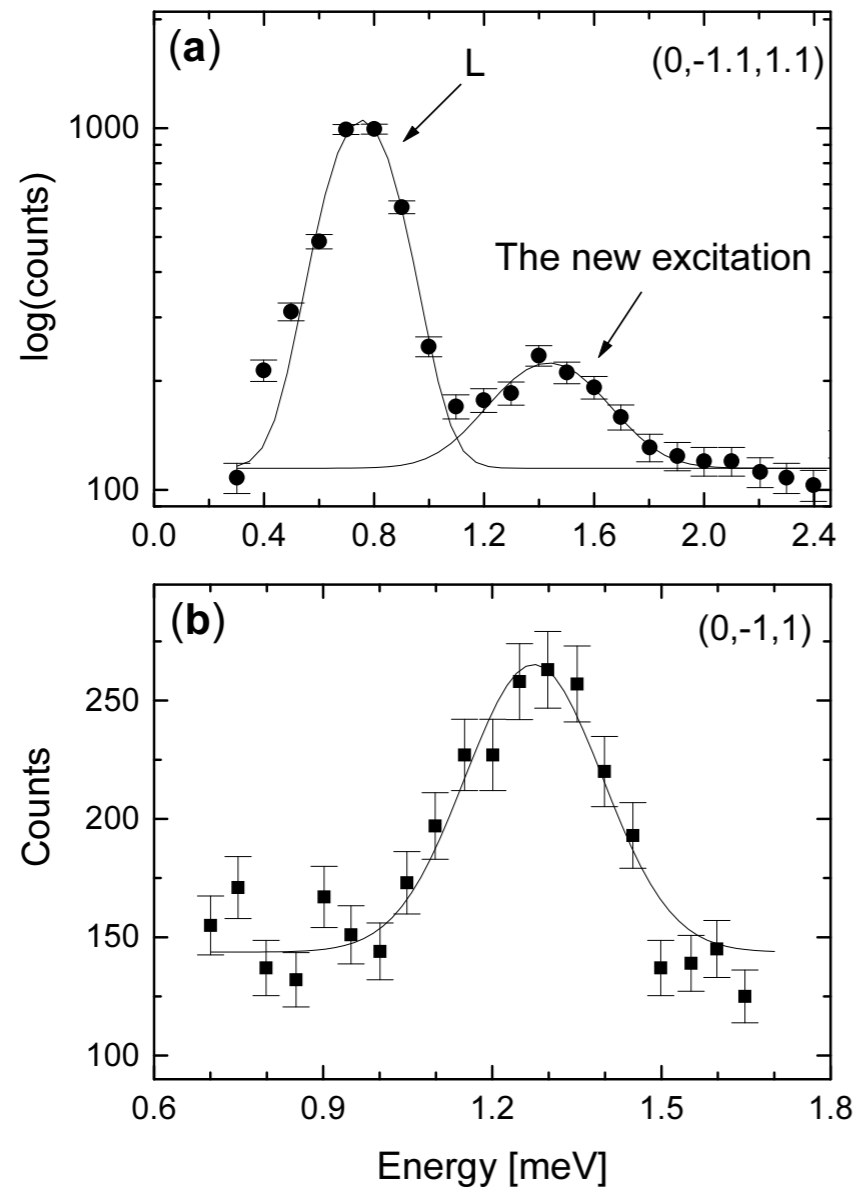
Collaborators: S. Gazit, H. Nonne, A. Auerbach, D. Arovav

Helium 4 - Phase diagram



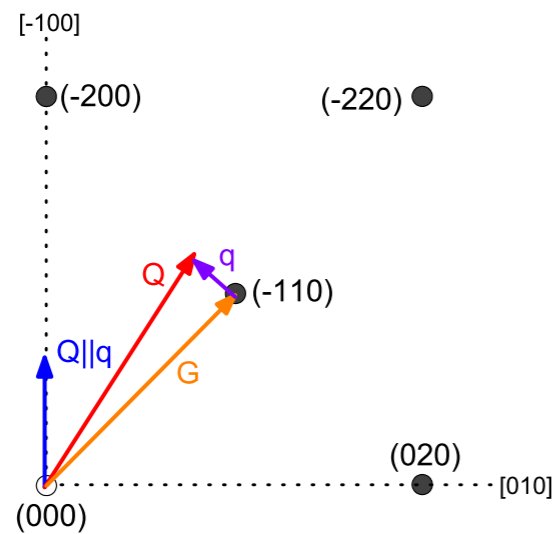
Inelastic neutron scattering

Optical mode observed!

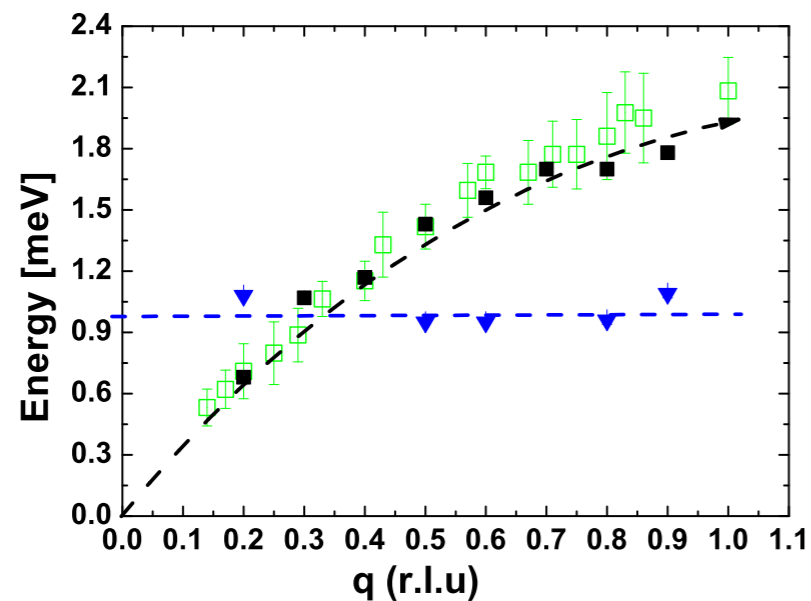


Multiple optical modes?

Look at different directions and polarizations

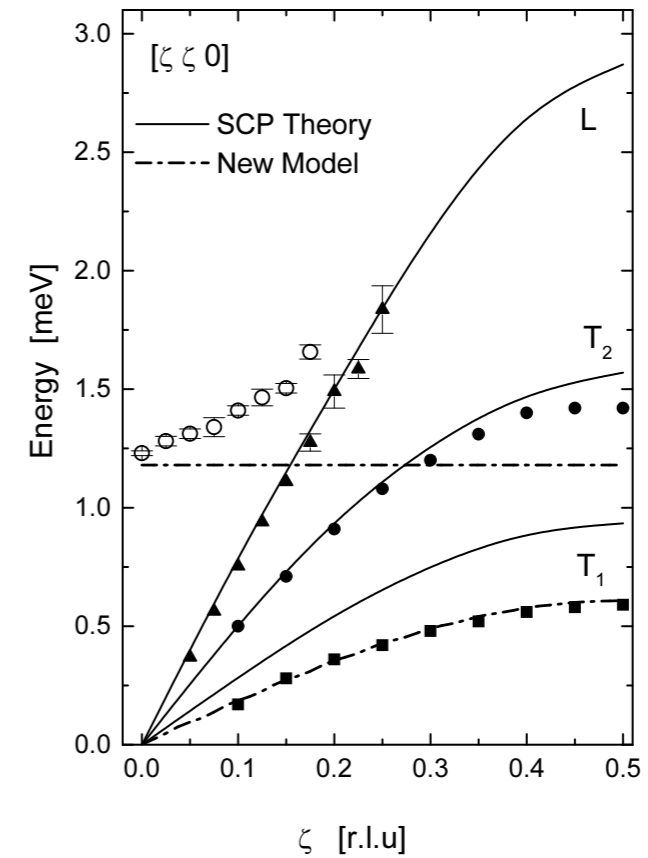


$\tau(100)$



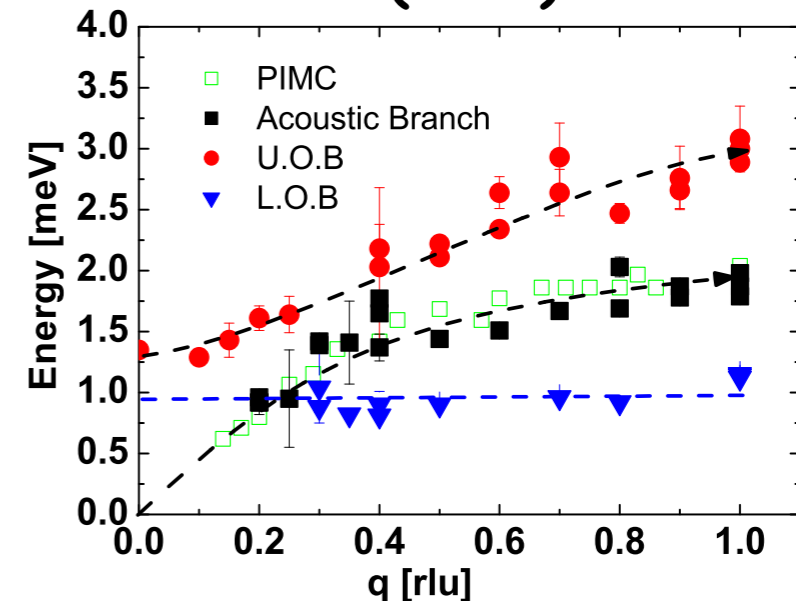
Pelleg et al, PRB **73**, 180301R ('06)

