Strain-mediated dynamical magnetoelectricity in multiferroic $BiFeO_3$

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Electromagnon
Orsay, France

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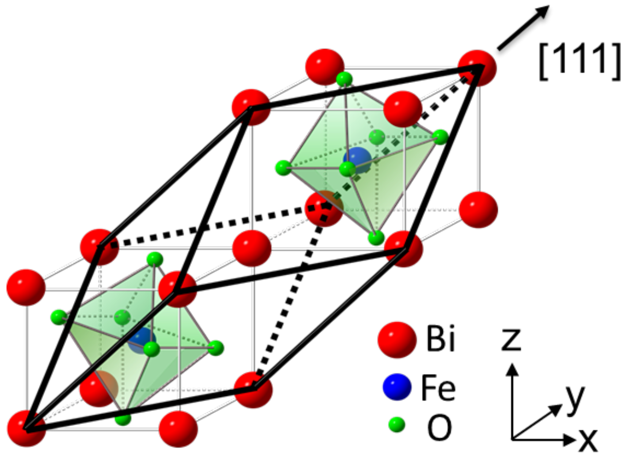
OBJECTIVES

- Dynamical MagnetoElectric Properties of $BiFeO_3$.
- Investigate Electromagnons in $BiFeO_3$.
- Reveal alternatives to electromagnons.

