

Plasmons in interacting arrays of metallic nanoparticles

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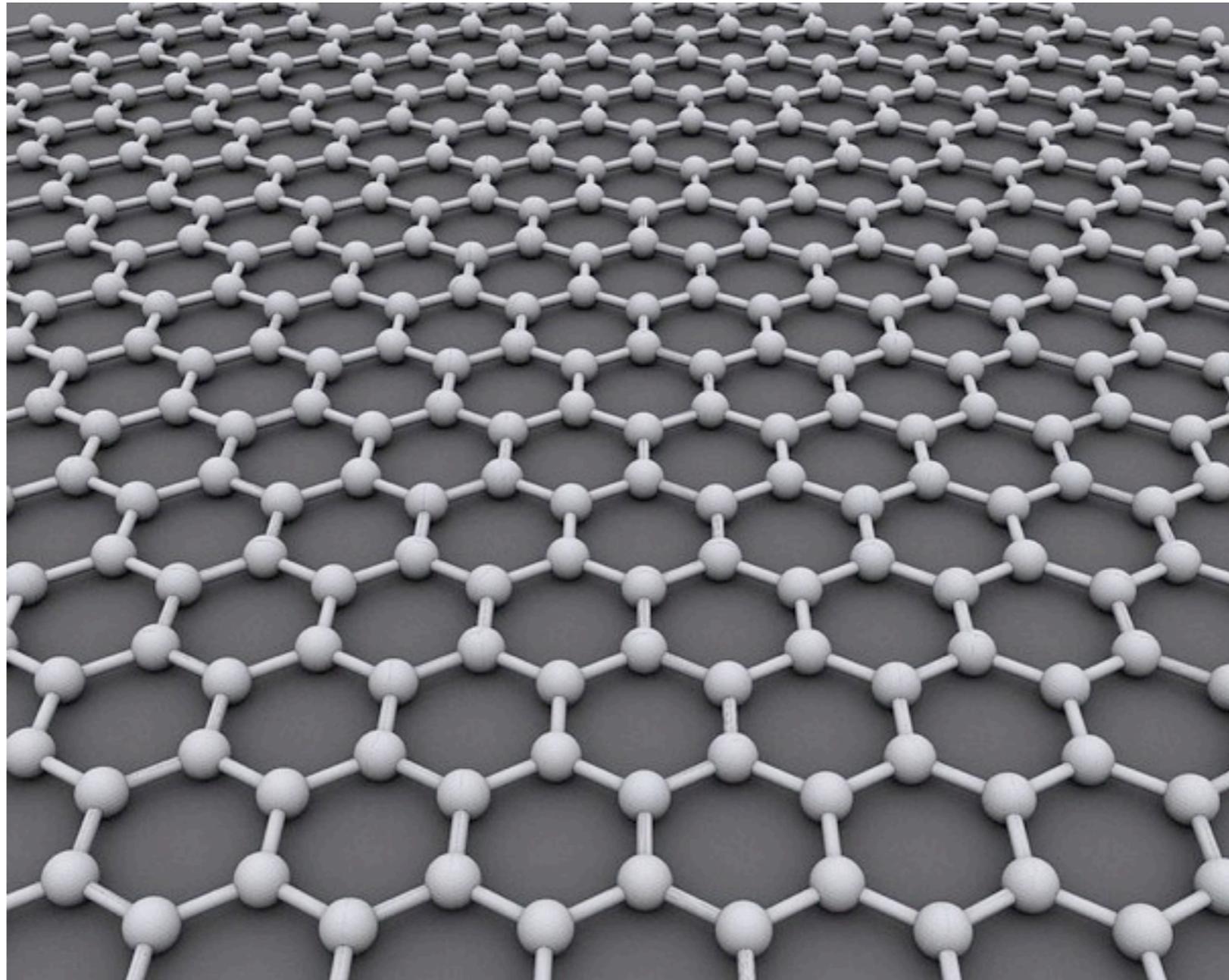


Bill Barnes (University of Exeter)



Ortwin Hess (Imperial College London)

Graphene

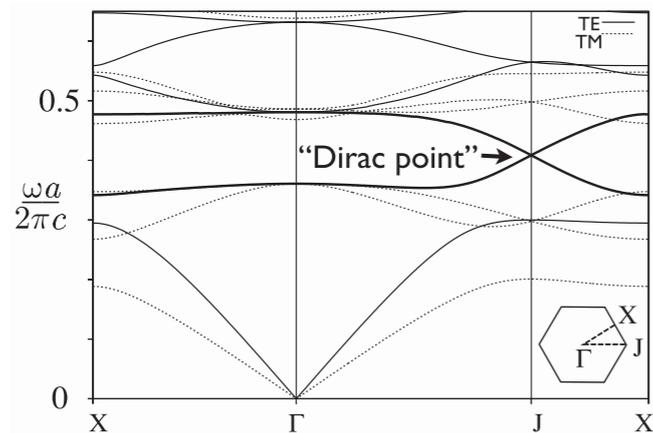
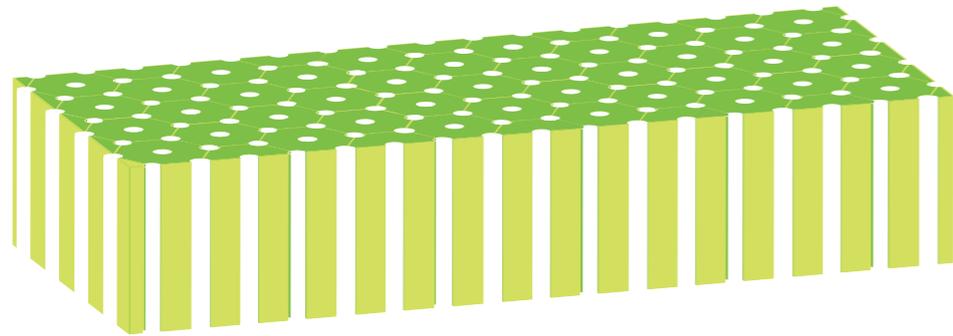


electrons behave as massless Dirac fermions
(honeycomb lattice + Bloch theorem)

Artificial graphene

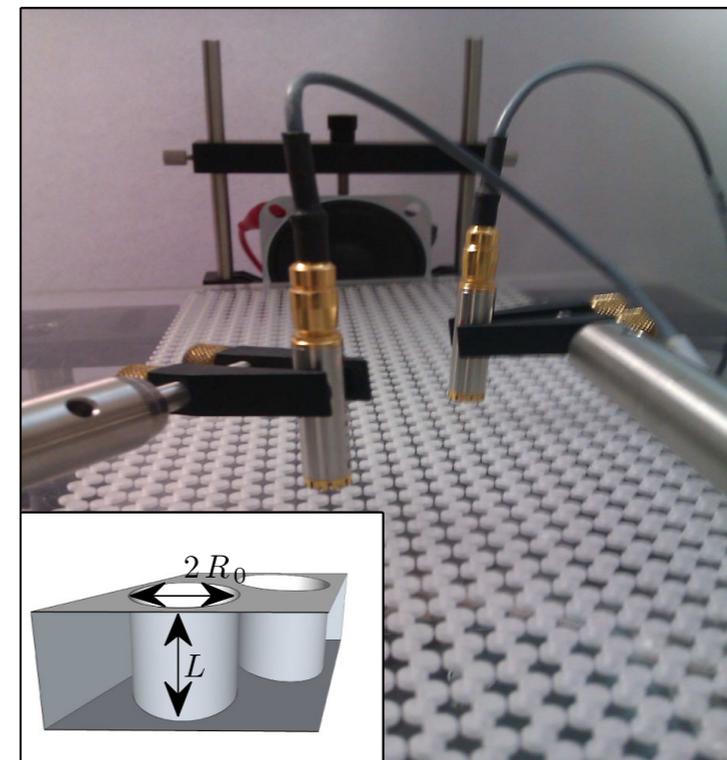
Photonic crystals

Haldane & Raghu, PRL 2008
 Sepkhanov, Bazaliy, Beenakker, PRA 2007



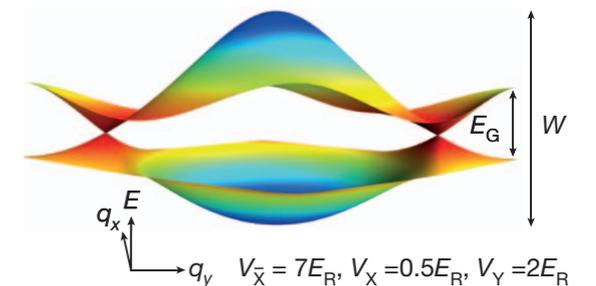
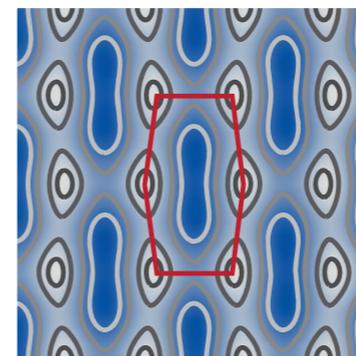
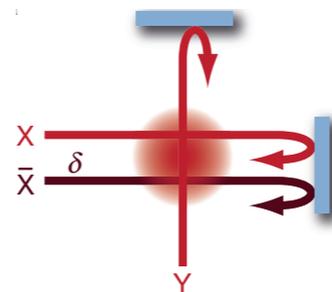
Acoustic waves

