Dynamical response of noncollinear spin systems from DFPT at constrained magnetic induction

Miquel Royo and Massimiliano Stengel

Institut de Ciència de Materials de Barcelona (ICMAB-CSIC)

Adiabatic Density-Functional Perturbation Theory

In a *strict* adiabatic (Born-Oppenheimer) approximation, electrons adapt instantaneously to any change due to a perturbation (atomic displacements).

Valid in trivial insulators and semiconductors

0.1 to few eV

Electronic excitations
(fast variables)

few to tens THz

Phonon excitations
(slow variables)

Adiabatic app. in practice

Static perturbation

Form factor

$$\Delta V(\mathbf{r}) = \lambda_i^{\mathbf{q}} \varphi_i^{q}(\mathbf{r}) e^{-i\mathbf{q}\cdot\mathbf{r}} + \mathrm{c.c}$$

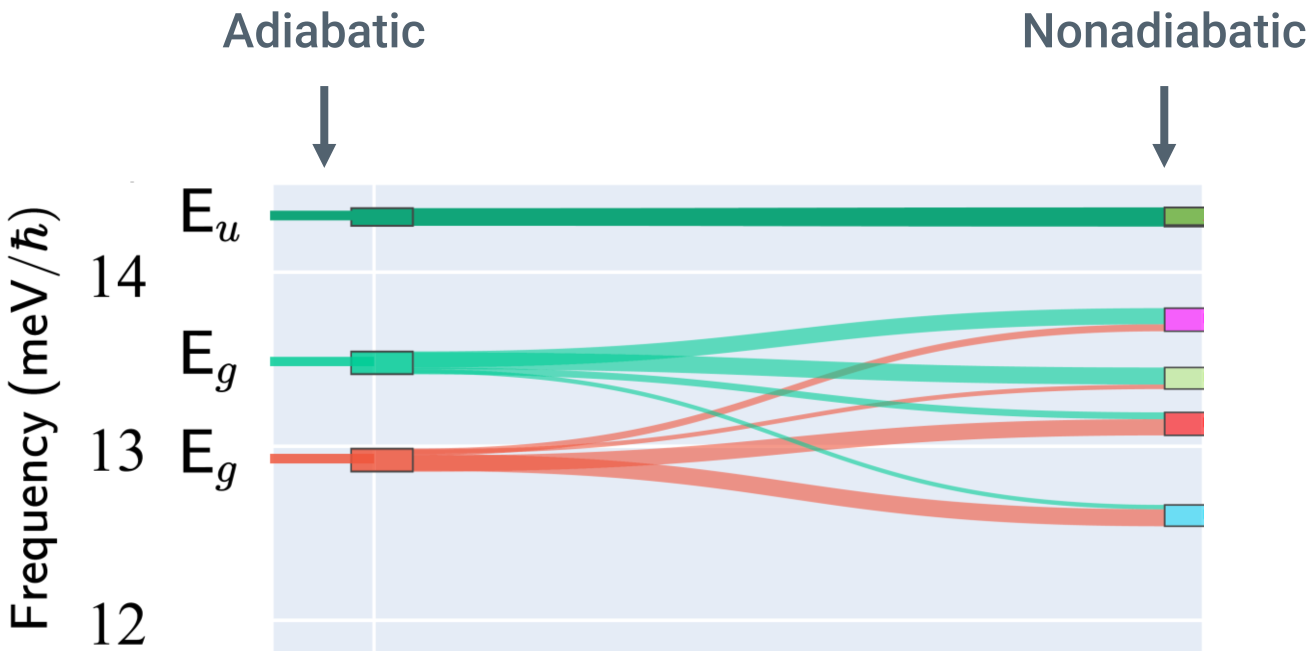
Amplitude

Slow dynamics from classical (harmonic) EOM

$$m_\kappa \ddot{u}^0_{\kappa\alpha}(t) = -\Phi^l_{\kappa\alpha\kappa'\beta} u^l_{\kappa'\beta}(t)$$

Unsuitability of the Adiabatic Approximation in Magnetic Insulators

Magnetism is not transferred to the response



CrI3 phonon frequencies at Γ

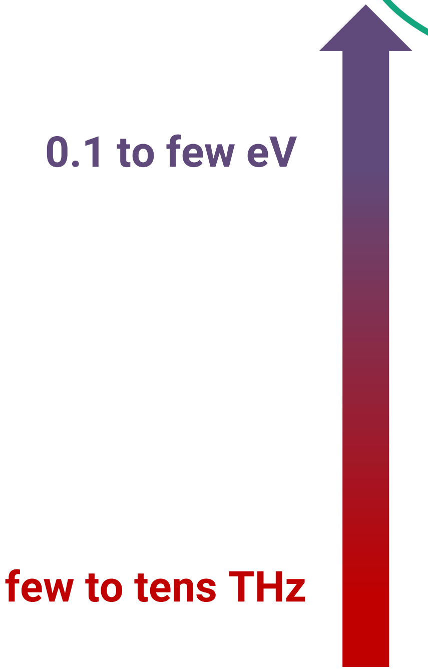
(Adapted from Bonini *et al.* PRL 2023)

Spin-wave excitations (magnons)

Low-energy excitations with electronic origin

fast or slow variables?

Niu & Kleinman. PRL 1998
Ren *et al.* PRX 2024



Time and Space Periodic Perturbations

Dynamic Perturbation:

$$\Delta V(\mathbf{r},t) = \underbrace{\lambda_i^{\mathbf{q},\omega}}_{\text{Amplitude}} \overbrace{\varphi_i^{q}}^{\text{Form factor}}(\mathbf{r})e^{-i(\mathbf{q}\cdot\mathbf{r}+\omega t)+\eta t} + \text{c.c.}$$

Generalized Susceptibility $\chi_{\lambda_1\lambda_2}$: (Equivalent to $E^{\lambda_1\lambda_2}$)

$$\chi_{\lambda_1,\lambda_2}(\mathbf{q},\omega) = \chi^{\text{Kubo}}_{\lambda_1,\lambda_2}(\mathbf{q},\omega) + \chi^{\text{geom}}_{\lambda_1,\lambda_2}(\mathbf{q}) \longrightarrow \text{Depends on } \lambda_1\lambda_2$$

$$\chi^{\text{Kubo}}_{\lambda_1,\lambda_2}(\mathbf{q},\omega) = \int [d^3k] \sum_{nm} \frac{f_{n\mathbf{k}} - f_{m\mathbf{k}+\mathbf{q}}}{\epsilon_{n\mathbf{k}} - \epsilon_{m\mathbf{k}+\mathbf{q}} + \omega + i\eta} \langle u^{(0)}_{n\mathbf{k}} | \hat{H}^{\lambda_1\dagger}_{\mathbf{k},\mathbf{q}} | u^{(0)}_{m\mathbf{k}+\mathbf{q}} \rangle \langle u^{(0)}_{m\mathbf{k}+\mathbf{q}} | \underbrace{\hat{\mathcal{H}}^{\lambda_2}_{\mathbf{k},\mathbf{q}}(\omega)}_{\hat{H}^{\lambda_2}_{\mathbf{k},\mathbf{q}} + V^{\lambda_2}_{\mathbf{q}}[n^{\lambda_2}_{\mathbf{q},\omega}(\mathbf{r})]} | u^{(0)}_{n\mathbf{k}} \rangle$$

