LATTICE DYNAMICS WITH BROKEN TIME REVERSAL SYMMETRY

John Bonini
Shang Ren*, David Vanderbilt, Massimiliano Stengel, Cyrus E. Dreyer, and Sinisa Coh

***See Shang's poster!**

FLATIRON INSTITUTE
Center for Computational Quantum Physics
SIMONS FOUNDATION

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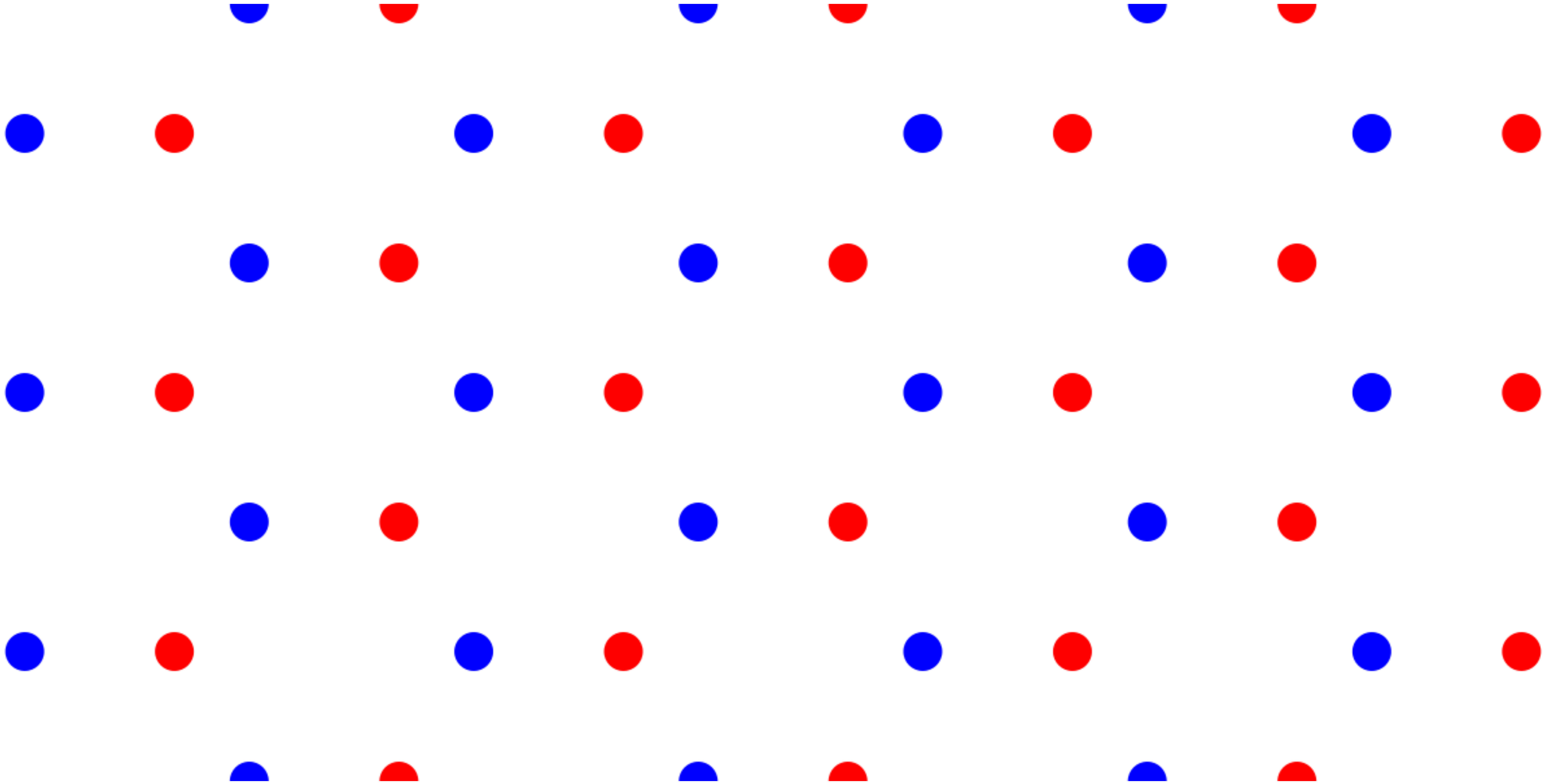
LATTICE



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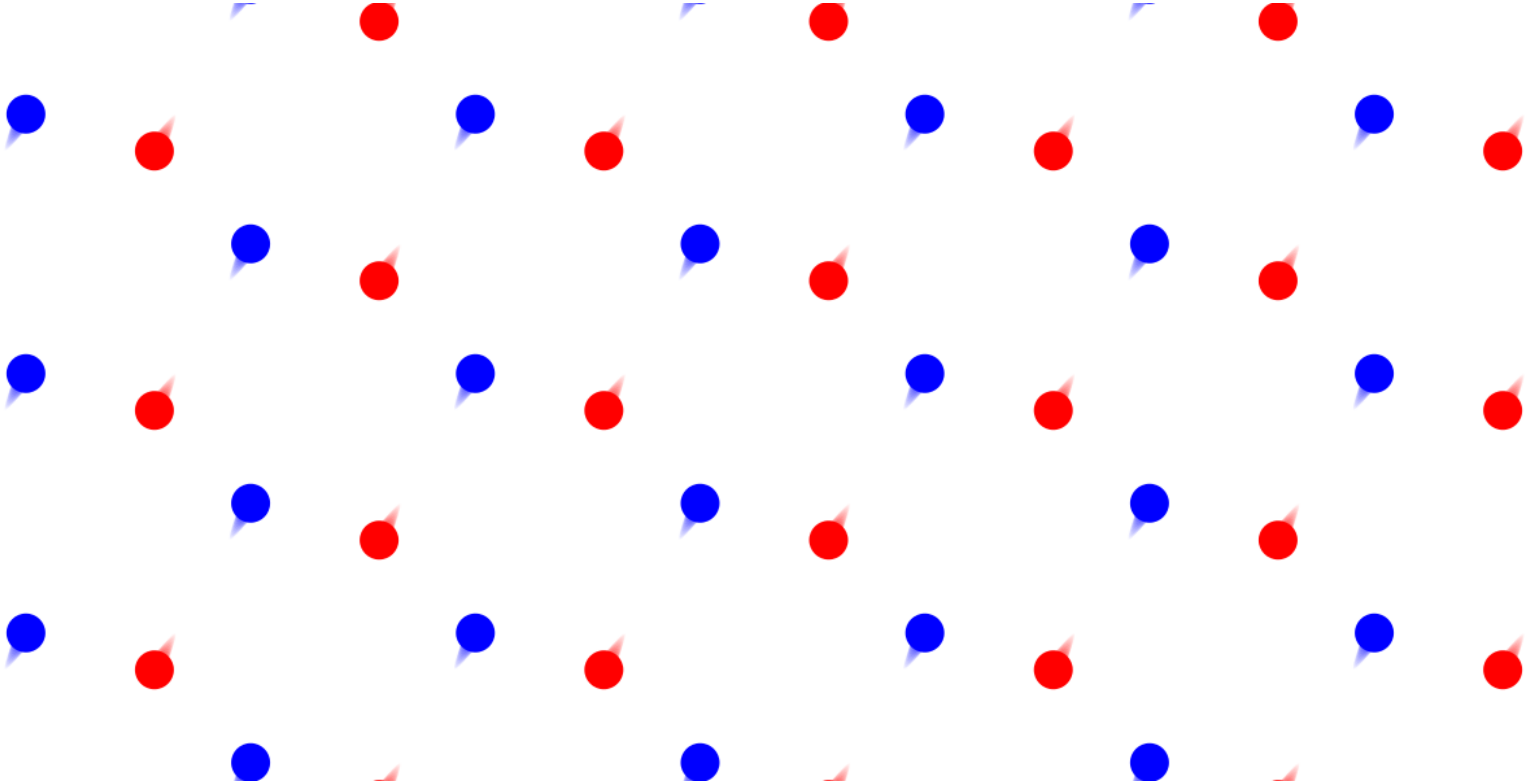

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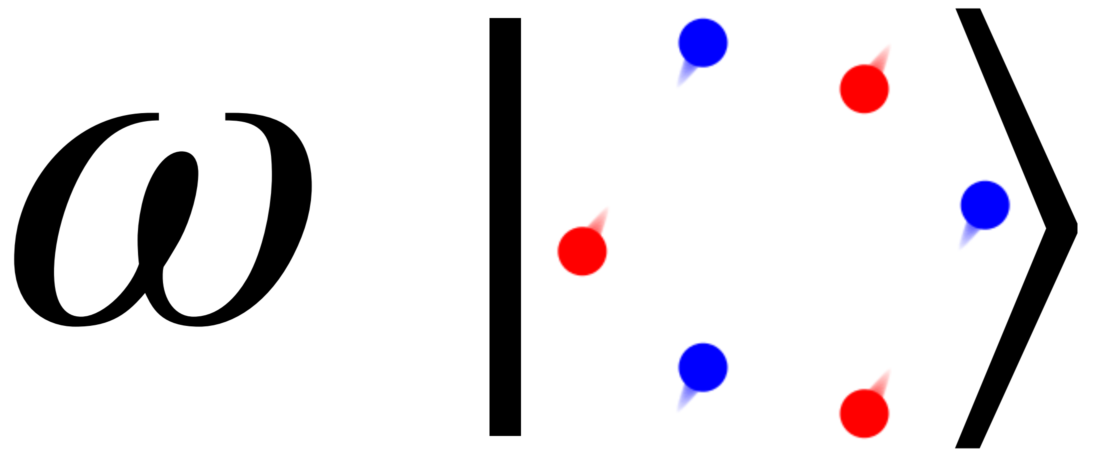

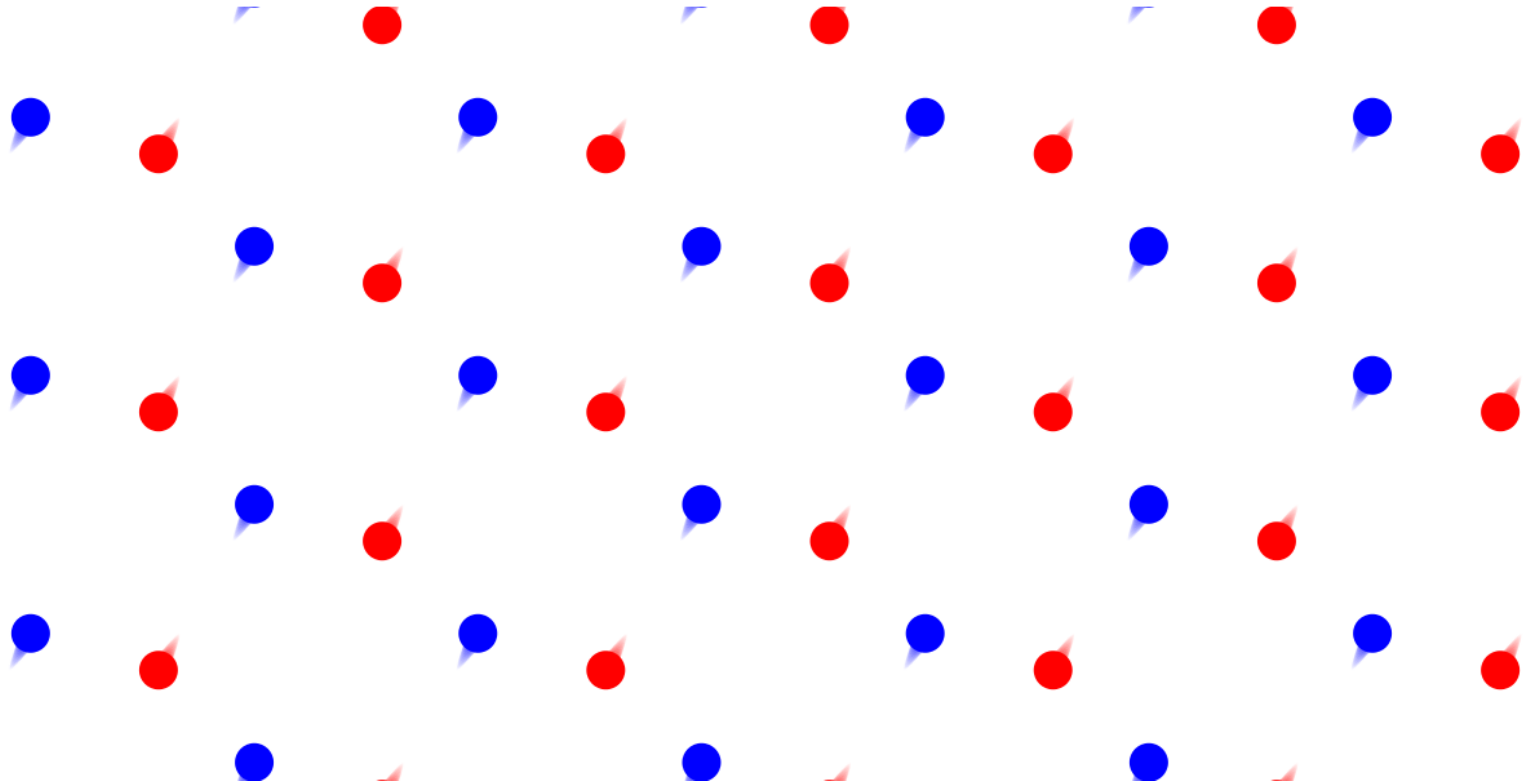

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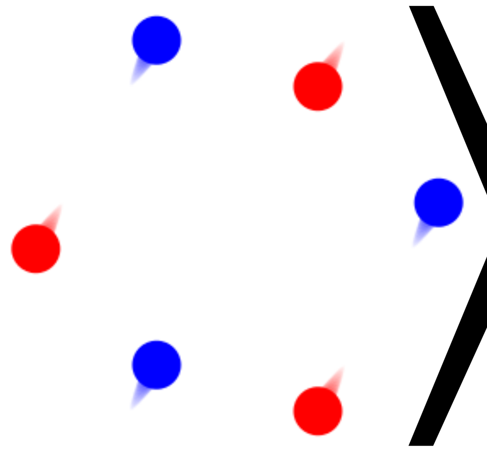
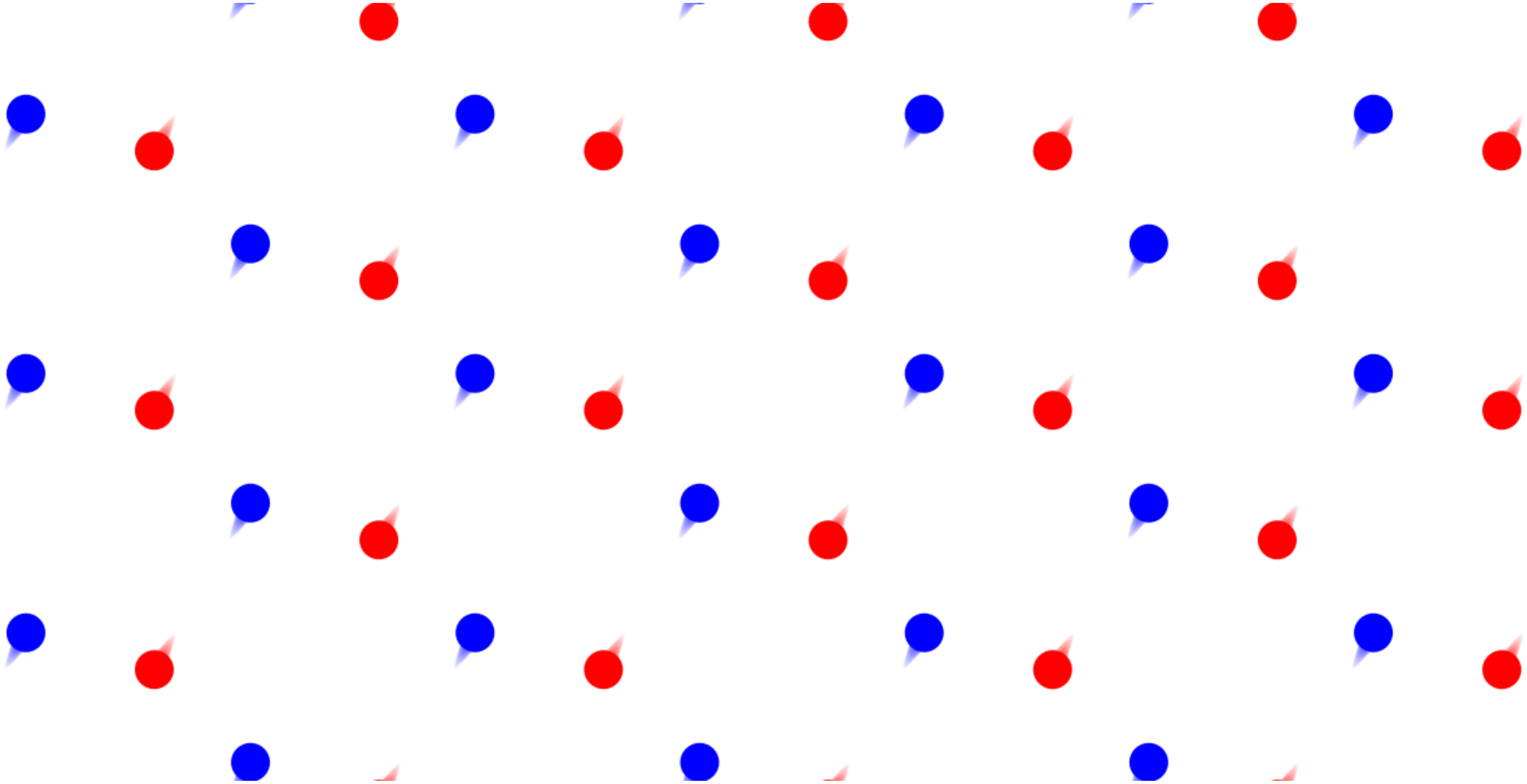
$\omega \; |$  $\rangle$



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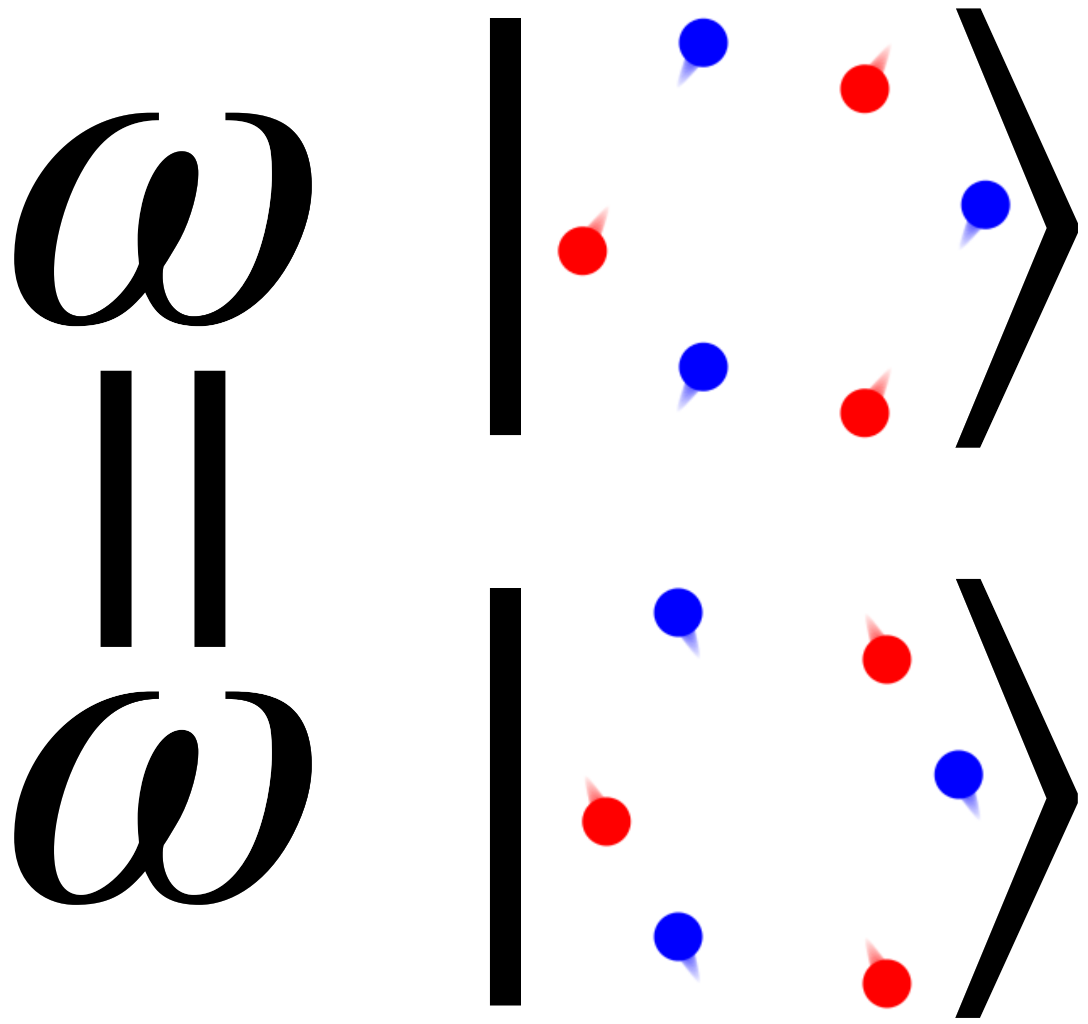

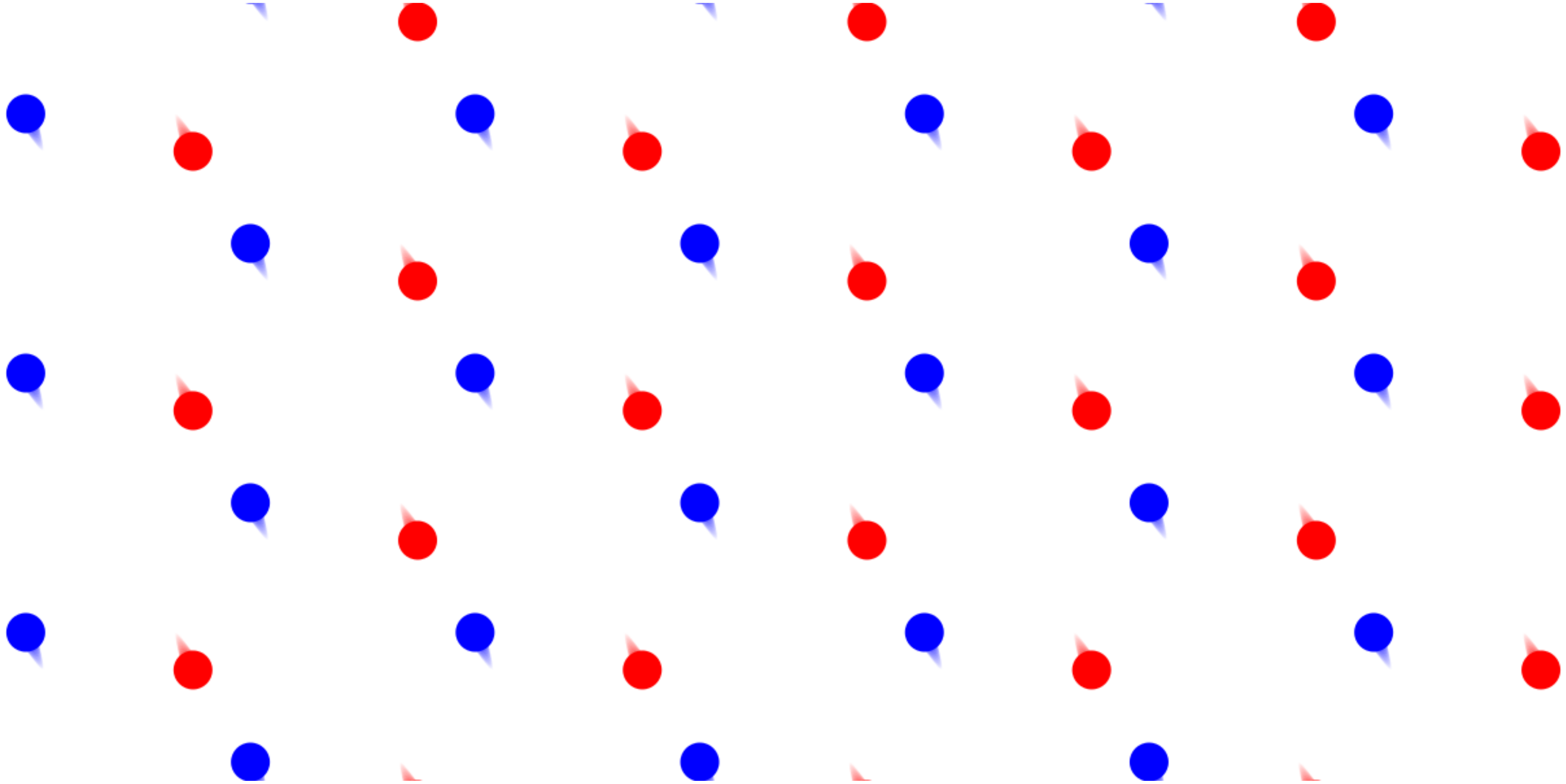

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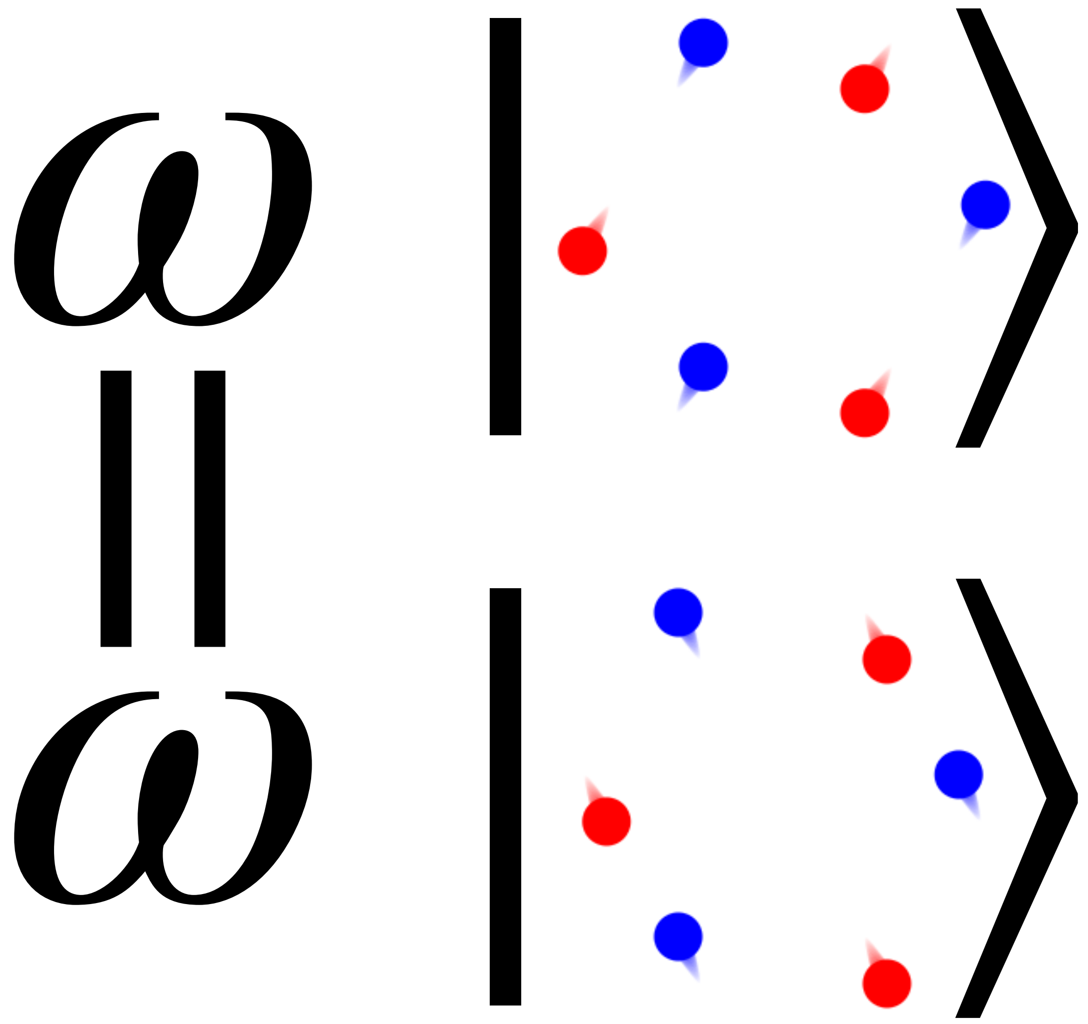