Torrent & Sánchez-Dehesa, PRL 2012



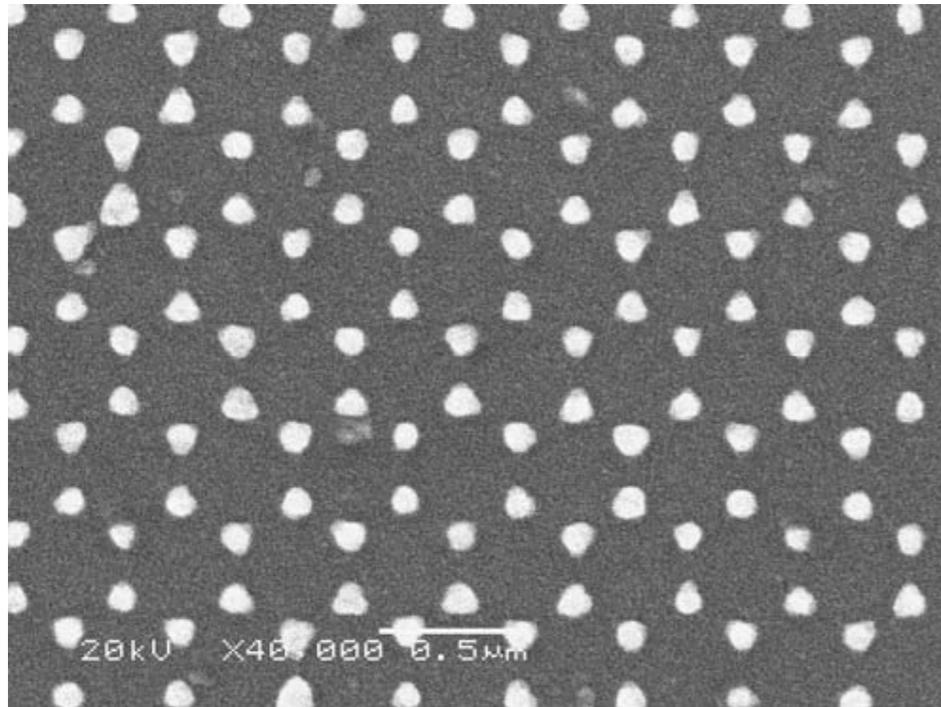
Cold atoms

Esslinger group, Nature 2012

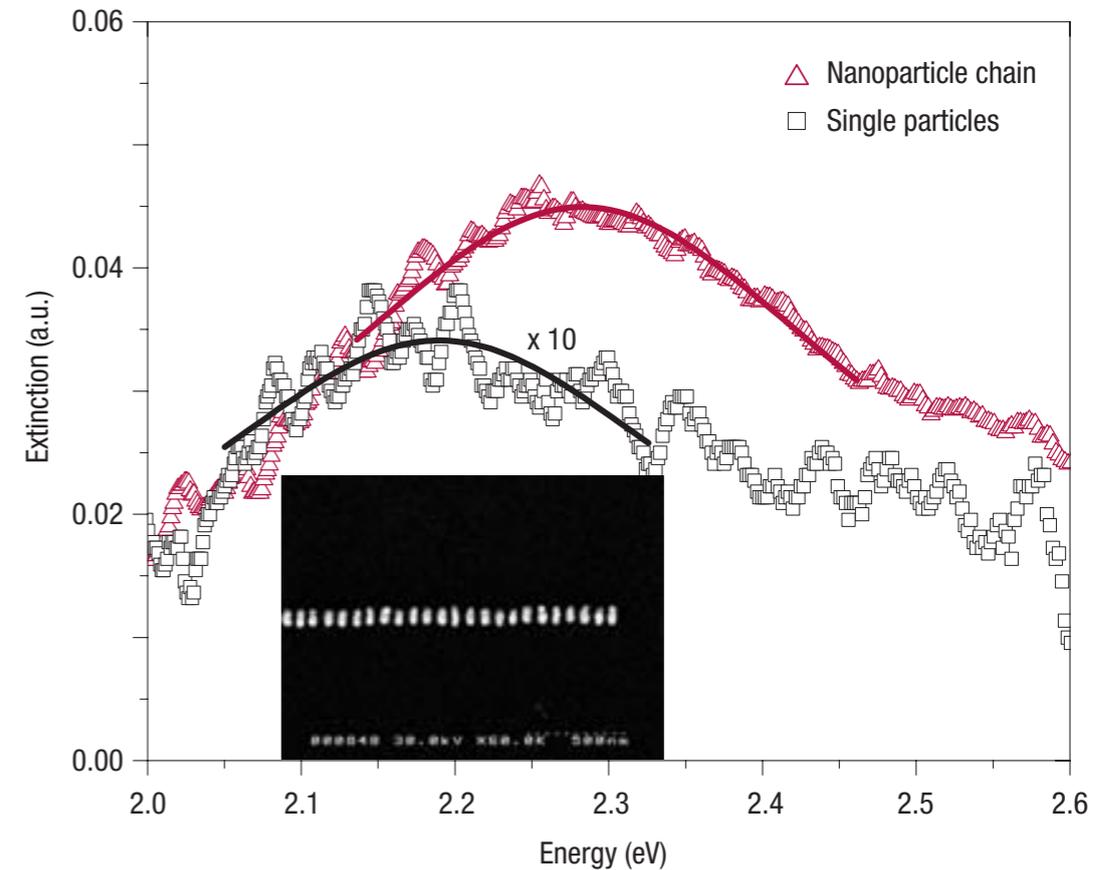


Plasmonic analogue of graphene

Plasmonic metamaterial

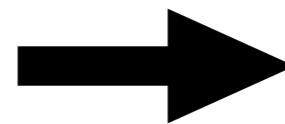


Zhu *et al.*, Plasmonics 2009
Han *et al.*, PRL 2009



Maier *et al.*, Nature Materials 2003

individual nanoparticle
localized surface plasmon



nanoparticle array
collective plasmon
(can propagate over macroscopic distances)

- ▶ subwavelength optics
- ▶ plasmonic "circuitry"

Individual nanoparticle

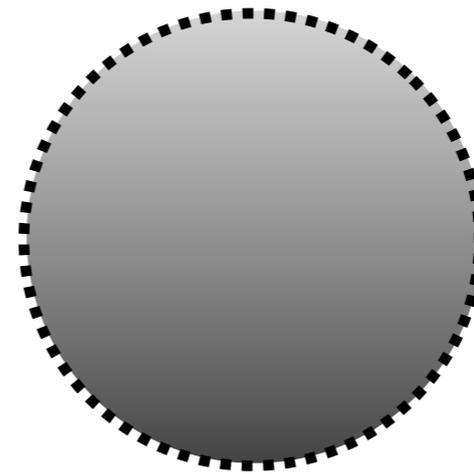
Localized surface plasmon:

➡ dipolar collective excitation of the electronic center of mass

$$\mathbf{E}(t) = \mathbf{E}_0 \cos(\omega t)$$



$$2r \ll \lambda$$

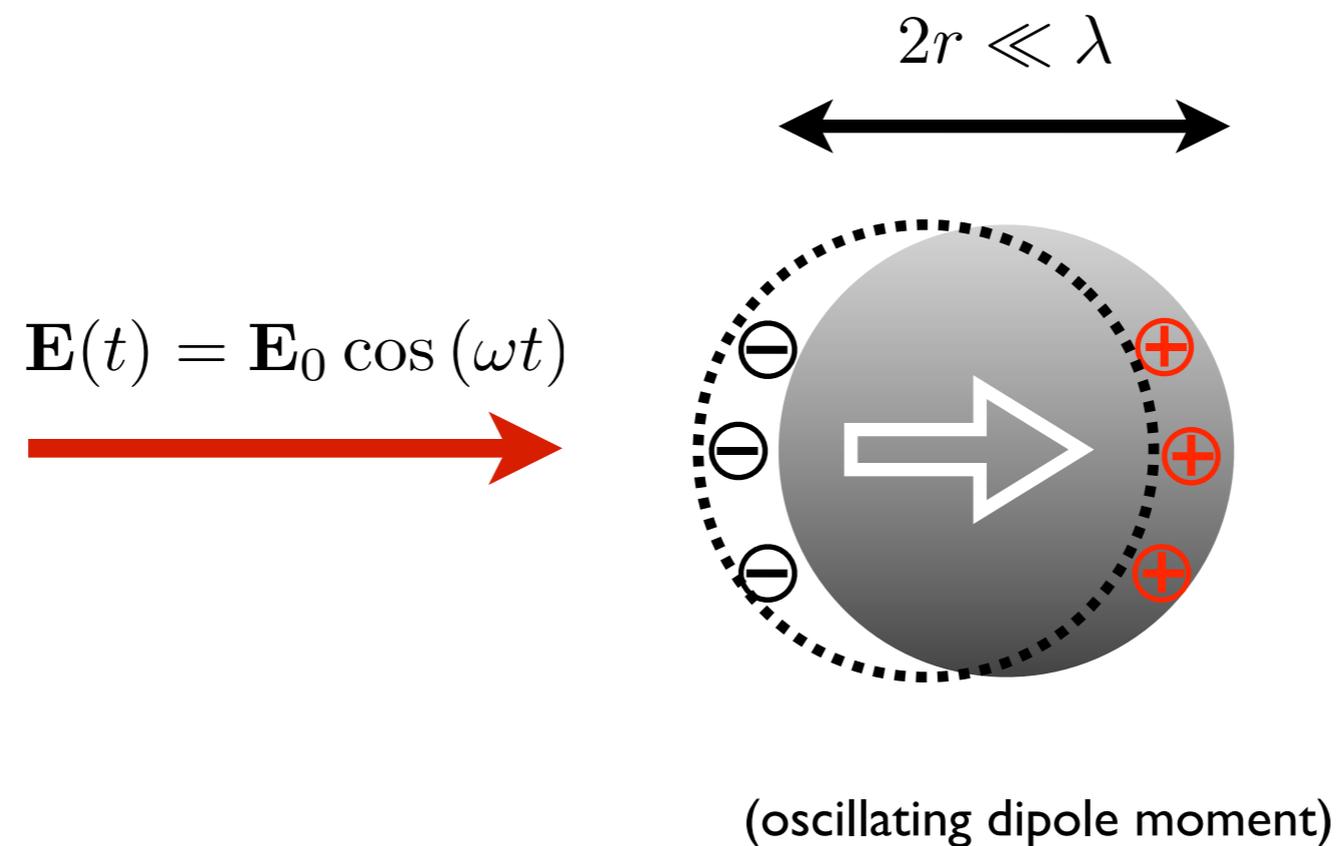


(oscillating dipole moment)

Individual nanoparticle

Localized surface plasmon:

➡ dipolar collective excitation of the electronic center of mass



Individual nanoparticle

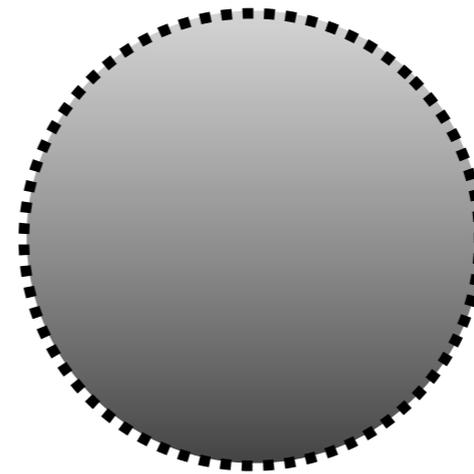
Localized surface plasmon:

