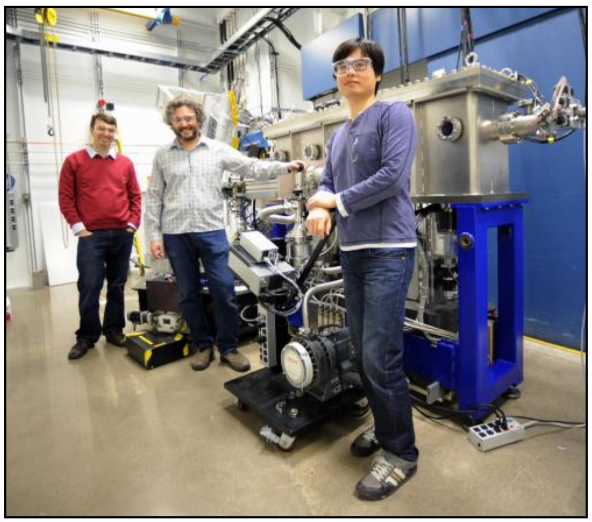
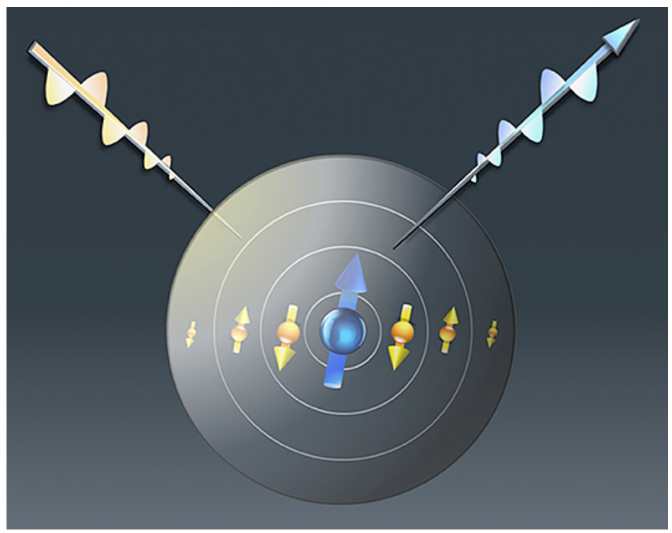
Introduction to Inelastic X-Ray Scattering

Jason N. Hancock
University of Connecticut
jason.hancock@uconn.edu





Neutron and X-ray Scattering School
Argonne National Laboratory, Argonne, IL

Outline

- Introductions
- Dispersion relations in materials physics
- Elastic waves and classification of phonons
- Phonon inelastic X-ray scattering examples
- Resonance phenomena in multi-electron atoms
- Resonant inelastic X-ray scattering examples

UConn Condensed Matter Physics

Alexander Balatsky
Theoretical physics
Superconductivity
Quantum materials

Gayanath Fernando
Theoretical physics
Electronic structure
Quantum materials

Boris Sinkovic
Thin films synthesis
Electron spectroscopy

Lea F. Dos Santos
Theoretical physics
Quantum chaos

Elena Dormidontova
Soft Matter Theory

Jason Hancock
THz, Infrared, X-ray
Applied physics

Ilya Sochnikov
Low-T transport
Scanning SQUID

Pavel Volkov
(Harvard until 2023)
Materials theory
Twistronics

Niloy Dutta
Photonics
Applied physics

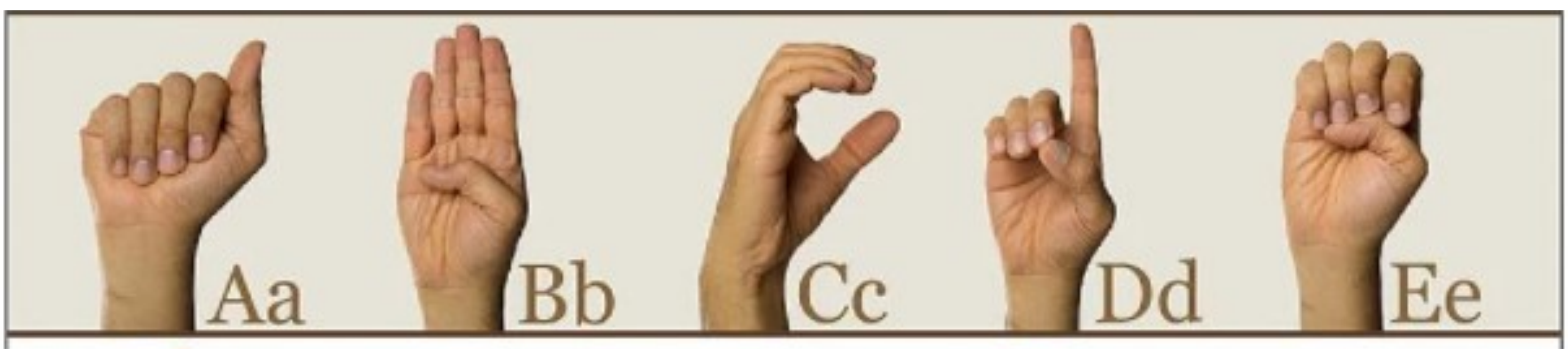
Menka Jain
Thin film synthesis

Barrett Wells
PLD films, Muons,
Neutrons, ARPES

Plus strong connections with: AMO group, UConn Institute for Materials Science, New UConn Tech Park

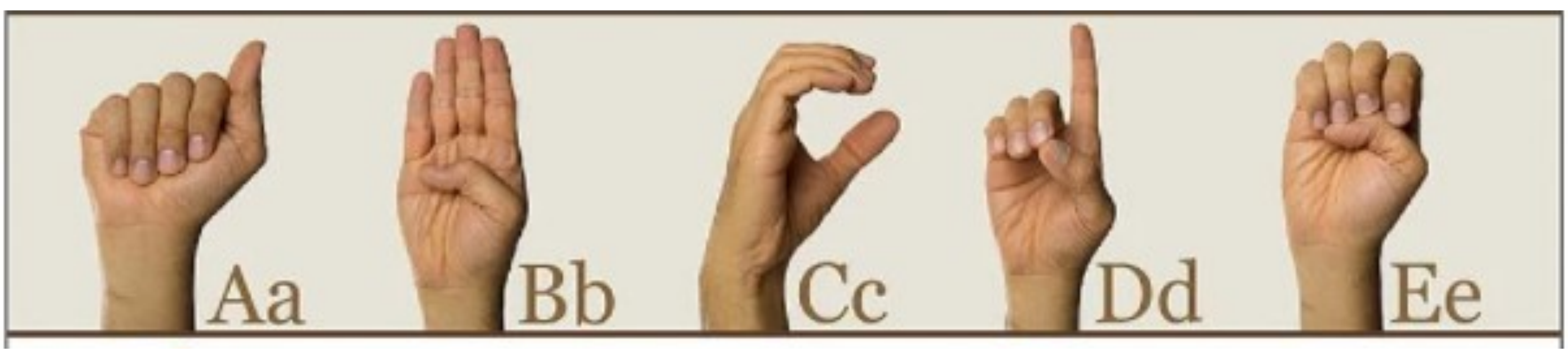
My PhD research does/will likely use…

A	B	C	D	E
X-rays	Neutrons	Both X-rays and Neutrons	Neither	I am a theorist

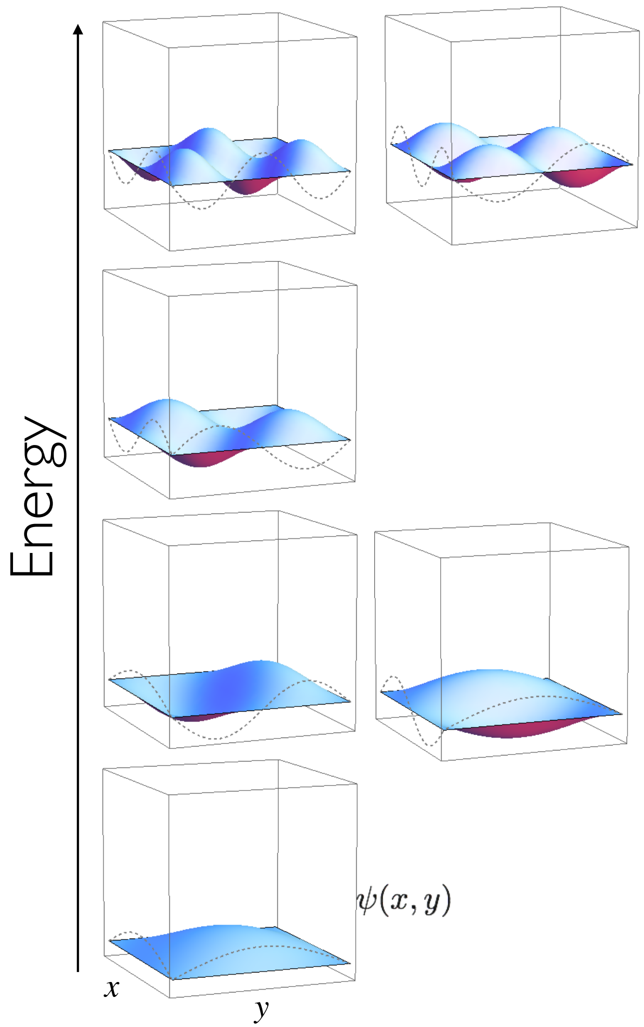


My PhD research does/will likely regard…

A	B	C	D	E
Mostly structural studies	Spectroscopy and electronic structure	Advanced imaging	Still figuring it out	None of these



Particle-in-a-box: the simplest quantum problem



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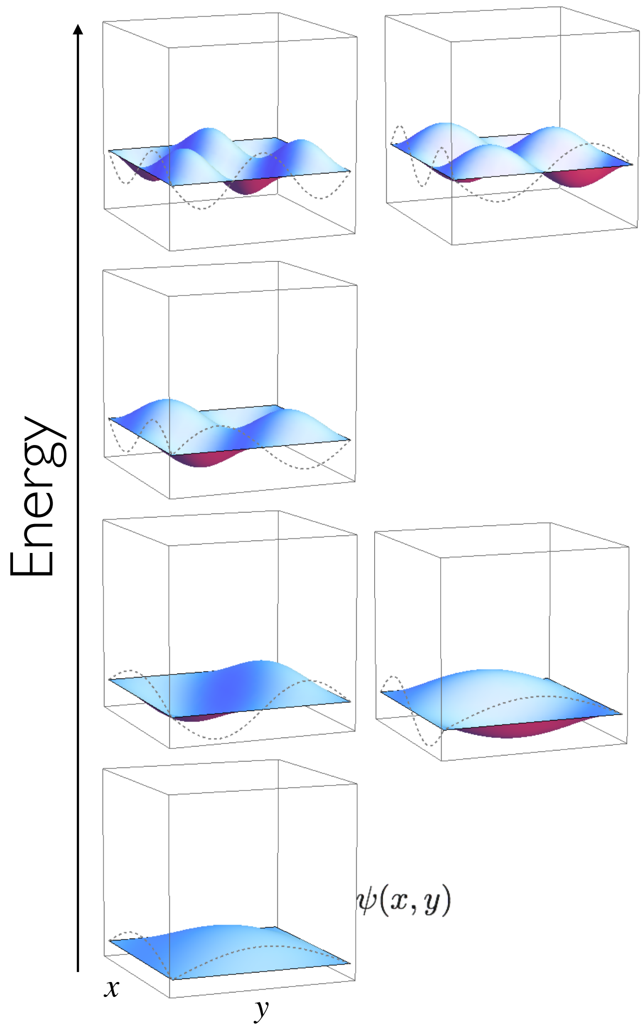

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Kinetic energy

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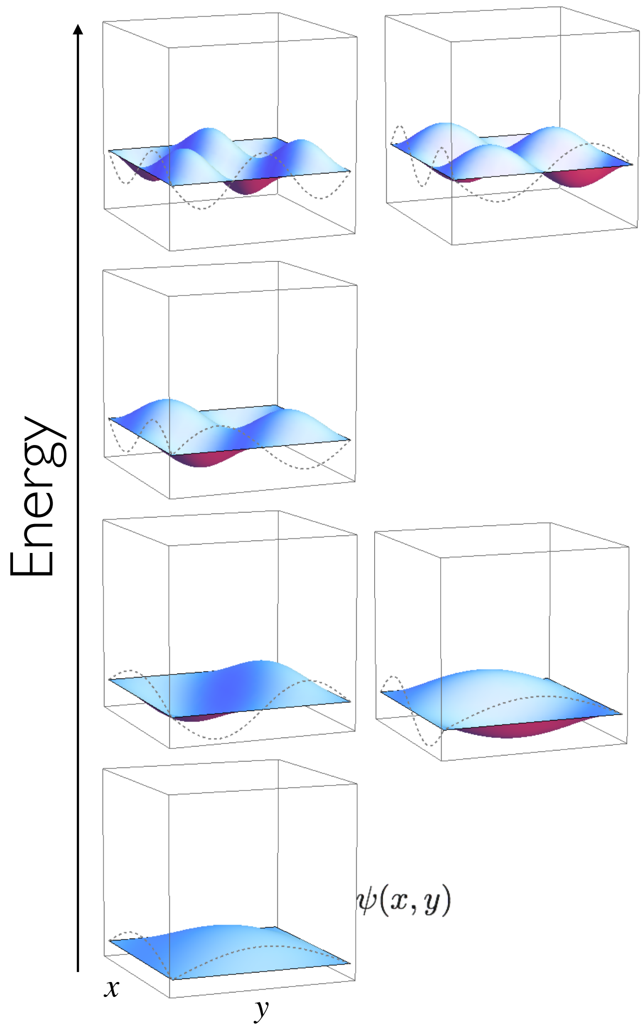

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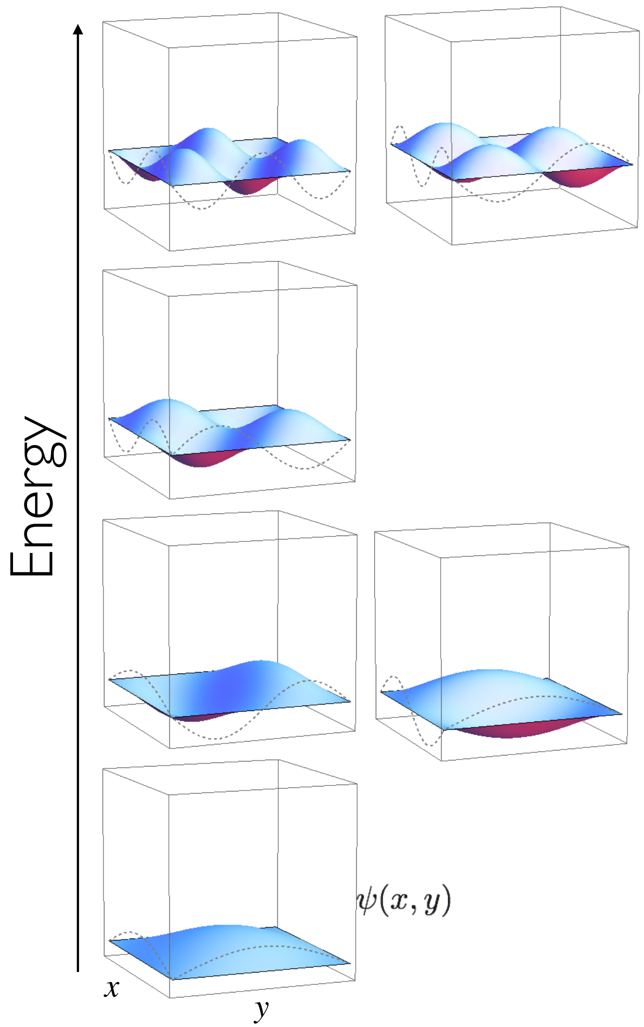

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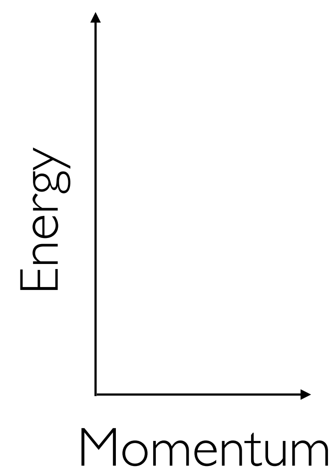

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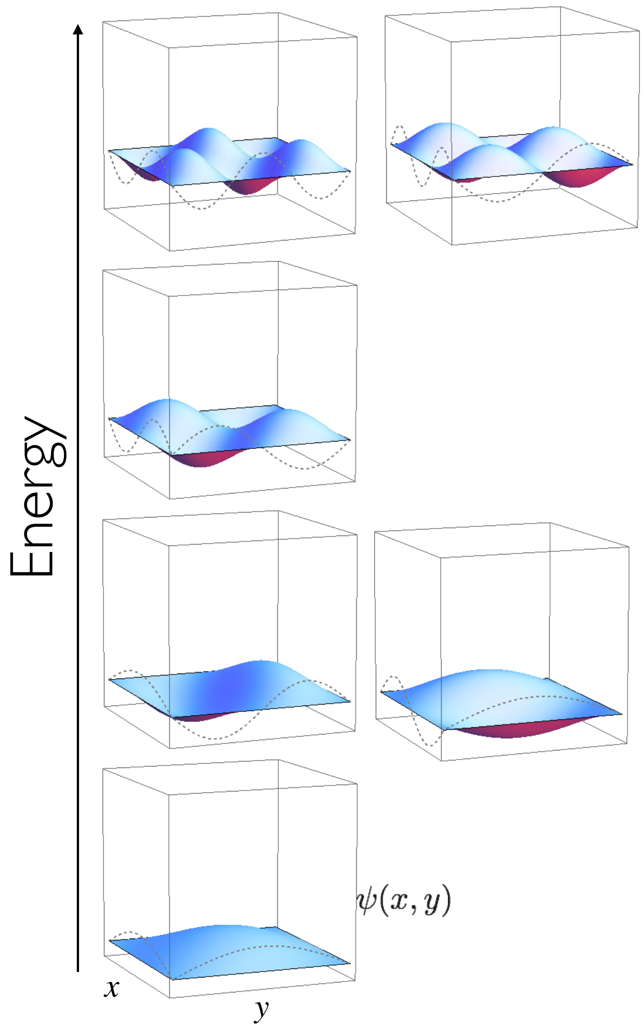

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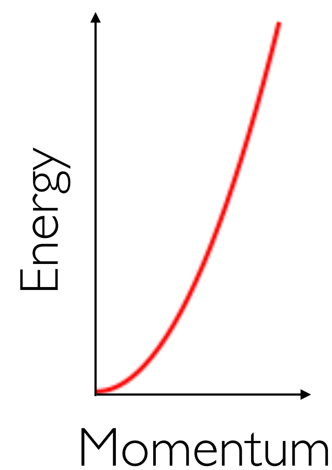

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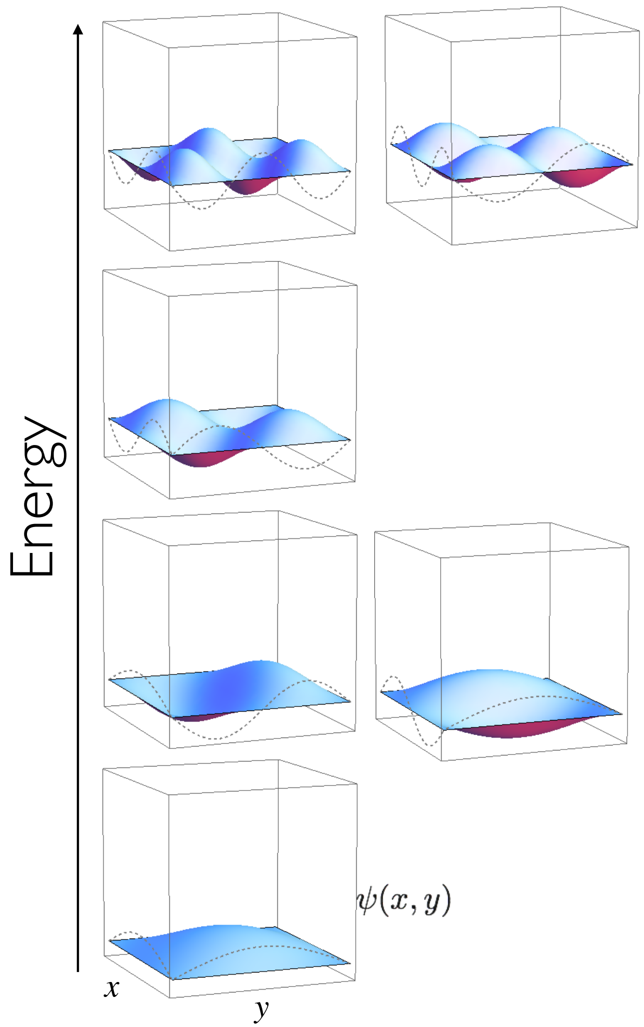

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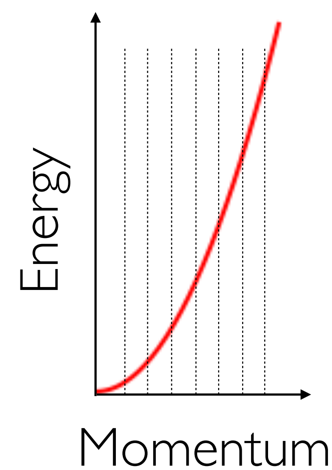

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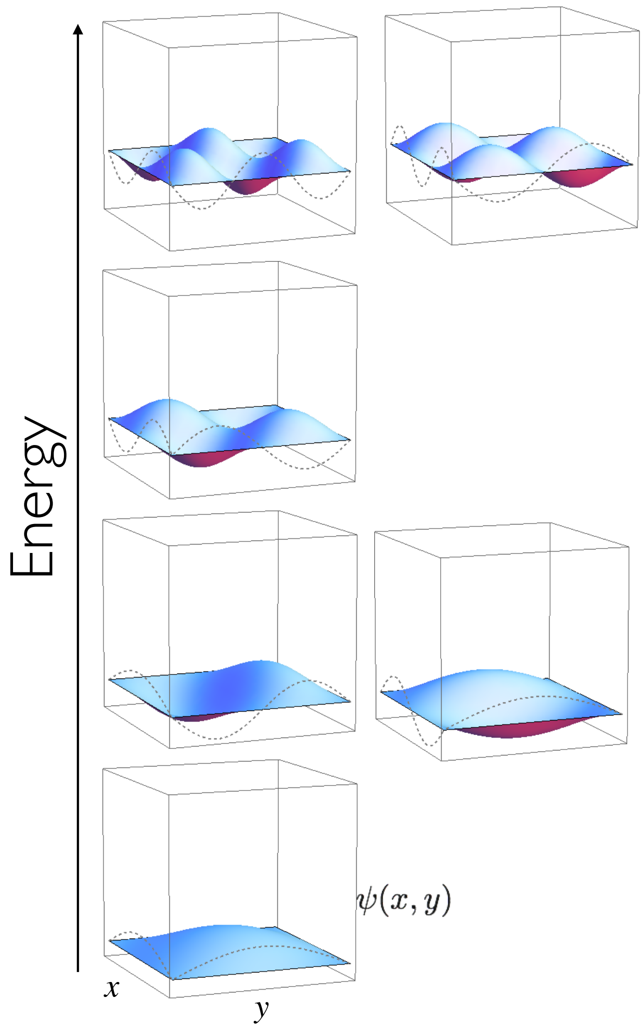

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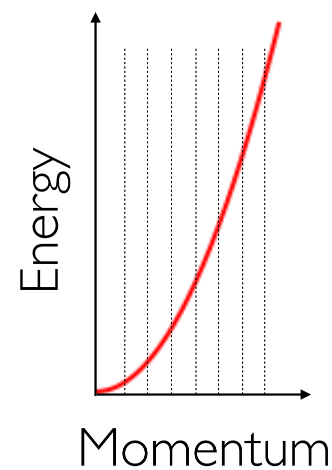

Dispersion relation

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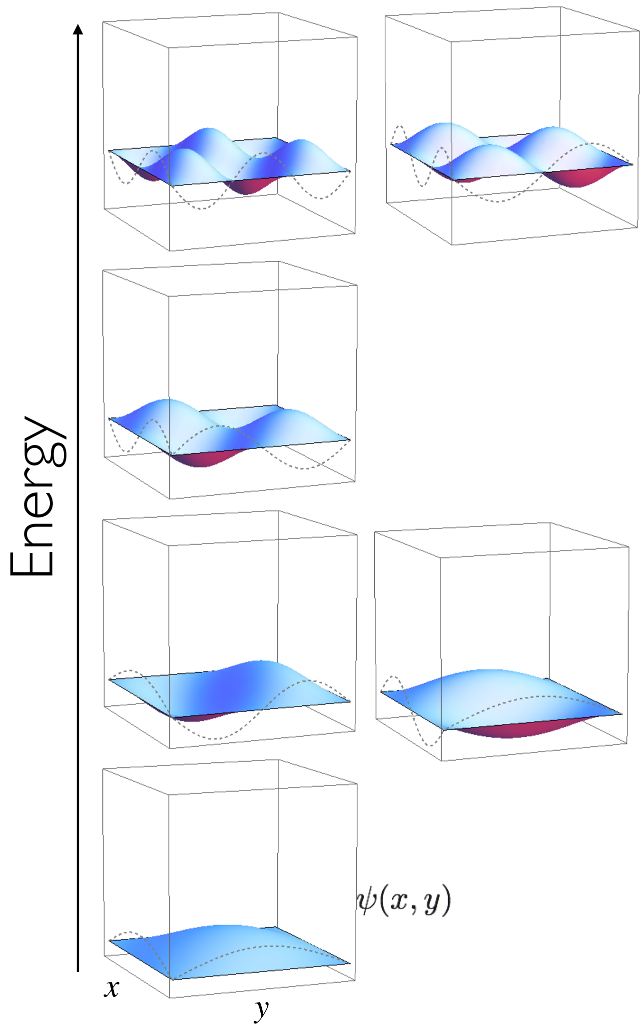

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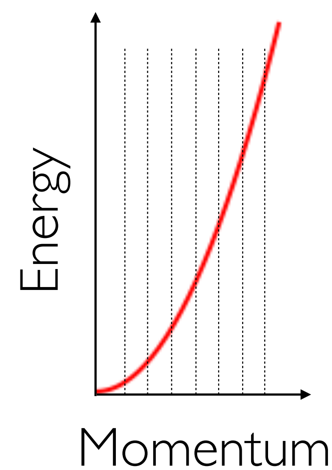
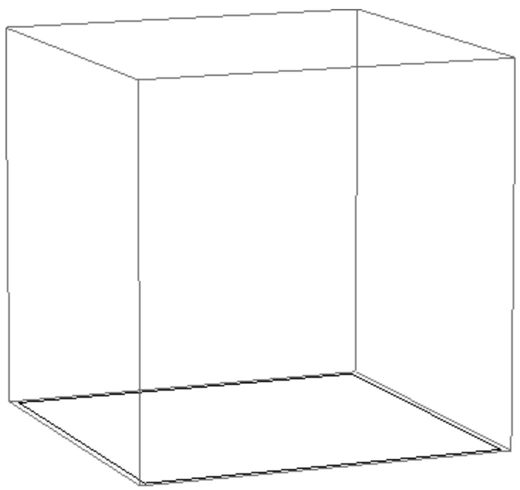


$$E \rightarrow i\hbar\frac{\partial}{\partial t}$$

$$p \rightarrow -i\hbar\frac{\partial}{\partial x}$$

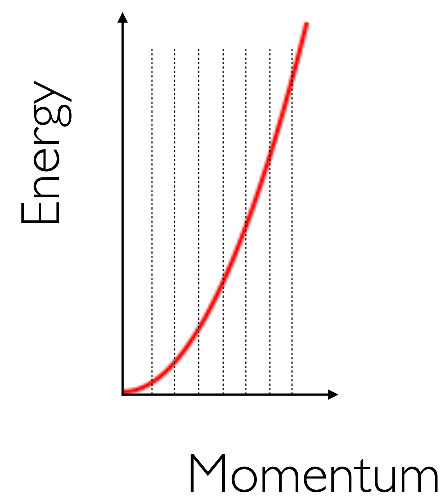
Dispersion relation

Particle in a box to the band theory of solids



$$H\psi = E\psi$$

$$H = \frac{p^2}{2m} + V(\vec{r})$$



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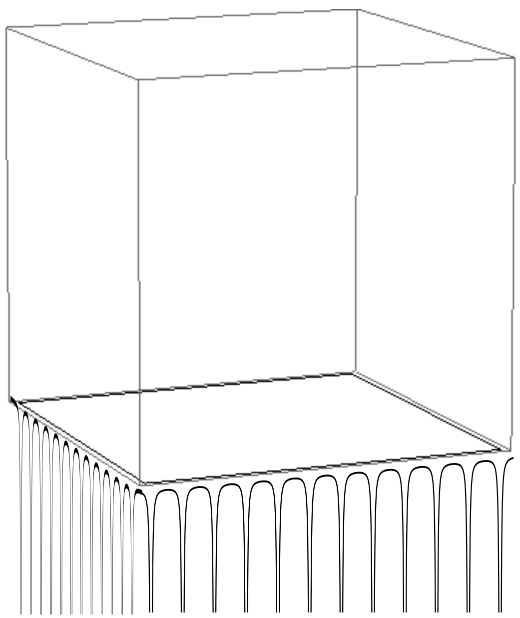

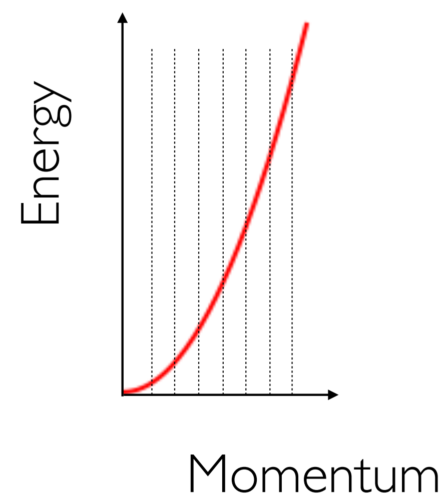

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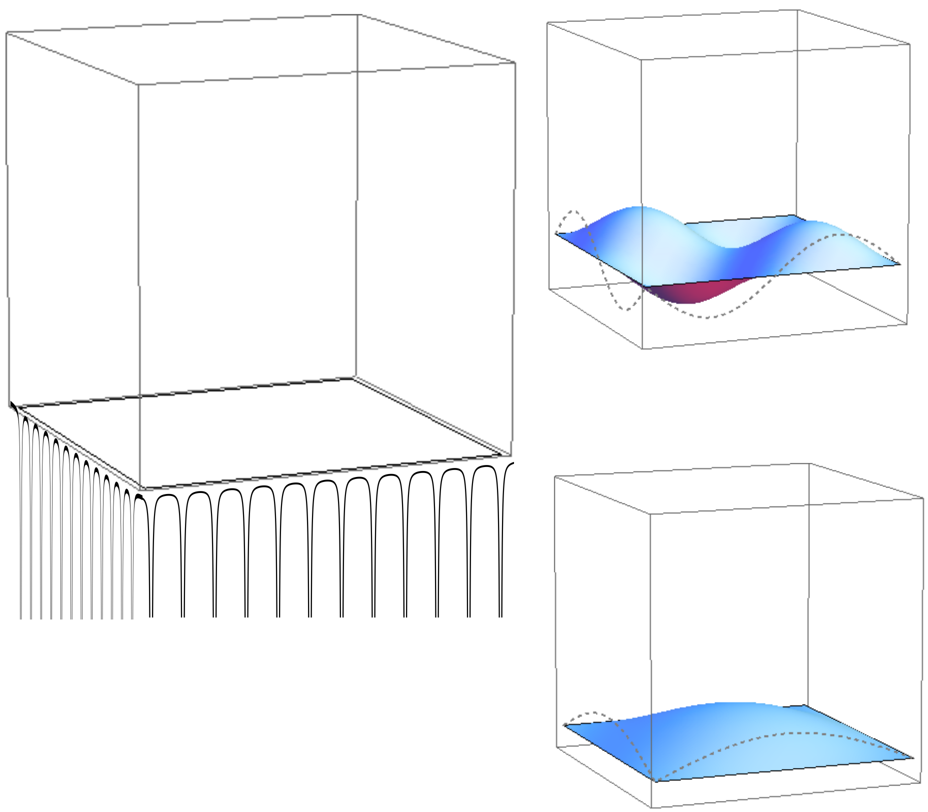

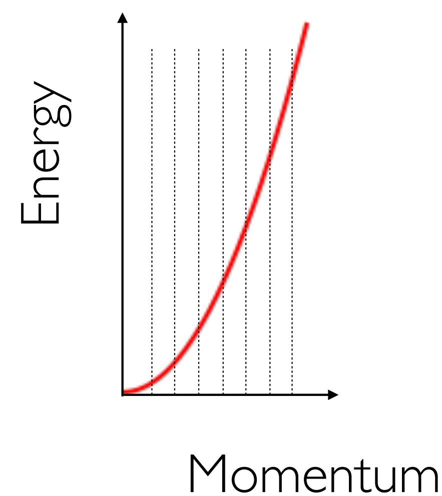

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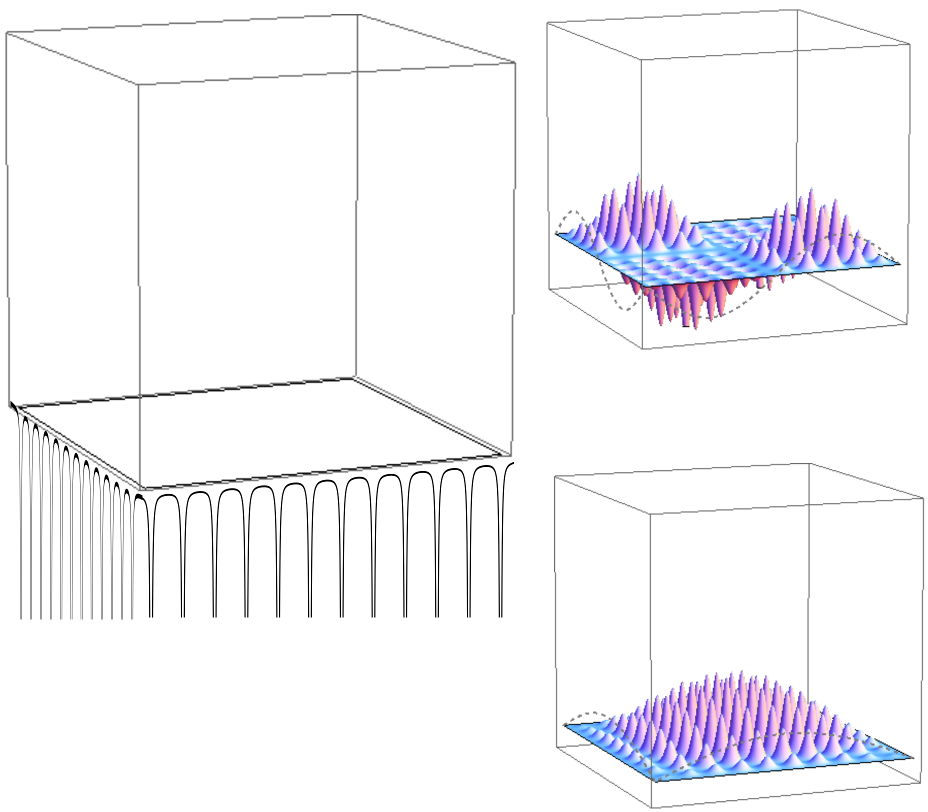

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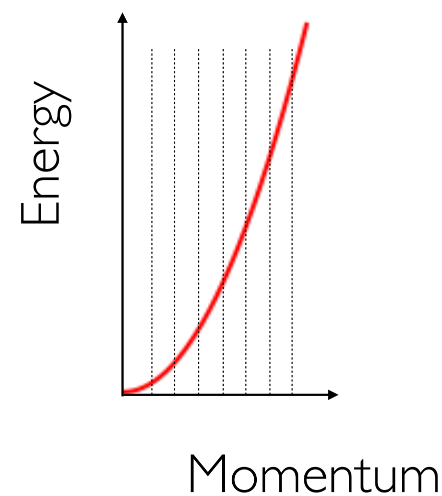

