Institut Néel - CNRS
Institut Laue Langevin

Qu’est un électromagnon ?
(What is an electromagnon?)

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GdR MEETICC Electromagnons - Orsay, 20-21 novembre 2019

Ouline

- Electromagnon in the literature
- Electro-active magnon from an ab-initio point of view
  - Back to basics
  - Within the BO approximation
  - Breaking the BO approximation (entangled phonon-magnon state)
- Conclusion

In the literature

Definitions found in the literature

- mixed magnon-phonon mode
- electroactive magnon
- elementary excitations if multiferroic compounds

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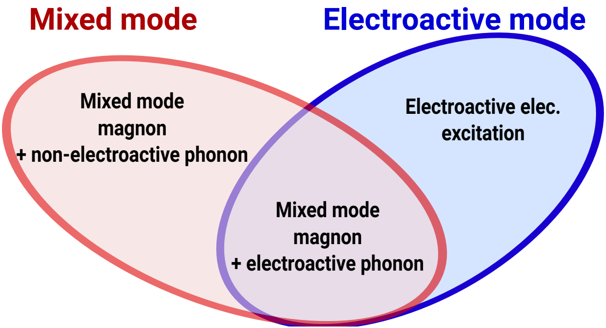
Two non equivalent definitions



In the literature

Definitions found in the literature

- mixed magnon-phonon mode
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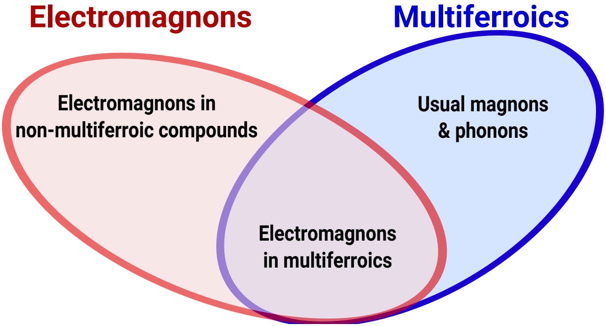
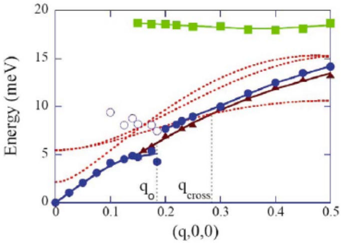
- elementary excitations if multiferroic compounds

Not specific to multiferroics



In the literature

Non-electroactive mixed phonon-magnon mode : $YMnO_3$



Phonons dispersion.

S. Petit *et al*, Pramana **71**, 869 (2008).

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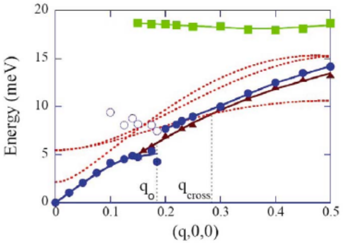

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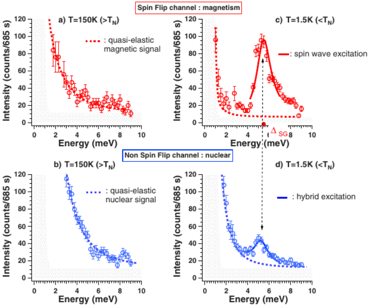

INS SF, NSP

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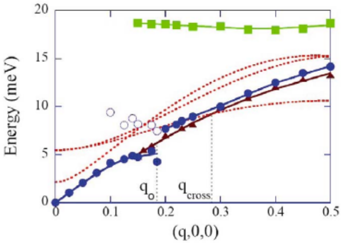

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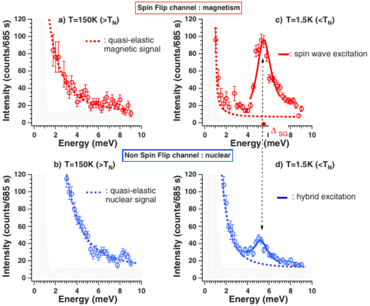

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No electromagnon observed in Raman & THz

C. Toulouse *et al*, PRB **89**, 094415 (2014)

In the literature

$YMnO_3$

- Mixed phonon-magnon mode
- Non electroactive

$\Longrightarrow$ $\exists$ symmetry forbidden phonon-magnon mixed modes

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$\Longrightarrow$ Magnons can be excited by light ($\vec{H}$)
Electromagnons ?