STRUCTURE OF $BiFeO_3$ AS A PROTOTYPE OF MULTIFERROICS

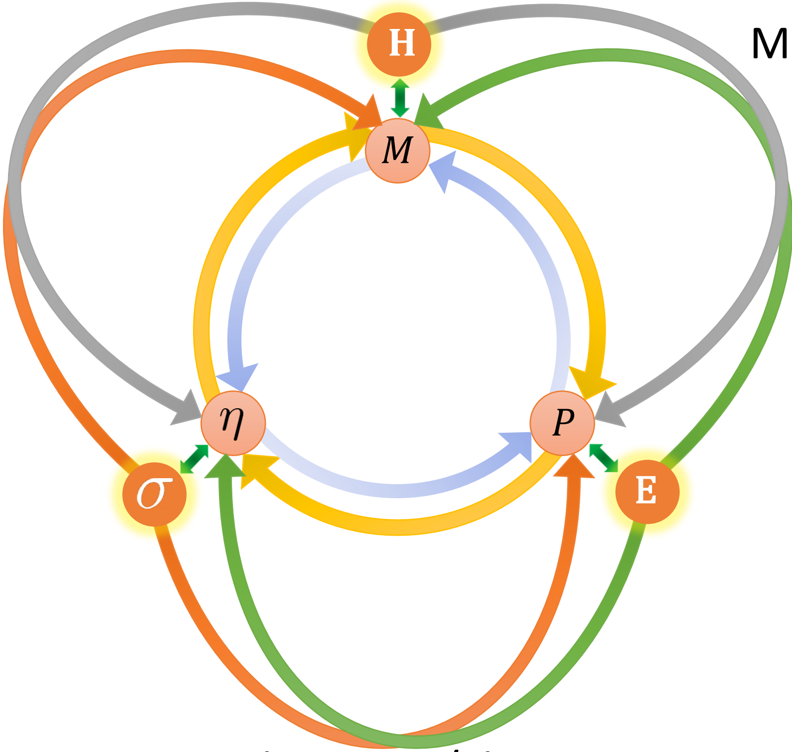


H. Naganuma, (ed. Lallart, M.) InTech, 2011

CROSS COUPLING BETWEEN DEGREES OF FREEDOM

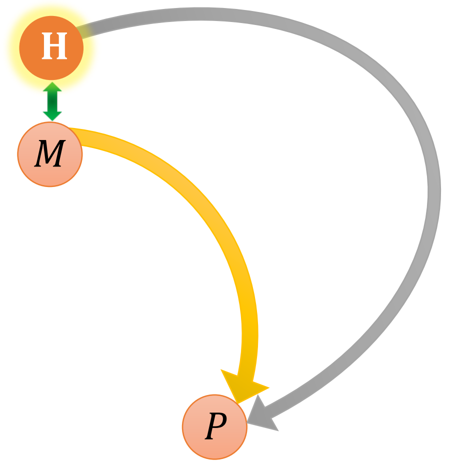
Piezomagneticity/Magnetostriction

Magnetoelectricity



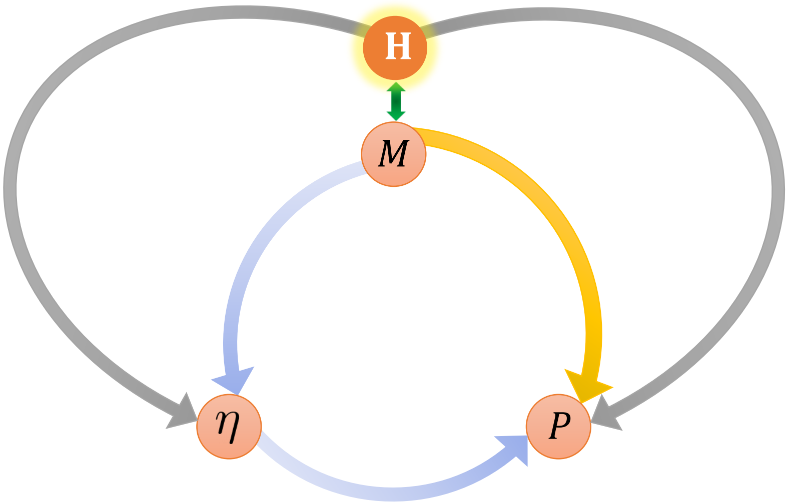
Piezoelectricity /Electrostriction

CROSS COUPLING BETWEEN DEGREES OF FREEDOM



DIRECT MAGNETOELECTRICITY

CROSS COUPLING BETWEEN DEGREES OF FREEDOM



INDIRECT (STRAIN-INDUCED) MAGNETOELECTRICITY

Quadratic MagnetoElectric Constant

$$F(\vec{E},\vec{H}) = F_0 - P_i^S E_i - M_i^S H_i$$
$$-\tfrac{1}{2}\epsilon_0\epsilon_{ij}E_iE_j - \tfrac{1}{2}\mu_0\mu_{ij}H_iH_j - \alpha_{ij}E_iH_j$$
$$-\tfrac{1}{2}\beta_{ijk}E_iH_jH_k - \tfrac{1}{2}\gamma_{ijk}H_iE_jE_k - \cdots$$

$$\Rightarrow$$

$$P_i(\vec{E},\vec{H}) = -\frac{\partial F}{\partial E_i}$$
$$= P_i^S + \epsilon_0\epsilon_{ij}E_j + \alpha_{ij}H_j$$
$$+ \frac{1}{2}\beta_{ijk}H_jH_k + \gamma_{ijk}H_iE_j - \cdots$$

Quadratic MagnetoElectric Constant

$$F(\vec{E},\vec{H}) = F_0 - P_i^S E_i - M_i^S H_i$$
$$-\tfrac{1}{2}\epsilon_0\epsilon_{ij}E_iE_j - \tfrac{1}{2}\mu_0\mu_{ij}H_iH_j - \alpha_{ij}E_iH_j$$
$$-\tfrac{1}{2}\beta_{ijk}E_iH_jH_k - \tfrac{1}{2}\gamma_{ijk}H_iE_jE_k - \cdots$$

$$P_i(\vec{E},\vec{H}) = -\frac{\partial F}{\partial E_i}$$
$$= \boxed{P_i^S} + \epsilon_0\epsilon_{ij}E_j + \boxed{\alpha_{ij}H_j}$$
$$+ \boxed{\frac{1}{2}\beta_{ijk}H_jH_k} + \gamma_{ijk}H_iE_j - \cdots$$

- In presence of only a magnetic field $P_i = P_i^s + \alpha_{ij}H_j + \frac{1}{2}\beta_{ijk}H_jH_k$

α_{ij} and β_{ij}: Linear and Quadratic magnetoelectric (ME) Coefficients

- If the magnitude of the applied magnetic field is large enough, the effect of linear ME coefficient can be neglected in comparison with the quadratic coefficient

$$\boxed{P_i = P_i^s + \frac{1}{2}\beta_{ijk}H_jH_k}$$

ATOMISTIC SIMULATIONS

- Technique: Effective Hamiltonian[1]