Variational Time-Dependent DFPT For Insulators

Variational second-order energy functional: (like Gonze's $E^{\lambda_1\lambda_2}$)

$$\chi_{\lambda_1,\lambda_2}(\mathbf{q},\omega) = \chi^{\mathrm{SCF}}_{\lambda_1,\lambda_2}(\mathbf{q},\omega) + \underbrace{\tilde{\chi}_{\lambda_1,\lambda_2}(\mathbf{q},\omega)}_{\text{Resonant}} + \underbrace{\tilde{\chi}^{*}_{\lambda_1,\lambda_2}(-\mathbf{q},-\omega)}_{\text{Antiresonant}} + \chi^{\mathrm{geom}}_{\lambda_1,\lambda_2}(\mathbf{q})$$

$$\tilde{\chi}_{\lambda_1,\lambda_2}(\mathbf{q},\omega) = \int [d^3k] \sum_m \Big[\langle \bar{u}^{\lambda_1}_{m\mathbf{k},\mathbf{q}}(\omega)| \left(H_{\mathbf{k}+\mathbf{q}} + a\hat{P}_{\mathbf{k}+\mathbf{q}} - \epsilon_{n\mathbf{k}} - \omega - i\eta \right) |u^{\lambda_2}_{m\mathbf{k},\mathbf{q}}(\omega)\rangle$$
$$+ \langle \bar{u}^{\lambda_1}_{m\mathbf{k},\mathbf{q}}(\omega)|\hat{Q}_{\mathbf{k}+\mathbf{q}} H^{\lambda_2}_{\mathbf{k},\mathbf{q}}|u^{(0)}_{m\mathbf{k}}\rangle + \langle u^{(0)}_{m\mathbf{k}}|(H^{\lambda_1}_{\mathbf{k},\mathbf{q}})^{\dagger}\hat{Q}_{\mathbf{k}+\mathbf{q}}|u^{\lambda_2}_{m\mathbf{k},\mathbf{q}}(\omega)\rangle$$

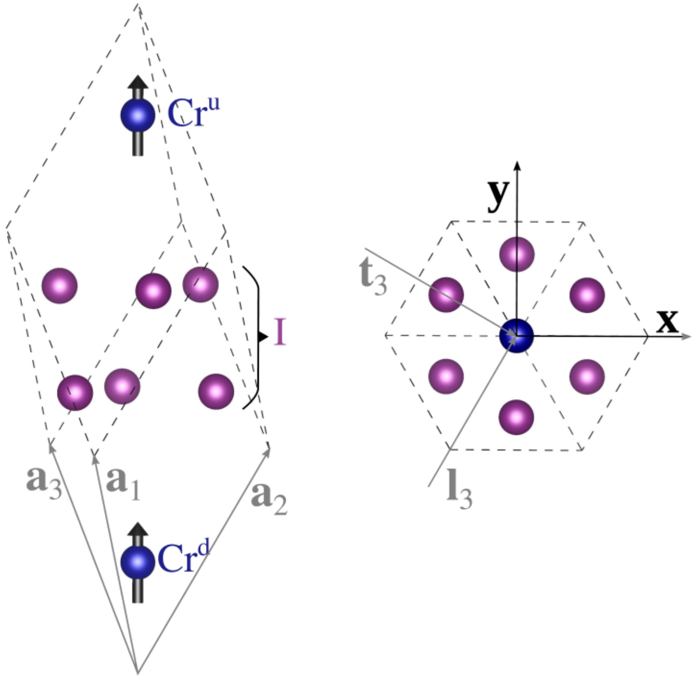
Coupled through SFC term: (due to broken TRS)

$$\chi^{\mathrm{SCF}}_{\lambda_1,\lambda_2}(\mathbf{q},\omega) = \int d^3r \int d^3r' [\bar{n}^{\lambda_1}_{\mathbf{q}\omega}]^{*}(\mathbf{r}) K_{\mathbf{q}}(\mathbf{r},\mathbf{r}') n^{\lambda_2}_{\mathbf{q}\omega}(\mathbf{r}') \qquad n^{\lambda_2}_{\mathbf{q},\omega}(\mathbf{r}) = \tilde{n}^{\lambda_2}_{\mathbf{q},\omega}(\mathbf{r}) + \left[\tilde{n}^{\lambda_2}_{-\mathbf{q},-\omega}(\mathbf{r})\right]^{*}$$

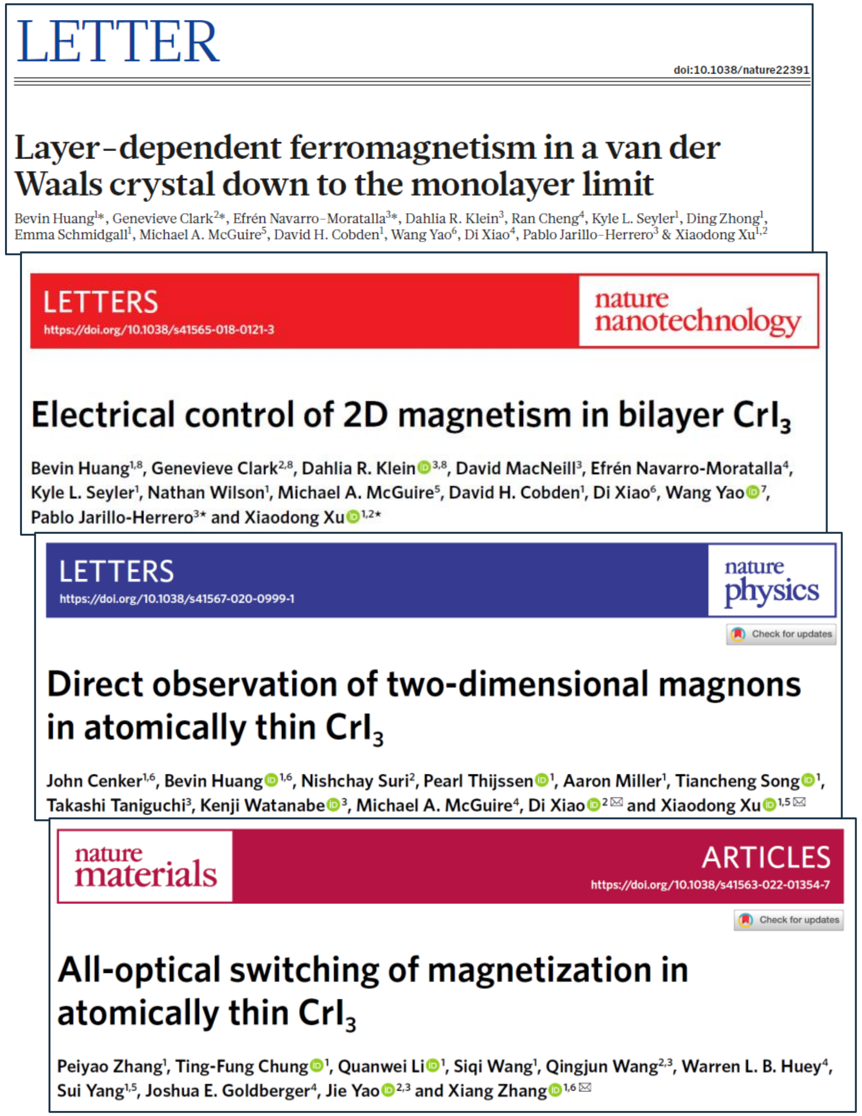
Dal Corso *et al.* PRB 1996 Calandra *et al.* PRB 2010 Lin *et al.* arXiv 2024

Target System: Ferromagnetic CrI_3

Hexagonal vdW insulating ferromagnet ($R\bar{3}$)



Complex interplay between electronic, lattice and magnetic degrees of freedom
(Delugas et al. PRB 2023, Ren *et al.* PRX 2024, etc…)



LETTER
doi:10.1038/nature22391

Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit

Bevin Huang[1*], Genevieve Clark[2*], Efrén Navarro-Moratalla[3*], Dahlia R. Klein[3], Ran Cheng[4], Kyle L. Seyler[1], Ding Zhong[1], Emma Schmidgall[1], Michael A. McGuire[5], David H. Cobden[1], Wang Yao[6], Di Xiao[4], Pablo Jarillo-Herrero[3] & Xiaodong Xu[1,2]

