Observing FE and AFM domain using SHG in Spin-Spiral Multiferroics

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Neumann's Principle

Established in 9th century, and states:

The symmetry elements of any physical property of a crystal must include all the symmetry elements of the point group of the crystal.

➡ Symmetry operations of the crystal class $\subseteq$ Symmetry operations of any physical property

➡ In other words,
the tensor representing a physical property must always be invariant with regard to every symmetry operation of the crystal class.

Symmetry ➡ Relation between tensor components ➡ Reduces the number of independent tensor components

Ferroelectricity

In general, *long-range order reduce the symmetry of a crystal*.

Spontaneous polarization violates inversion symmetry since it reverses when **r** is replaced by –**r**.

Thus, ferroelectricity always violates spatial-inversion symmetry.

Broken symmetry operations lead to domains.

Recently discovered: If a magnetically ordered state violates the spatial inversion symmetry, it can lead to spontaneous electric polarization.

Spiral-spin ordering is one such magnetically order state

Spiral-Spin Magnetism

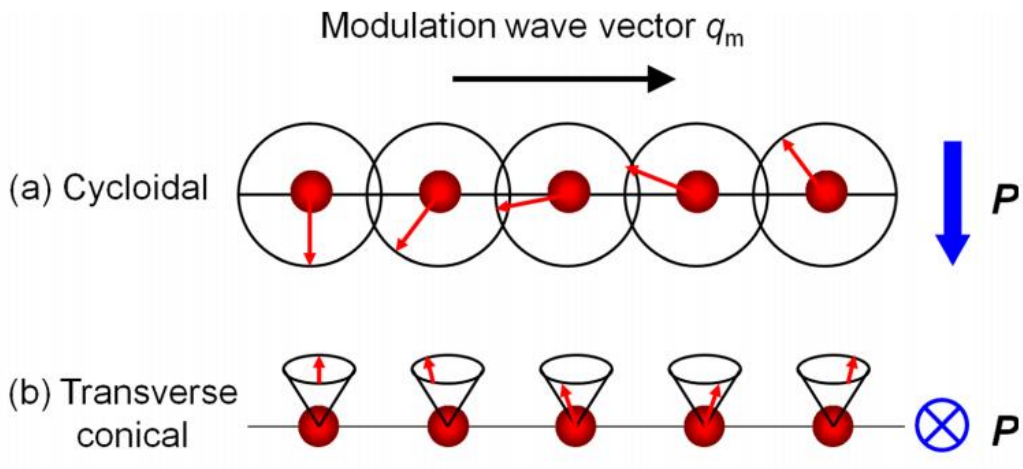
Simultaneously breaking of spatial-inversion and time-reversal symmetries

Ferroelectricity is induced by the inherent magnetic configuration
Inverse effect of the Dzyaloshinskii-Moriya (DM) interaction

$$\boldsymbol{P} \propto \boldsymbol{e_{ij}} \times (\boldsymbol{S_i} \times \boldsymbol{S_j})$$

$\boldsymbol{e_{ij}}$ is the unit vector connecting the neighboring sites *i* and *j*

This product transforms like electric polarization and connects magnetic spiral order to FE order.



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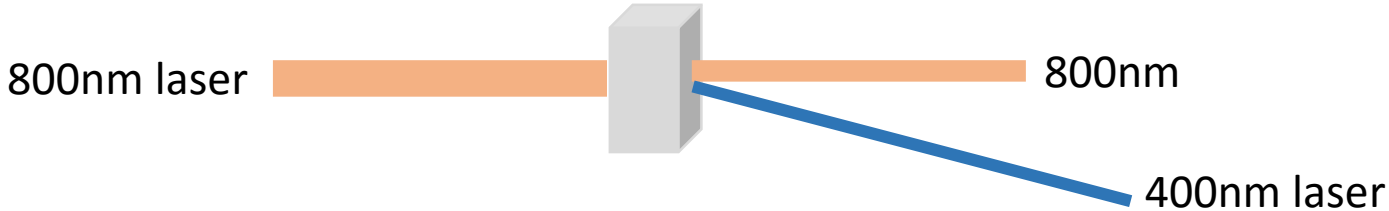
Brief overview of SHG



$$\boldsymbol{P_k(E)} = C + \varepsilon_o \chi_{ik}^{(1)} E_k + \varepsilon_o \chi_{ijk}^{(2)} E_i E_j + \varepsilon_o \chi_{ijlk}^{(3)} E_i E_j E_l + \ \ldots$$

Constant DC term

1st order Radiation at ω

2nd order perturbation

$$P_k(E) = \ \varepsilon_o \chi_{ijk}^{(2)} \quad \left(E_i E_j e^{-i2\omega t} + \ E_i^* E_j^* e^{i2\omega t} + \ E_i^* E_j + E_j^* E_i\right)$$

Cos (2ω)