✓ Accuracy of first principles methods

✓ Not limited to T= 0 K

✓ Not limited to small cells

- Simulation type: Molecular Dynamics[2] (MD)
- Δt: 0.5 fs, cells:12x12x12,
- Temperature: 1 K (In order to have better statistics T=1 K)
- DC Magnetic field along $[11\bar{2}]$
- AC Magnetic field along $[11\bar{2}]$ (ω: to be changed in each simulation)
- Material: $BiFeO_3$ (Bulk)

[1] *Adv. Funct. Mater.* **23**, 234 (2003) by S. Prosandeev *et al*.

[2] *Phys Rev. Lett.* **109**, 067203 (2012) by D. Wang *et al*.

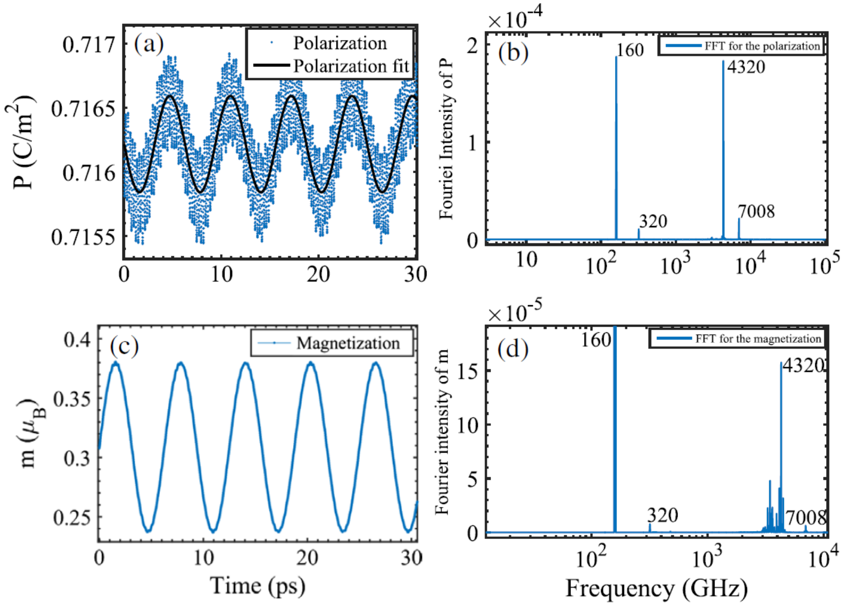
ATOMISTIC SIMULATIONS

1. **Clamped simulations:** frozen supercell lattice vectors (strain fixed)

2. **Unclamped simulations:** homogeneous strain allowed to change (strain relaxed)

Case 1: Strain Fixed

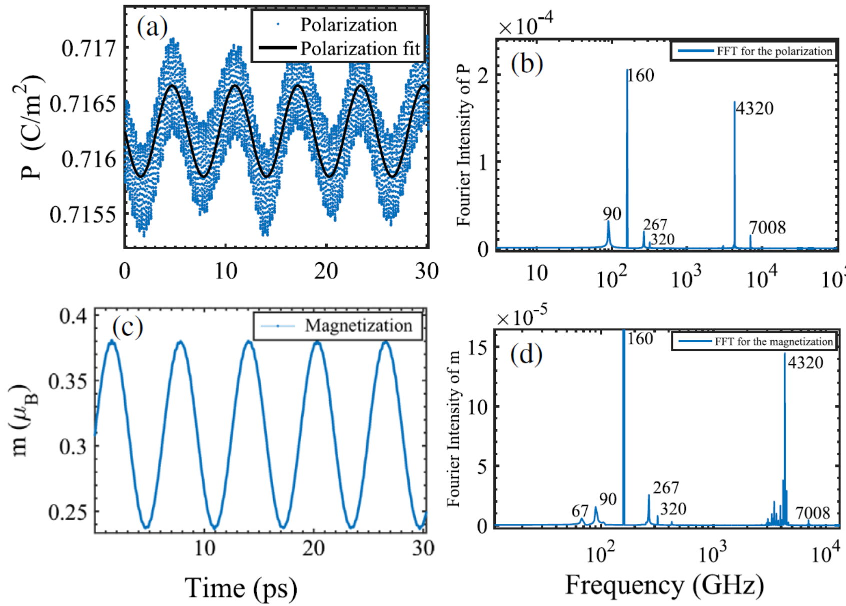
- Polarization follows magnetic field (**magnetoelectricity** verified!)
- Existence of dual vibrations with the frequencies of phonons around 4320 and 7000 GHz (**electromagnonic** nature of BFO!)
- Existence of the second harmonic