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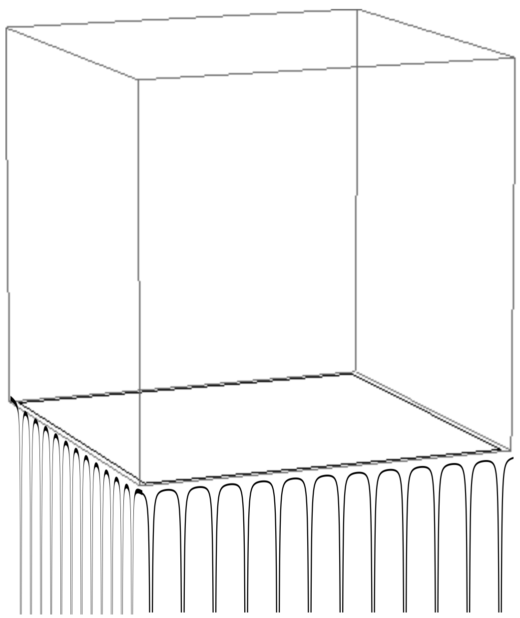
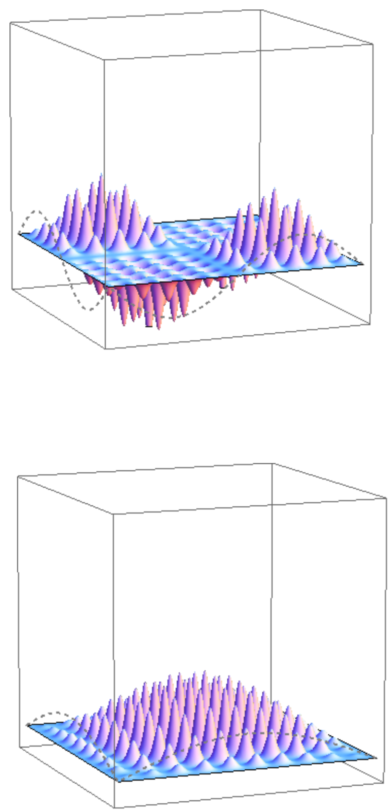
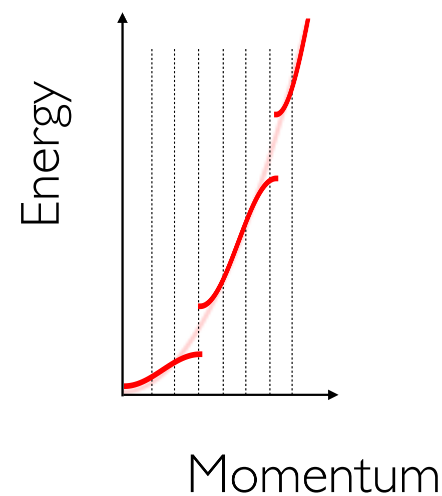
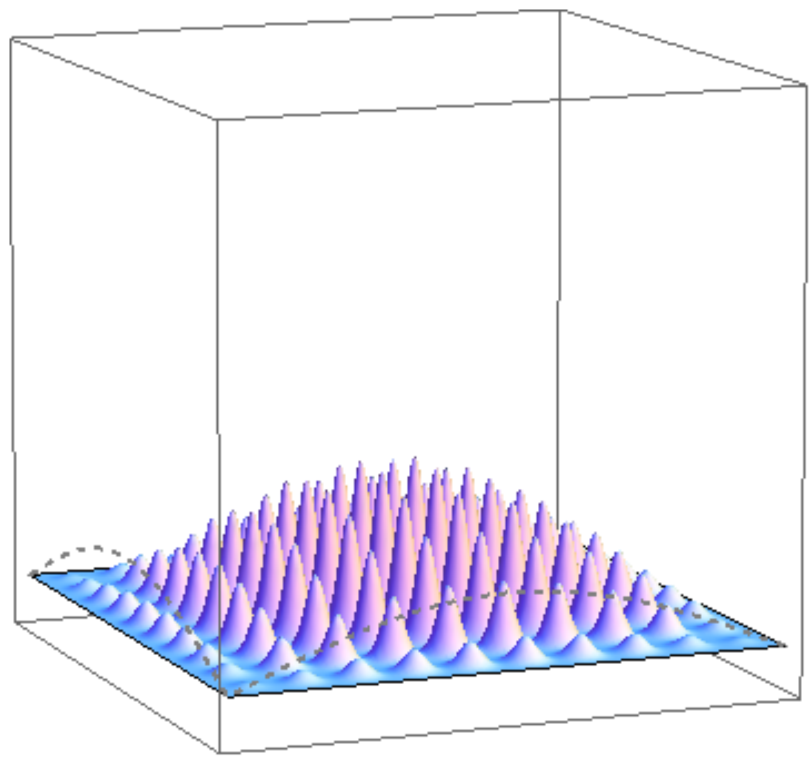
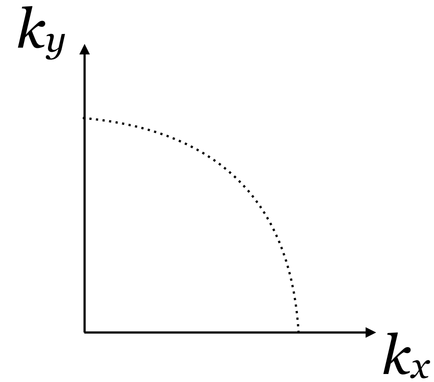






Many electron states





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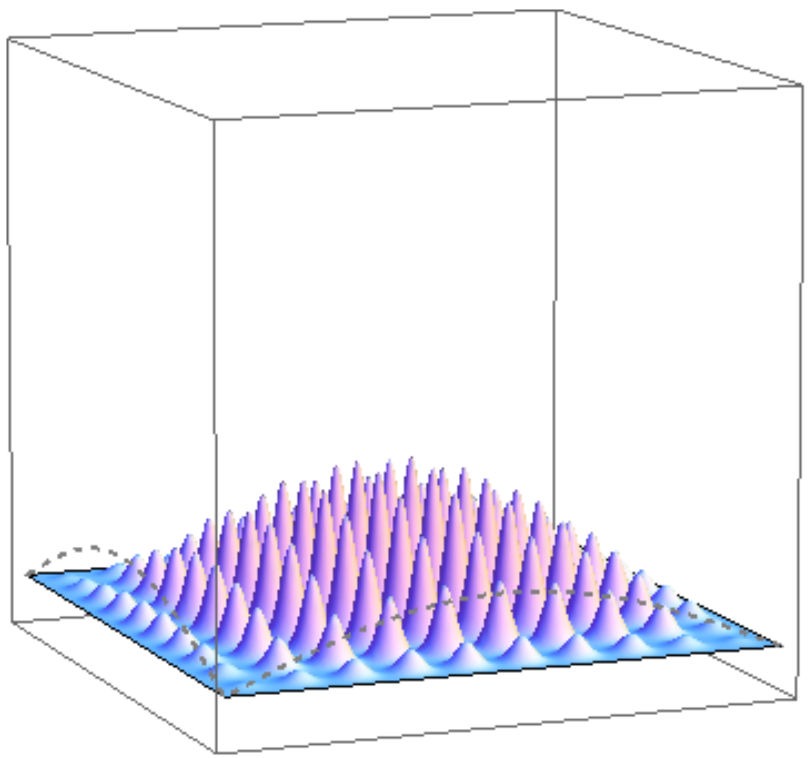

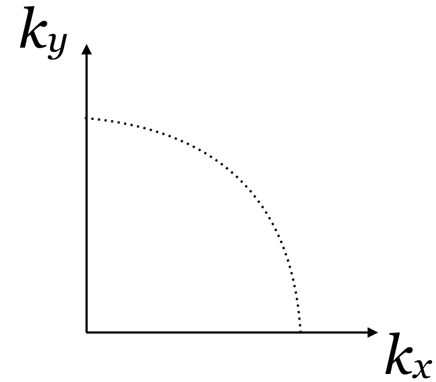

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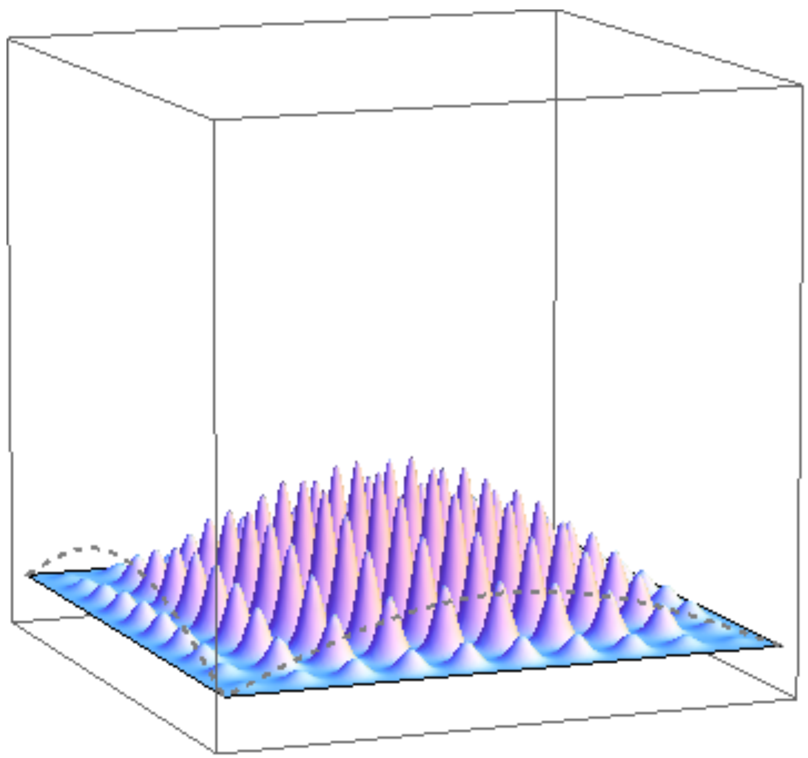

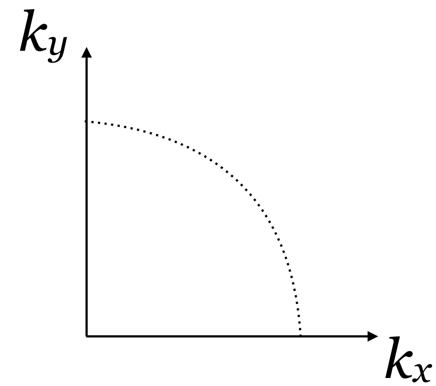

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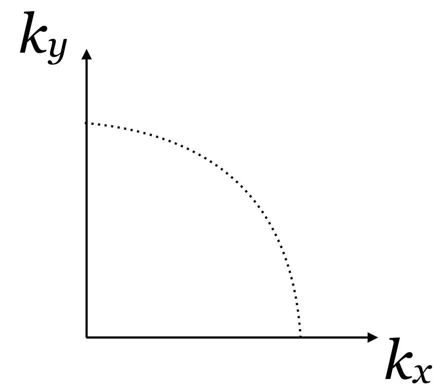

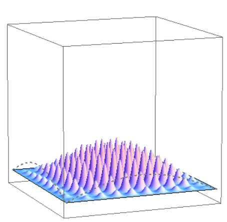

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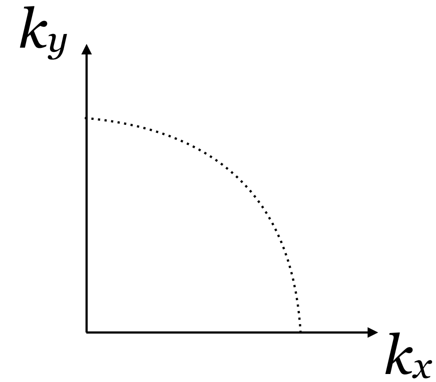

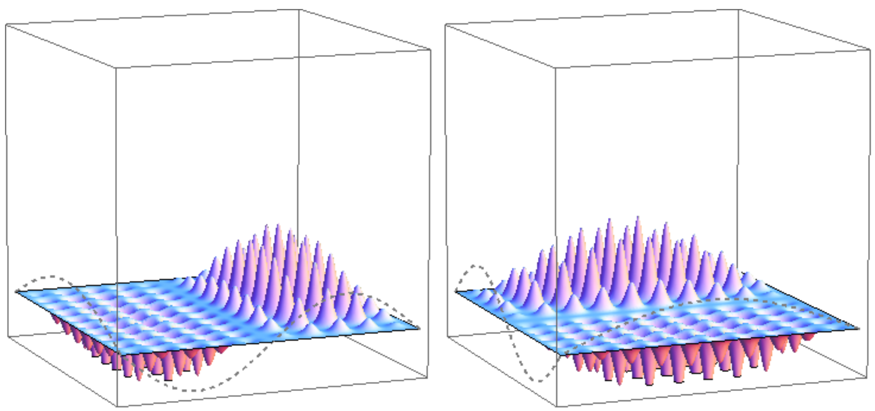

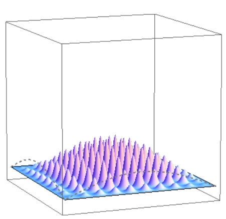

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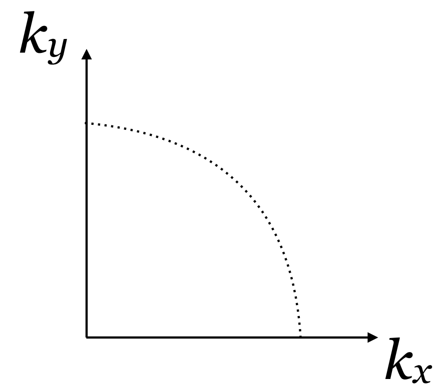

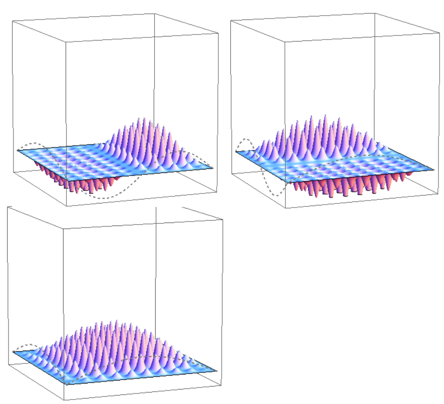

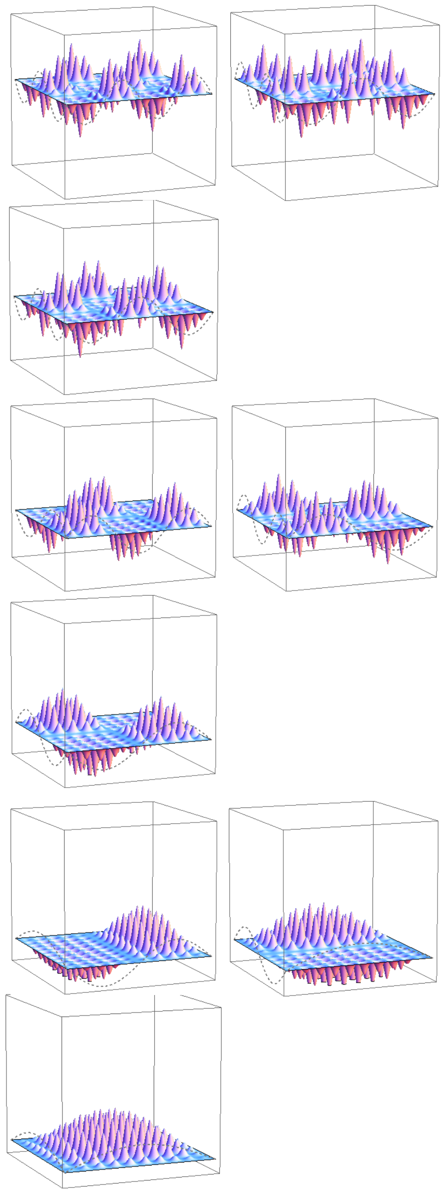

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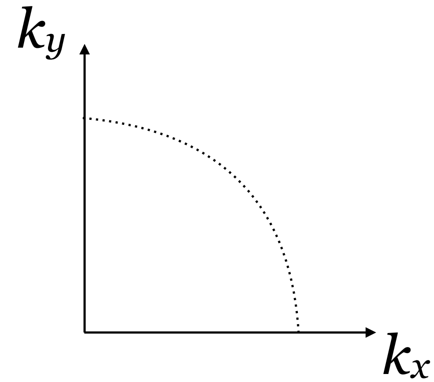

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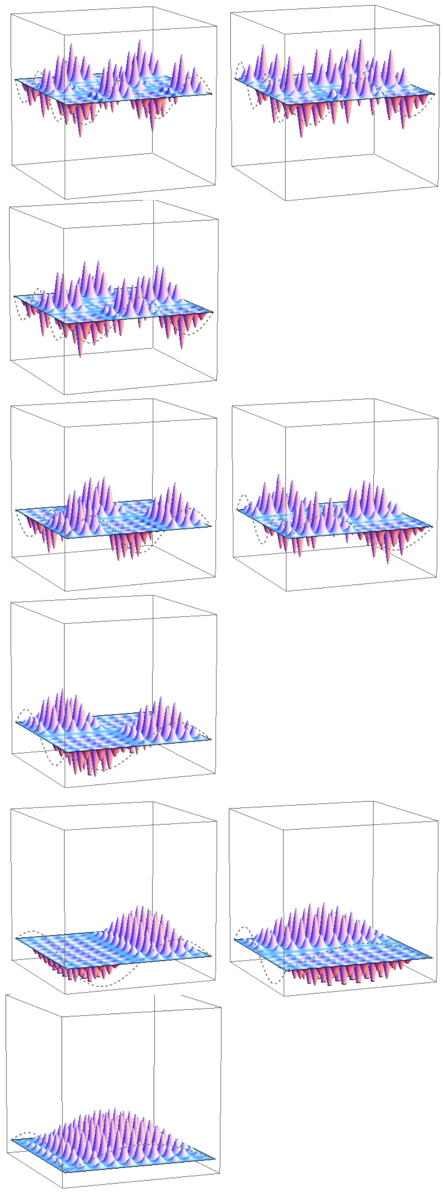

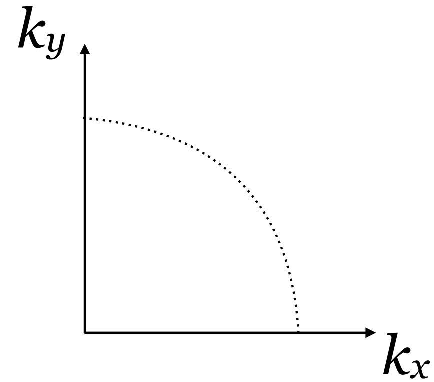

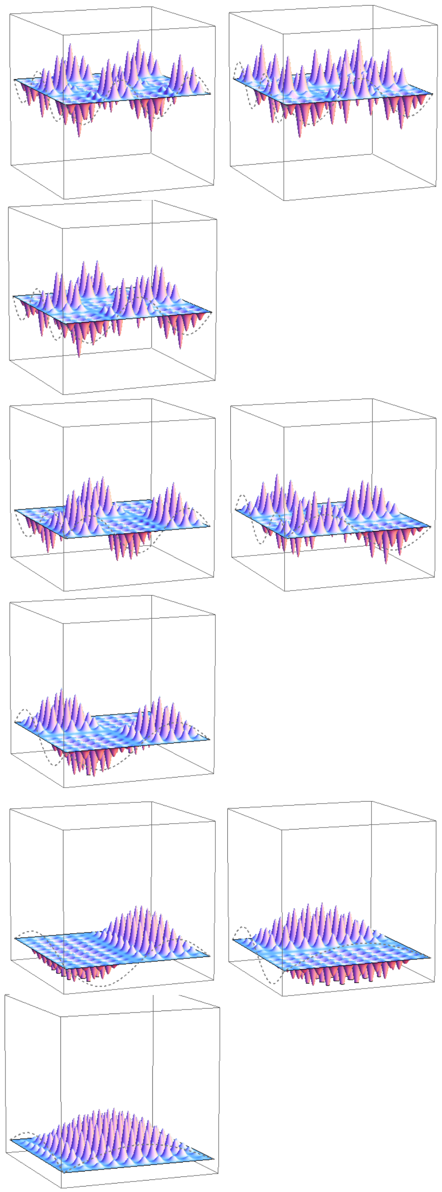

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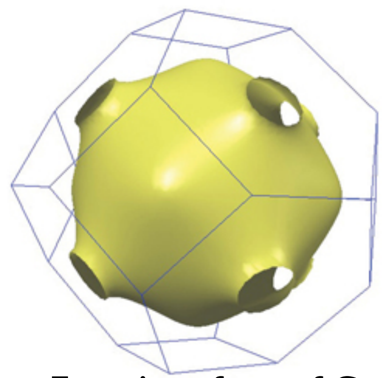
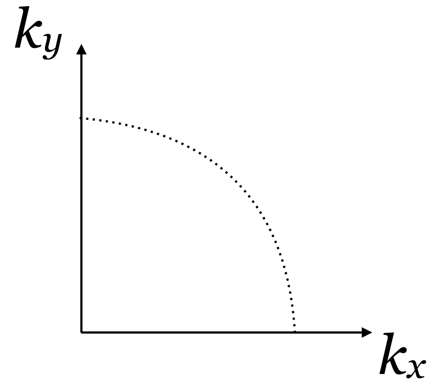


Fermi surface of Cu



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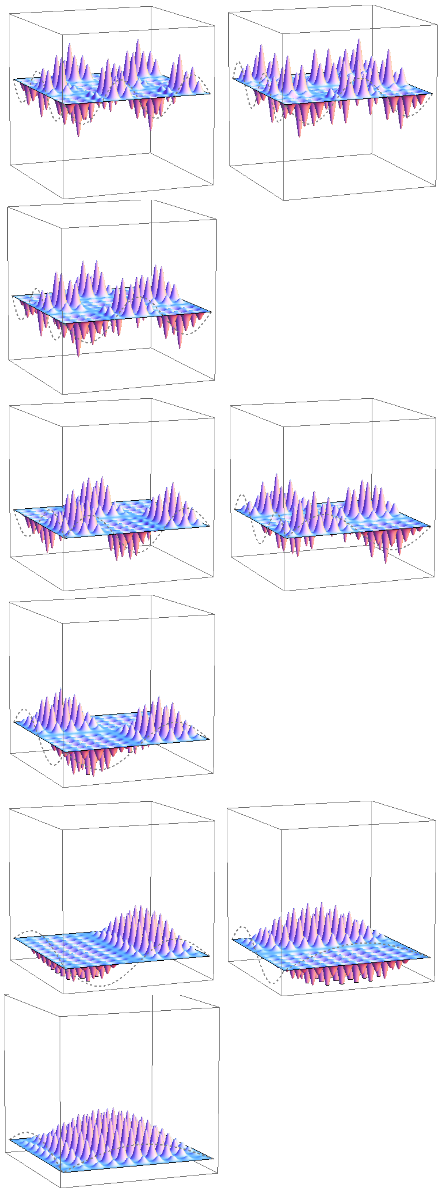

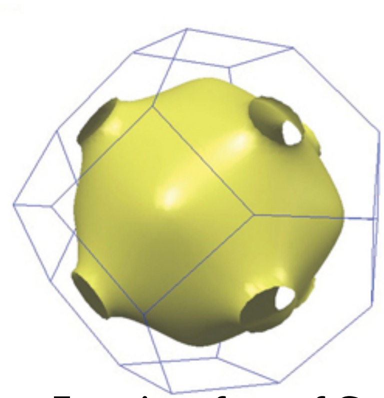

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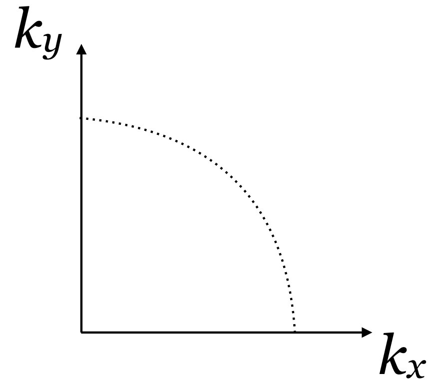

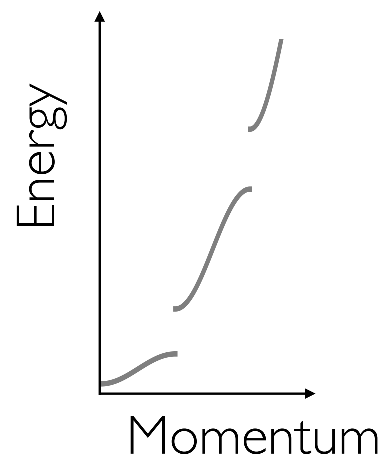

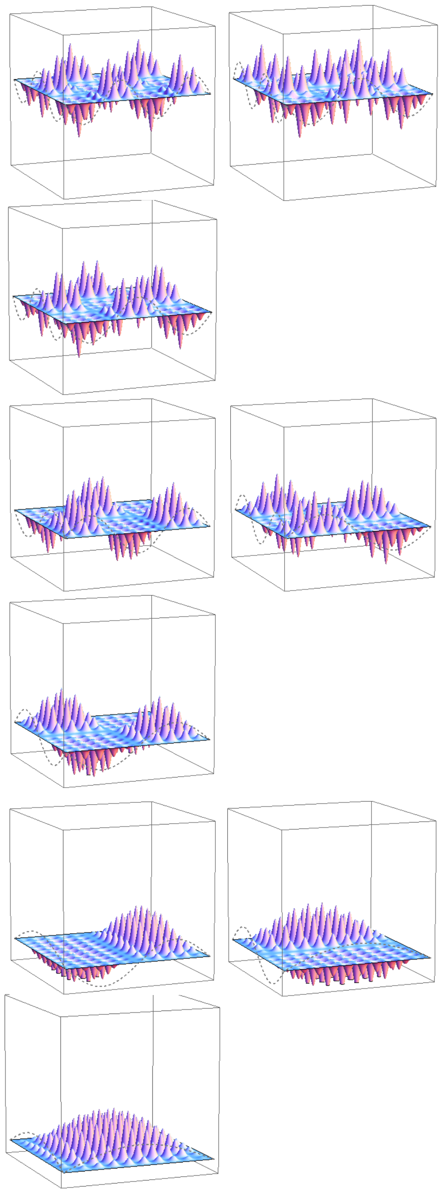

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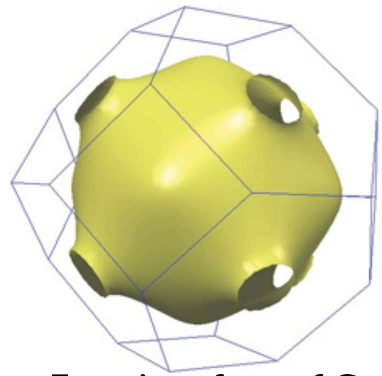

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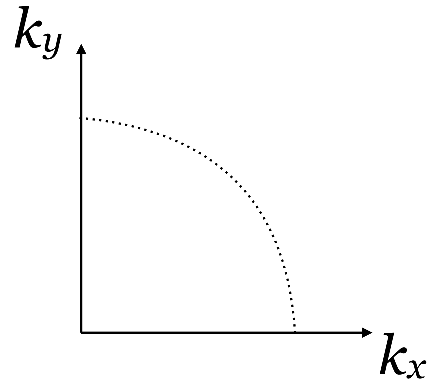

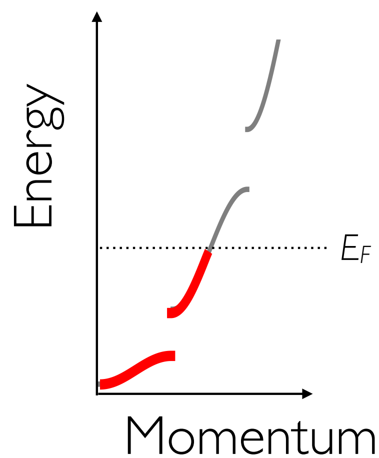

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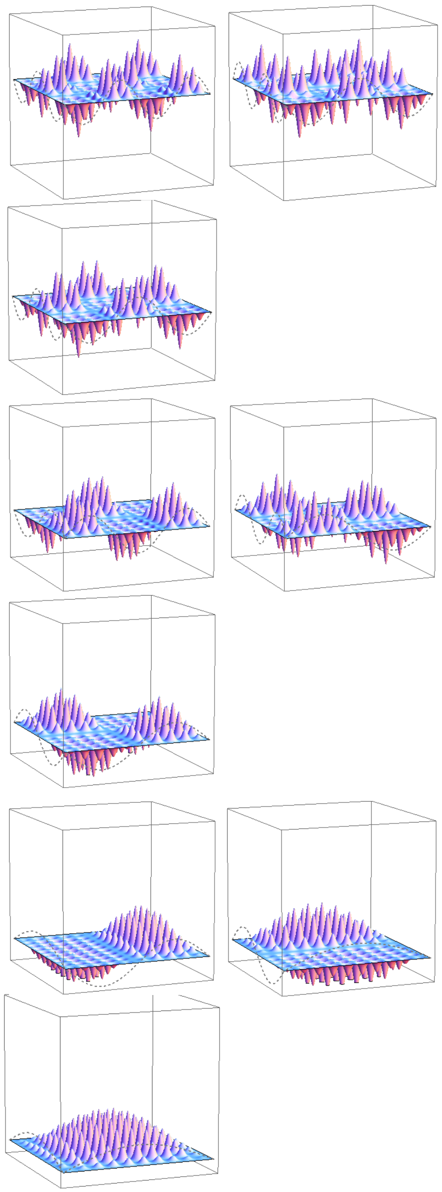

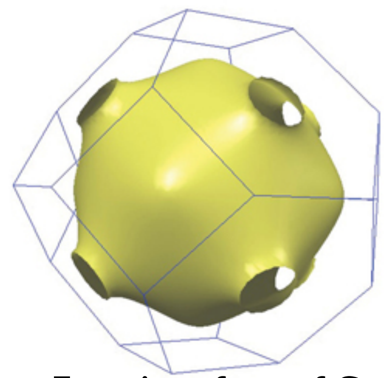

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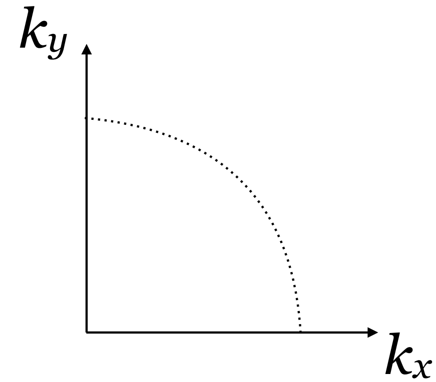
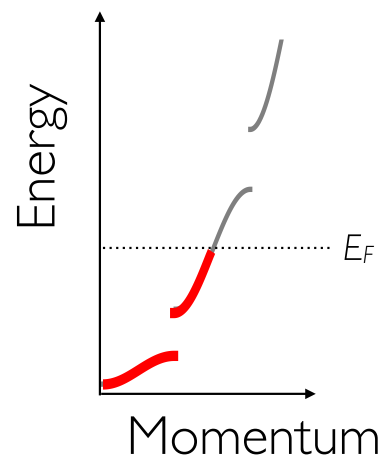
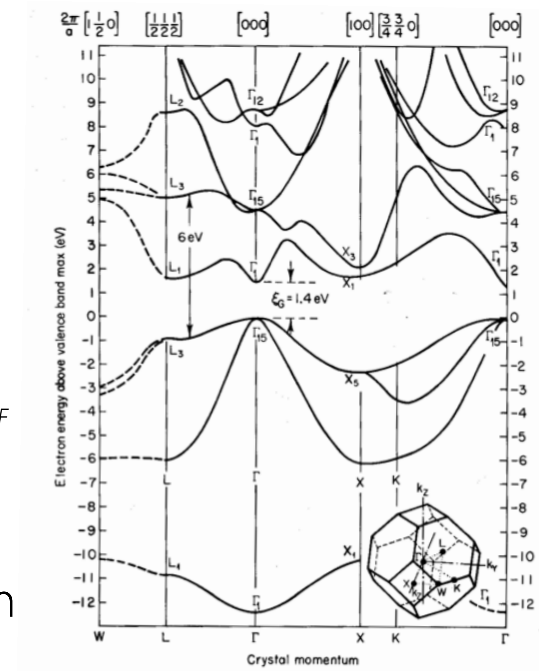




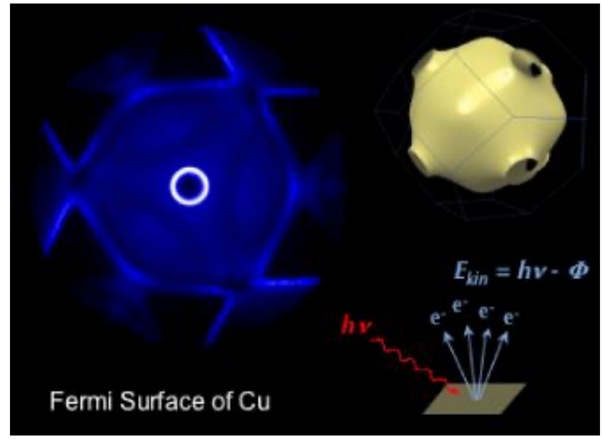


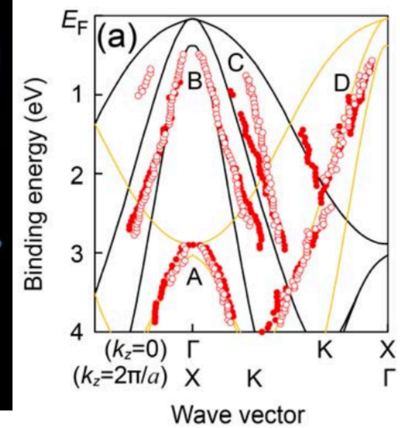
Dispersion relation of GaAs

Uncorrelated electron systems

Electron-electron interactions do not dominate material behavior

Well-established theoretical framework describes most properties of semiconductors (Si,Ge,GaAs), good metals (Cu,Ag,Al,Au)





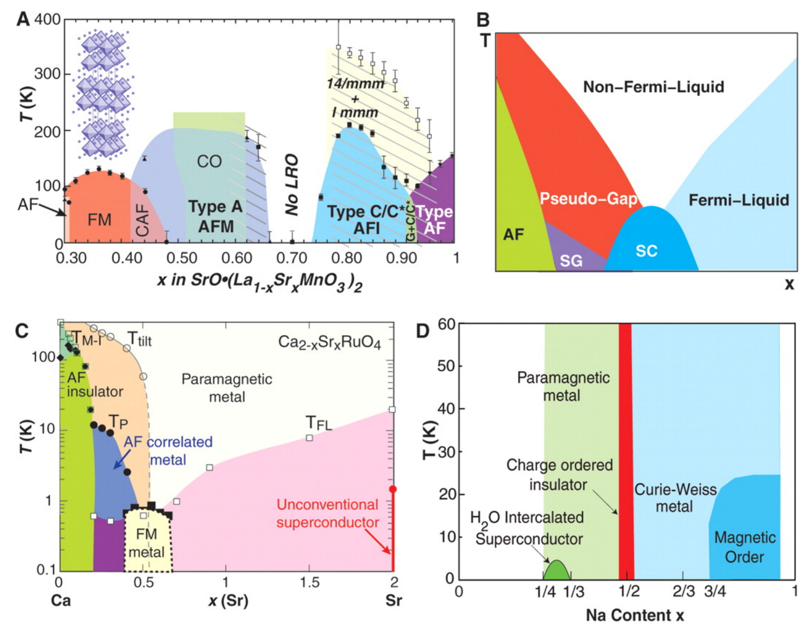
Correlated electron systems

Electron-electron interactions dominate material behavior

Often includes magnetism, more exotic behavior

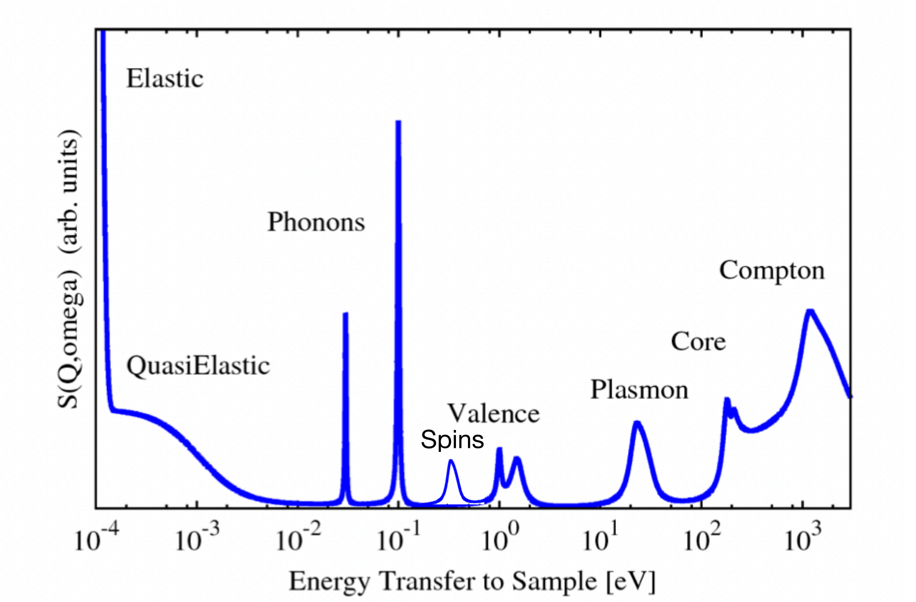
Theoretical treatment is very limited

High potential for applications and interesting basic science



Collective excitations in materials

- “Fields” of spin, charge, nuclear displacement self organize according to interactions and confining potential
- Disturbances in the pattern form excited states relevant to material behavior



Inelastic X-ray scattering can probe many different fundamental excitations

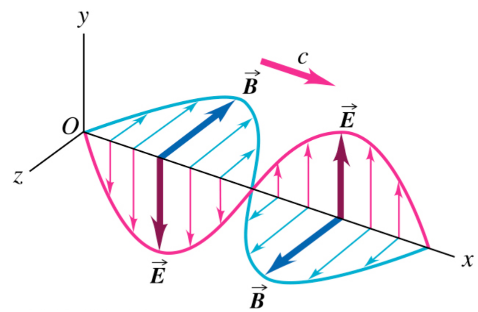
The Electromagnetic Field

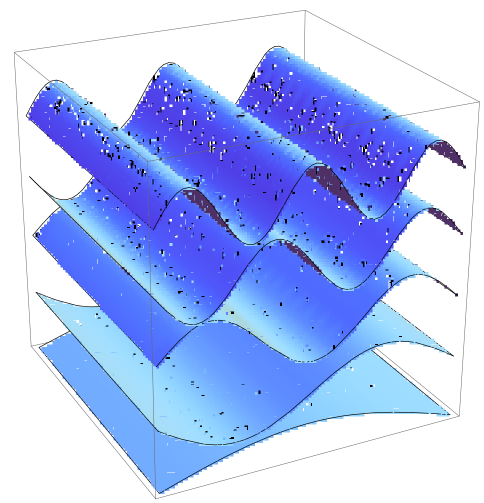
- Photons are bosons, independent, luminal
- Described by Maxwell's equations at low energy, QED at high energy
- Fully understood, quantized, modes are described as

$$|v\rangle = |n_0, \dots n_{\mathbf{k}} \dots\rangle$$

Vector potential:

$$\mathbf{A}(\mathbf{r},t) = \sum_{\mathbf{k}} \sum_{\mu=-1,1} \left(\mathbf{e}^{(\mu)}(\mathbf{k}) a_{\mathbf{k}}^{(\mu)}(t)\, e^{i\mathbf{k}\cdot\mathbf{r}} + \bar{\mathbf{e}}^{(\mu)}(\mathbf{k}) \bar{a}_{\mathbf{k}}^{(\mu)}(t)\, e^{-i\mathbf{k}\cdot\mathbf{r}} \right)$$