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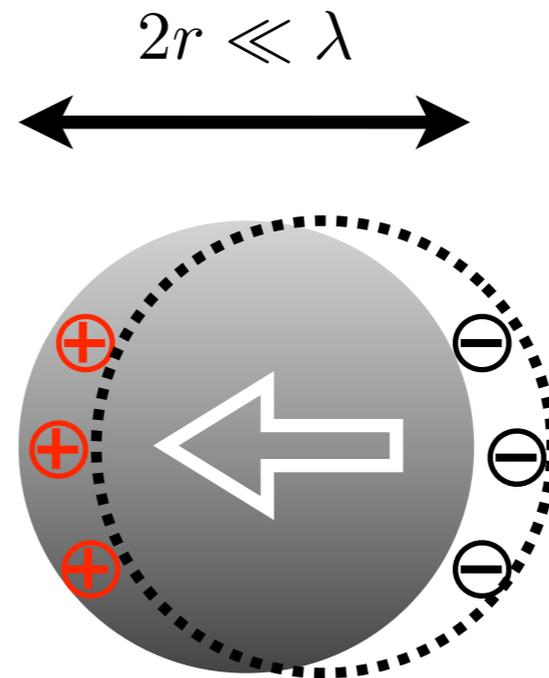
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Individual nanoparticle

Localized surface plasmon:

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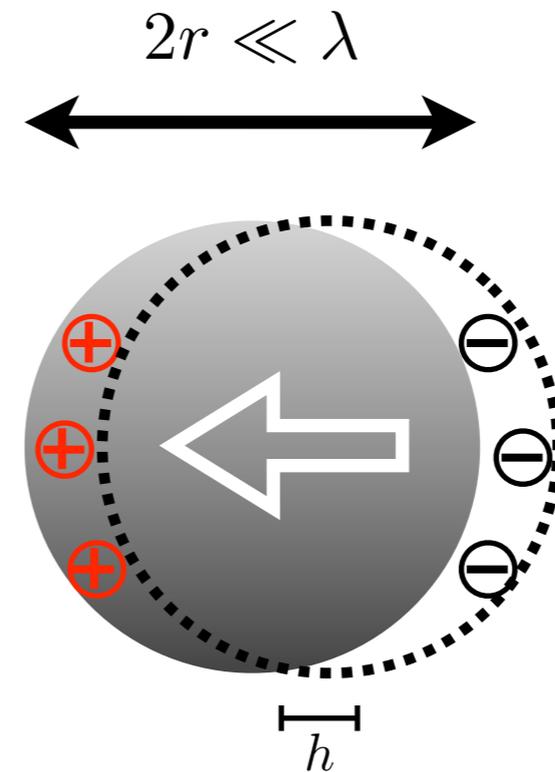
(oscillating dipole moment)

Individual nanoparticle

Localized surface plasmon:

➡ dipolar collective excitation of the electronic center of mass

$$\mathbf{E}(t) = \mathbf{E}_0 \cos(\omega t)$$



(oscillating dipole moment)

$$H_{\text{LSP}} = \frac{\Pi^2}{2M} + \frac{M}{2} \omega_0^2 h^2$$

Mie frequency: $\omega_0 = \frac{\omega_p}{\sqrt{1 + 2\epsilon_m}}$ (visible, near-IR)

plasma frequency: $\omega_p = \sqrt{\frac{4\pi n_e e^2}{m_e}}$

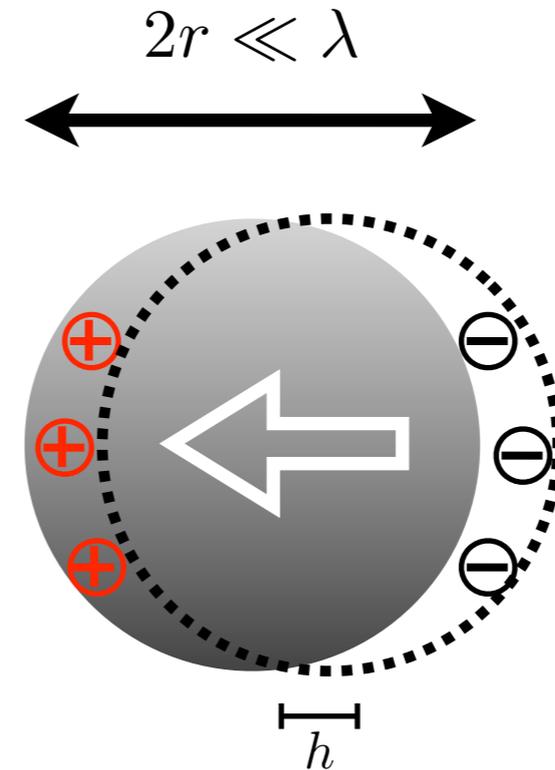
$$M = N_e m_e$$

Individual nanoparticle

Localized surface plasmon:

➡ dipolar collective excitation of the electronic center of mass

$$\mathbf{E}(t) = \mathbf{E}_0 \cos(\omega t)$$



(oscillating dipole moment)

$$b \sim h + i\Pi$$

$$H_{\text{LSP}} = \hbar\omega_0 \left(b^\dagger b + \frac{1}{2} \right)$$

Mie frequency: $\omega_0 = \frac{\omega_p}{\sqrt{1 + 2\epsilon_m}}$ (visible, near-IR)

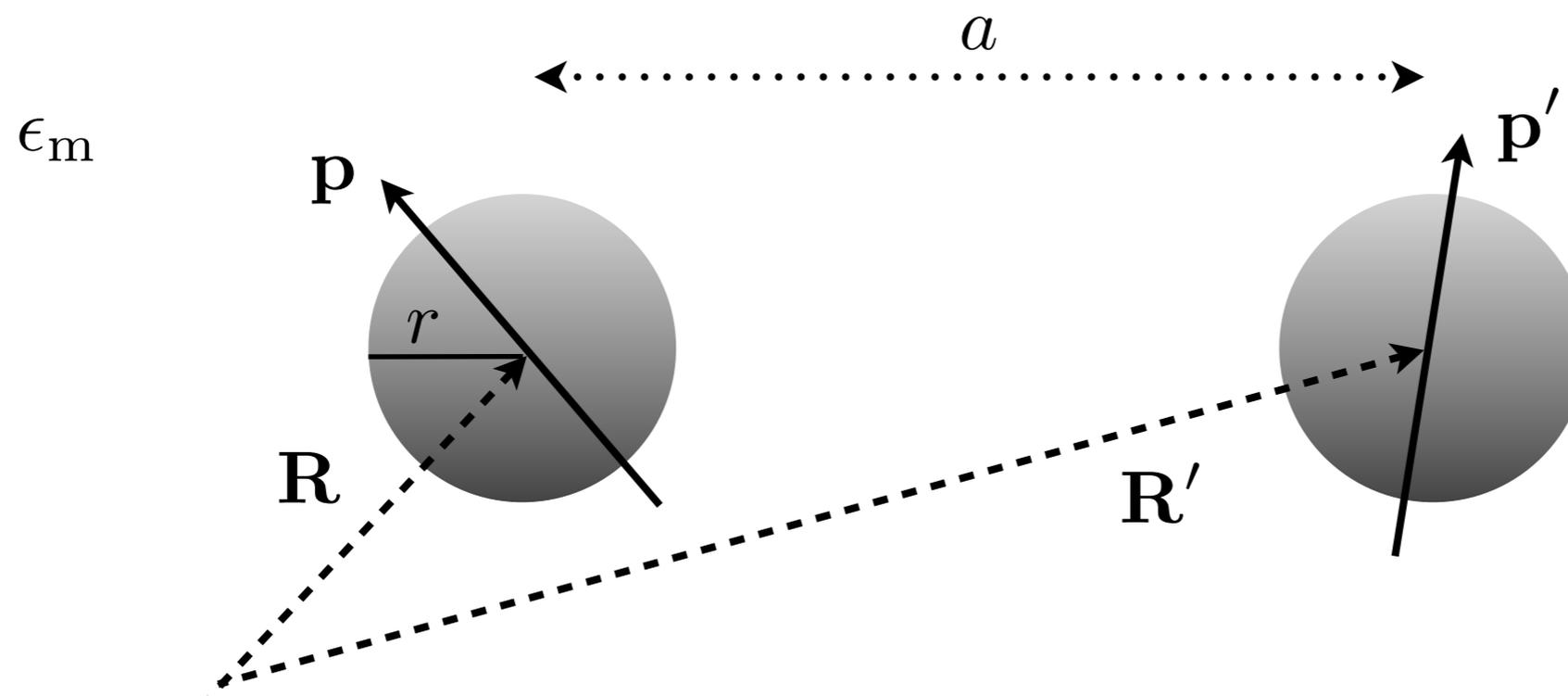
plasma frequency: $\omega_p = \sqrt{\frac{4\pi n_e e^2}{m_e}}$

$$M = N_e m_e$$

Two nanoparticles

Dipole-dipole interaction:

➡ quasistatic approximation for point-like dipoles ($r \lesssim a/3 \ll \lambda$)

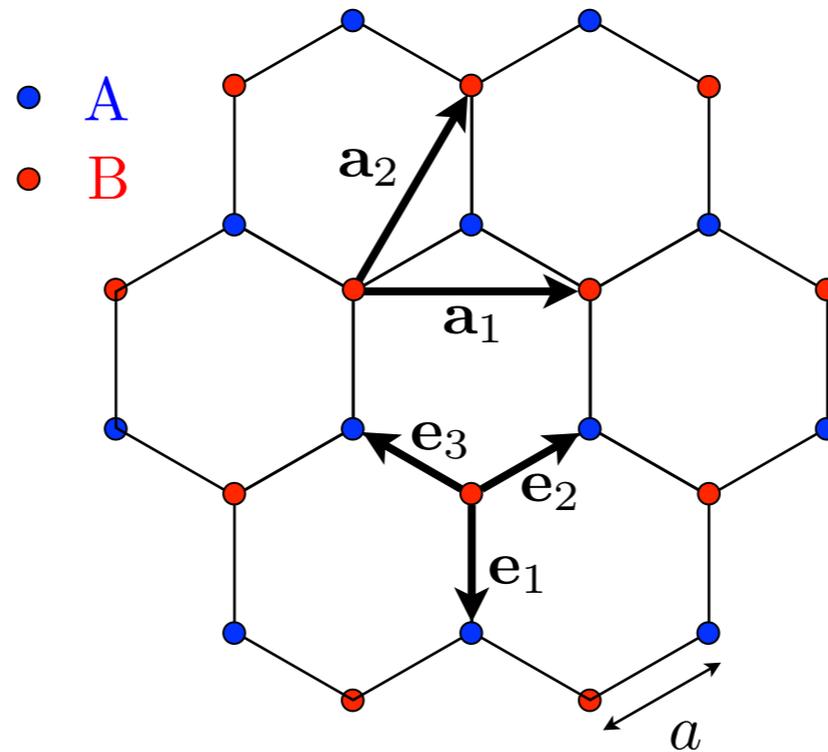


$$\mathcal{V} = \frac{\mathbf{p} \cdot \mathbf{p}' - 3(\mathbf{p} \cdot \mathbf{n})(\mathbf{p}' \cdot \mathbf{n})}{\epsilon_m |\mathbf{R} - \mathbf{R}'|^3}$$

$$\mathbf{p} = -eN_e h(\mathbf{R}) \hat{\mathbf{p}}$$

$$\mathbf{n} = \frac{\mathbf{R} - \mathbf{R}'}{|\mathbf{R} - \mathbf{R}'|}$$

Honeycomb plasmonic lattice:



$$H_0 = \sum_{s=A,B} \sum_{\mathbf{R}_s} \left[\frac{\Pi_s^2(\mathbf{R}_s)}{2M} + \frac{M}{2} \omega_0^2 h_s^2(\mathbf{R}_s) \right]$$

$$H_{\text{int}} = \frac{(eN_e)^2}{\epsilon_m a^3} \sum_{\mathbf{R}_B} \sum_{j=1}^3 C_j h_B(\mathbf{R}_B) h_A(\mathbf{R}_B + \mathbf{e}_j)$$