S. Sayedaghaee *et al.*, *Phys. Rev. Lett.* **122**, 097601 (2019).

Case 2: Strain Relaxed

- Additional vibrations with the frequencies of about 90 and 267 GHz are observed
- Existence of what we coined **electroacoustic magnon** quasiparticles – that mix optical phonons, acoustic phonons and magnons



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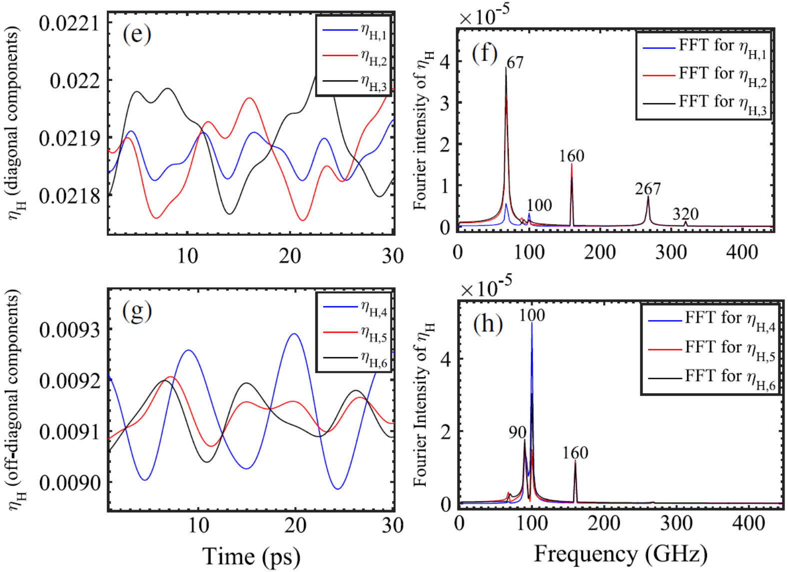
Case 2: Strain Relaxed



- The 267 GHz mechanical resonance corresponds to oscillations of diagonal strain components, (η_1, η_2, η_3) , whereas the origin of the frequency of 90 GHz is found in the shear components of the strain (η_4, η_5, η_6)

S. Sayedaghaee *et al.*, *Phys. Rev. Lett.* **122**, 097601 (2019).

Model

- Model Hamiltonian:

$$\mathcal{H} = \frac{1}{2}m_P\dot{P}^2 + \frac{1}{2}m_\eta\dot{\eta}^2 + \frac{1}{2}kP^2 + \frac{1}{4}bP^4 + \frac{1}{2}\epsilon P^2M^2 - EP + \frac{1}{2}Q\eta P^2 + \frac{1}{2}\lambda\eta M^2 + \frac{1}{2}c\eta^2$$

Bi-quadratic ME effect (ϵ); electrostriction (Q); elastic constant (c); mass (m_P, m_η); harmonic constant (k); anharmonic constant (b); electric field (E); magnetostriction (λ)

- Newtonian dynamical equations:

$$\begin{aligned}\left(\omega_P^2 - \omega^2 + i\Gamma_P\omega\right)\tilde{P} \quad = \quad & -\xi_P\int d\omega_1\int d\omega_2\tilde{P}(\omega-\omega_1)\tilde{P}(\omega_1-\omega_2)\tilde{u}(\omega_2) \\ & \underbrace{-\,\xi_{PM}\int d\omega_1\int d\omega_2\tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_2)\tilde{M}(\omega_2)}_{\text{(Direct ME coupling)}} \\ & \underbrace{+\,\frac{1}{2}\tilde{Q}_P\tilde{Q}_\eta\int d\omega_1\int d\omega_2\tilde{P}(\omega-\omega_1)\frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\tilde{P}(\omega_1-\omega_2)\tilde{P}(\omega_2)}_{\text{(Electrostriction)}} \\ & \underbrace{+\,\frac{1}{2}\tilde{Q}_P\tilde{\lambda}\int d\omega_1\int d\omega_2\tilde{P}(\omega-\omega_1)\frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\tilde{M}(\omega_1-\omega_2)\tilde{M}(\omega_2)}_{\text{Electrostriction + Magnetostriction}}\end{aligned}$$