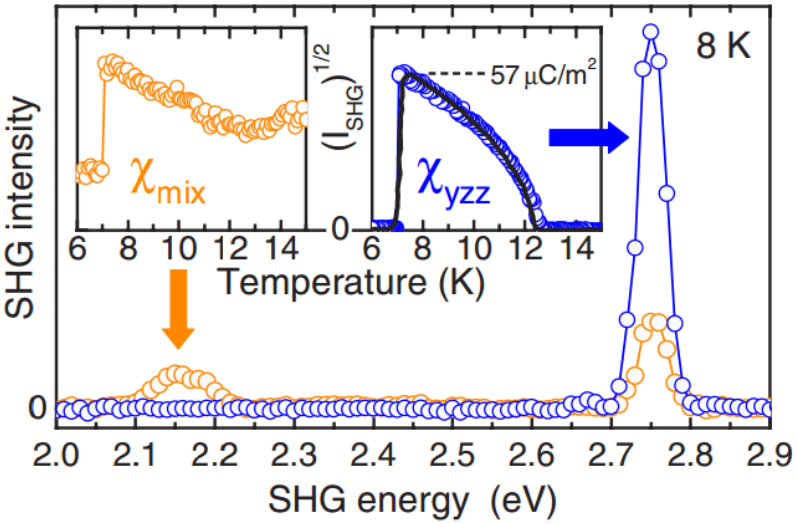
Second Harmonic Generation (SHG)

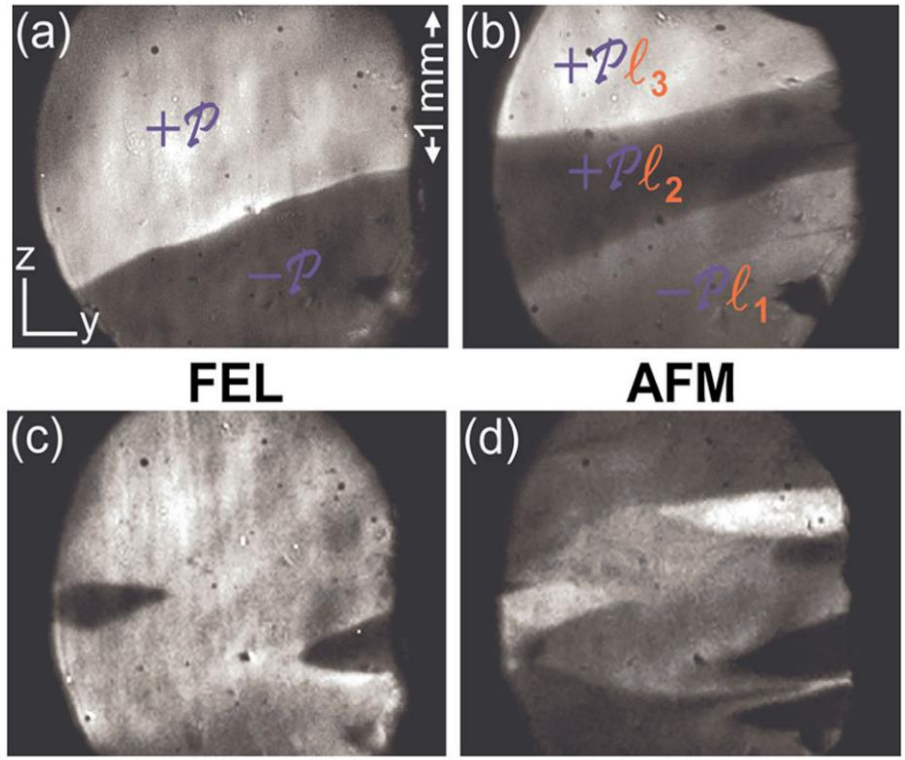
Oscillation at frequency 2ω

Radiation emitted at 2ω

Observing FE and AFM domain using SHG

Coexistence of FEL and AFM domains in zero-field-cooled **$MnWO_4$**(100). FEL and AFM domains are denoted by *P* and *l*, respectively.





(a) and (c) obtained at 8 K with SHG at 2.75 eV from $(\chi_{yzz} + \chi_{para})$

(b) and (d) obtained at 8 K with SHG at 2.15 eV from $(\chi_{AFM} + \chi_{para})$