LETTERS
https://doi.org/10.1038/s41565-018-0121-3
nature nanotechnology

Electrical control of 2D magnetism in bilayer CrI_3

Bevin Huang[1,8], Genevieve Clark[2,8], Dahlia R. Klein[3,8], David MacNeill[3], Efrén Navarro-Moratalla[4], Kyle L. Seyler[1], Nathan Wilson[1], Michael A. McGuire[5], David H. Cobden[1], Di Xiao[6], Wang Yao[7], Pablo Jarillo-Herrero[3*] and Xiaodong Xu[1,2*]

LETTERS
https://doi.org/10.1038/s41567-020-0999-1
nature physics
Check for updates

Direct observation of two-dimensional magnons in atomically thin CrI_3

John Cenker[1,6], Bevin Huang[1,6], Nishchay Suri[2], Pearl Thijssen[1], Aaron Miller[1], Tiancheng Song[1], Takashi Taniguchi[3], Kenji Watanabe[3], Michael A. McGuire[4], Di Xiao[2] and Xiaodong Xu[1,5]

nature materials
ARTICLES
https://doi.org/10.1038/s41563-022-01354-7
Check for updates

All-optical switching of magnetization in atomically thin CrI_3

Peiyao Zhang[1], Ting-Fung Chung[1], Quanwei Li[1], Siqi Wang[1], Qingjun Wang[2,3], Warren L. B. Huey[4], Sui Yang[1,5], Joshua E. Goldberger[4], Jie Yao[2,3] and Xiang Zhang[1,6]

Computational Set Up

- Noncollinear DFT and DFPT with SOC [ixcrot=3]
  (Verstraete *et al.* PRB 2008, Ricci *et al.* PRB 2019)
- LDA
- Fully Relativistic NCPSPs (pseudodojo)
- No symmetries [nsym=1]
- ecut=40 Ha, 6x6x6 **k**-point mesh
- Zone-center (**q**=0) linear-response
- Transparent frequency and nondissipative regimes
  $\hbar\omega = 0 - 40\ meV\ ;\ \eta = 0$

Considered Perturbations:

		$\boldsymbol{\tau}$	$\mathbf{E}$	$\mathbf{H}$
Atom displacement →	$\boldsymbol{\tau}$	$\boldsymbol{\Phi}$	$\mathbf{Z}^*$	$\mathbf{M}^*$
Electric field →	$\mathbf{E}$		ϵ	α
Zeeman field →	$\mathbf{H}$			χ

χ=spin susceptibility

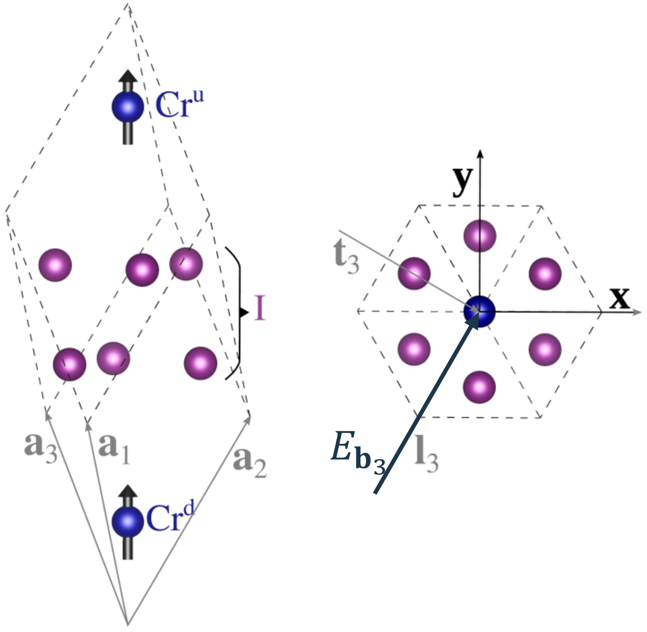
α=spin magnetoelectric

$\mathbf{M}^*$=magnetic charges

Linear response to electric field

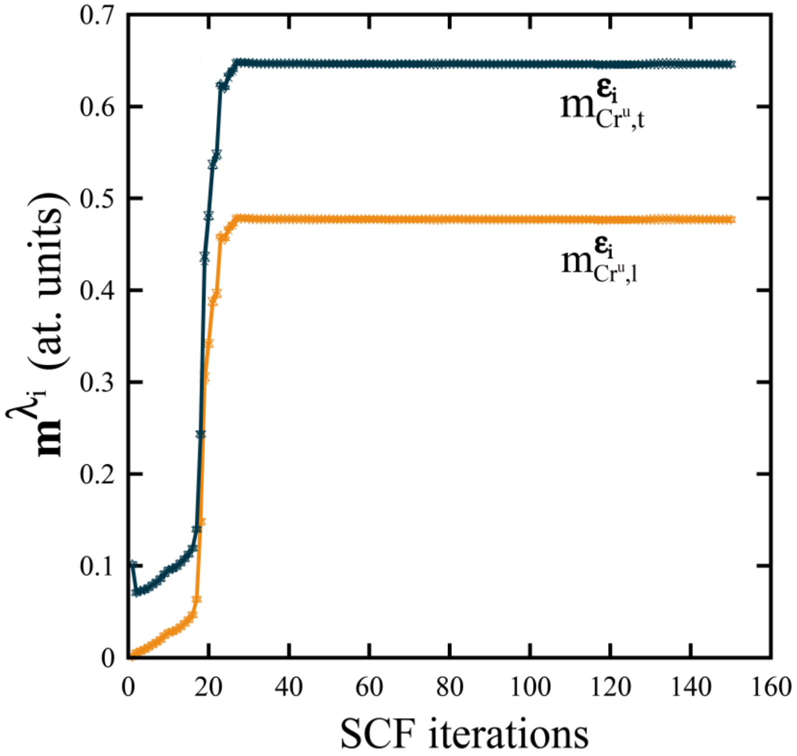
Self-consistency of induced local magnetic moments

$$\mathbf{m}_{\kappa}^{\lambda_i} = \int d^3r\, \mathbf{m}^{\lambda_i}(\mathbf{r}) f_{\kappa}(\mathbf{r} - \boldsymbol{\tau}_{\kappa})$$

