Transient magnetic scattering in thin Co/Pt multilayers upon femtosecond IR excitation

Outline

- Scientific background
- Experimental technique
- Magnetic resonant scattering in Co/Pt multilayers
- Transient magnetic scattering induced by IR laser pulses
- Simulation of transient magnetic scattering
- Conclusion

Scientific background

Motivation

Ultrafast magnetization dynamics excited by ultrashort laser pulses have attracted tremendous interest in the past years due to:

- **Intriguing physics, including fundamental interactions between spins, electrons, and lattice degrees of freedom far from equilibrium.**
- **High potential for fast energy-efficient memory applications**

Particularly important is unravelling the non-equilibrium spin dynamics on nanometer length scale where magnetic order emerges

Access to the nm scale in the ultrafast processes has been achieved due to development of XFEL's

This opportunity was recently realized for ferrimagnet

Scientific background

Ferromagnets

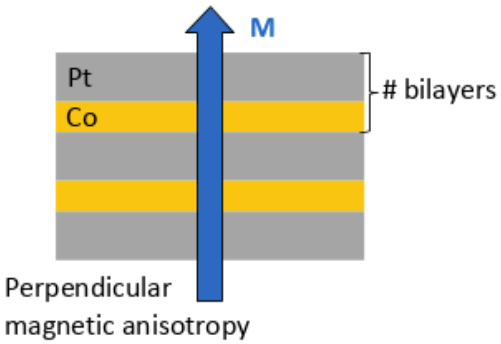
- For ferromagnetic materials the vast majority of studies were focused on the ultrafast dynamics of magnetic domain structure.
- TR-SAXS patterns were recorded in the low-q region, corresponding to magnetic domains over ~100 nm.
- The behavior of the non-equilibrium nanometer size magnetic structures was not studied so far.

Goal of the present work

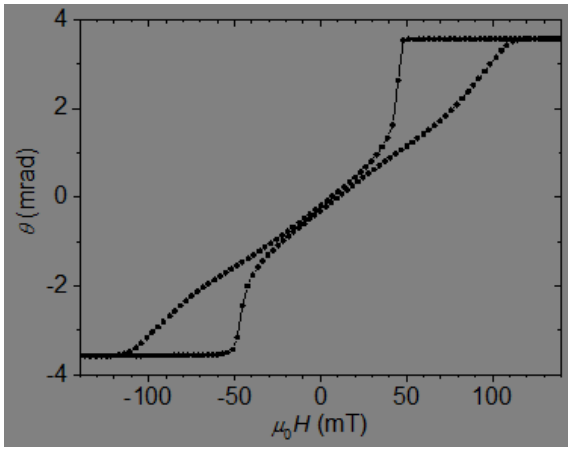
Our goal was to fill the gap and investigate the nanometer scale magnetic fluctuations in a ferromagnet, which arise after femtosecond IR pumping at the picosecond timescale.

Co/Pt samples

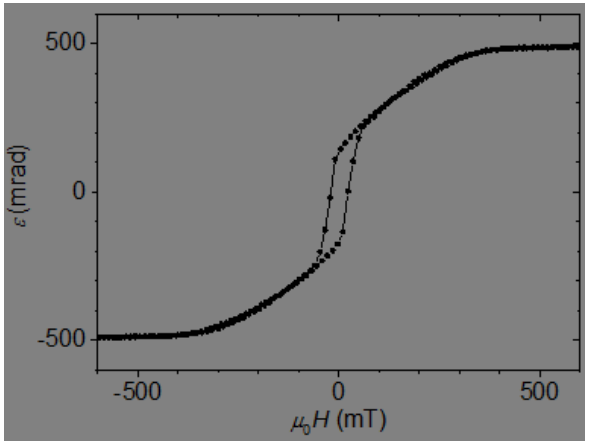
Magnetic properties of the (1.0 nm Co / 1.2 nm Pt)$_6$ sample