(θ, φ) : polarization of the dipoles

$$C_j = 1 - 3 \sin^2 \theta \cos^2 (\varphi - 2\pi[j - 1]/3)$$

► nearest-neighbor interactions only

Analogy with electrons in graphene

Bosonic ladder operators:

$$H_{\text{int}} = \hbar\Omega \sum_{\mathbf{R}_B} \sum_{j=1}^3 C_j b_{\mathbf{R}_B}^\dagger \left(a_{\mathbf{R}_B + \mathbf{e}_j} + a_{\mathbf{R}_B + \mathbf{e}_j}^\dagger \right) + \text{H.c.}$$

$$a_{\mathbf{R}} = \sqrt{\frac{M\omega_0}{2\hbar}} h_A(\mathbf{R}) + \frac{i\Pi_A(\mathbf{R})}{\sqrt{2\hbar M\omega_0}}$$

$$b_{\mathbf{R}} = \sqrt{\frac{M\omega_0}{2\hbar}} h_B(\mathbf{R}) + \frac{i\Pi_B(\mathbf{R})}{\sqrt{2\hbar M\omega_0}}$$

$$\Omega = \omega_0 \left(\frac{r}{a}\right)^3 \frac{1 + 2\epsilon_m}{6\epsilon_m} \ll \omega_0$$

➔ cf. tight-binding Hamiltonian for electrons in graphene!

Graphene	Plasmonic graphene
fermions (electrons)	bosons (plasmons)
AB sublattices linked by kinetic process (hopping of electrons)	AB sublattices linked by <i>interactions</i> (dipole-dipole)
equal hopping matrix elements t	<i>tunable</i> couplings $\hbar\Omega C_j$ (cf. strained graphene)
∅	$H_0 = \hbar\omega_0 \sum_{\mathbf{R}_A} a_{\mathbf{R}_A}^\dagger a_{\mathbf{R}_A} + \hbar\omega_0 \sum_{\mathbf{R}_B} b_{\mathbf{R}_B}^\dagger b_{\mathbf{R}_B}$
∅	anomalous term $\propto b_{\mathbf{R}_B}^\dagger a_{\mathbf{R}_B + \mathbf{e}_j}^\dagger$

Exact diagonalization

Starting Hamiltonian:

$$H = \hbar\omega_0 \sum_{\mathbf{q}} (a_{\mathbf{q}}^\dagger a_{\mathbf{q}} + b_{\mathbf{q}}^\dagger b_{\mathbf{q}}) + \hbar\Omega \sum_{\mathbf{q}} [f_{\mathbf{q}} b_{\mathbf{q}}^\dagger (a_{\mathbf{q}} + a_{-\mathbf{q}}^\dagger) + \text{H.c.}] \quad f_{\mathbf{q}} = \sum_{j=1}^3 C_j \exp(i\mathbf{q} \cdot \mathbf{e}_j)$$

Bogoliubov #1:

$$\alpha_{\mathbf{q}}^\pm = \frac{1}{\sqrt{2}} \left(\frac{f_{\mathbf{q}}}{|f_{\mathbf{q}}|} a_{\mathbf{q}} \pm b_{\mathbf{q}} \right)$$

$$\Rightarrow H = \sum_{\tau=\pm} \sum_{\mathbf{q}} \left[(\hbar\omega_0 + \tau\hbar\Omega|f_{\mathbf{q}}|) \alpha_{\mathbf{q}}^{\tau\dagger} \alpha_{\mathbf{q}}^\tau + \tau \frac{\hbar\Omega|f_{\mathbf{q}}|}{2} (\alpha_{\mathbf{q}}^{\tau\dagger} \alpha_{-\mathbf{q}}^{\tau\dagger} + \text{H.c.}) \right]$$

Bogoliubov #2:

$$\beta_{\mathbf{q}}^\pm = \cosh \vartheta_{\mathbf{q}}^\pm \alpha_{\mathbf{q}}^\pm - \sinh \vartheta_{\mathbf{q}}^\pm \alpha_{-\mathbf{q}}^{\pm\dagger}$$

$$\cosh \vartheta_{\mathbf{q}}^\pm = 2^{-1/2} [(1 \pm \Omega|f_{\mathbf{q}}|/\omega_0)/(1 \pm 2\Omega|f_{\mathbf{q}}|/\omega_0)^{1/2} + 1]^{1/2}$$

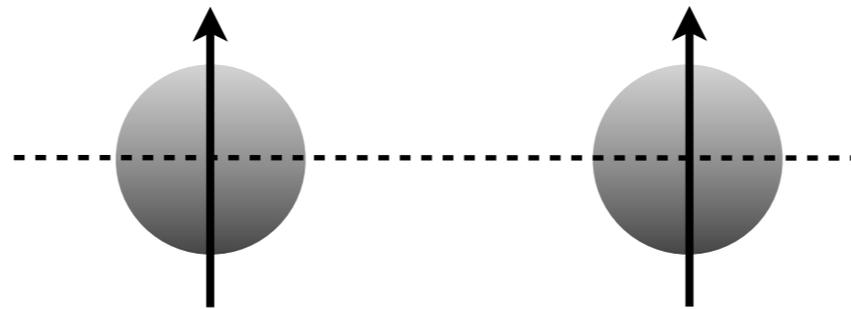
$$\sinh \vartheta_{\mathbf{q}}^\pm = \mp 2^{-1/2} [(1 \pm \Omega|f_{\mathbf{q}}|/\omega_0)/(1 \pm 2\Omega|f_{\mathbf{q}}|/\omega_0)^{1/2} - 1]^{1/2}$$

$$\Rightarrow H = \sum_{\tau=\pm} \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}}^\tau \beta_{\mathbf{q}}^{\tau\dagger} \beta_{\mathbf{q}}^\tau$$

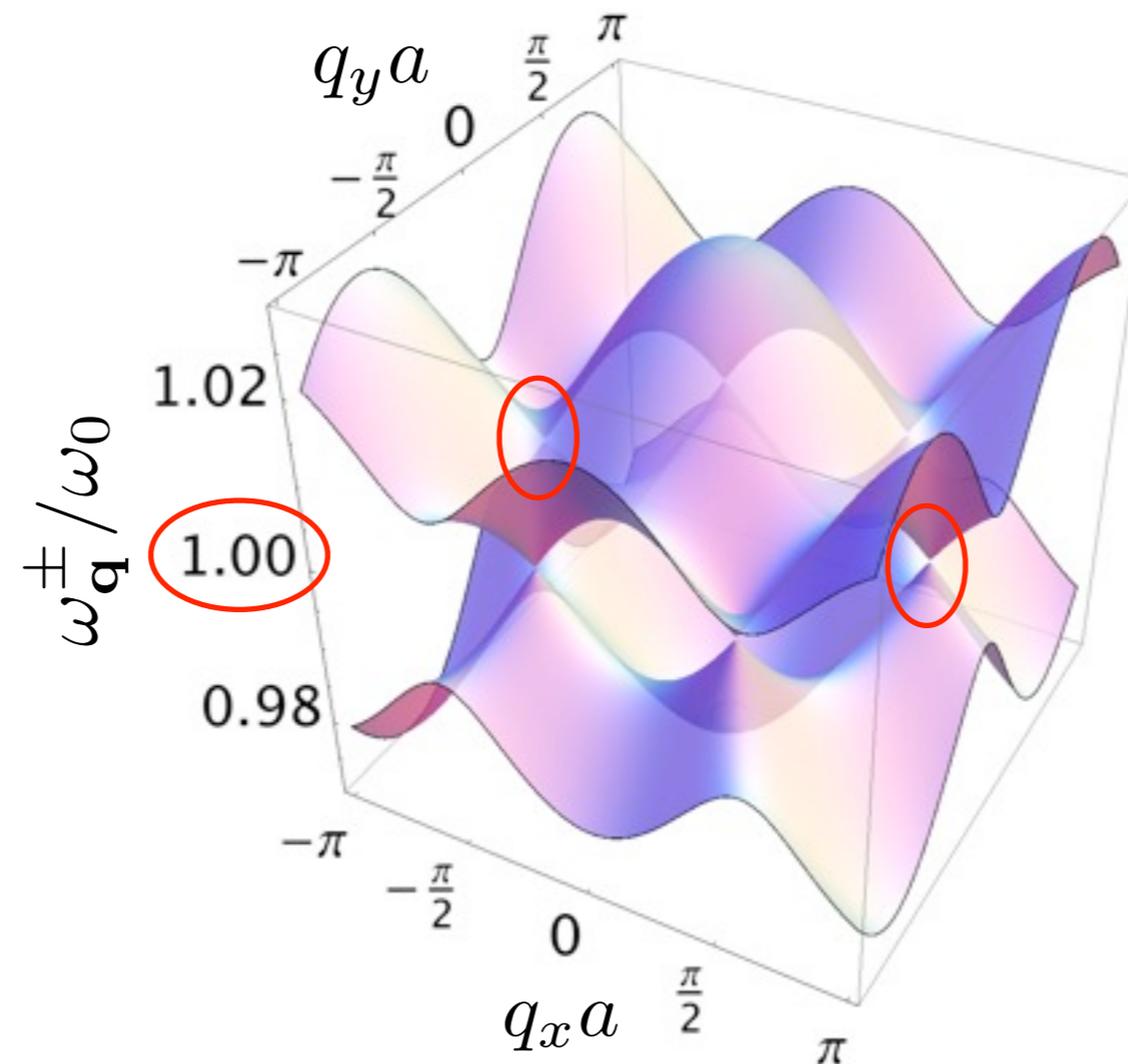
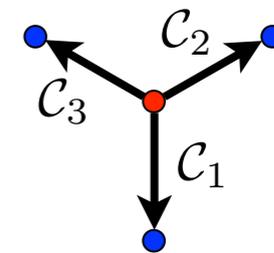
$$\omega_{\mathbf{q}}^\pm = \omega_0 \sqrt{1 \pm 2 \frac{\Omega}{\omega_0} |f_{\mathbf{q}}|}$$