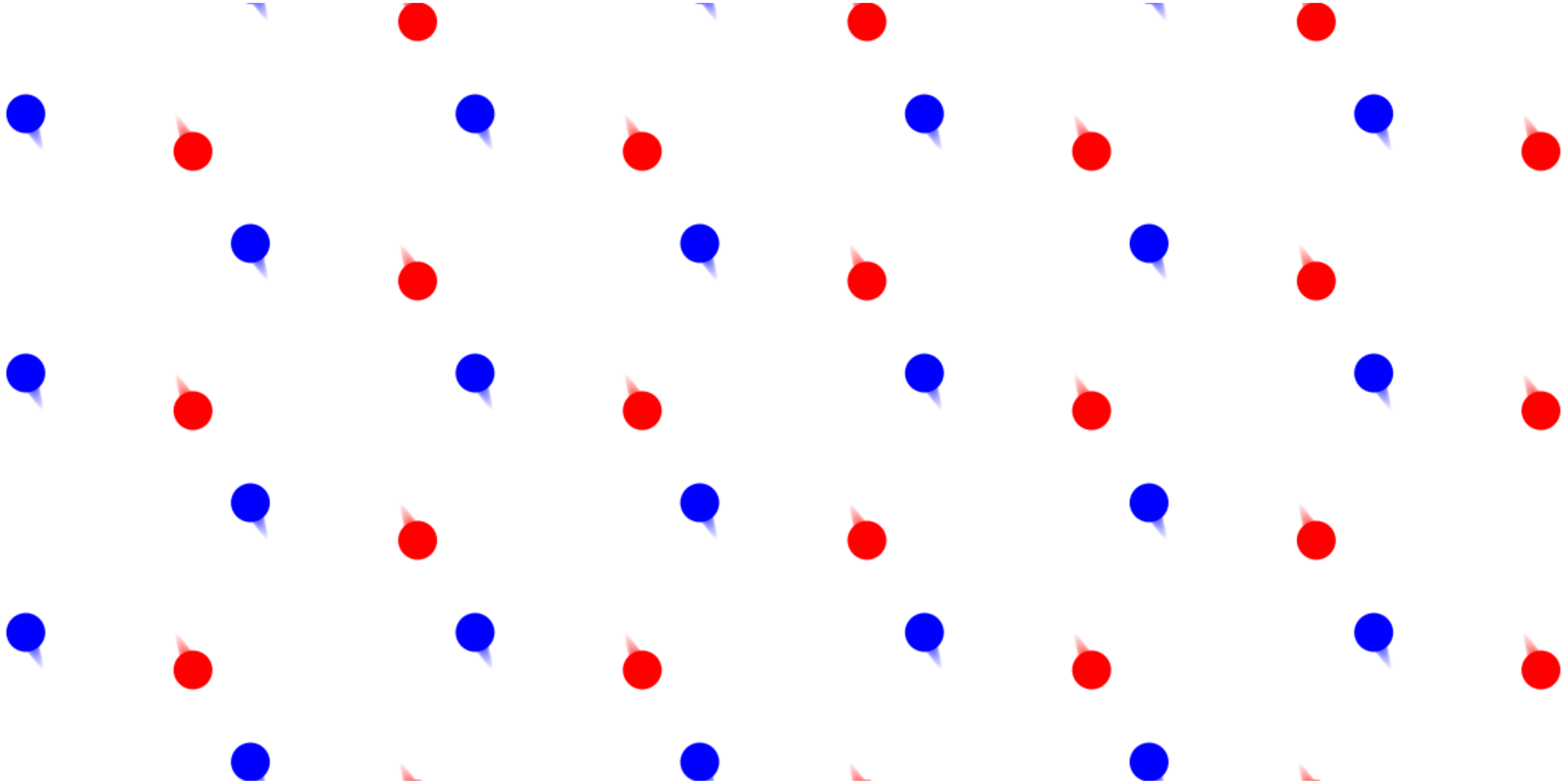

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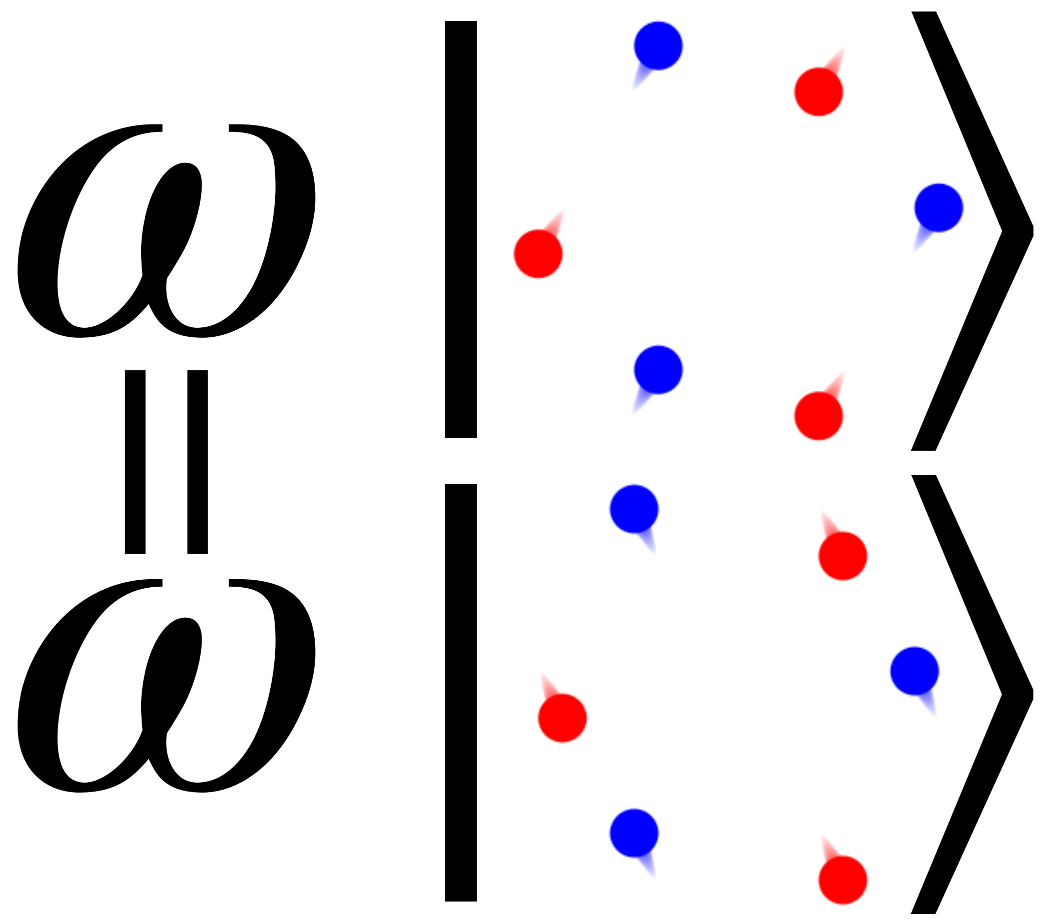

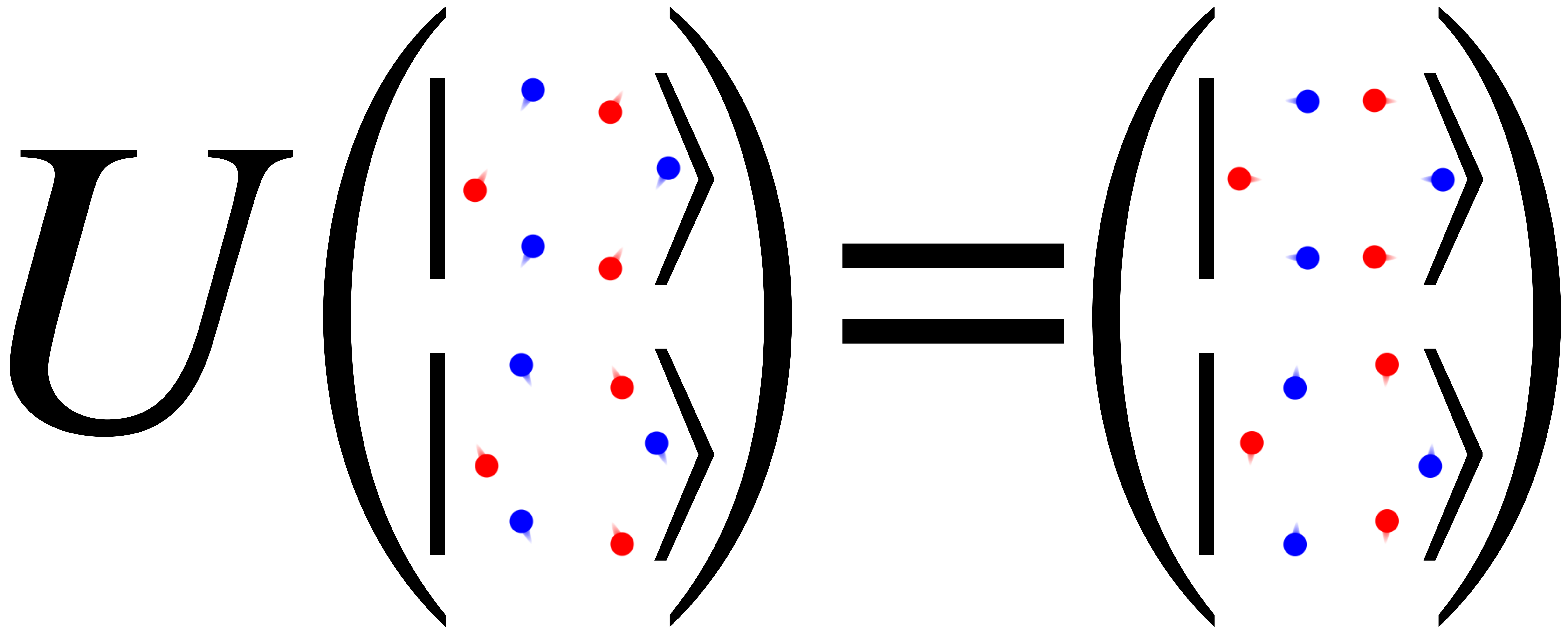

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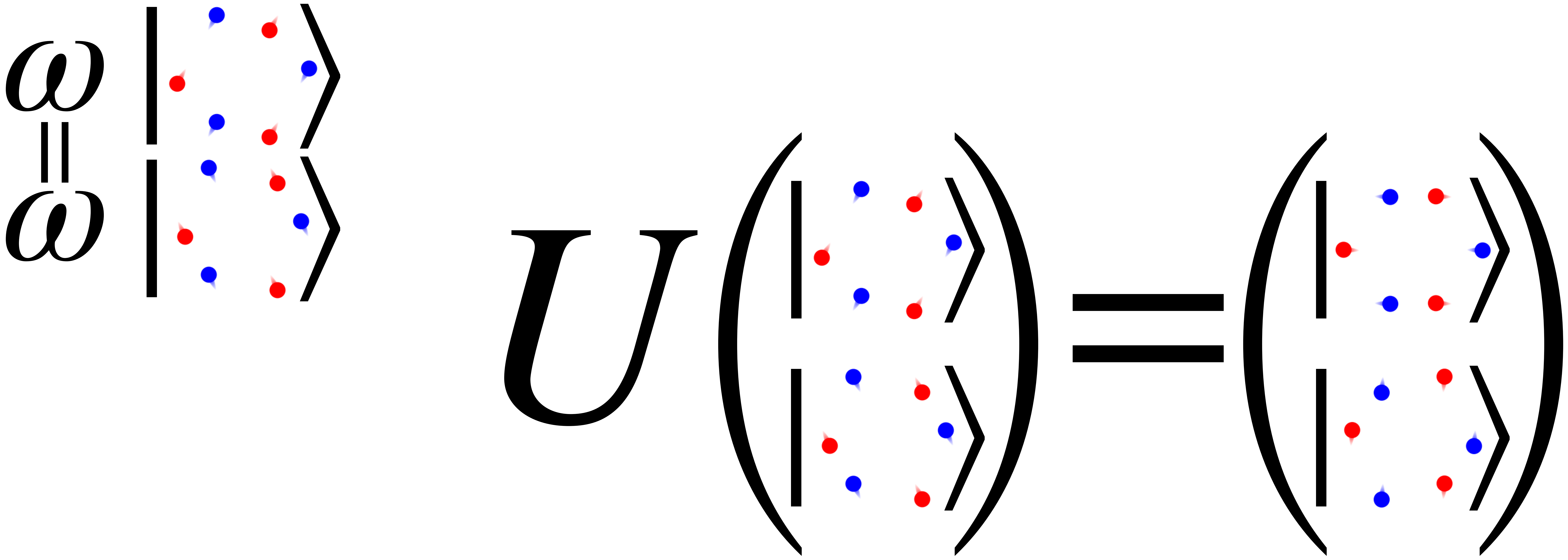

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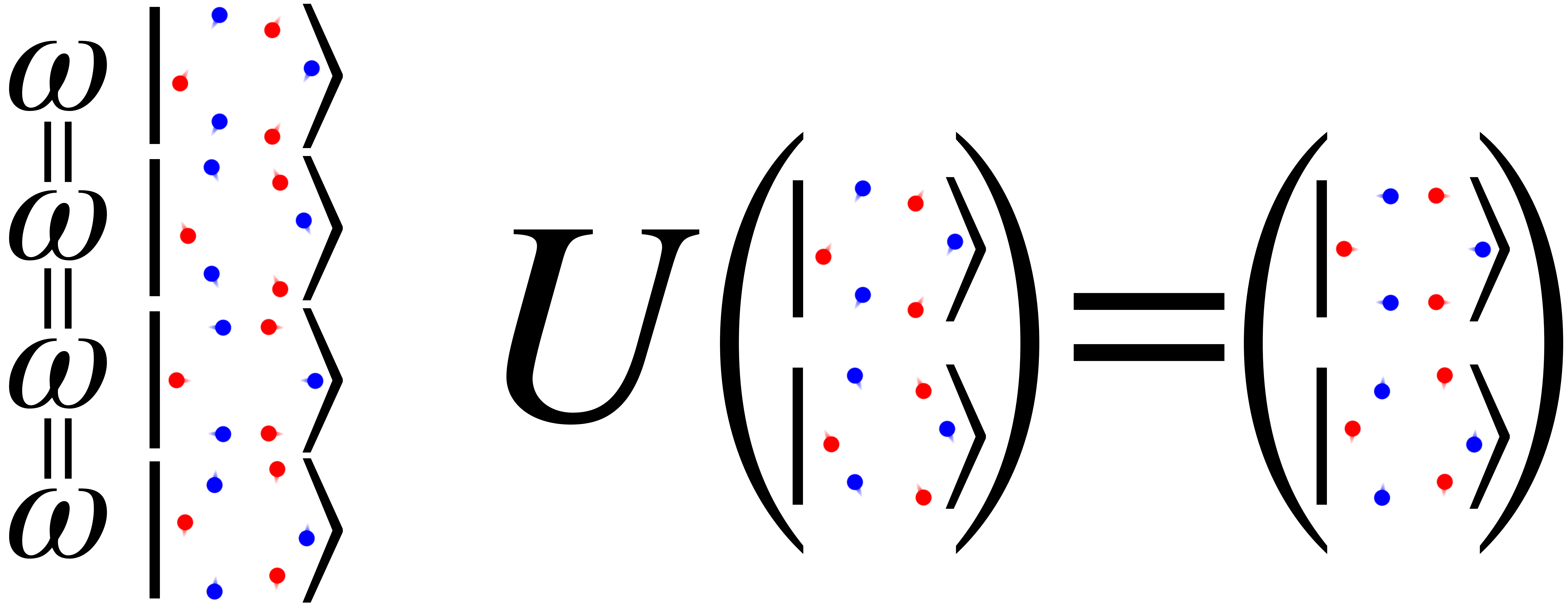

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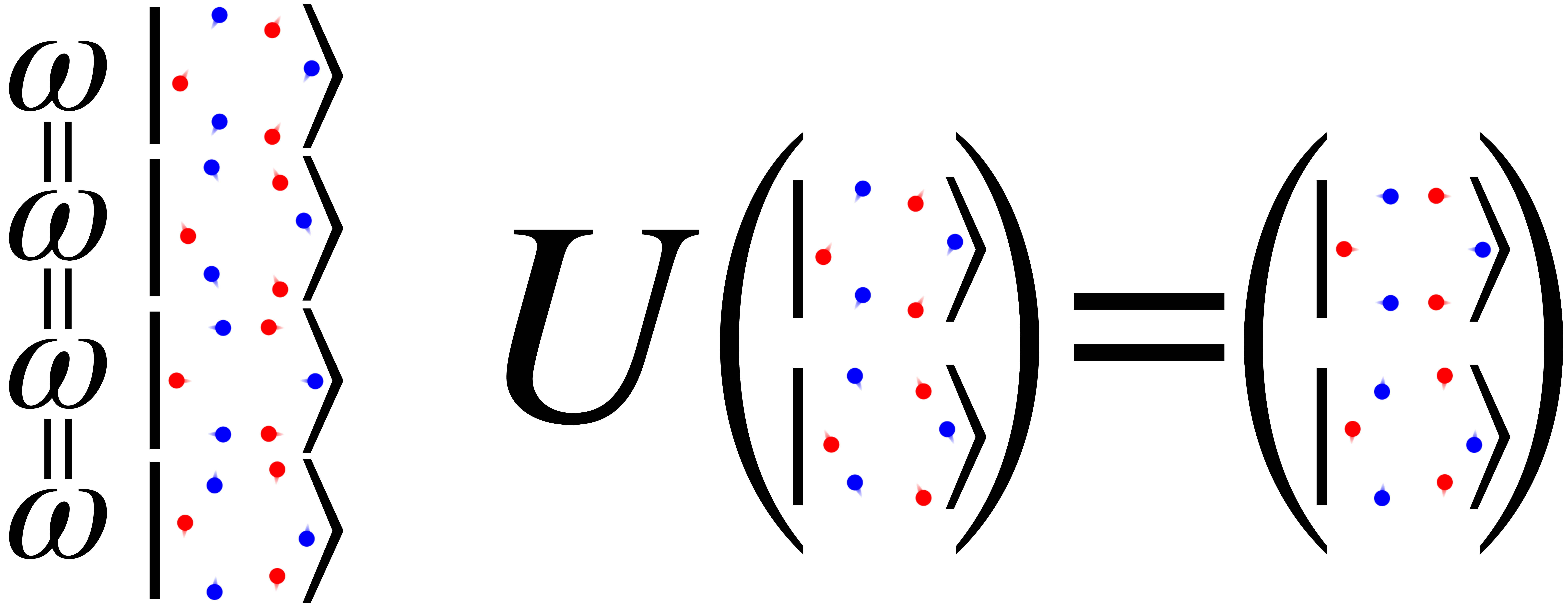

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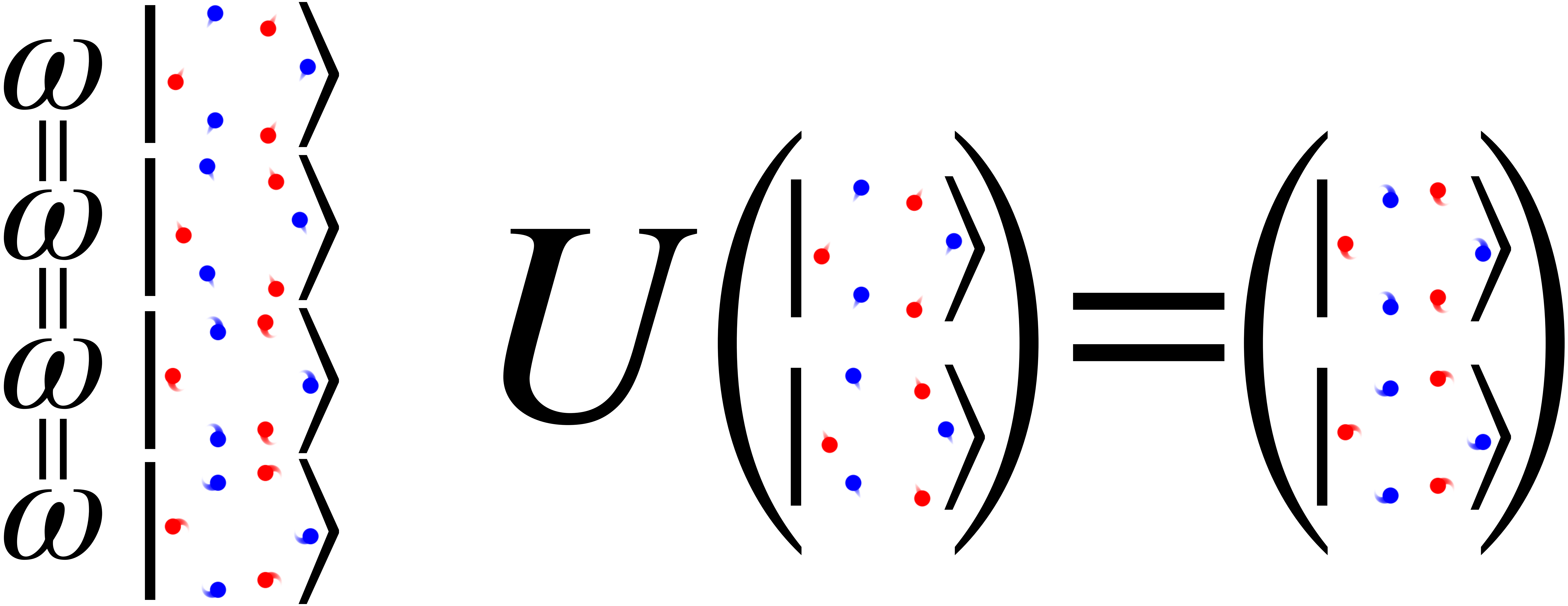

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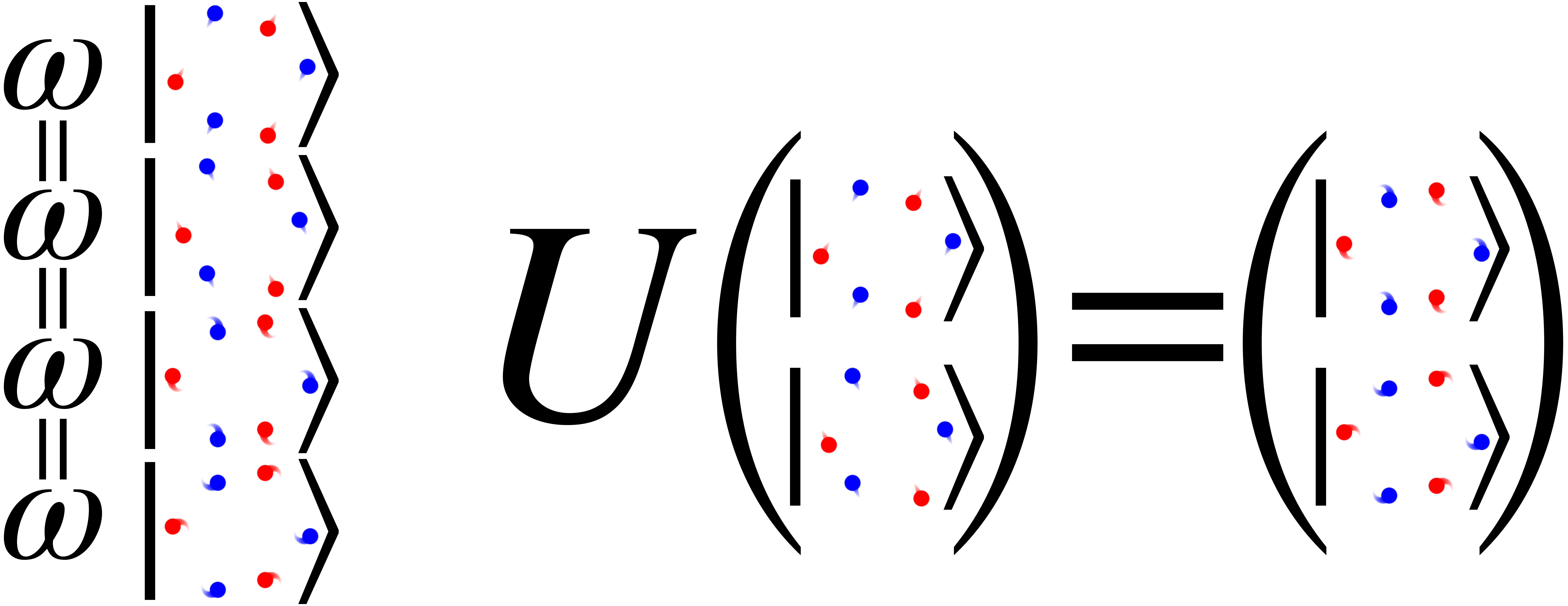

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"Chiral phonons"

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Recent interest in chiral phonons

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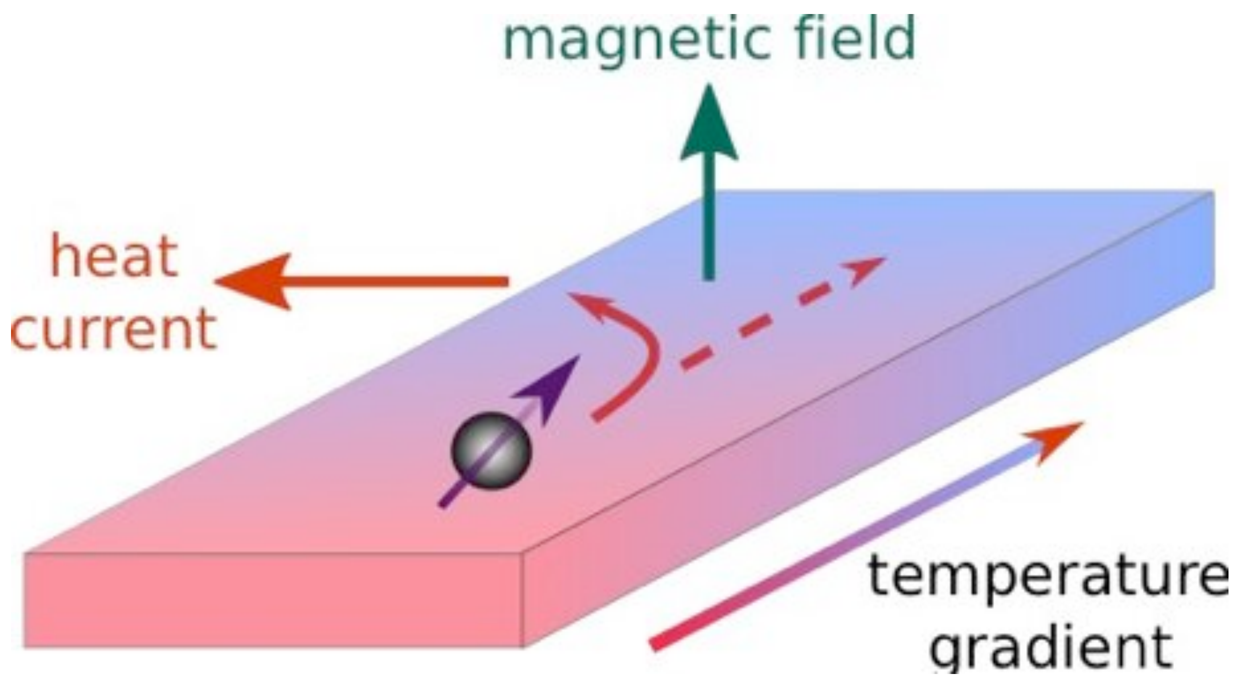
Thermal Hall conductivity



Large phonon thermal Hall conductivity in the antiferromagnetic insulator Cu_3TeO_6

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Anomalous Thermal Hall Effect in an Insulating van der Waals Magnet

Heda Zhang,[1,*] Chunqiang Xu,[1,2,*] Caitlin Carnahan,[3,*] Milos Sretenovic,[1] Nishchay Suri,[3] Di Xiao,[3,4,5] and Xianglin Ke[1]

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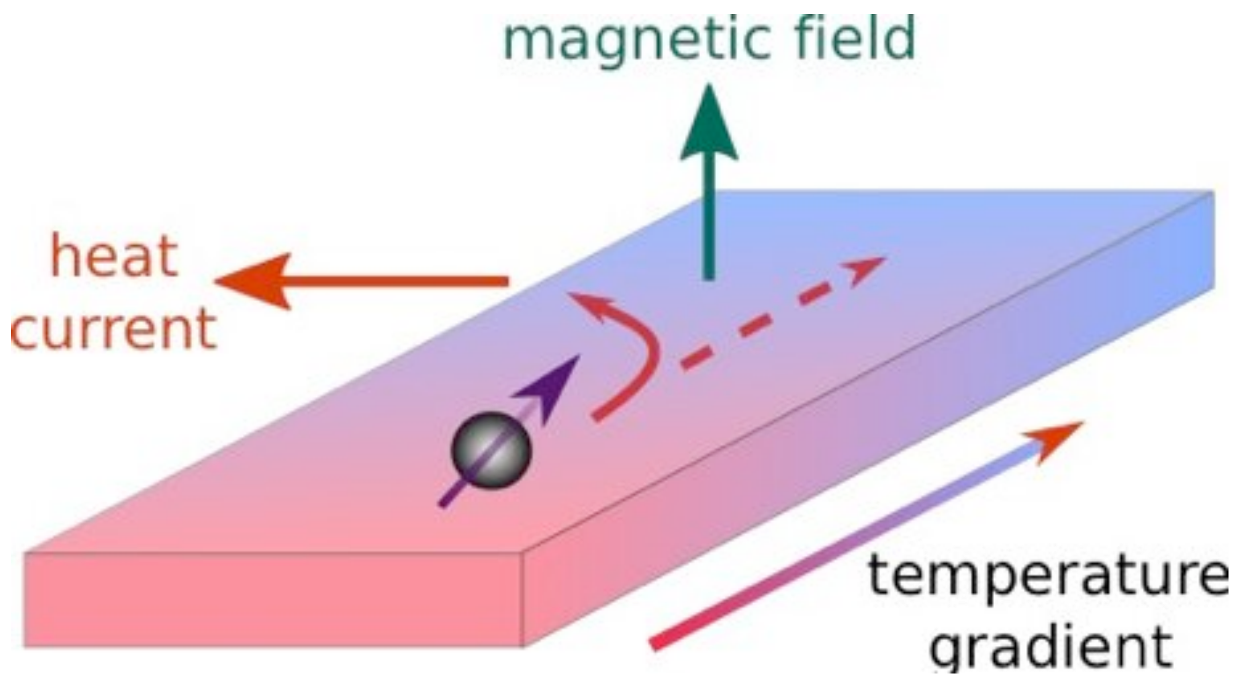

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Phonon magnetic moments

Phonon Magnetic Moment from Electronic Topological Magnetization

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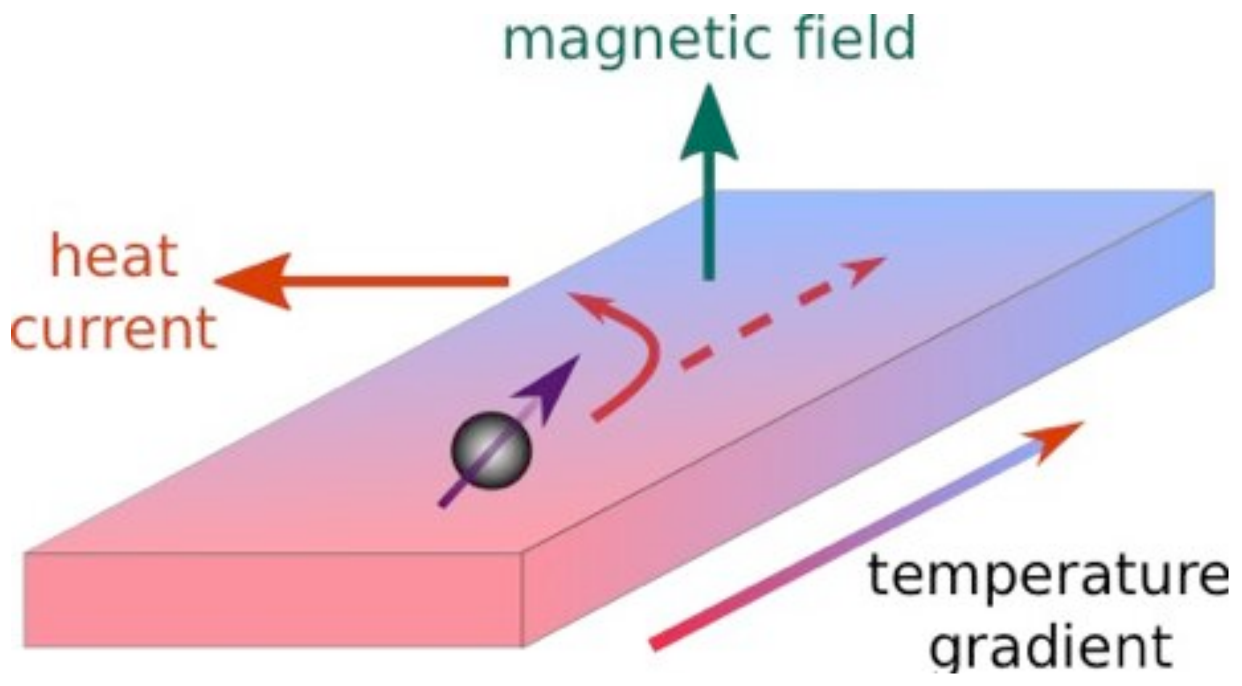

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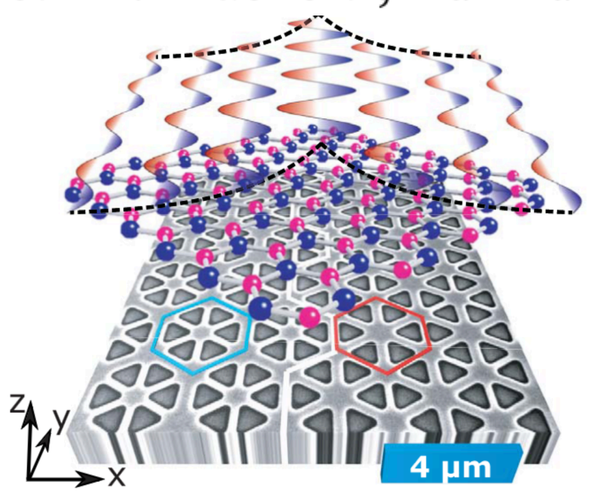