Easy axis out-of-plane behavior



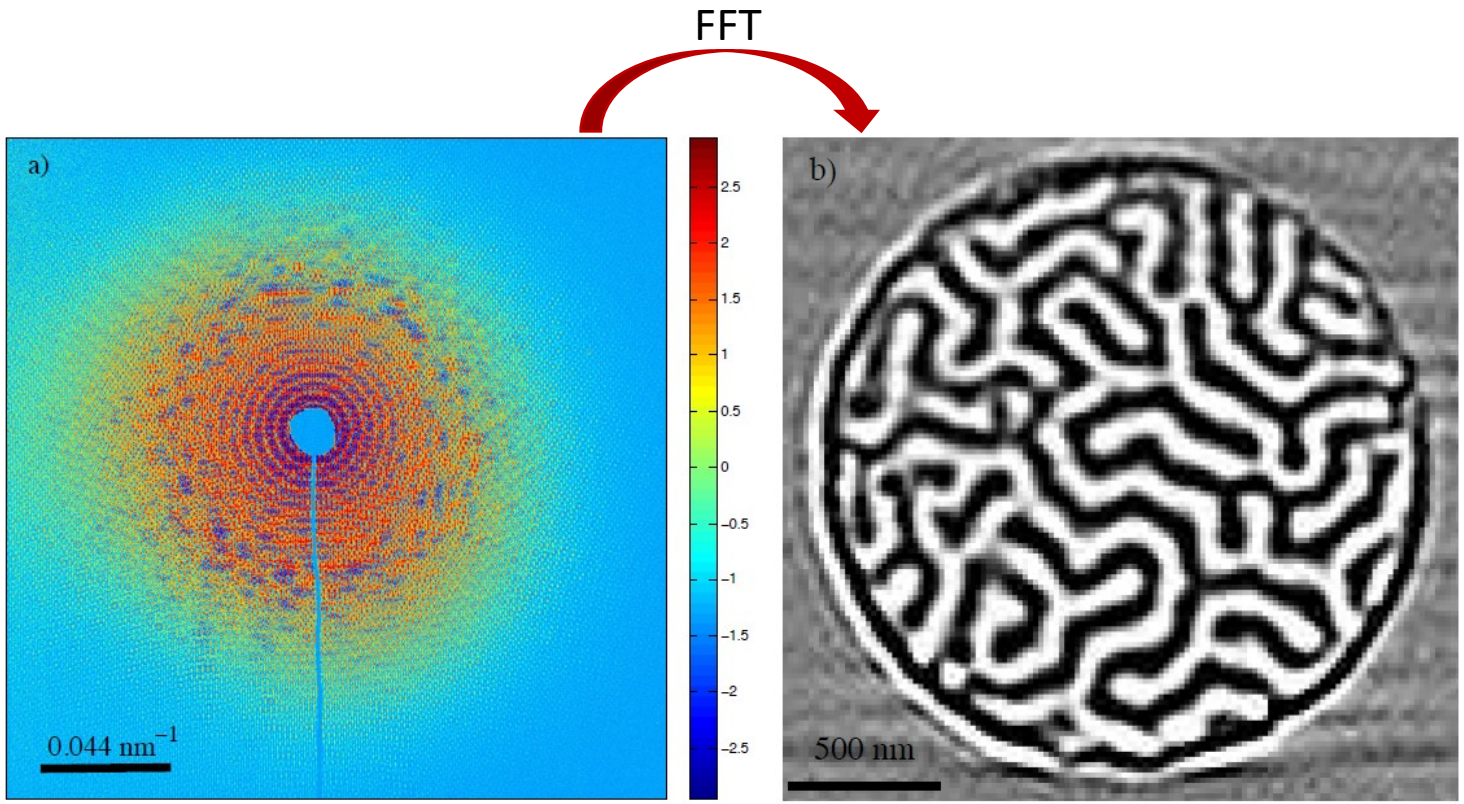
Hard axis in-plane behavior



Sample preparation details and magnetic properties are described in:
G. Winkler, A. Kobs et al., J. Appl. Phys. 117, 105306 (2015).

Domain structure of the Co/Pt sample

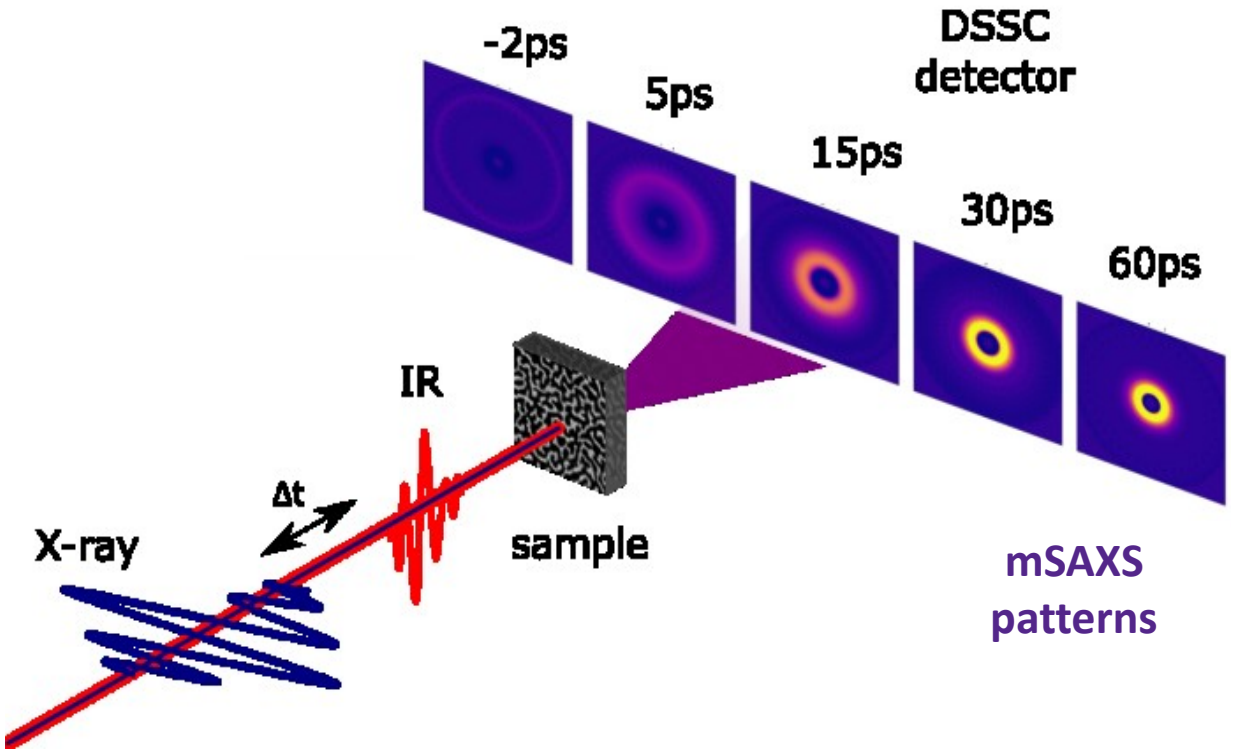
Magnetic imaging by Fourier transform holography (at FERMI FEL)