Plasmon dispersion

$$\theta = 0$$



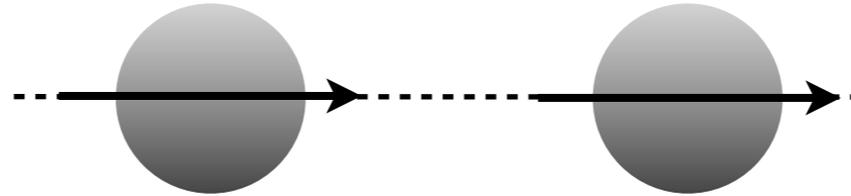
$$c_1 = c_2 = c_3 = 1$$



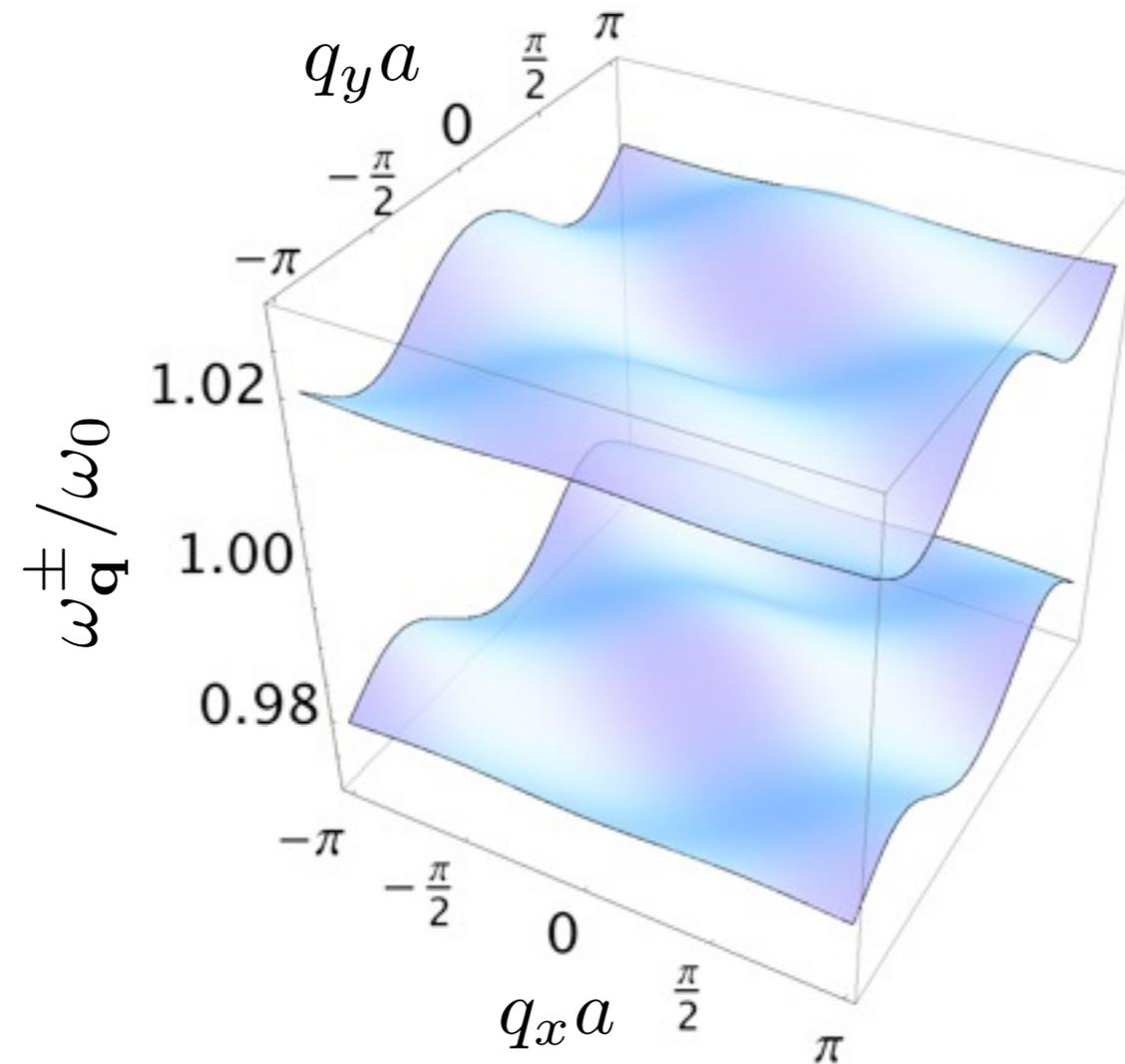
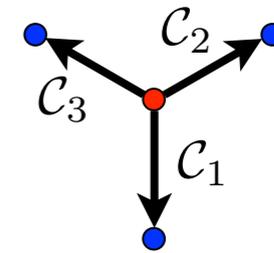
➔ two Dirac cones

Plasmon dispersion

$$\theta = \frac{\pi}{2}$$



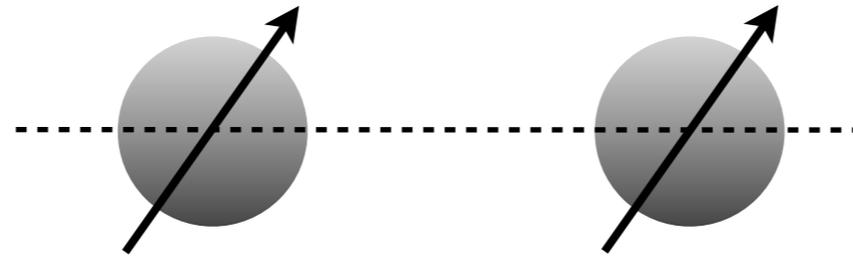
$$C_1 = -2, \quad C_2 = C_3 = 1/4$$



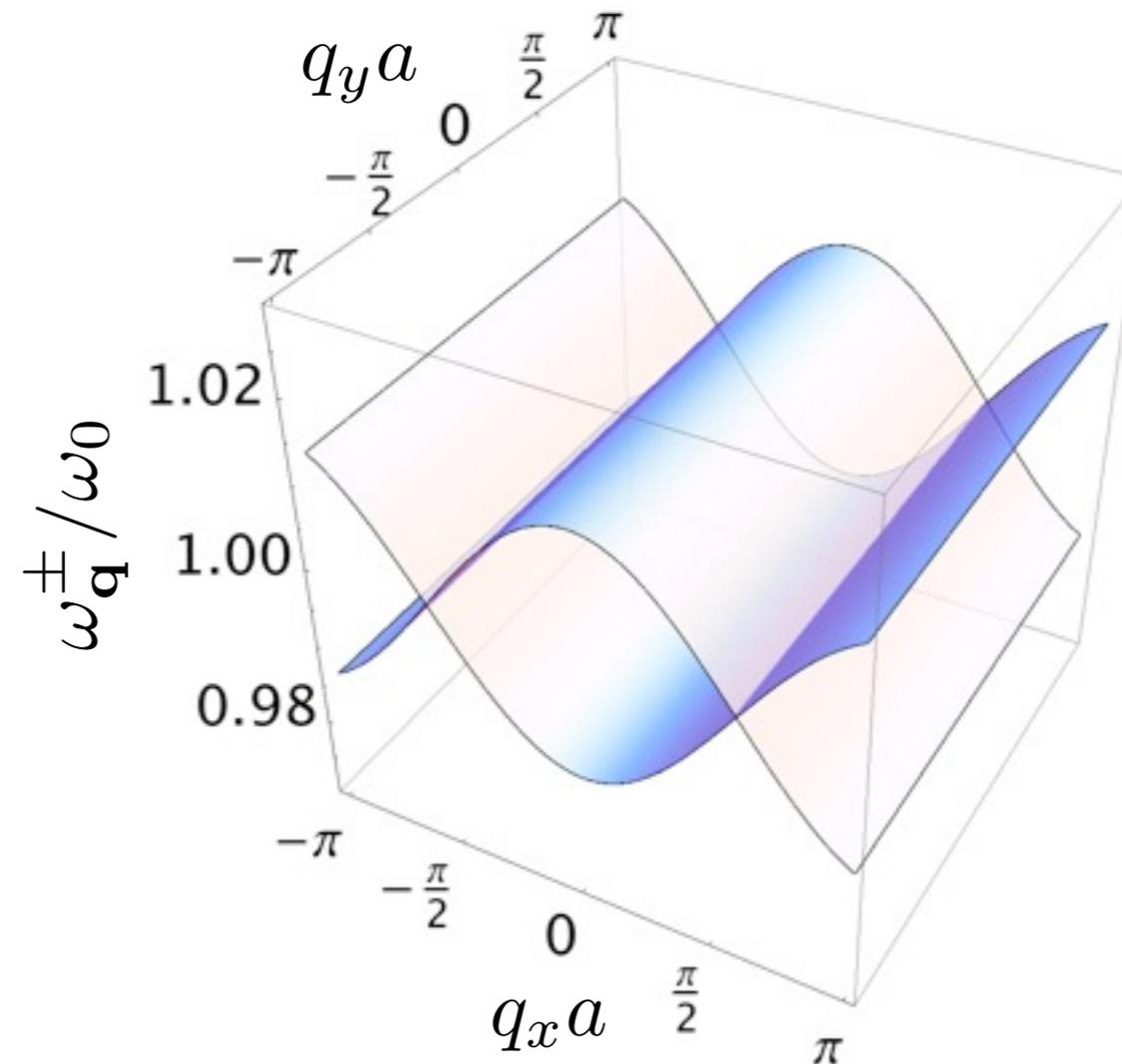
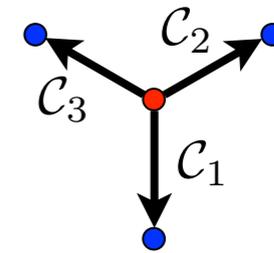
➔ gapped modes