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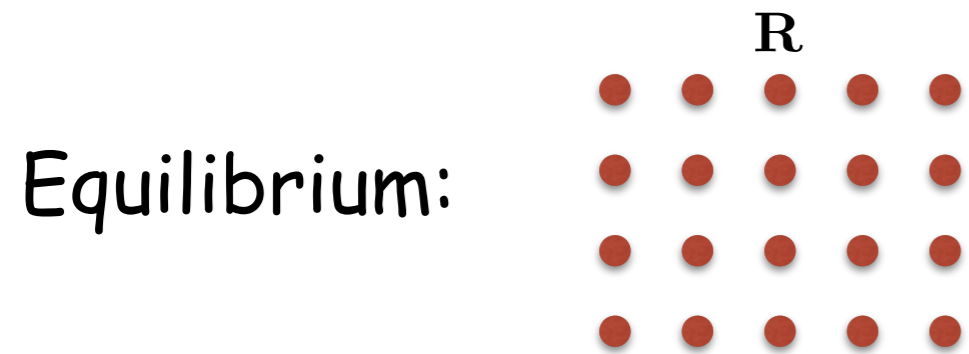
Markovic et al., PRL **88**, 195301 ('02)

L(100)

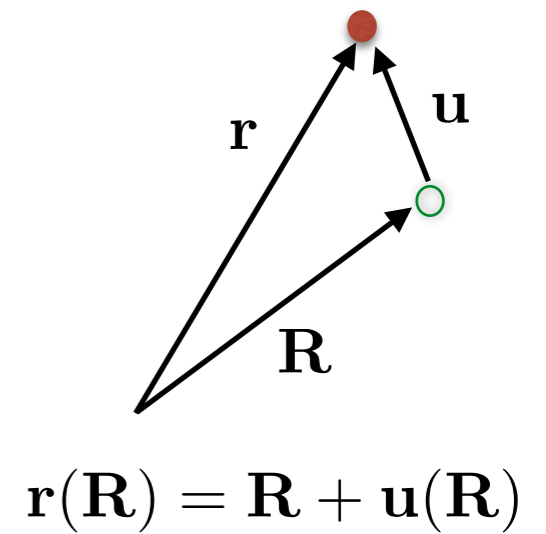


Pelleg et al., JLTP **151**, 1164 ('08)

Harmonic theory of solids



Fluctuations:



Small fluctuations $\sqrt{\langle \mathbf{u}^2 \rangle} \ll \Delta R$

$$U_{\text{harm}} = \frac{1}{2} \sum_{\mathbf{R}\mathbf{R}'} \sum_{\mu\nu} u_{\mu}(\mathbf{R}) D_{\mu\nu}(\mathbf{R} - \mathbf{R}') u_{\nu}(\mathbf{R}')$$

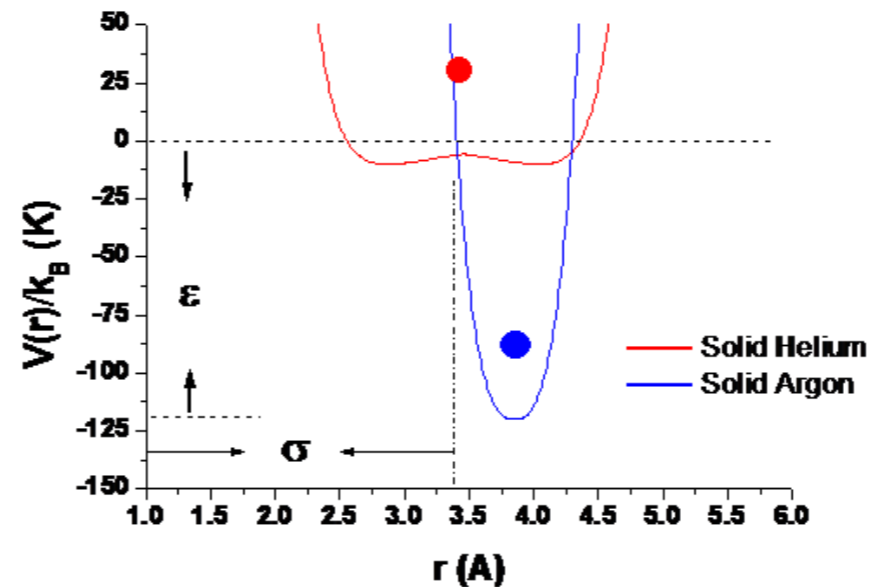
Monatomic Bravais lattice \Leftrightarrow acoustic phonons only

Corrections to harmonic theory: $U_{\text{anh}} \sim u^3 + u^4 + \dots$

Lindemann criterion: $\sqrt{\langle \mathbf{u}^2 \rangle} = 0.1 \Delta R \Leftrightarrow$ melting

Helium - A quantum solid

Atoms do not sit at minimum of V :



Large zero point motion:

H. Glyde, "Helium, Solid"

Rare-gas crystal	Debye temperature θ_D (K)	Melting temperature T_M (K)	Debye zero point energy $E_{ZD} = \frac{9}{8}\theta_D$	Lindemann parameter $\delta = \langle u^2 \rangle^{1/2} / R$
$^3\text{He}(\text{bcc})$	19	0.65	21	0.368
$^4\text{He}(\text{bcc})$	25	1.6	28	0.292
Ne	66	24.6	74	0.091
Ar	84	83.8	95	0.048
Kr	64	161.4	72	0.036
Xe	55	202.0	62	0.028

Harmonic theory does not give correct acoustic phonon velocities

Large quantum fluctuations

⇒ restoring force is non-linear



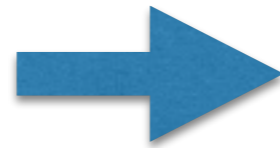
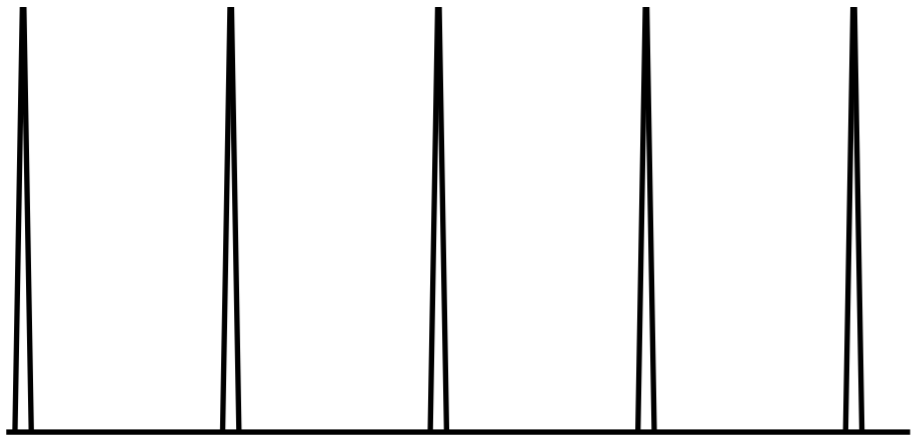
$$m\ddot{u}_i = -\kappa(u_i - u_{i+1}) - \kappa(u_i - u_{i-1}) + \gamma(u_i - u_{i+1})^2 + \gamma(u_i - u_{i-1})^2 + \dots$$

⇒ Non-linear equations can in principle give multiple solutions
(more phonons than number of degrees of freedom)

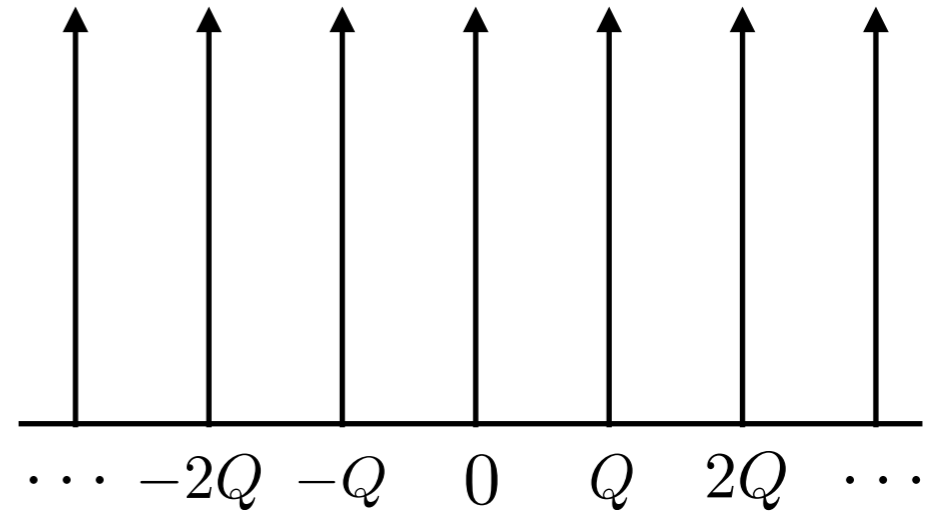
⇒ How to construct a linear theory for optical modes?