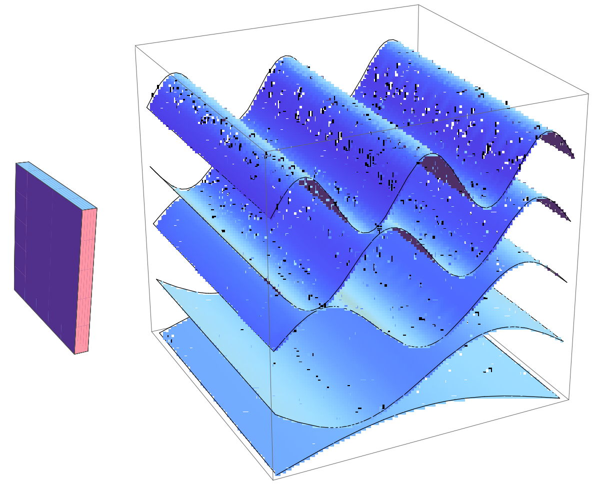


Spectroscopy, general remarks

- Sample can exchange energy, momentum with EM field

$$|\psi_g\rangle \otimes |v\rangle \rightarrow |\psi_e\rangle \otimes |v'\rangle$$

(sample) (vacuum)



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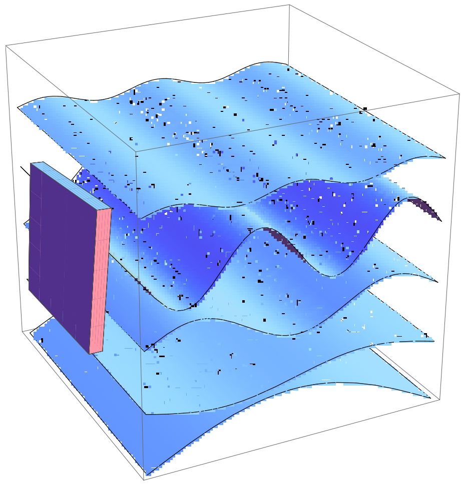

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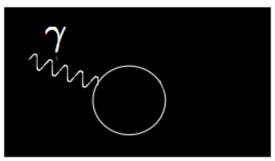
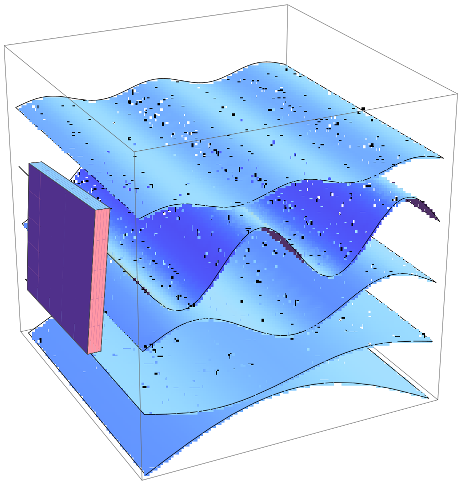


One photon
Optical spectroscopy
X-ray absorption

$$|n_0, ...n_{\mathbf{k}}...\rangle \rightarrow |n_0, ...n_{\mathbf{k}} - 1...\rangle$$



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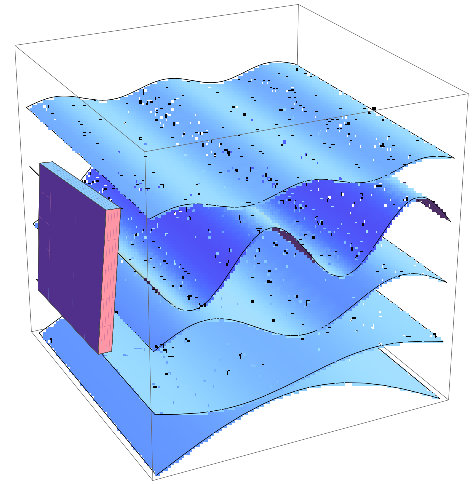

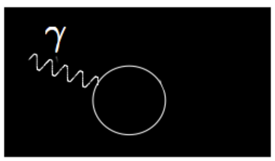

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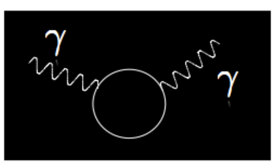

Two photon
Raman
Inelastic X-ray Scattering

$$|n_0, ...n_{\mathbf{k}}...\rangle \rightarrow |n_0, ...n_{\mathbf{k}} - 1...n_{\mathbf{k'}} + 1...\rangle$$

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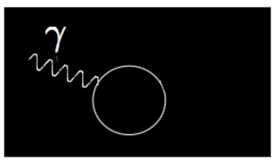

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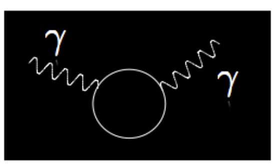

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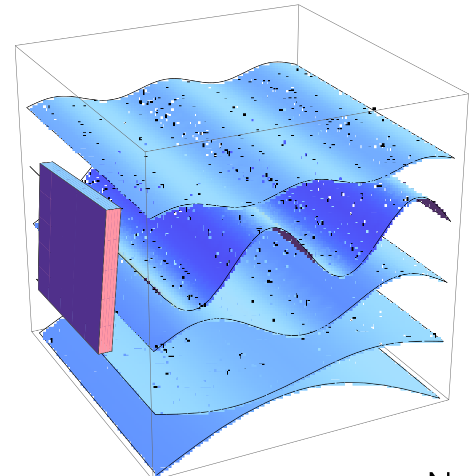
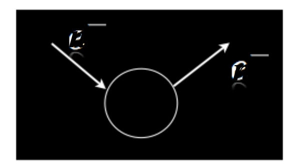
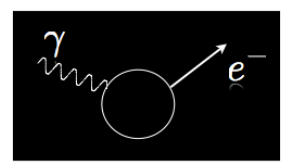


Related spectroscopies:

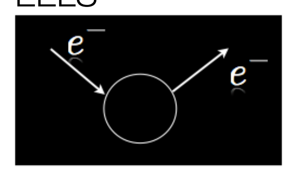
Neutron



ARPES



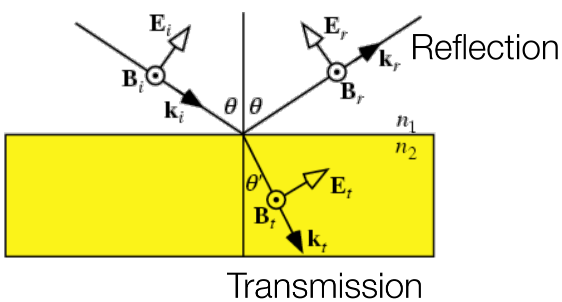
EELS

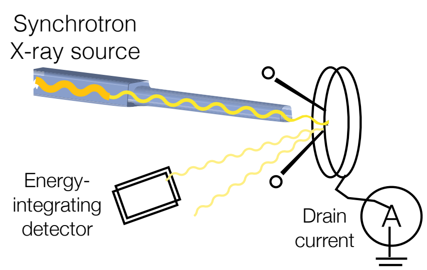


Photon spectroscopies

One-photon

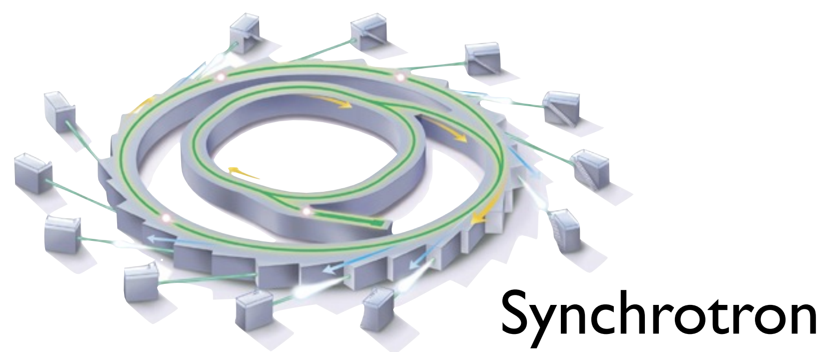
- Can measure from reflection or transmission in time or frequency domain
- Use Fresnel's equations to determine $n + ik$, . $\sigma_1 - i\sigma_2, \epsilon_1 + i\epsilon_2$ and relate to fundamental behavior
- X-ray absorption collected differently, through total electron or total fluorescence yield





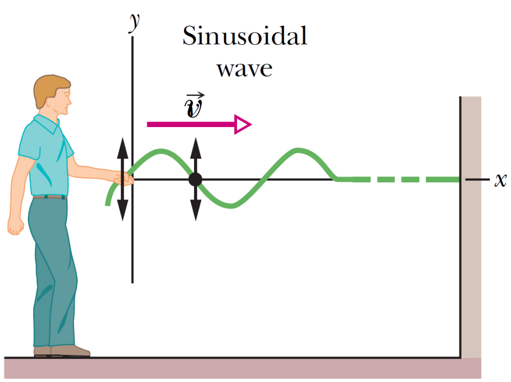
Two-photon

- IXS and RIXS (also REXS, Raman)
- Probes electronic excitations in a momentum-resolved way
- Technical advances making rapid progress



Elastic waves

- Materials can distort in wavelike patterns, typical of transverse waves on a string



A common wave equation: $$\frac{\partial^2 f}{\partial x^2} = \frac{1}{v^2}\frac{\partial^2 f}{\partial t^2}$$

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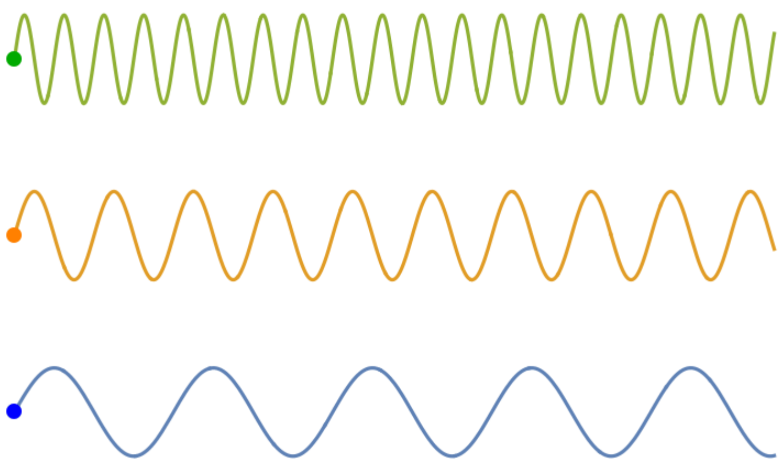
All waves move at the same speed, regardless of wavelength



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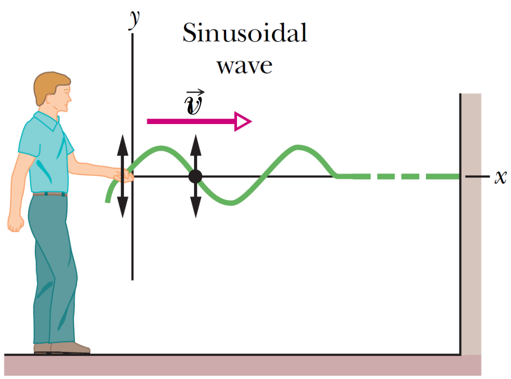

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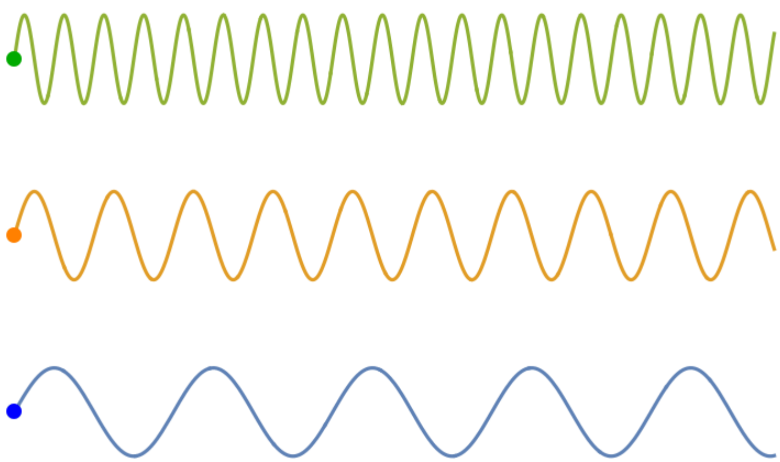

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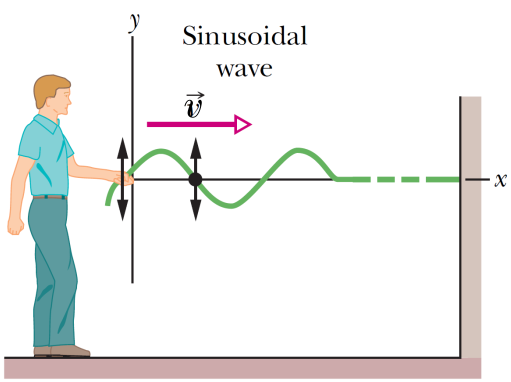

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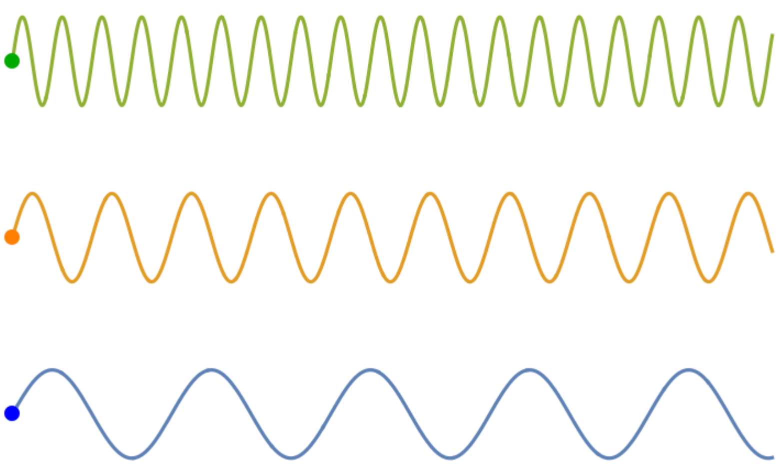

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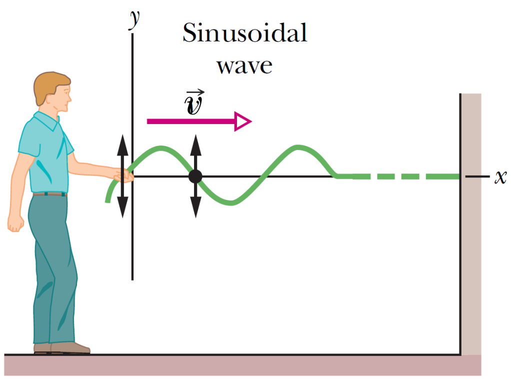

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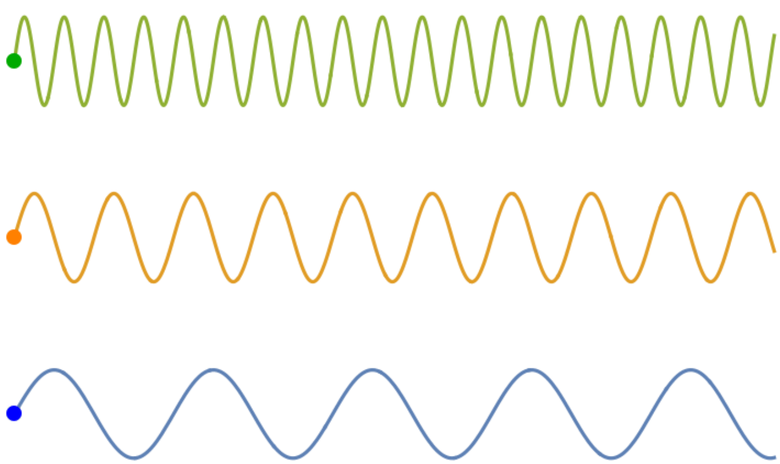

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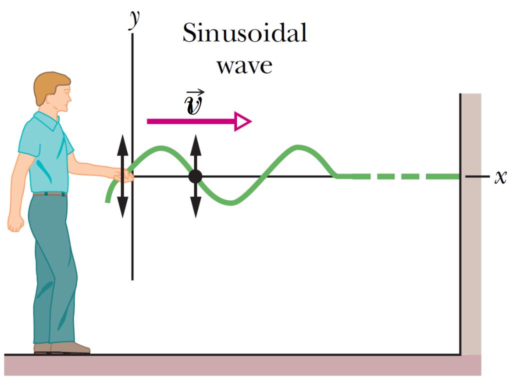

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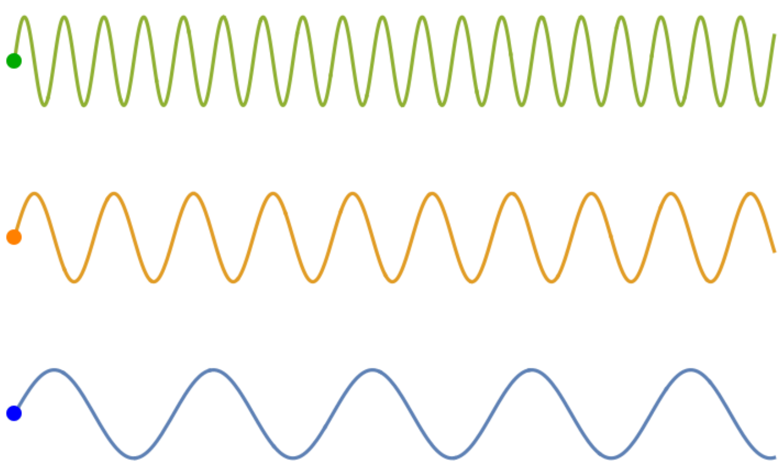

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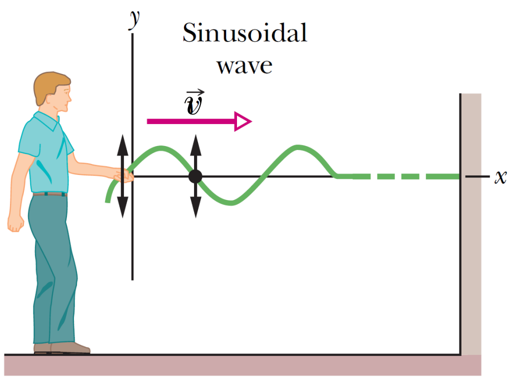

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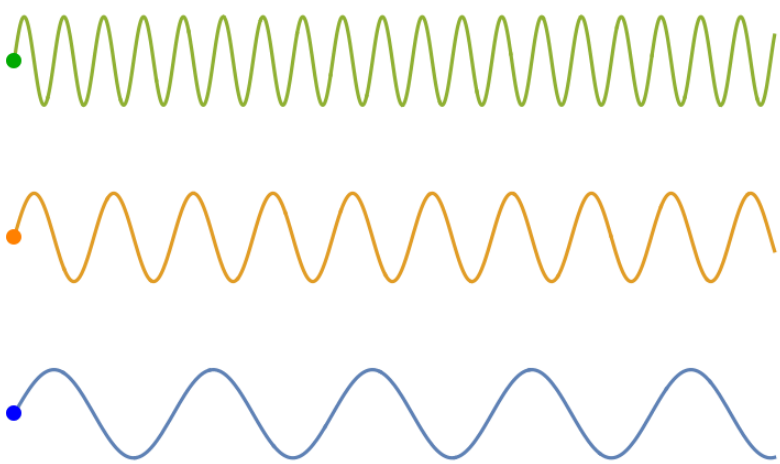
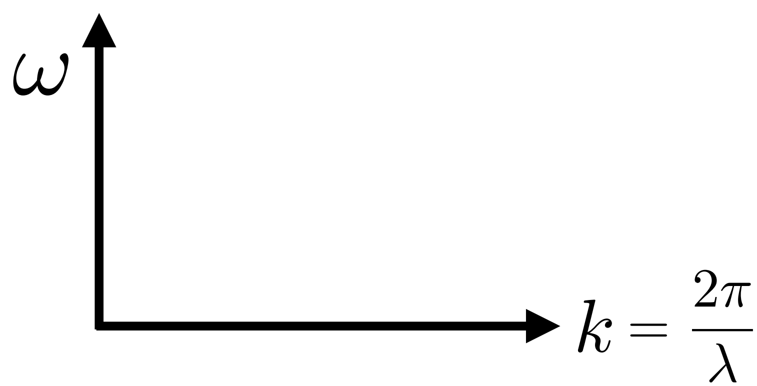


Dispersion relation for waves on a string



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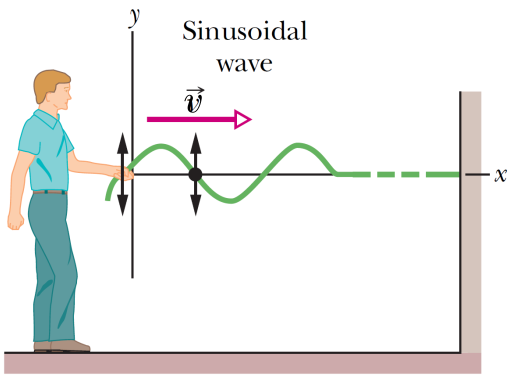

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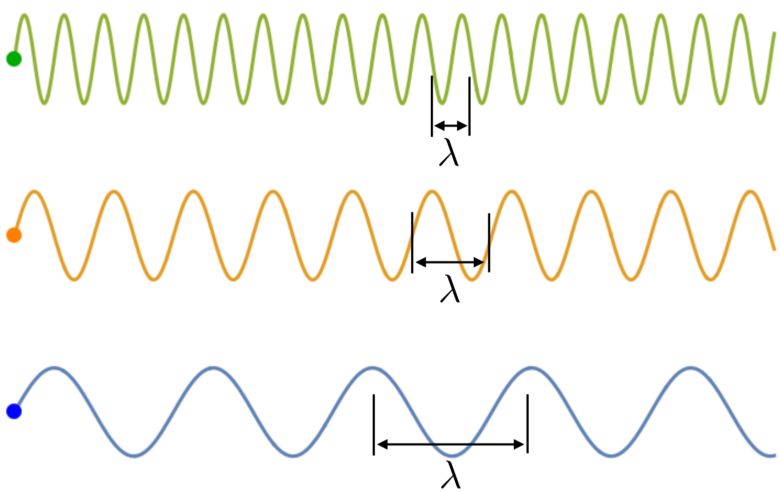

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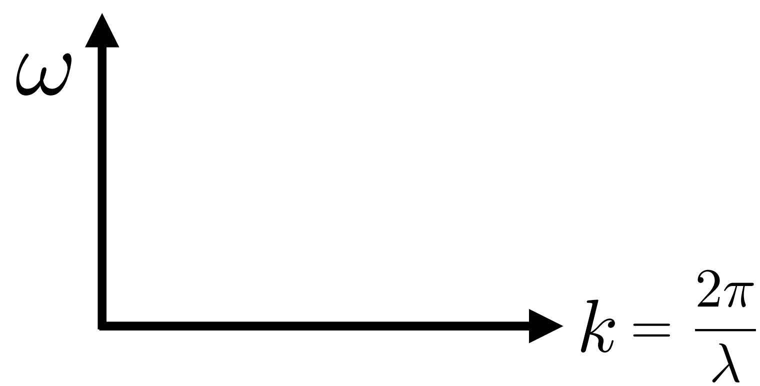

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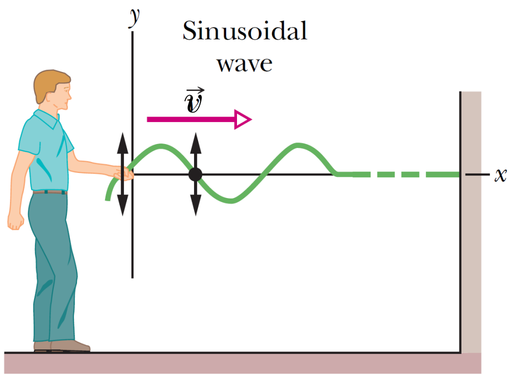

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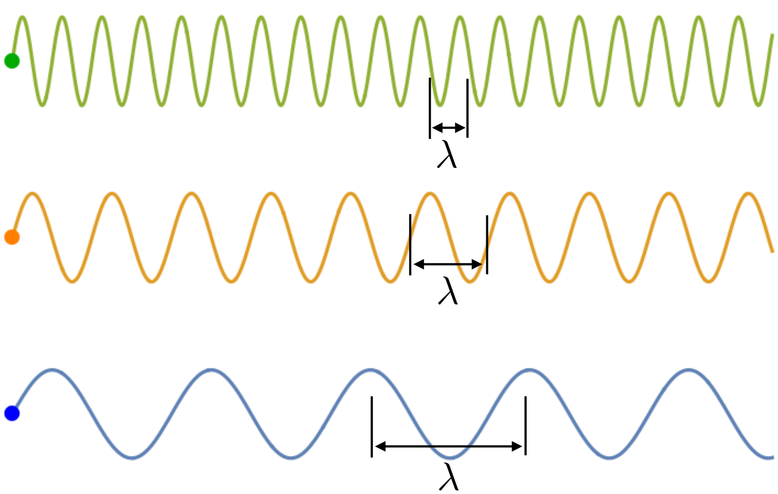

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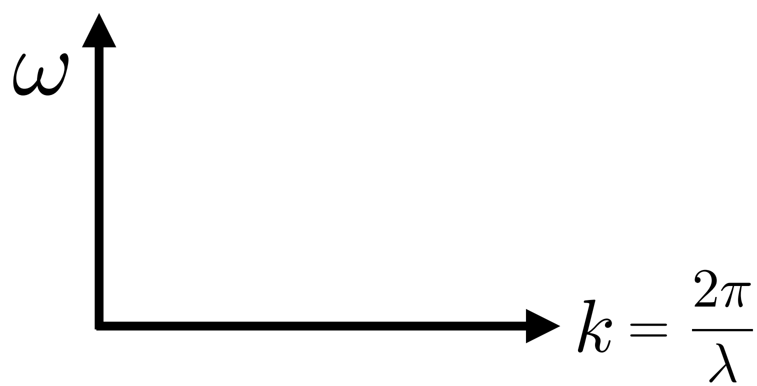

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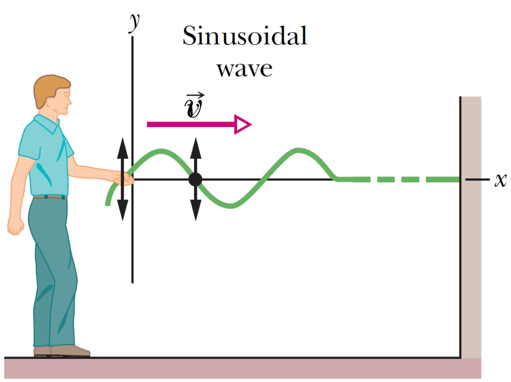

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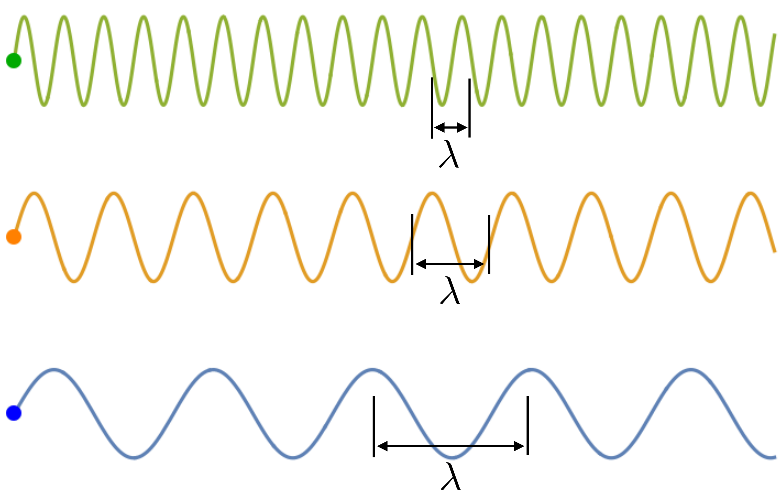

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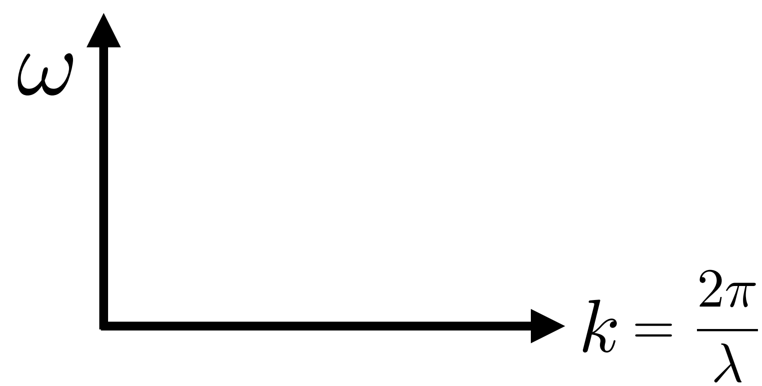

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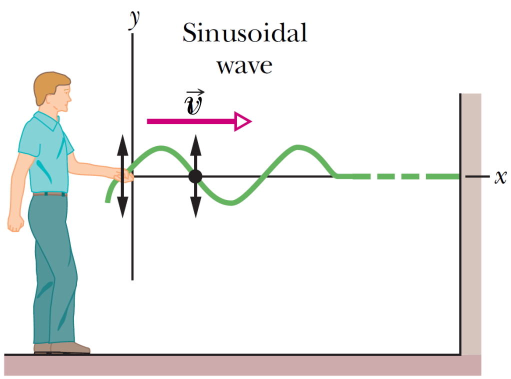

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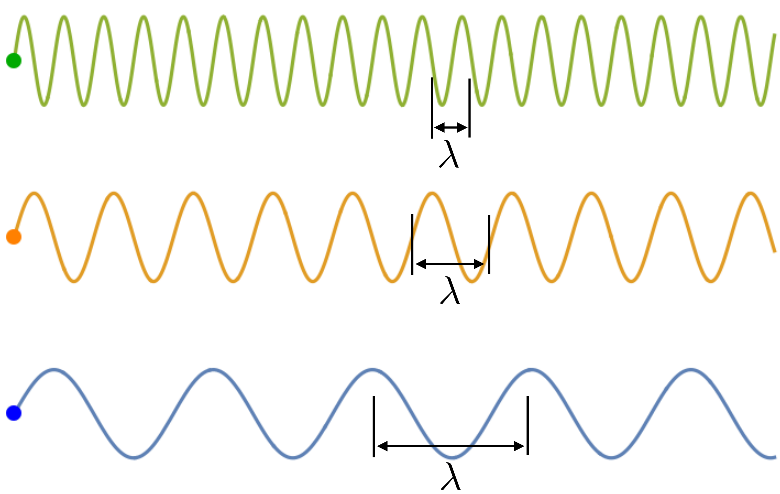

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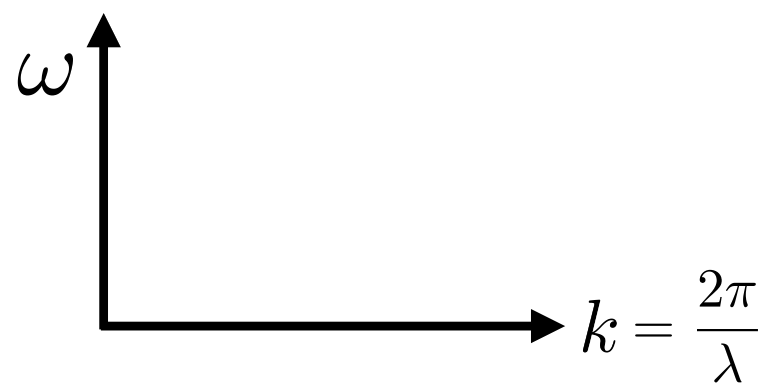

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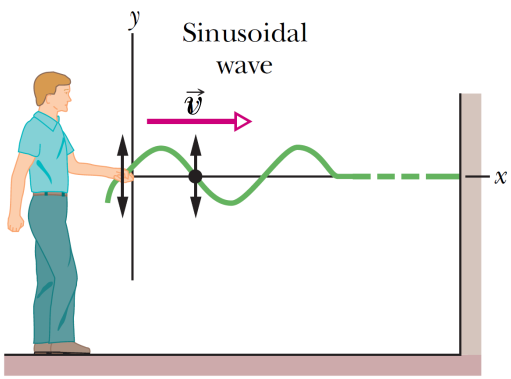

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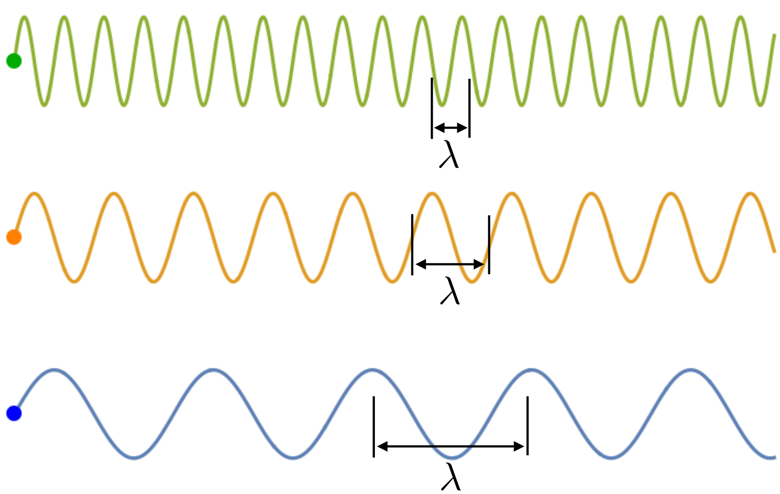

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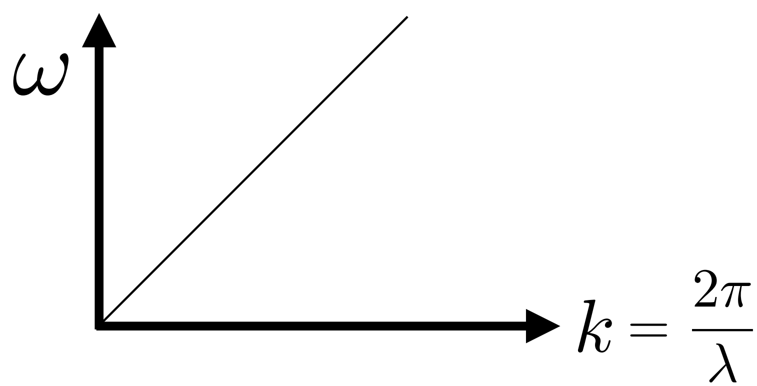

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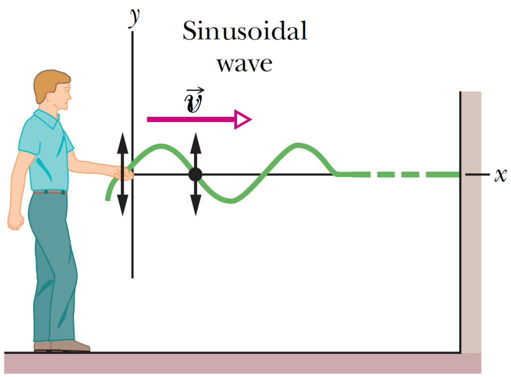

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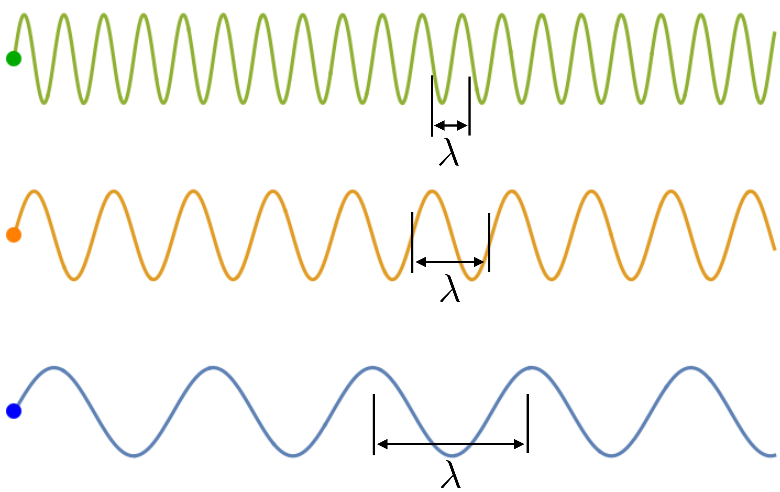