Plasmon dispersion

$$\theta = \arcsin \sqrt{\frac{1}{3}}$$

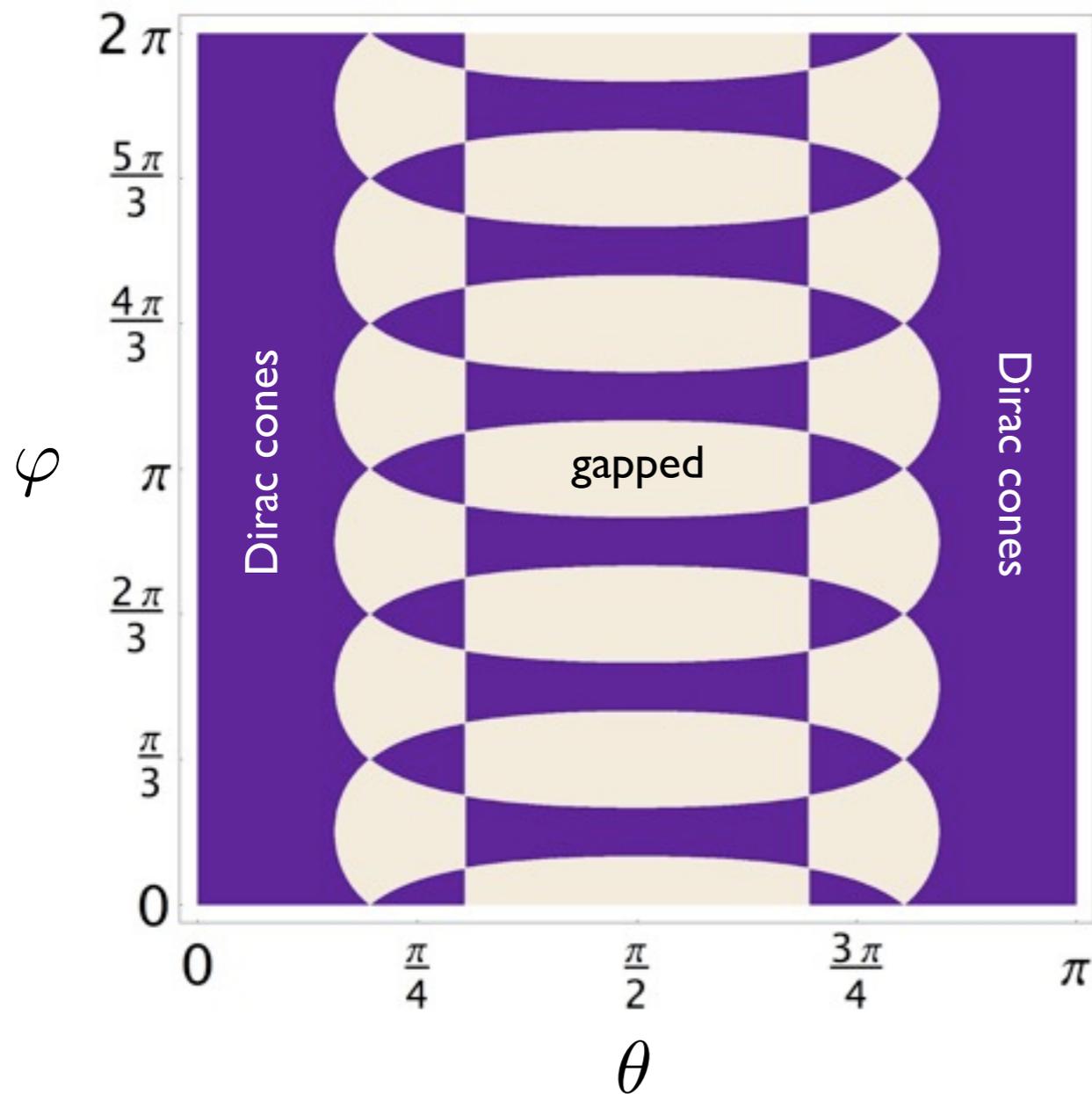


$$c_1 = 0, \quad c_2 = c_3 = 3/4$$



➔ Dirac "lines"

Plasmon dispersion



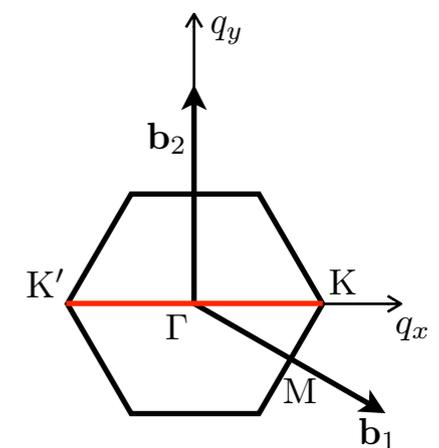
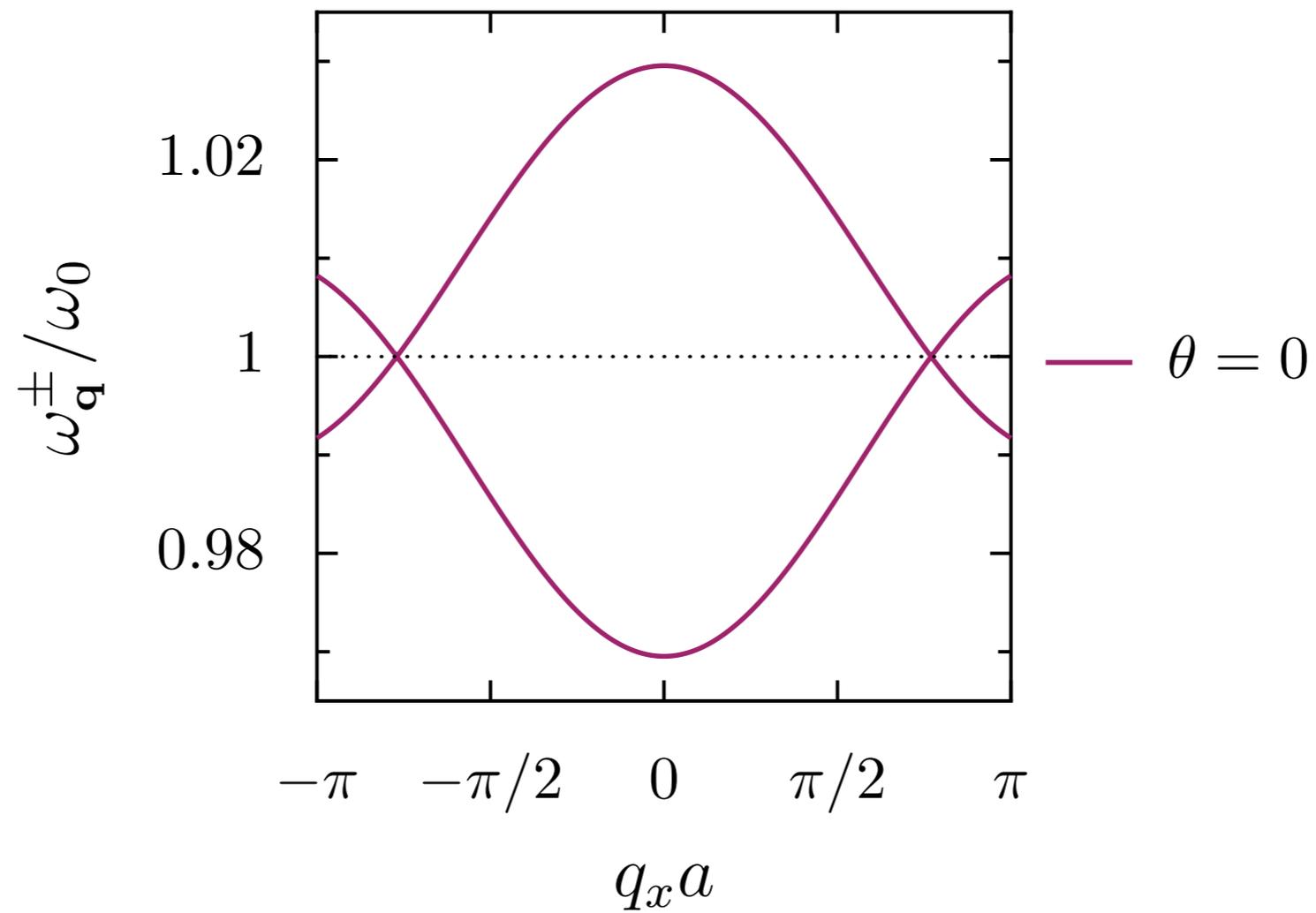
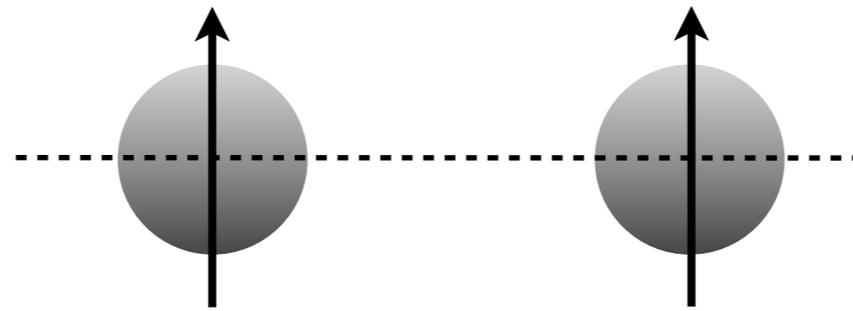
$$\omega_{\mathbf{q}}^{\pm} = \omega_0 \pm \Omega |f_{\mathbf{q}}|$$