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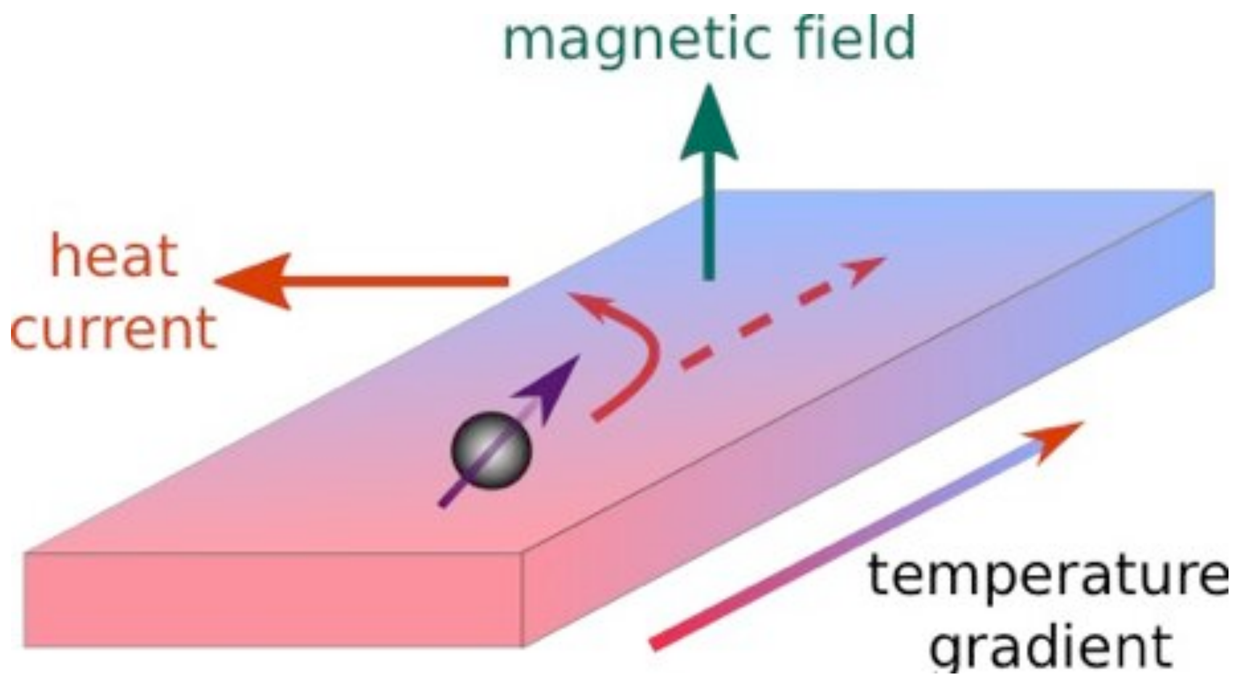

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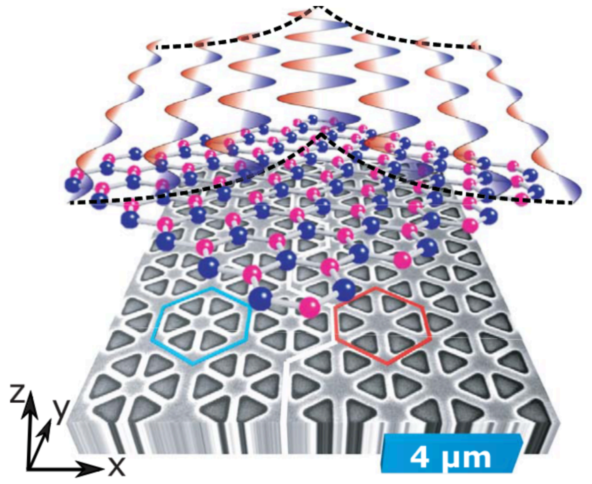

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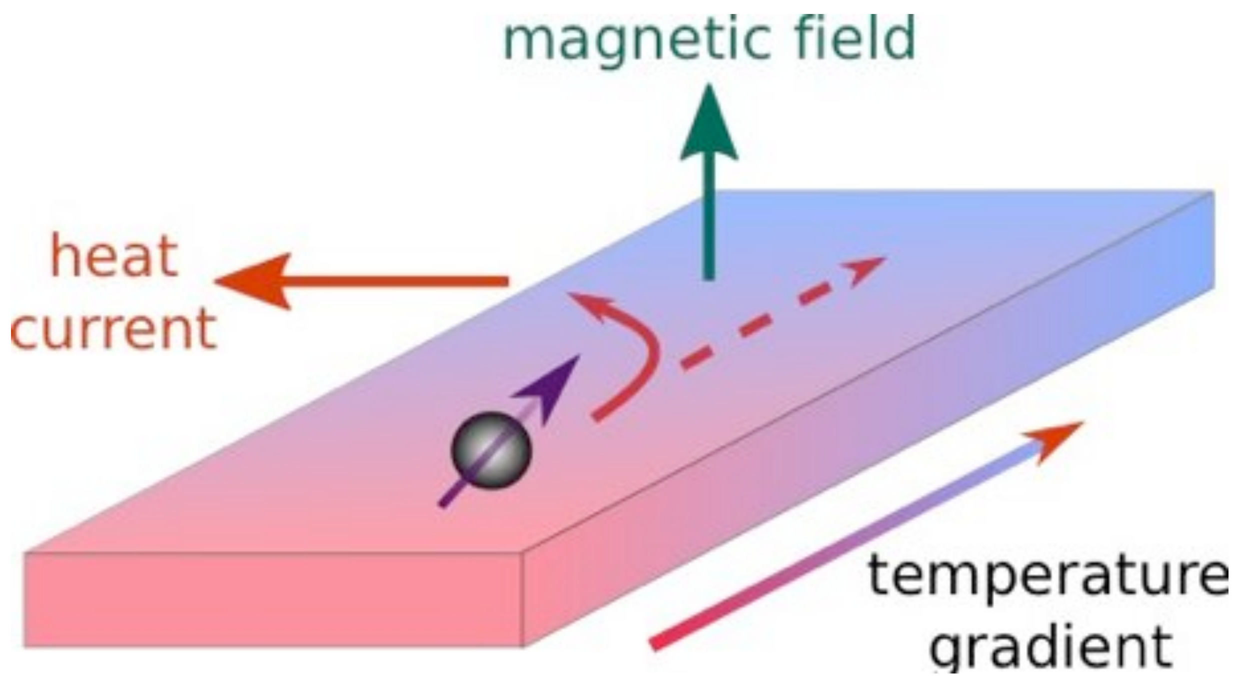

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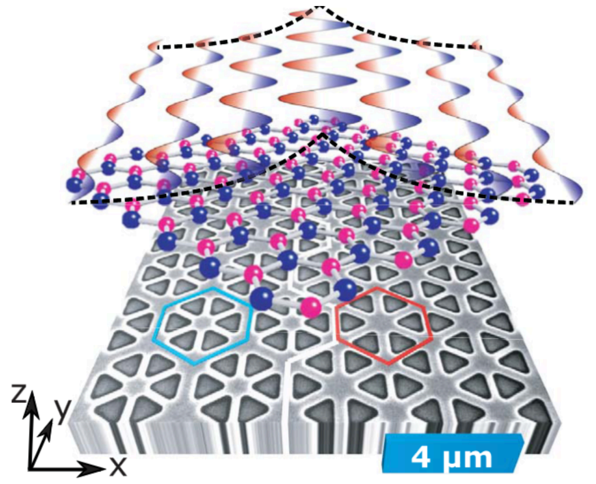

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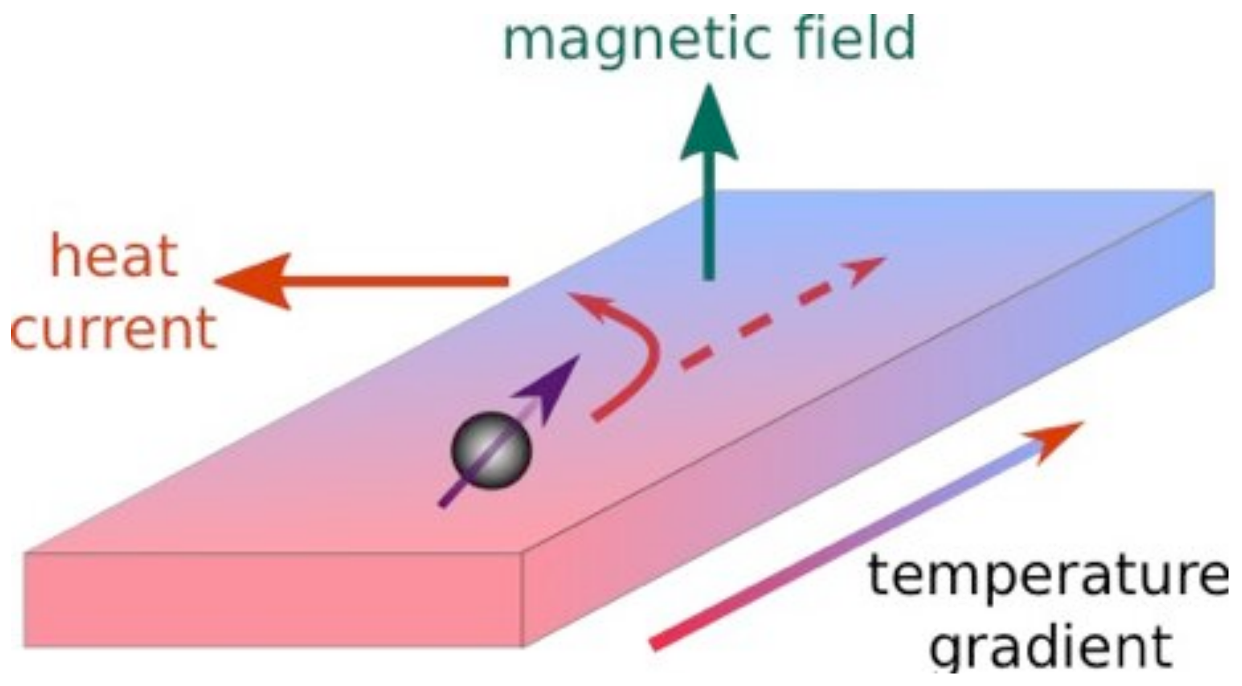

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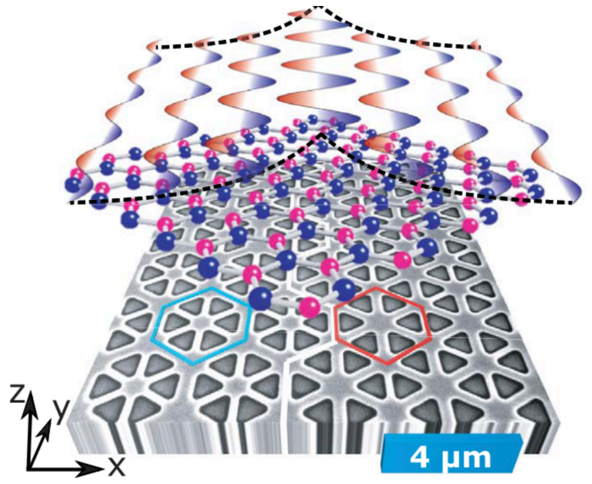

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How can we treat such physics with ab initio methods?

Interatomic force constants

$$F_i = \mathrm{Tr}\left[(\partial_{R_i} H)\rho(R_1, R_2 \ldots)\right]$$

Interatomic force constants

Standard phonon procedure:

$$F_i \approx -C_{ij}\Delta R_j$$

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Interatomic force constants

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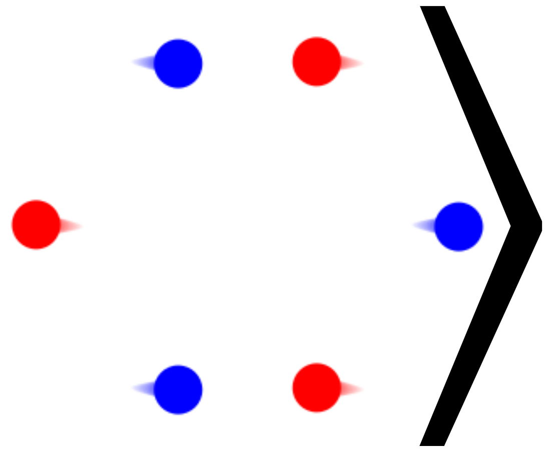
$\omega_x \Big|$  $\Big\rangle$

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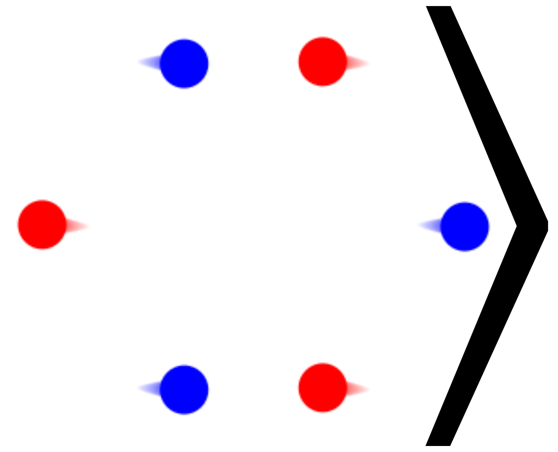
$\omega_x \,|$  $\rangle$

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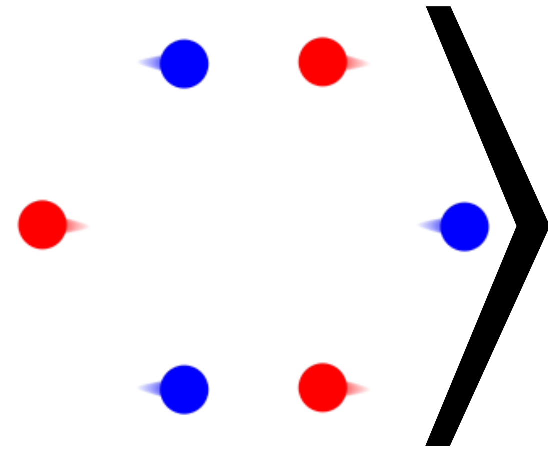
$\omega_x |$  $\rangle$

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$$m_i \frac{d^2 R_i}{dt^2} = f_i(R_1, R_2, \ldots) = m_i \frac{d^2 R_i}{(-dt)^2}$$

Forces as a function of only $\{R_i\}$ result in TR symmetric dynamics

Interatomic force constants

Standard phonon procedure:

$$F_i \approx - C_{ij} \Delta R_j$$

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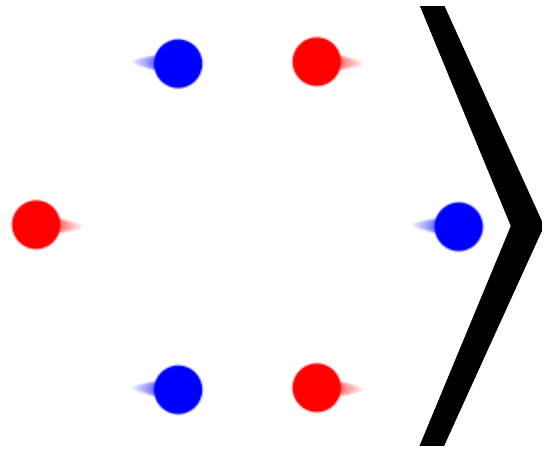
$\omega_x \Big|$  $\Big\rangle$

$$F_i = \mathrm{Tr} \left[\sum_{\alpha \in \{\uparrow\uparrow, \uparrow\downarrow, \downarrow\uparrow, \downarrow\downarrow\}} (\partial_{R_i} H)_\alpha \rho_\alpha (R_1, R_2 \ldots) \right]$$

$$m_i \frac{d^2 R_i}{dt^2} = f_i(R_1, R_2, \ldots) = m_i \frac{d^2 R_i}{(-dt)^2}$$

Forces as a function of only $\{R_i\}$ result in TR symmetric dynamics

Even in magnetic systems

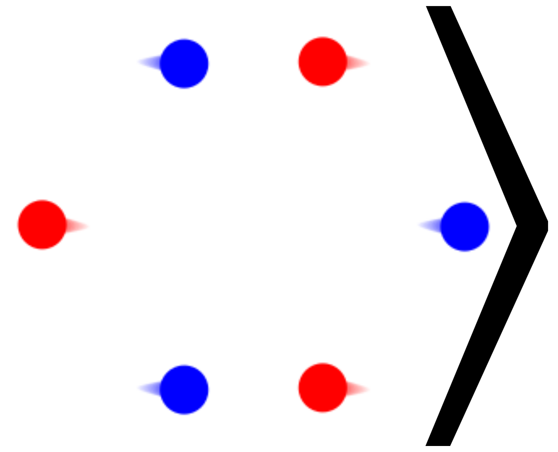
Interatomic force constants

Standard phonon procedure:

$$F_i \approx -C_{ij}\Delta R_j$$

$$m_i \frac{d^2R_i}{dt^2} = -\frac{1}{2}\sum_{ij} C_{ij}\Delta R_j$$

$$\mathbf{M}\omega_n^2 |n\rangle = \mathbf{C}|n\rangle$$

ω_x |⟩

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Forces as a function of only $\{R_i\}$ result in TR symmetric dynamics

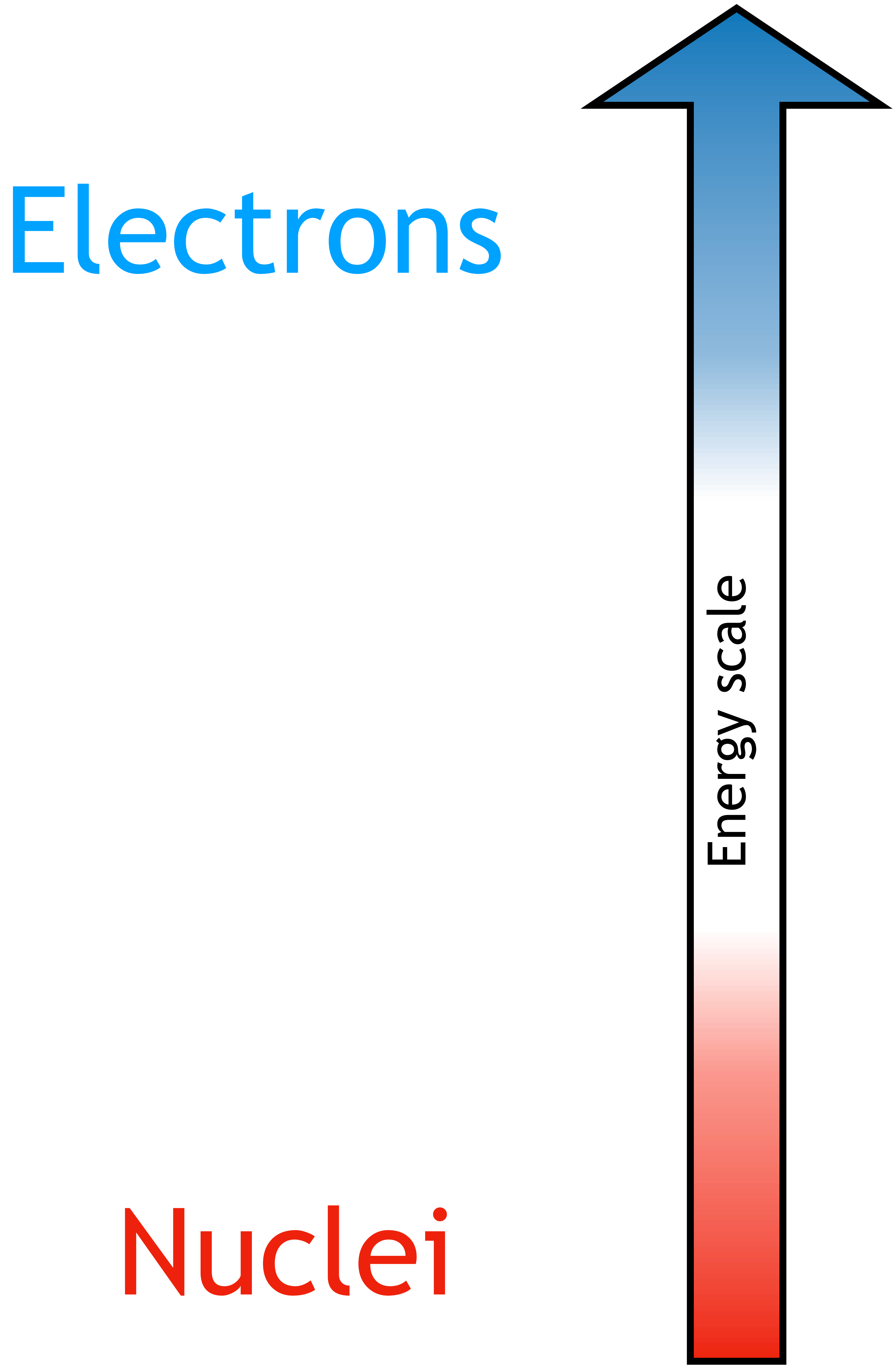
Even in magnetic systems

We must go beyond static forces!

Beyond static forces

$$f_i(R_1, R_2, \ldots) = \nabla_{R_i} \epsilon(R_1, R_2, \ldots)$$

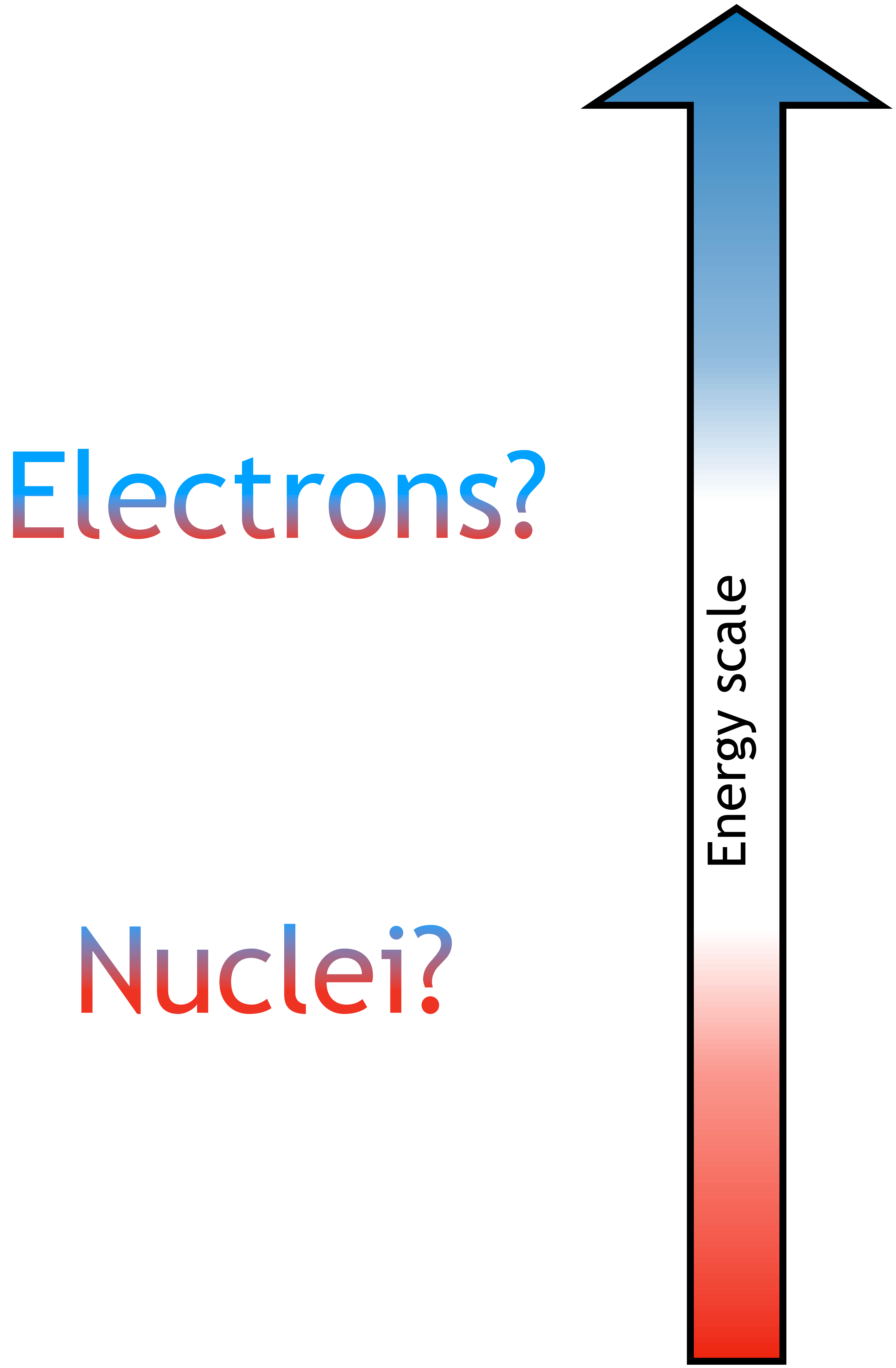
$\epsilon(R_1, R_2, \ldots)$: Low energy Born-Oppenheimer surface



Beyond static forces

$$f_i(R_1, R_2, \ldots) = \nabla_{R_i}\epsilon(R_1, R_2, \ldots)$$

$\epsilon(R_1, R_2, \ldots)$: Low energy Born-Oppenheimer surface insufficient for capturing TR broken dynamics



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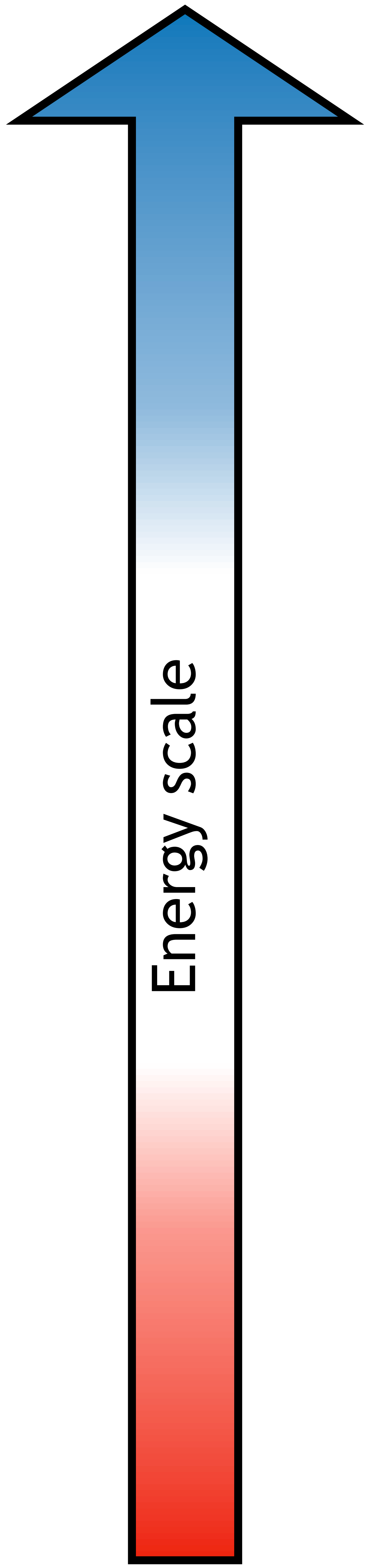
$\epsilon(R_1, R_2, \ldots)$: Low energy Born-Oppenheimer surface insufficient for capturing TR broken dynamics

Even in **insulating** systems with large electronic gaps

Electrons?

Nuclei?



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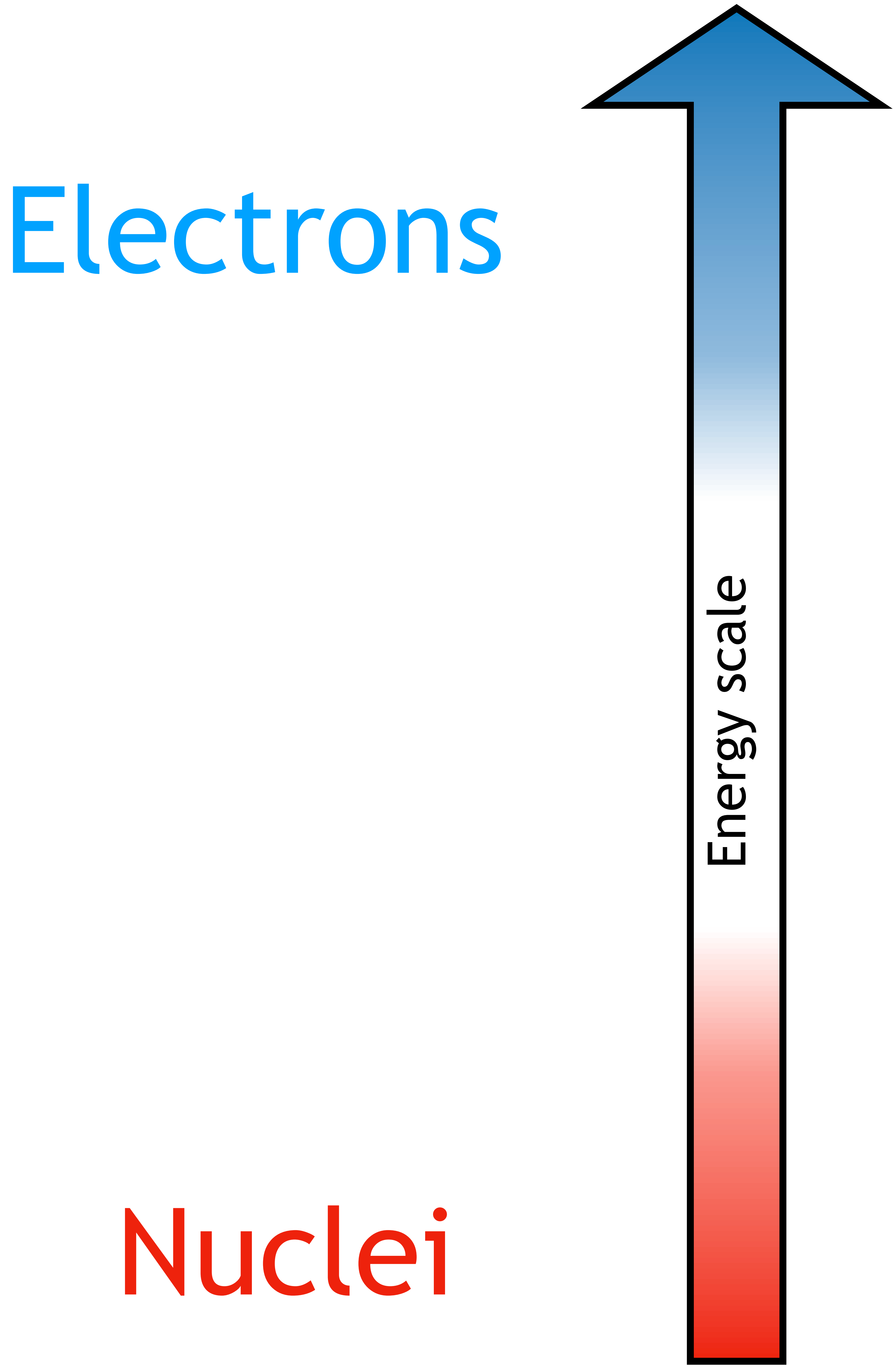
Even in **insulating** systems with large electronic gaps

Approaches:

Velocity-force constants:

Continue adiabatic perturbation expansion to include derivatives with $\dot{R}_i$