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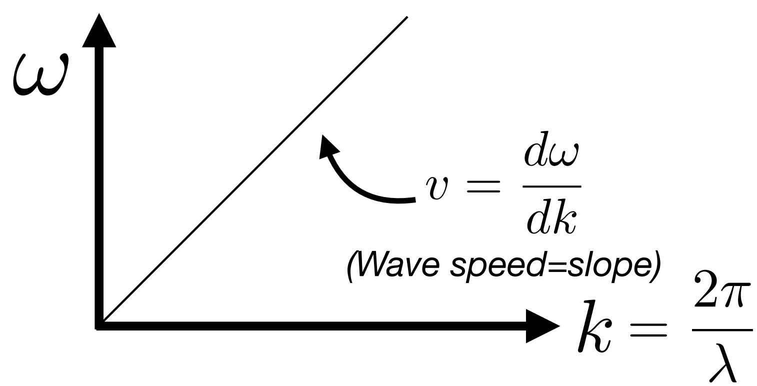

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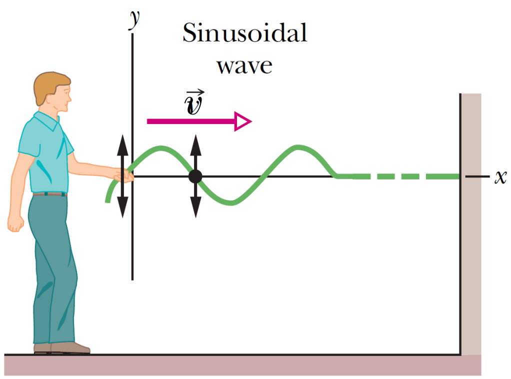

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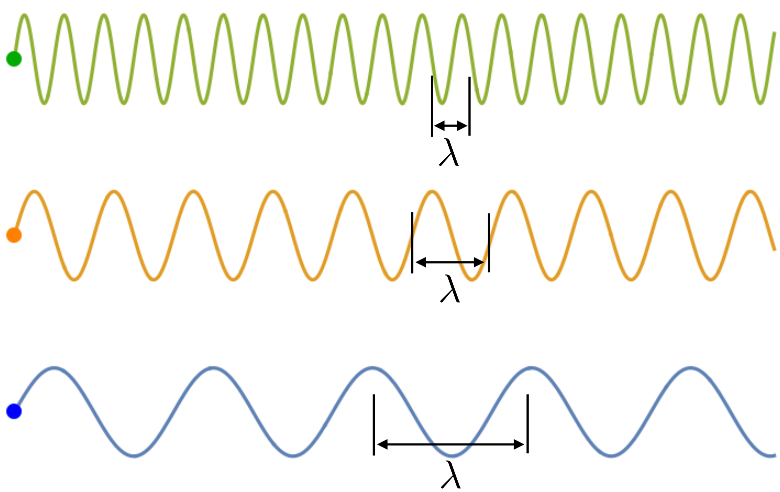

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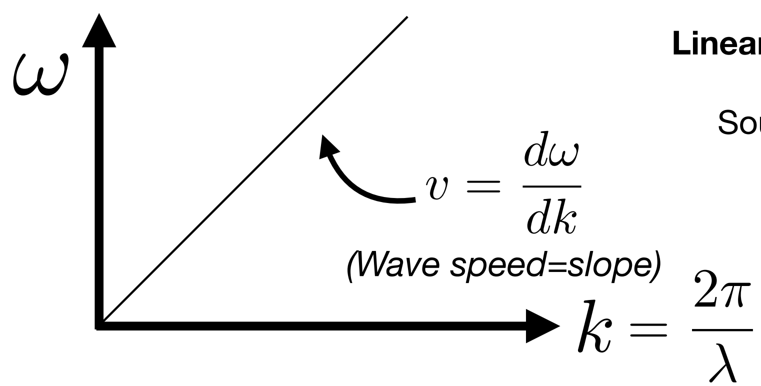

Linear dispersion also found in:
X-rays and light
Sound at long wavelengths
Shallow water waves
Gravitational waves

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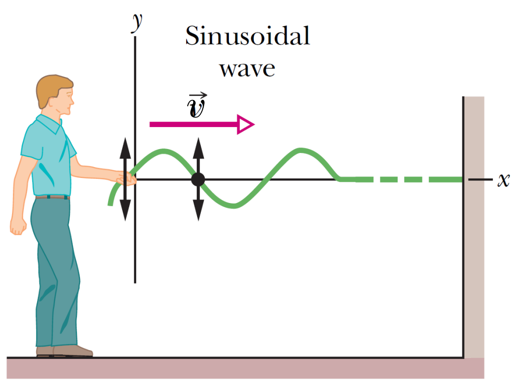

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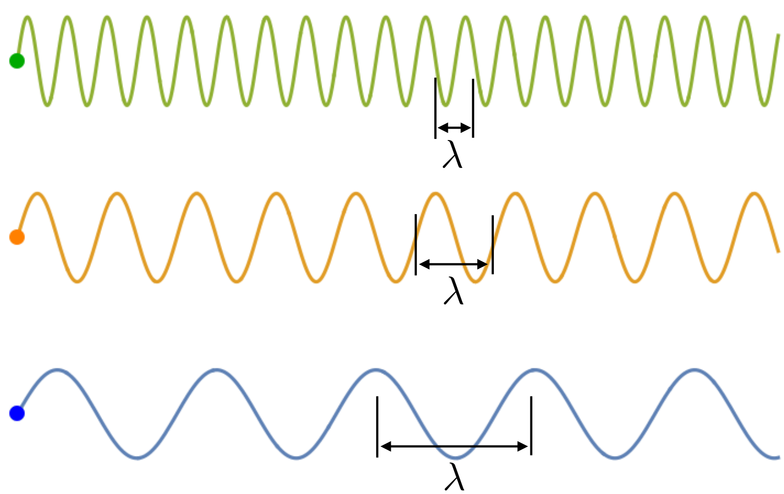
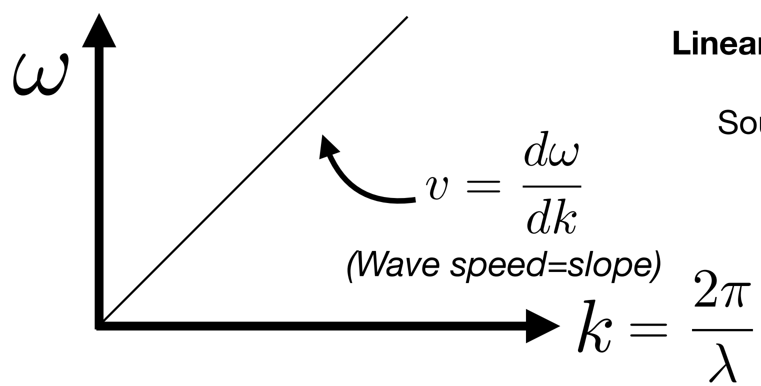


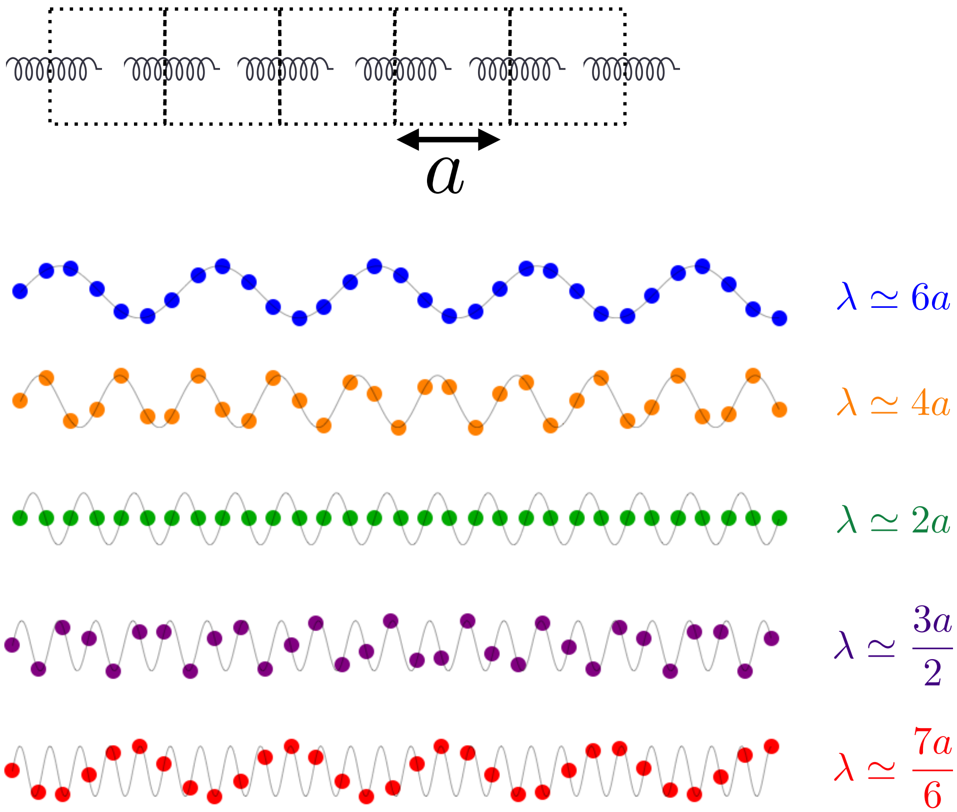
Dispersion relation for waves on a string

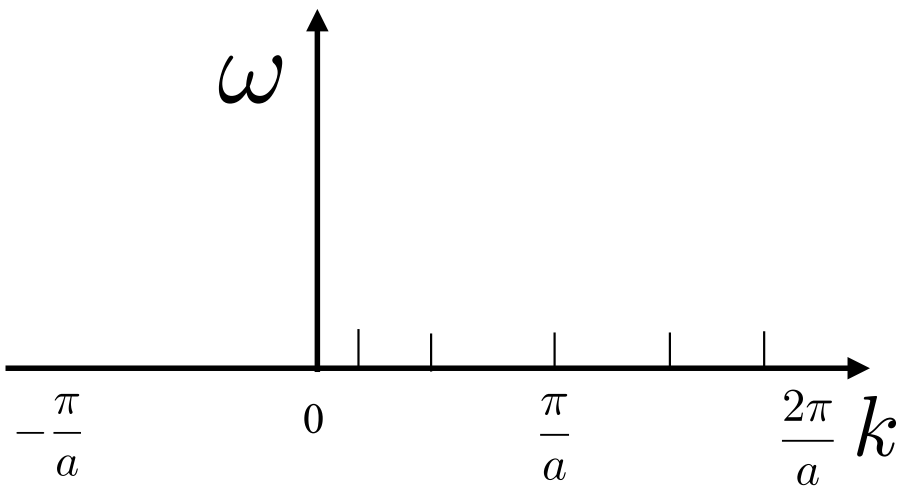


Linear dispersion also found in:
X-rays and light
Sound at long wavelengths
Shallow water waves
Gravitational waves

What about atomic scale disturbances?

- Consider *mona*tomic 1D chain and wavelengths approaching lattice parameter a





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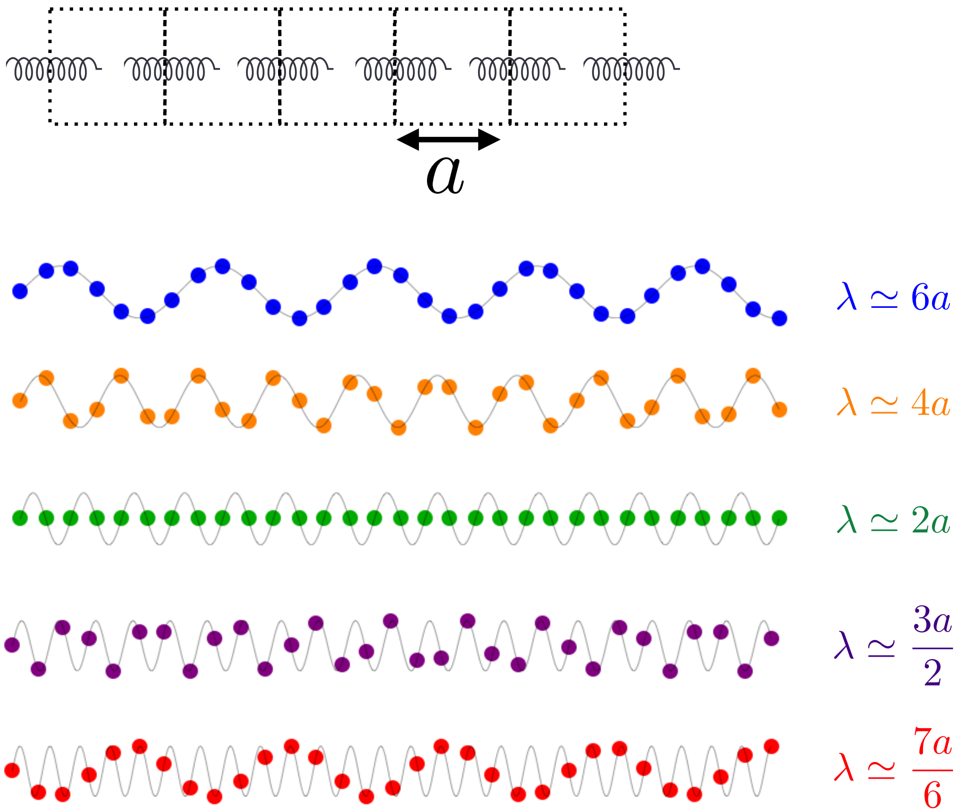

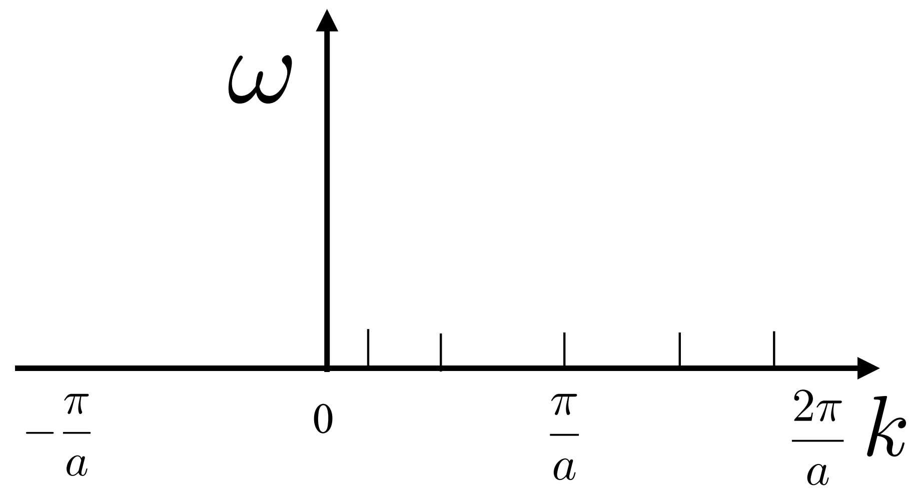

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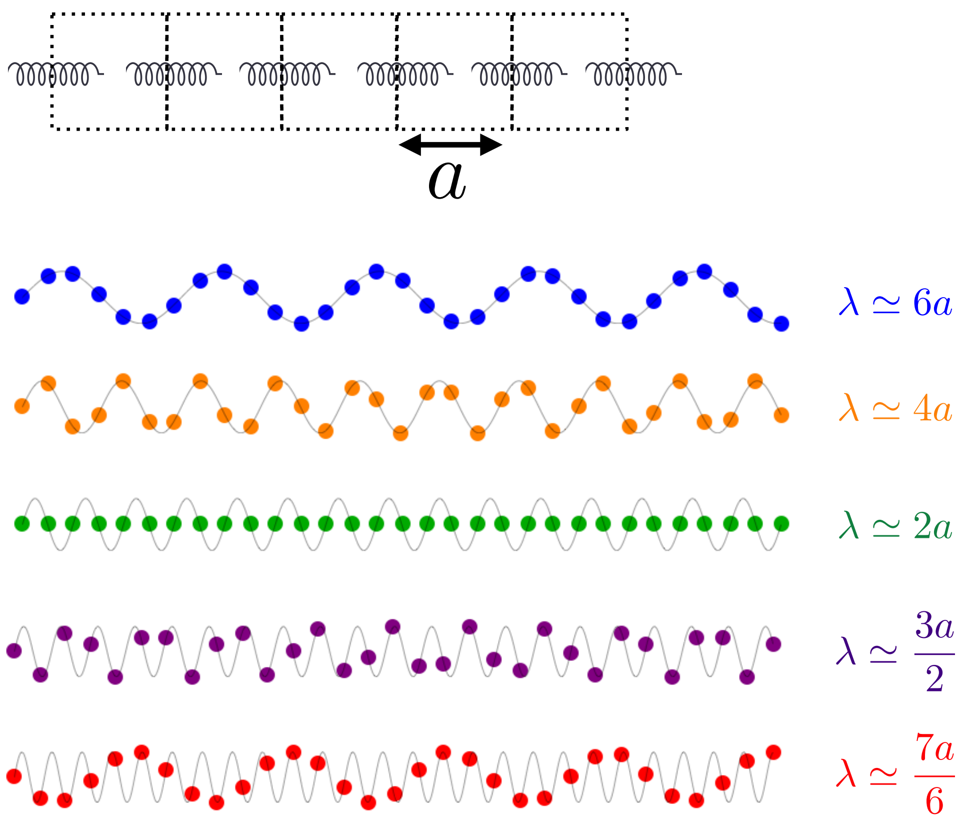

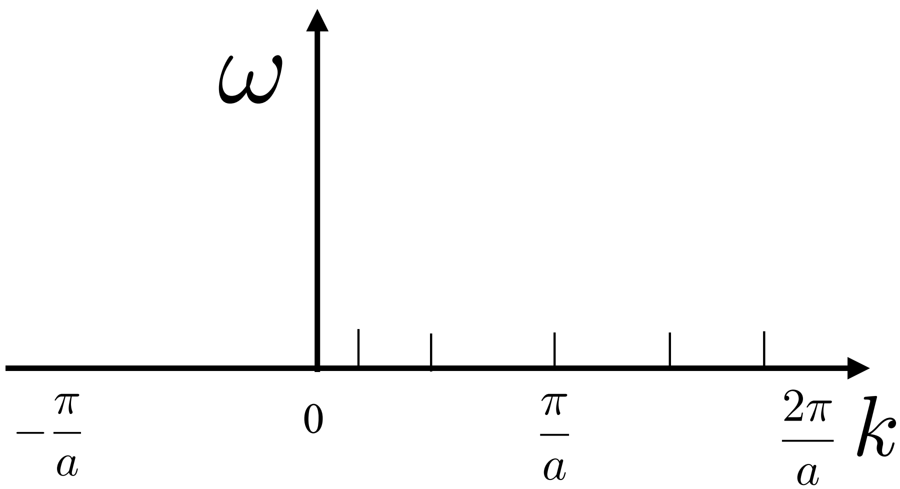

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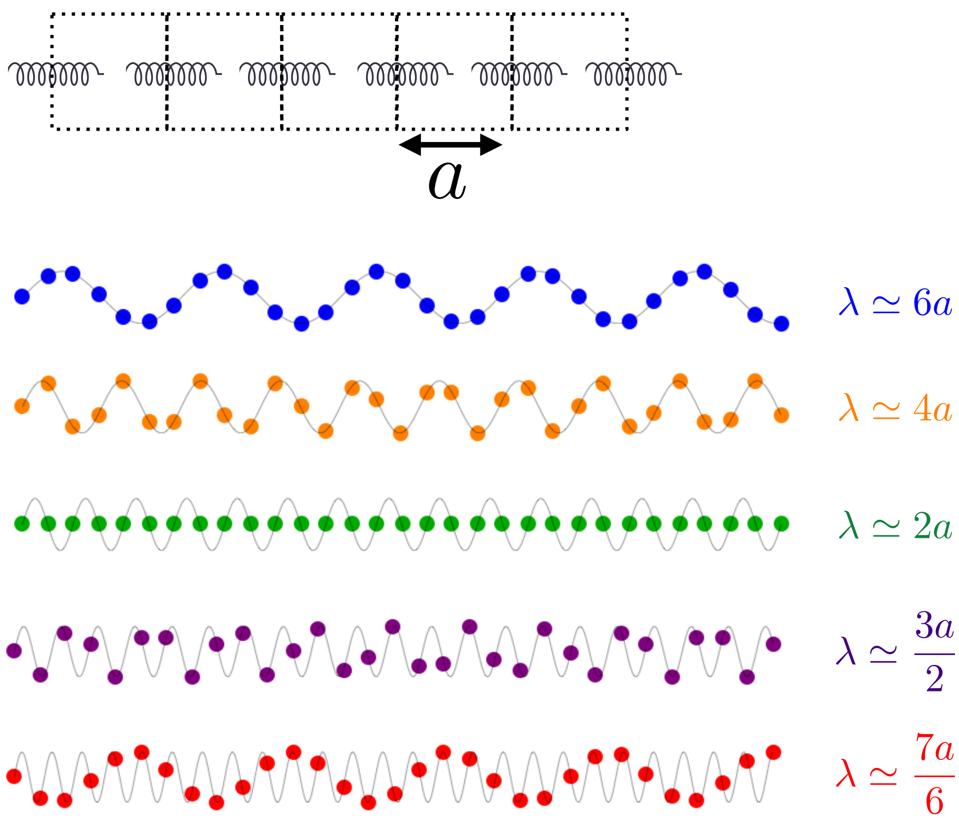

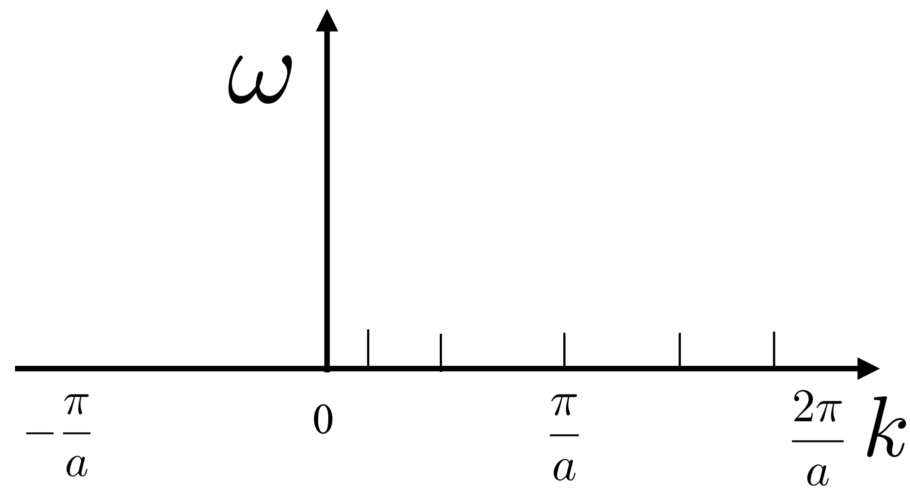

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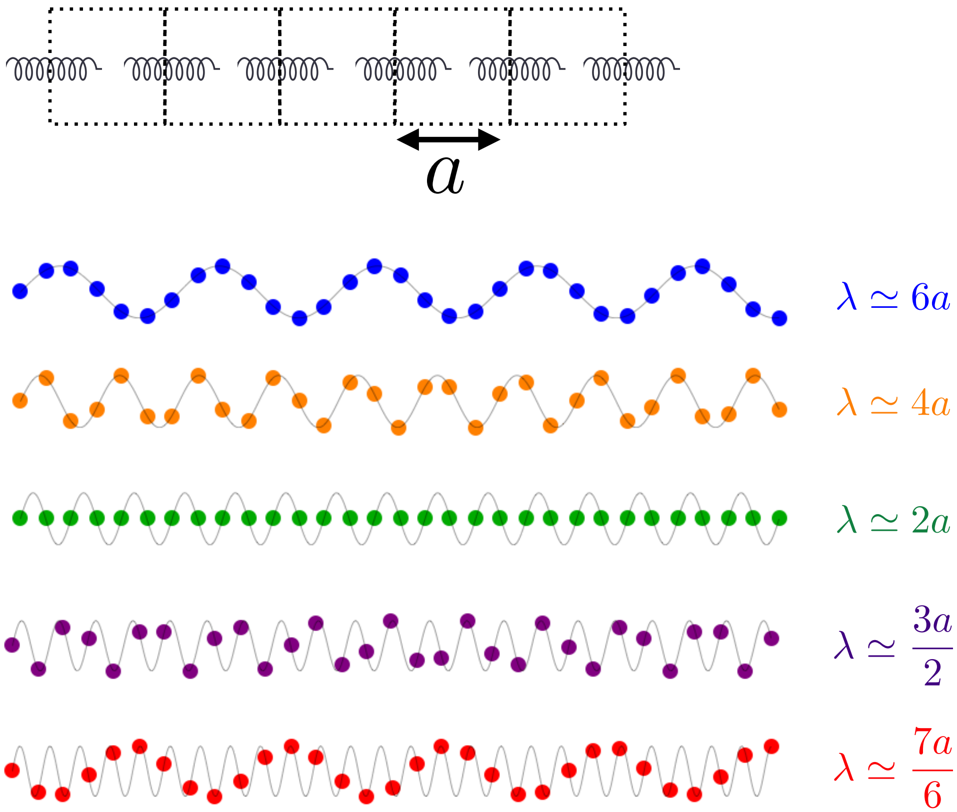

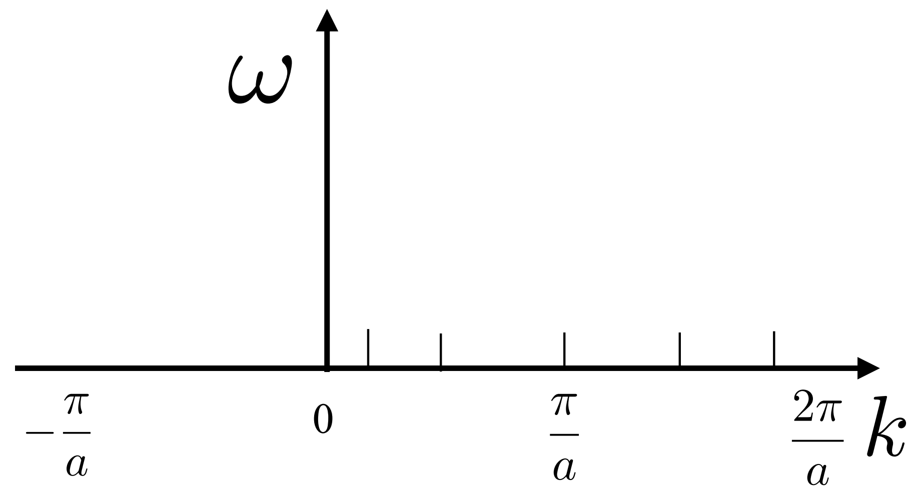

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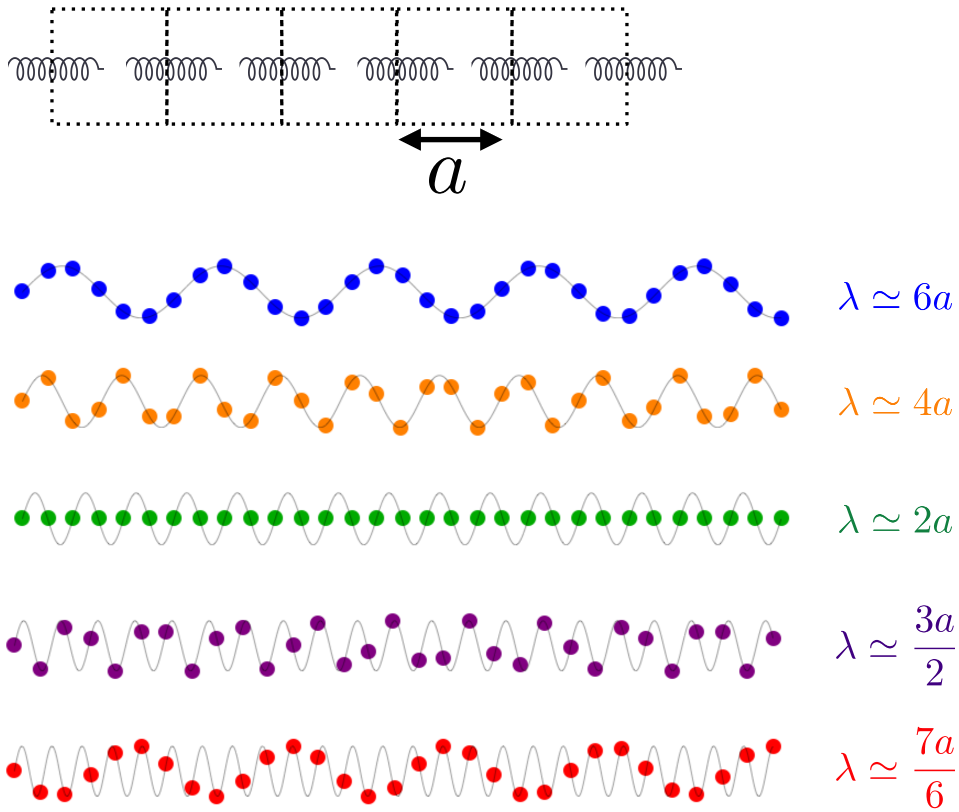

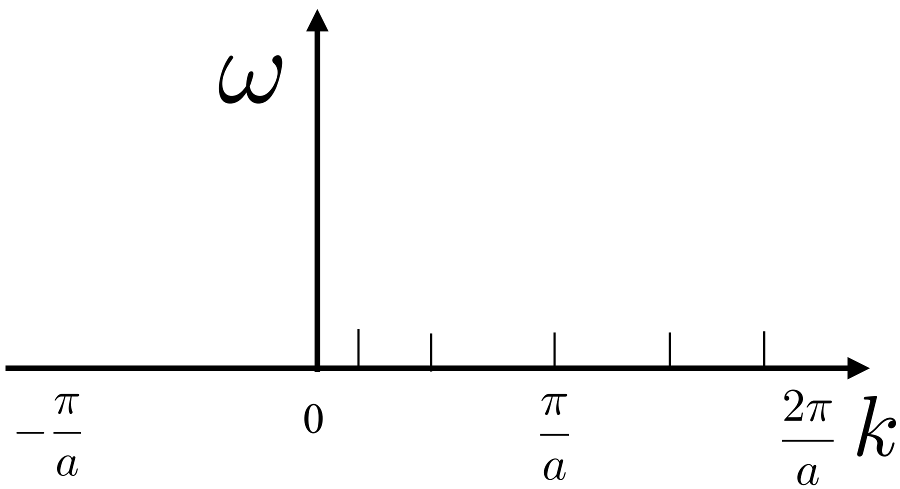

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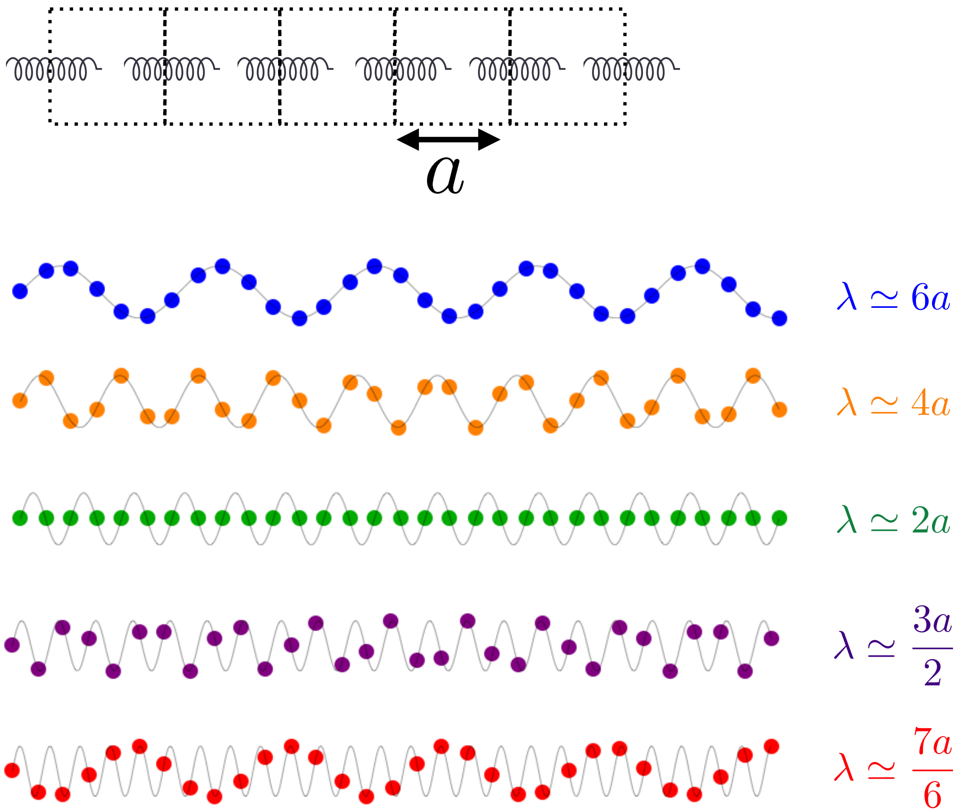

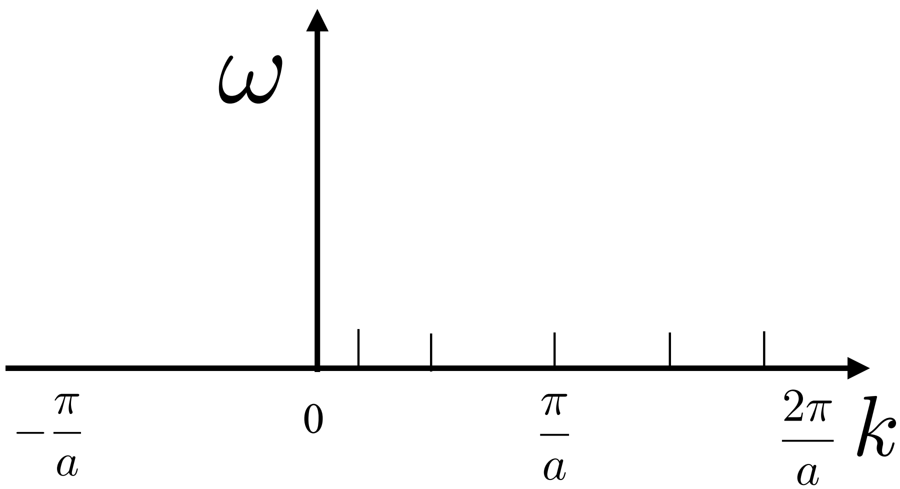

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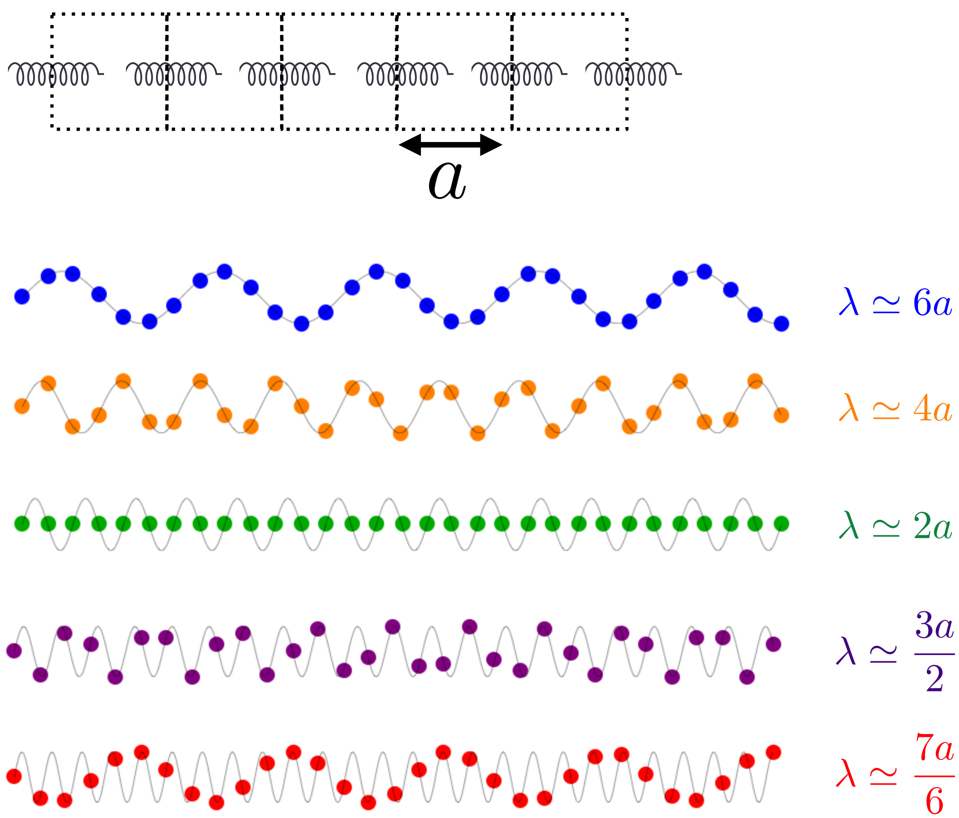