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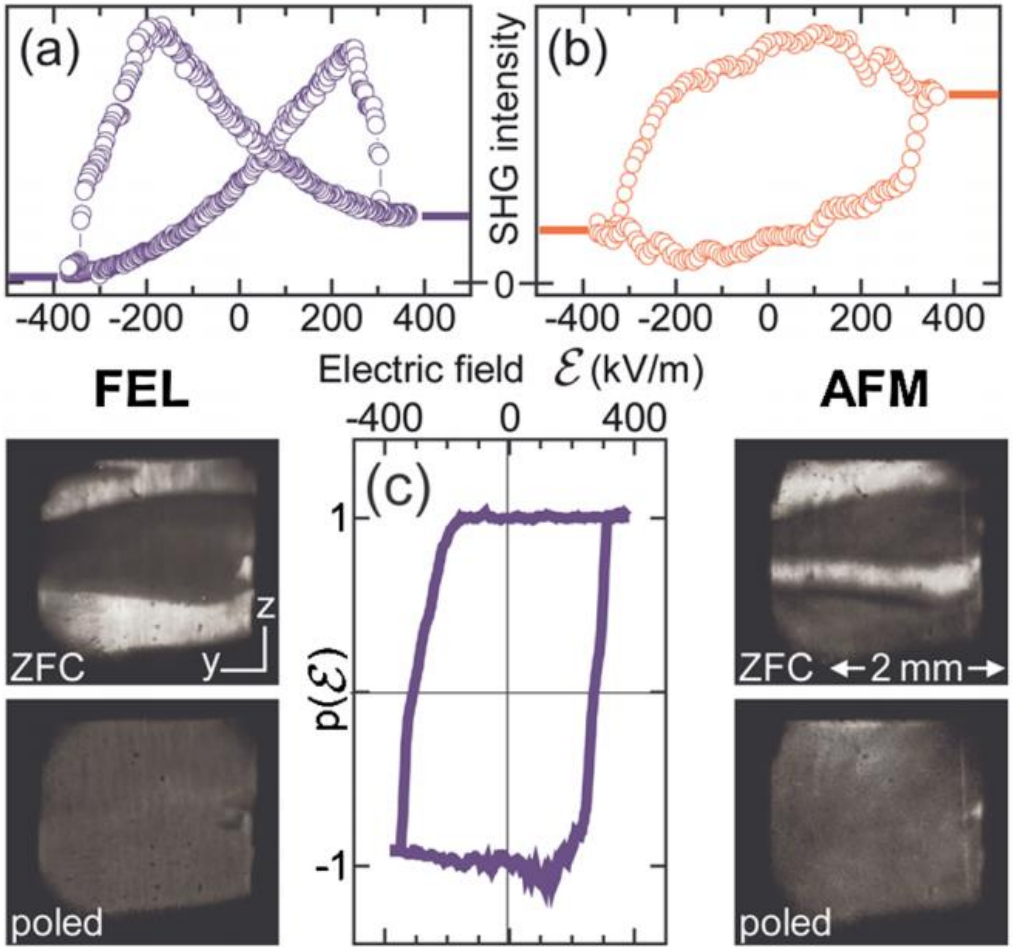
Observing FE and AFM domain using SHG

Control of AFM domain structures by electric fields



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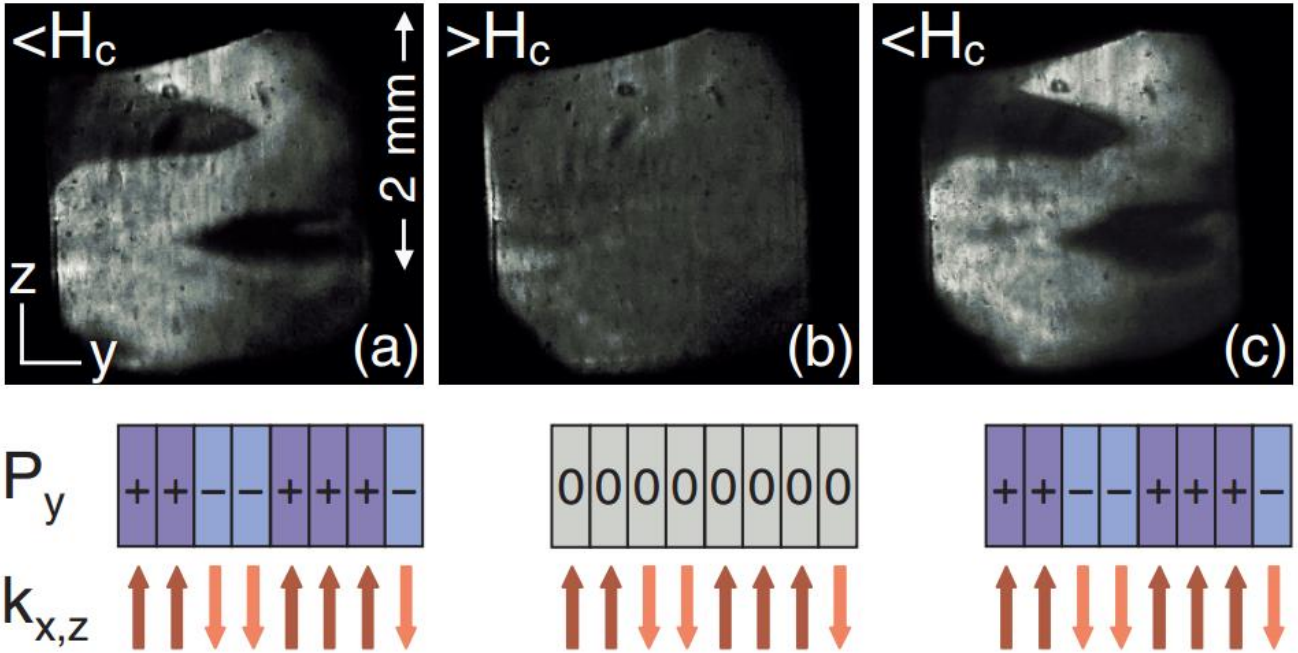
Observing FE and AFM domain using SHG

Control of FEL domain structures by magnetic fields



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Thank You