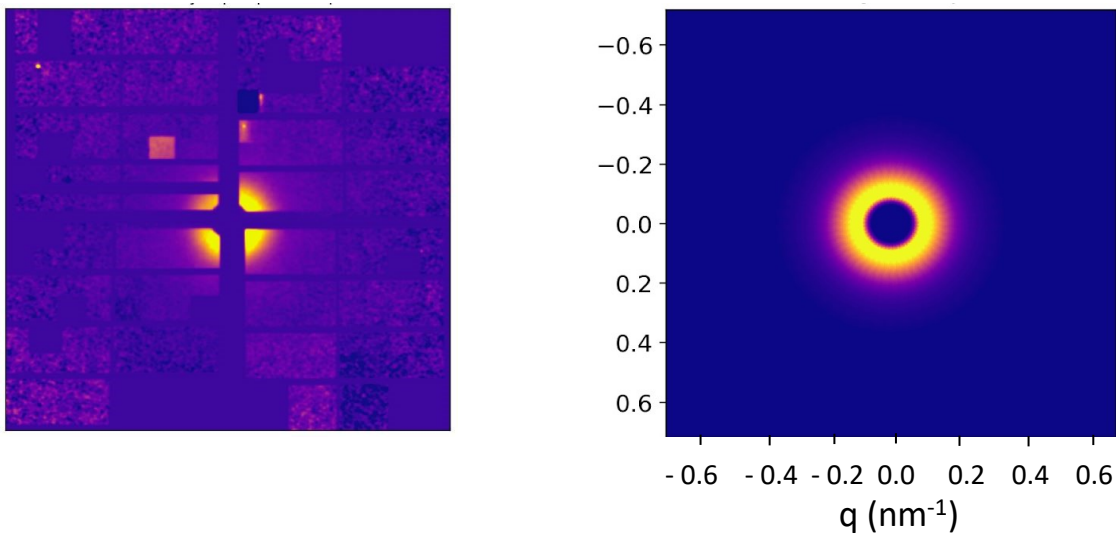
L. Müller, G. Grübel et al., Synchr. Rad. News, **26**, 30 (2013)

Experiments at SCS@EuXFEL

- Pump-probe experiment in transmission geometry
- TR-mSAXS at the Co L_3 edge (778 eV)
- A femtosecond 800 nm IR laser was used for pumping