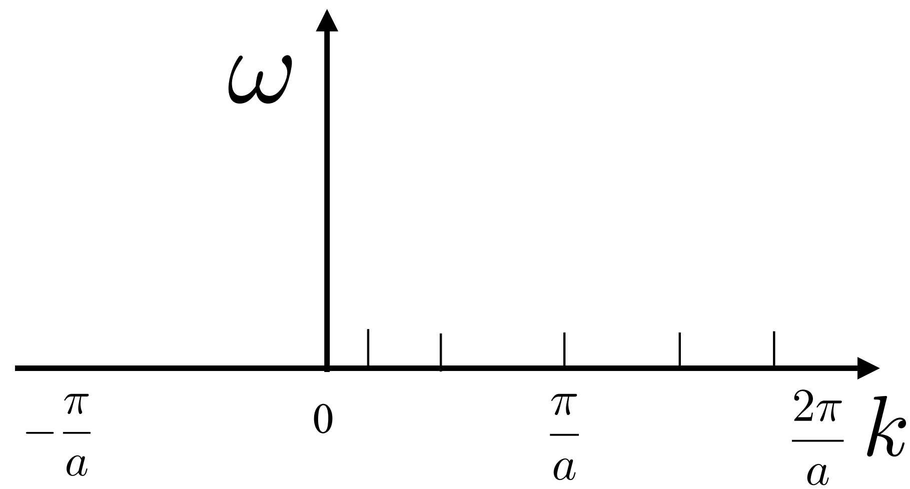

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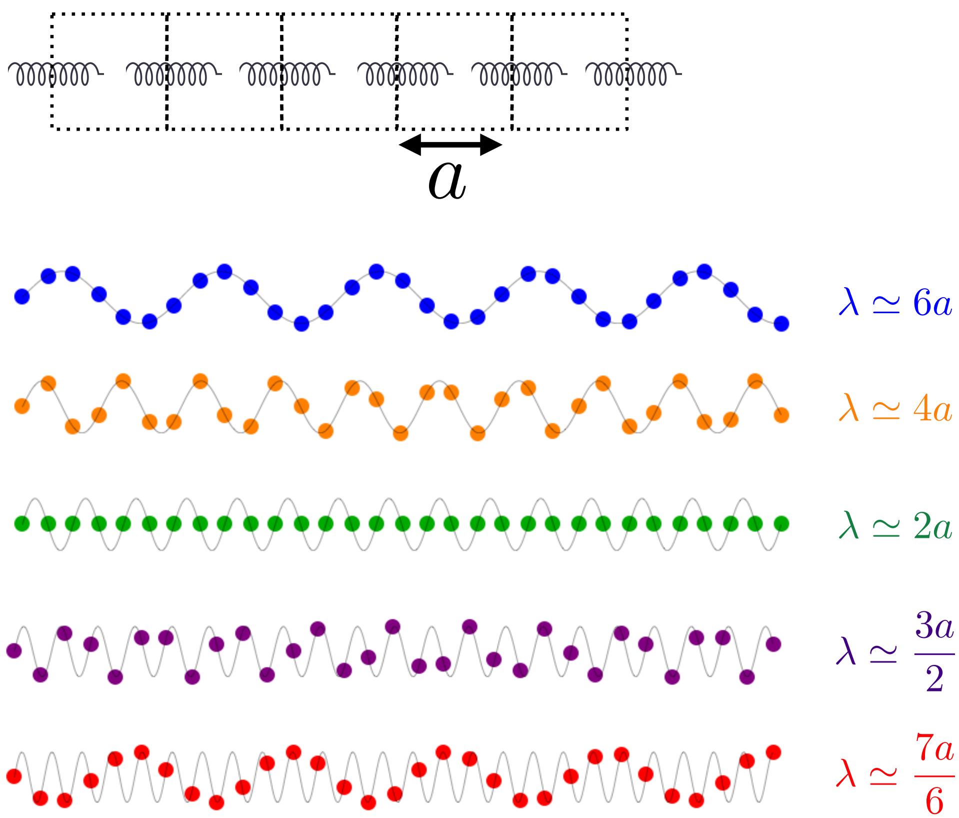

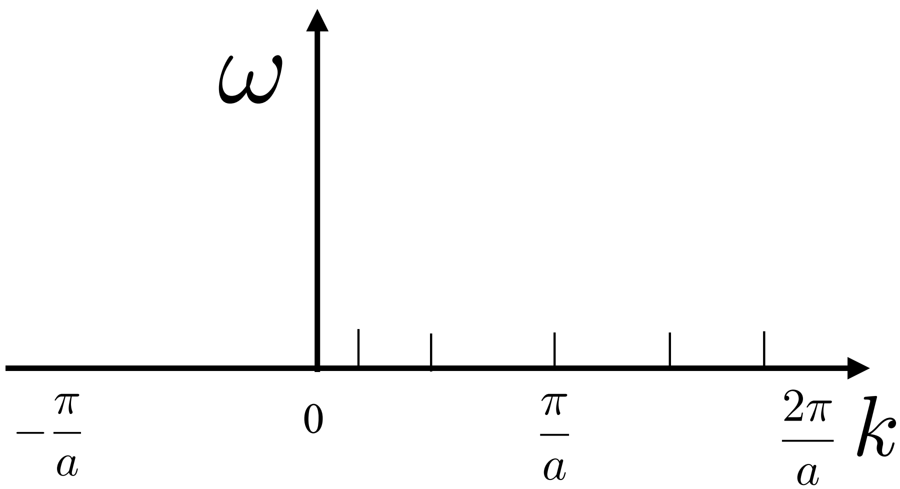

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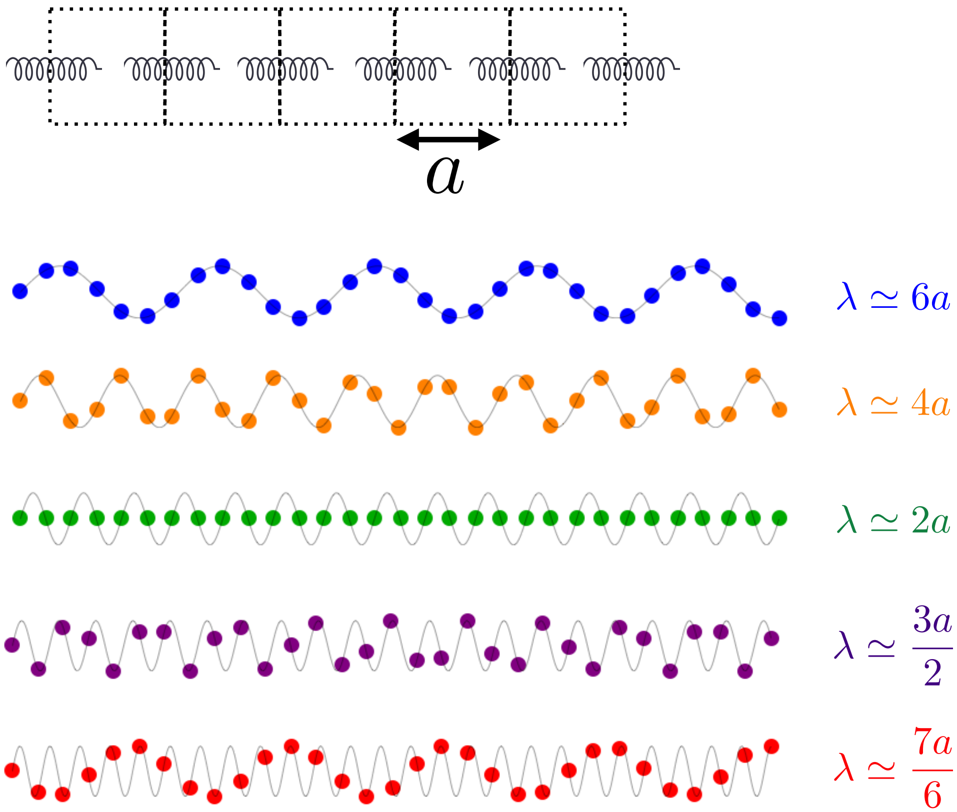

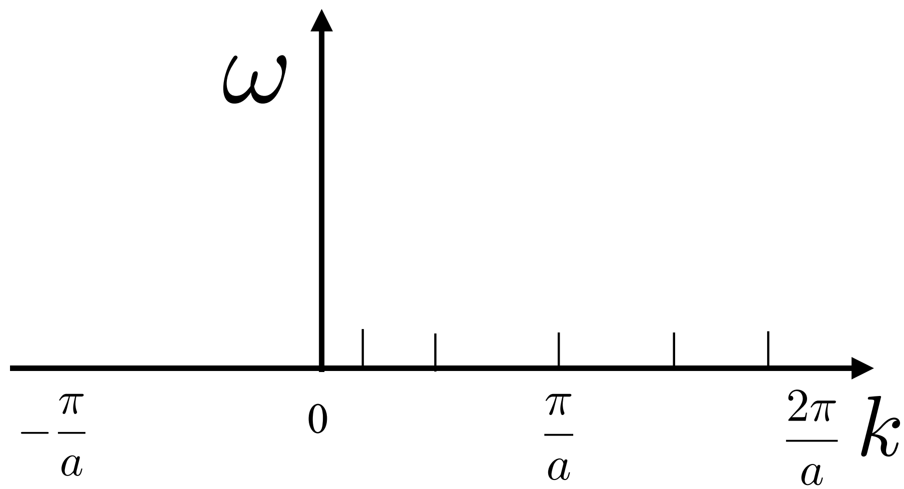

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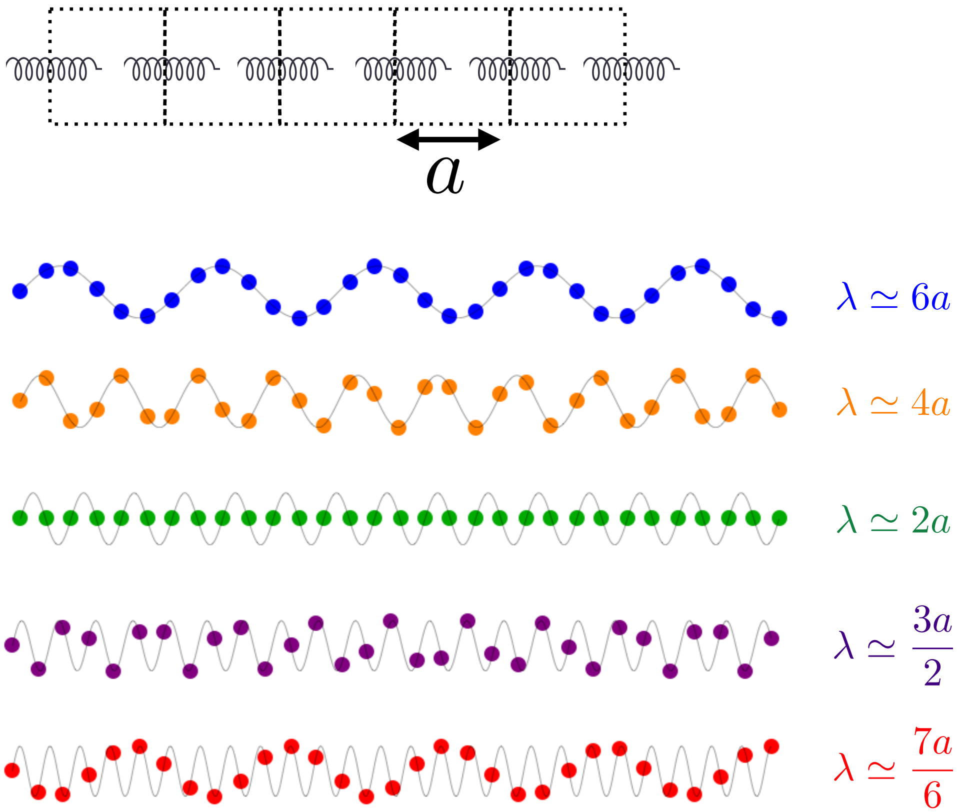

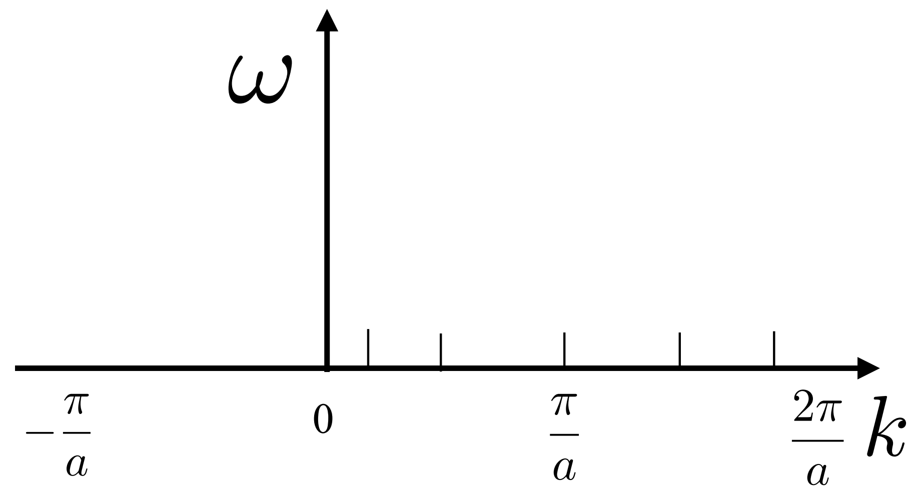

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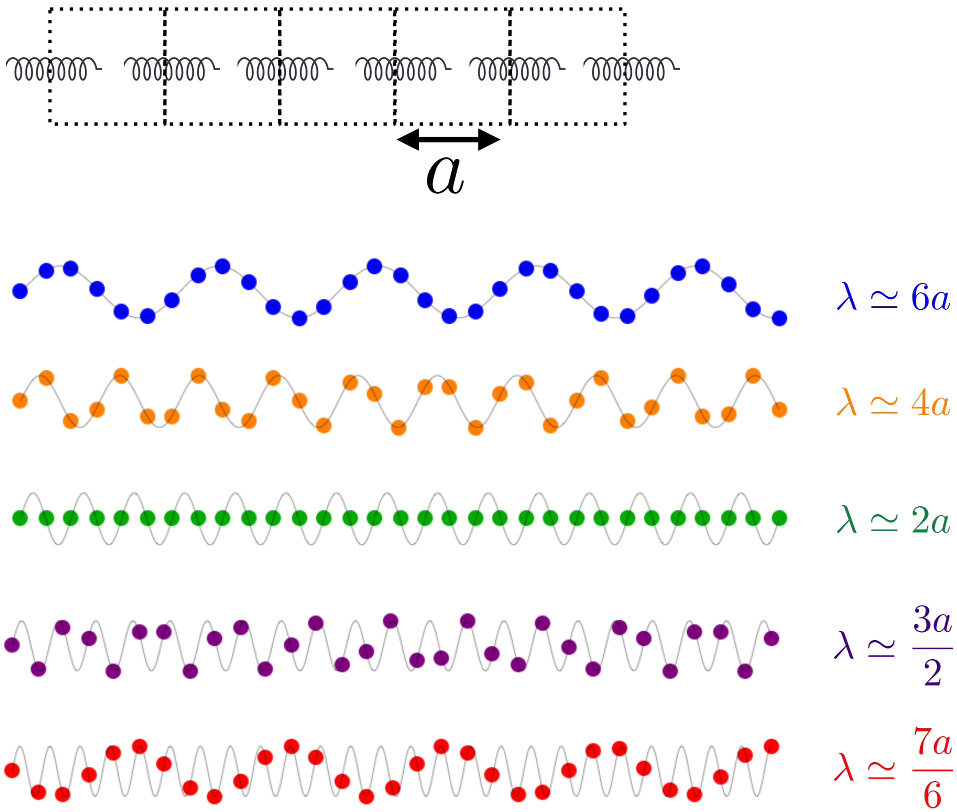

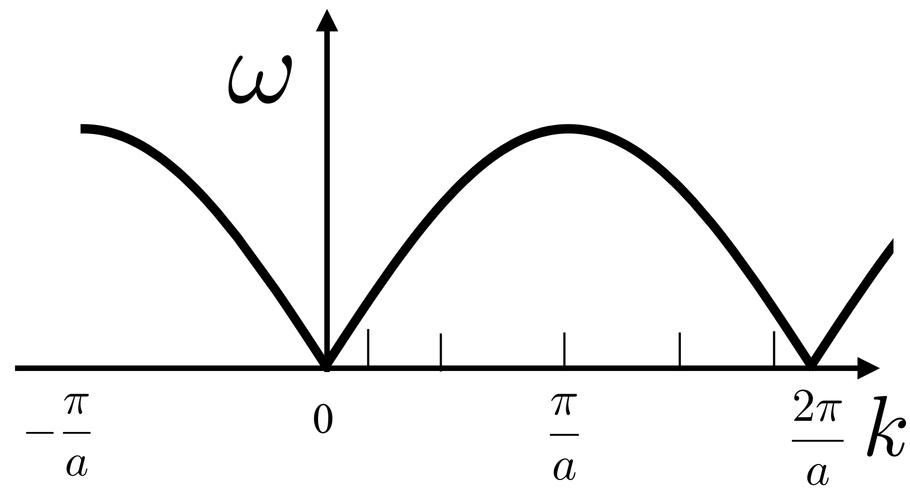

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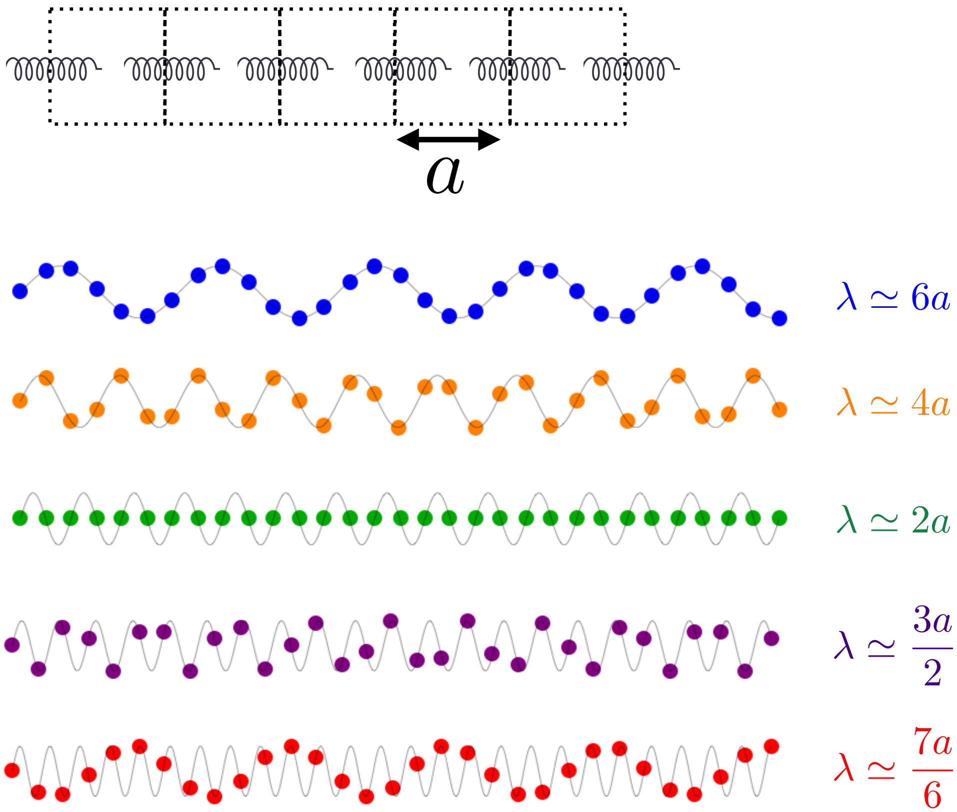

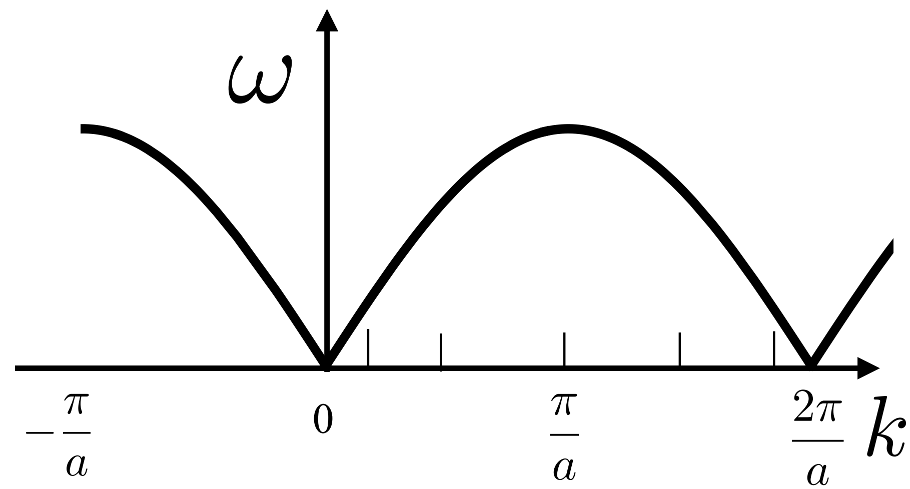

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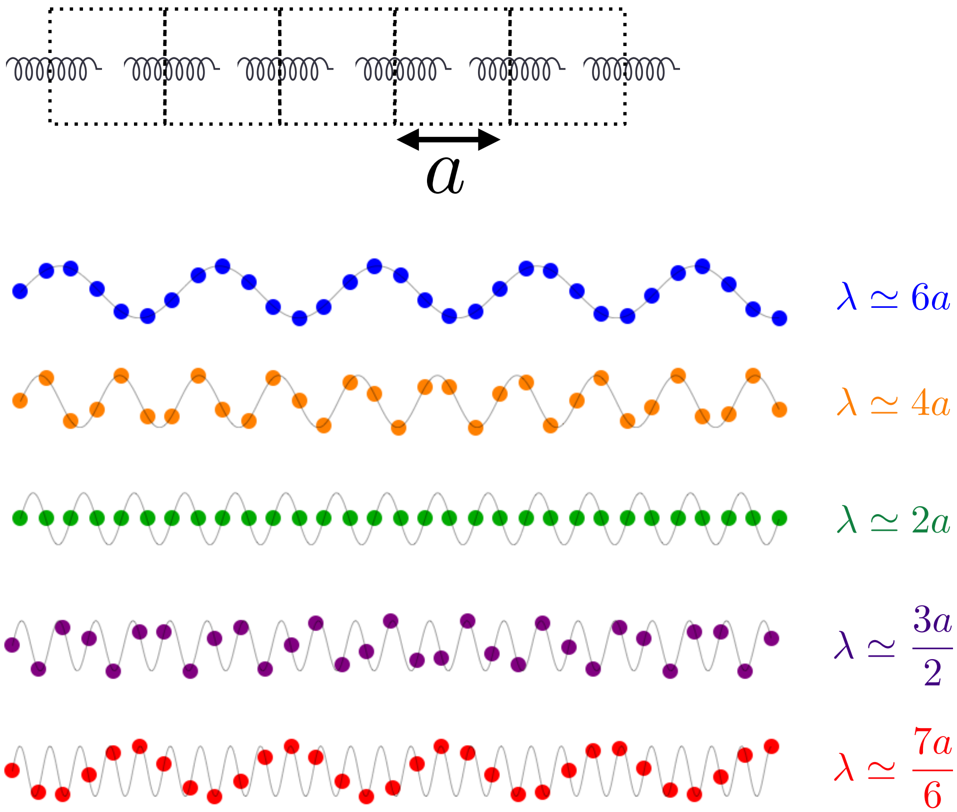

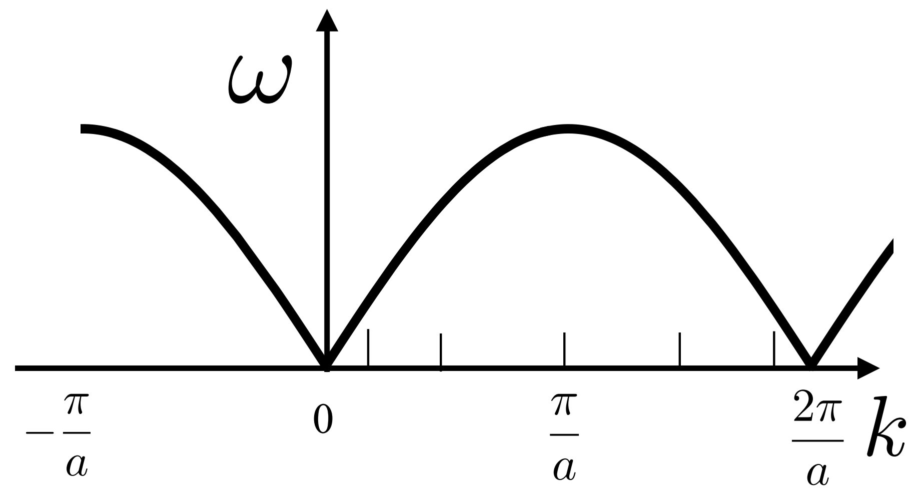

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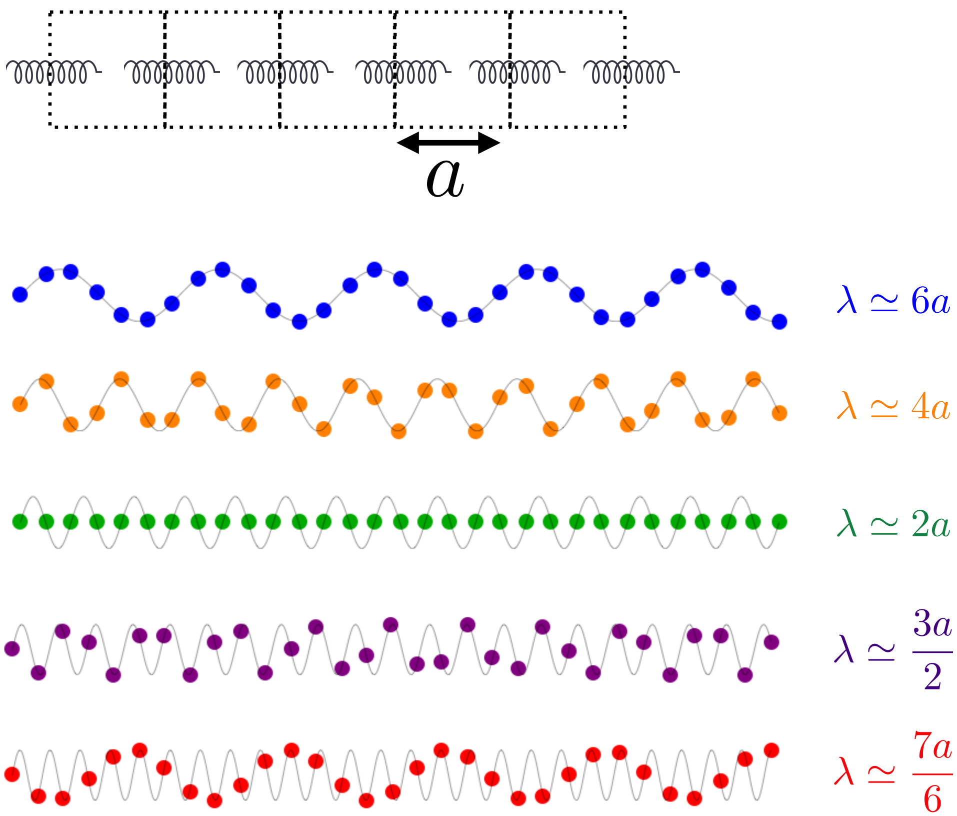

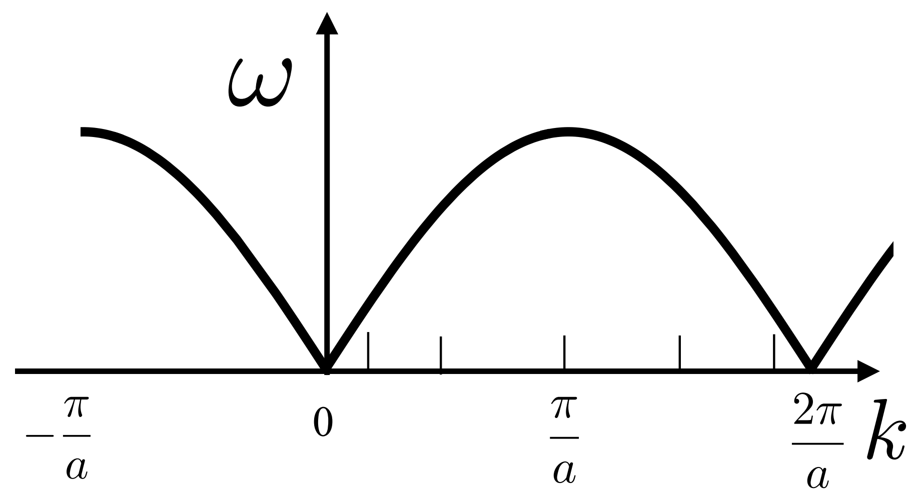

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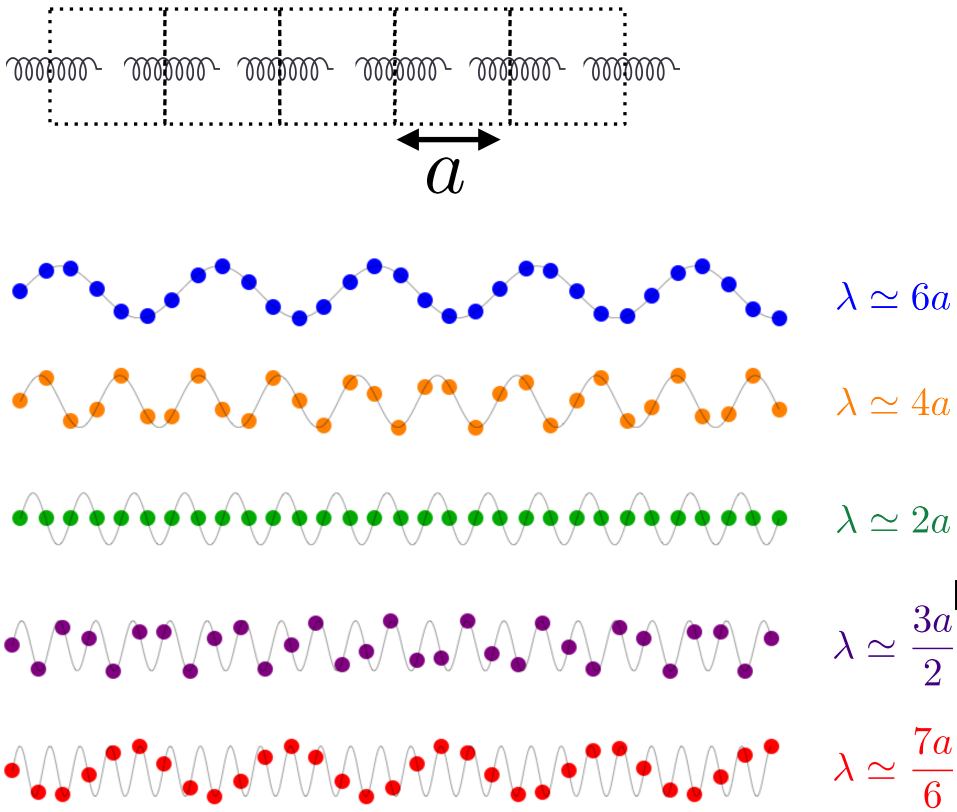
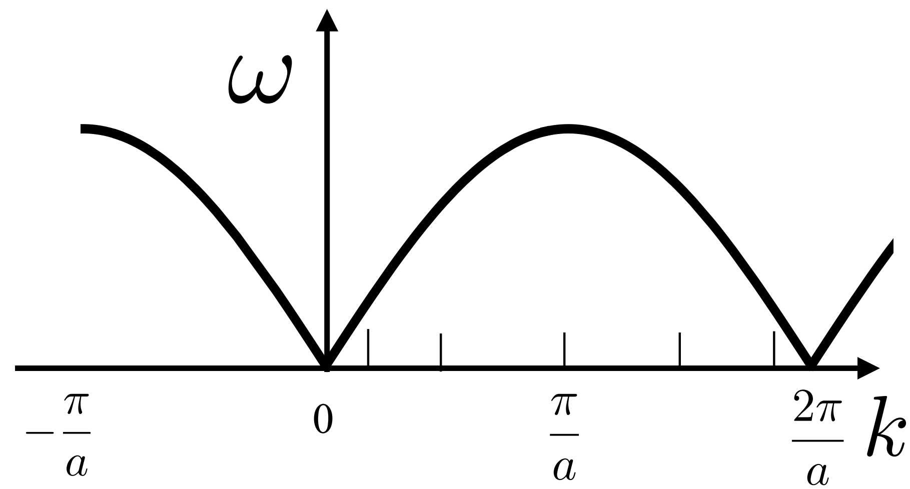




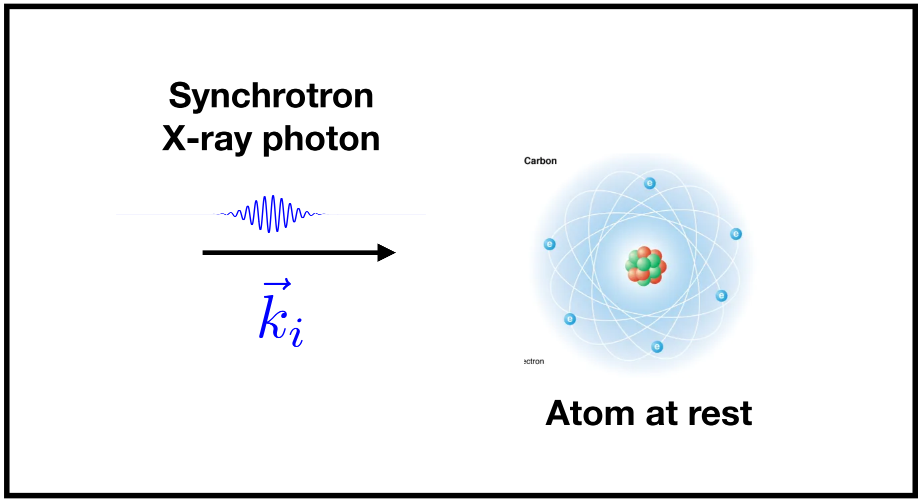
Lattice waves are periodic in momentum, just like the lattice

Quantum mechanics applied to lattice vibrations lead to the concept of “phonons”

Loosely, each k value refers to a different "phonon mode”

How do X-rays scatter from atoms?