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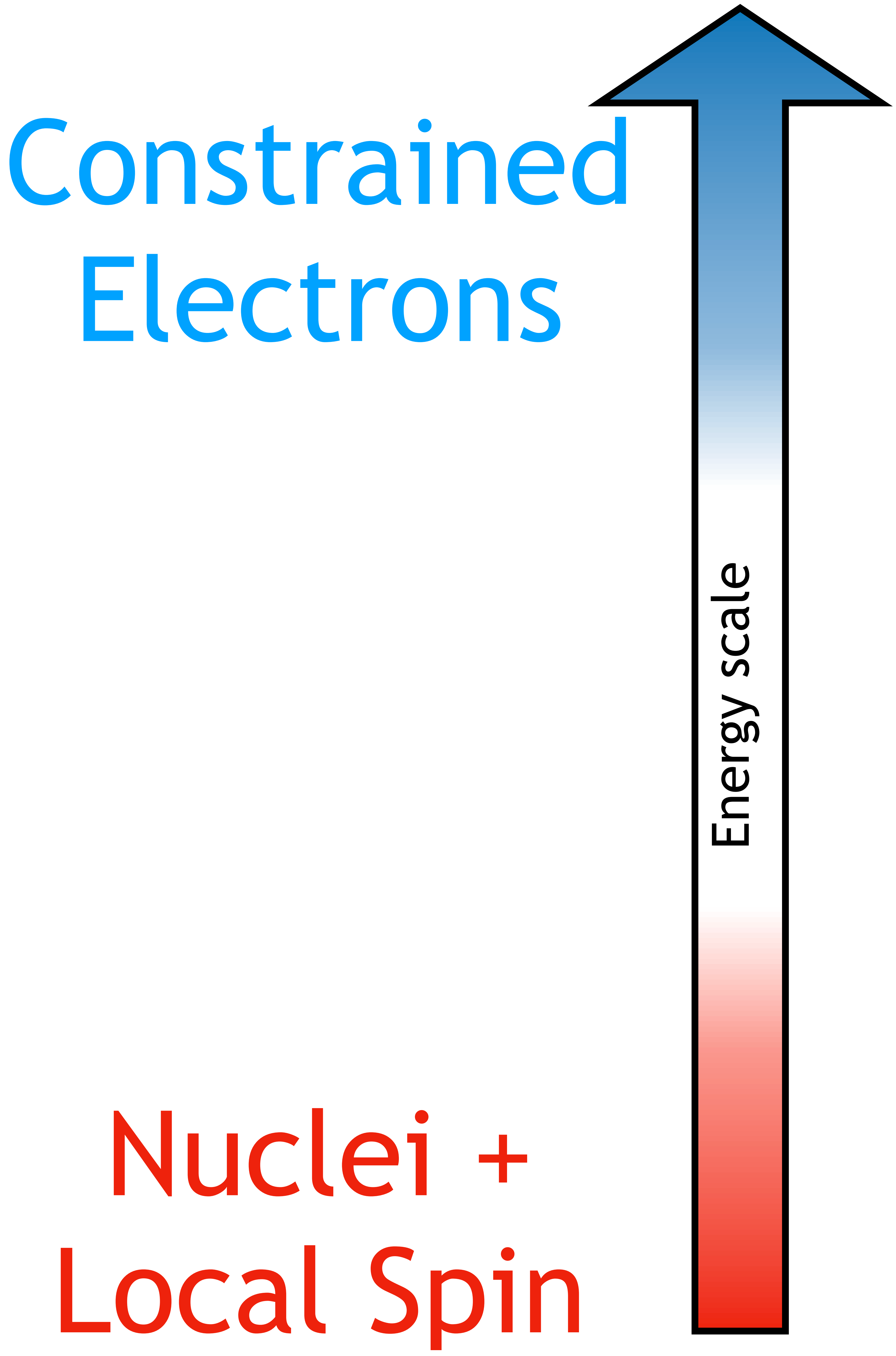
Even in **insulating** systems with large electronic gaps

Approaches:

Velocity-force constants:
Continue adiabatic perturbation expansion to include derivatives with $\dot{R}_i$

Beyond adiabatic phonon response:
Treat local spin and nuclei on the same footing



Velocity force constants

$$(T_{\text{nuc}} + T_e + V)\,|\,\Psi_{e-\text{nuc}}\rangle = W\,|\,\Psi_{e-\text{nuc}}\rangle$$

Velocity force constants

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Born-Oppenheimer:

$$|\Psi_{e-\text{nuc}}\rangle \approx |\psi_e(R_{\text{nuc}})\rangle\chi(R_{\text{nuc}})$$

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"Nuclear Berry Potential"

Velocity force constants

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"Nuclear Berry Potential"

Harmonic
Semi-classical EOM:

$$m_I\frac{d^2R_i}{dt^2} = -\sum_j C_{ij}\Delta R_j + \sum_j G_{ij}\frac{d\Delta R_j}{dt}$$

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"Nuclear Berry Potential"

Harmonic Semi-classical EOM:

"Nuclear Berry Curvature" $G_{ij} = 2\hbar\,\text{Im}\langle\frac{\partial\psi_e}{\partial R_i}|\frac{\partial\psi_e}{\partial R_j}\rangle$

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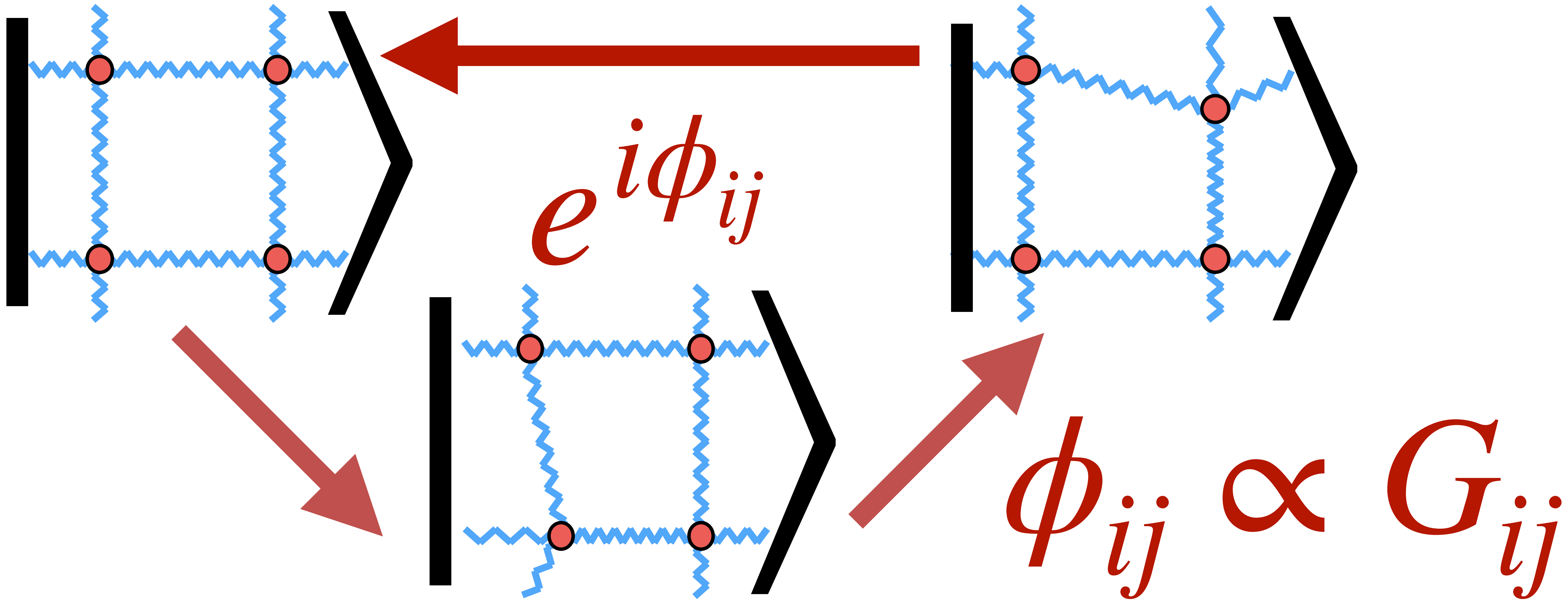

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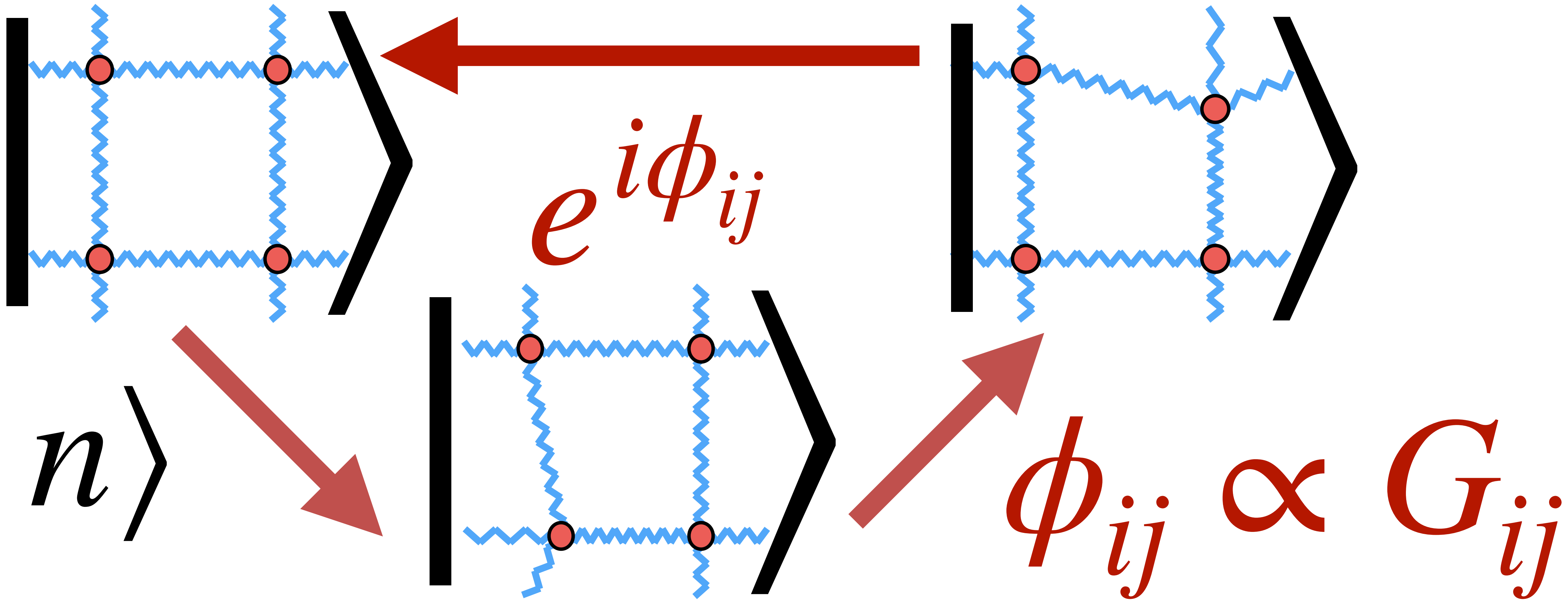
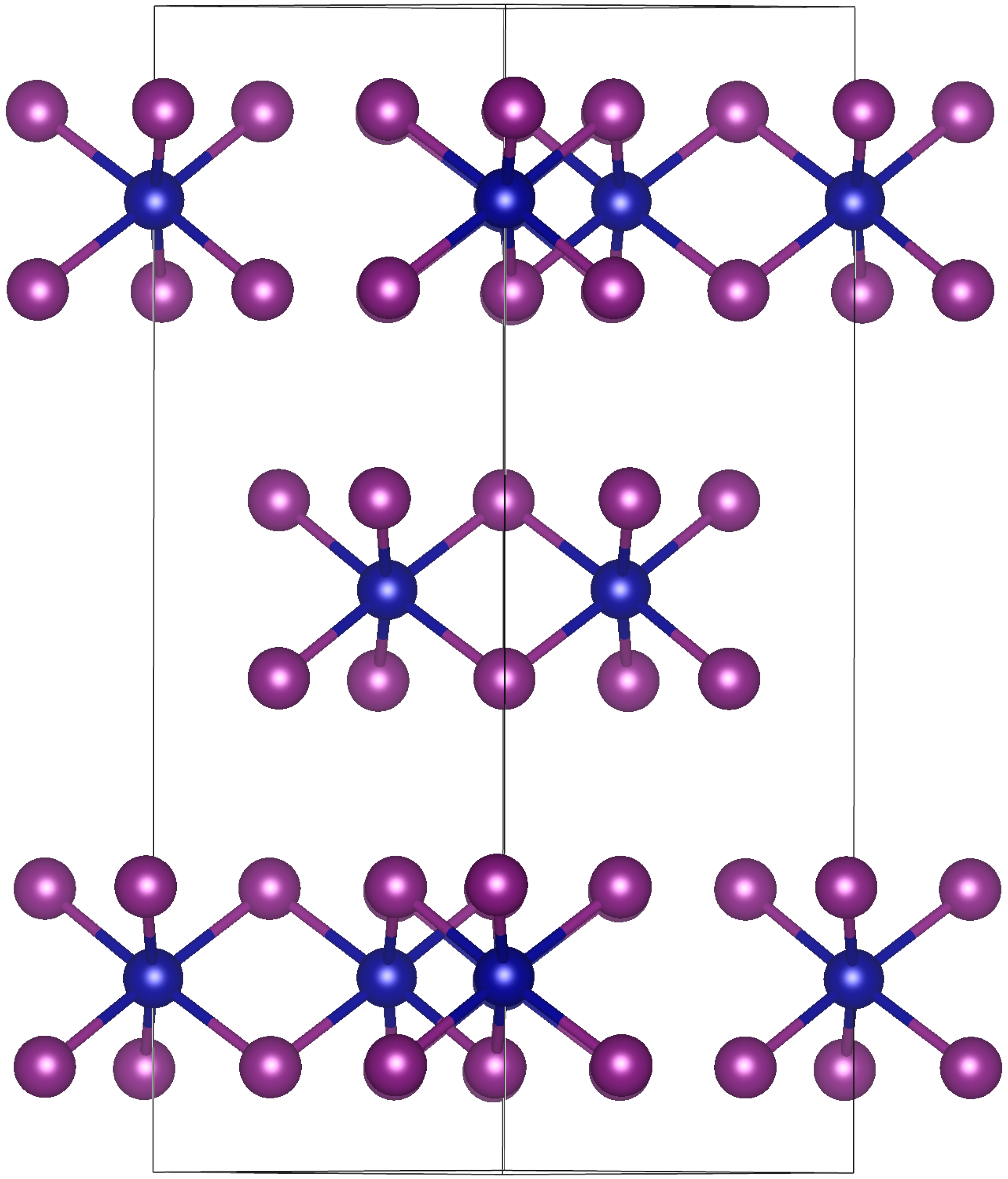
$$\mathbf{M}\omega_n^2|n\rangle = (\mathbf{C} + i\omega_n\mathbf{G})|n\rangle$$



Example system: Γ modes of CrI_3

CrI_3

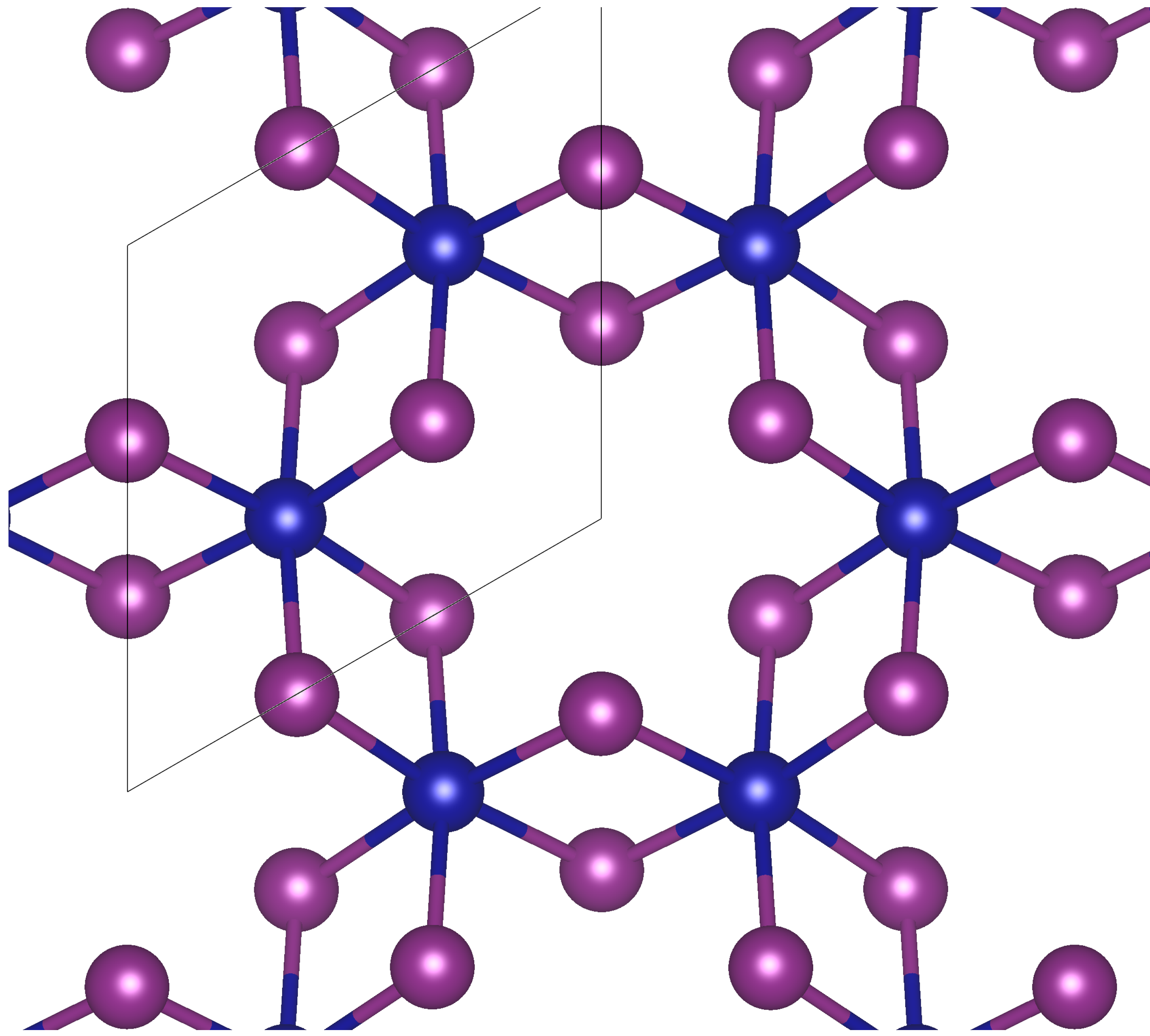
- **Ferromagnetic** insulator (~1eV gap) w/**strong SOC**



Cr

I

Single layer top view:



Example system: Γ modes of CrI_3

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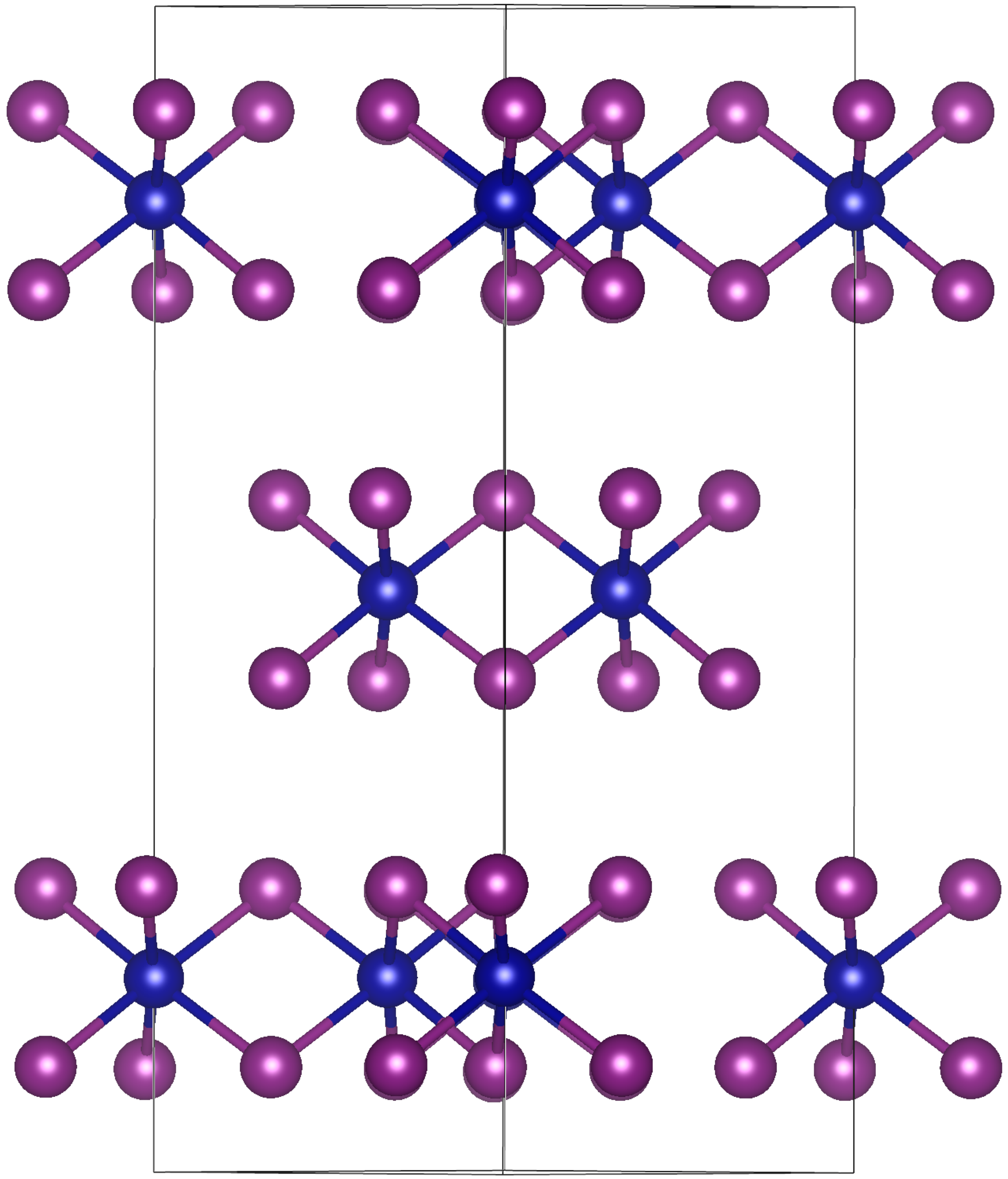
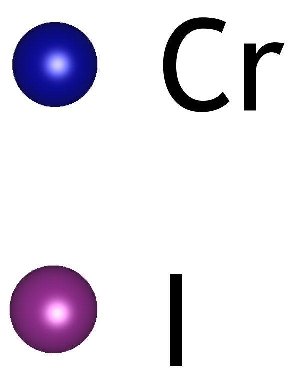
- **Ferromagnetic** insulator (~1eV gap) w/**strong SOC**

$R\overline{3}$ space group

- At Γ space group has **2D irreps**
  - →degenerate phonon modes





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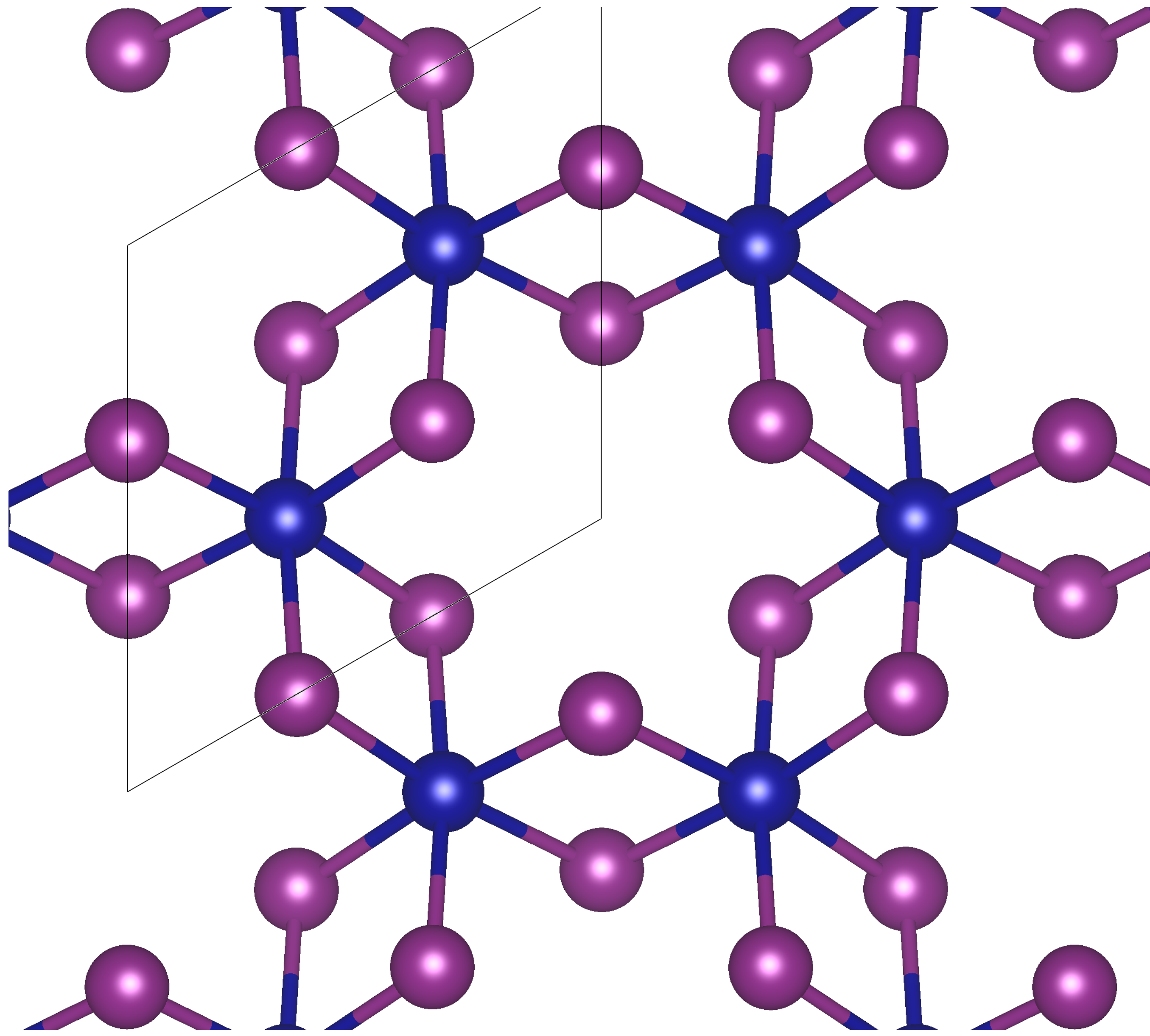


Example system: Γ modes of CrI_3

CrI_3

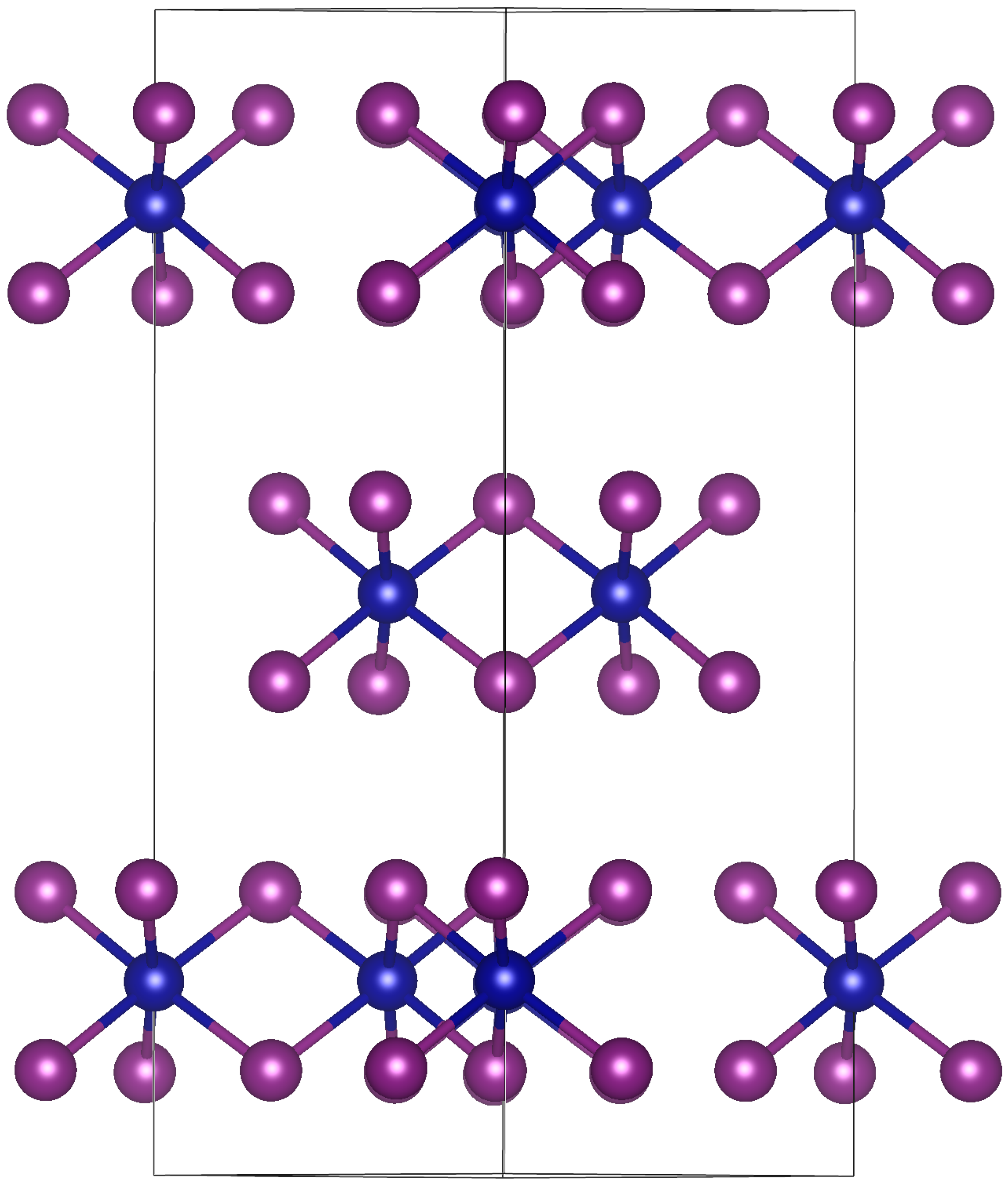
Ferromagnetic insulator
(~1eV gap) w/**strong SOC**