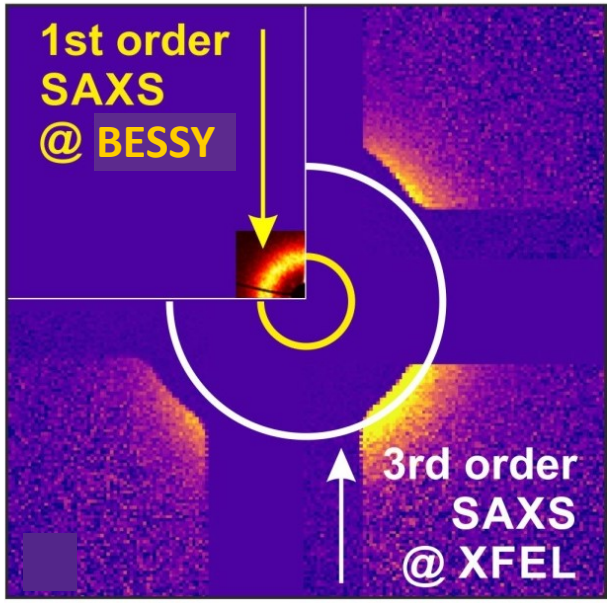
mSAXS data treatment



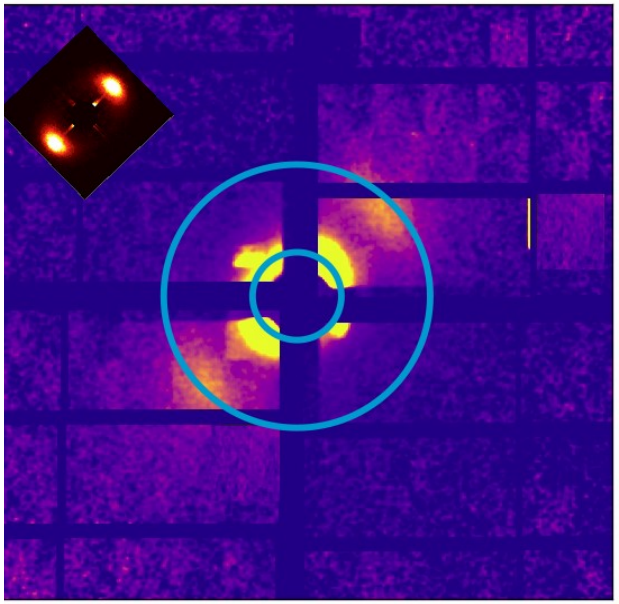
- Each frame of DSSC detector was first normalized to the signal of XGM monitor
- Then dark image was subtracted from each frame and averaging over thousands frames of the run was carried out
- Finally, the image was cleaned of dead pixels
- For labyrinth-like domains, the patterns were azimuthally averaged

Comparison of mSAXS data obtained in two SCS@EuXFEL beamtimes

L = 530 mm, Proposal # 2313
(PI I. Pronin)



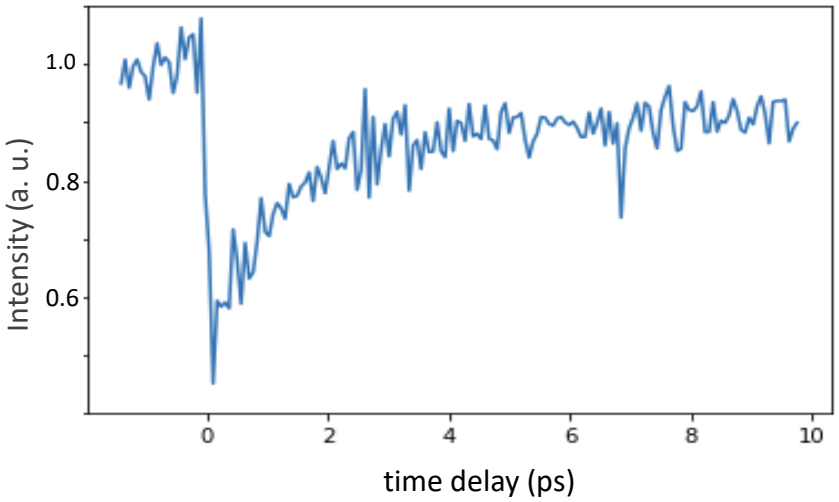
L = 2000 mm, Proposal #2212
(PI E. Jul)



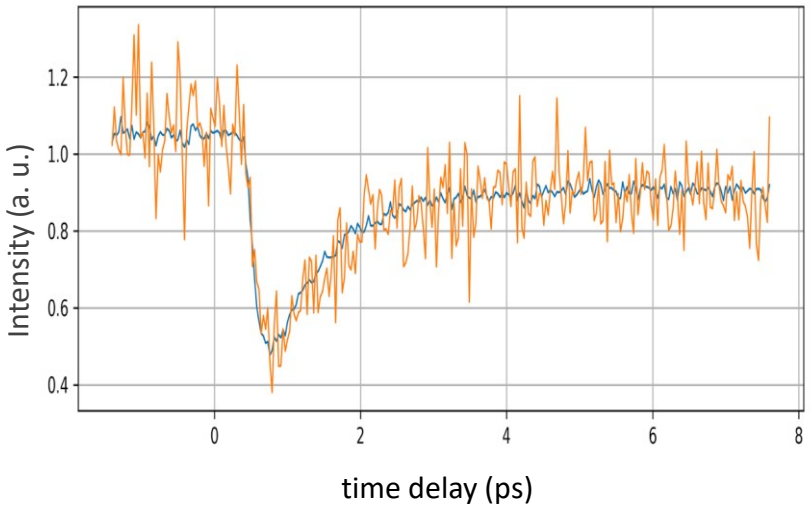
- The results of the experiments are in a good agreement
- The magnetic nature of scattering rings is confirmed by their sharp weakening at X-ray energy below the Co L_3 absorption edge