Model

- $\beta(0,\omega_0)$ couples the dc and ac fields

$$\beta(0,\omega_0) = -\xi_{PM}\frac{\chi_M(0)\chi_M(\omega_0)\tilde{P}(0)}{\tilde{\omega}_P^2-\omega_0^2+i\Gamma_P\omega_0}+\frac{\tilde{Q}_P\tilde{\lambda}}{2}\frac{\chi_M(0)\chi_M(\omega_0)\tilde{P}(0)}{(\tilde{\omega}_P^2-\omega_0^2+i\Gamma_P\omega_0)(\omega_\eta^2-\omega_0^2+i\Gamma_\eta\omega_0)}$$

- $\beta(\omega_0,\omega_0)$ couples the ac field with itself

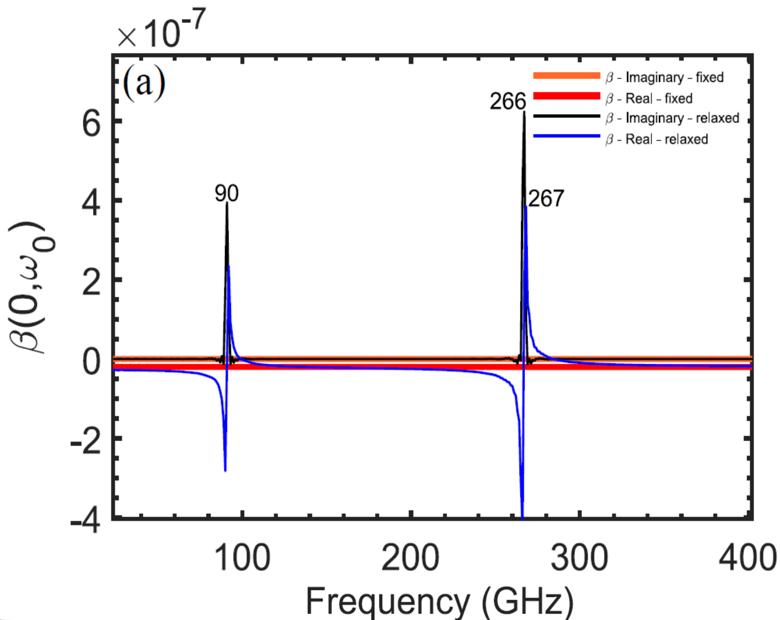
$$\beta(\omega_0,\omega_0) = -\xi_{PM}\frac{\chi_M(\omega_0)^2\tilde{P}(0)}{\tilde{\omega}_P^2-4\omega_0^2+i\Gamma_P 2\omega}+\frac{\tilde{Q}_P\tilde{\lambda}}{2}\frac{\chi_M(\omega_0)^2\tilde{P}(0)}{(\tilde{\omega}_P^2-4\omega_0^2+i\Gamma_P 2\omega_0)(\omega_\eta^2-4\omega_0 2^2+i\Gamma_\eta 2\omega)}$$