A different point of view

Idealized crystal:

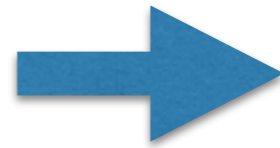
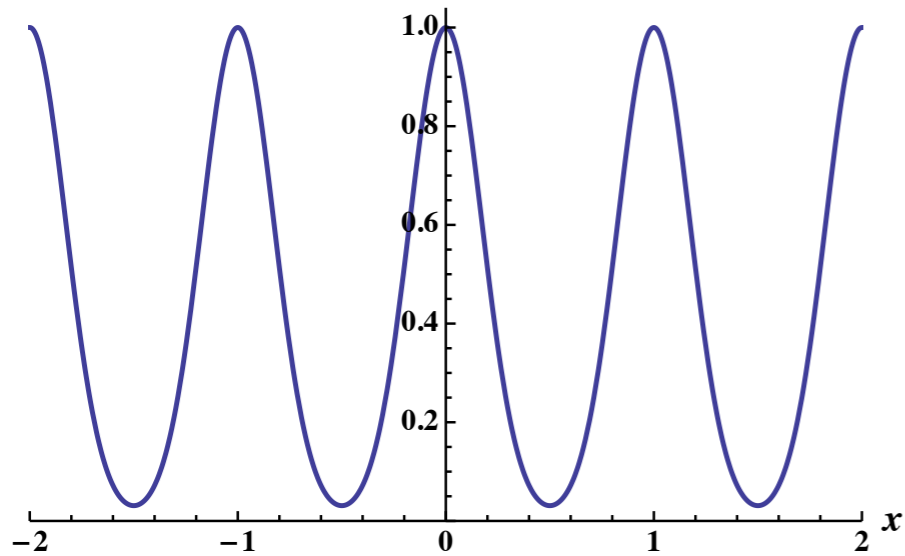


Bragg peaks:

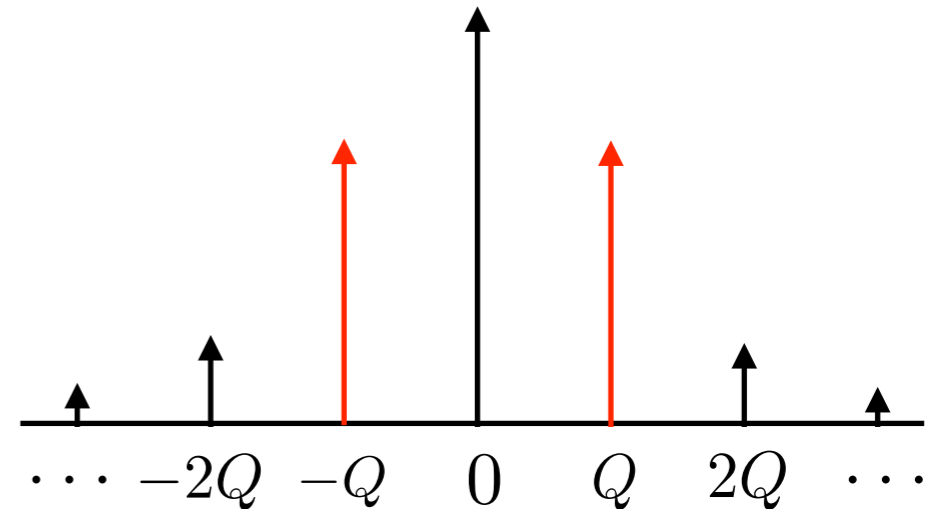


A different point of view

Helium:



Bragg peaks:

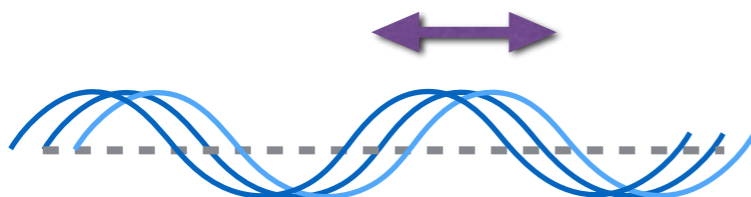


Focus on dynamics of principal Bragg vectors

Can we think of solid He-4 as a charge density wave (CDW)?

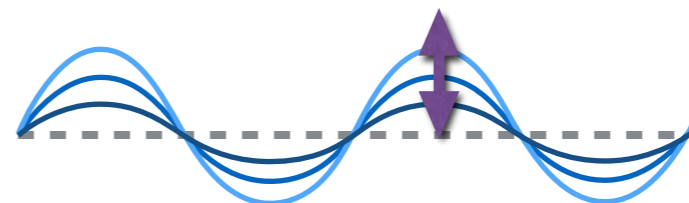
A CDW allows naturally for gapped modes:

"phason"



$$\omega \sim cq$$

"amplitudon"



$$\omega \sim \sqrt{m^2 + c^2q^2}$$

Ginzburg-Landau theory for 3D CDW

Density modulation: $\rho(\mathbf{r}) = n(\mathbf{r}) - n_0$

Assume order parameter is small (large fluctuations):

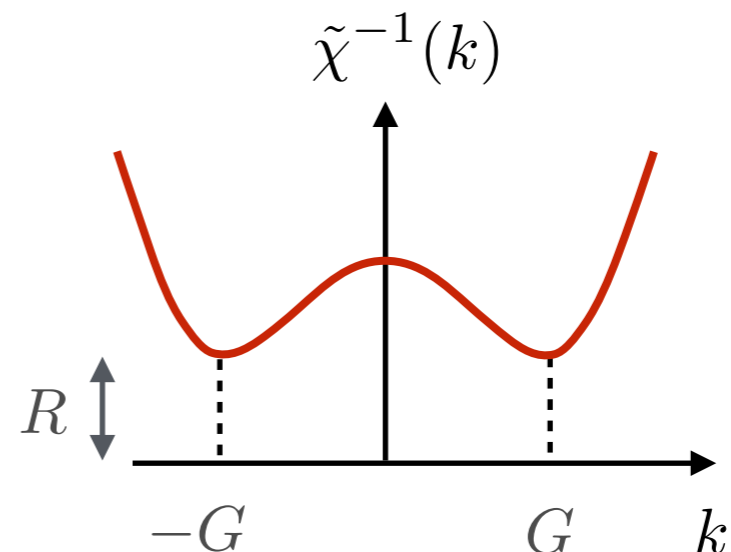
$$F_{\text{GL}} = \frac{1}{2} \int d\mathbf{r}_1 d\mathbf{r}_2 \rho(\mathbf{r}_1) \chi^{-1}(\mathbf{r}_1 - \mathbf{r}_2) \rho(\mathbf{r}_2) - B \int d\mathbf{r} \rho(\mathbf{r})^3 + C \int d\mathbf{r} \rho(\mathbf{r})^4$$

In Fourier space:

$$F_{\text{GL}} = \frac{1}{2} \int d\mathbf{k} \frac{1}{\tilde{\chi}(\mathbf{k})} |\rho(\mathbf{k})|^2 - B \int d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3 \rho(\mathbf{k}_1) \rho(\mathbf{k}_2) \rho(\mathbf{k}_3) \delta(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3) \\ + C \int d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3 d\mathbf{k}_4 \rho(\mathbf{k}_1) \rho(\mathbf{k}_2) \rho(\mathbf{k}_3) \rho(\mathbf{k}_4) \delta(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3 + \mathbf{k}_4)$$

Static susceptibility:

$$\tilde{\chi}^{-1}(\mathbf{k}) = R + a (\mathbf{k}^2 - G^2)^2$$



"Should all crystals be BCC?"

Alexander and McTague, PRL **41**, 702 (1978)

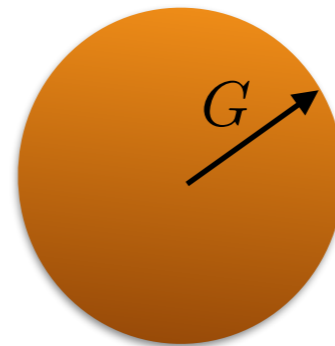
Baym, Bethe, and Pethick, Nuc. Phys. A**175**, 225 (1971)

Minimize F_{GL} , order by order:

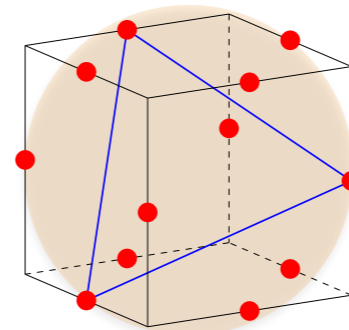
$$F_{GL} = \frac{1}{2} \int d\mathbf{k} \frac{1}{\tilde{\chi}(\mathbf{k})} |\rho(\mathbf{k})|^2 - B \int d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3 \rho(\mathbf{k}_1) \rho(\mathbf{k}_2) \rho(\mathbf{k}_3) \delta(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3) + C \int d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3 d\mathbf{k}_4 \rho(\mathbf{k}_1) \rho(\mathbf{k}_2) \rho(\mathbf{k}_3) \rho(\mathbf{k}_4) \delta(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3 + \mathbf{k}_4)$$

1. "Condense" on sphere $k=G$

$$\rho(\mathbf{k}) \neq 0 \quad |\mathbf{k}| = G$$



2. Maximize number of triangles with zero total momentum



"Should all crystals be BCC?"