Comparison of TR-mSAXS data obtained in two SCS@EuXFEL beamtimes

3rd order scattering, Proposal #2313

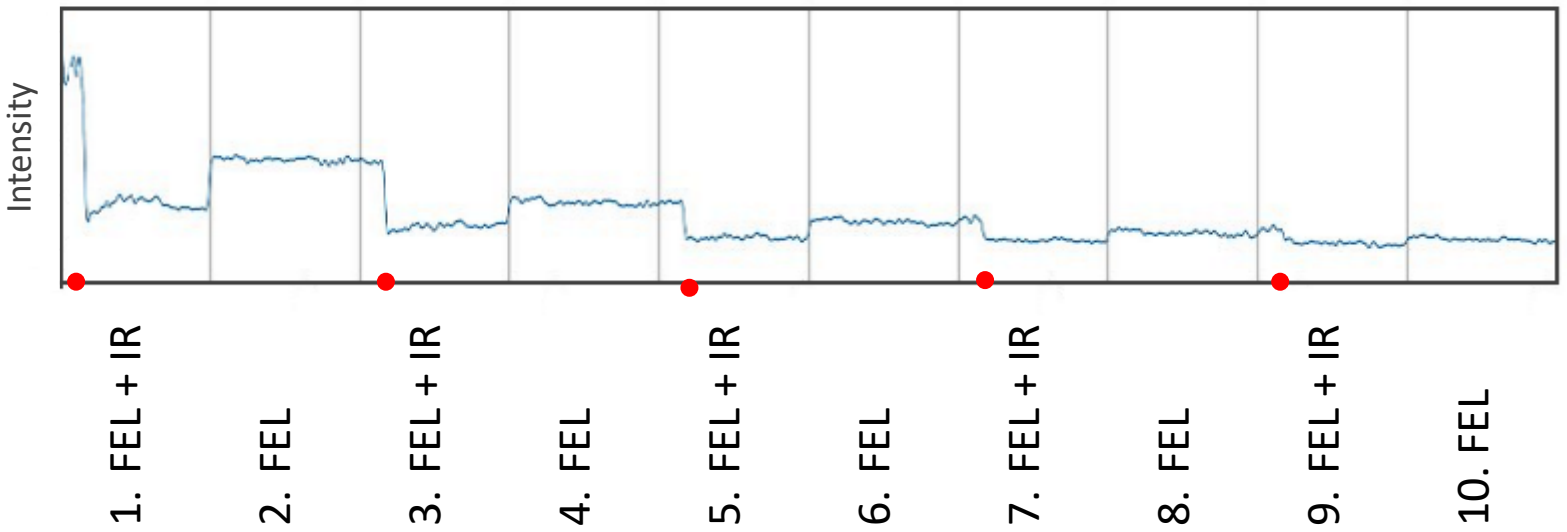


1st and 3rd order, Proposal #2212



- **Ultrafast drop of the magnetization has bee observed in both experiments with a *fs* time constant**
- **The drop is followed by a slow recovery on the *ps* time scale**

Frame-by-frame analysis of the intensity of magnetic scattering

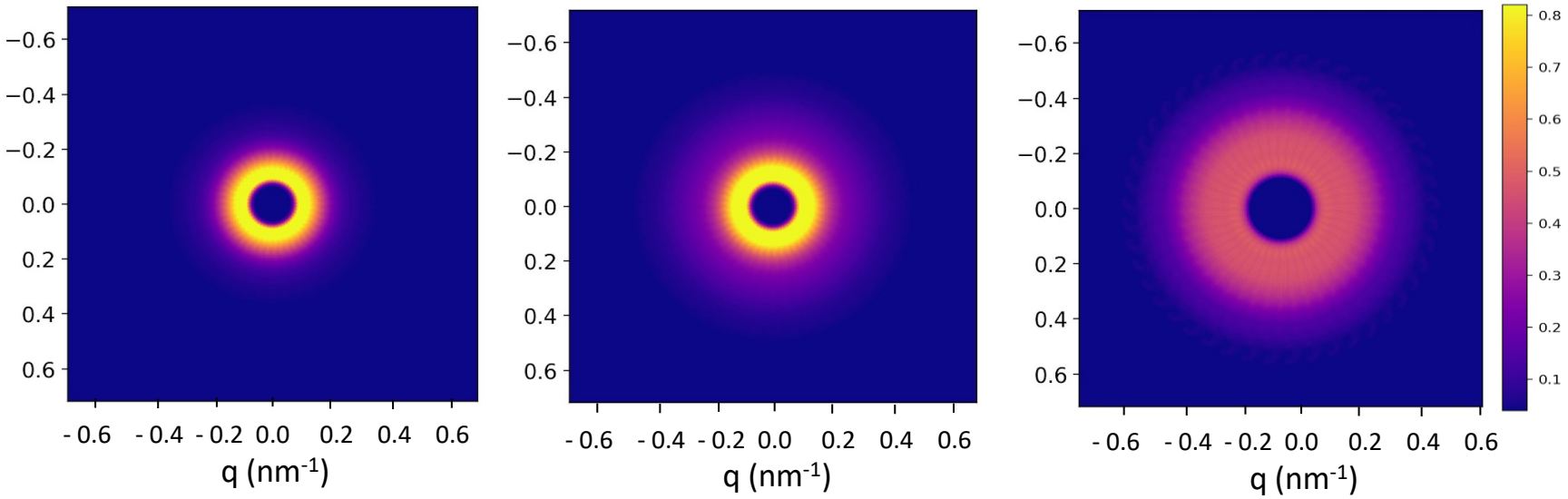


- **Temporal evolution of magnetic system during a train is illustrated by the dependence of the scattering intensity on the delay time and the frame number**
- **The frames are not equivalent. The strong scattering at the first frame loses its contrast in the subsequent frames.**