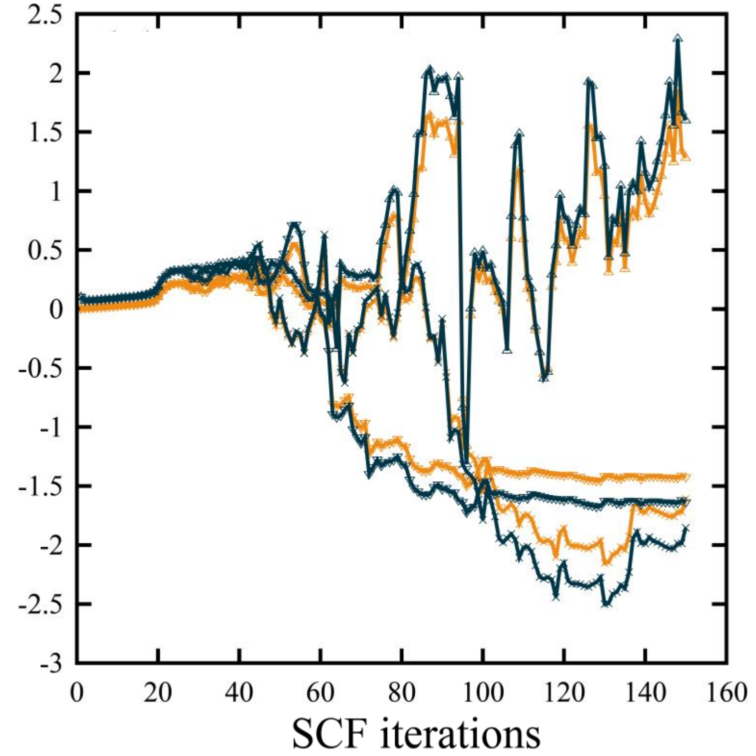


Electric field along $\mathbf{b}_1, \mathbf{b}_2, \mathbf{b}_3$

$\hbar\omega = 0\ meV$



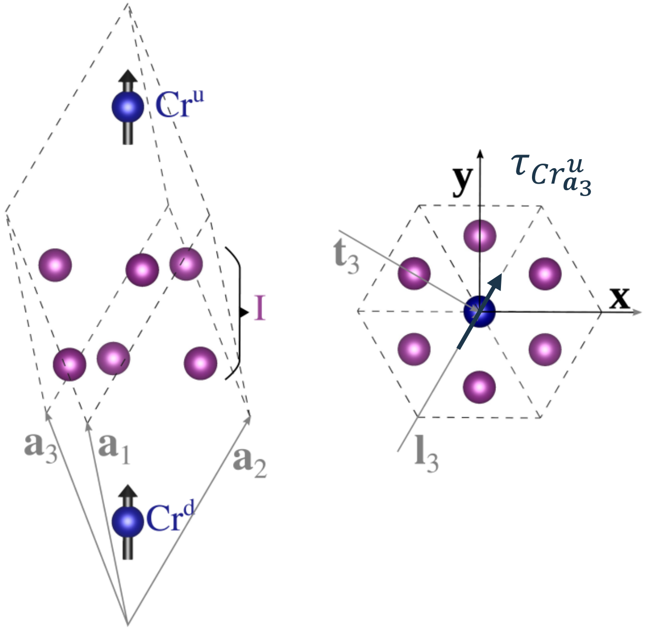
$\hbar\omega \approx 33\ meV$



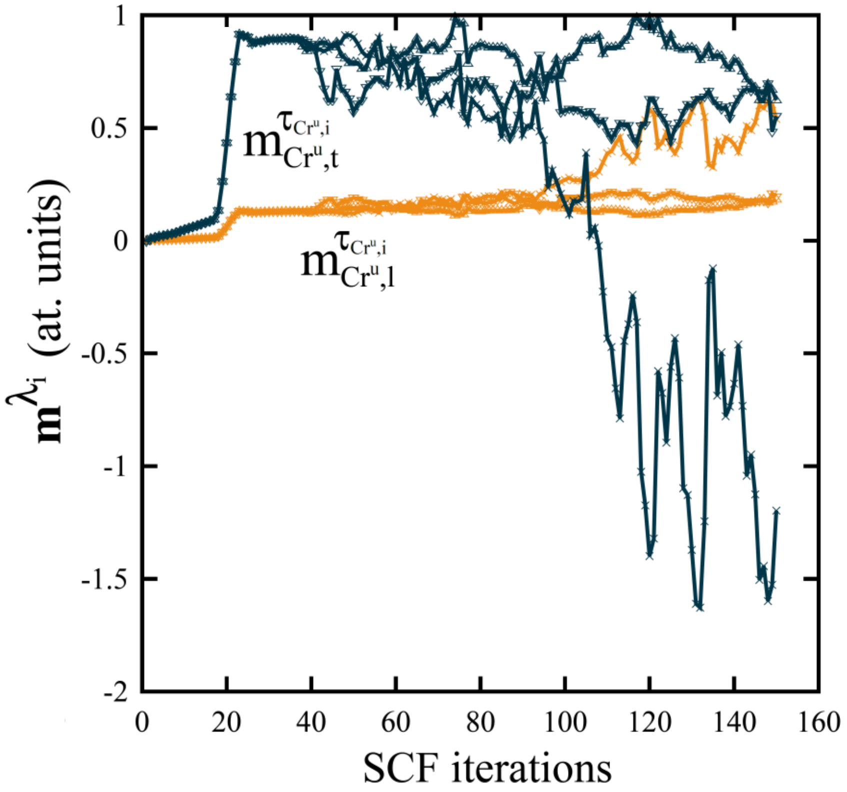
Linear response to atomic displacements and Zeeman fields

Self-consistency of induced local magnetic moments

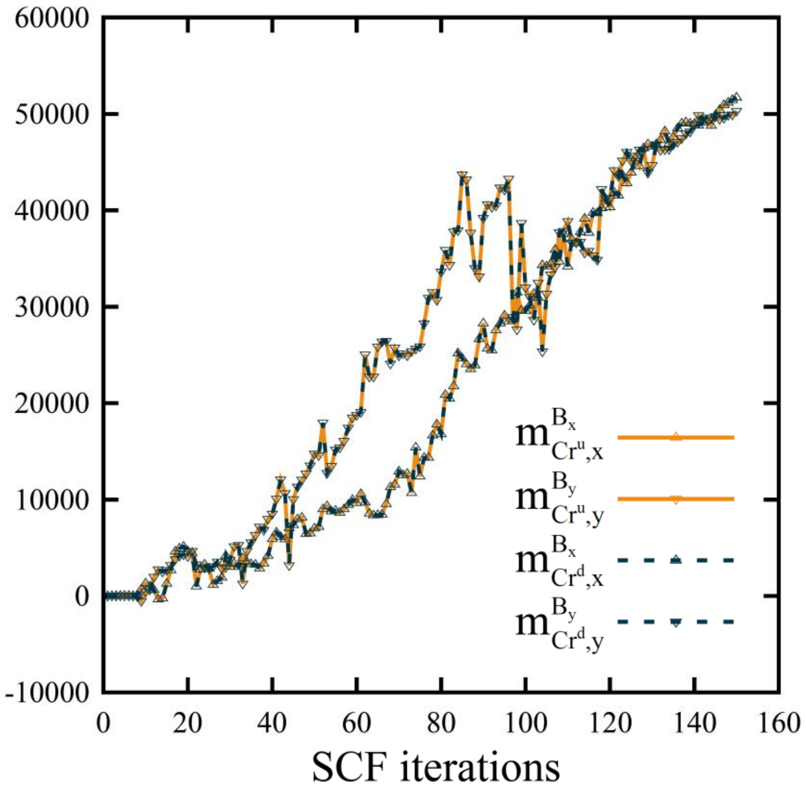
$$\mathbf{m}_\kappa^{\lambda_i} = \int d^3r\, \mathbf{m}^{\lambda_i}(\mathbf{r}) f_\kappa(\mathbf{r} - \boldsymbol{\tau}_\kappa)$$



Atomic displacement at $\hbar\omega = 0\ meV$



Zeeman field at $\hbar\omega = 0\ meV$

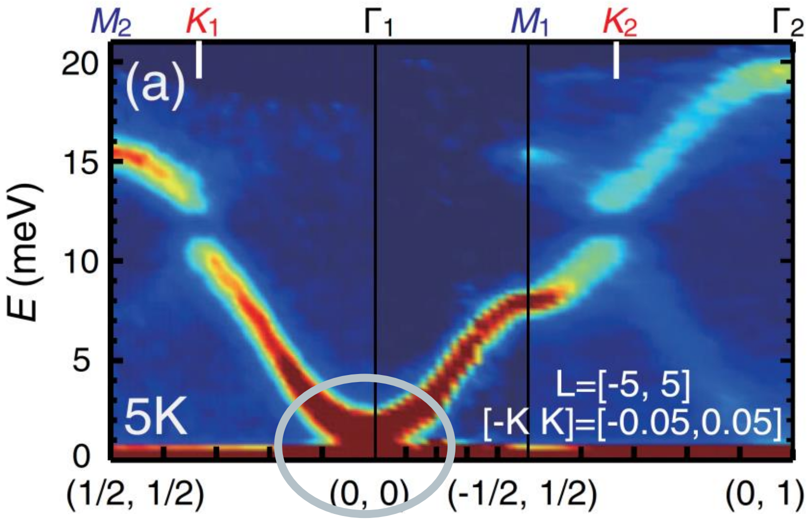


Cr^u displaced along $\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3$

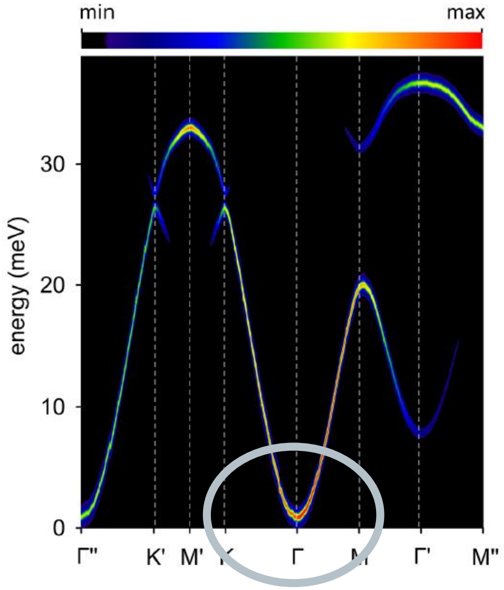
Zeeman field along $\boldsymbol{x}, \boldsymbol{y}$