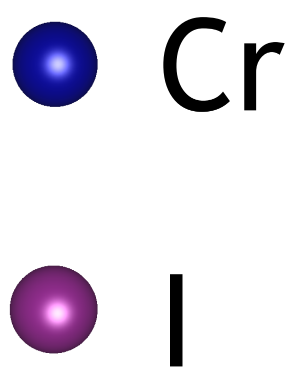
$R\bar{3}$ space group

At Γ space group has **2D irreps**
→degenerate phonon modes

Time reversal is broken

Magnetic space group has only **1D irreps**
→no symmetry enforced degeneracy





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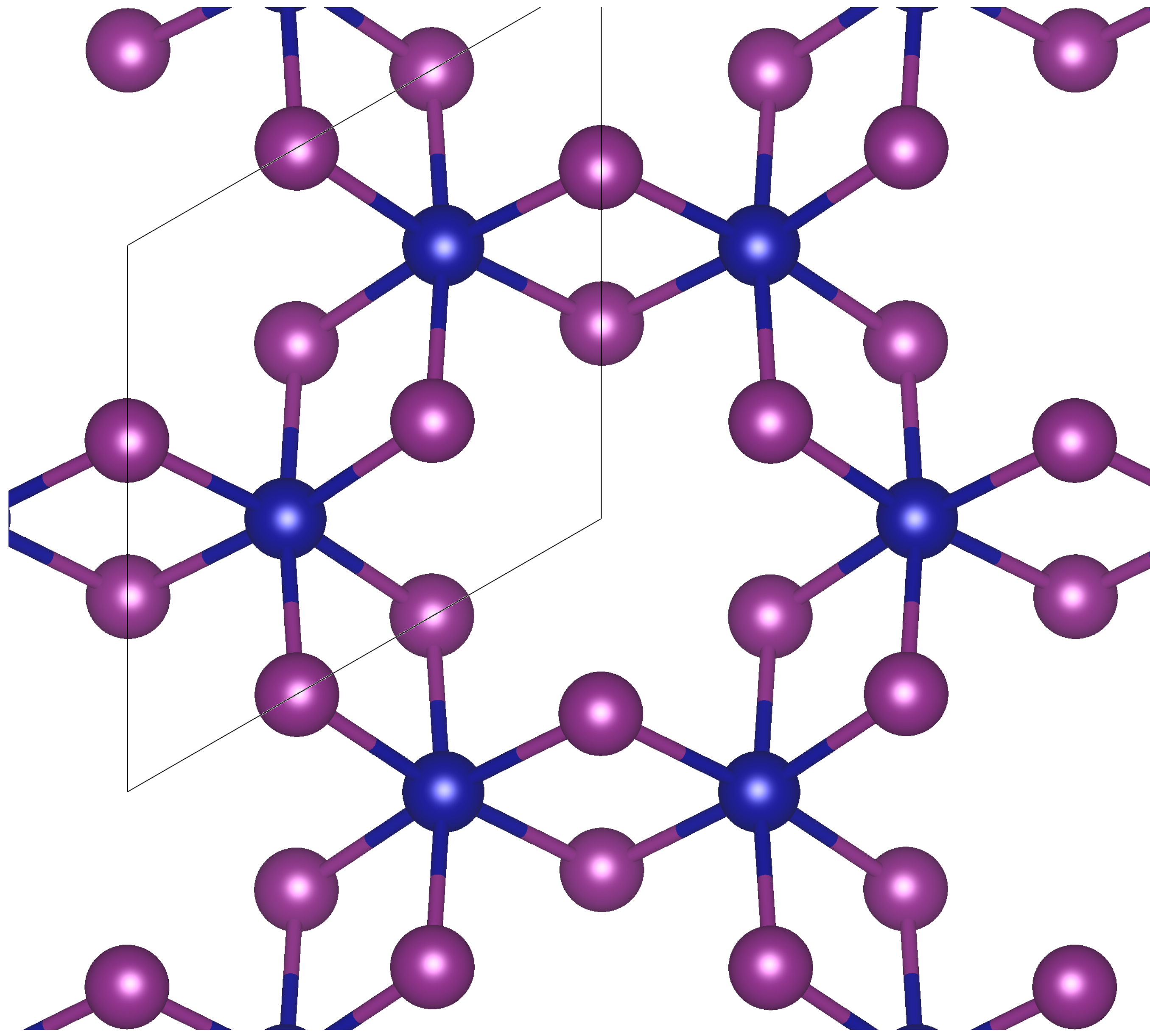
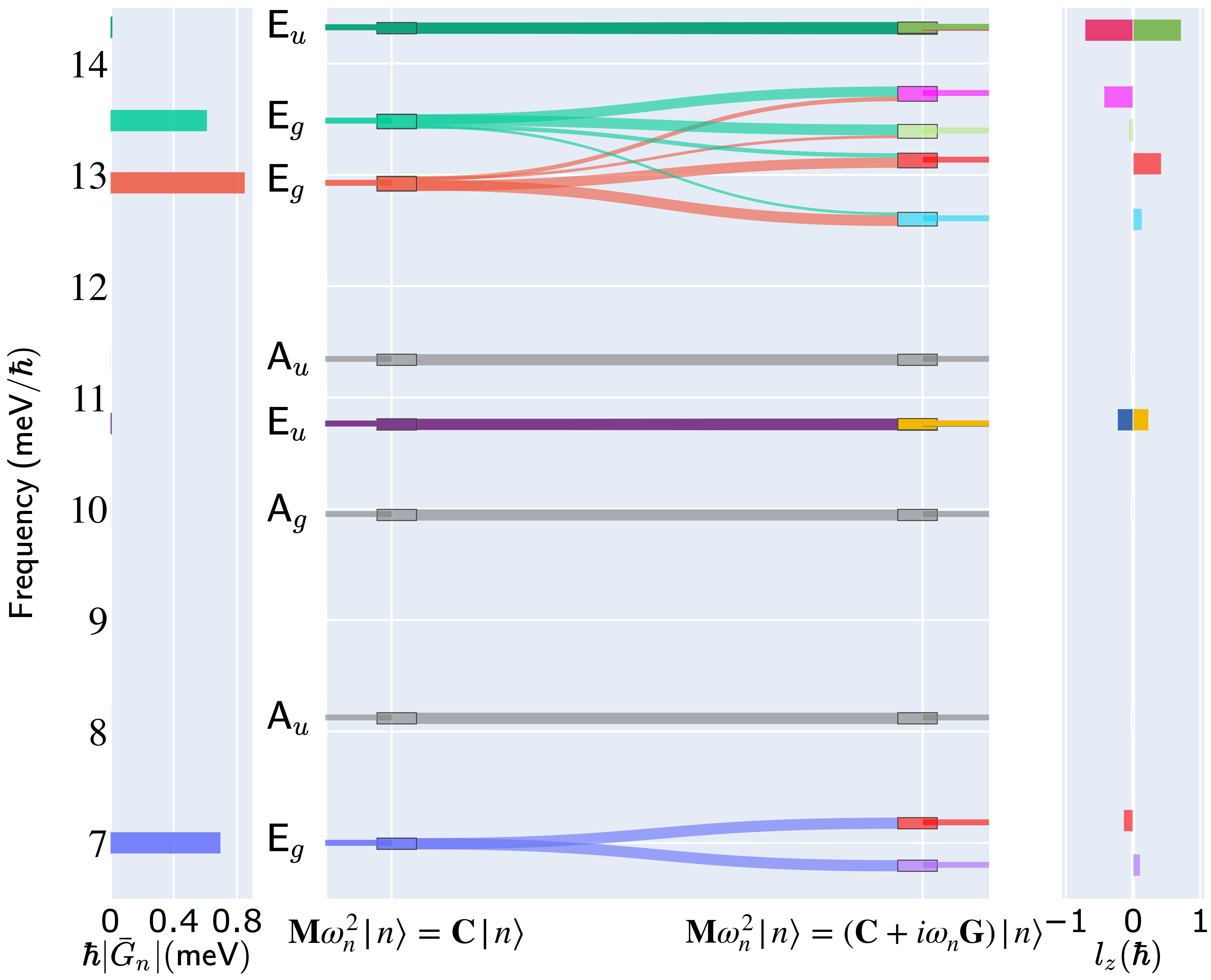


Adiabatic theory: Results for CrI_3 Γ phonons

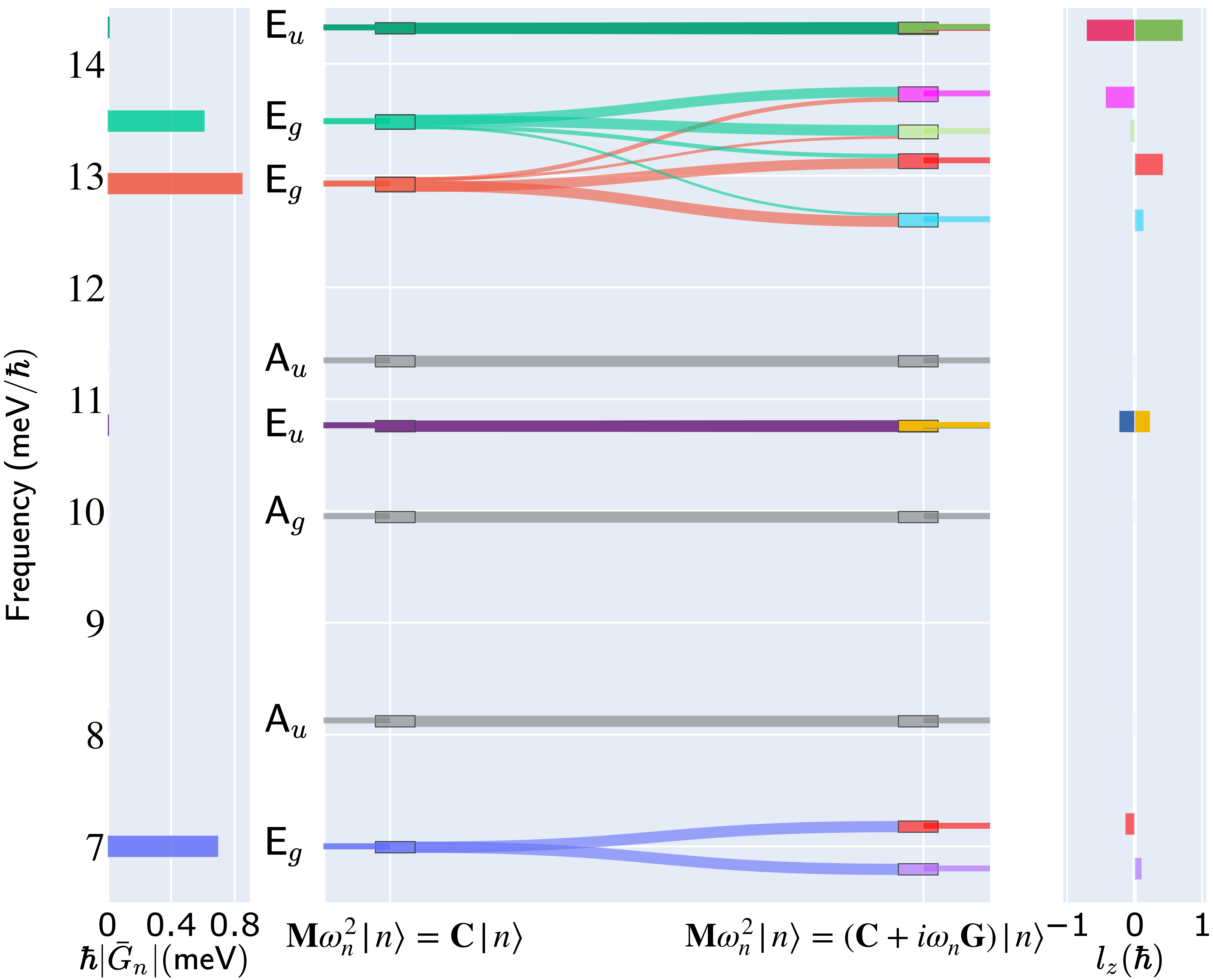


Mode degeneracies now correctly correspond to the irreps of the *magnetic symmetry group*

Degenerate pairs split to chiral phonons with angular momentum

Large splittings for E_g modes

Adiabatic theory: Results for CrI₃ Γ phonons



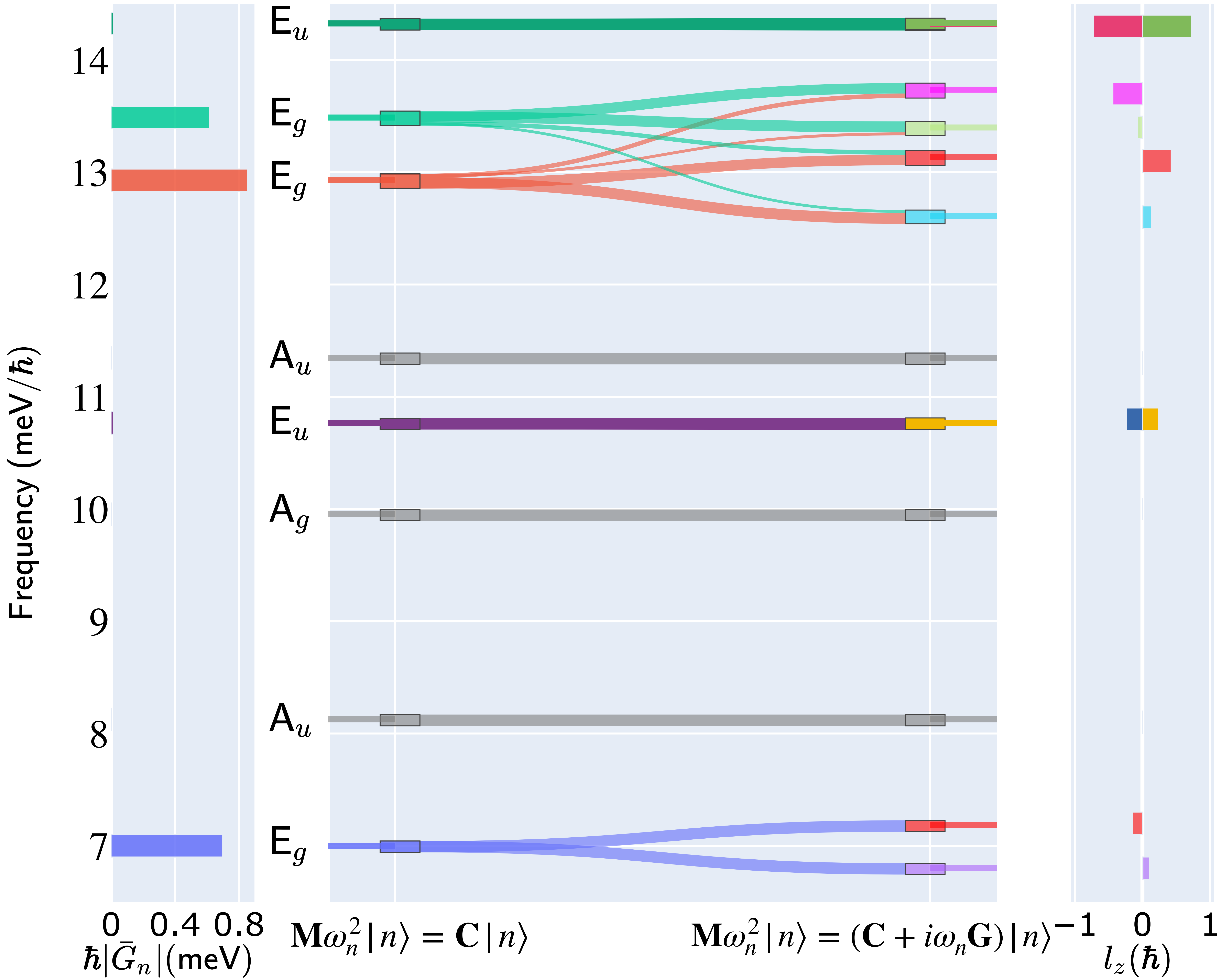
Mode degeneracies now correctly correspond to the irreps of the *magnetic symmetry group*

Degenerate pairs split to chiral phonons with angular momentum

Large splittings for E_g modes

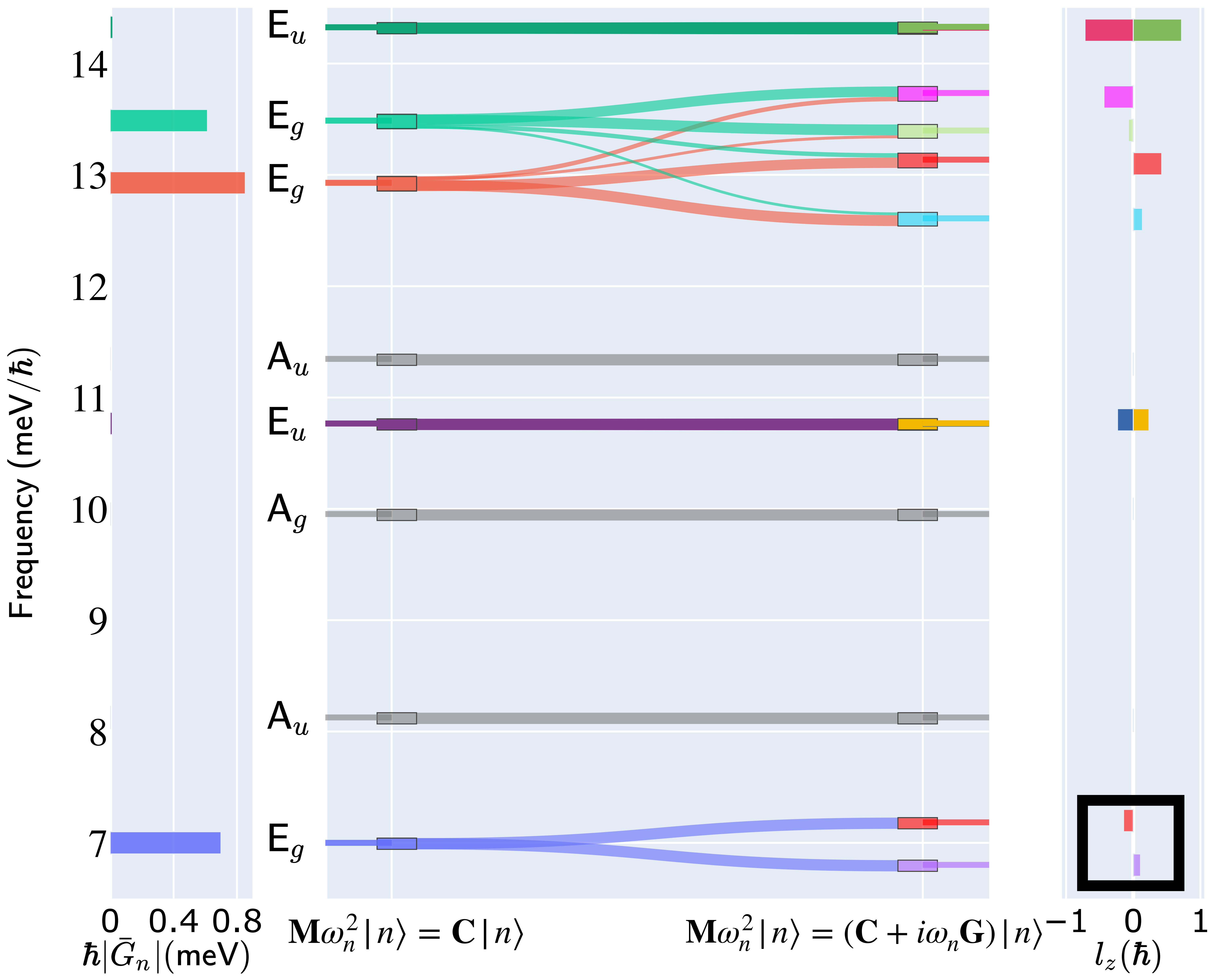
…unbelievably large

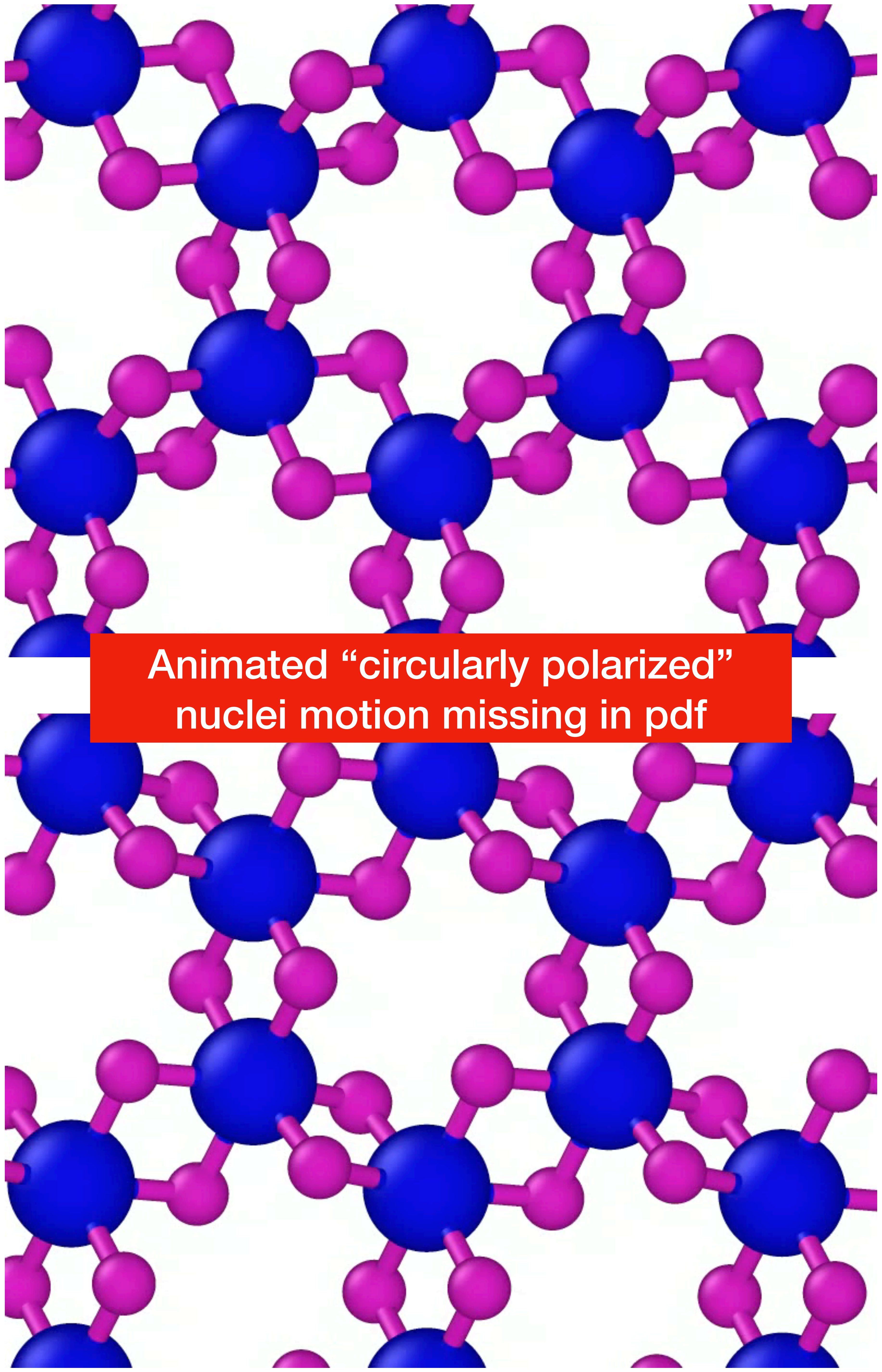
Adiabatic theory: Results for CrI_3



Animated "circularly polarized" nuclei motion missing in pdf

Adiabatic theory: Results for CrI_3





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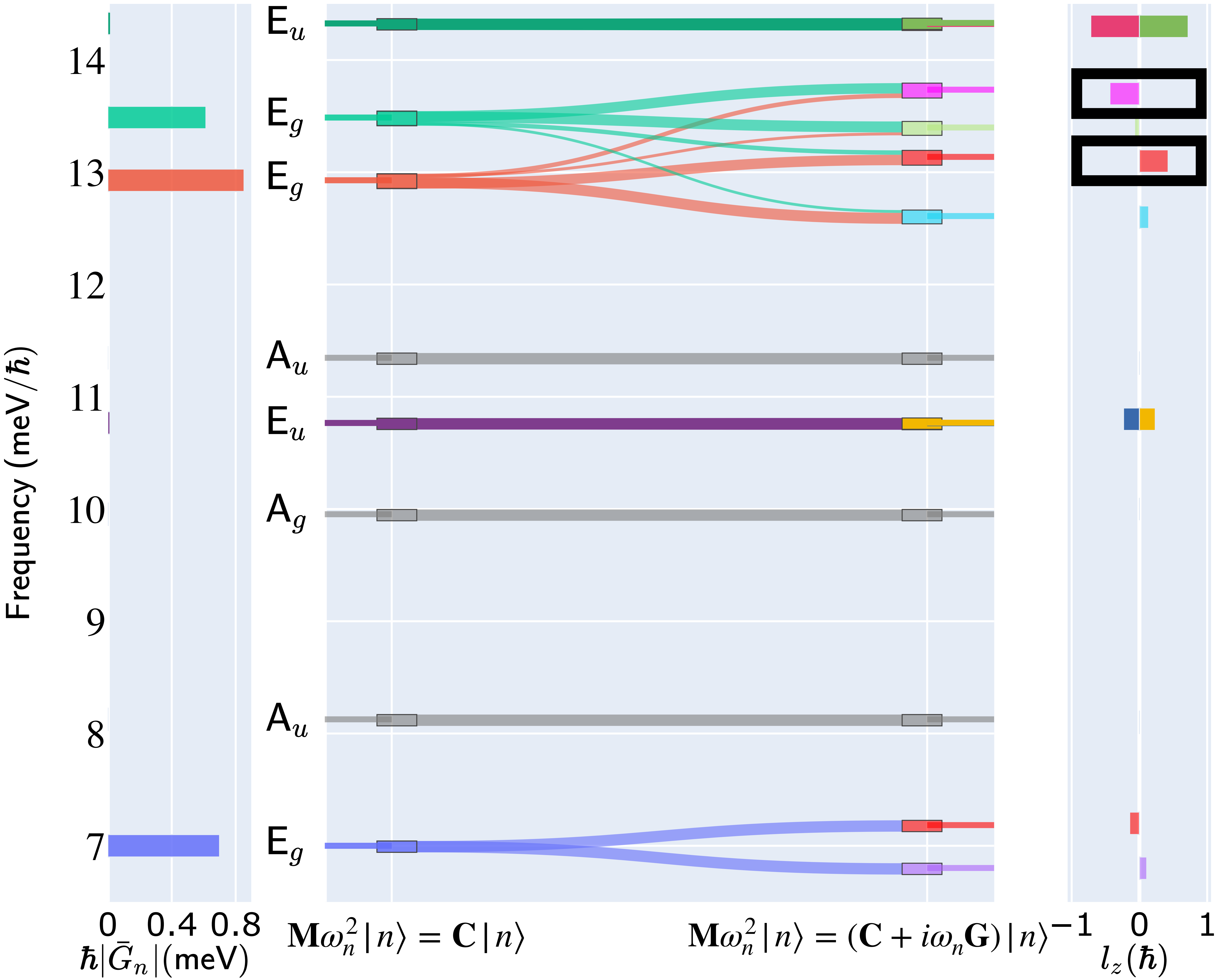
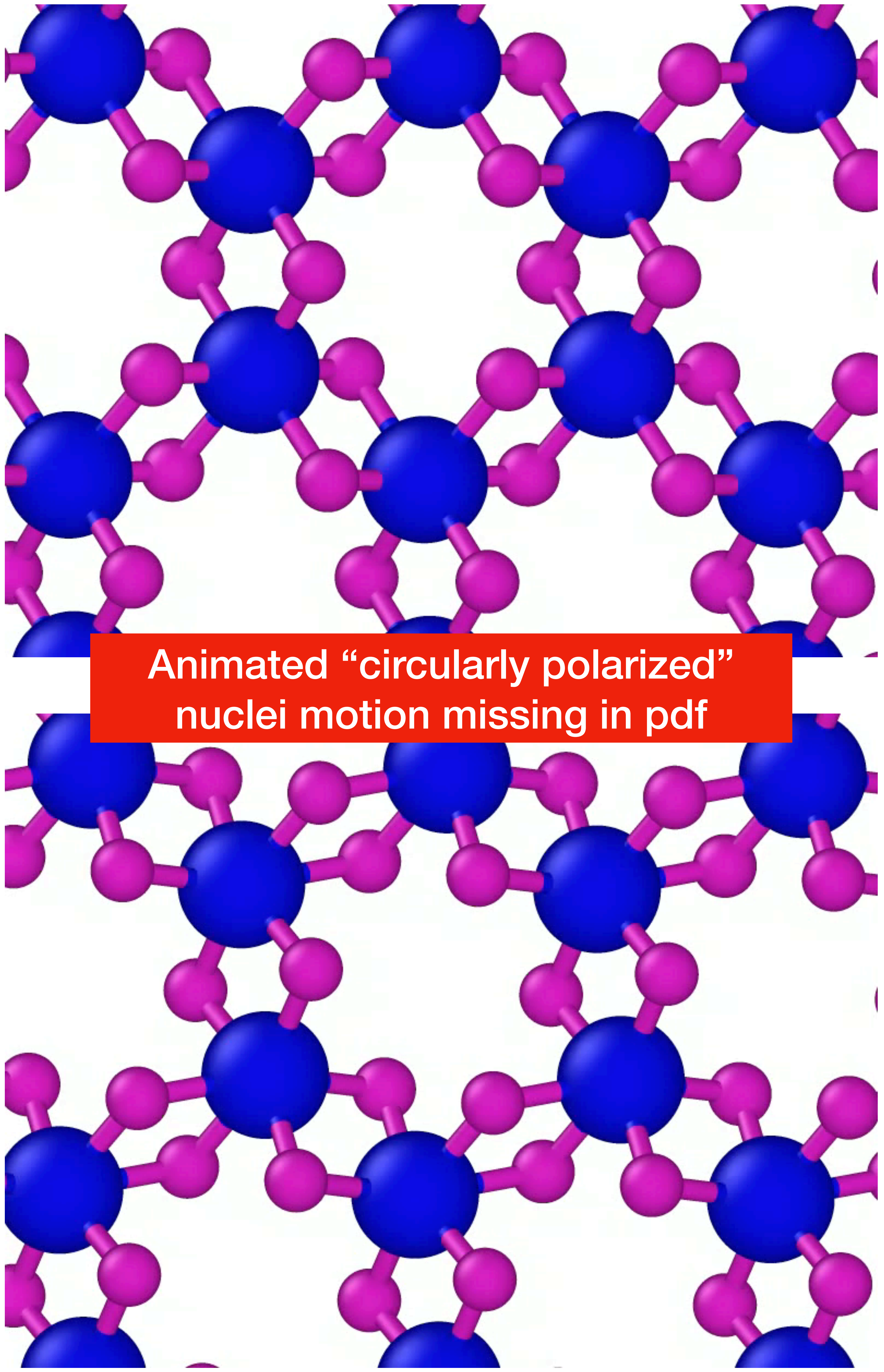
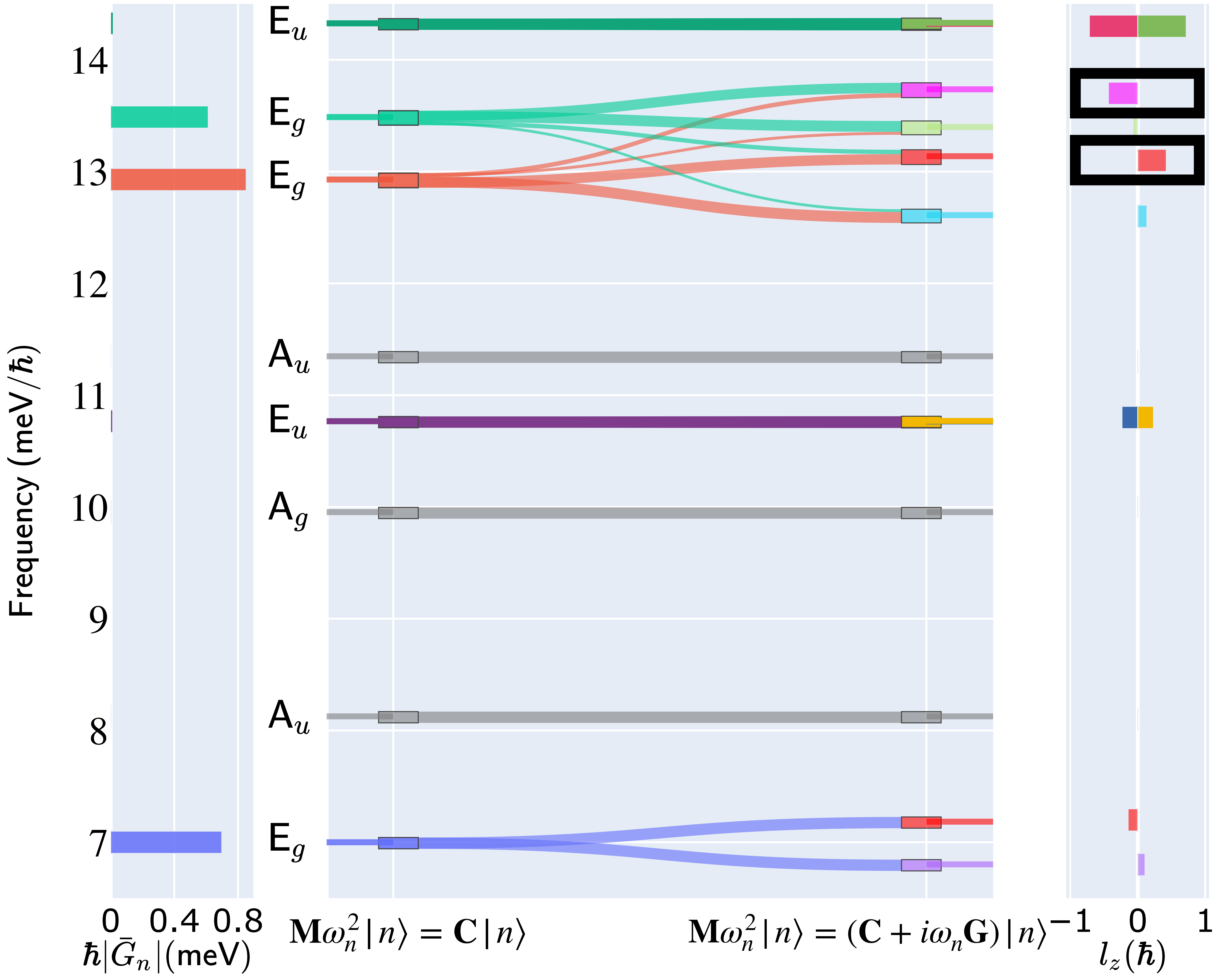
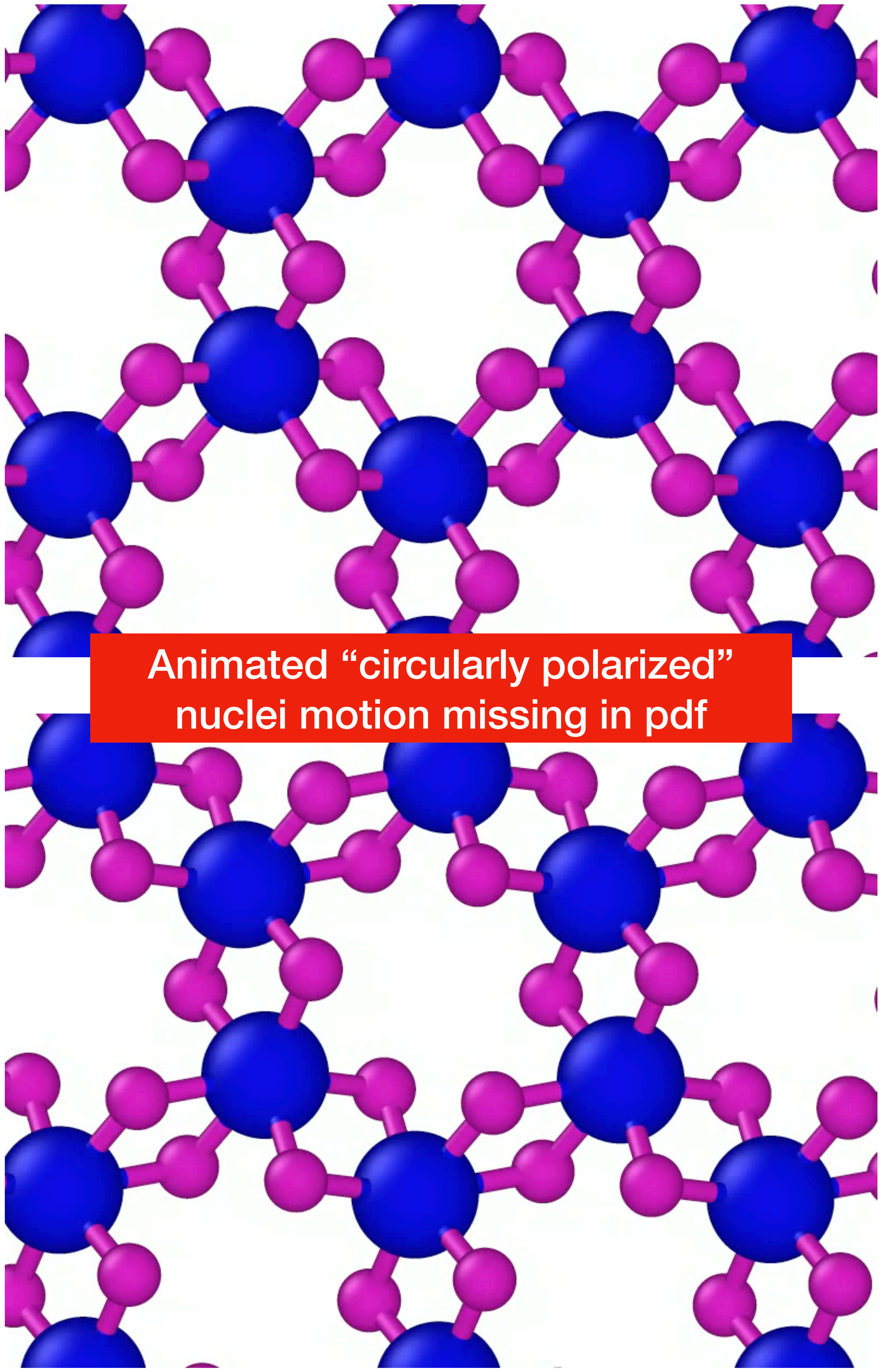