Low-q and high-q rings in TR-SAXS patterns taken after femtosecond IR excitation

IR fluence 5.6 mJ/cm^2, Delay 15 ps

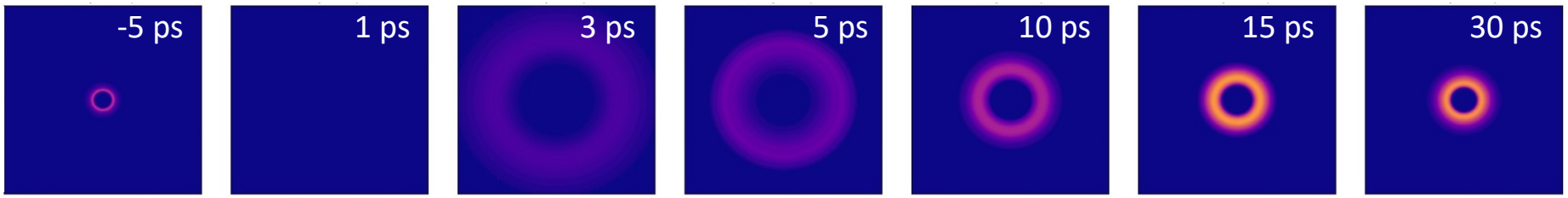
Unpumped **Pumped** **Pumped - Unpumped**



The difference pattern demonstrates an additional high-q scattering ring of red color, that appears in the pumped pattern in 15 ps after the IR pulse.

Time evolution of high-q scattering

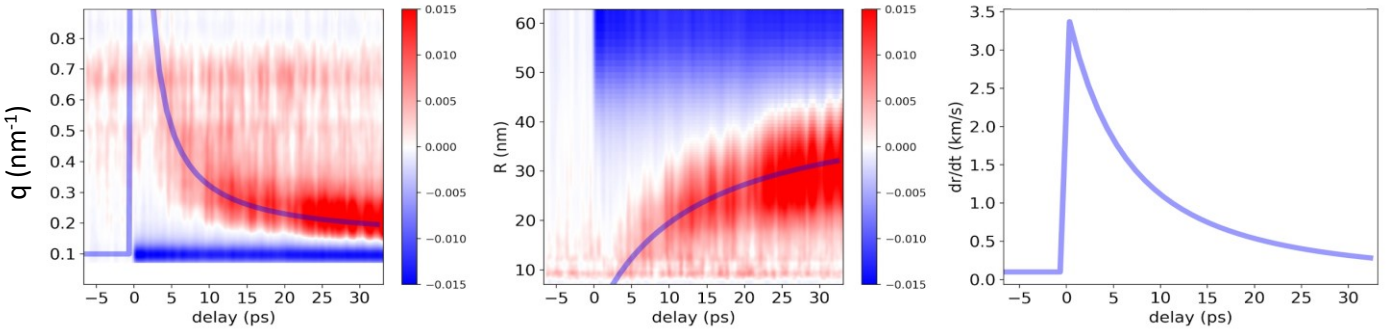
- The low-q ring is a fingerprint of the equilibrium magnetic structure in the pre-pump state
  - IR pulse strongly perturbs magnetic domains. In 1 ps after pumping the low-q ring is hardly visible