Model

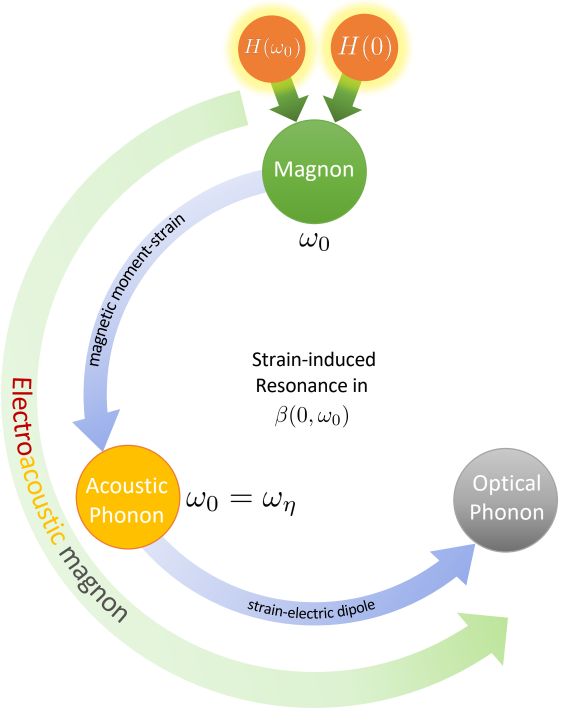
$$\beta(0,\omega_0) = -\xi_{PM}\frac{\chi_M(0)\chi_M(\omega_0)\tilde{P}(0)}{\tilde{\omega}_P^2-\omega_0^2+i\Gamma_P\omega_0} + \frac{\tilde{Q}_P\tilde{\lambda}}{2}\frac{\chi_M(0)\chi_M(\omega_0)\tilde{P}(0)}{(\tilde{\omega}_P^2-\omega_0^2+i\Gamma_P\omega_0)(\omega_\eta^2-\omega_0^2+i\Gamma_\eta\omega_0)}$$

- $\beta(0,\omega_0)$ generates a signal at ω_0.

- Resonances:
  - $\omega_0 \approx \omega_{magnon}$
  - $\omega_0 \approx \omega_{phonon}$
  - $\omega_0 \approx \omega_\eta$



Magnetoelectric response

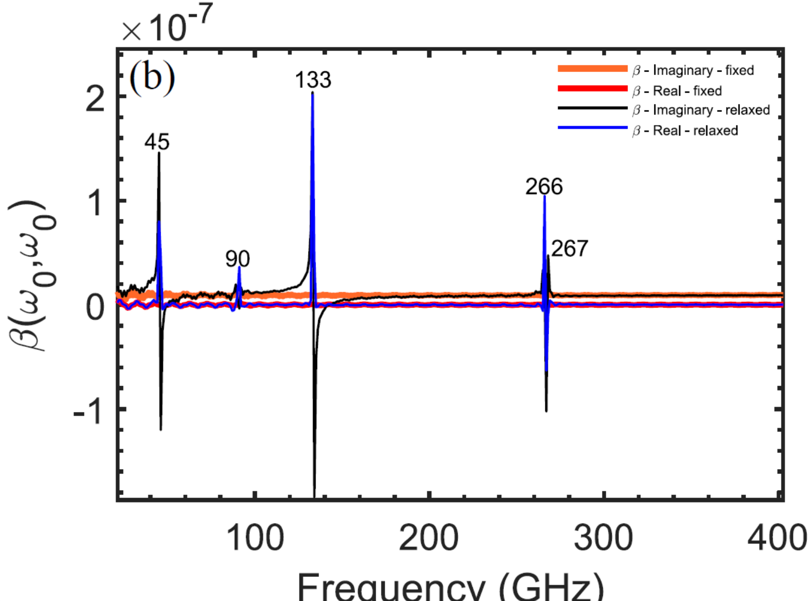


Model

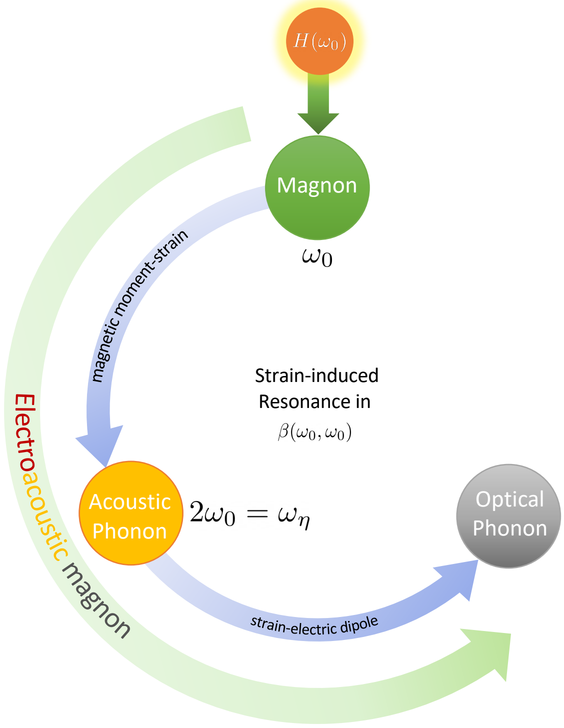
$$\beta(\omega_0,\omega_0) = -\xi_{PM}\frac{\chi_M(\omega_0)^2\tilde{P}(0)}{\tilde{\omega}_P^2 - 4\omega_0^2 + i\Gamma_P 2\omega_0} + \frac{\tilde{Q}_P\tilde{\lambda}}{2}\frac{\chi_M(\omega_0)^2\tilde{P}(0)}{(\tilde{\omega}_P^2 - 4\omega_0^2 + i\Gamma_P 2\omega_0)(\omega_\eta^2 - 4\omega_0^2 + i\Gamma_\eta 2\omega_0)}$$