gapless modes: $|f_{\mathbf{q}}| = 0$

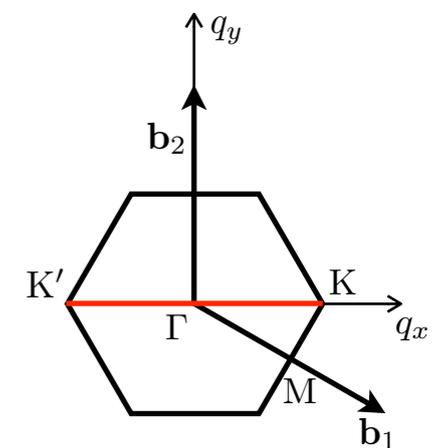
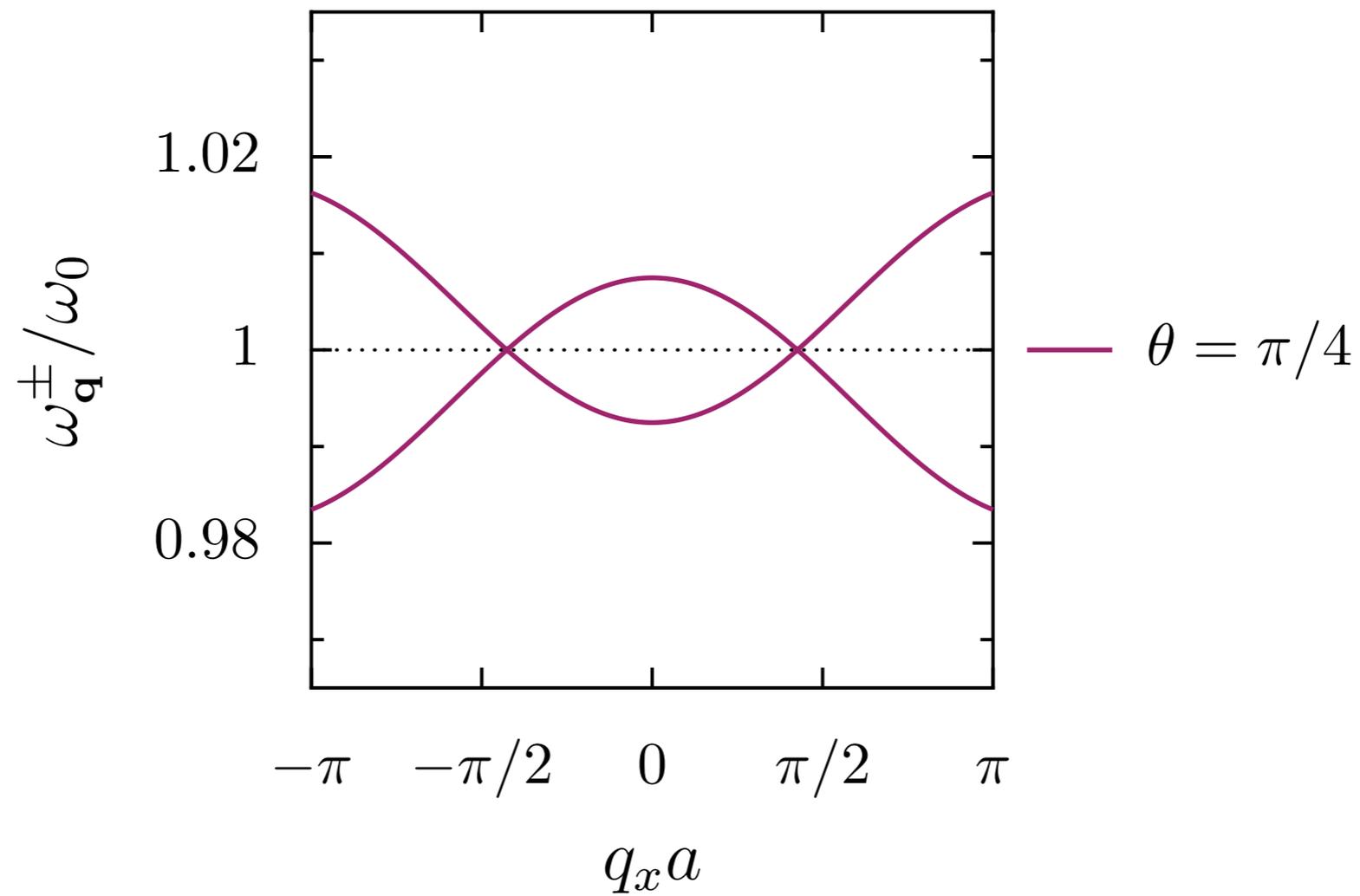
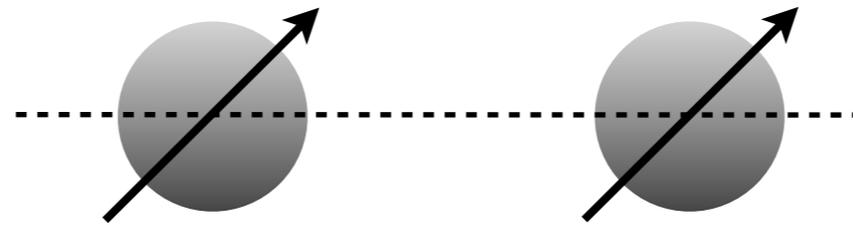
$$0 \leq \frac{(\mathcal{C}_2 + \mathcal{C}_3)^2 - \mathcal{C}_1^2}{4\mathcal{C}_2\mathcal{C}_3} \leq 1$$

➔ fully tunable spectrum (polarization)