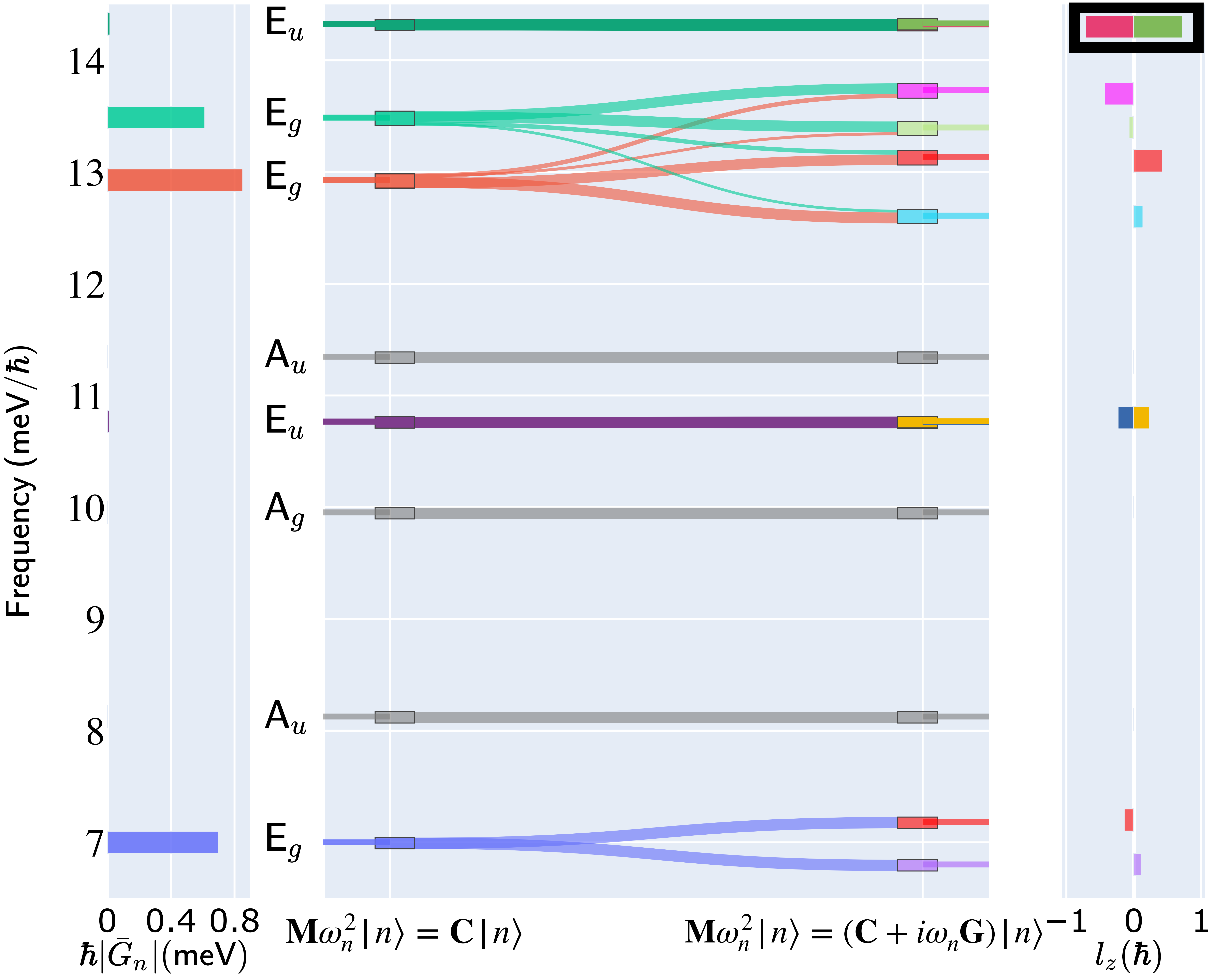


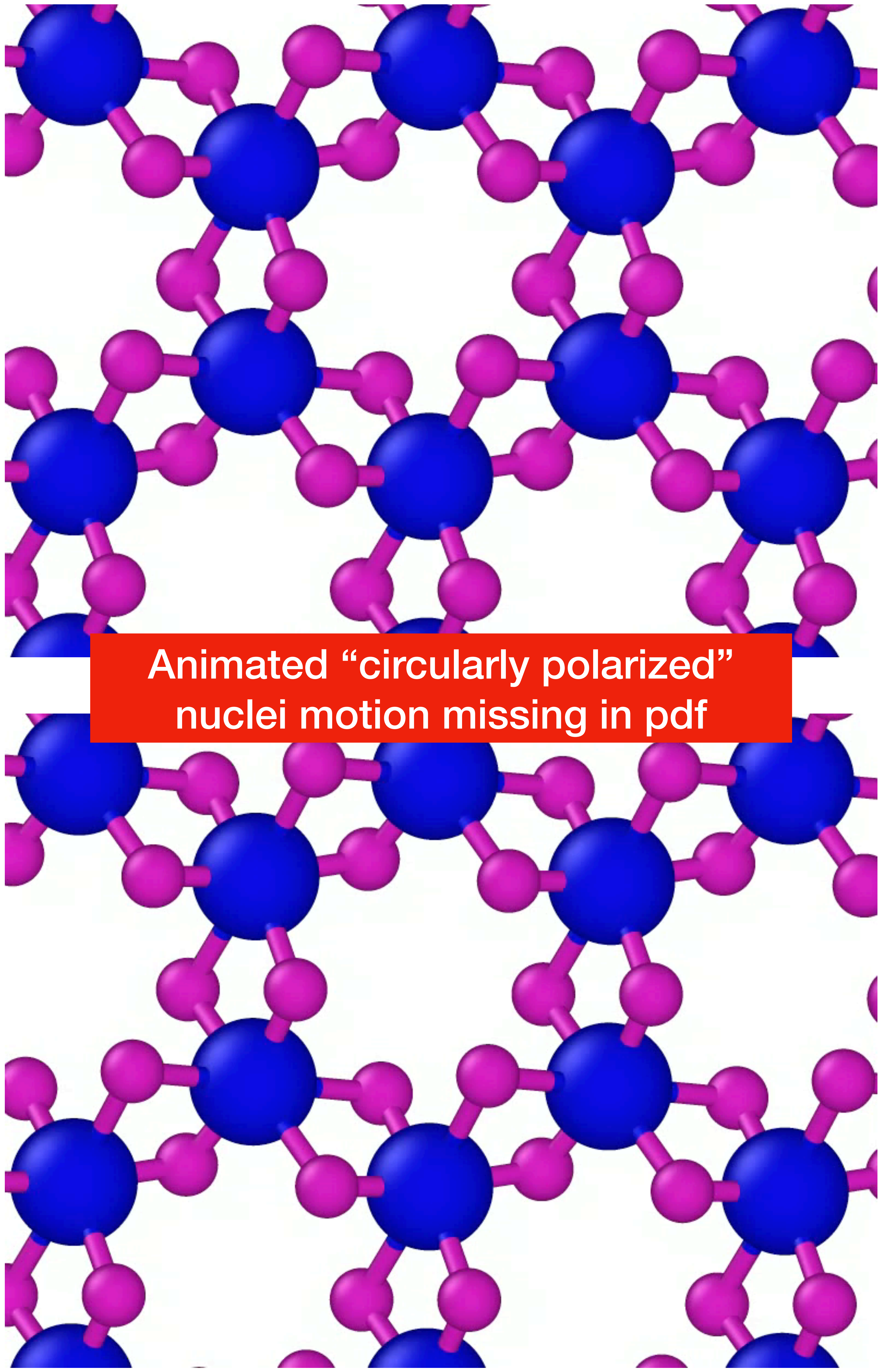
Adiabatic theory: Results for CrI_3



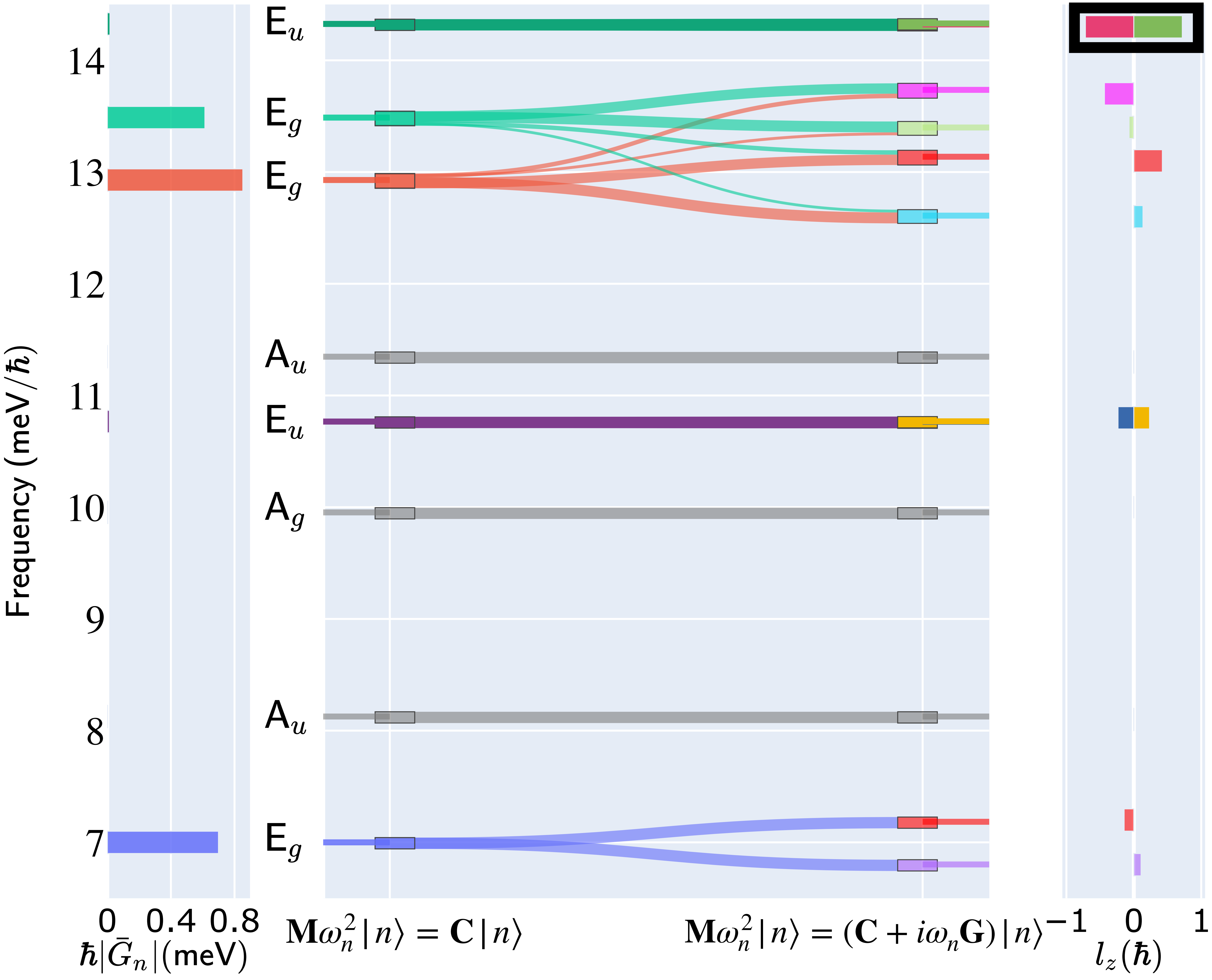


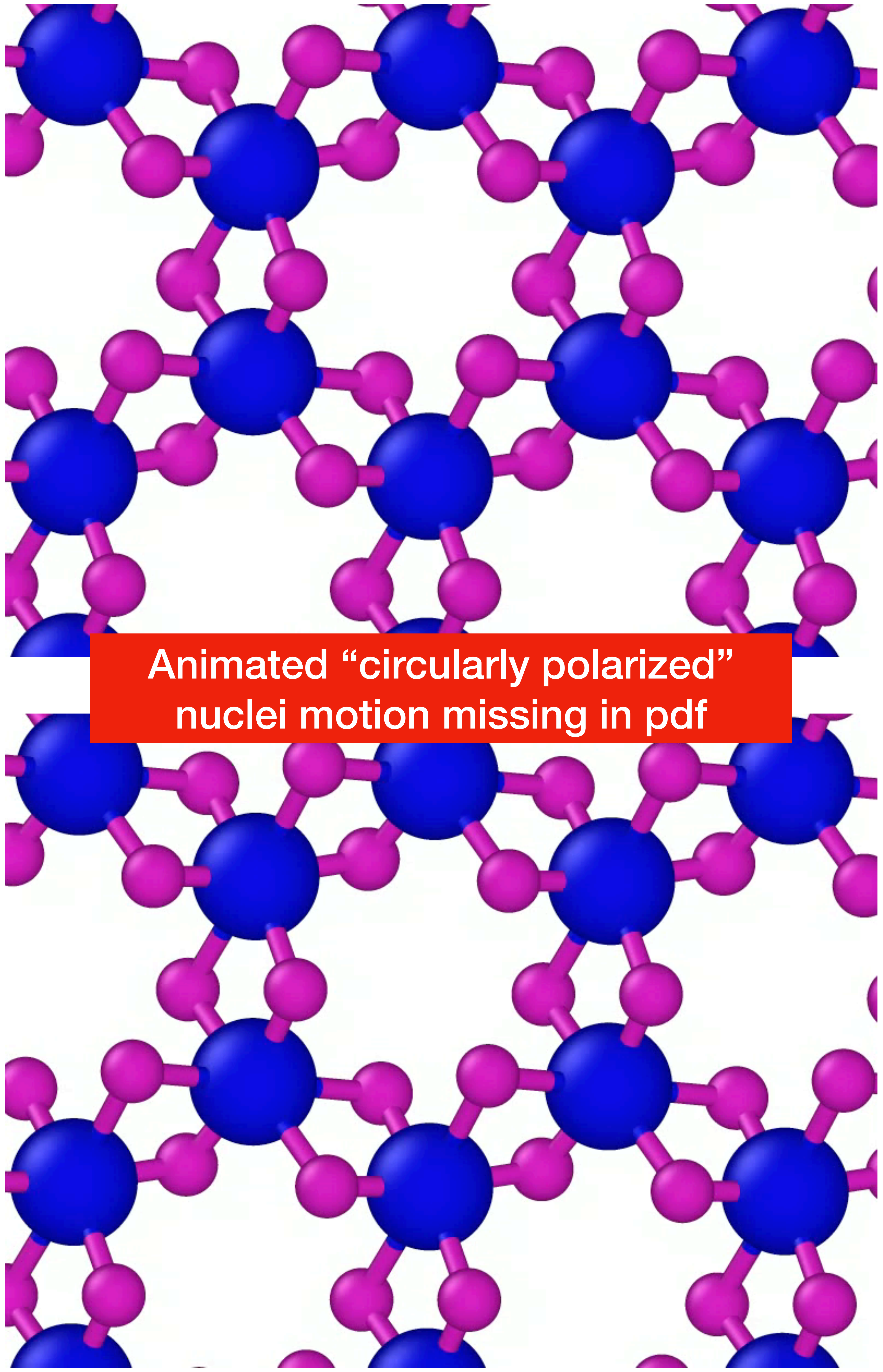
Adiabatic theory: Results for CrI_3





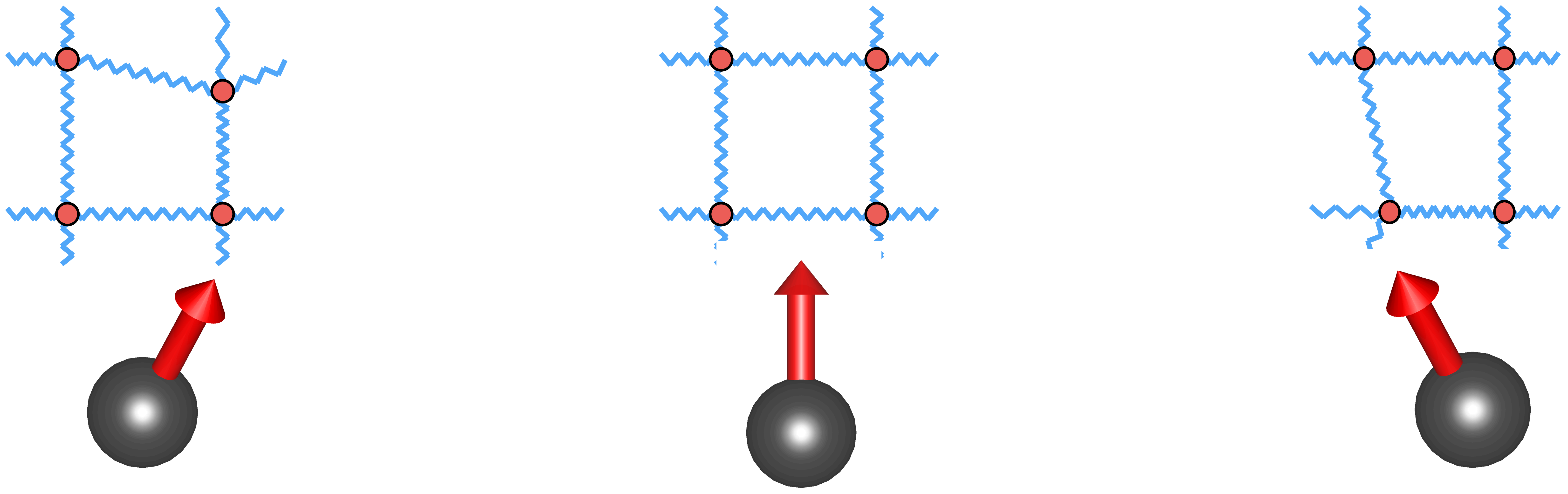
Adiabatic theory: Results for CrI₃





Magnon-phonon coupling

Lattice distortions couple to canting of local magnetization of Cr sites



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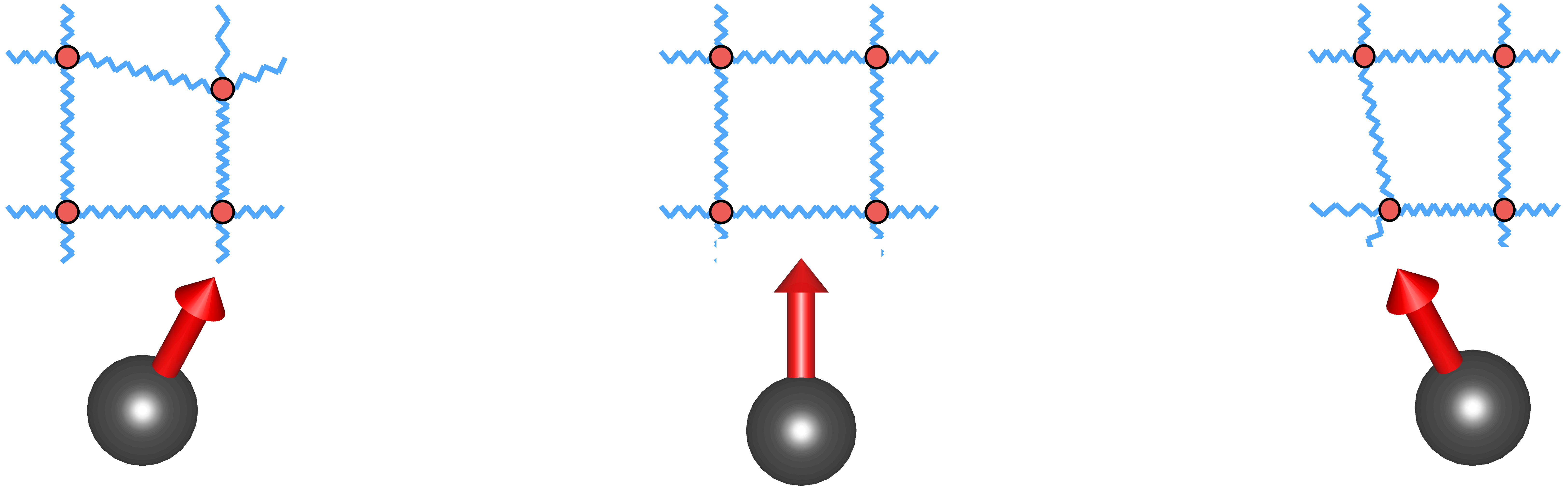
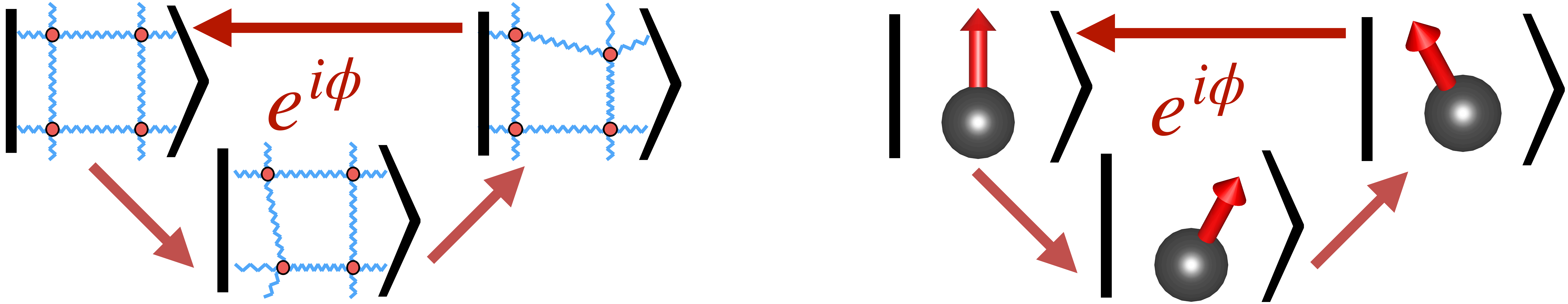
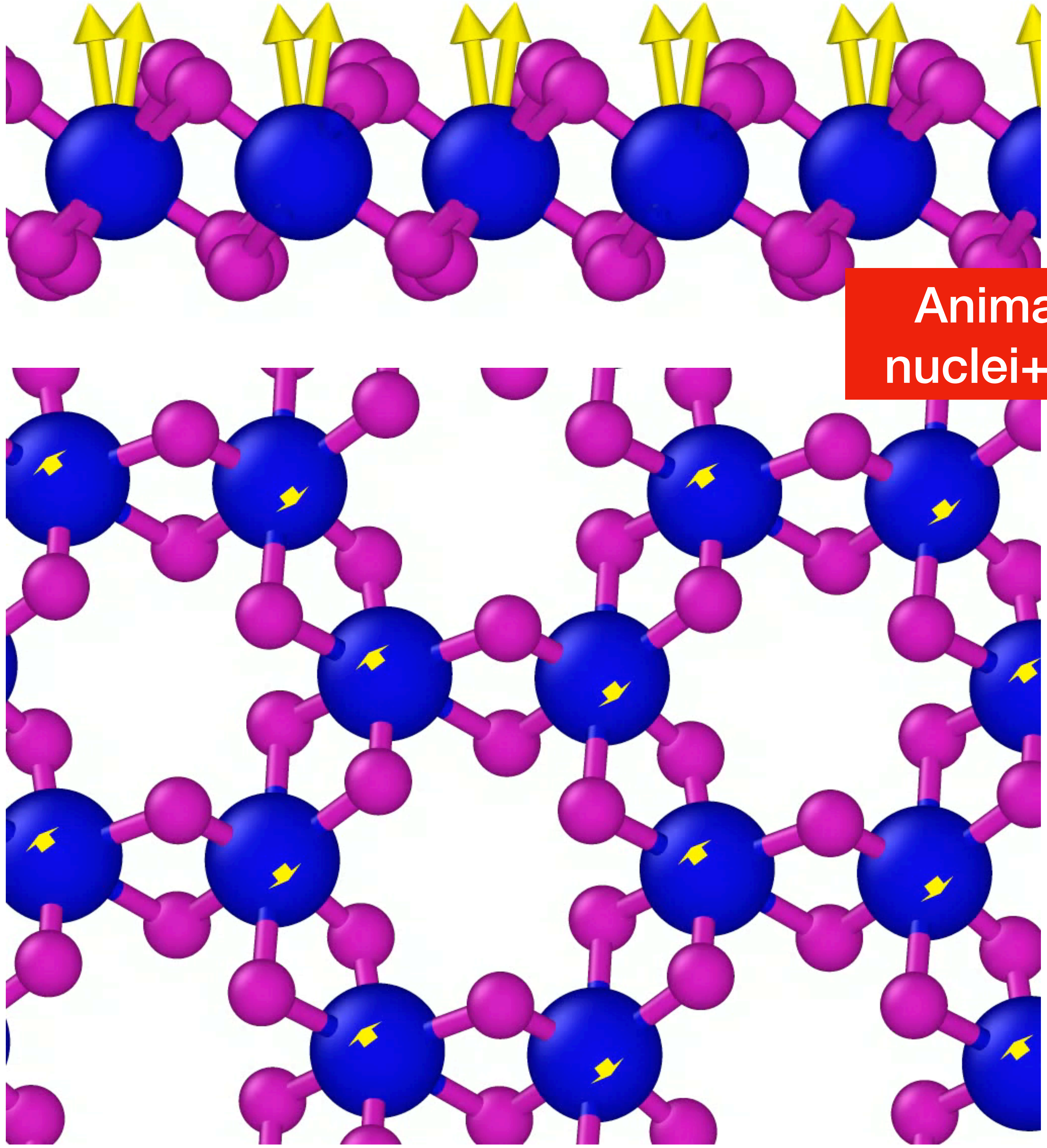


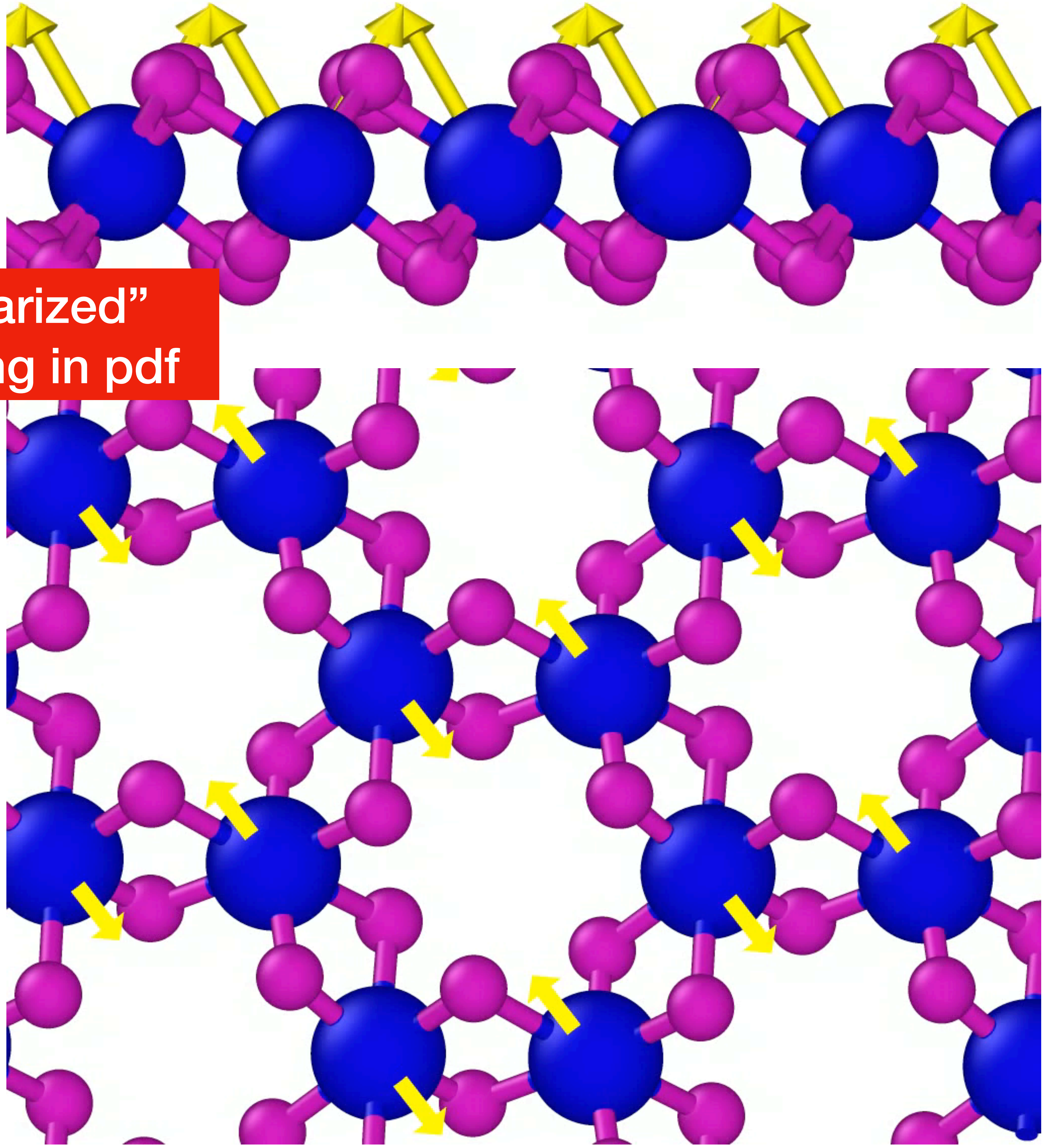
G matrix elements are almost entirely recovered by treating local moments as 3/2 spins and computing corresponding the spin Berry curvature