- $\beta(\omega_0, \omega_0)$ generates a signal at $2\omega_0$ (Second Harmonic Generation).

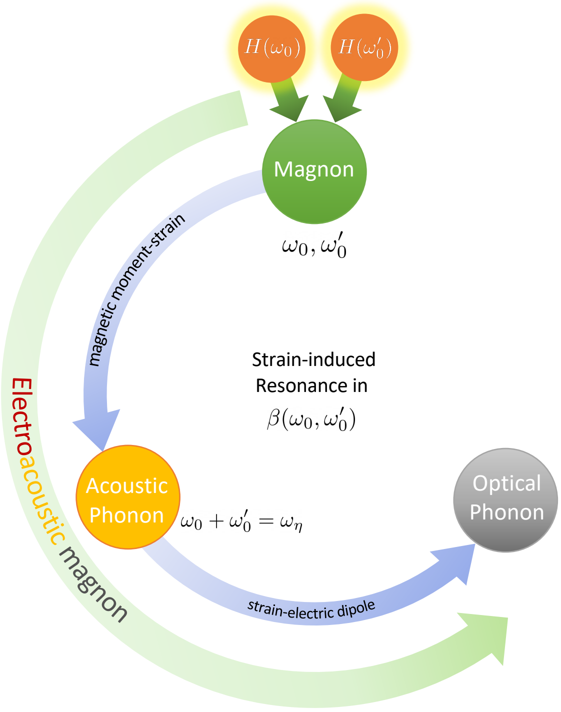
- Resonances:
  - $\omega_0 \approx \omega_{magnon}$
  - $\omega_0 \approx \frac{\omega_{phonon}}{2}$
  - $\omega_0 \approx \frac{\omega_\eta}{2}$