Before collision



- Off resonance, an X-ray can cause an atom to recoil in a momentum conserving collision

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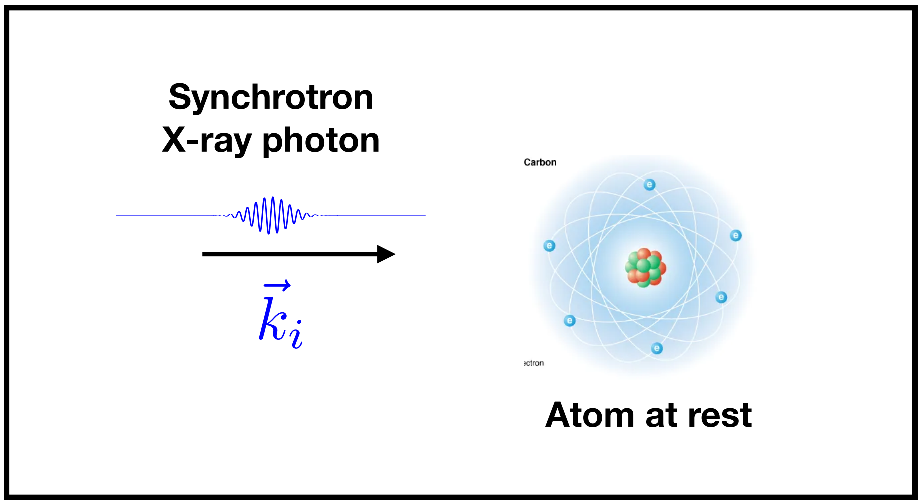
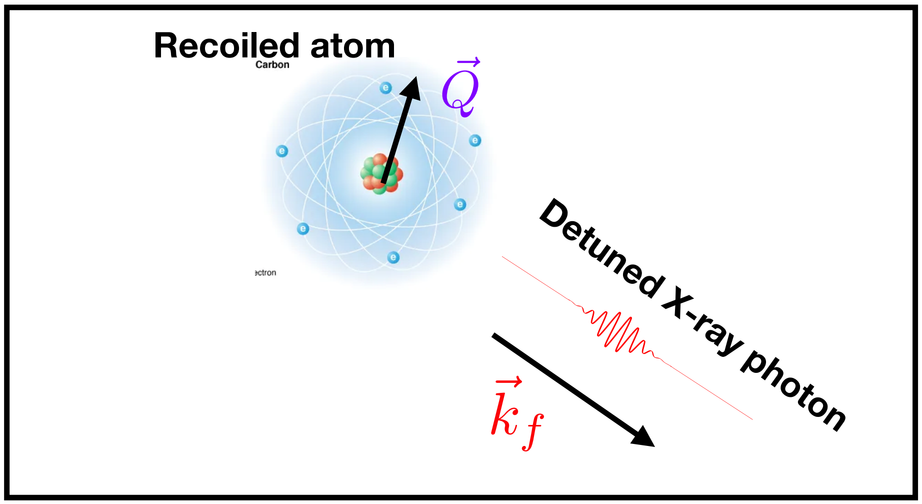


After collision



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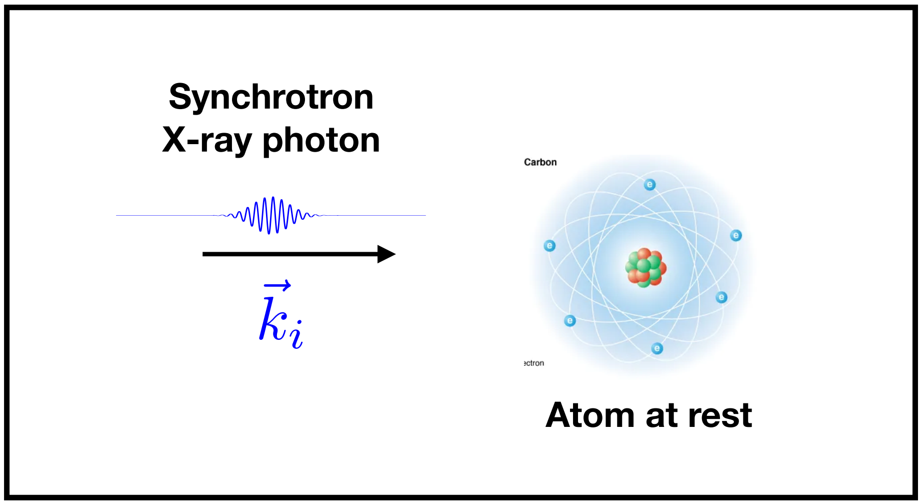

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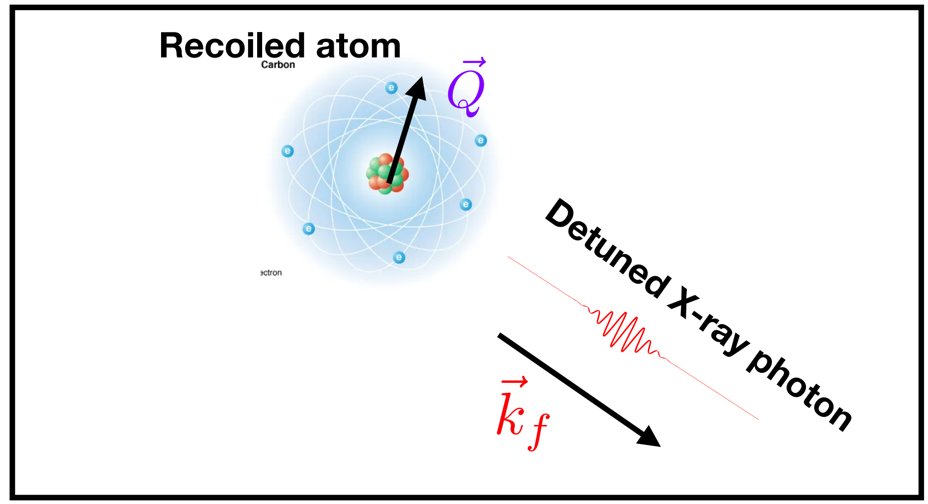
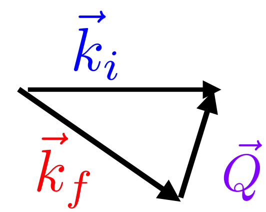


$\vec{Q} = \vec{k}_f - \vec{k}_i$ is the momentum delivered to the atom



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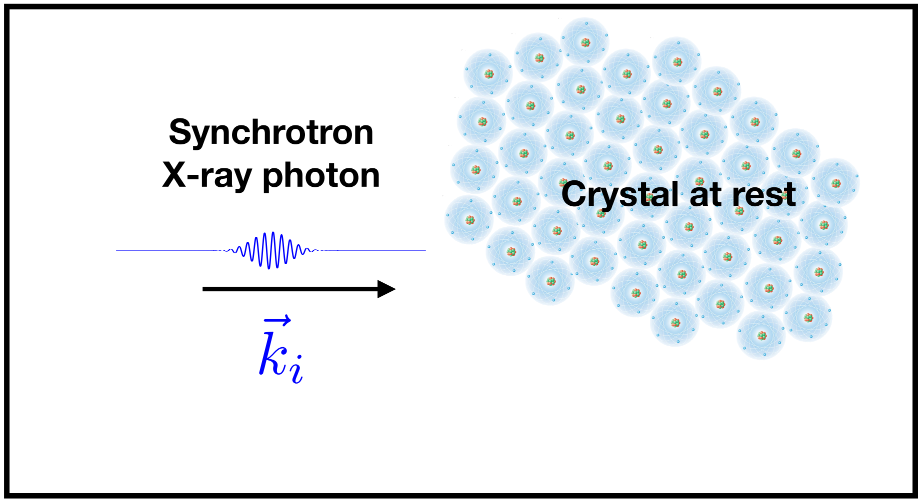

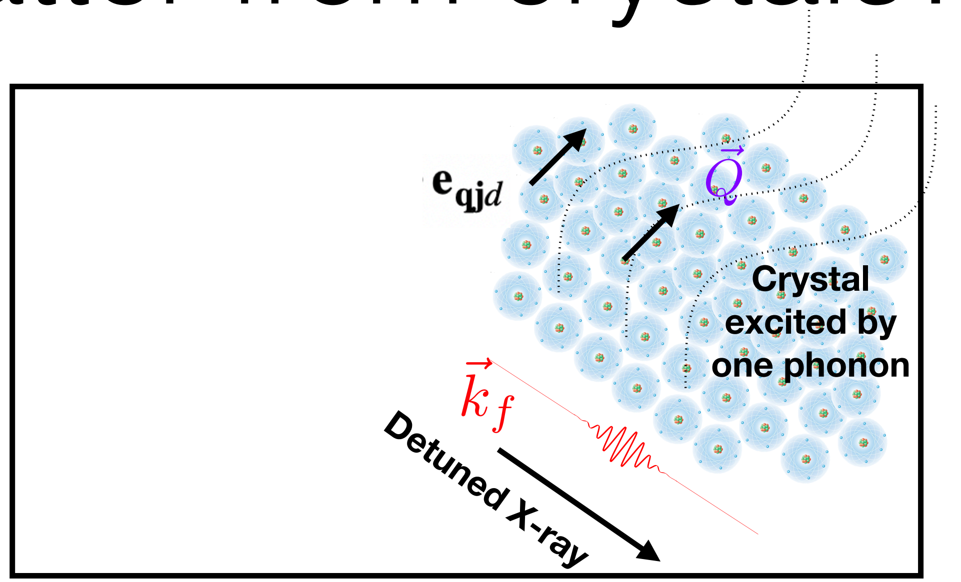

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- Probability of scattering off of a given phonon is strong when the displacements are along Q

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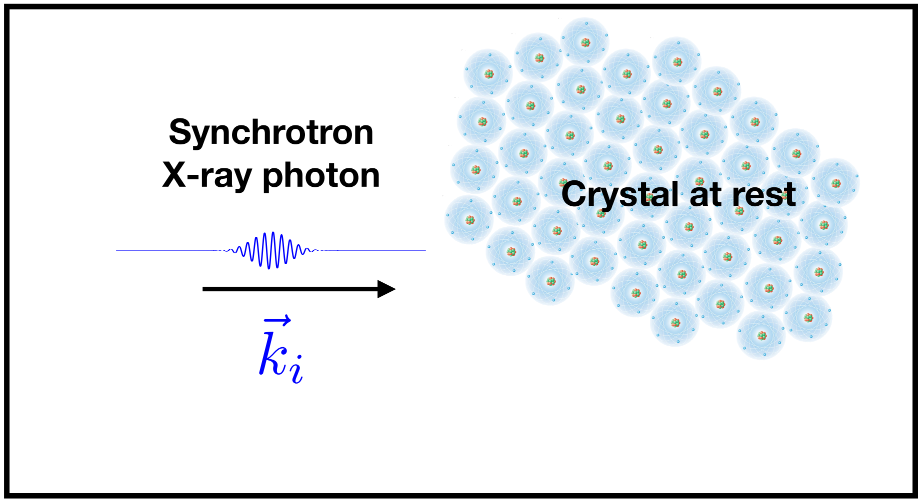

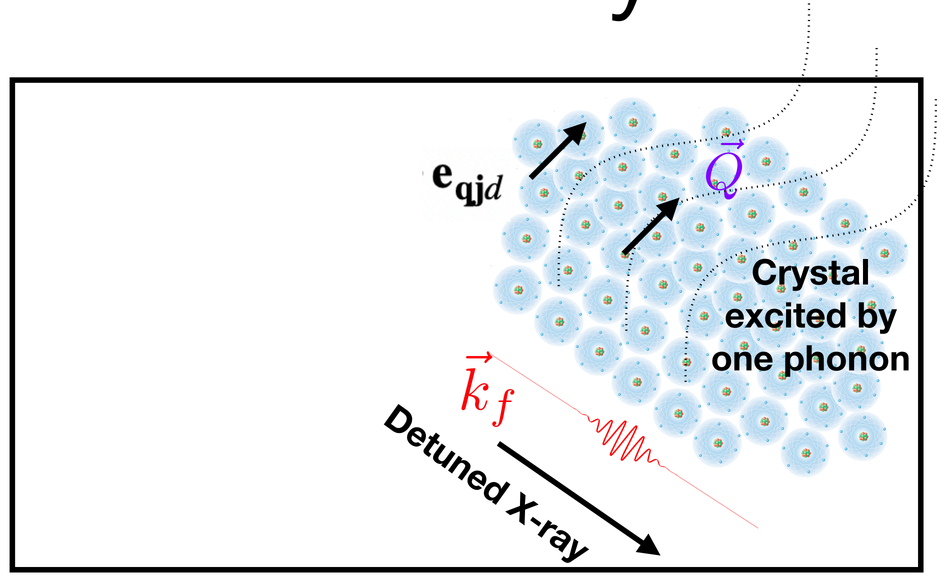

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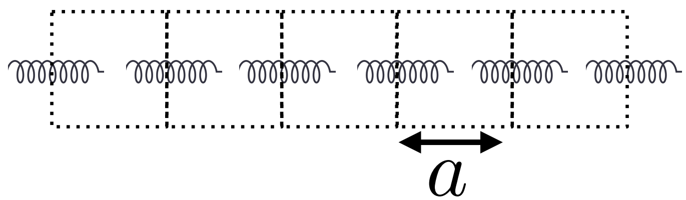
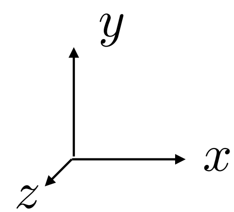
Polarization of phonons

- Atoms can move in three directions, different dispersion for different directions

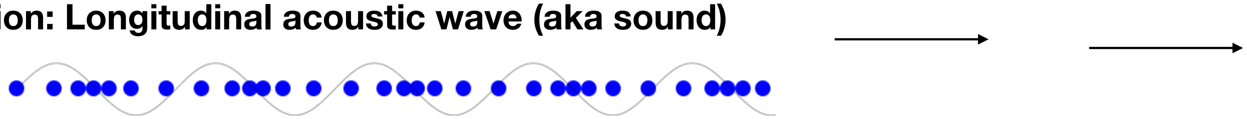




Displacement direction $\vec{k}$

Y or Z directions: Transverse acoustic waves

X-direction: Longitudinal acoustic wave (aka sound)



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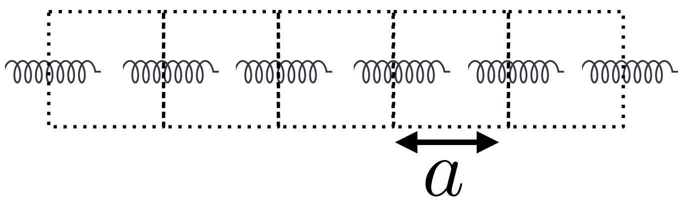

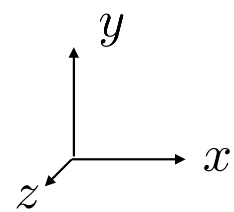

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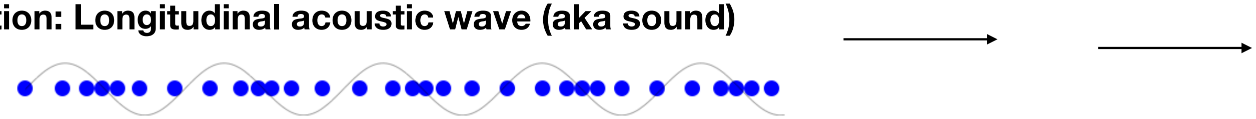

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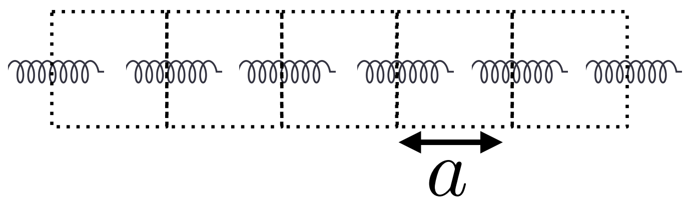

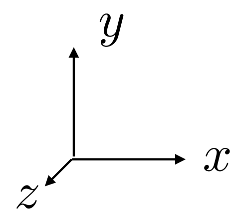

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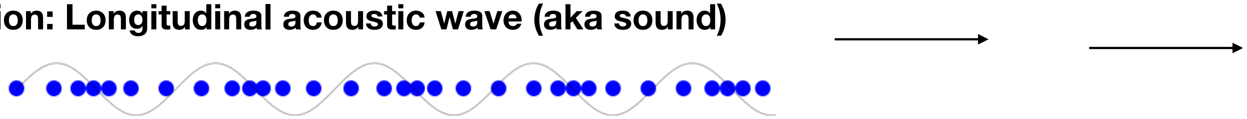

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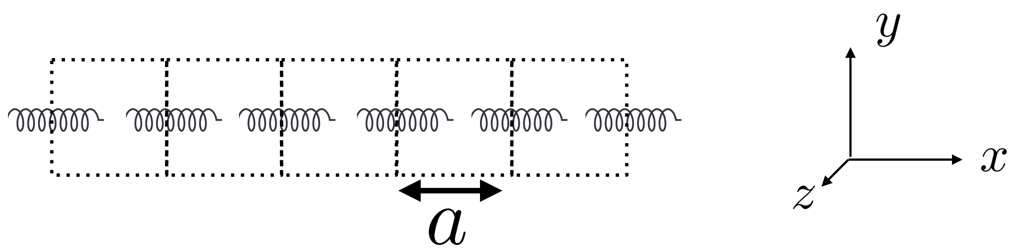

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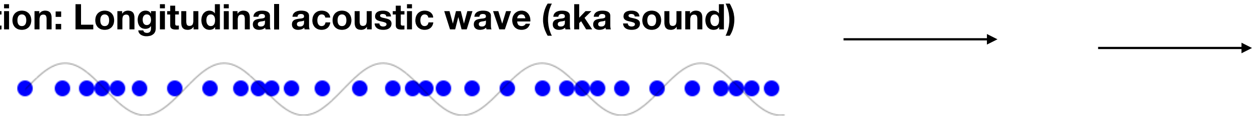

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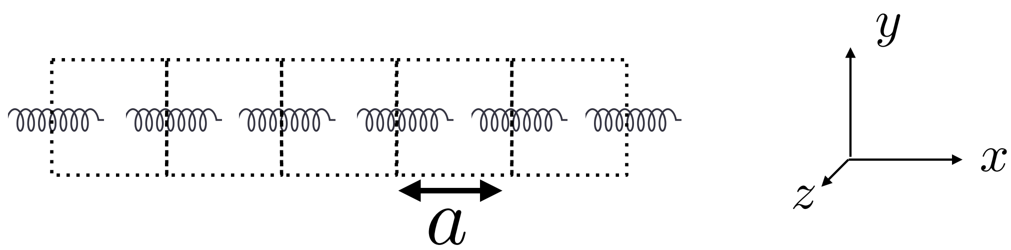
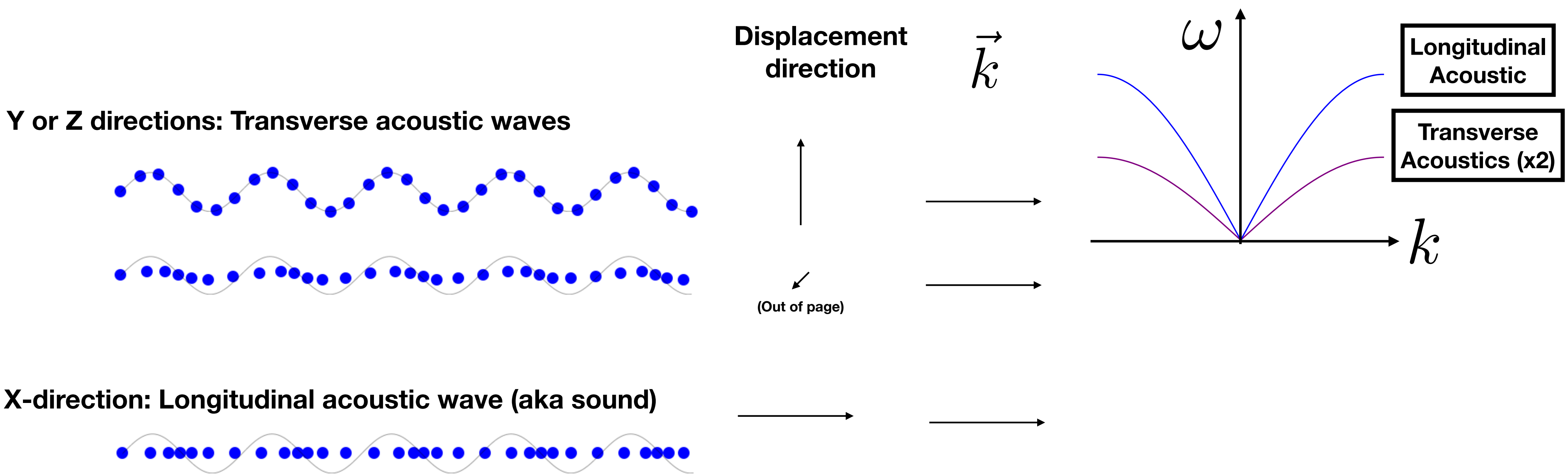
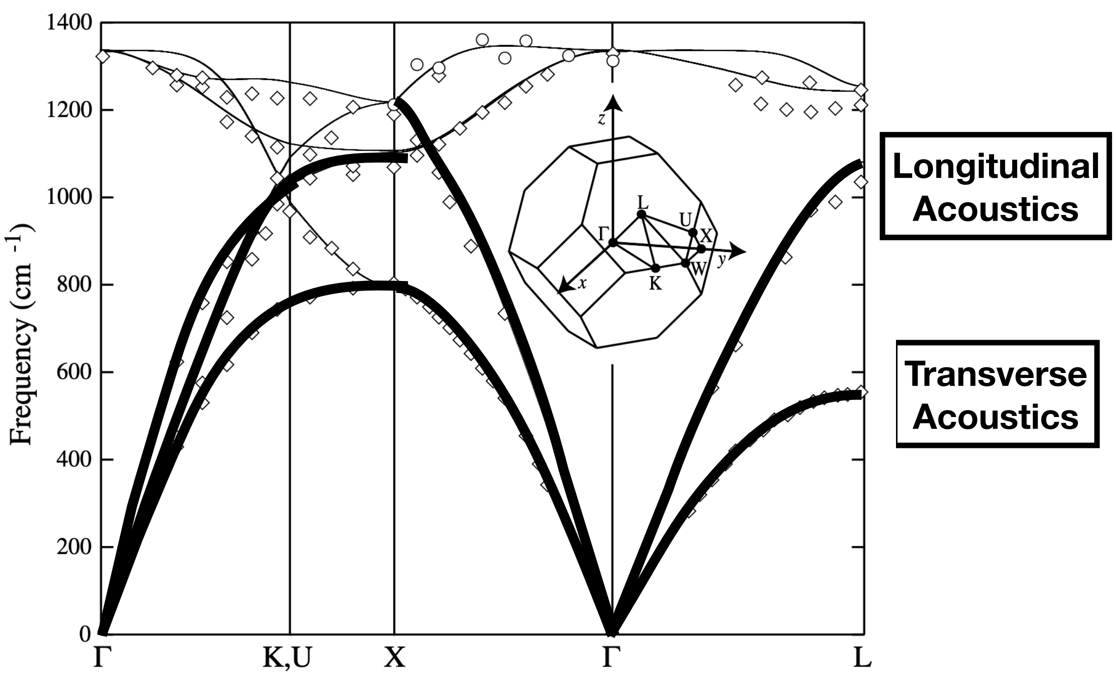


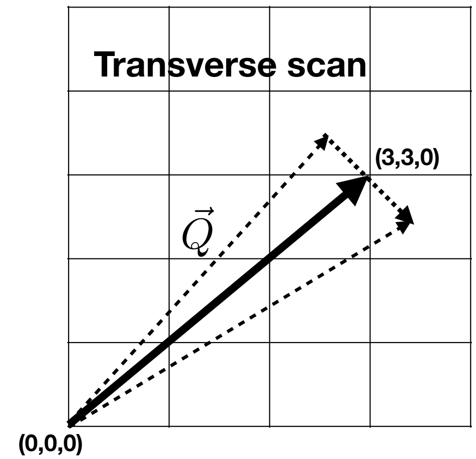


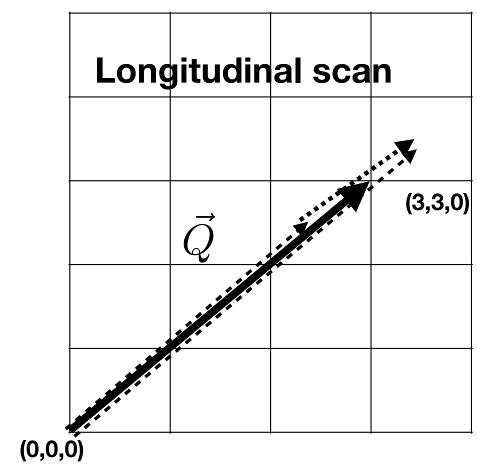
Measuring acoustic phonons with IXS

- Q sets the direction of atomic displacements

- Polarization of atomic displacement wrt reduced q can be changed by scanning different directions

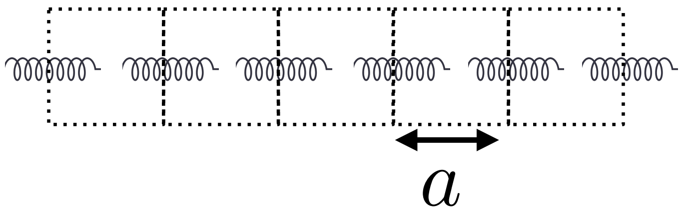


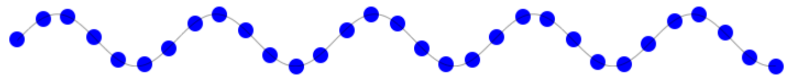


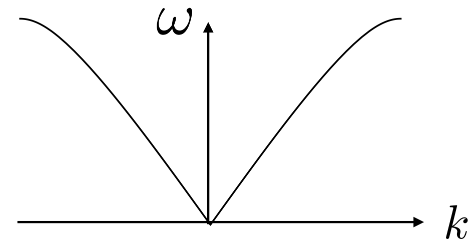


What about more complicated unit cells?

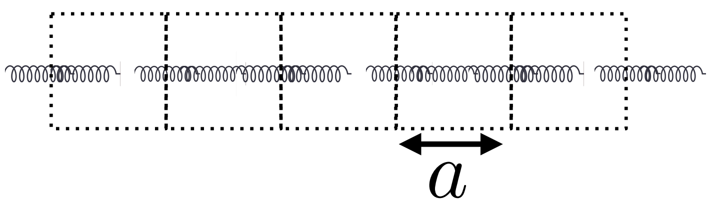
- *Mon*atomic 1D chain

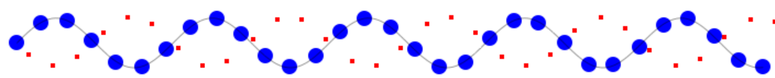




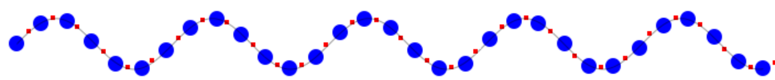


- *Di*atomic 1D chain





Red atom beats against blue atom



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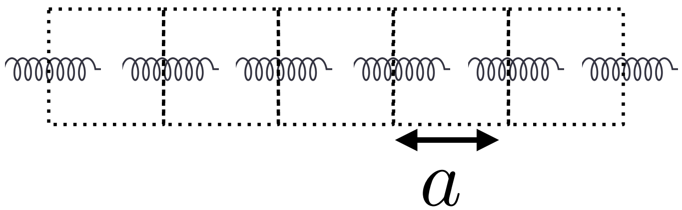
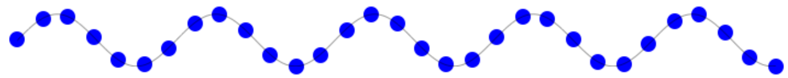
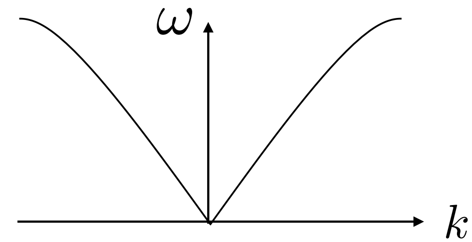
What about more complicated unit cells?

- *Mon*atomic 1D chain







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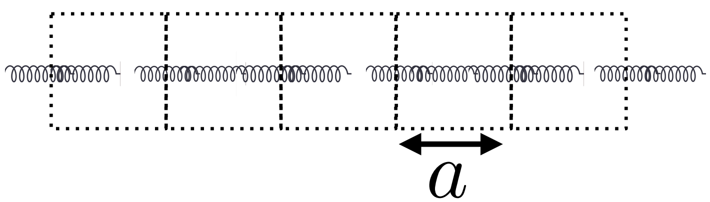

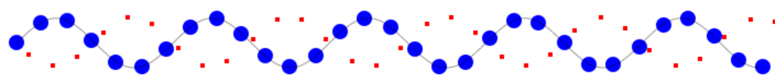

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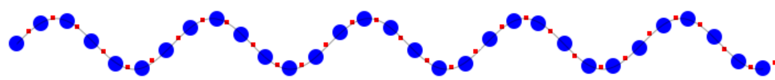

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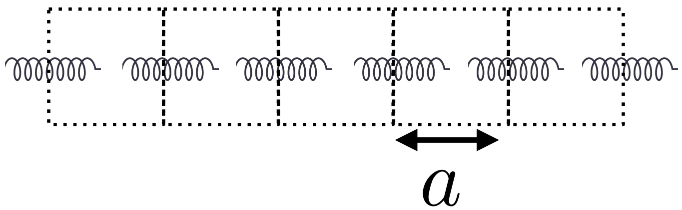

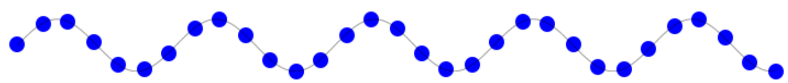

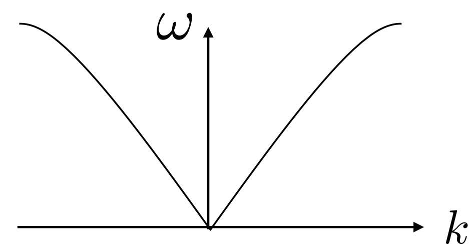

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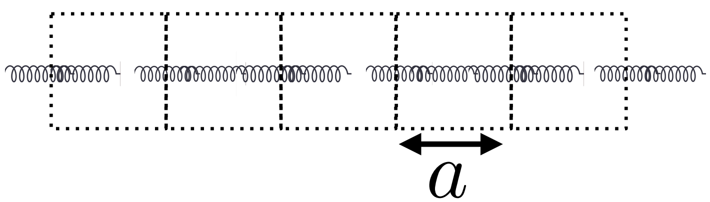

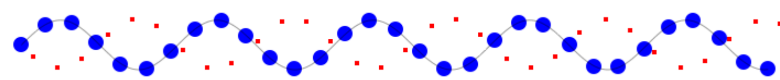

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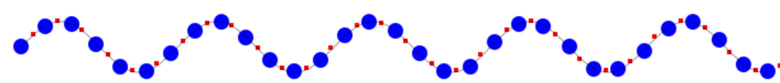

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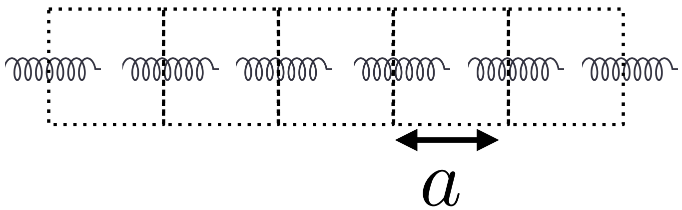

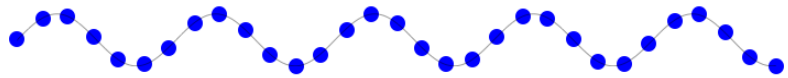

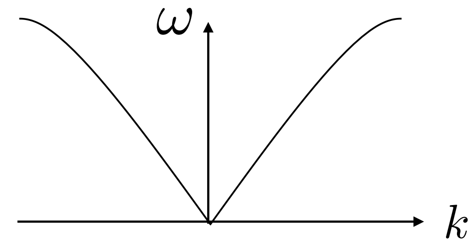

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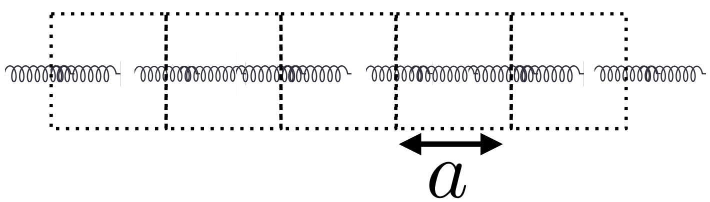

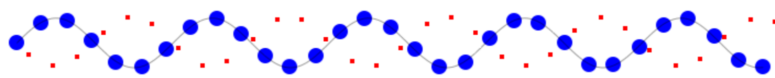

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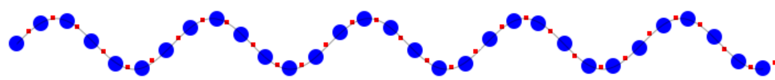

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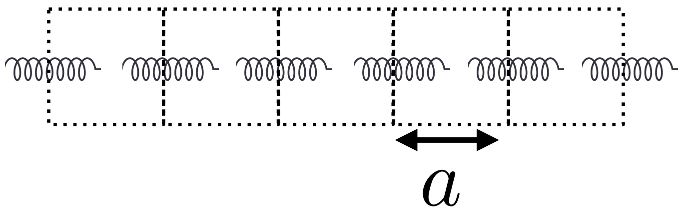

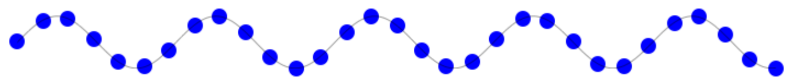

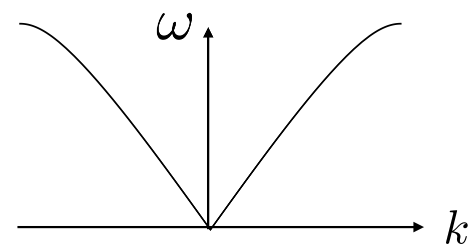

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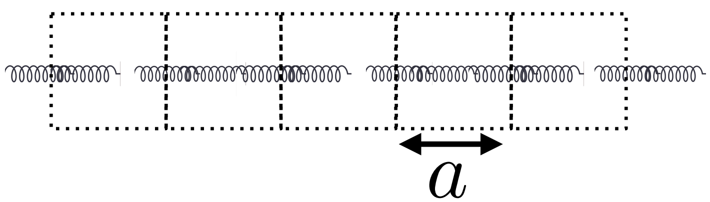

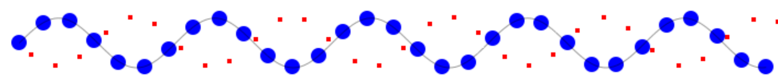

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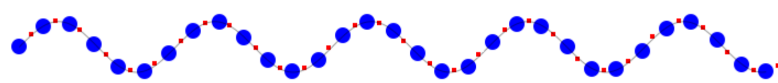

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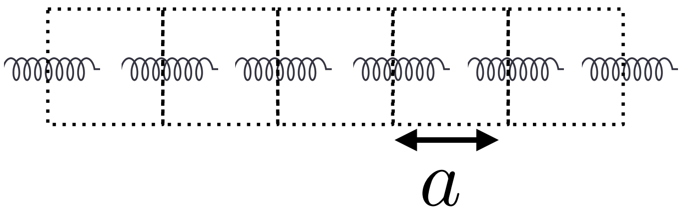

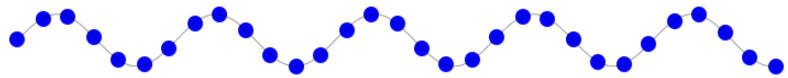

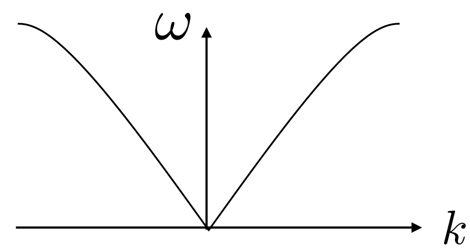

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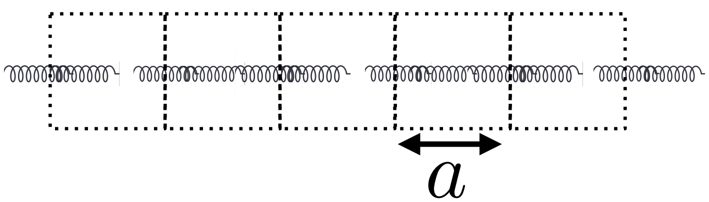

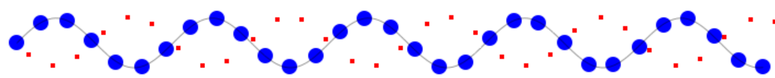

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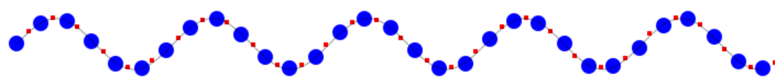

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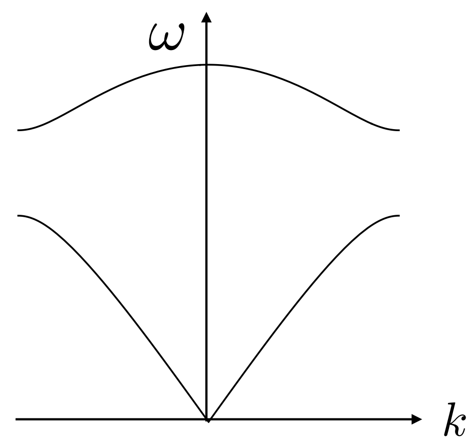

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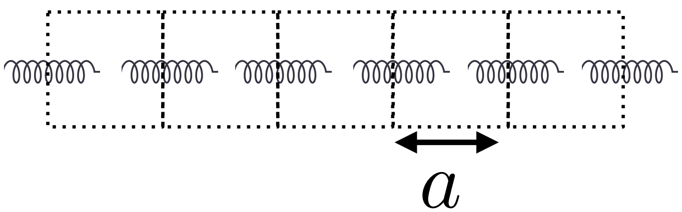

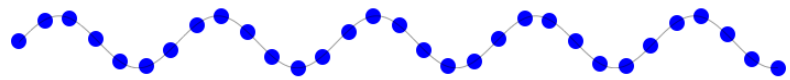

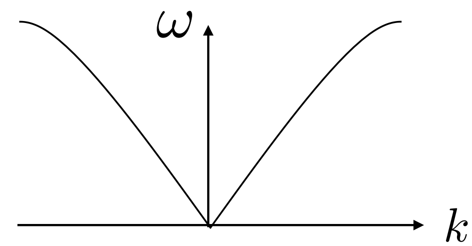

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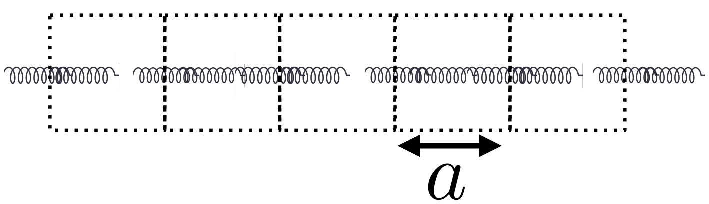

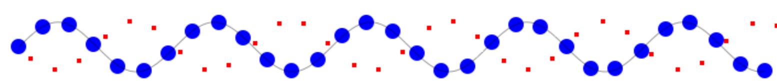

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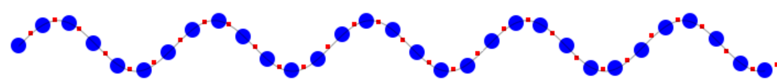

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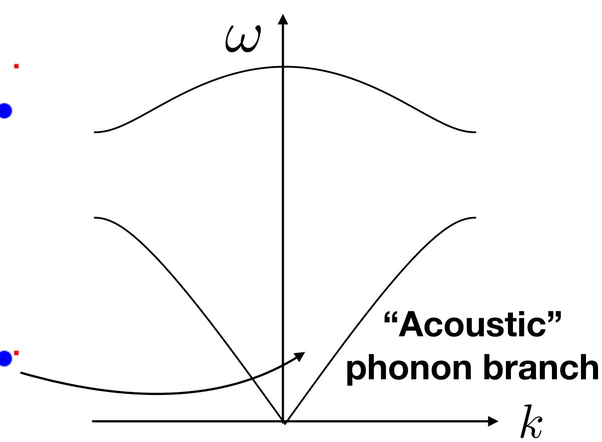

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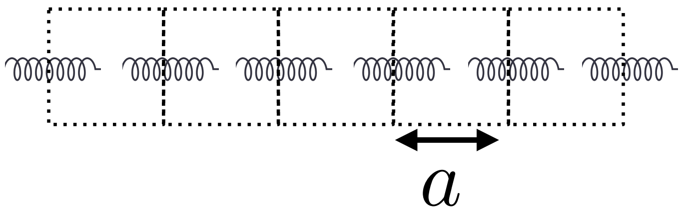