Magnon-phonon coupling



Animated “circularly polarized” nuclei+spin motion missing in pdf



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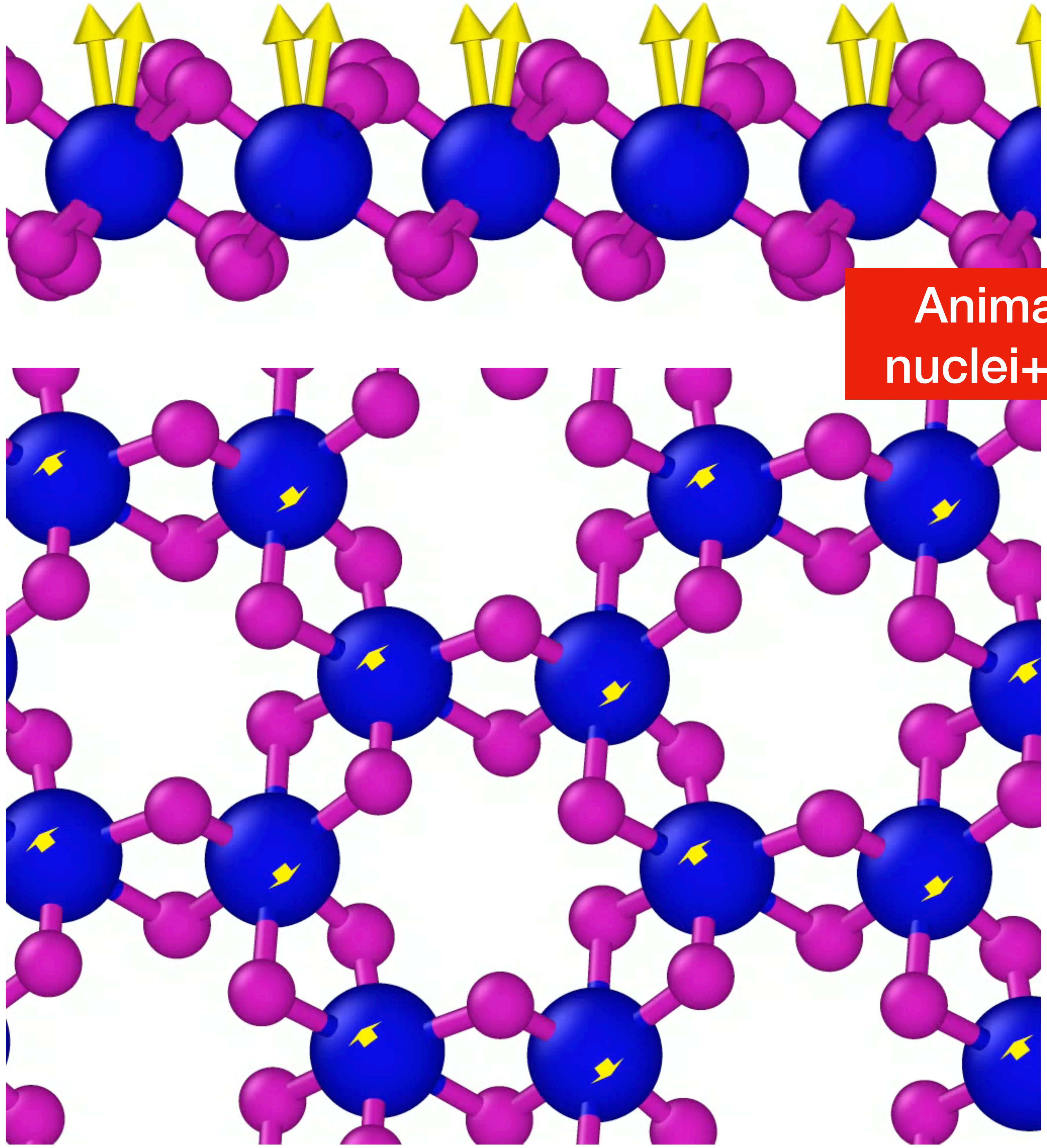

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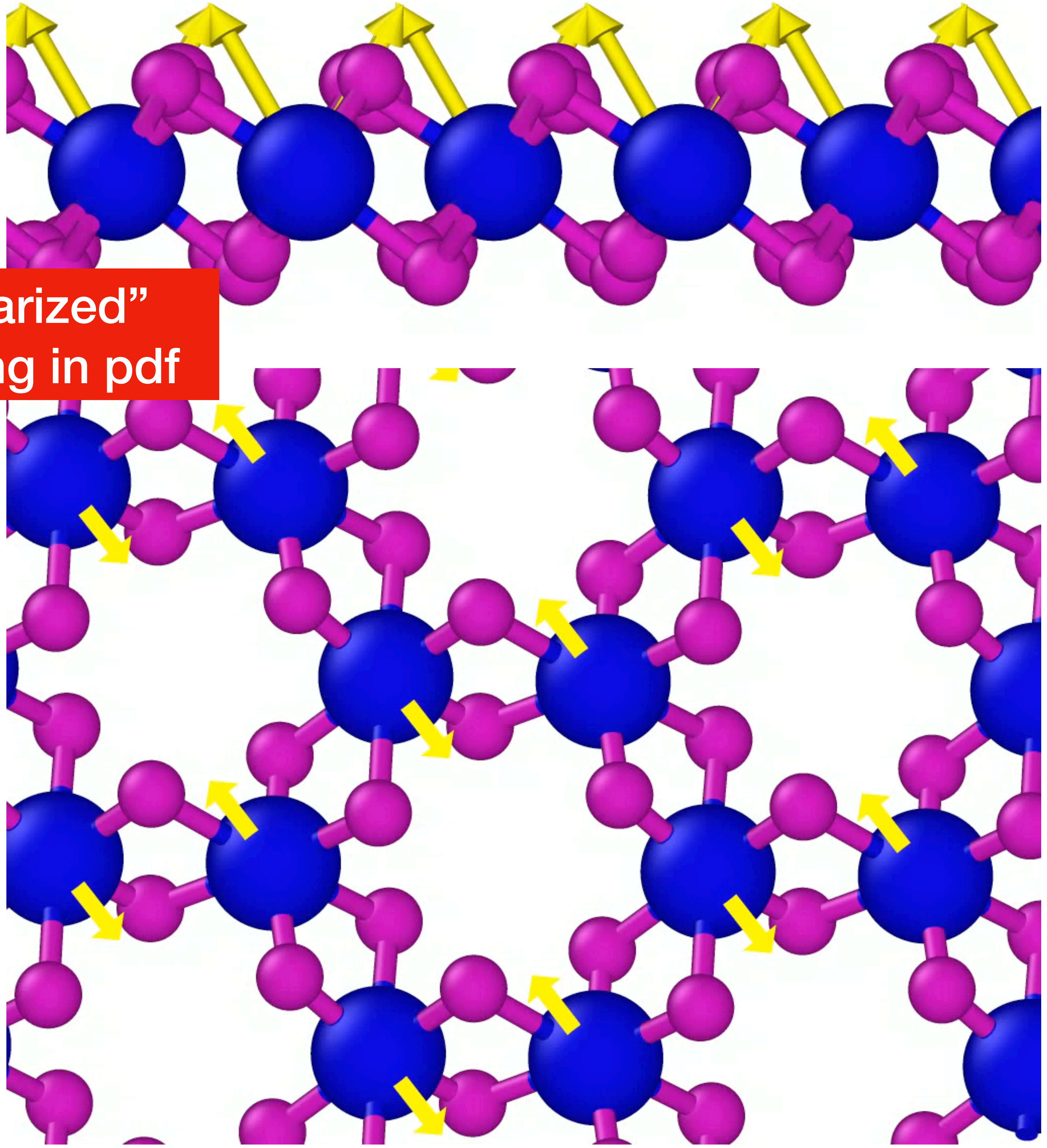

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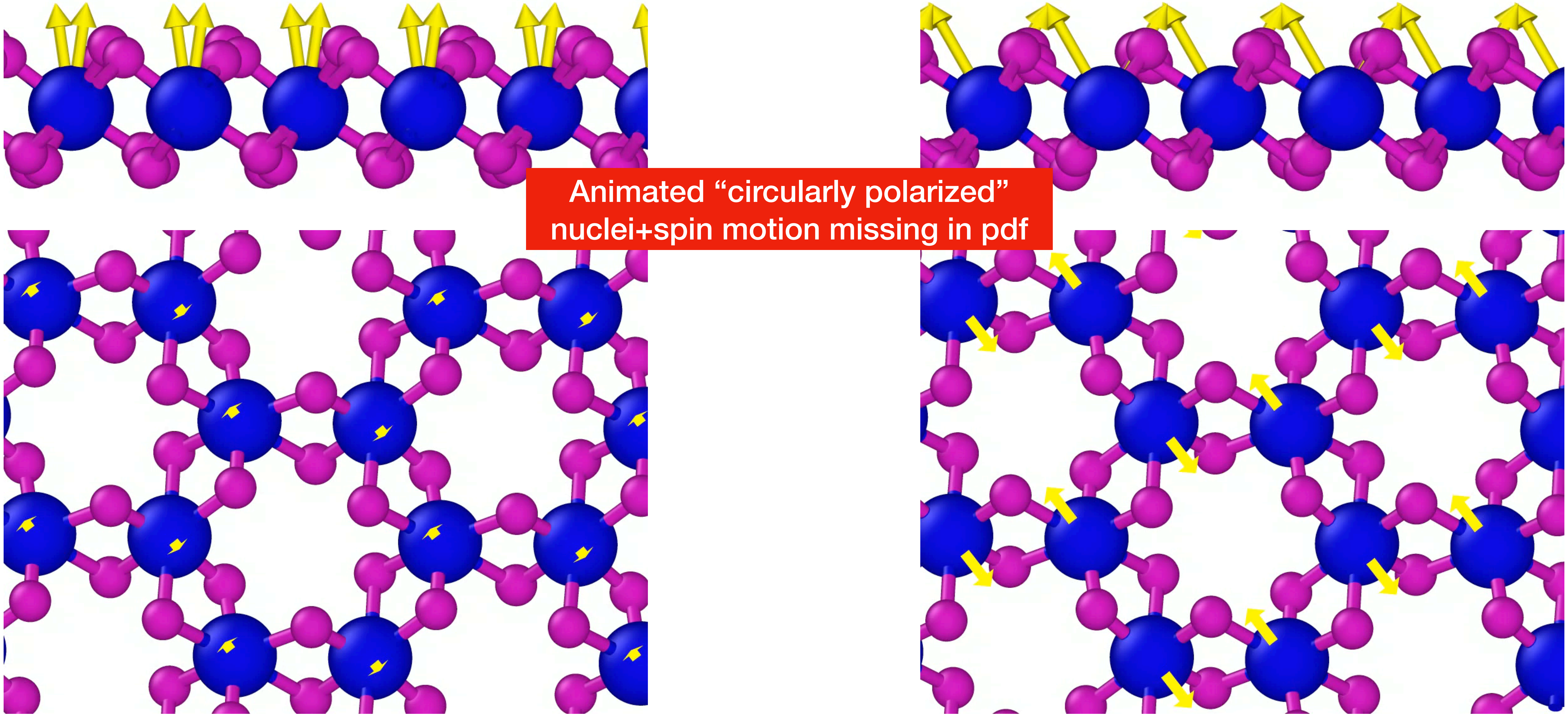

The adiabatic theory assumed electrons to be **fast** with respect to phonons.

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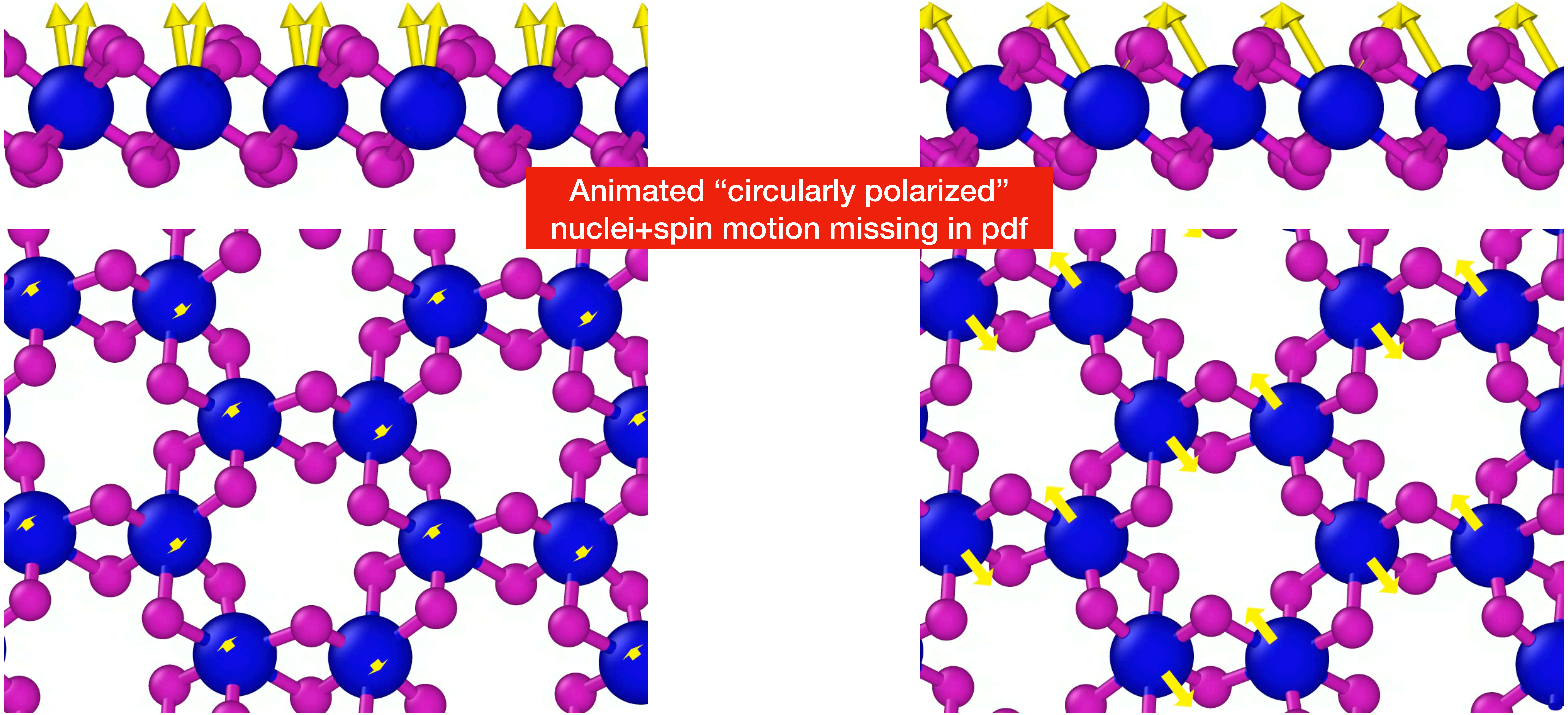


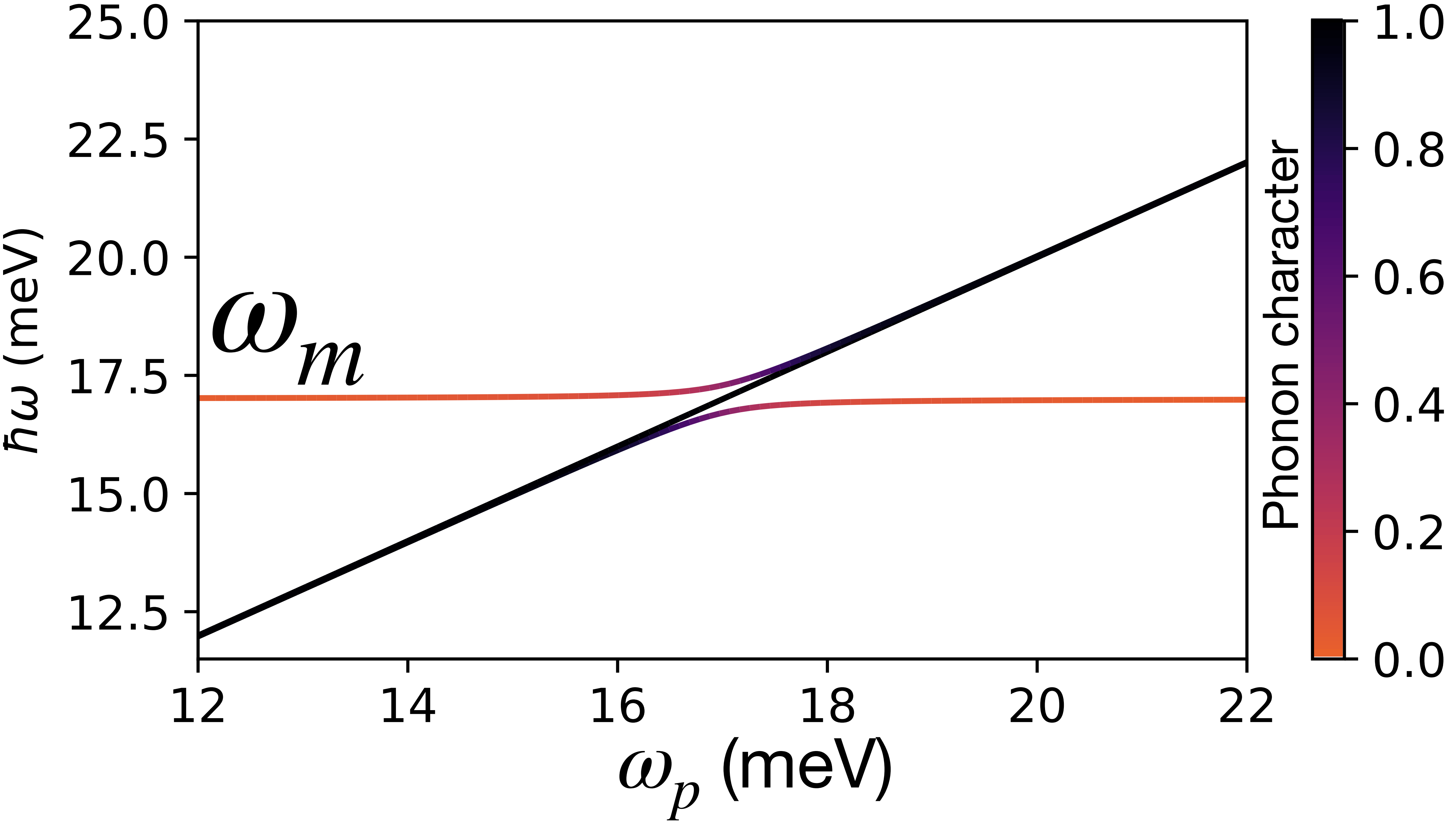
The adiabatic theory assumed electrons to be **fast** with respect to phonons.

Time scale for the spin canting is set by the magnon frequencies
same order (optical magnon) or **slower** (acoustic magnon) than the phonons!

Magnon-phonon coupling: simple model

$$H_{\rm sp} = \frac{1}{2}(p_x^2 + p_y^2) + \omega_p(x^2 + y^2)$$

$$+\frac{1}{2}S\omega_m(s_x^2 + s_y^2) + \gamma(xs_x + ys_y)$$



(meV/ħ)

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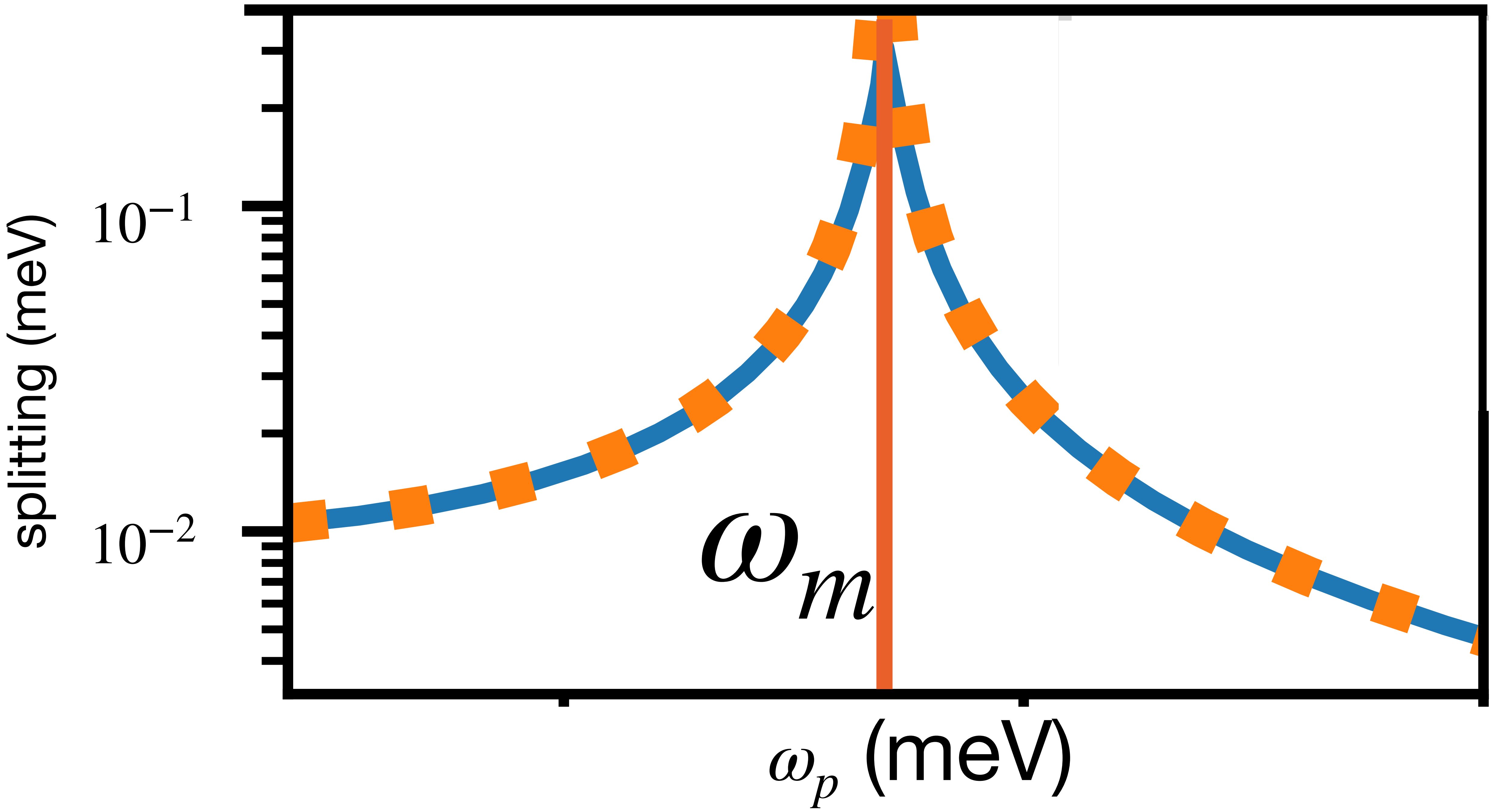
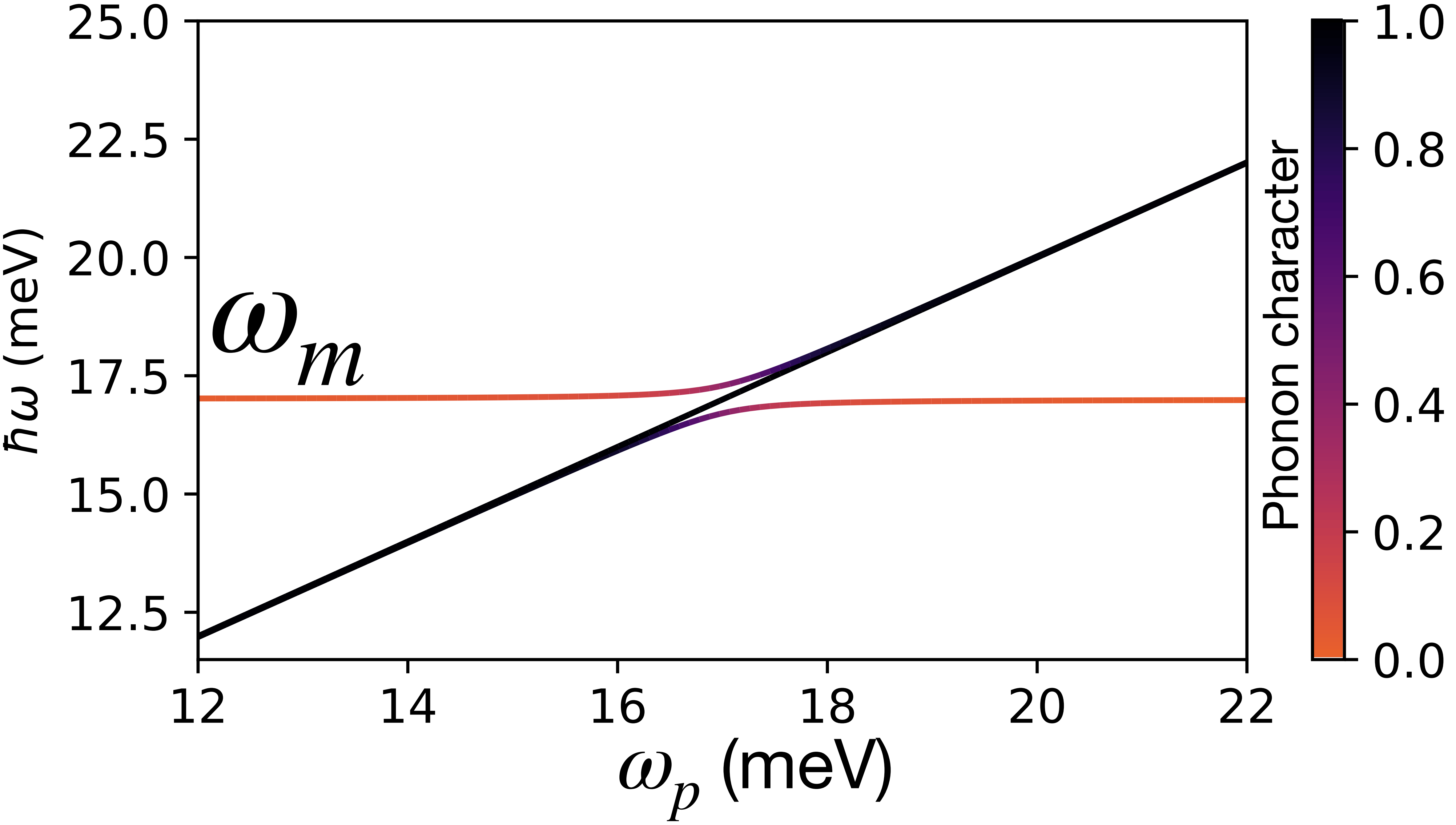


(meV/$\hbar$)



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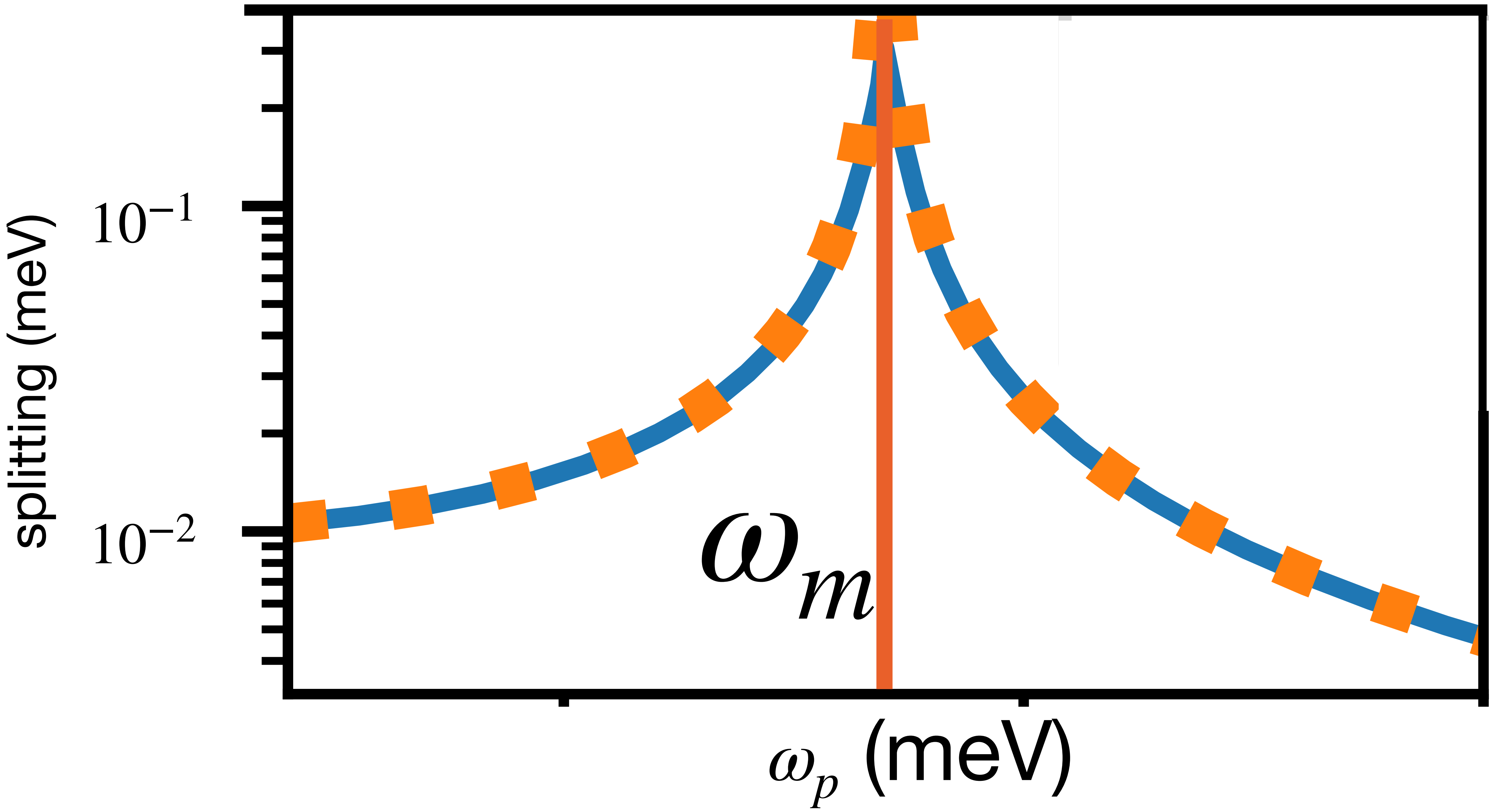