Alexander and McTague, PRL **41**, 702 (1978)

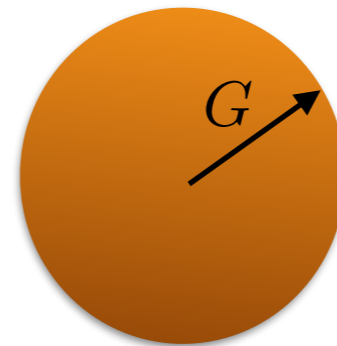
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Minimize F_{GL} , order by order:

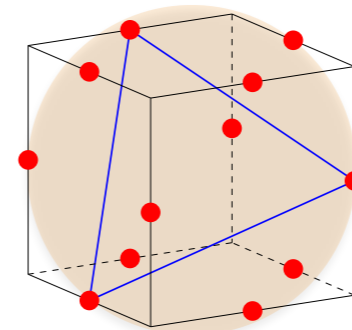
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1. "Condense" on sphere $k=G$

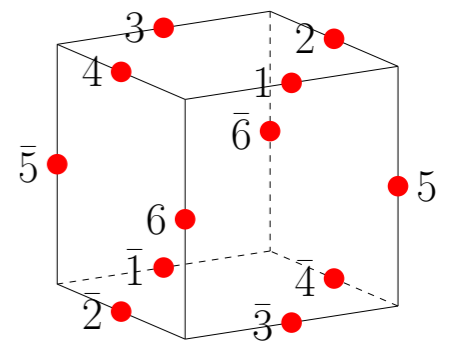
$$\rho(\mathbf{k}) \neq 0 \quad |\mathbf{k}| = G$$



2. Maximize number of triangles with zero total momentum



BCC wins:



Collective modes

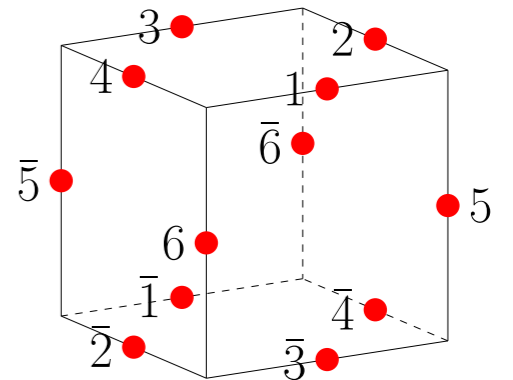
Dynamical Ginzburg-Landau:

$$L = \frac{1}{2} \int d^3r \left(\frac{\partial \rho}{\partial t} \right)^2 - F_{\text{GL}}$$

Fluctuations about mean-field:

$$\rho(\mathbf{r}, t) = \sum_i (\bar{\rho}_i + \psi_i(\mathbf{r}, t)) e^{i\mathbf{G}_i \cdot \mathbf{r}}$$

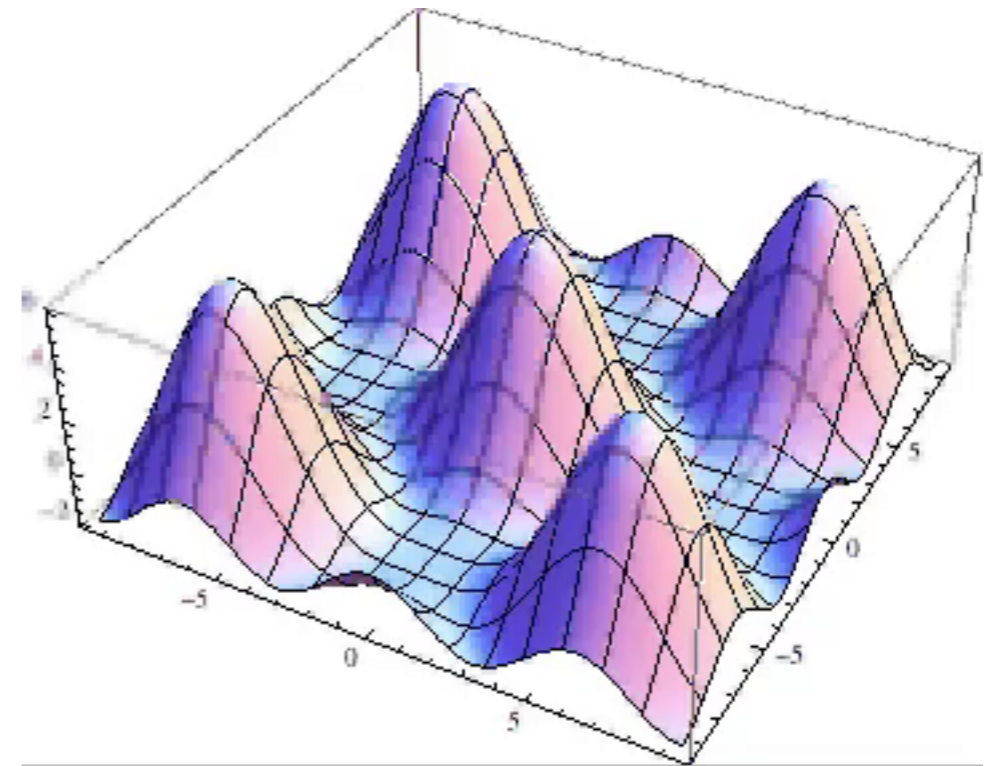
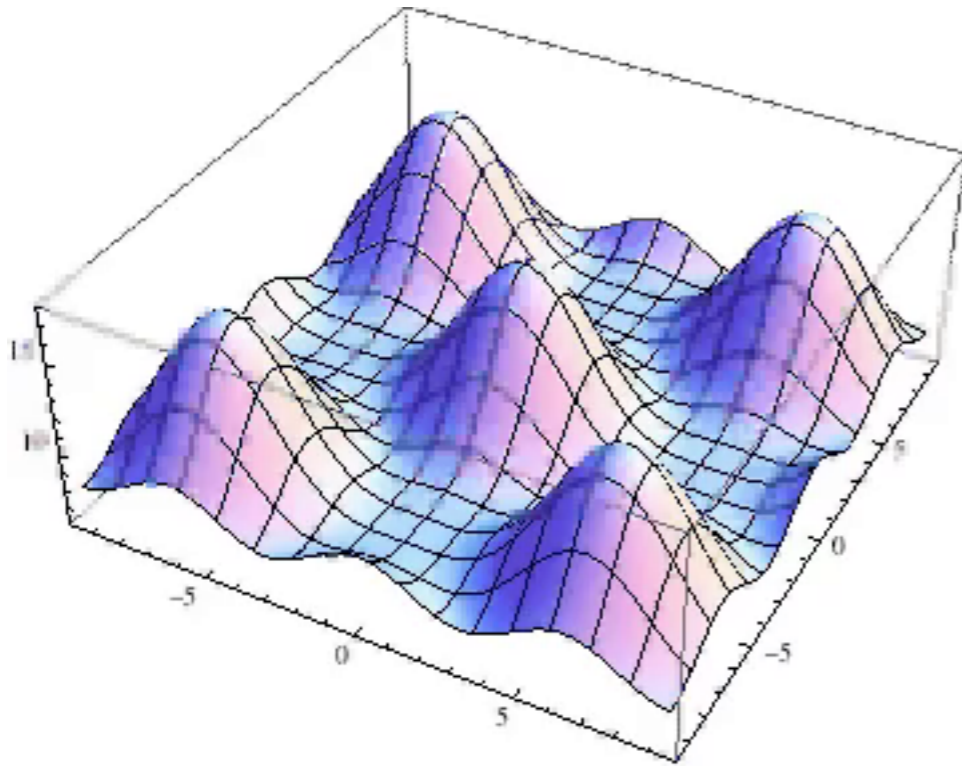
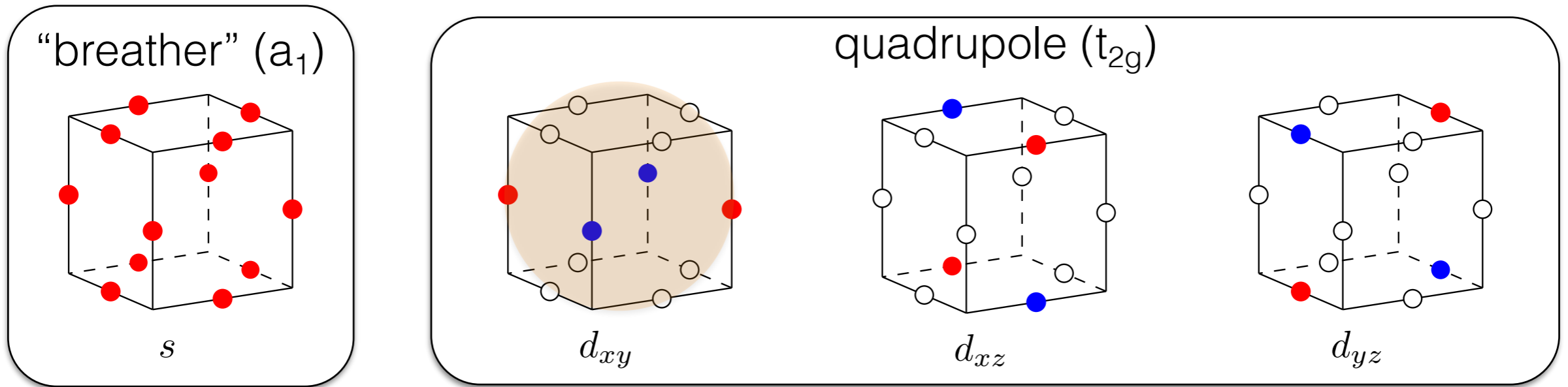
$$\psi_i(\mathbf{r}, t) = \psi_{-i}^*(\mathbf{r}, t)$$