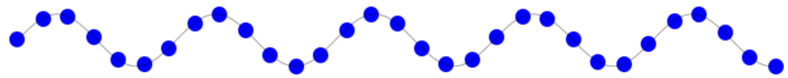

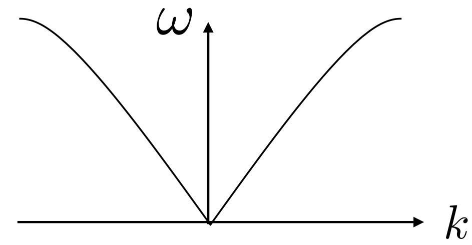

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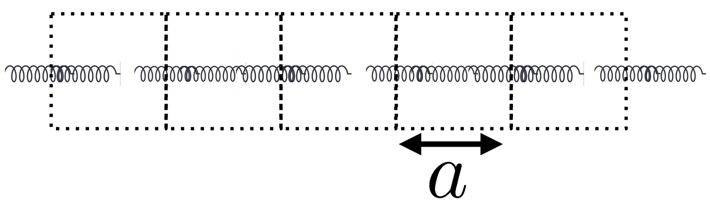
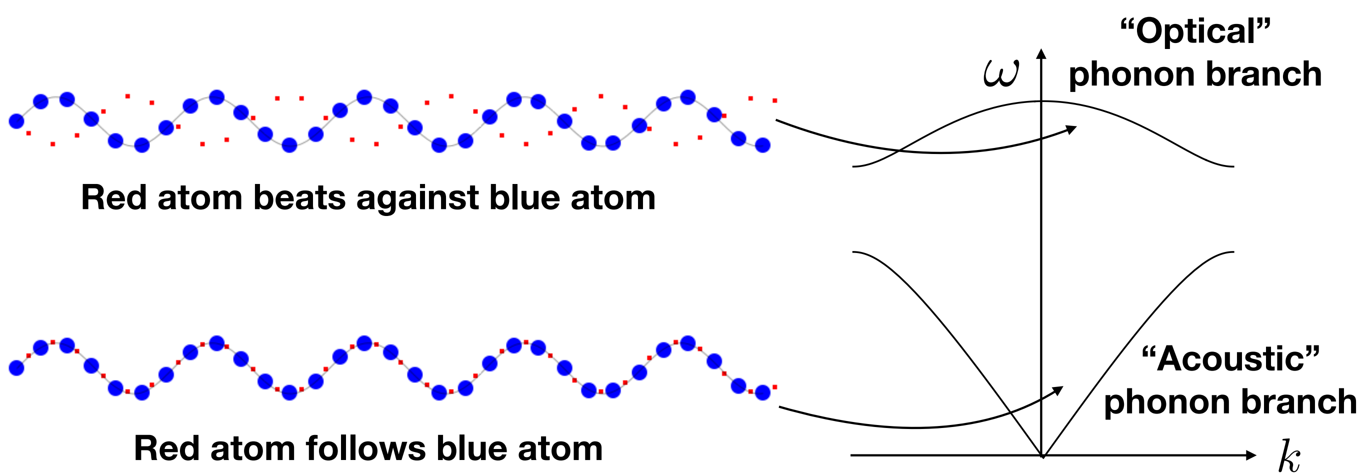
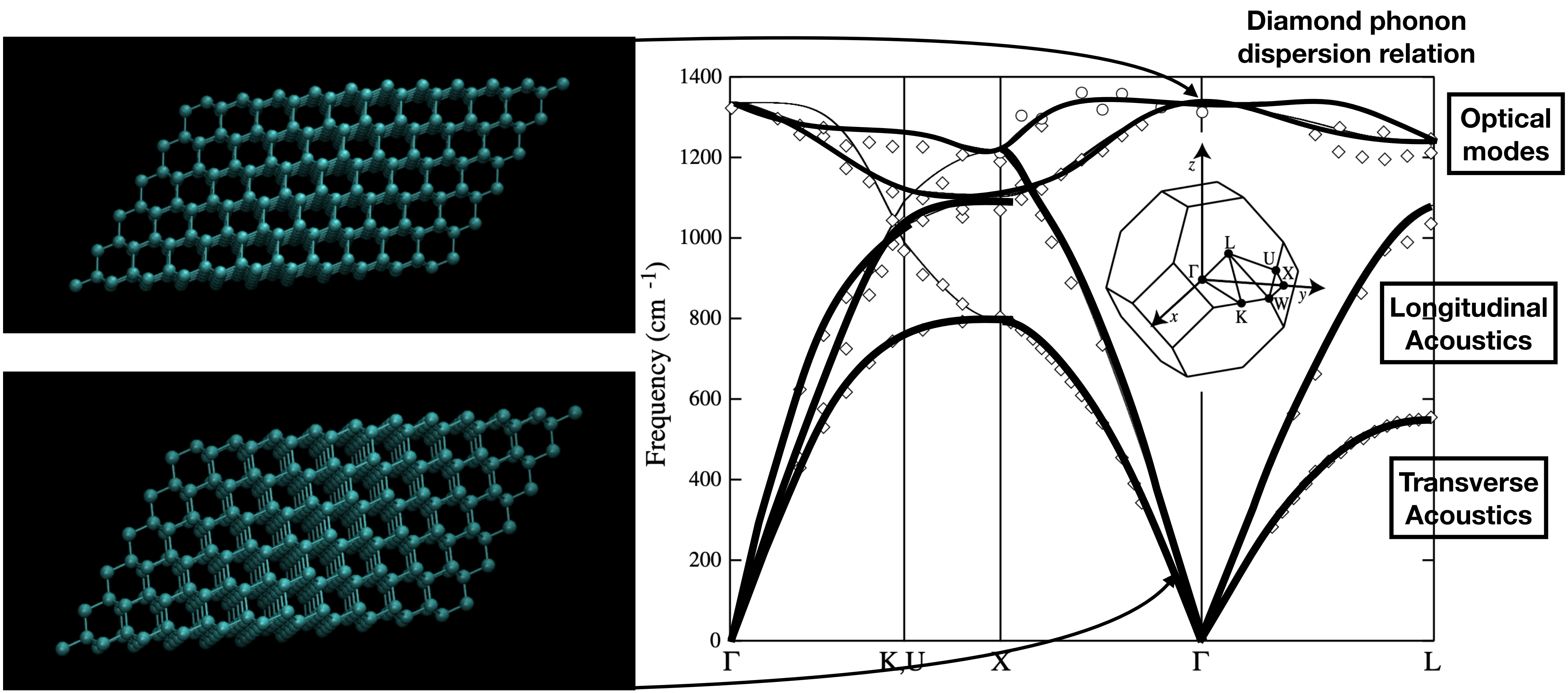




Phonons throughout the Brillouin zone



Gonze, et al, Z. Krist. 220 (2005) 458

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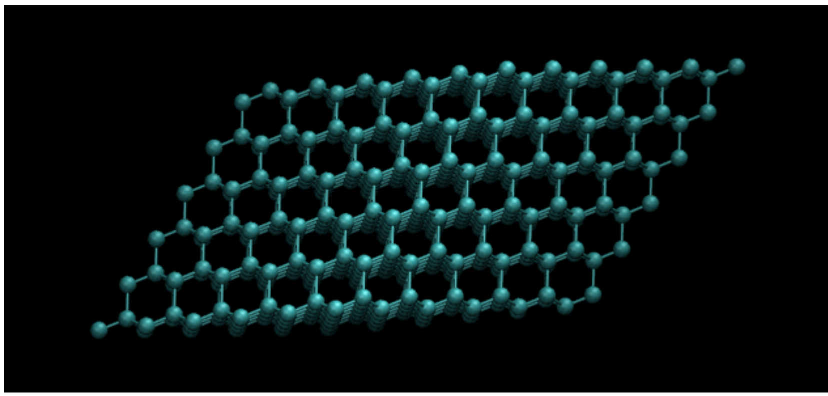
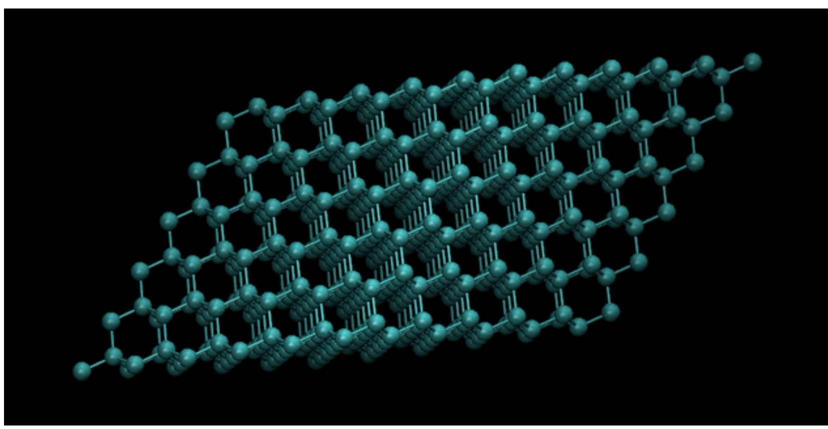
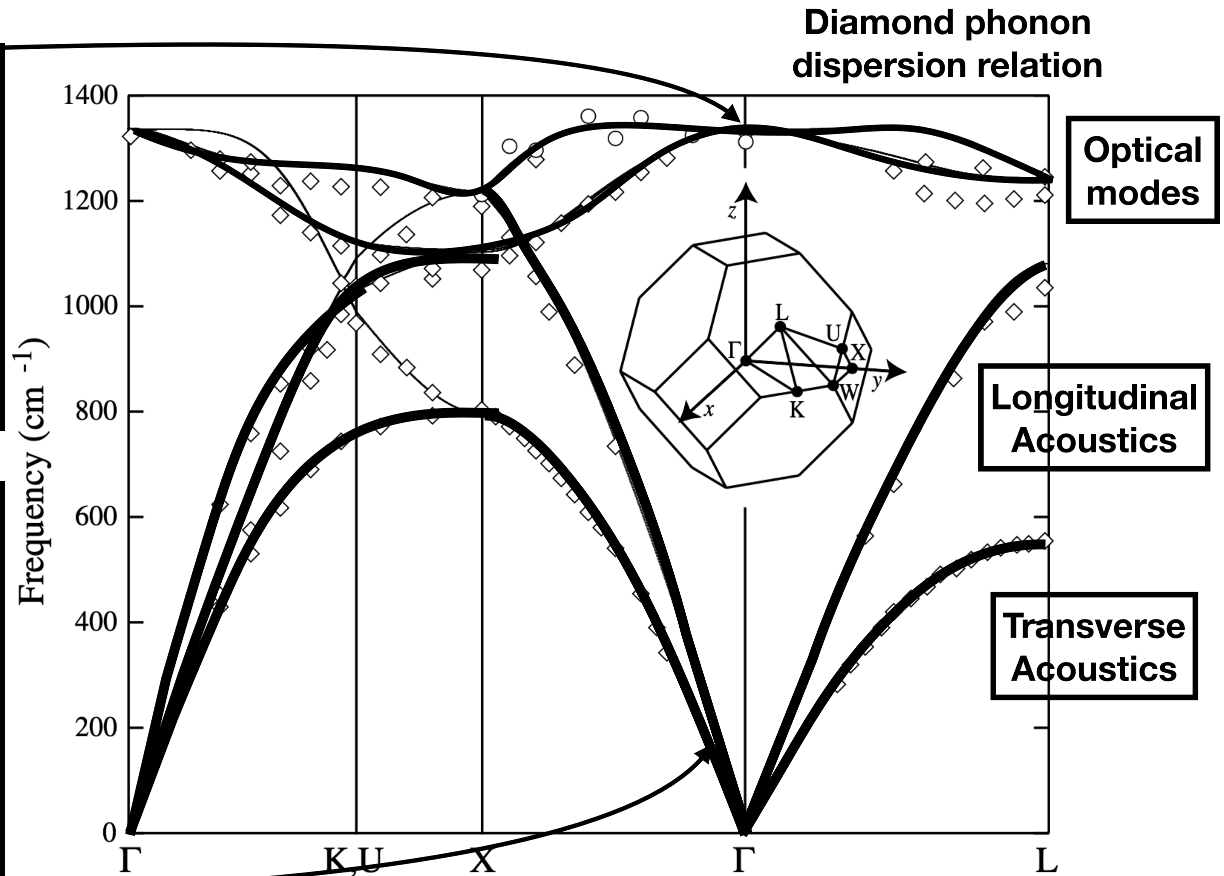
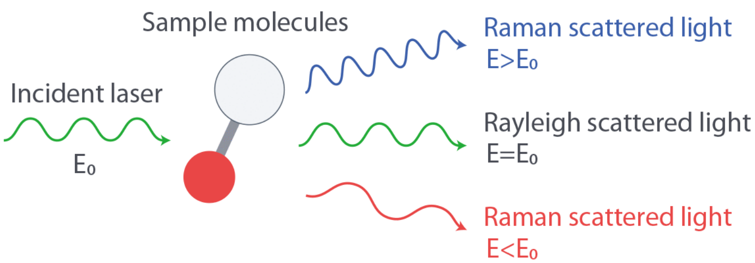


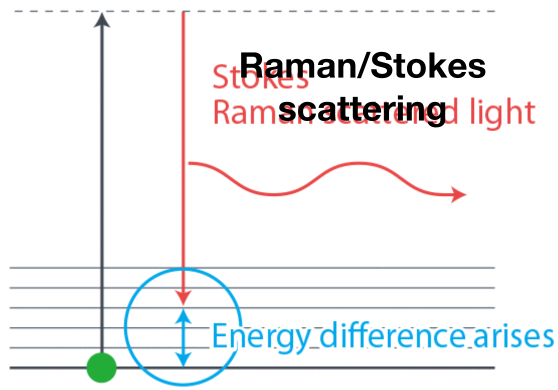




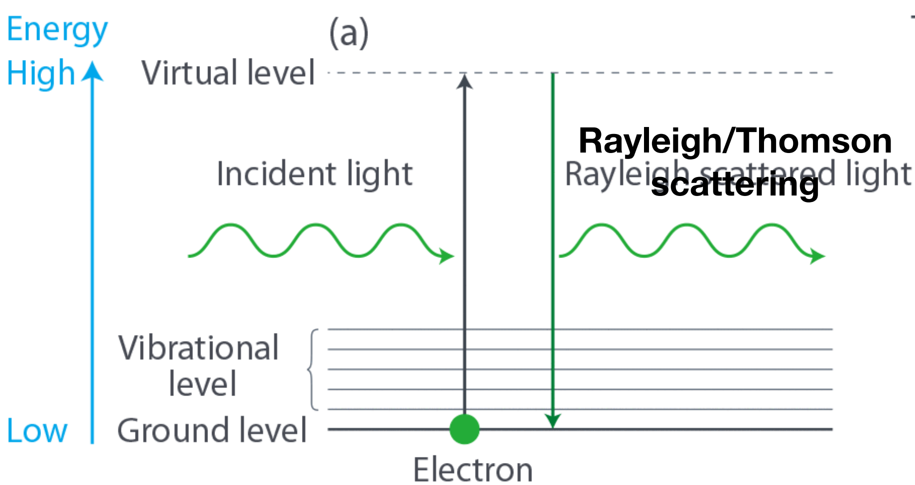
Gonze, et al, Z. Krist. 220 (2005) 458

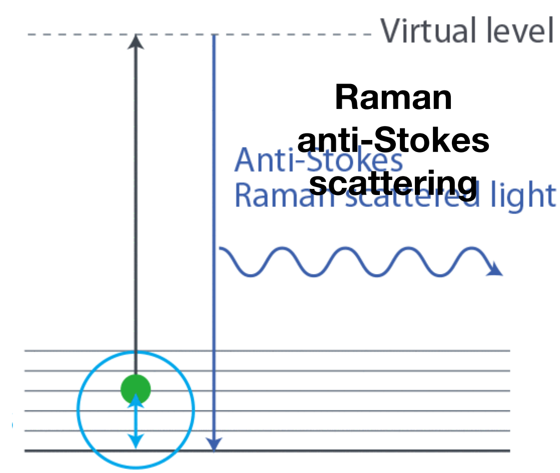
Raman=inelastic light scattering





Discovered 1928
Nobel 1930

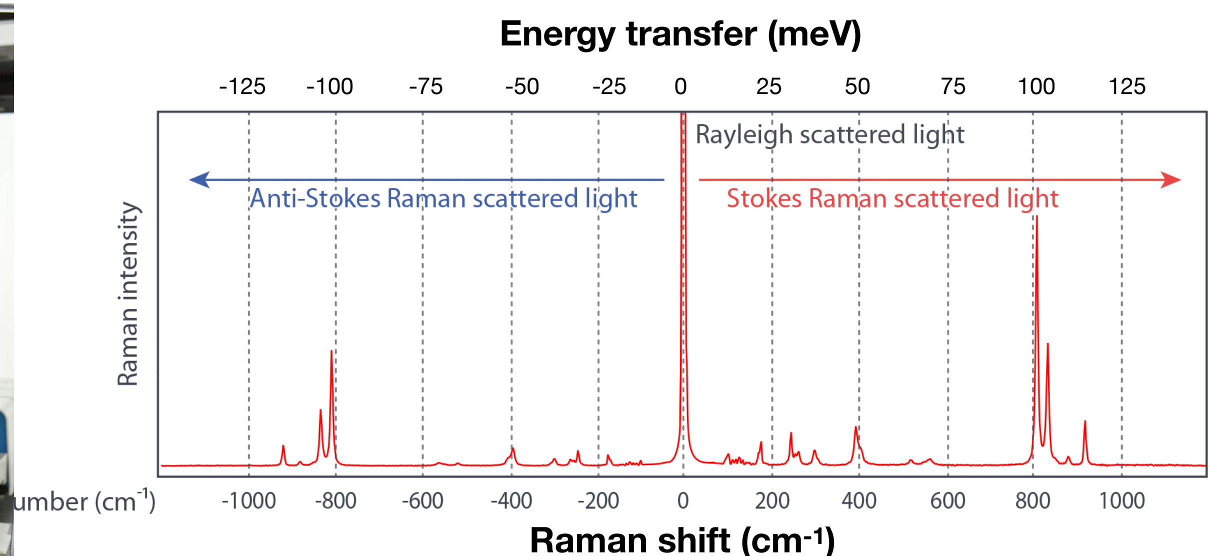




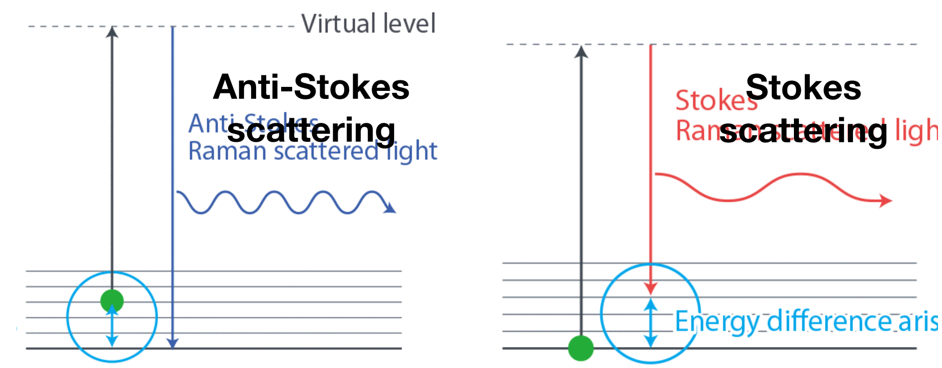
Strong limitation: momentum of light is very small

https://www.nanophoton.net/

Energy transfer and Raman shift

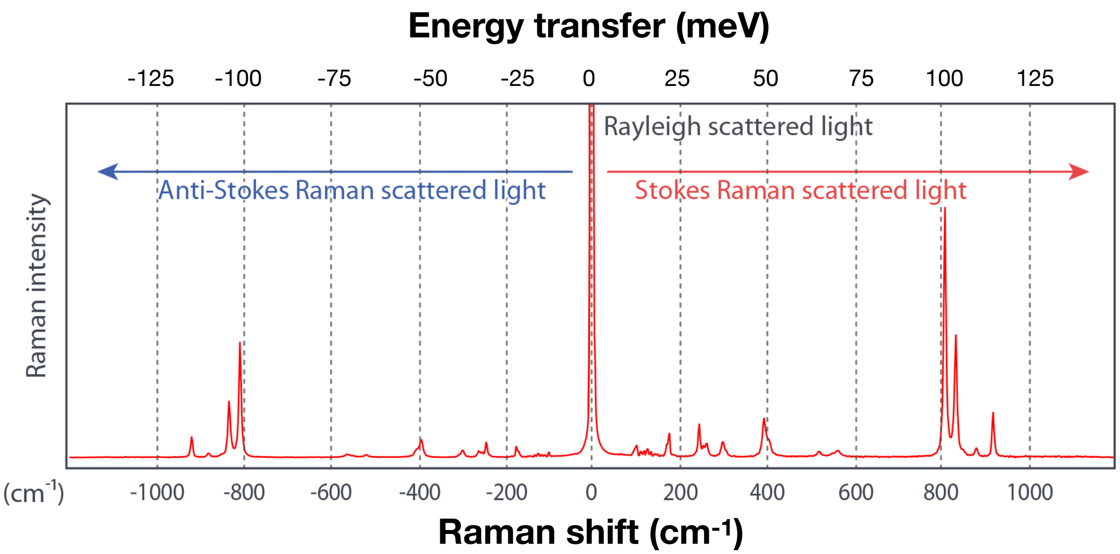


- Laser source=high resolution
- Lattice excitations easy to see
- Electronic and magnetic excitations possible



Light or X-ray gains energy **Light or X-ray loses energy**

Energy transfer and Raman shift



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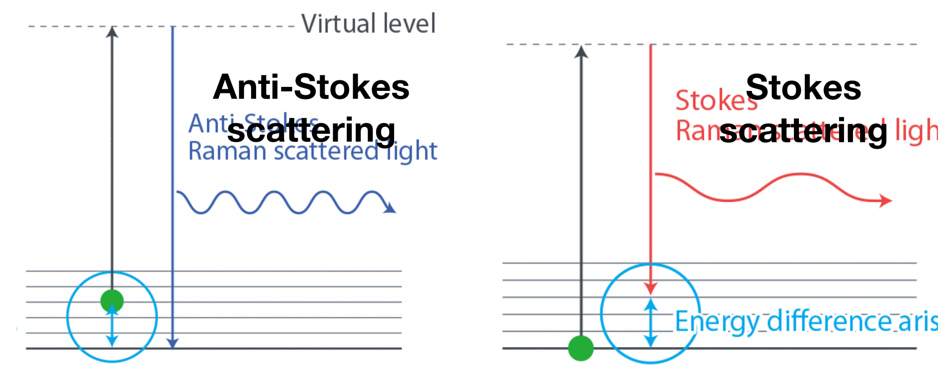

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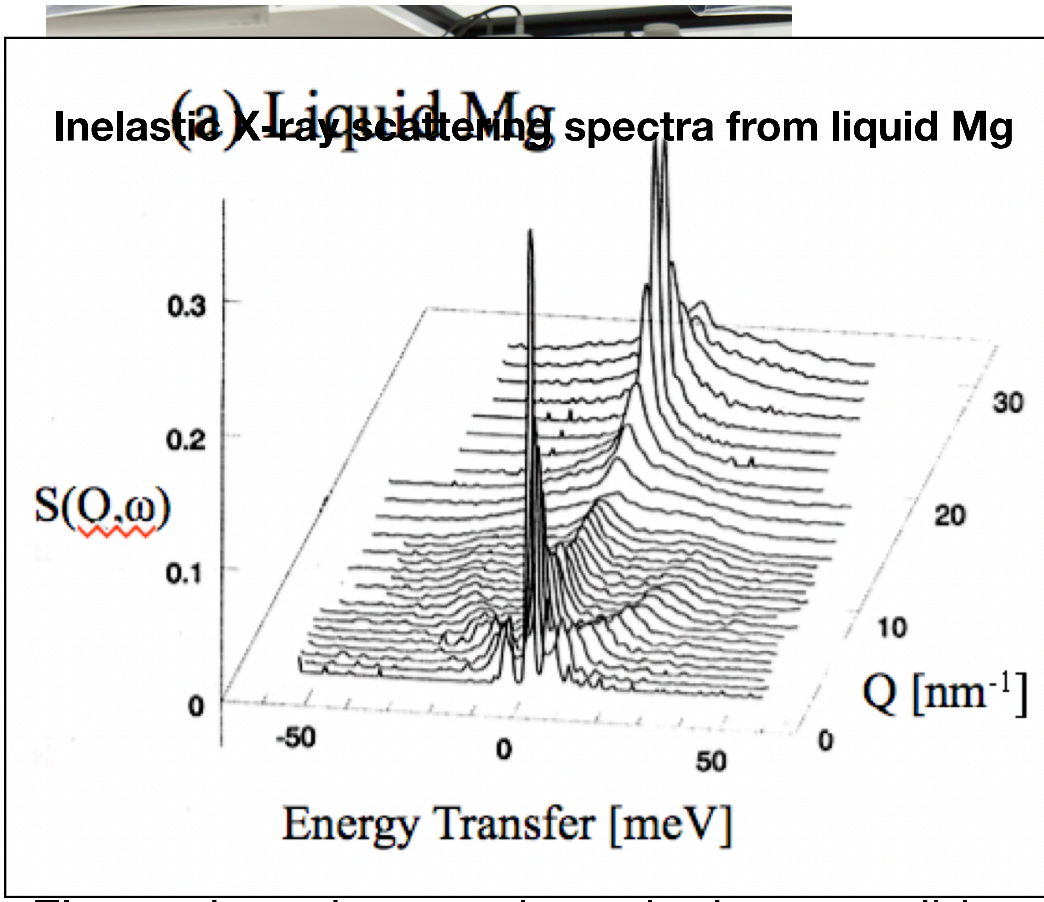
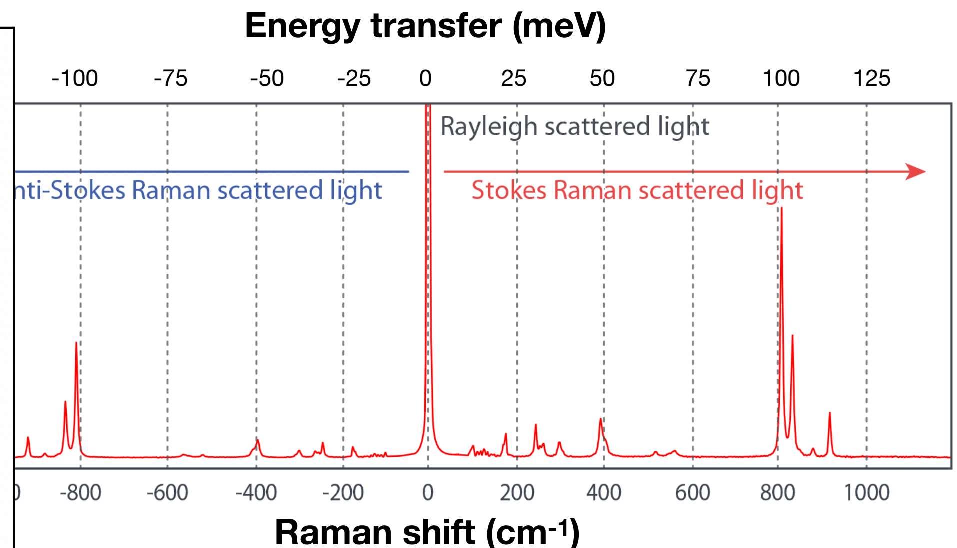
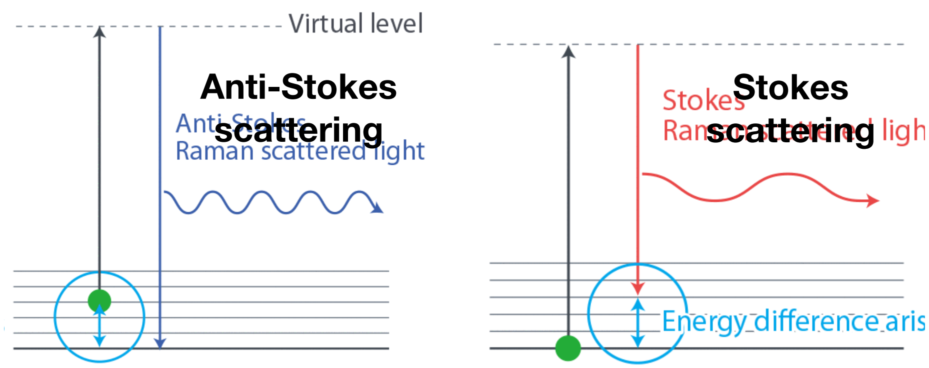
Energy transfer and Raman shift

Inelastic X-ray scattering spectra from liquid Mg







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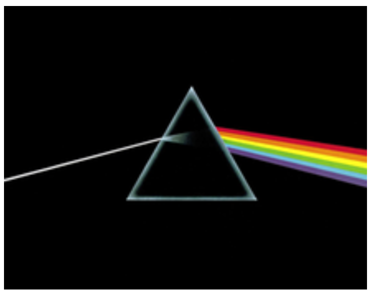
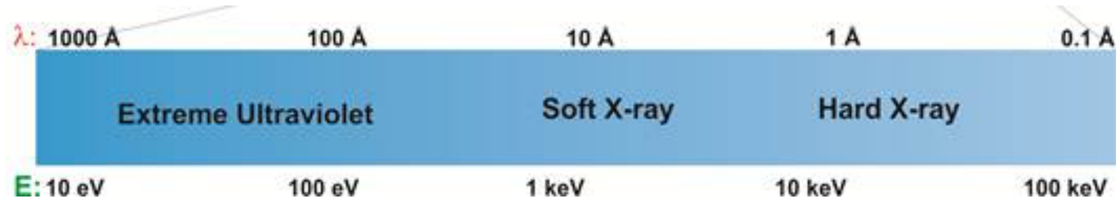
How can we disperse X-rays according to energy/wavelength?

Prism?





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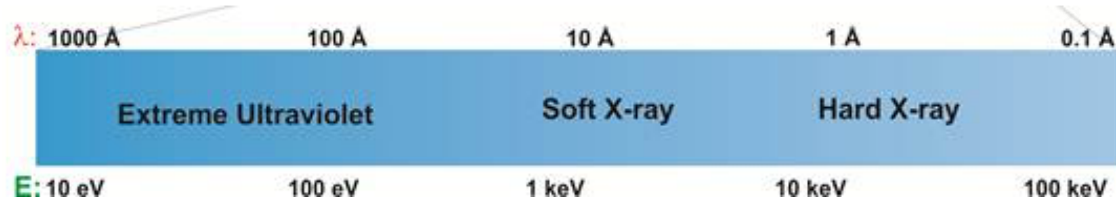

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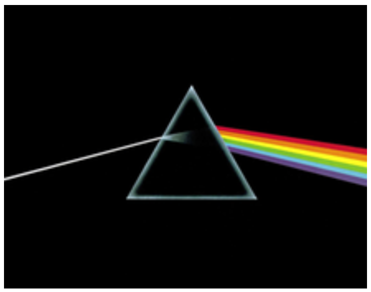

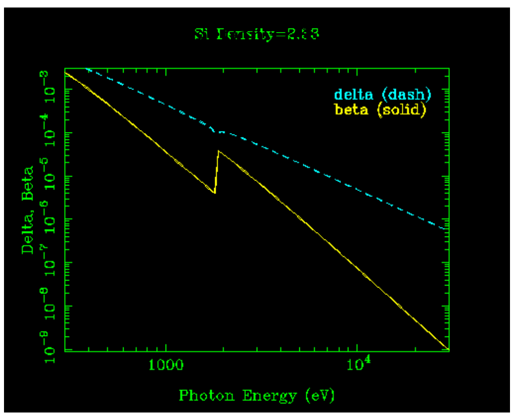

Inefficient
Index changes too small to be effective

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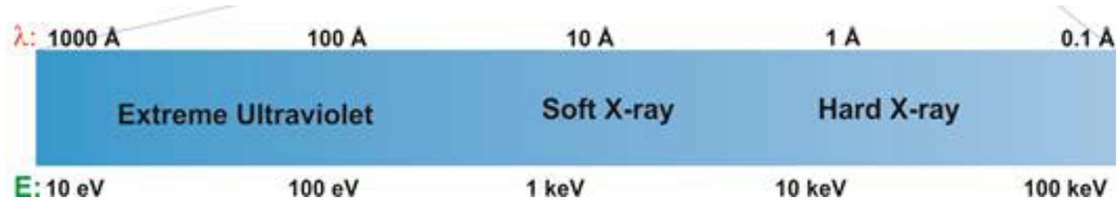

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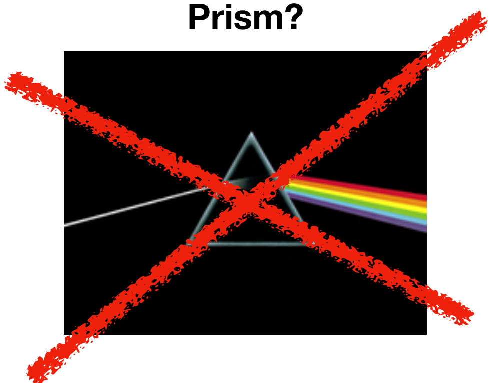

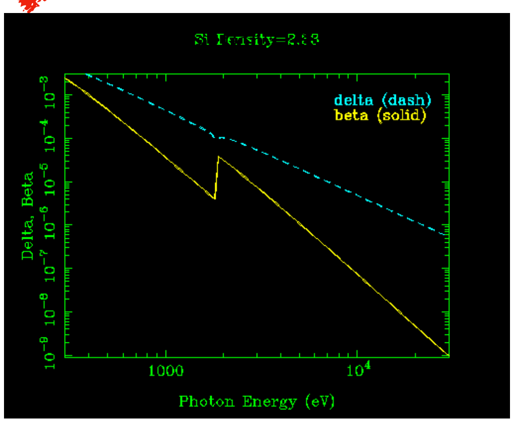

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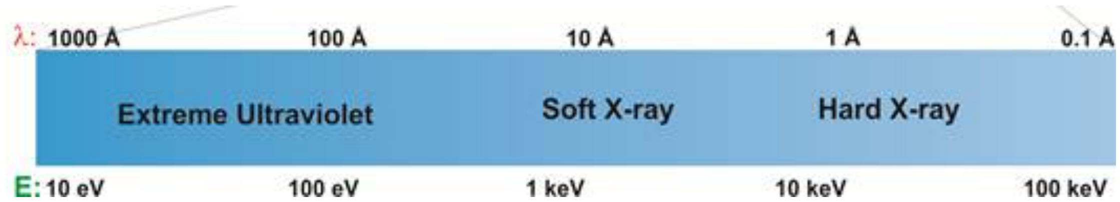

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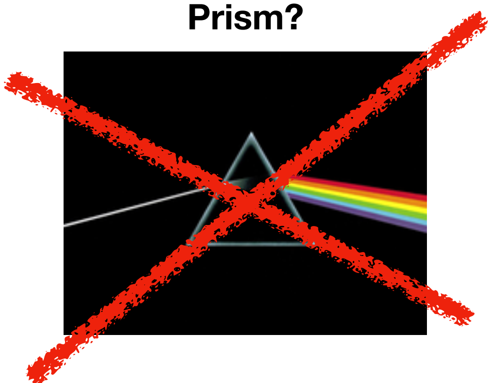

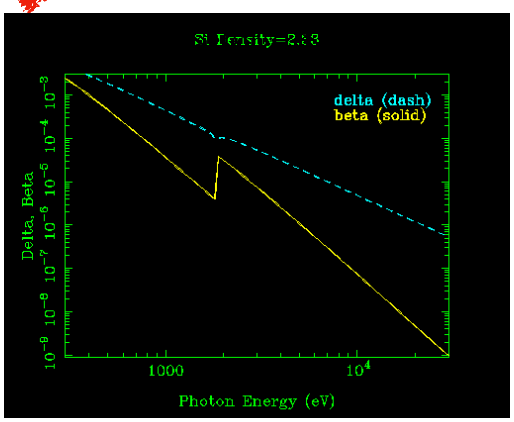

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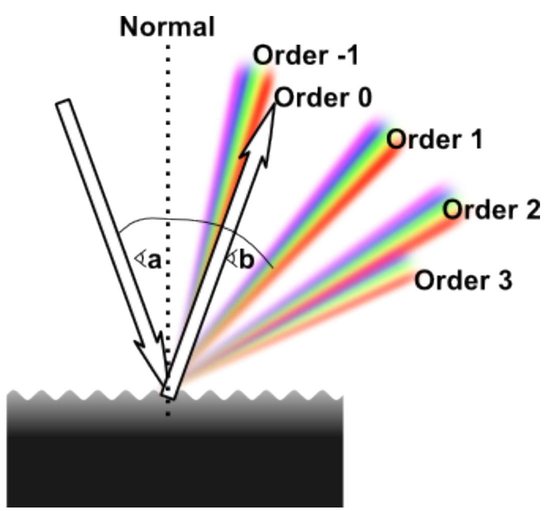
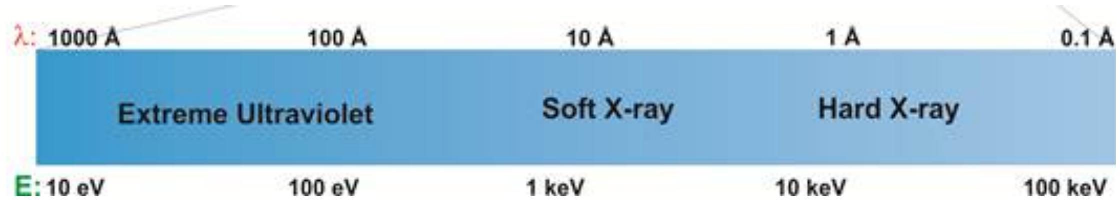
Grating?