Spin-Wave (Magnon) Excitation Spectra of CrI_3

Inelastic Neutron Scattering
(Chen et al. PRX 2021)



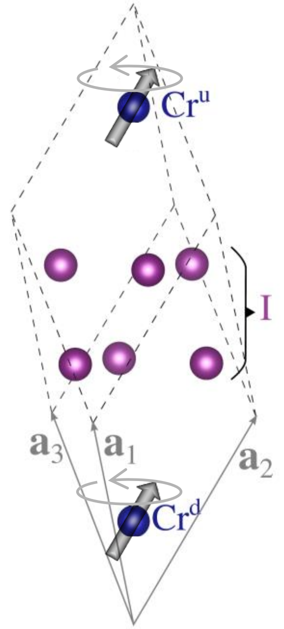
First principles
(Gorni et al. PRB 2022)



Acoustic magnon (q=0)

$E_{\mathrm{exp}} \approx 0.3\ meV$

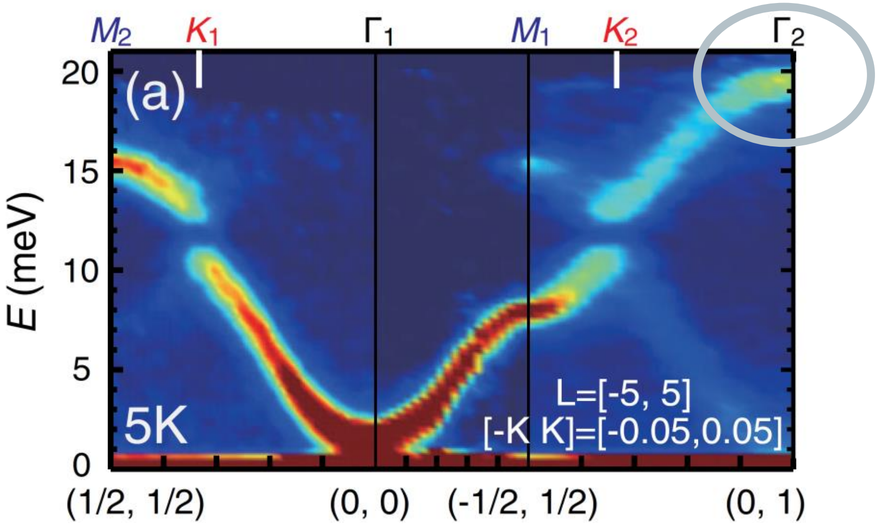
$E_{\mathrm{DFT}} \approx 1.3\ meV$



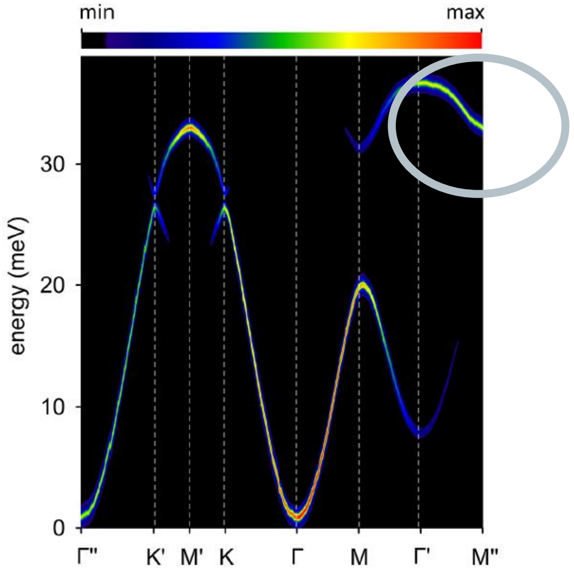
E_g symmetry
Couples with **H** and with Raman active phonon modes

Spin-Wave (Magnon) Excitation Spectra of CrI_3

Inelastic Neutron Scattering
(Chen et al. PRX 2021)



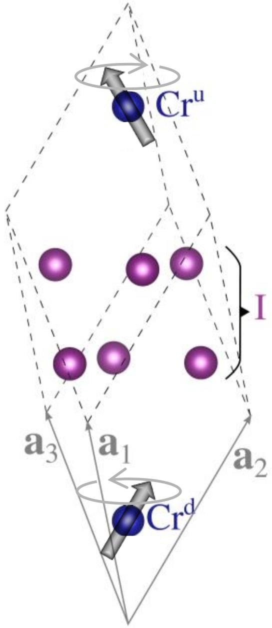
First principles
(Gorni et al. PRB 2022)



Optical magnon (q=0)

$E_{\mathrm{exp}} \approx 17\ meV$

$E_{\mathrm{DFT}} \approx 33\ meV$



E_u **symmetry**
Couples with **E** and with IR active phonon modes

Preconditioning the Linear Response of Noncollinear Spin Systems

Penalty term:

$$\bar{E}^{\lambda_1\lambda_2}(\mathbf{q},\omega) = E^{\lambda_1\lambda_2}(\mathbf{q},\omega) + \overbrace{\frac{\alpha}{2}\sum_l [m_l^{\lambda_1}(\mathbf{q},\omega)]^* m_l^{\lambda_2}(\mathbf{q},\omega)}^{\text{stiffens local spin cantings}}$$

$\alpha \longrightarrow$ *Penalty amplitude*

$l \longrightarrow (\kappa, \beta)$

$\kappa \longrightarrow$ *Magnetic sites*

$\beta \longrightarrow$ *Cartesian coord.*

- Shifts magnons to higher frequencies to facilitate an efficient convergence
- Real response-functions must not depend on the penalty and need to be recovered

Thermodynamic Ground-State Magnetic Functionals

Constrained-B:

Penalty amplitude

$$\bar{E}(B_l) = E_{\mathrm{KS}} + \frac{\alpha}{2}\sum_l (B_l - m_l)^2$$

Local magnetic inductions

Constrained-H:

$$\bar{F}(H_l) = E_{\mathrm{KS}} - \sum_l \frac{H_l^2}{2\alpha} - \sum_l H_l m_l$$

Local magnetic field strengths $H_l = \frac{\partial \bar{E}}{\partial B_l} = \alpha(B_l - m_l)$

Constrained-M:

Lagrange multipliers

$$\tilde{E}(M_l) = E_{\mathrm{KS}} + \sum_l \Lambda_l (m_l - M_l)$$

Target local magnetic moments

Constrained-H:

$$\tilde{F}(H_l) = E_{\mathrm{KS}} - \sum_l H_l m_l$$

GS functional under a (local) Zeeman field (Bousquet et al. PRL 2011)

Electric functionals case: Stengel, Spalding and Vanderbilt Nat. Phys 2008

From Constrained-B to Constrained-H Quantities

Local magnetic susceptibility

$$\chi_{lp} = -\frac{\partial^2 \tilde{F}}{\partial H_l \partial H_p}$$

l, p,... → (κ, β)

$$\boxed{\boldsymbol{\chi} = \bar{\boldsymbol{\chi}} \left(\mathbf{I} - \alpha \bar{\boldsymbol{\chi}}\right)^{-1}}$$

