Coexistence of ferromagnetism and superconductivity in iron based pnictides: a time resolved magnetooptical study (supplemental information)

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Optical response in the conical helimagnetic state

Assuming that within a single plane Eu^{2+} spins are ordered ferromagnetically Eq. (1) can be applied to each plane separately. Neglecting for simplicity the crystallographic details and assuming a conical helimagnetic phase with $\mathbf{M} = M_0[\cos(q_0 z)\sin(\theta), \sin(q_0 z)\sin(\theta), \cos(\theta)]$, where q_0 represents the helix propagation wavevector and θ the local magnetization angle relative to the c-axis, we introduce following Ref. [1] a local coordinate system,

$$\begin{aligned}\hat{x}' =&[\hat{x}\cos(q_0 z) + \hat{y}\sin(q_0 z)]\cos(\theta) - \hat{z}\sin(\theta),\\ \hat{y}' =&-\hat{x}\sin(q_0 z) + \hat{y}\cos(q_0 z),\\ \hat{z}' =&[\hat{x}\cos(q_0 z) + \hat{y}\sin(q_0 z)]\sin(\theta) + \hat{z}\cos(\theta).\end{aligned} \tag{S1}$$

Here ˆ represents the Cartesian unit vectors. The modes of a spin-operators linearized Hamiltonian are then represented by,[2]

$$\begin{aligned}\delta M_{x'}(z) \propto \sum_m s_{x'}(\mathbf{q} + mq_0\hat{z})e^{i[(\mathbf{q}+mq_0\hat{z})\cdot\mathbf{r} - \omega(\mathbf{q})t]},\\ \delta M_{y'}(z) \propto \sum_m s_{y'}(\mathbf{q} + mq_0\hat{z})e^{i[(\mathbf{q}+mq_0\hat{z})\cdot\mathbf{r} - \omega(\mathbf{q})t]},\end{aligned} \tag{S2}$$

where m are integers and $s_{i'}(\mathbf{q})$ and $\omega(\mathbf{q})$ depend on the particular choice of Hamiltonian.[1, 2] Here nonzero m terms need to be introduced because in the presence of an in-plane external magnetic field higher harmonics appear in the modulation.[2] An in-plane anisotropy also introduces higher harmonic terms. Transforming back to the crystal coordinate system and taking into account only the oscillating part by omiting the terms containing $\delta M_{z'}$ we obtain:

$$\begin{aligned}\delta M_x(z) \propto& e^{-i\omega(\mathbf{q})t}\cos(\theta)[\cos(q_0\hat{z})\sum_m s_{x'}(\mathbf{q}+mq_0\hat{z})e^{i(\mathbf{q}+mq_0\hat{z})\cdot\mathbf{r}} - \sin(q_0\hat{z})\sum_m s_{y'}(\mathbf{q}+mq_0\hat{z})e^{i(\mathbf{q}+mq_0\hat{z})\cdot\mathbf{r}}],\\ \delta M_y(z) \propto& e^{-i\omega(\mathbf{q})t}\cos(\theta)[\sin(q_0\hat{z})\sum_m s_{x'}(\mathbf{q}+mq_0\hat{z})e^{i(\mathbf{q}+mq_0\hat{z})\cdot\mathbf{r}} + \cos(q_0\hat{z})\sum_m s_{y'}(\mathbf{q}+mq_0\hat{z})e^{i(\mathbf{q}+mq_0\hat{z})\cdot\mathbf{r}}],\\ \delta M_z(z) \propto& e^{-i\omega(\mathbf{q})t}\sin(\theta)\sum_m s_{x'}(\mathbf{q}+mq_0\hat{z})e^{i(\mathbf{q}+mq_0\hat{z})\cdot\mathbf{r}}.\end{aligned} \tag{S3}$$