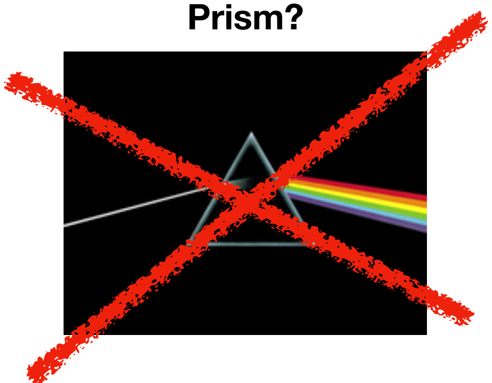
If feature sizes can be on order wavelength

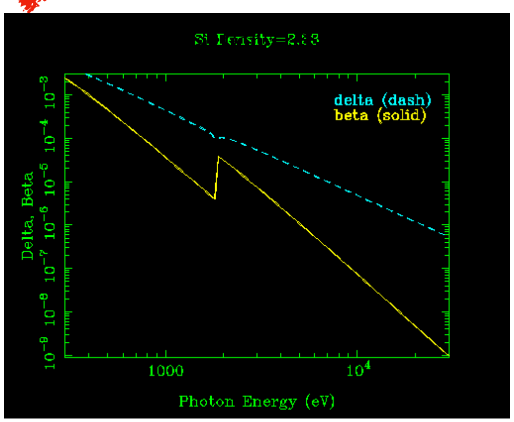
Good choice for lasers up to soft X-ray ~ 1 nm

How can we disperse X-rays according to energy/wavelength?



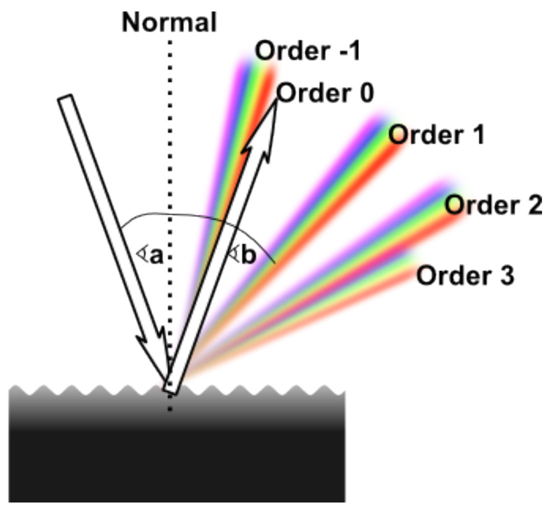
Prism?





Inefficient
Index changes too small to be effective

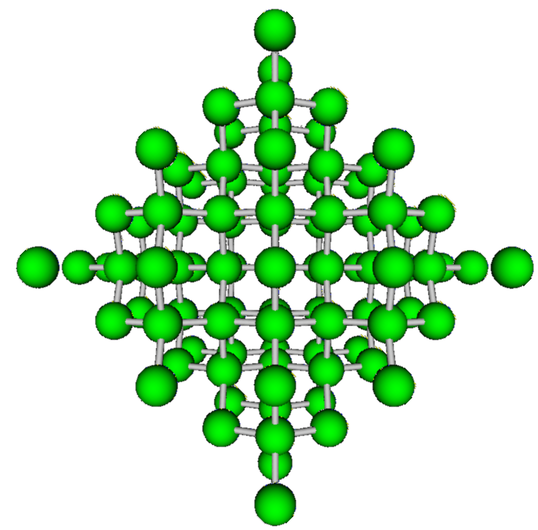
Grating?



If feature sizes can be on order wavelength

Good choice for lasers up to soft X-ray ~ 1 nm

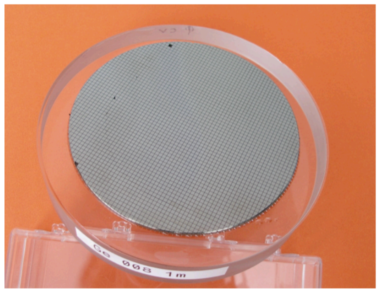
Crystal?



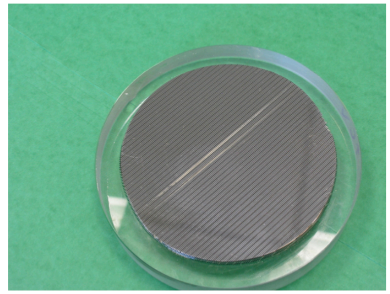
Feature size (atoms) of order X-ray wavelength

Hard X-ray << 1nm

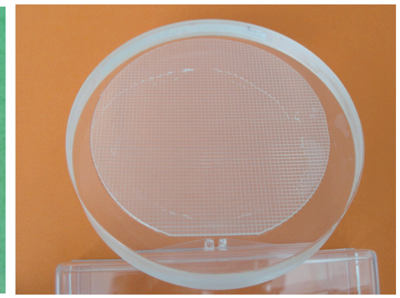
Diffracting X-rays crystals



Si



Ge

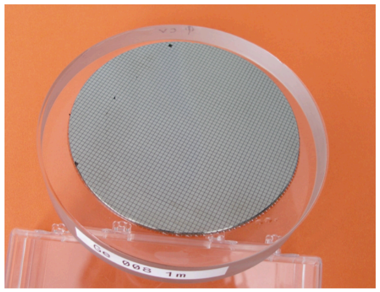


$LiNbO_3$

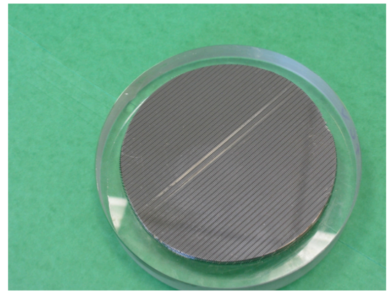
- Collecting inelastically scattered photons requires high quality, large, diced, bent crystal analyzers

- APS is a world leader in analyzer development

Diffracting X-rays crystals



Si



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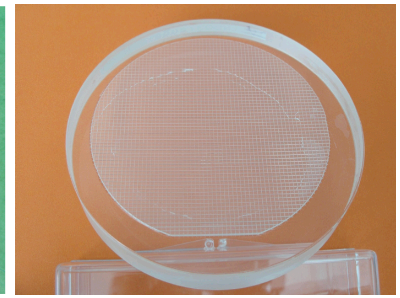
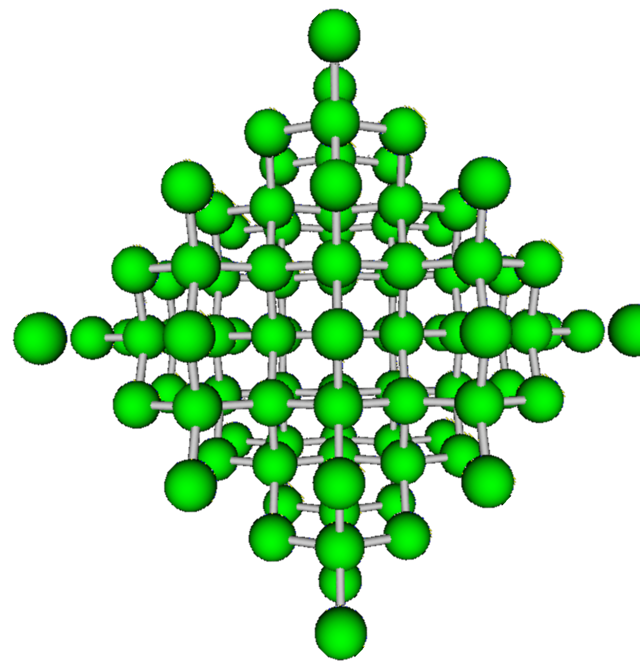


$LiNbO_3$



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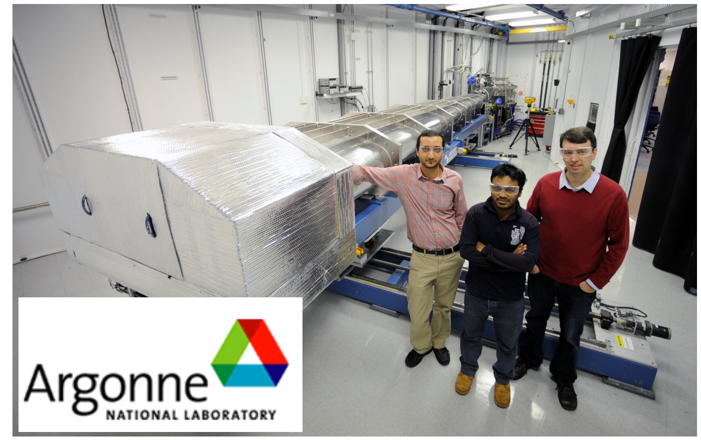
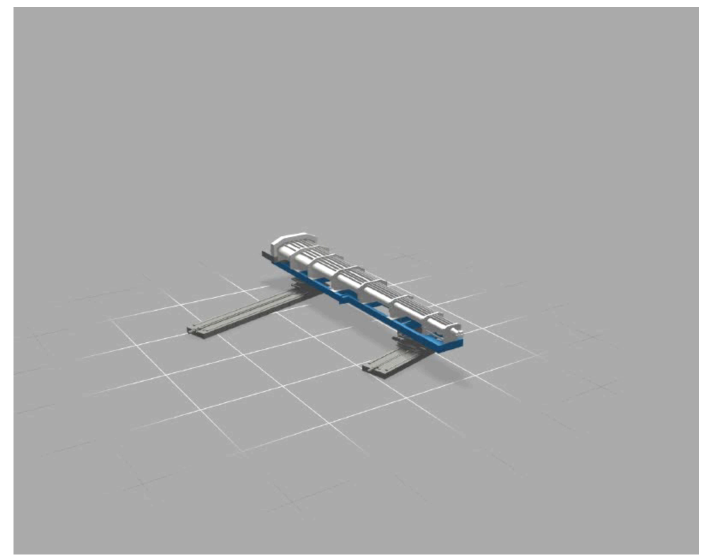
High **E**nergy **R**esolution **I**nelastic **X**-ray Scattering

- Sector 30, Advanced Photon Source, Argonne National Lab
- 23,724 eV incident energy
- <1 meV incident bandwidth
- Resolving power $E_i/\Delta E_i = 2\times10^7$
- 9 analyzers sample, 9 momenta transfers simultaneously, 9m arm
- 1.5 meV energy resolution
- 20μm x 5μm spot size
- Measures energy and momentum distribution of lattice vibrations $S(\vec{q},\omega)$





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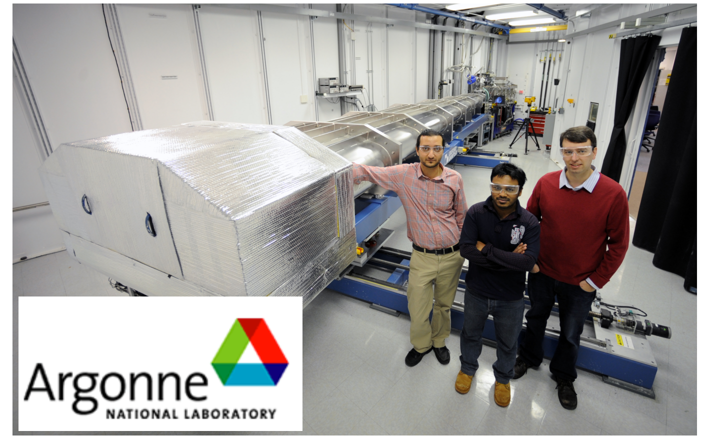
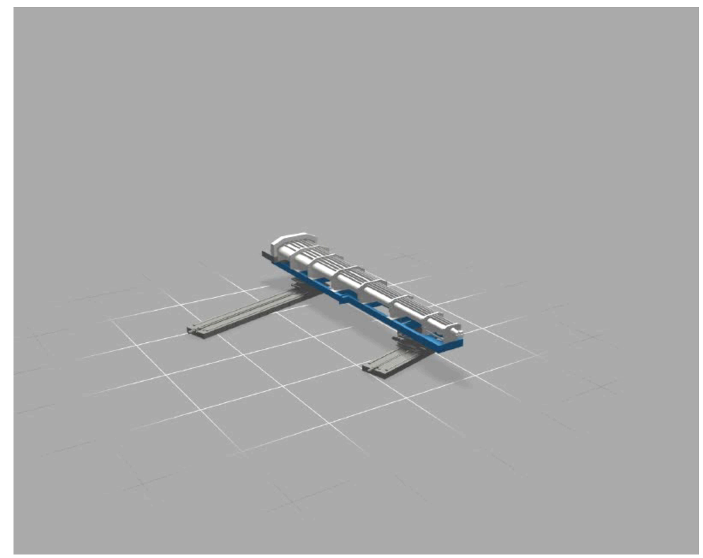
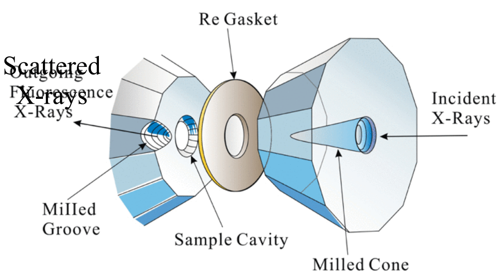


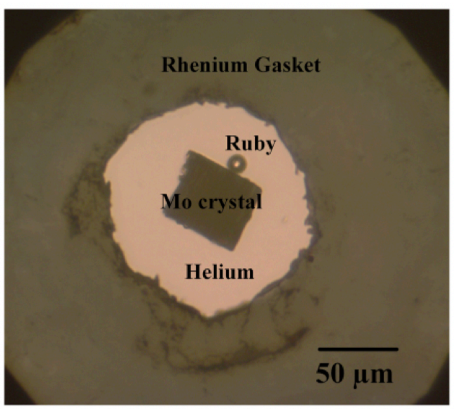


PHYSICAL REVIEW B **105**, 094105 (2022)

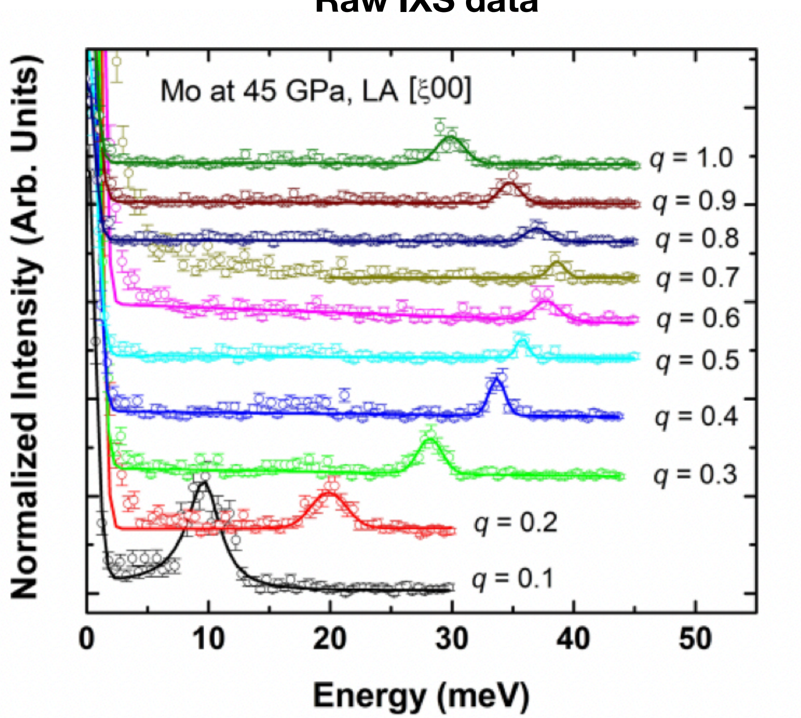
Kohn anomaly and elastic softening in body-centered cubic molybdenum at high pressure

Sample and environment

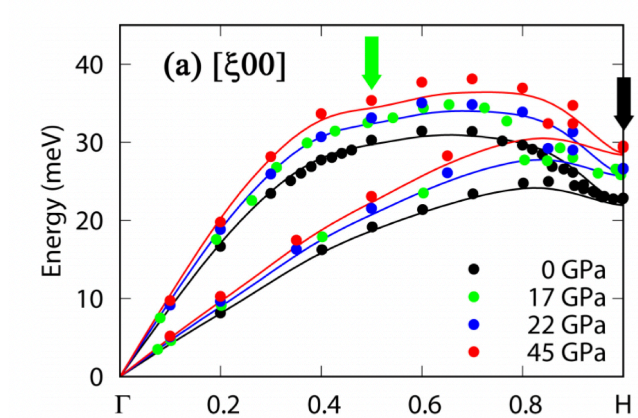




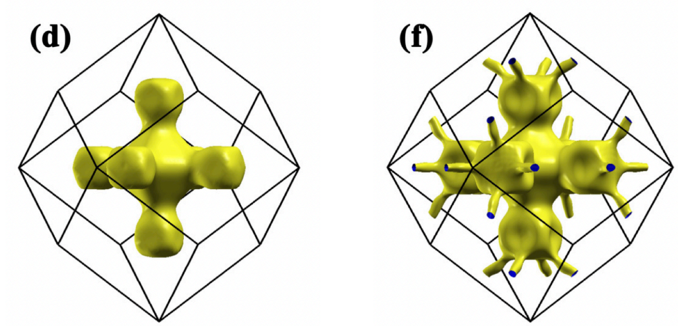
Raw IXS data



Pressure-dependent dispersion



Interpretation - fermi surface changes at high pressure

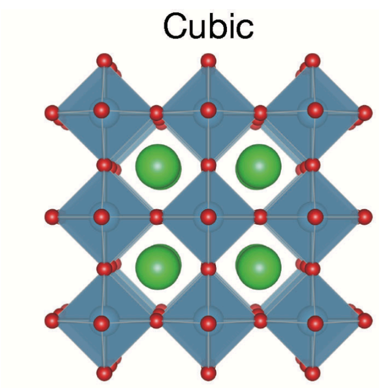


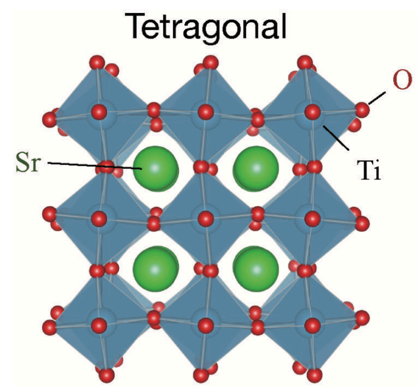
Low pressure FS *High pressure FS*

PHYSICAL REVIEW B **78**, 104121 (2008)

Central peak and narrow component in x-ray scattering measurements near the displacive phase transition in $SrTiO_3$

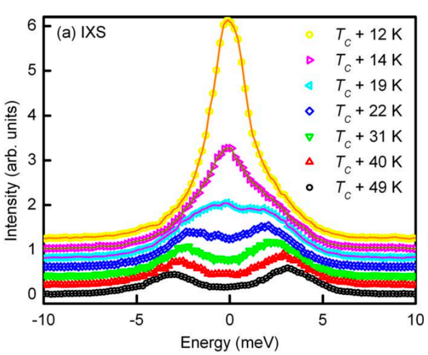
- Structural transitions can be described as the “freezing” of a phonon mode

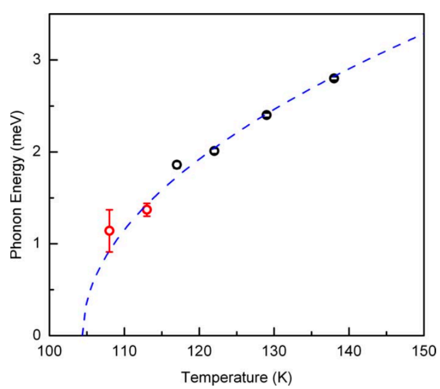




High temperature structure

Low temperature structure

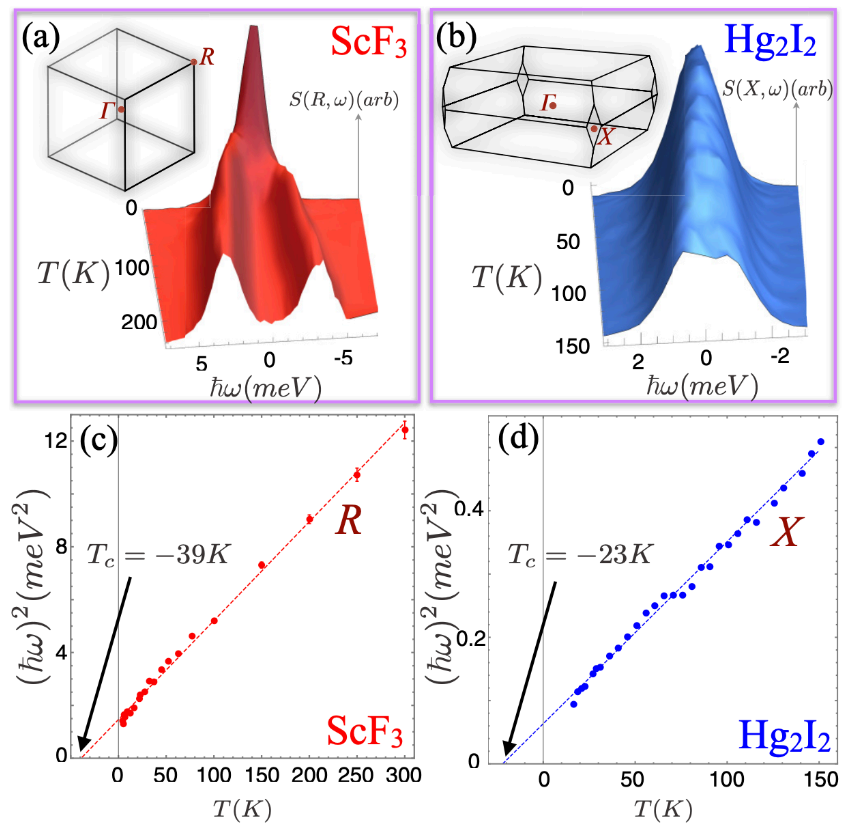


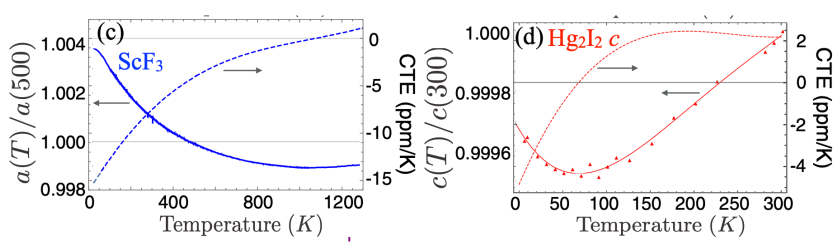


Journal of the Ceramic Society of Japan 127(6):404-408 (2019)

PHYSICAL REVIEW MATERIALS **1**, 070603(R) (2017)

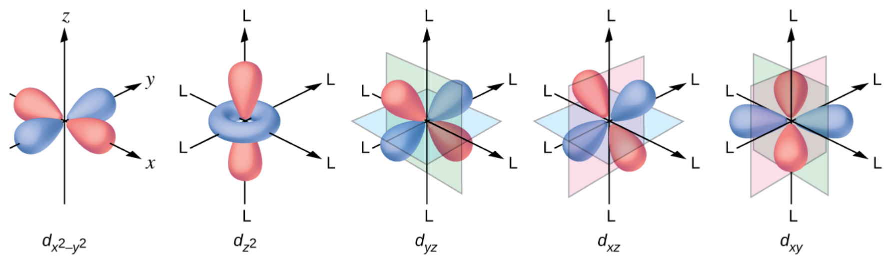
Negative thermal expansion near two structural quantum phase transitions

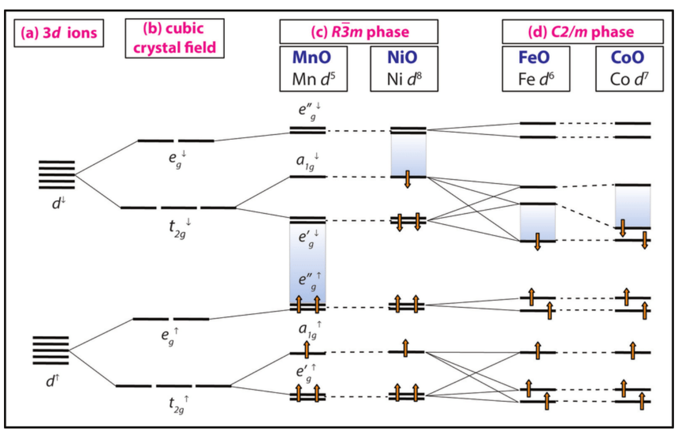


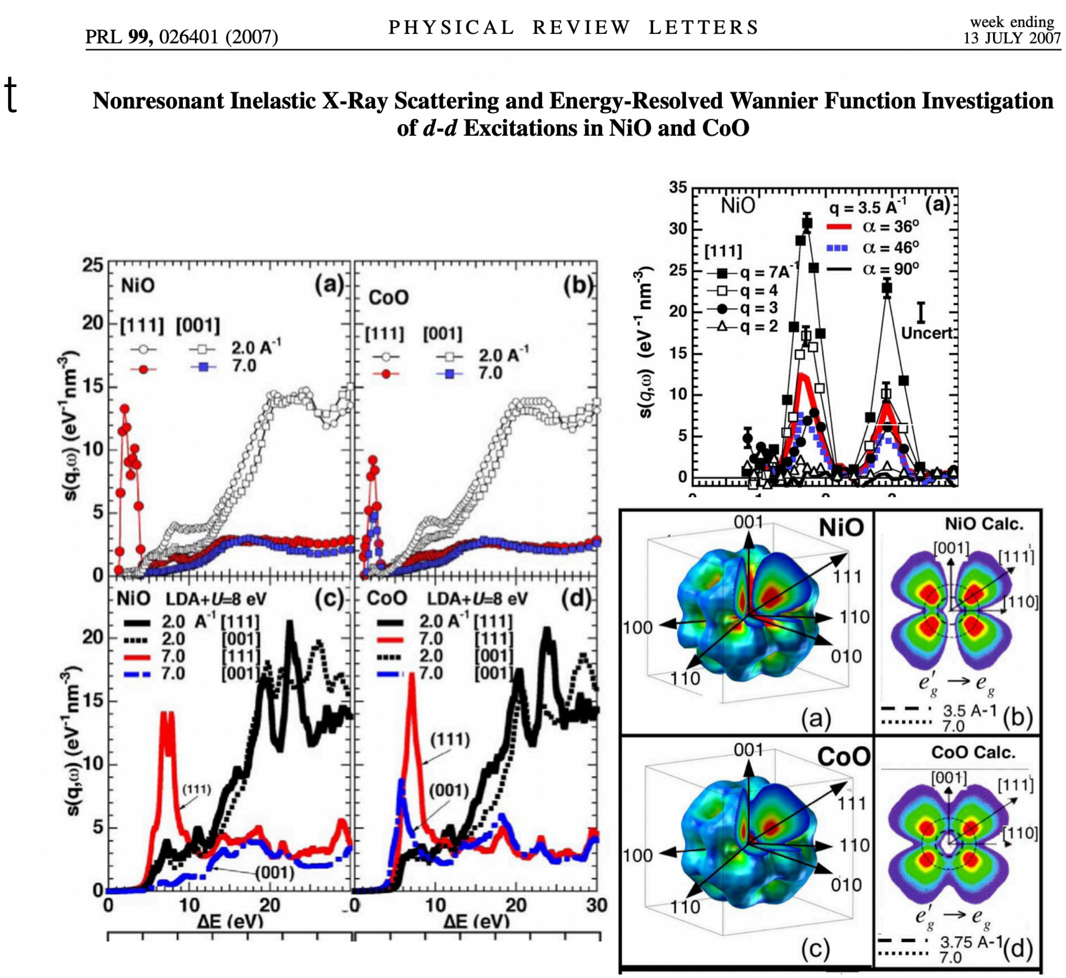


Non-phonon IXS-active excitations

- Different d electron orbitals have different energy when placed in a crystal
- Gives rise to "crystal field excitations"







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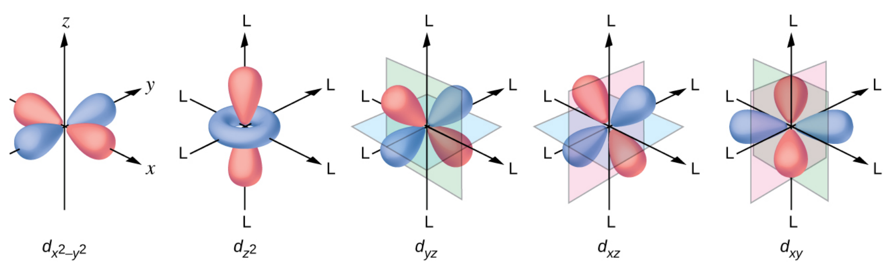
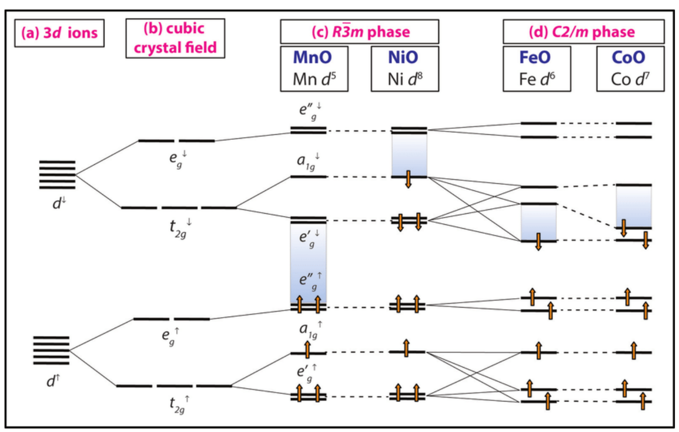
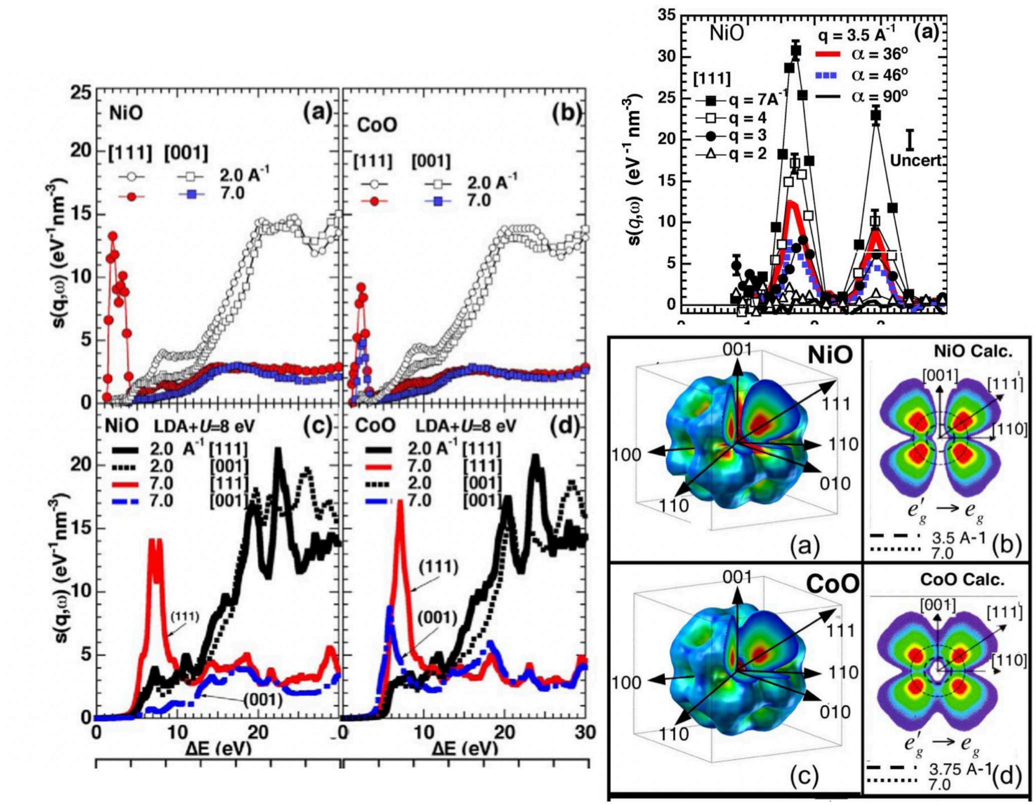




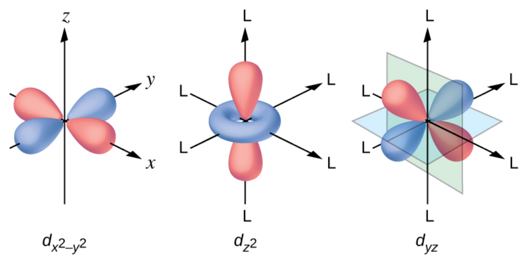
PRL 99, 026401 (2007) PHYSICAL REVIEW LETTERS week ending 13 JULY 2007

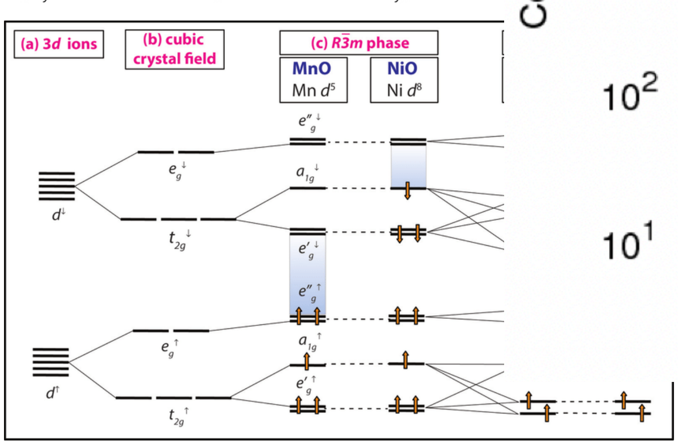
Nonresonant Inelastic X-Ray Scattering and Energy-Resolved Wannier Function Investigation of *d-d* Excitations in NiO and CoO

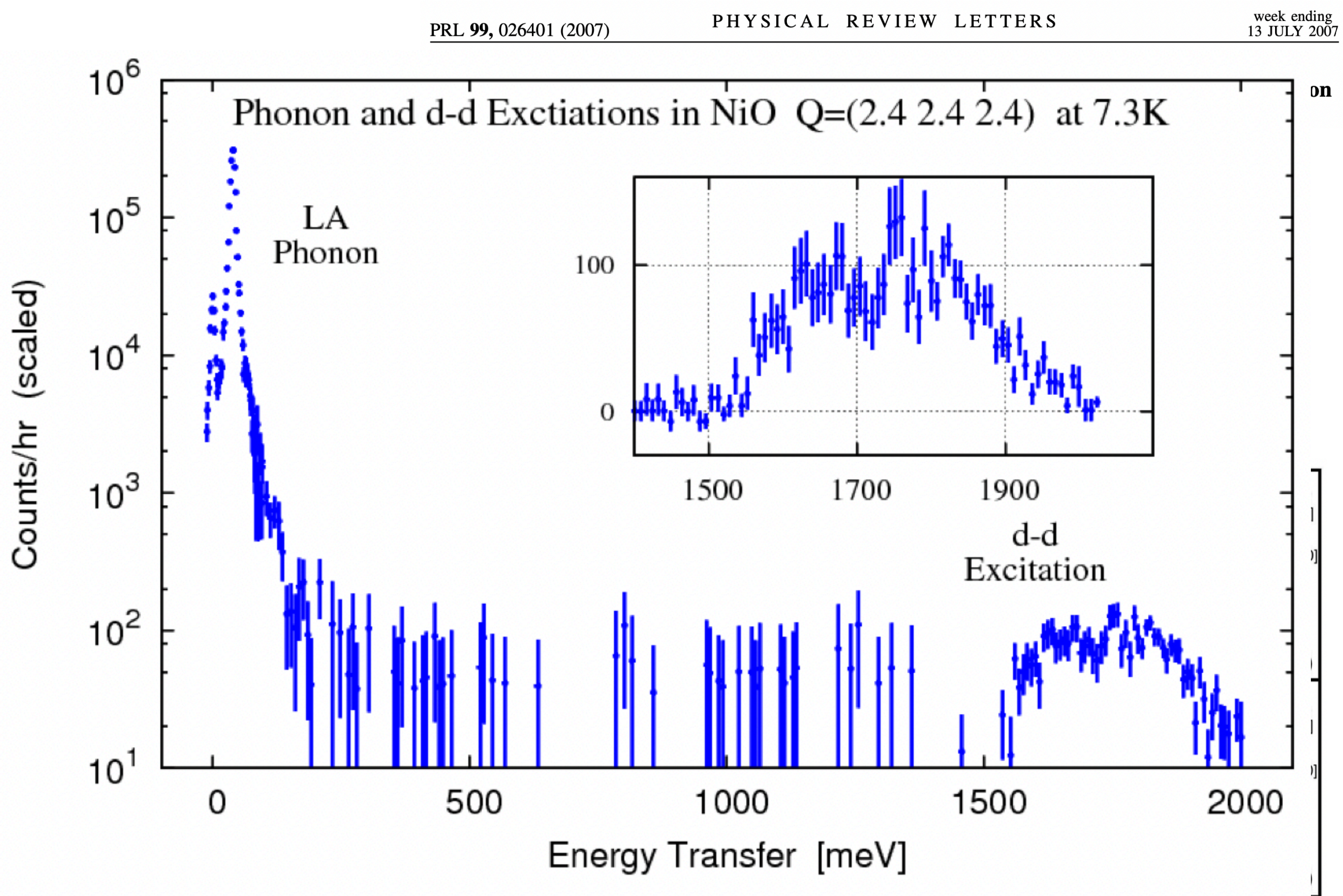


Non-phonon IXS-active excitations

- Different d electron orbit… energy when placed in a…
- Gives rise to "crystal fiel…







arXiv:1504.01098v7

Thank you!

Feedback

Lecture – 8:30 – 9:30
Inelastic X-ray Scattering - Jason Hancock
https://forms.office.com/g/mXiC92R1B4