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$\Longrightarrow$ Magnons can be excited by light ($\vec{H}$) Electromagnons ?

$Ba_2Mg_2Fe_{12}O_{22}$

- Electromagnon in PE phase

N. Kida *et al*, PRB **80**, 220406R (2009)

$\Longrightarrow$ Not multiferroic elementary excitation

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    (entangled phonon-magnon state)
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From an ab-initio point of view : Generalities

Back to the basics

Mixed phonon-magnon mode $\Longrightarrow$ breaking the BO approximation

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The BO approximation

$$\hat{H} \quad = \quad \hat{T}^{e} + \hat{V}^{ee} + \hat{V}^{eN} + \hat{T}^{N} + \hat{V}^{NN}$$

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Decoupling of nuclear & electronic motions

- $$\hat{H} \longrightarrow \left\{ \begin{array}{lcl} \hat{H}^e(\vec{r}_i, \vec{R}_n) & = & \hat{T}^e + \hat{V}^{ee} + \hat{V}^{eN} \\ \hat{H}^N(\vec{R}_n, E^e(\vec{R}_n)) & = & \hat{T}^N + \hat{V}^{NN} + E^e(\vec{R}_n) \end{array} \right.$$

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- $$\begin{aligned} \hat{H}^e(\vec{r}_i, \vec{R}_n) \left|\psi_j(\vec{r}_i, \sigma_i, \vec{R}_n)\right\rangle &= E^e_j(\vec{R}_n) \left|\psi_j(\vec{r}_i, \sigma_i, \vec{R}_n)\right\rangle \\ \hat{H}^N_j(\vec{R}_n, E^e_j(\vec{R}_n)) \left|\xi_{j\nu}(\vec{R}_n, E^e_j)\right\rangle &= \left(E^e_j(\vec{R}_n) + E^N_{j\nu}\right) \left|\xi_{j\nu}(\vec{R}_n, E^e_j)\right\rangle \\ \hat{H} \left|\Psi_{j\nu}\right\rangle &= \left(E^e(\vec{R}_n) + E^N\right) \left|\psi_j(\vec{r}_i, \sigma_i, \vec{R}_n)\right\rangle \left|\xi_{j\nu}(\vec{R}_n, E^e_j)\right\rangle \end{aligned}$$

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Validity of the BO approximation

$$\hat{T}^N|\psi(\vec{r}_i,\vec{R}_n)\rangle \quad \simeq \quad 0$$

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Mixed phonon-magnon mode (2 magnons)

$$\begin{aligned} \hat{H}\,|\Psi\rangle &= E\,|\Psi\rangle \\ |\Psi(\vec{r}_i,\sigma_i,\vec{R}_n)\rangle &= \sum_{\nu} c_{j,\nu} \left|\psi_j(\vec{r}_i,\sigma_i,\vec{R}_n)\right\rangle \left|\xi_{j\nu}(\vec{R}_n, E^e_j)\right\rangle + \sum_{\mu} c_{l,\mu} \left|\psi_l(\vec{r}_i,\sigma_i,\vec{R}_n)\right\rangle \left|\xi_{l\mu}(\vec{R}_n, E^e_l)\right\rangle \end{aligned}$$

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From an ab-initio point of view : within BO

Application of an electric field within the dipolar approx.

$$\hat{H} + \hat{\mathcal{V}} = \hat{H} - \frac{1}{\hbar\omega}\left[\hat{H}_0\,,\, \widehat{\vec{d}} \cdot \vec{\mathcal{E}}_0\right] \quad \text{with} \quad \widehat{\vec{d}} = \widehat{\vec{d}_e} + \widehat{\vec{d}_N}$$

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Action of $\hat{\mathcal{V}}$ on GS : 2 possibilities

	Phonons transition	Electronic/magnon transition
Init. state	$\vert\psi_0\xi_{00}\rangle$	

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Coupling	$\langle\xi_{0\nu}\|\widehat{\vec{d}_N}\cdot\vec{\mathcal{E}}_0\|\xi_{00}\rangle$	