IR pump pulse

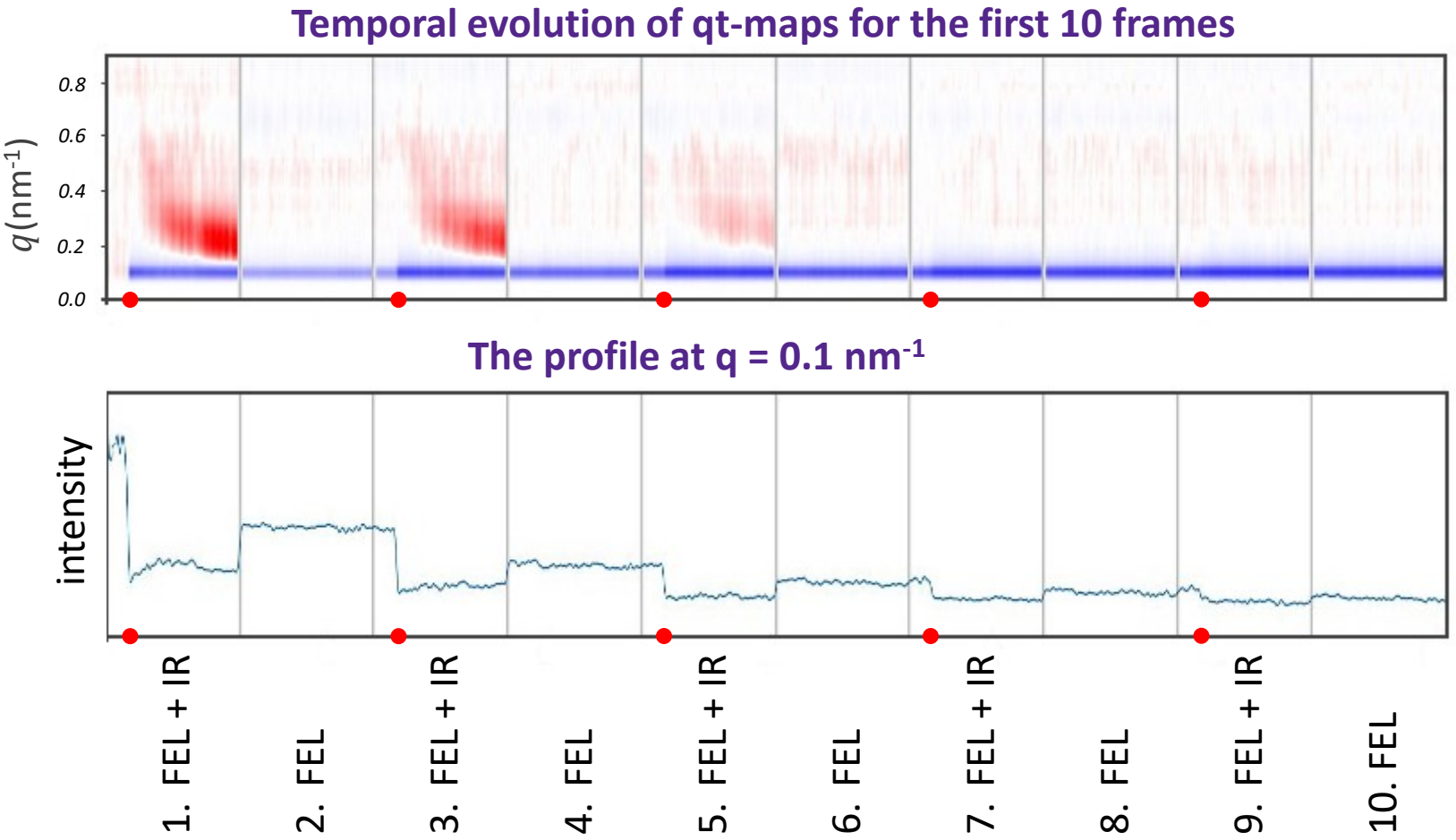
- The high-q scattering ring appears in 2 ps after pumping
- The ring shrinks over the delay time increase
- The ring width is proportional to its diameter

Temporal evolution of fluctuations responsible for the high-q scattering



- The appearance of high-q ring can be attributed to the IR induced nucleation of numerous small fluctuations
- The correlation length of the fluctuations can be estimated by the well-known formula: $R = 2\pi/q$
- The experimental dependence $q = q(t)$ is fitted by $q = 1.8 /(t - t_0) + 0.15$, where t (ps), q ($nm^{-1}$)
- The correlation length of fluctuations is growing nonlinearly with the delay
- The velocity of fluctuations expansion drops rapidly with increasing the delay time

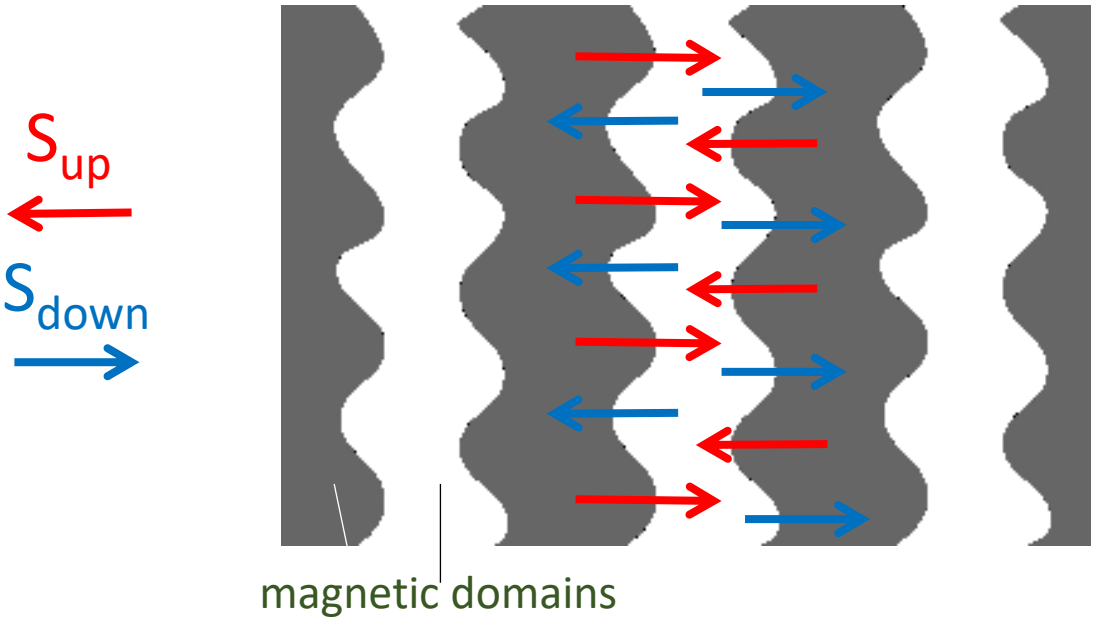
Frame-by-frame analysis of the intensity of transient scattering