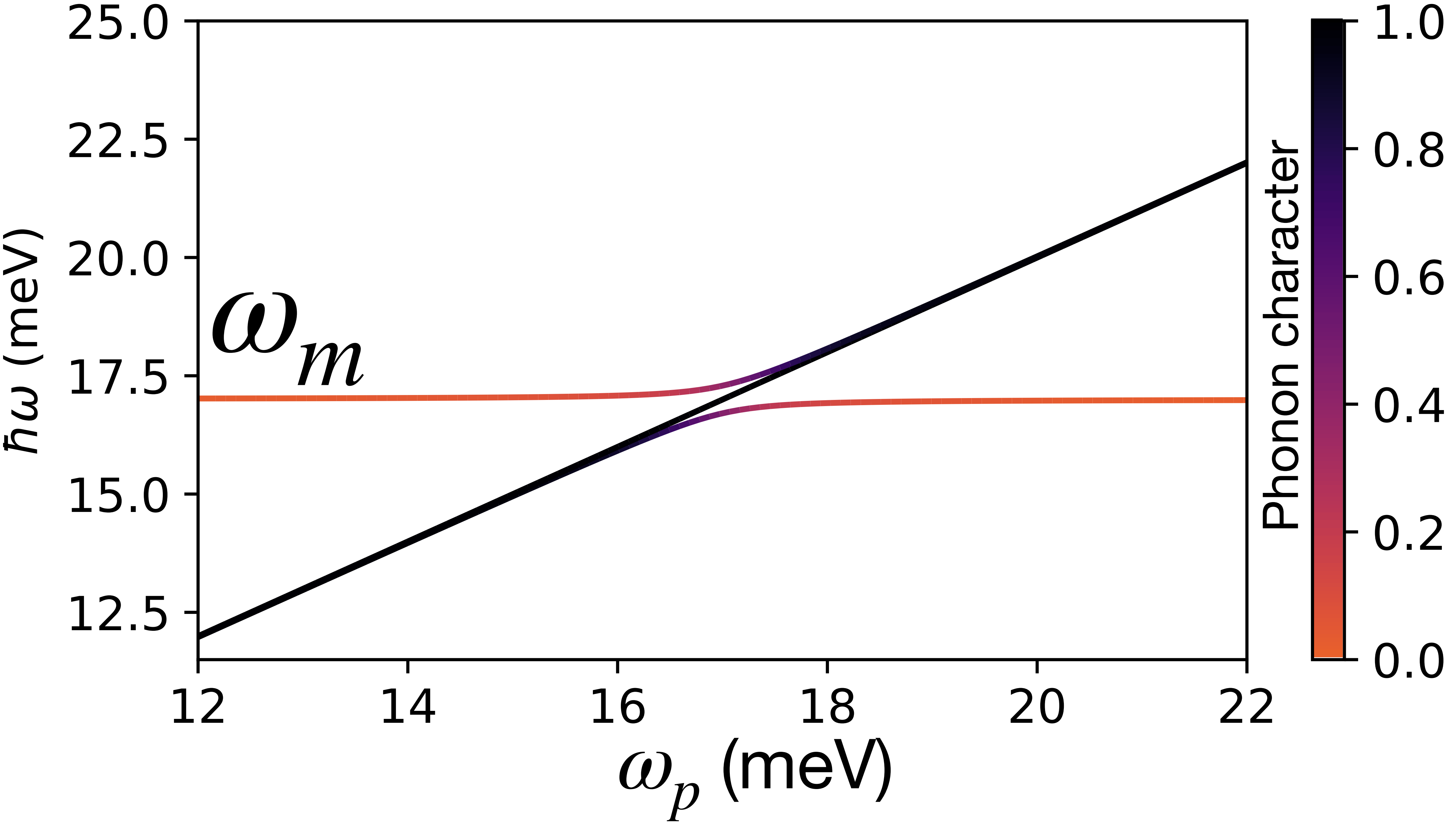

(meV/$\hbar$) Irrep	ω_p	Splitting: Adiabatic	Splitting: H_{sp}
E_g	6.9999	0.3820	0.0007
Couple to acoustic	12.9287	0.5270	0.0003
$\omega_m = 0.3$	13.4876	0.3368	0.0001
	29.8521	0.0244	3×10^{-6}
E_u	10.7667	0.0043	0.0046
Couple to optical	14.3259	0.0090	0.0311
$\omega_m = 17$	27.8168	0.0349	0.0118

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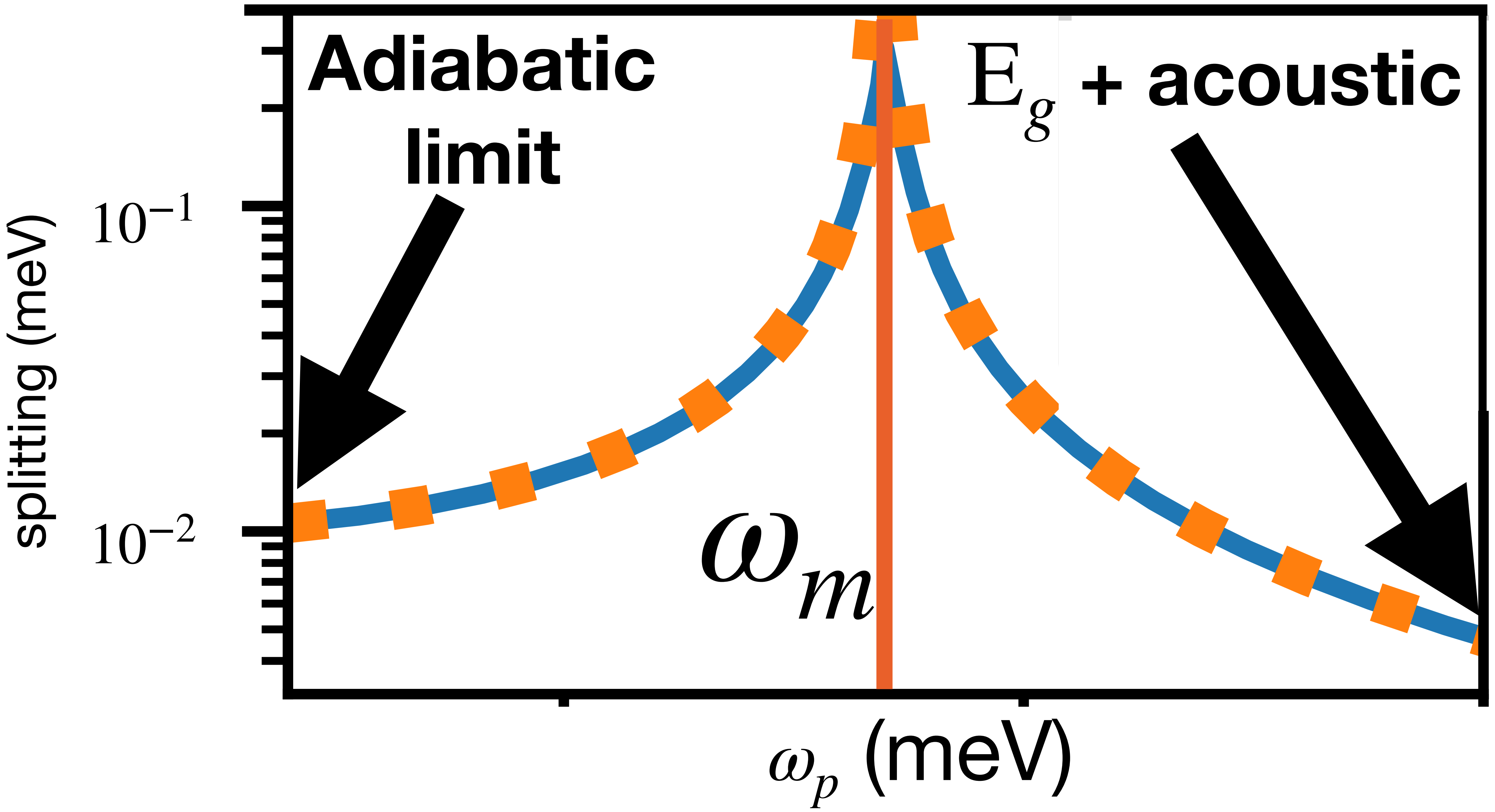

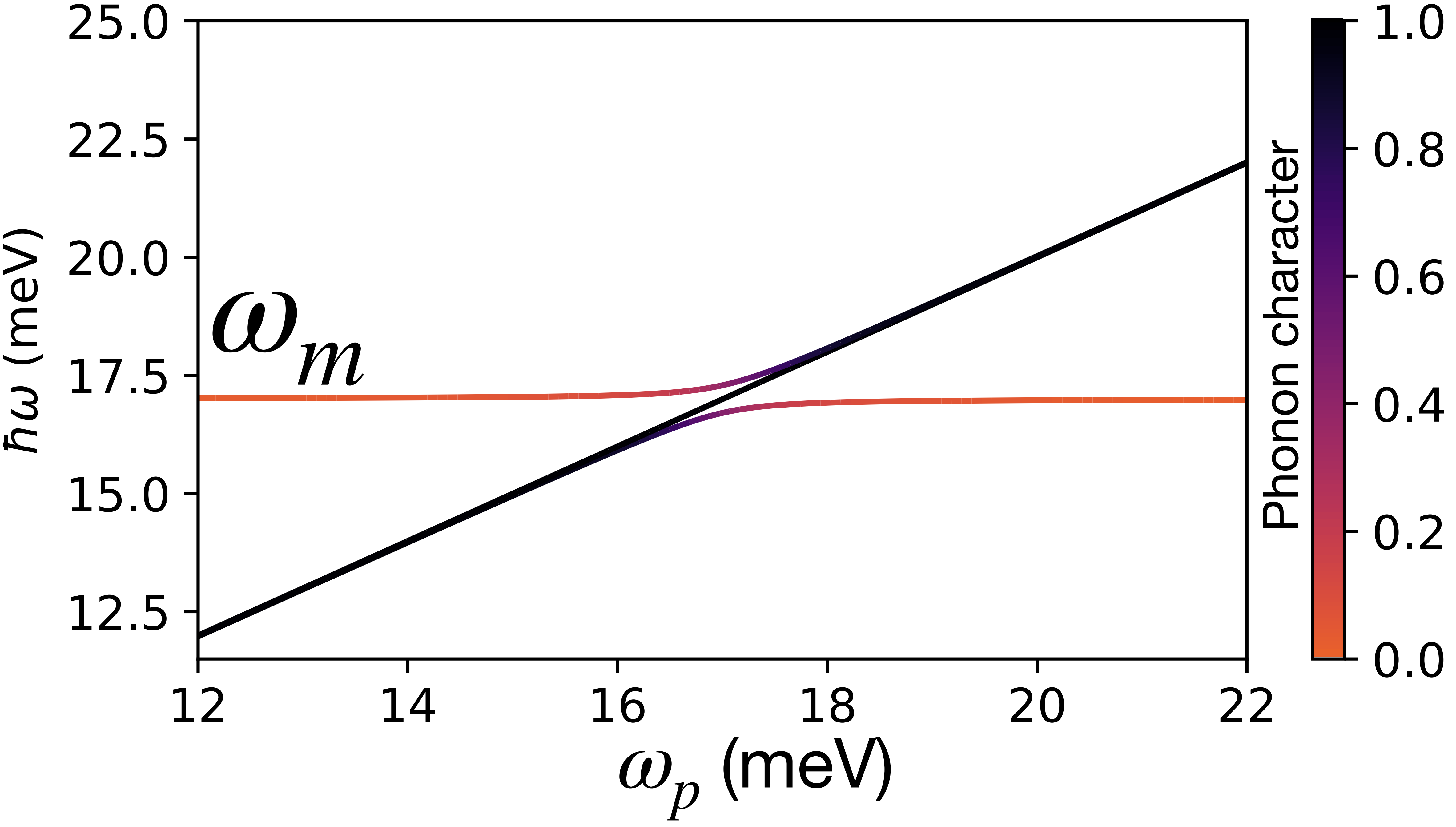

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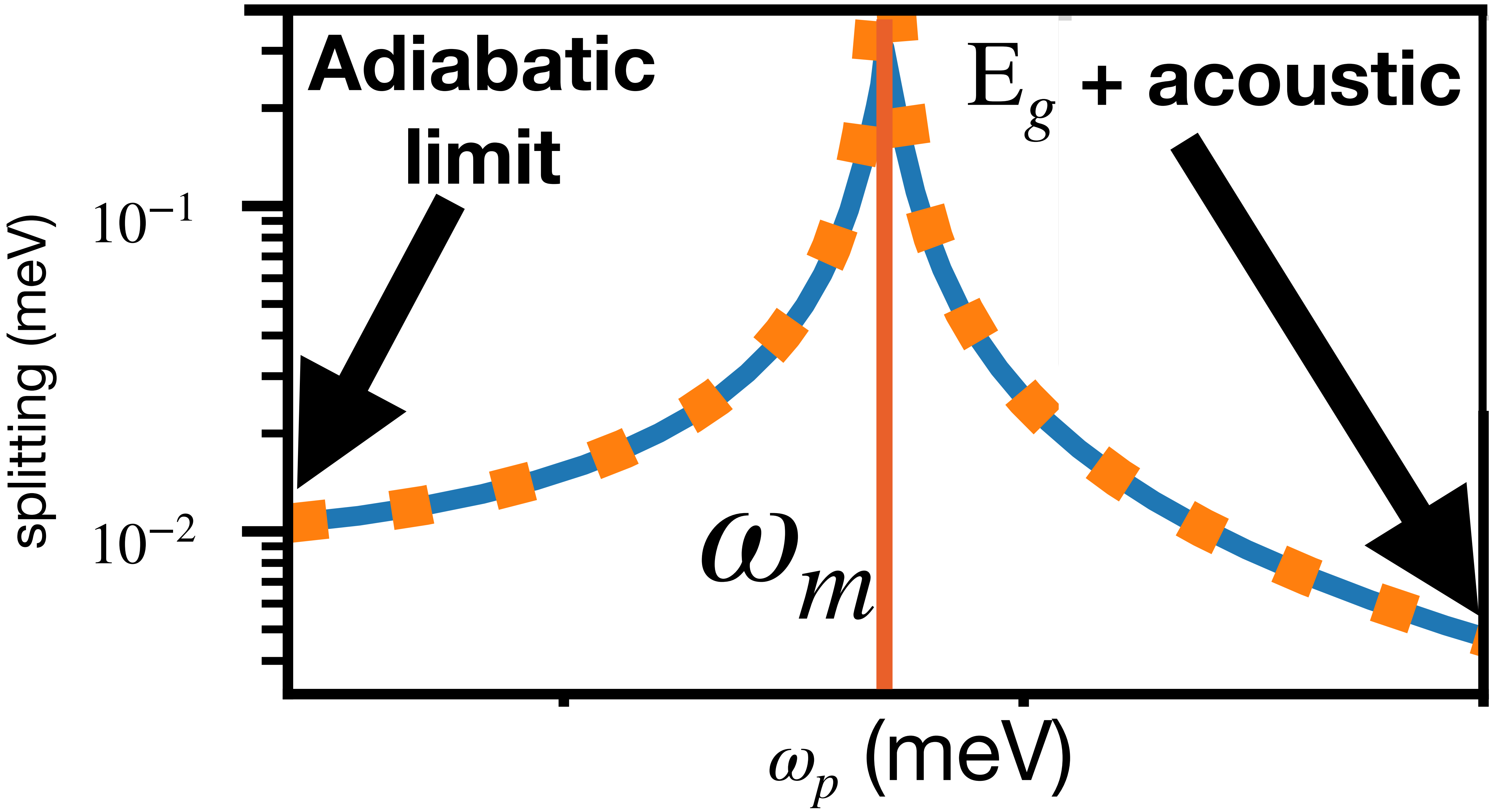

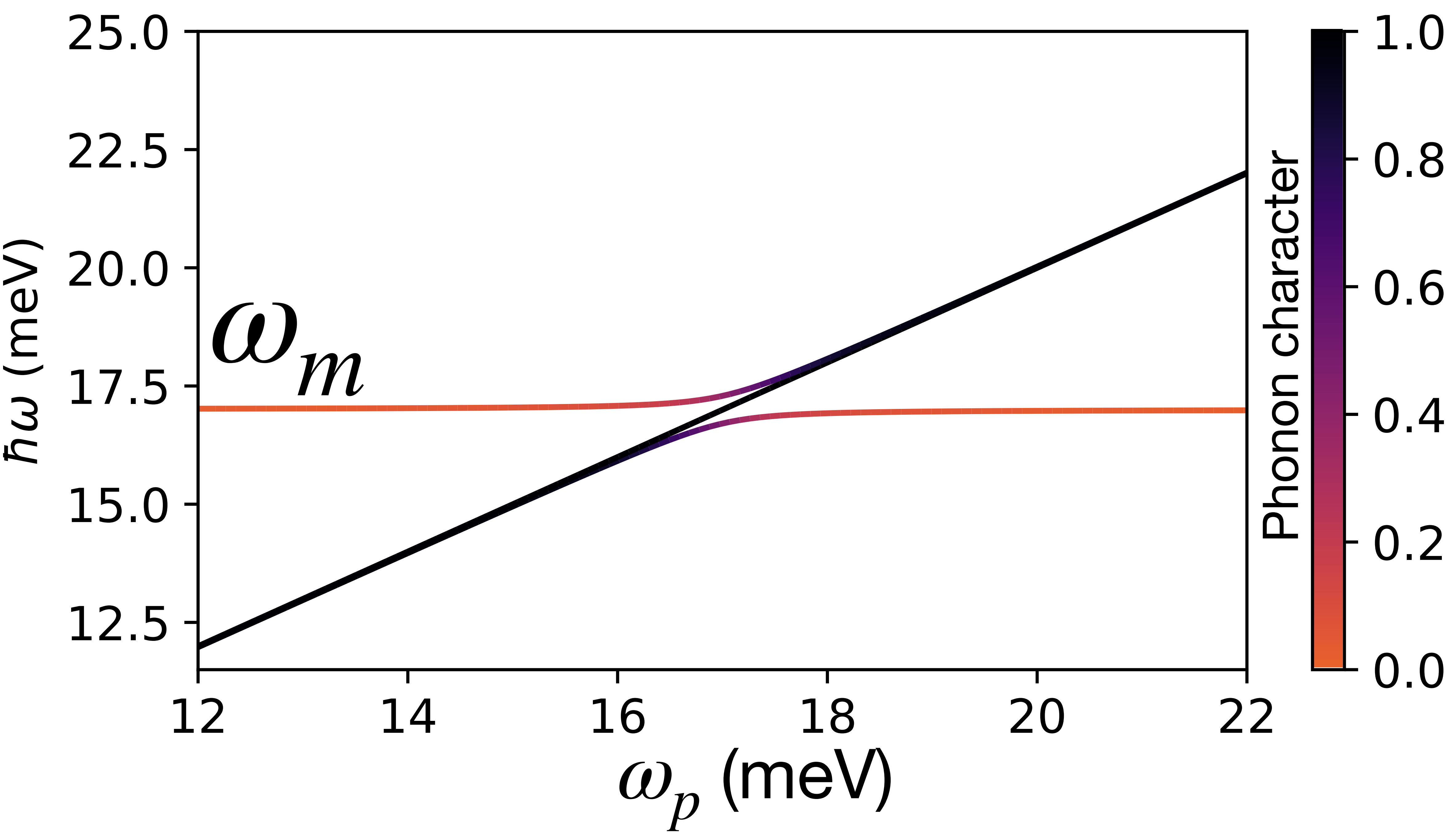

Physical Review Letters
130 (8), 086701 (2023)

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Generalized adiabatic response

$$[T_e + V(Q)]\,|\,\psi_e(Q)\rangle = \epsilon(Q)\,|\,\psi_e(Q)\rangle$$

Slow degrees of freedom: nuclei positions, spin canting

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Lagrangian:

$$\mathscr{L} = \frac{1}{2}\sum_i M_i \ddot{Q}_i^{\,2} - \epsilon(Q) + \hbar \sum_i \dot{Q}_i \langle \psi_e(Q)\,|\,i\frac{\partial}{\partial Q_i}\,|\,\psi_e(Q)\rangle$$

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Harmonic semi-classical:

$$\mathbf{M}\omega_n^2\,|\,n\rangle = (\mathbf{K} + i\omega_n \mathbf{G})\,|\,n\rangle$$

Spin + Phonon Hessian

Spin + Phonon Berry Curvature

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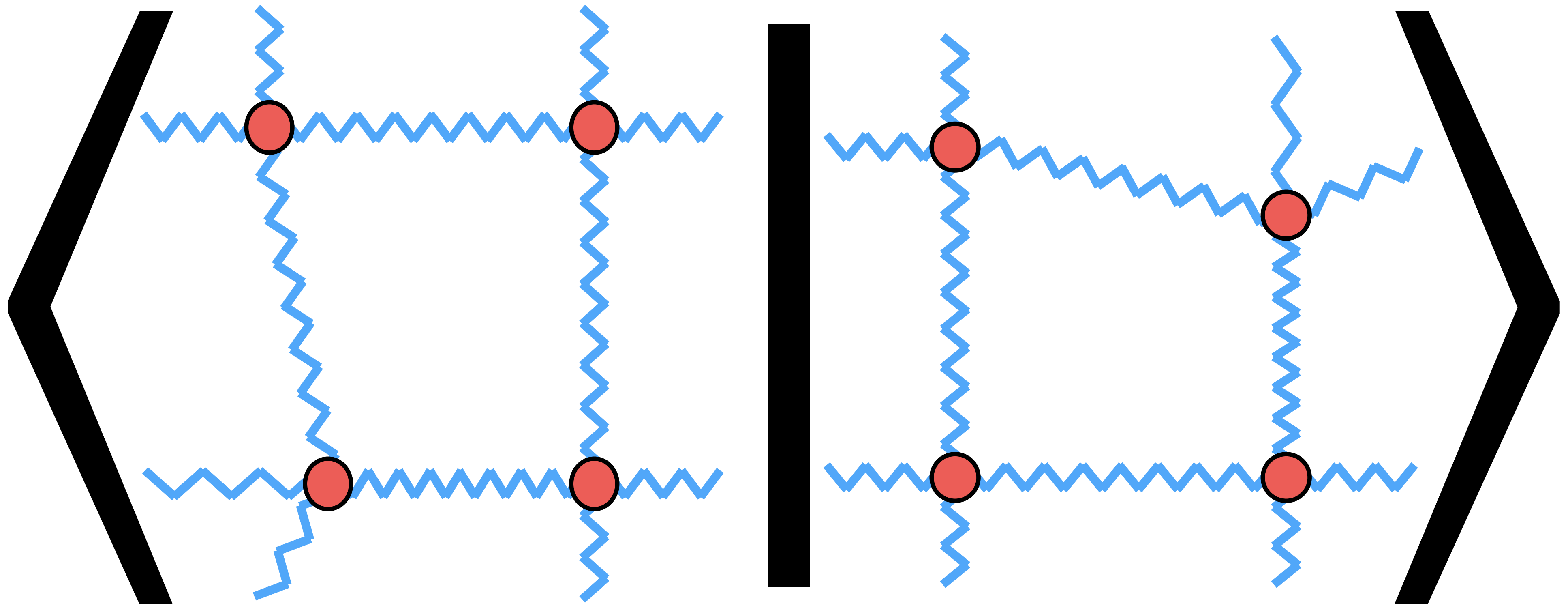
Paper in preparation
including “anti-chiral” phonons in antiferromagnets
See Shang Ren’s poster!

Outlook

Density functional perturbation theory (DFPT) implementation

Currently using finite differences



Beyond Γ point

Resonance with acoustic modes

Compute Thermal Hall conductivity, other observables

DFPT would be useful

Local spins

Currently constraining magnetization in "sphere" around site

More systematic approaches to identify low energy local spin degrees of freedom

Connect model Hamiltonians to first principles for beyond semi-classical treatment

Summary

Broken time reversal (TR) symmetry in the electronic sector can break TR in the lattice dynamics

Requires terms beyond static forces

Nuclear Berry curvature approach yields results consistent with magnetic space group, but can fail as a quantitative method

Developed and implemented general adiabatic formalism for coupled magnons+phonons

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Thank you collaborators!


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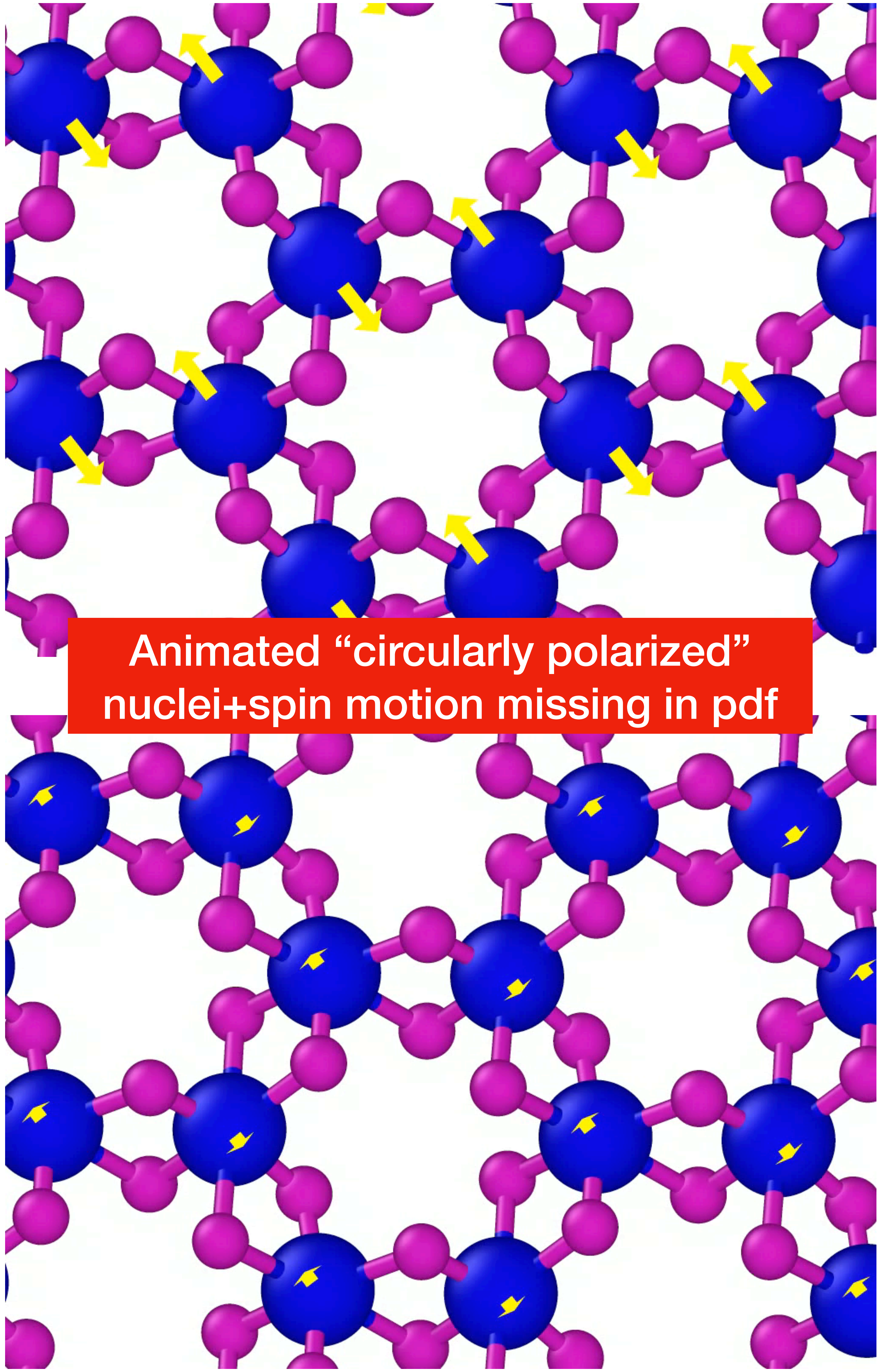

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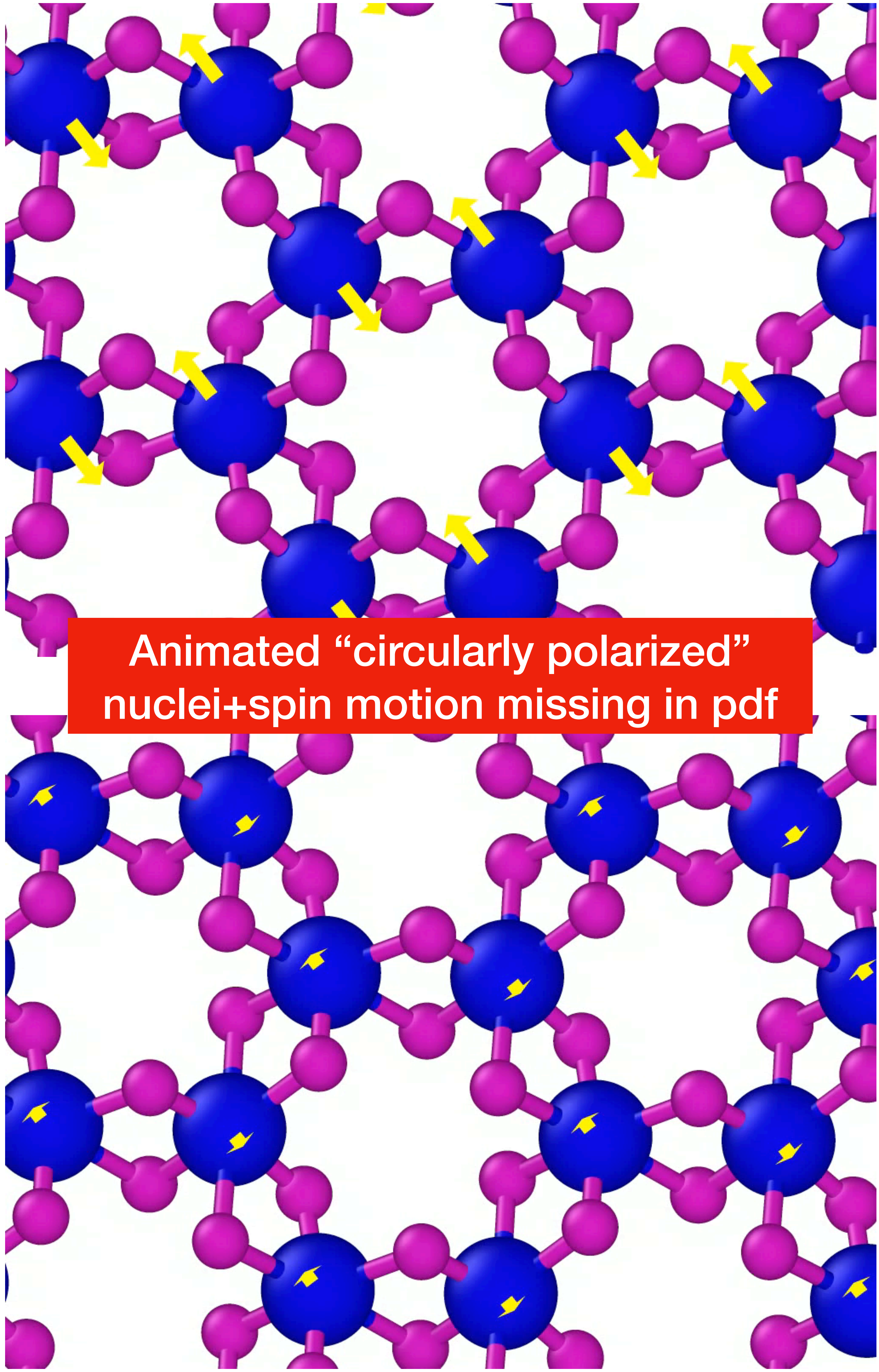

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FLATIRON INSTITUTE
Center for Computational Quantum Physics

Metric term

$$\Lambda(R) = \frac{\hbar^2}{2M_I} \langle \partial_{I\alpha} \psi(R) | Q | \partial_{I\alpha} \psi(R) \rangle \,,$$

$$Q = 1 - |\psi(R)\rangle\langle\psi(R)| \,,$$

$$G_{ij} = 2\hbar \mathrm{Im} \langle \frac{\partial \psi_e}{\partial R_i} \mid \frac{\partial \psi_e}{\partial R_j} \rangle$$

Doesn't break time reversal

Factor of nuclei mass M_I means this is typically much smaller than $\epsilon(R)$

2nd order derivatives (for phonons) involve high order derivatives of $|\psi_e\rangle$