Linearize Euler-Lagrange equations

6 pairs of reciprocal lattice vectors \Rightarrow 12 modes

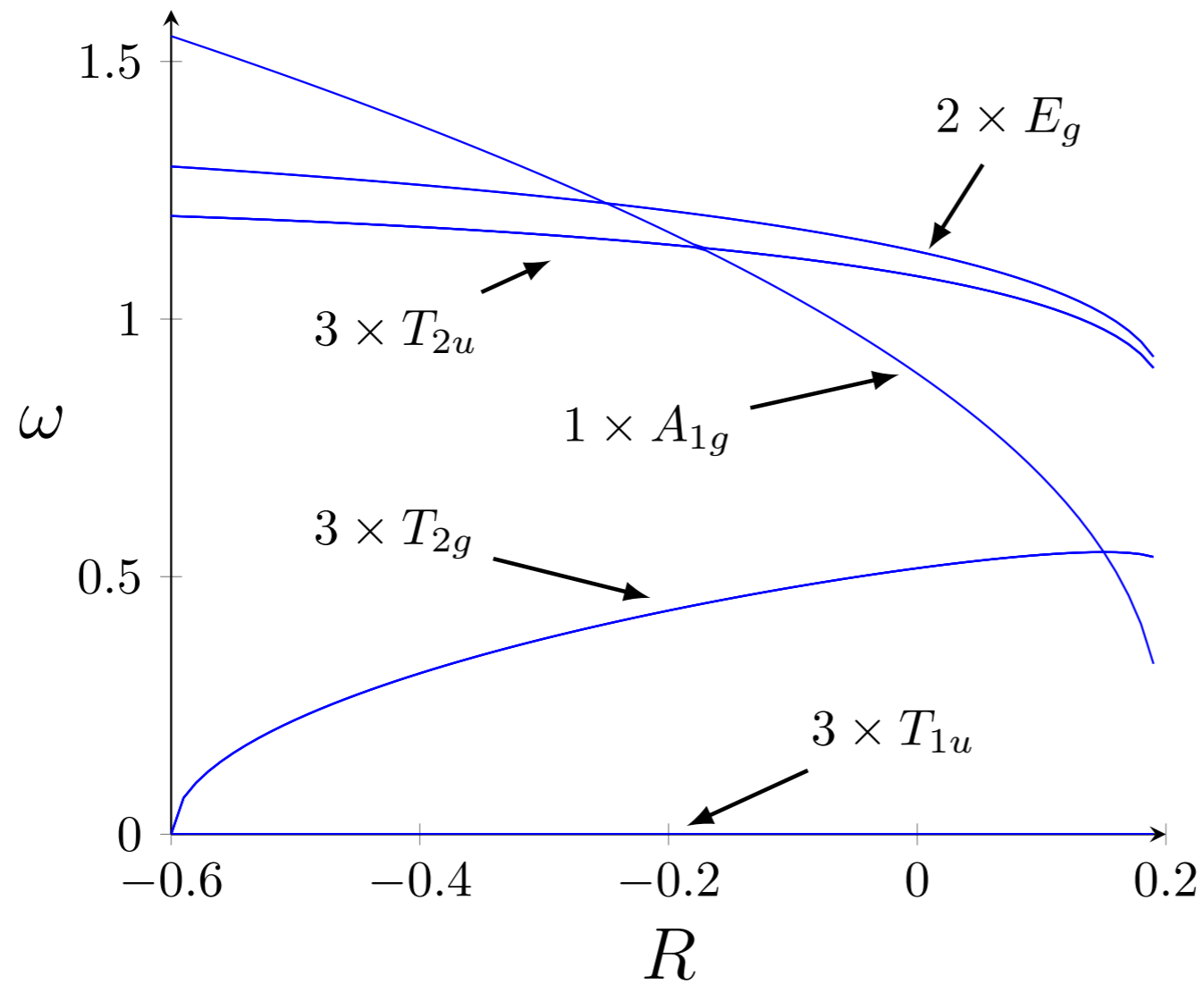
Visualizing the optical modes



d_{xy} “quadrupolon” has vanishing z-axis spring constant \Rightarrow flat band

Which gapped mode is lowest?

Breather or quadruplon, depends on GL parameters:



Neutron scattering

Dynamical structure factor: $S(\mathbf{q}, \omega) = \text{Im} \{ \langle \delta\rho_{\mathbf{G}}(\mathbf{q}, \omega) \delta\rho_{-\mathbf{G}}(-\mathbf{q}, -\omega) \rangle \}$

Compute by quantizing Ginzburg-Landau action. Result:

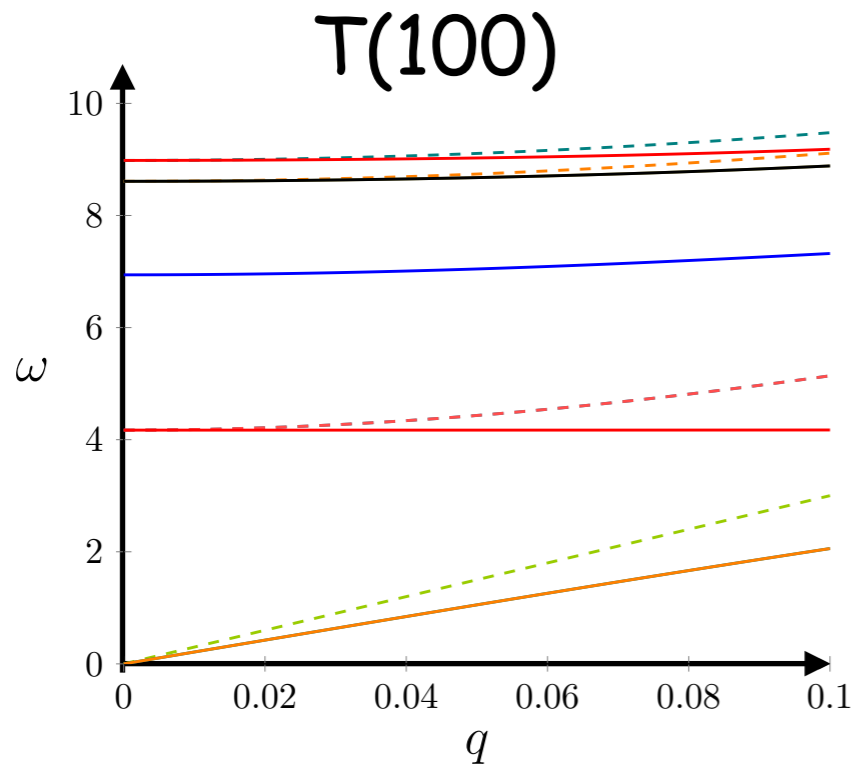
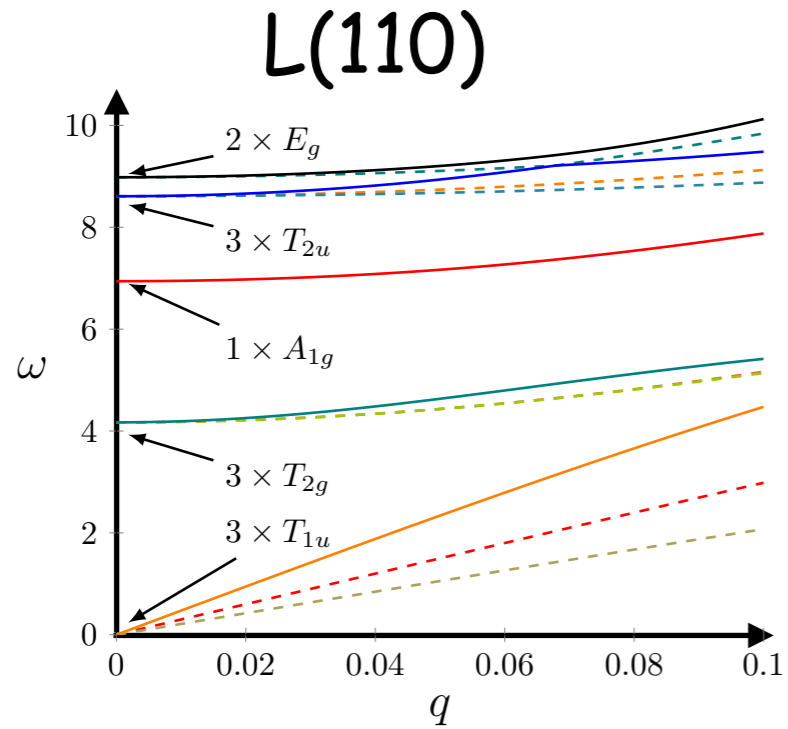
$$S(\mathbf{G} + \mathbf{q}, \omega) = \sum_{\alpha} \frac{M_{\mathbf{G},\alpha}(\mathbf{q})}{2\omega_{\alpha}(\mathbf{q})} [(1 + n_B(\omega_{\alpha}))\delta(\omega - \omega_{\alpha}(\mathbf{q})) - n_B(\omega_{\alpha})\delta(\omega + \omega_{\alpha}(\mathbf{q}))]$$

Note — sum rule is satisfied:

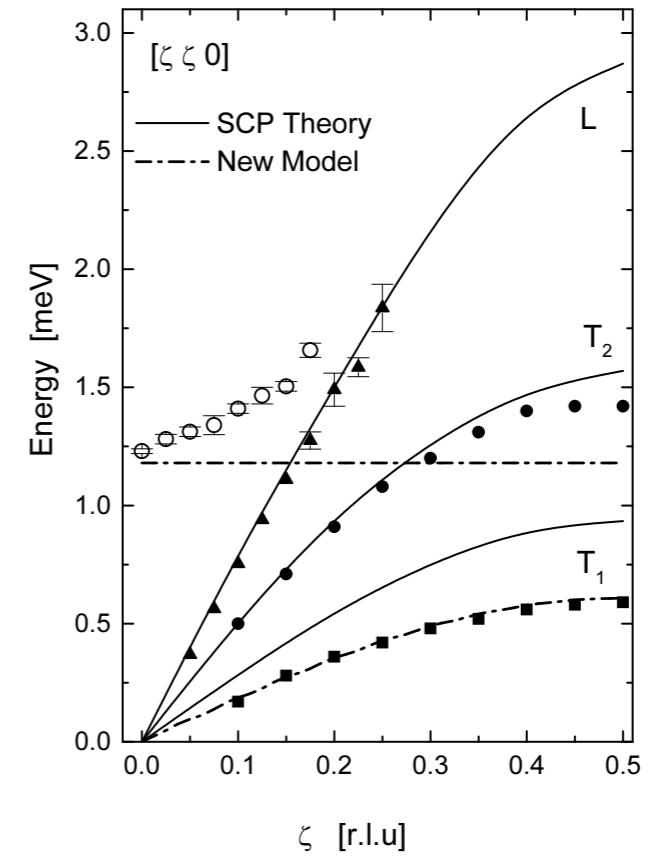
$$\int_{-\infty}^{\infty} d\omega \omega S(\mathbf{G} + \mathbf{q}, \omega) = 1$$

Hence, despite having more phonons than predicted by harmonic theory, the overall spectral weight remains the same. We have not introduced spurious degrees of freedom.