Plasmon dispersion



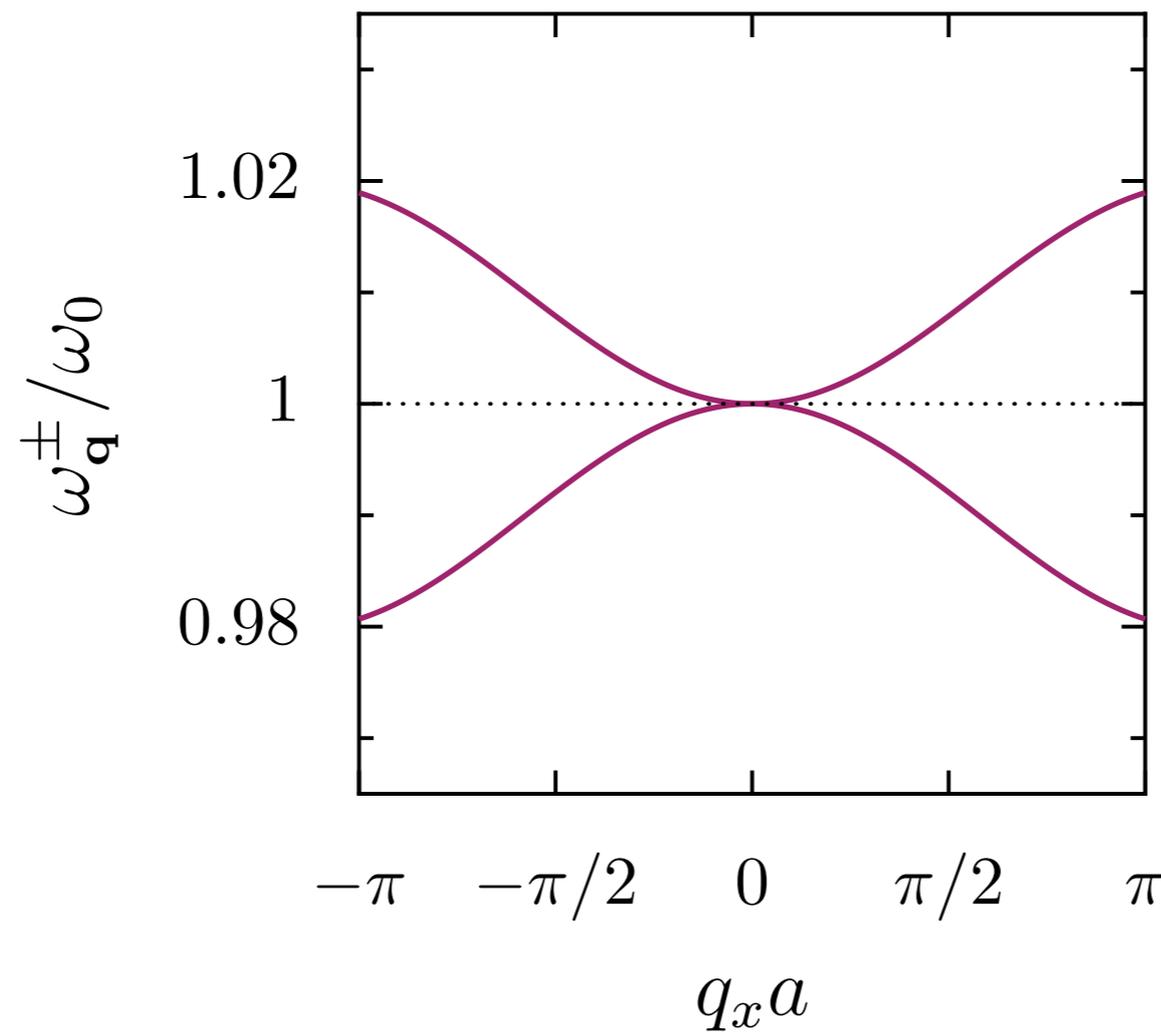
Plasmon dispersion



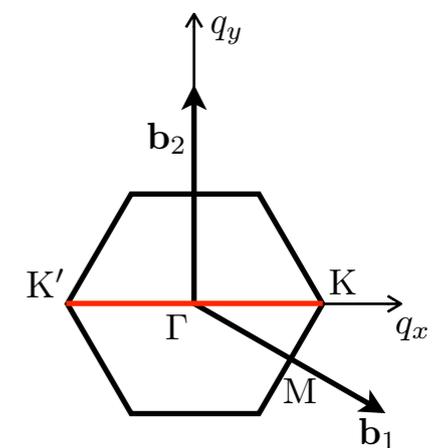
Plasmon dispersion



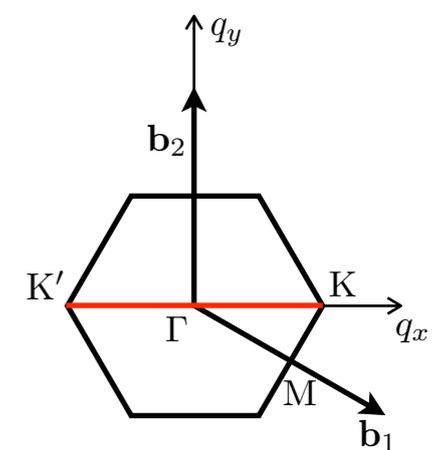
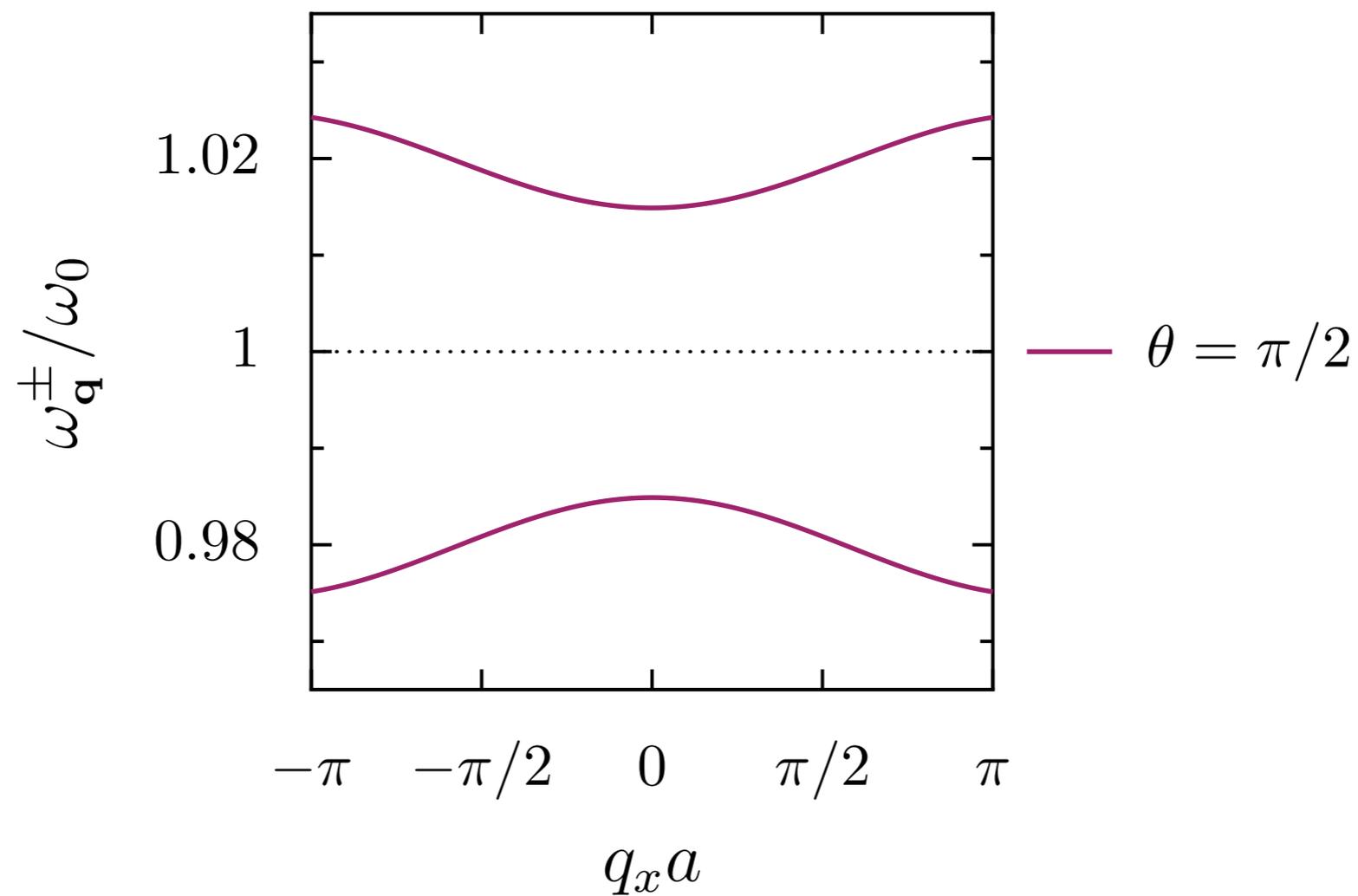
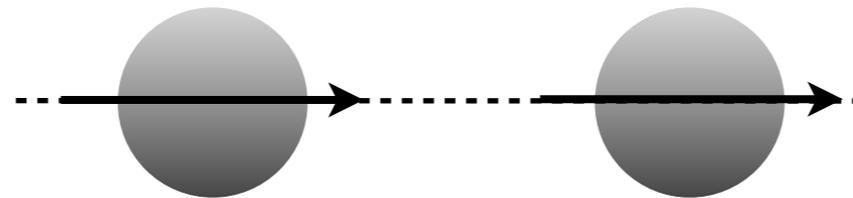
topological transition



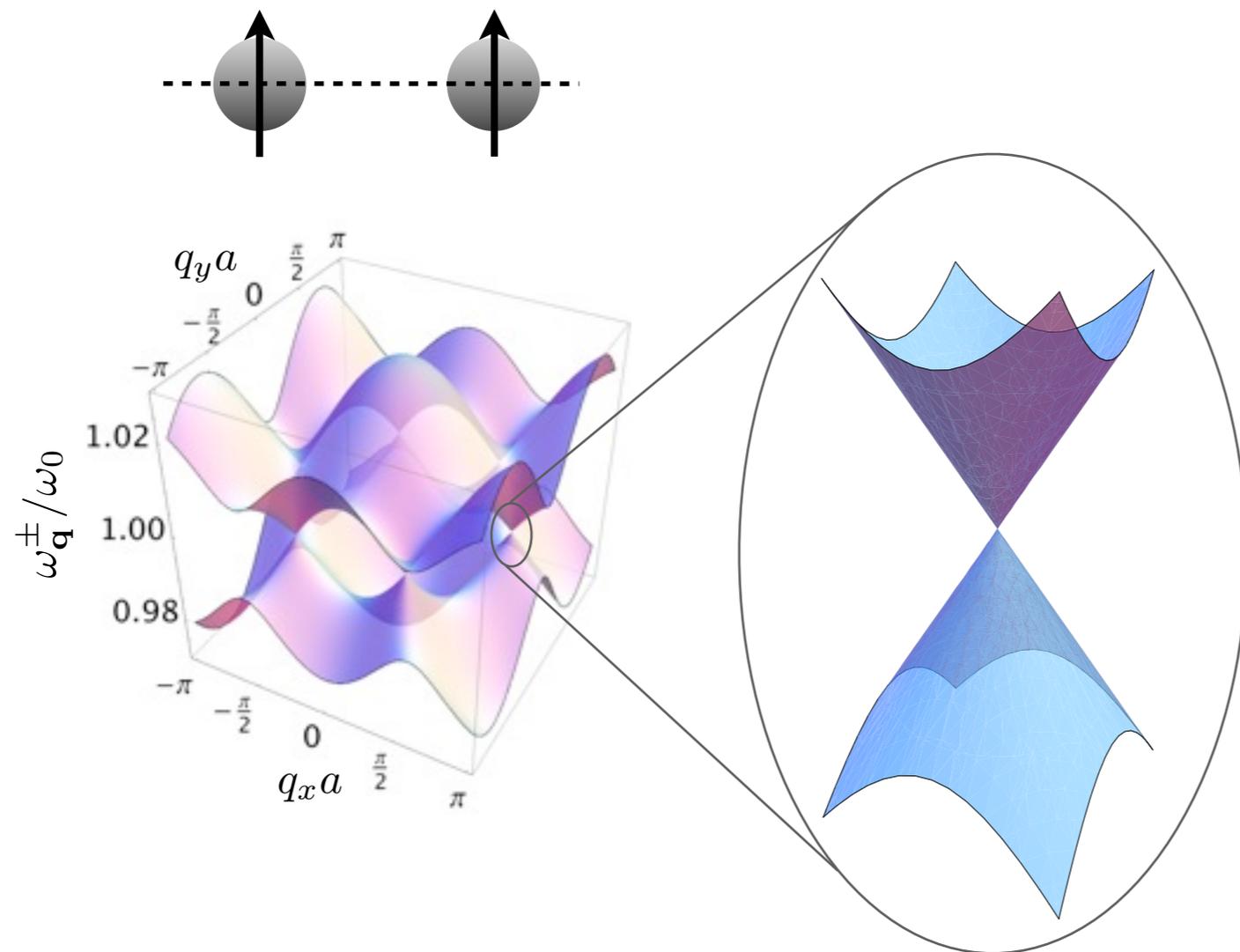
— $\theta = \arcsin \sqrt{\frac{2}{3}}$



Plasmon dispersion



Dirac-like plasmons



Close to K point:

$$\omega_{\mathbf{k}}^{\pm} \simeq \omega_0 \pm v|\mathbf{k}|$$

group velocity: $v = 3\Omega a/2 \approx c/100$

$$\mathcal{H}_{\mathbf{k}}^{\text{eff}} = \hbar\omega_0\mathbb{1} - \hbar v\tau_z \otimes \boldsymbol{\sigma} \cdot \mathbf{k}$$

spinor eigenstates:

$$\psi_{\mathbf{k},\text{K}}^{\pm} = \frac{1}{\sqrt{2}}(1, e^{\mp i\xi_{\mathbf{k}}}, 0, 0)$$

chirality (helicity) $\boldsymbol{\sigma} \cdot \hat{\mathbf{k}} = \pm\mathbb{1}$

➔ Collective plasmons should show similar effects to electrons in graphene

- absence of backscattering
- Klein paradox
- Berry phase of π
- ...

How do plasmons couple to light in periodic arrays of nanoparticles?

PHYSICAL REVIEW

VOLUME 103, NUMBER 5

SEPTEMBER 1, 1956

Atomic Theory of Electromagnetic Interactions in Dense Materials*

U. FANO

National Bureau of Standards, Washington, D. C.

(Received May 8, 1956)

PHYSICAL REVIEW

VOLUME 112, NUMBER 5

DECEMBER 1, 1958

Theory of the Contribution of Excitons to the Complex Dielectric Constant of Crystals*†

J. J. HOPFIELD‡

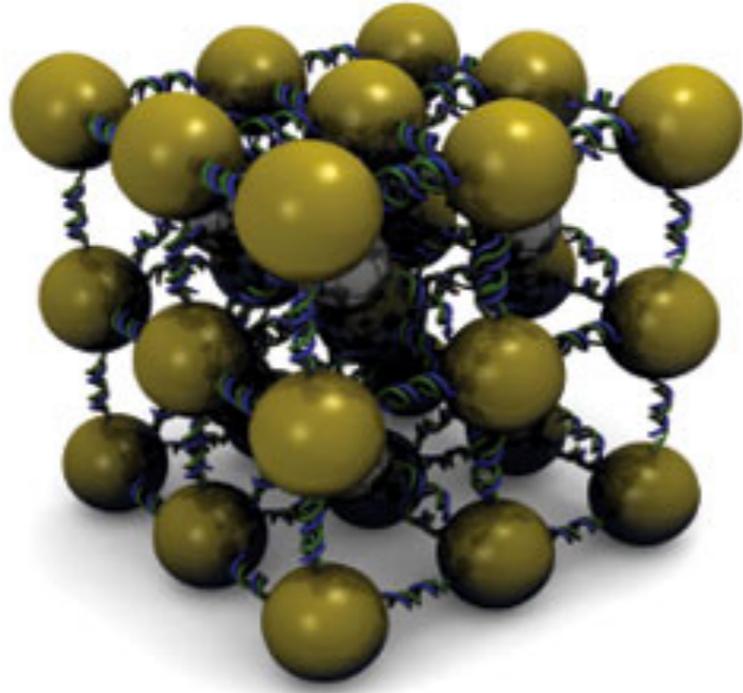
Physics Department, Cornell University, Ithaca, New York

(Received July 16, 1958)

➡ plasmon + photon = plasmon polariton

translational invariance: $\mathbf{k}_{\text{photon}} = \mathbf{k}_{\text{plasmon}}$

Simple cubic lattice



Tan *et al.*, Nature Nanotech. 2011

unpublished

Plasmons in honeycomb lattices of metallic nanoparticles:

- massless Dirac-like *bosons*
- similar properties as electrons in graphene
- fully *tunable* spectrum

GW, C. Woollacott, W.L. Barnes, O. Hess, E. Mariani
Dirac-like plasmons in honeycomb lattices of metallic nanoparticles
Phys. Rev. Lett. **110**, 106801 (2013)

Plasmon polaritons in 3d (cubic) arrays:

- polaritonic band gap can be modified w/ light polarization
- tunable optical properties

GW, E. Mariani
Tunable plasmon polaritons in interacting arrays of metallic nanoparticles
unpublished