Penalized susceptibility

Penalty amplitude

Induced local magnetic moments and fields

$$\boxed{\left.\frac{\partial m_l}{\partial \lambda_a}\right|_{B_l} = -\bar{\chi}_{lp} \left.\frac{\partial H_p}{\partial \lambda_a}\right|_{M_p}}$$

$$\boxed{\left.\frac{\partial m_l}{\partial \lambda_a}\right|_{H_l} = -\chi_{lp} \left.\frac{\partial H_p}{\partial \lambda_a}\right|_{M_p}}$$

Second-order susceptibilities

$$\boxed{\tilde{E}_{ab} = \bar{E}_{ab} + \left.\frac{\partial H_l}{\partial \lambda_a}\right|_{M_l} \bar{\chi}_{lp} \left.\frac{\partial H_p}{\partial \lambda_b}\right|_{M_p}}$$

Frozen magnetic

$$\boxed{\tilde{F}_{ab} = \tilde{E}_{ab} - \left.\frac{\partial H_l}{\partial \lambda_a}\right|_{M_l} \chi_{lp} \left.\frac{\partial H_p}{\partial \lambda_b}\right|_{M_p}}$$

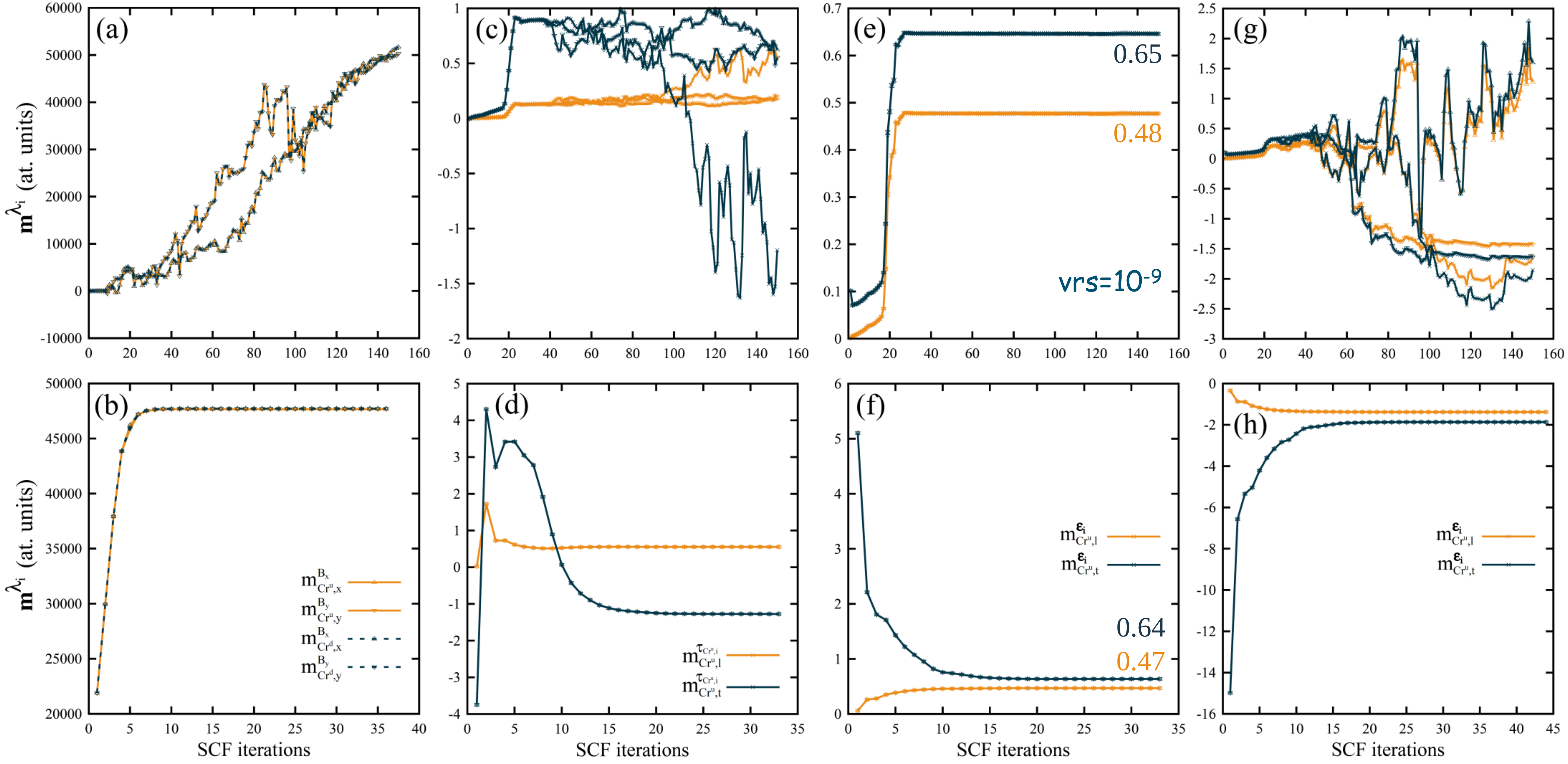
Spin relaxed

Convergence Improvement

Zeeman ($\hbar\omega = 0$) | Atomic disp. ($\hbar\omega = 0$) | E field ($\hbar\omega = 0$) | E field ($\hbar\omega \approx 33\ meV$)

DFPT

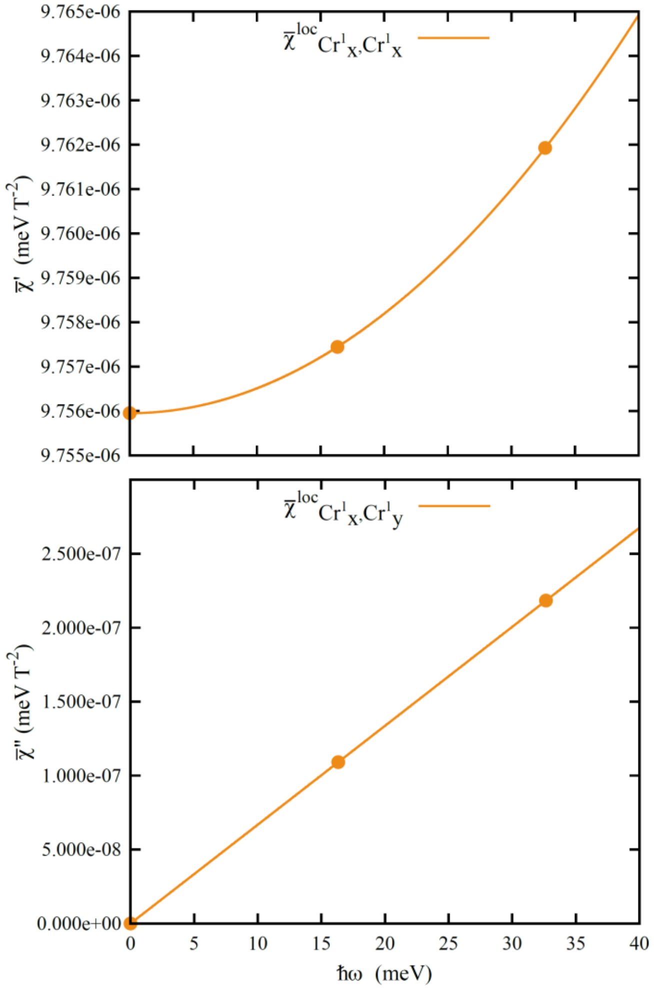
Constrained-B DFPT (tolvrs=10^{-14})