Comparison to experiment

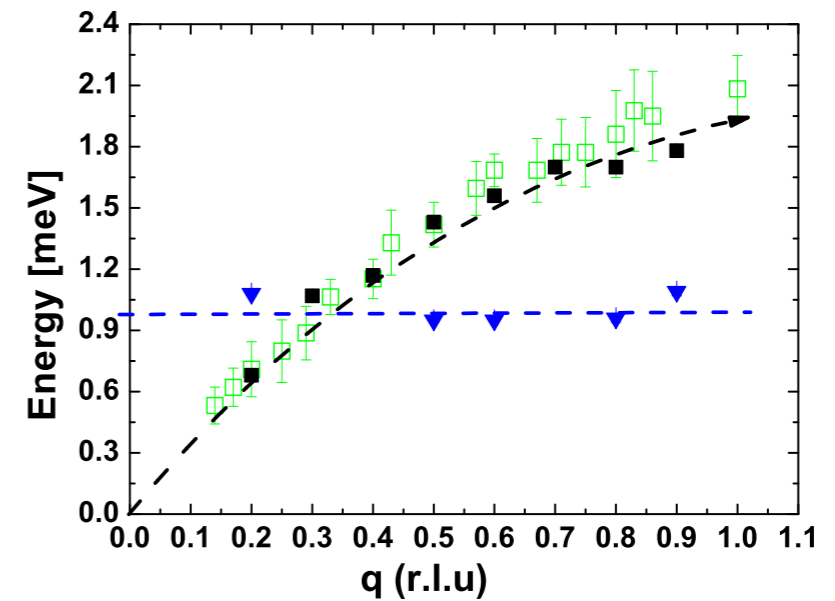


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Markovic et al., PRL 88, 195301 ('02)

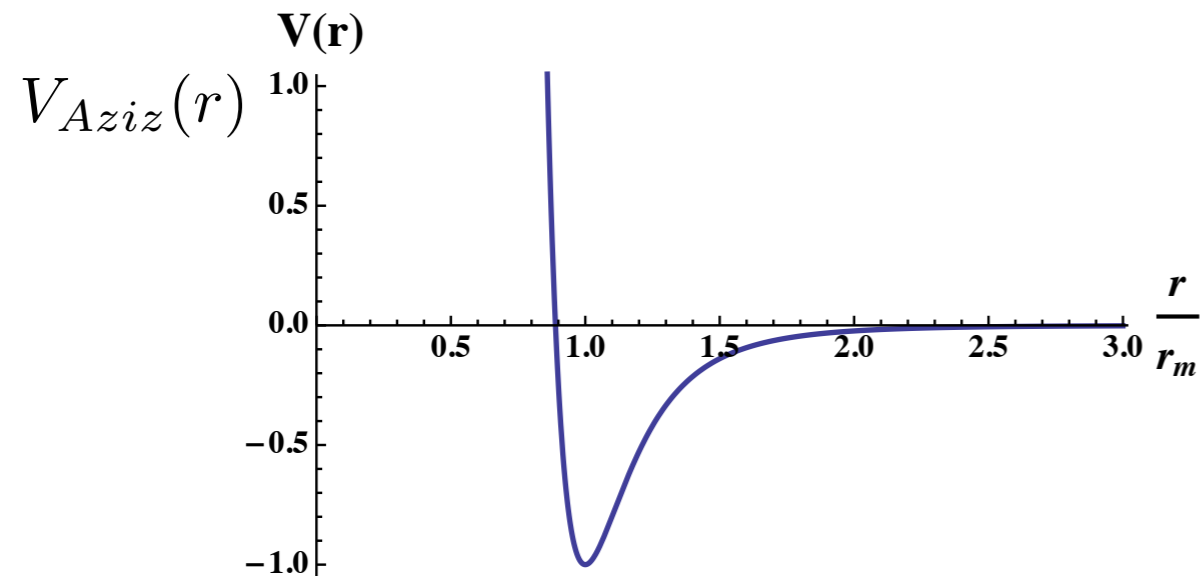
T(100)



Pelleg et al, PRB 73, 180301R ('06)

Quantum Monte Carlo

AB-initio simulations



$$H = -\frac{1}{2m} \sum_{i=1}^N \nabla_i^2 + \sum_{i < j} V_{Aziz}(r_i - r_j)$$

$$n_0 = 0.02854 \text{ \AA}^{-3}$$

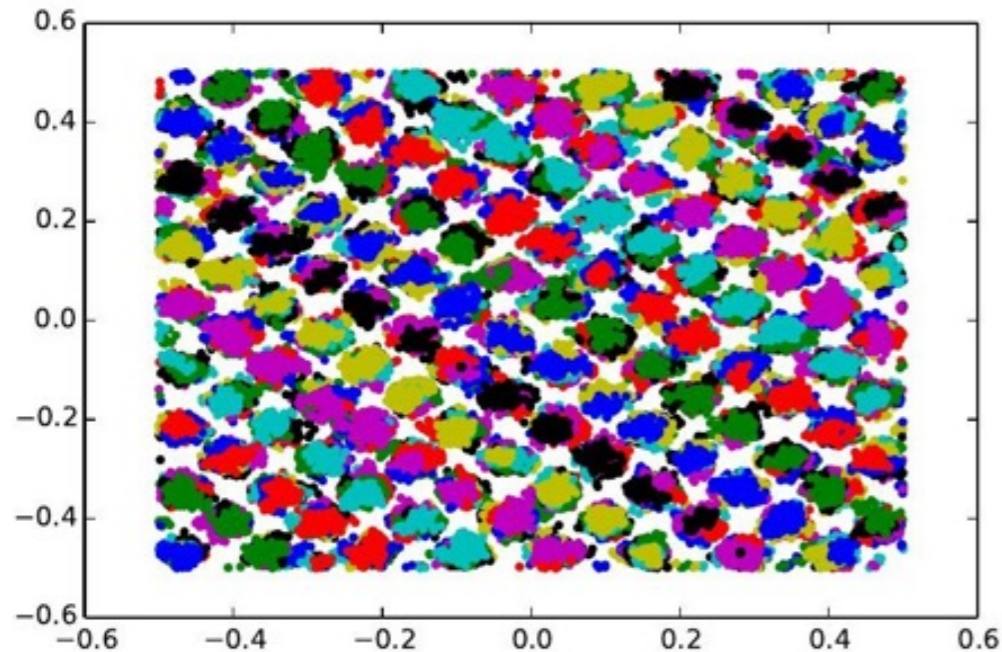
$$T = 1.6 \text{ K}$$

Place 2000 atoms in a box with periodic BC

Simulate using continuous space path integral QMC

Quantum Monte Carlo

2000 He4 Atoms



BCC phase

large zero point motion

Structure factor:

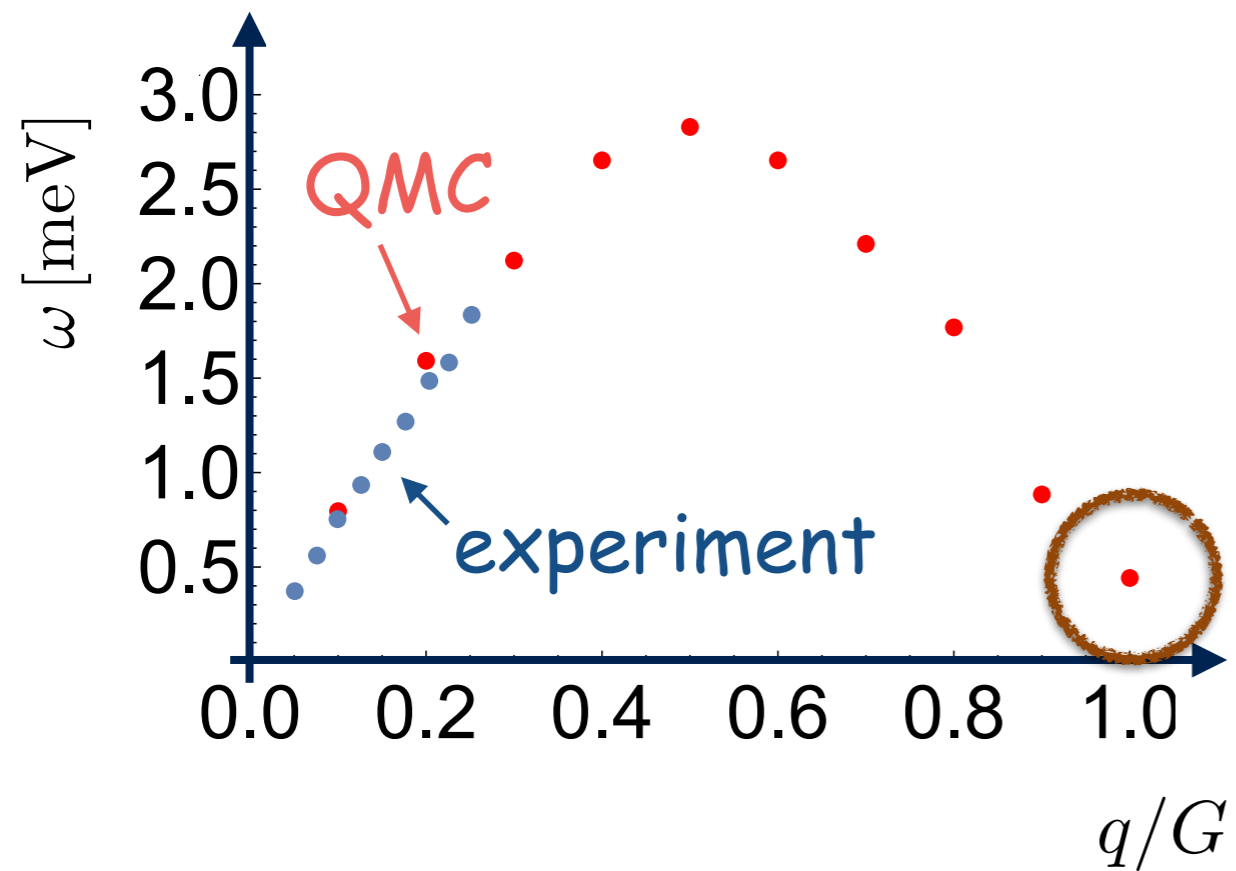
$$S(\mathbf{q}, \omega) = \langle \rho(\mathbf{q}, \omega) \rho(-\mathbf{q}, -\omega) \rangle$$

$$\rho(\mathbf{q}, t) = \sum_n e^{i\mathbf{q} \cdot \mathbf{r}_n(t)}$$

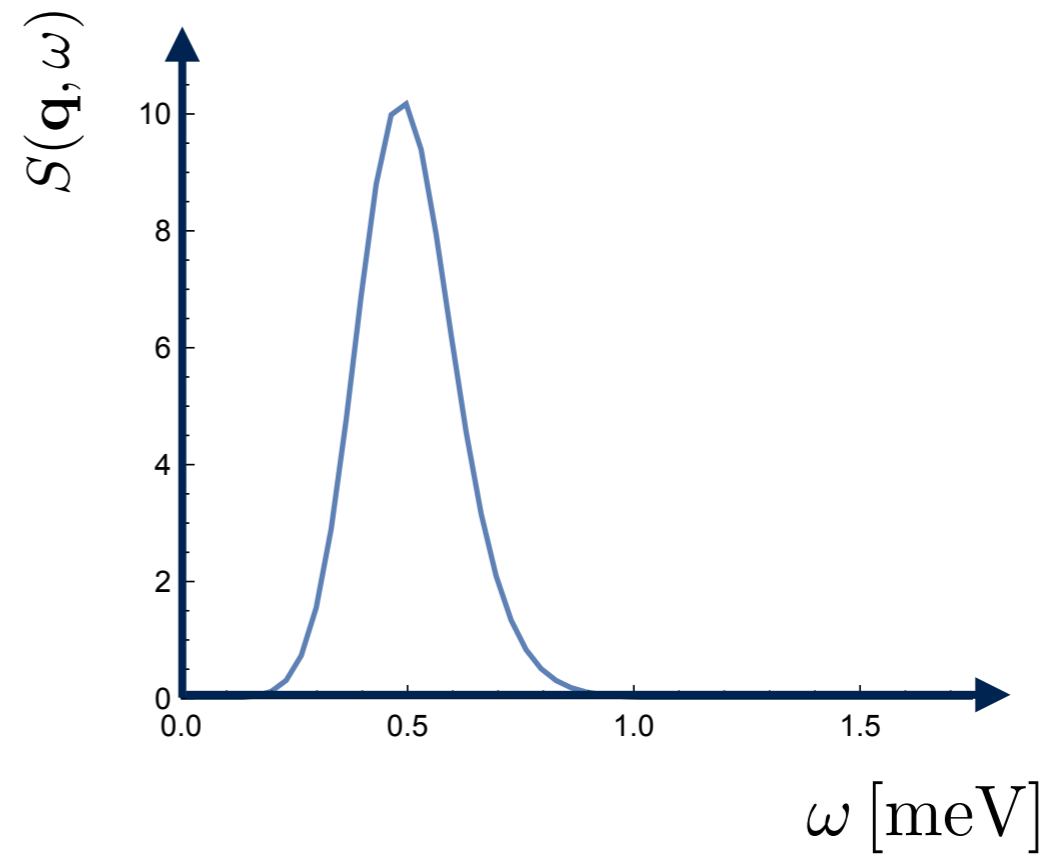
QMC simulations are performed along the imaginary time axis, perform numerical analytical continuation to real time.

QMC results

L(110) dispersion

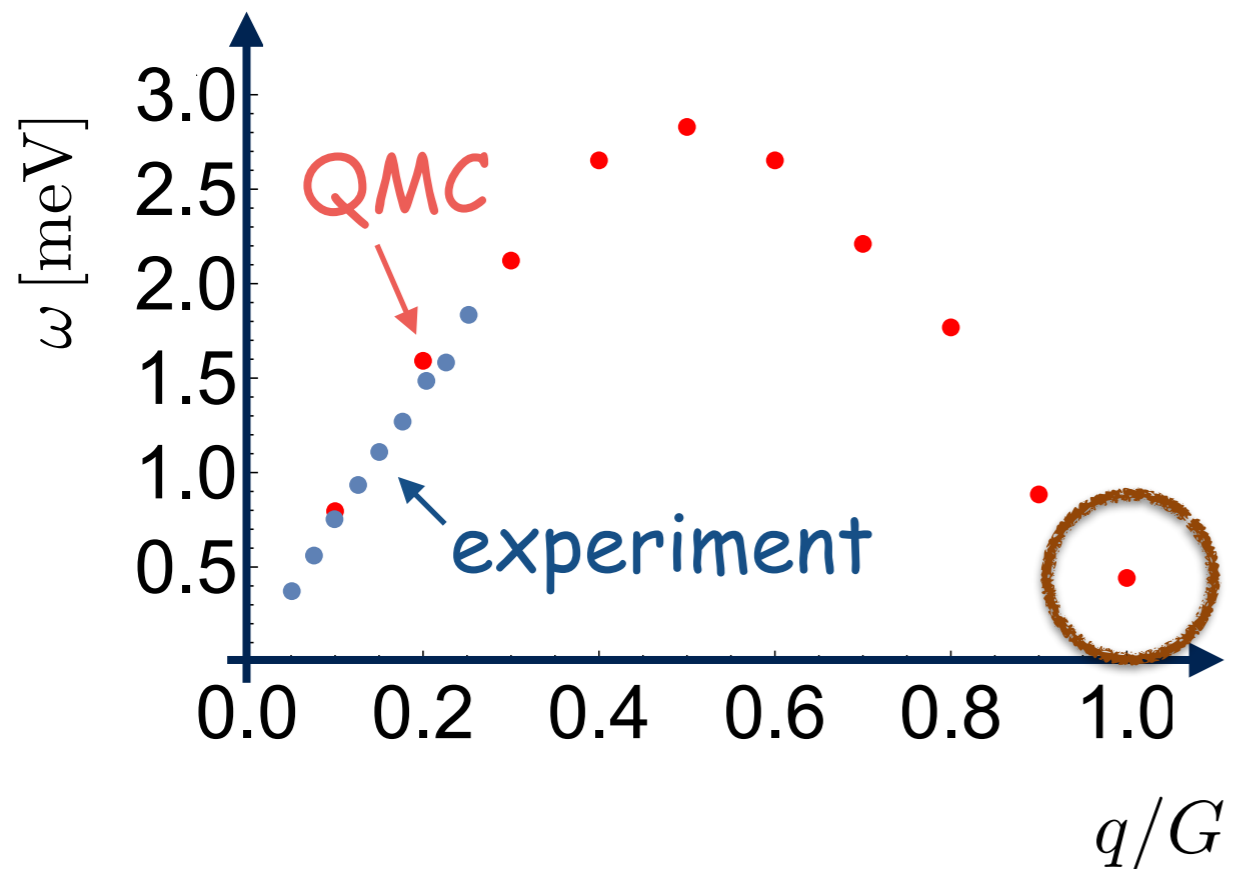


At Bragg vector:

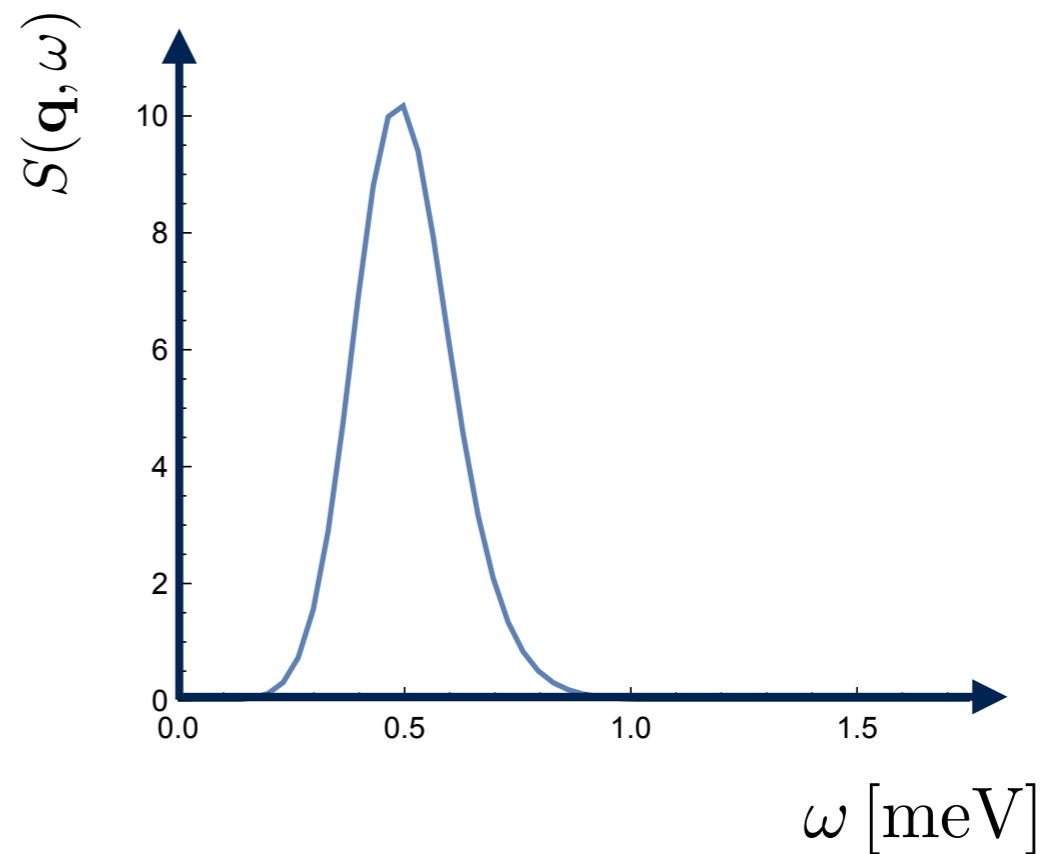


QMC results

L(110) dispersion



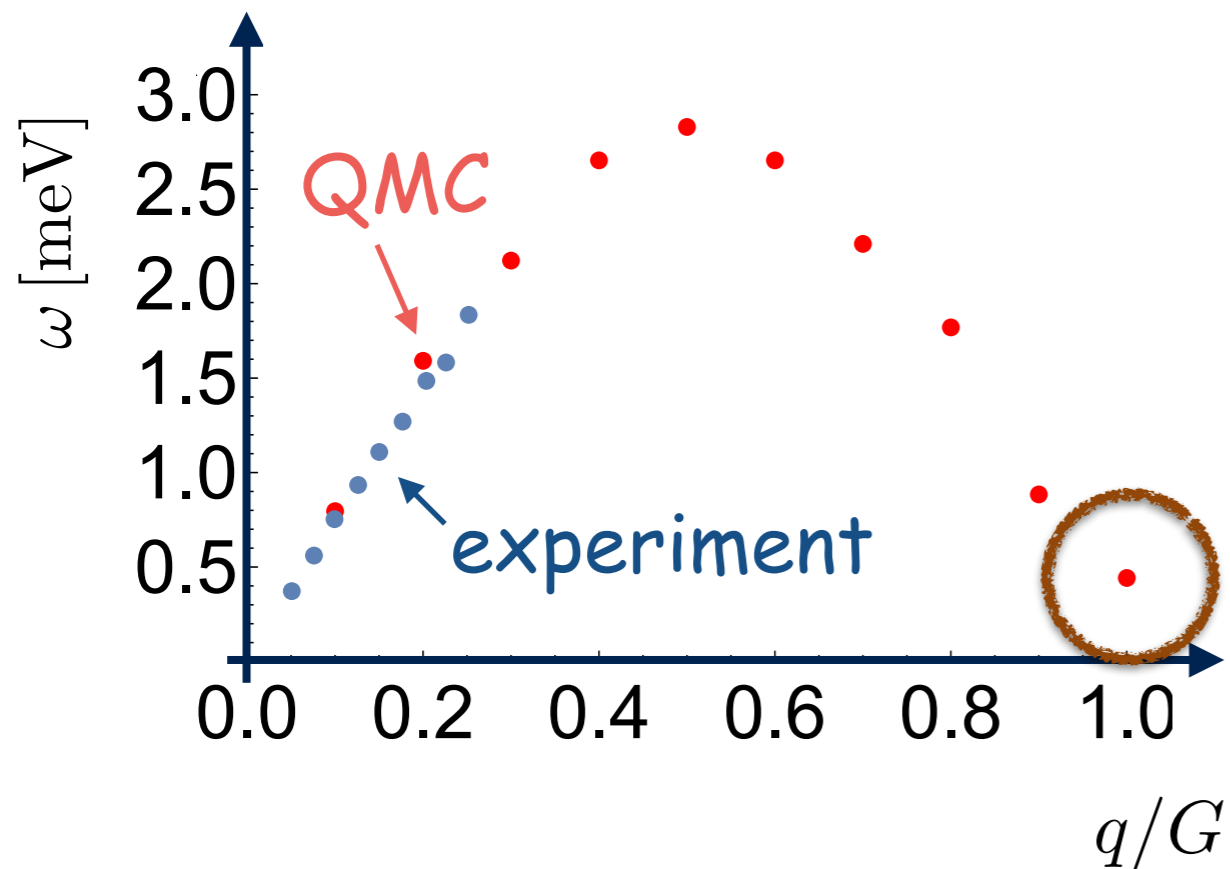
At Bragg vector:



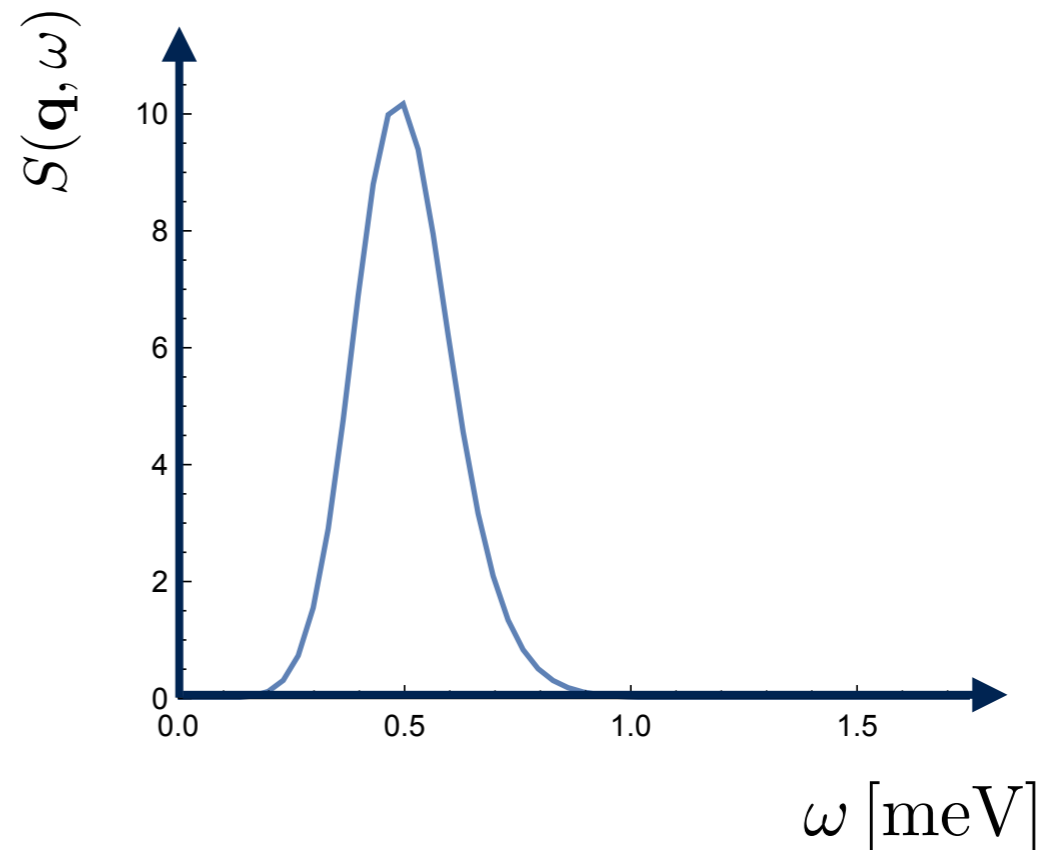
Confirms presence of optical mode (e.g. not due to crystal domains).

QMC results

L(110) dispersion



At Bragg vector:



Confirms presence of optical mode (e.g. not due to crystal domains).

Small energy. Difficulty with analytical continuation?

Lowest optical mode missed by experiment?

Summary:

- * In Helium, harmonic theory fails due to large zero point motion
- * New "harmonic theory" for the optical modes
- * QMC finds low energy optical mode — could it be there?
- * Prediction: such modes should appear in other quantum solids (solid Helium-3, 2d and 3d CDWs, etc)