Both panels exhibit similar dynamics, indicating a magnetic nature of high-q scattering

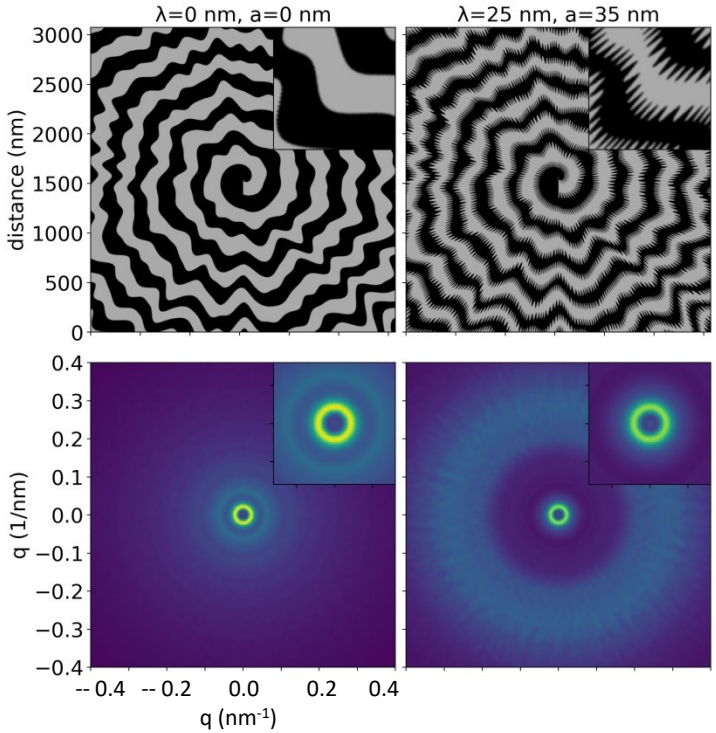
Simple model of the excited magnetic system

- At fluence of ~ 5 mJ/cm^2, a single IR pulse does not completely destroy the magnetic domains
- The low-q and high-q scattering rings exist in the ps delay range, so the model should take into account both initial domains and high frequency fluctuations
- Transient scattering is attributed to magnetic fluctuations caused by IR induced counter penetrating spin currents at the domain boundaries

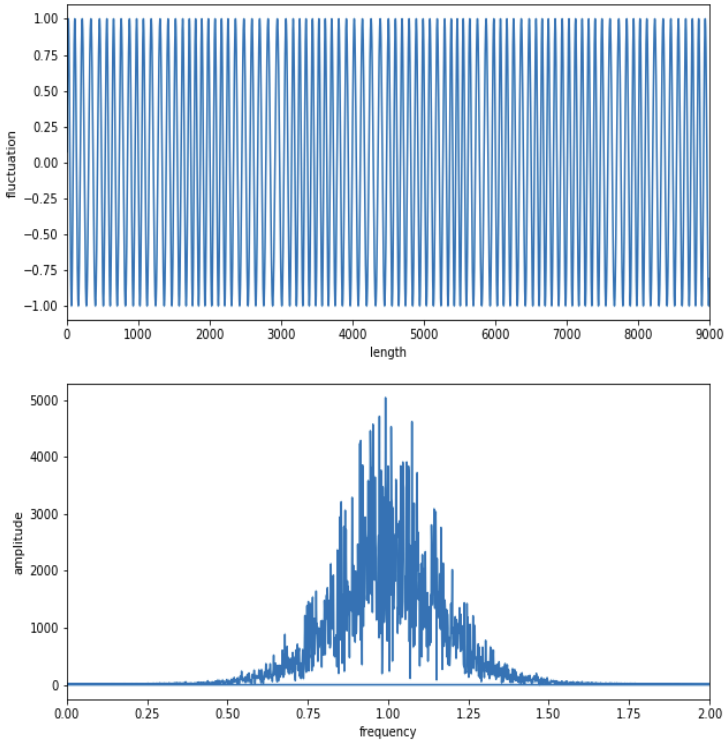


Simulation of mSAXS patterns (II)

Spiral domain patterns with and without high frequency fluctuations



Oscillations with a frequency floating ±10% around the center value were used to simulate wide scattering rings in SAXS.



Conclusions

- We have for the first time observed transient magnetic scattering in Co/Pt multilayers upon femtosecond IR excitation. The scattering occurs shortly after the pump on a picosecond time scale.
- A simplified model of magnetic structure that might cause the observed mSAXS patterns has been proposed. The transient high-q scattering is attributed to short-range magnetic fluctuations caused by IR induced counter penetrating spin currents at the domain boundaries.
- The establishment of the micro mechanism of the observed phenomenon requires further research.

Experimental team

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