$\langle M_i \delta M_i \rangle$ therefore contains terms:

$$\begin{aligned}
\langle M_x \delta M_x \rangle &\propto \cos^2(\theta) \sum_m s_{x'}(q_z \hat{z} + m q_0 \hat{z}) \langle \cos^2(q_0 z) e^{i(q_z + m q_0) z} \rangle, \\
\langle M_y \delta M_y \rangle &\propto \cos^2(\theta) \sum_m s_{x'}(q_z \hat{z} + m q_0 \hat{z}) \langle \sin^2(q_0 z) e^{i(q_z + m q_0) z} \rangle, \\
\langle M_z \delta M_z \rangle &\propto \cos(\theta)\sin(\theta) \sum_m s_{x'}(q_z \hat{z} + m q_0 \hat{z}) \langle e^{i(q_z + m q_0) z} \rangle. \qquad \text{(S4)}
\end{aligned}$$

Here $\langle\rangle$ represents the average over Eu^{2+} planes. $\langle M_x \delta M_x \rangle$ and $\langle M_y \delta M_y \rangle$ are nonzero only when (i) $q_z + m q_0 = \pm 2 q_0$ or (ii) $q_z + m q_0 = 0$. In case (i) $\langle M_x \delta M_x \rangle = -\langle M_y \delta M_y \rangle$ leading to an anisotropic in-plane response while in case (ii) $\langle M_x \delta M_x \rangle = \langle M_y \delta M_y \rangle$ leading to the isotropic response. The frequencies present in the in-plane isotropic response are therefore $\omega(0 + m q_0)$.

Due to terms $\langle e^{i(q_z + m q_0) z} \rangle$ the term $\langle M_z \delta M_z \rangle$ also leads to isotropic response at frequencies $\omega(0 + m q_0)$.

The TR-MOKE response, on the other hand, is determined by $\delta\epsilon_{xy} \propto i \langle \delta M_z \rangle$ and contains terms $\langle e^{i(q_z + m q_0) z} \rangle$ that are nonzero for $q_z + m q_0 = 0$, which is identical to (ii). In a single magnetic domain sample the in-plane isotropic modes should therefore appear also in the c-axis TR-MOKE response.

Optical response in the canted antiferromagnetic state

For convenience we switch to the standard definition of the order parameters in weak ferromagnets.[3] Assuming that the magnetization at $H = 0$ is oriented along the c axis, the total magnetization displacement, $\delta\mathbf{M}$, of the quasi-FM mode would lie on an ellipse perpendicular to $\mathbf{M}$ with the AFM vector displacements, $\delta\mathbf{L}$, linear along $\mathbf{M}$, [4] while for the quasi-AFM mode $\delta\mathbf{M}$ would be linear along $\mathbf{M}$ and $\delta\mathbf{L}$ on an ellipse lying in the xy-plane. Looking at the symmetric part of the dielectric tensor for the orthorhombic case following Iida *et al.*[4] and assuming $\mathbf{L}||a$ and $\mathbf{H}||b$ it follows:

$$\begin{aligned}
\epsilon_{ii} = &\epsilon_{0,ii} + a_{iizz} M_z^2 + a_{iiyy} M_y^2 +, \\
&+ b_{iixx} L_x^2 + c_{iizx} M_z L_x. \qquad \text{(S5)}
\end{aligned}$$

Here the terms $c_{iizx} M_z L_x$ are due to the Dzyaloshinskii-Moriya interaction and are incompatible with our samples crystallographic structure ($Fmmm$).[5] The modulation of the dielectric tensor obtained from (S5) to the linear order in displacements is given by:

$$\begin{aligned}
\delta\epsilon_{ii} = &2 a_{iizz} M_z \delta M_z + 2 a_{iiyy} M_y \delta M_y + \\
&+ 2 b_{iixx} L_x \delta L_x + c_{iizx} (L_x \delta M_z + M_z \delta L_x). \qquad \text{(S6)}
\end{aligned}$$

For both magnetic modes the nearly-isotropic response can come from the term $2 a_{iizz} M_z \delta M_z$ only since $a_{xxzz} \sim a_{yyzz}$ due to the small orthorhombicity. The nearly isotropic in-plane response can therefore only be associated with δM_z. Since both modes contribute to δM_z at a finite in-plane magnetic field, when $\mathbf{M}$ is tilted away from the c-axis along the magnetic field, both should occur concurrently in the transient reflectivity response.

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