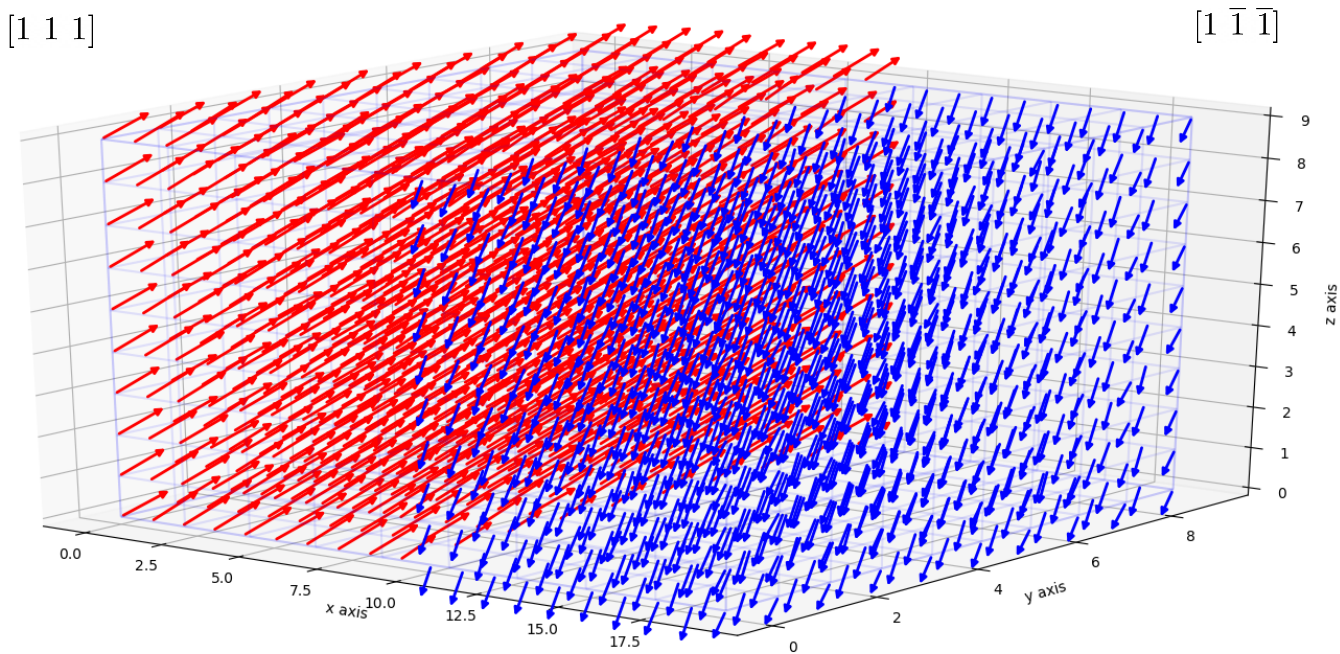
Magnetoelectric response



Magnetoelectric response

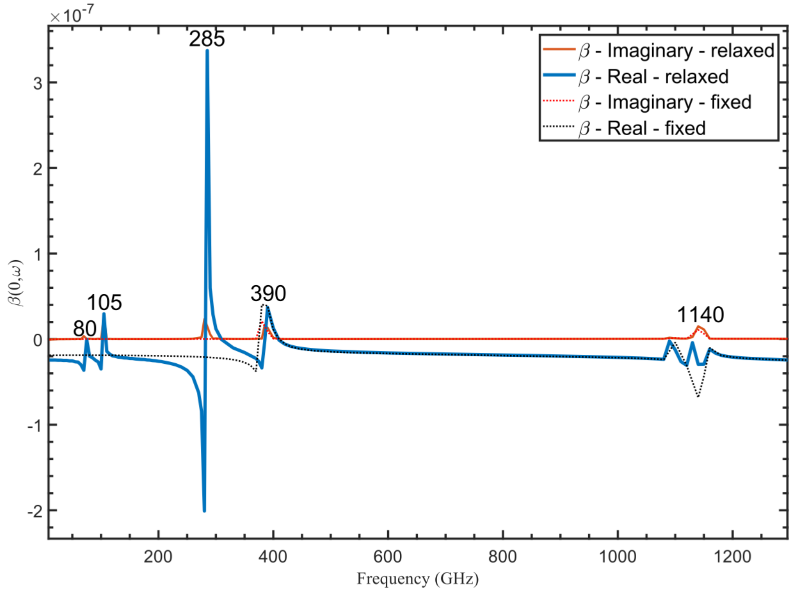


For the case of BFO adopting 109° multidomains



	D1		D2	
Polarization	$[1\ 1\ 1]$	$\rightarrow$	$[1\ \bar{1}\ \bar{1}]$	109° structure
Magnetization	$[\bar{2}\ 1\ 1]$		$[\bar{2}\ 1\ 1]$	
AFM	$[0\ 1\ \bar{1}]$		$[0\ 1\ \bar{1}]$	

Results for Magnetoelectric response



- 109˚ multidomain structure: monodomain frequencies still observed.
- Specific additional vibrations at 390 and 1150 GHz, independent of strain fixed or relaxed → localized electromagnons in the DW.

CONCLUSIONS

Mechanical strain leads to the formation of new quasiparticles (electroacoustic magnons) that takes part in the enhancement of magnetoelectric responses for both cases of monodomain and multidomain structures.

Design and development of novel devices by (1) tailoring the shape and size of the samples to tune the resonance frequency of the electroacoustic magnons; (2) using localized electromagnons (in the multidomain case)

Due to the generality of our theoretical model, it can be applied to a wide scope of materials as well as non-linear physics.