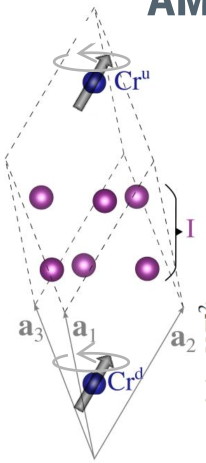
Frequency Interpolation of Penalized Quantities

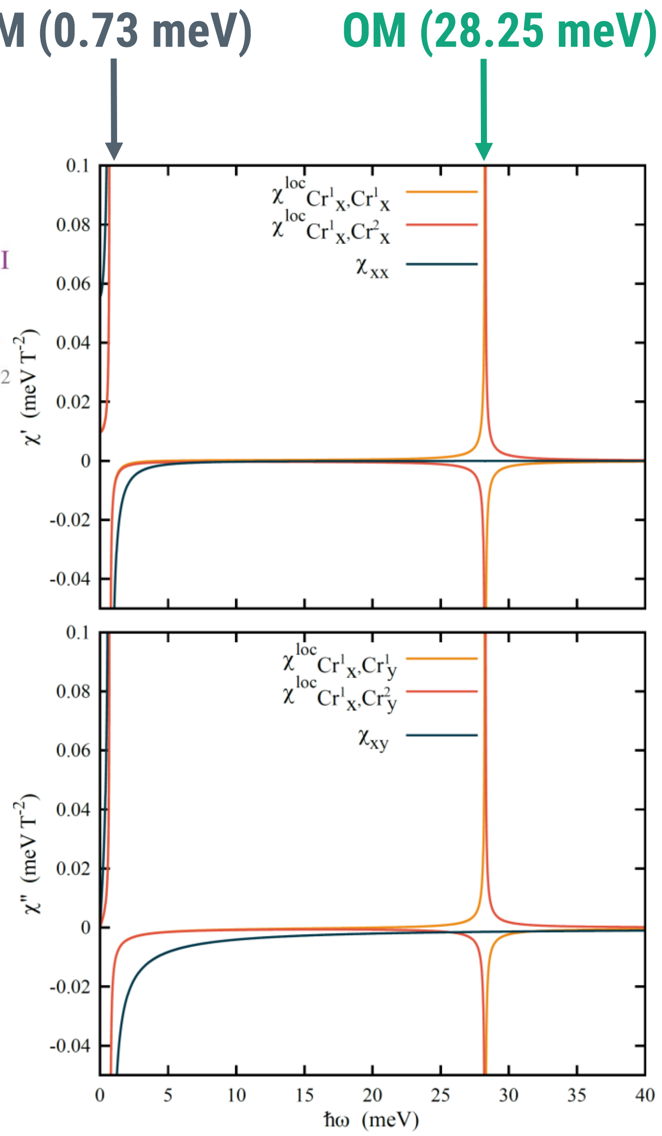
Local magnetic susceptibility

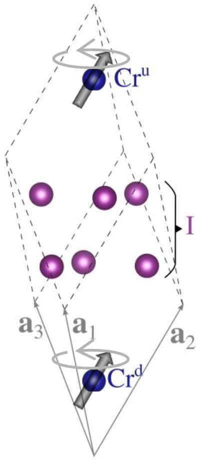
Constrained-B
(resonance free)



Constrained-H







Nonadiabatic Lattice Dynamics

A nonadiabatic phonon must be calculated at the phonon eigenfrequency

Phonon Green's function:

$$\mathbf{G}(\mathbf{q},\omega) = \left[(\omega + i\eta)^2\mathbf{I} - \underbrace{\mathbf{D}(\mathbf{q},\omega)}_{\text{Interpolated dynamical matrix}}\right]^{-1}$$

Spectral function:

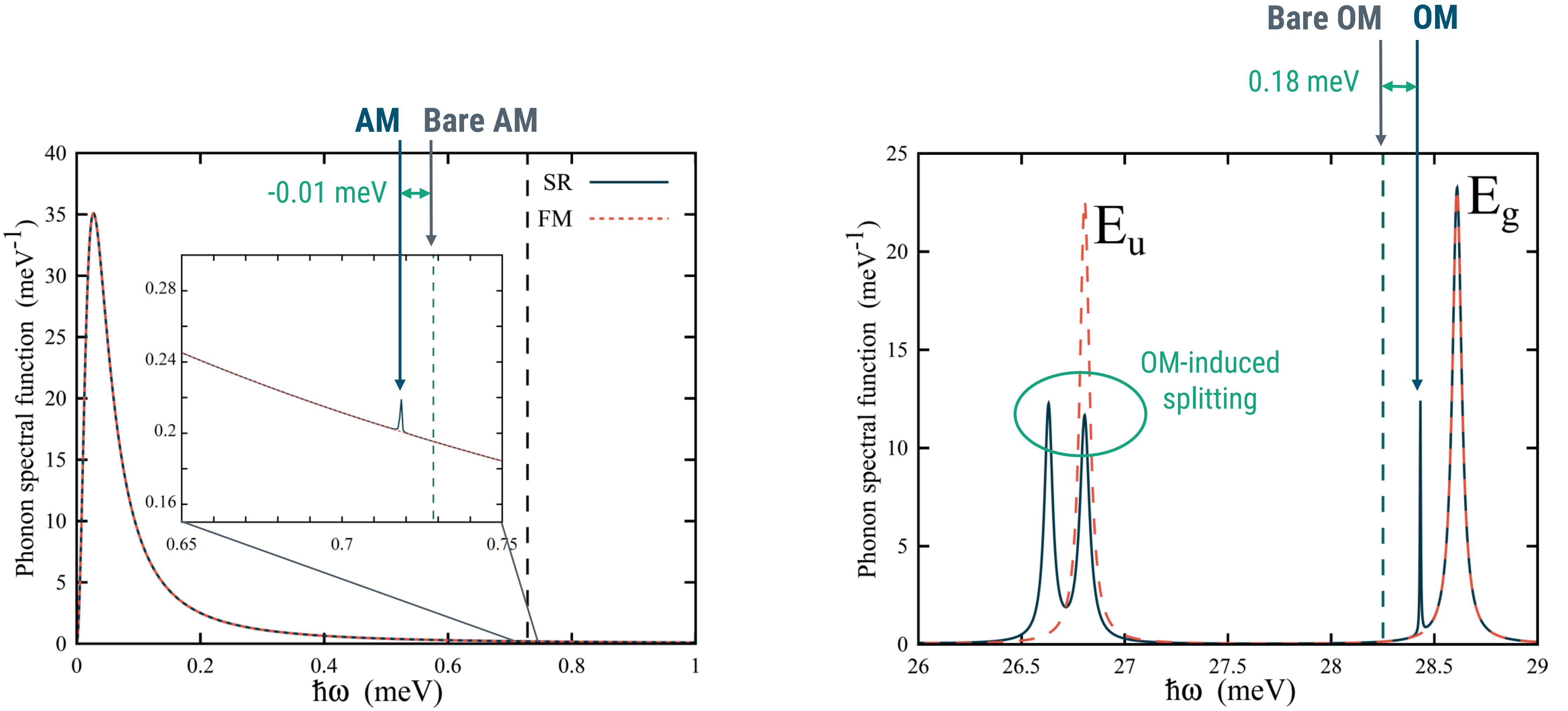
$$A(\mathbf{q},\omega) = -\frac{2\omega}{\pi}\mathrm{Tr}\,\mathrm{Im}\mathbf{G}(\mathbf{q},\omega)$$

(Berges *et al.* PRX 2023)

Lattice contributions to the response:

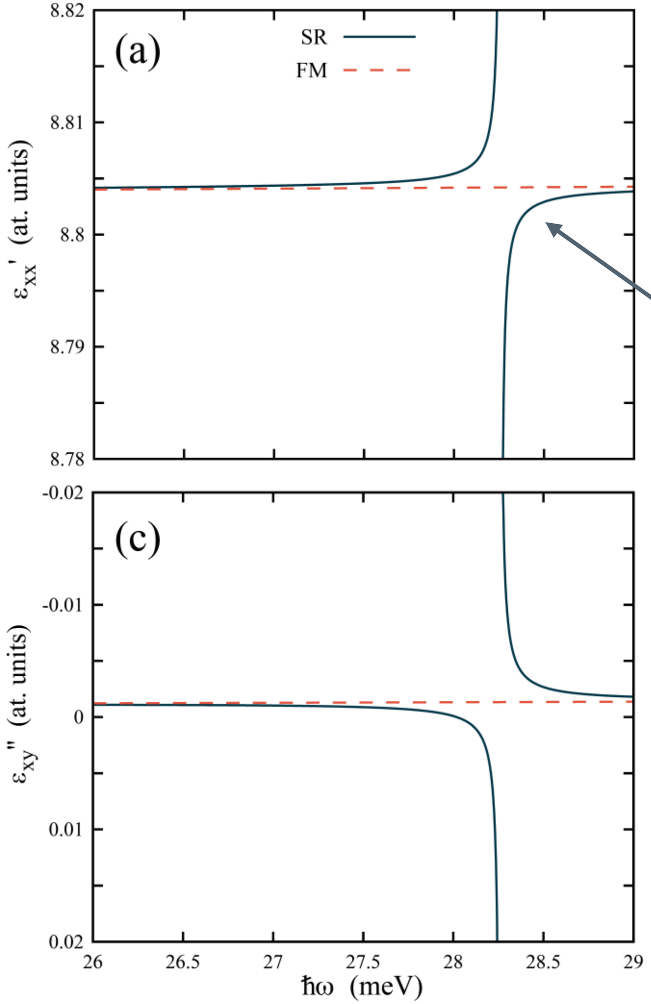
$$\underbrace{\chi^{\mathrm{RI}}_{\lambda_1,\lambda_2}}_{\text{Relaxed ion}}(\mathbf{q},\omega) = \underbrace{\chi^{\mathrm{CI}}_{\lambda_1,\lambda_2}}_{\text{Clamped ion}}(\mathbf{q},\omega) - \underbrace{\chi_{\lambda_1,\tau_{\kappa\alpha}}(\mathbf{q},\omega)M_\kappa^{-1/2}G_{\kappa\alpha,\kappa'\beta}(\mathbf{q},\omega)M_{\kappa'}^{-1/2}\chi_{\tau_{\kappa'\beta},\lambda_2}(\mathbf{q},\omega)}_{\text{Lattice mediated}}$$

Nonadiabatic Lattice Dynamics



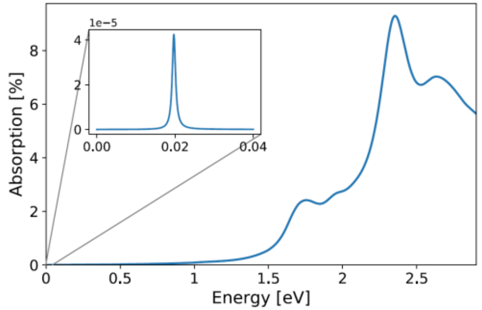
Dielectric Response of CrI_3

Clamped ion



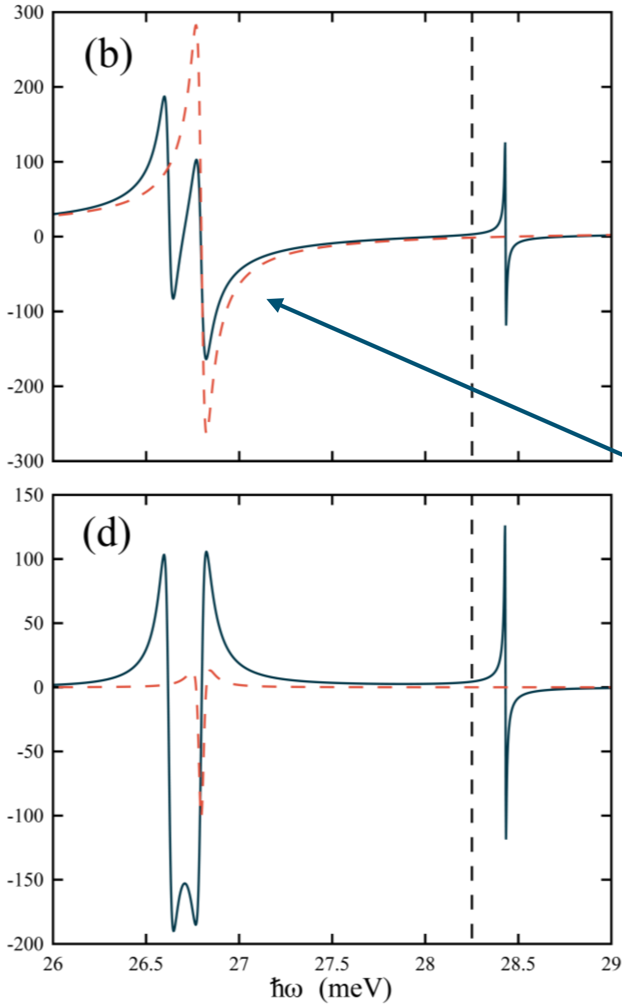
Weak OM resonance

TDDFT IR spectrum



(Olsen PRL 2021)

Relaxed ion

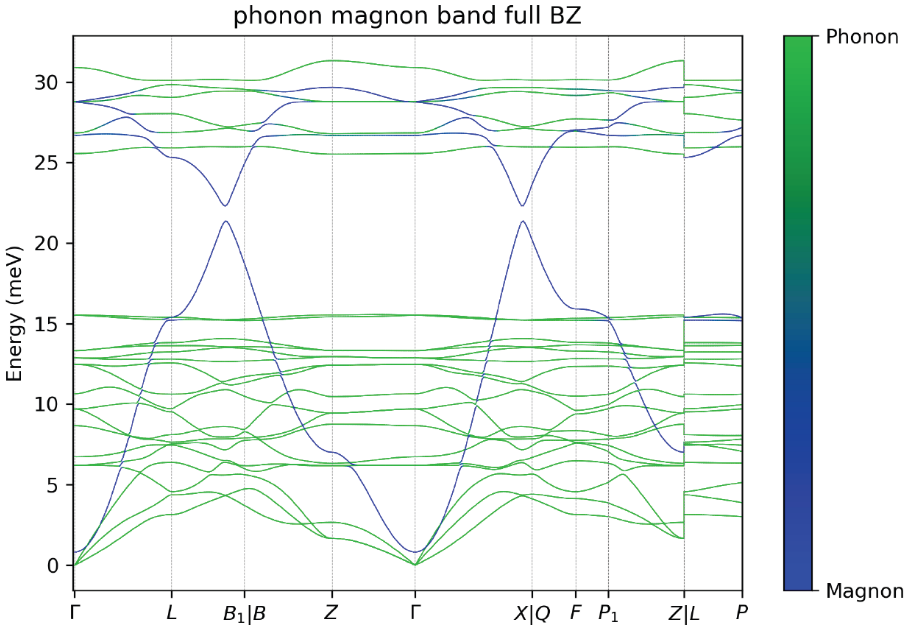


Shifted and amplified OM resonance

Additional magnon-phonon coupling signatures

Summary and outlook

- TD-DFPT (and DFPT) challenging in noncollinear magnets
- Constrained-**B** approach converges properly by stiffening magnons
- Simultaneous influence of magnons and phonons demonstrated
- Finite-**q** regime: magnon-phonon bands
- Nonlinear and spatial-dispersion properties



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ICMAB
INSTITUT DE CIÈNCIA DE MATERIALS DE BARCELONA
EXCELENCIA SEVERO OCHOA
CSIC
CONSEJO SUPERIOR DE INVESTIGACIONES CIENTÍFICAS

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