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$\hbar\omega$	$E_{0\nu}^N - E_{00}^N$	$[E_j^e + E_{j\mu}^N] - [E_0^e + E_{00}^N]$
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$\hbar\omega$	$E^N_{0\nu} - E^N_{00}$	$[E^e_j + E^N_{j\mu}] - [E^e_0 + E^N_{00}]$
Coupling	$\langle\xi_{0\nu}\vert\widehat{\vec{d}_N}\cdot\vec{\mathcal{E}}_0\vert\xi_{00}\rangle$	$\langle\psi_j\vert\widehat{\vec{d}_e}\cdot\vec{\mathcal{E}}_0\vert\psi_0\rangle$

From an ab-initio point of view : within BO

Spin-charge decoupling

Usual representation

- Nuclear, electronic motions decoupling (BO approximation)
  $|\Psi(\vec{r}_i,\sigma_i,\vec{R}_n)\rangle = |\psi(\vec{r}_i,\sigma_i,\vec{R}_n)\rangle \; |\xi(\vec{R}_n)\rangle$
- Decoupling of electronic and spin degrees of freedom
  $|\Psi(\vec{r}_i,\sigma_i,\vec{R}_n)\rangle = \underbrace{|\zeta(\sigma_i,\vec{r}_i,\vec{R}_n)\rangle}_{\textit{spin}} \; \underbrace{|\phi(\vec{r}_i,\vec{R}_n)\rangle}_{\textit{charge}} \; \underbrace{|\xi(\vec{R}_n)\rangle}_{\textit{phonon}}$

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Action of electric field

- $\langle\psi_j|\widehat{\vec{d}_e}\cdot\vec{\mathcal{E}}_0|\psi_0\rangle$ acts on charge part
  (Ex : modification of space part of magn. orb.)

How can electric field act on magnons ?

From an ab-initio point of view : within BO

Usual spin Hamiltonians

- Heisenberg : $\sum_{i,j} J_{ij} \widehat{\vec{S}_i} \cdot \widehat{\vec{S}_j} \rightarrow$ unique space part for all spin states
  - unique definition of supporting magnetic orbital

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- SO effects
    - single ions anisotropy
    - DM interaction

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Different space parts according to spin states

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Spin Hamiltonian : not really a spin-space decoupling

(no factorisation of WF)

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  Different space parts according to spin states
  Spin Hamiltonian : not really a spin-space decoupling
  (no factorisation of WF)

Action on magnons ?

- $\langle \psi_j | \widehat{\vec{d}_e} \cdot \vec{\mathcal{E}}_0 | \psi_0 \rangle$ acts on charge part
- Action on magnons $\Longrightarrow$ OK if no spin-charge decoupling

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Excitation toward a mixed phonon-magnon state

$$|\psi_0\xi_{00}\rangle \longrightarrow |\Psi\rangle = \sum_\nu c_{j,\nu} \left|\psi_j\right\rangle \left|\xi_{j\nu}\right\rangle + \sum_\mu c_{l,\mu} \left|\psi_l\right\rangle \left|\xi_{l\mu}\right\rangle$$

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- $\widehat{\vec{d}^e} \cdot \vec{\mathcal{E}}_0$ on electronic part : two terms
  $\Longrightarrow$ potential increase of the efficiency

From an ab-initio point of view : beyond BO

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- Action on magnon $\Longrightarrow$ breaking of spin-charge separation

$$|\psi_0\xi_{00}\rangle \longrightarrow |\Psi\rangle \neq \underbrace{\left(\sum_\nu c_{j,\nu} \left|\phi_j\right\rangle \left|\xi_{j\nu}\right\rangle + \sum_\mu c_{l,\mu} |\phi_l\rangle \left|\xi_{l\mu}\right\rangle\right)}_{\textit{entangled charge-phonons}} \underbrace{|\zeta_m\rangle}_{\textit{spin}}$$

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$$|\psi_0\xi_{00}\rangle \longrightarrow |\Psi\rangle = \left(\sum_{\nu} c_{j,\nu,p} |\zeta_p\rangle\left|\phi_j\right\rangle \left|\xi_{j\nu}\right\rangle + \sum_{\mu} c_{l,\mu,q} |\zeta_q\rangle|\phi_l\rangle \left|\xi_{l\mu}\right\rangle\right)$$

Conclusion

Electro-active magnon

- Not a specific to mutiferroics

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- Does not requires a mixed phonon-magnon state

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Mixed (entangled) phonon-magnon excited state

- Not necessarily electro-active

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Mixed (entangled) phonon-magnon excited state

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- Cannot be alone, associated with mixed phonon-magnon GS or other excited state

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