Magnetic Excitations in Functional Oxide Thin Films: Beyond Single Magnons

A. Scientific Context of the Project

1. Description

Context

Magnons are the fundamental units of magnetic oscillations within magnetic materials and represent a change of 1 unit of spin angular momentum[1]. When a magnon propagates through a magnetic medium, no electrical charge transport is involved and hence no electrical losses take place. This is the key advantage of using magnons as information carriers. Prior research has centered around magnons with energies in the GHz range. However, another class of magnons with substantially higher energies, in the THz regime, has only begun to be explored[2]. Higher energy magnons arise in magnetic perovskites and could be uniquely manipulated by chemical substitution and substrate strain. The magnon frequency has an important impact on the performance of magnon-based devices because the larger the excitation frequency, the "faster" the magnons can be. This means that the use of high frequency magnons would provide a great opportunity for the design of ultrafast devices. Nevertheless, a fundamental understanding of the attainable magnon frequencies, their interaction with other degrees of freedom (such as lattice and orbit) and their dynamics is so far missing.

Purpose and Adequacy to Call

We have recently discovered a novel cascade of high energy-magnons, the multi-magnons[3], that propagate double, triple, quadruple and quintuple the energy and the spin angular momentum of conventional single-magnons known from neutron scattering[4]. While the presence of multi-magnons has previously been noted as a tail contributing to the asymmetric line shape of single magnons[5], the identification of distinct multi-magnon peaks is a novel observation (Fig. 1). This observation presents an opportunity to delve into the new physics underlying multi-magnons. We showed that the mechanisms to excite multi-magnons must rely on spin non-conserving interactions where the lattice and orbit degrees of freedom act as reservoirs of angular momentum enabling these higher multiple excitations[3]. Building upon this initial observation, our primary aim is to investigate the impact of spin non-conserving interactions on multi-magnons within thin films of magnetic perovskites. This research seeks to address two pivotal questions:

(1) Can we manipulate the energy and dispersion of multi-magnons and enhance their efficiency through tailored material design, affording control over spin non-conserving interactions?
(2) What are the dynamics governing multi-magnons on timescales ranging from tens of femtoseconds to few picoseconds. Do these dynamics correlate with the lattice dynamics of the magnetic thin film?

These questions will be addressed using multiscale characterization tools with a focus on X-ray spectroscopy at synchrotron facilities and X-ray free electron lasers (XFELs). This work will provide invaluable information about pathways to design materials with long-lived high-energy magnons. On the longer term, a microscopic understanding of the efficiency of angular momentum transfer between spin and lattice degrees of freedom brought by spin non-conserving interactions could inspire the design of new efficient spintronic and magnonic devices.

This proposal is seamlessly aligned with the thematic axes 2 of the "Institut de science des matériaux" as our project is dedicated to unraveling spin physics in thin films through synchrotron and XFEL experiments which is a fundamental challenge in materials science creating knowledge.

Scientific Approach

Magnetic perovskites (ABO_3 with A = La, Gd and B = Cr, Fe) form a class of materials that can host high-energy multi-magnons. We have identified two spin non-conserving interactions that are magnon-phonon[6] and spin-orbit interactions[7] serving as leading terms with the potential to tune the properties of multi-magnons. We will explore the new physics related to these magnetic excitations by growing tailored epitaxial thin films of transition-metal oxide magnetic perovskites. Precise control of strain in the thin films, achieved through appropriate substrate selection, will allow us to manipulate the lattice symmetry and hence the magnon-lattice interaction. Previous studies show that the magnons and

phonons are close in energetics which certifies an enhanced magnon-lattice interaction and the possibility to tune it through strain and thickness[8]. Moreover, by chemical substitution from La($4f^0$, no orbital momentum) to Gd($4f^7$, max. orbital momentum), we will manipulate the spin-orbit interaction.

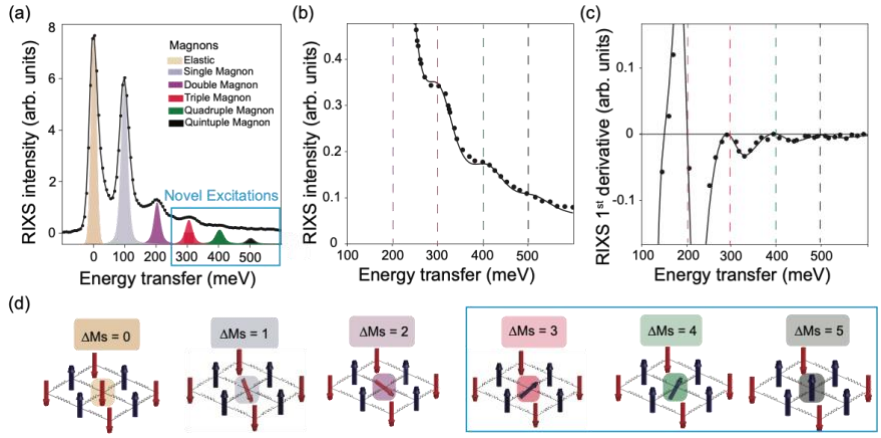

***Figure 1**: Fe 2p3d RIXS measurements.* (a) Single- and multi-magnons. The blue square highlights the novel excitations. (b) A zoom on the multi-magnons. (c) The 1st derivative of b. The peak positions can be clearly identified when the derivative is equal to zero. (d) A sketch illustrating the multi-magnons and the change of angular momentum involved. The *color coding corresponds to the peaks in a.*

To determine the multi-magnon energies, dispersion and lifetime, one can perform Resonant X-ray Inelastic Scattering (RIXS)[9]. While this technique is well known at synchrotrons, it is only recently that it has been applied for fs time-resolved studies at XFELs[10]. We will study the dynamics of the single- versus multi-magnons using a fs infrared laser pump and fs RIXS probe. The comparison between the response time of single- versus multi-magnons will shed light on the time scale at which such high-energy magnons involving exchange of angular momentum with other degrees of freedom can be controlled and the subsequent relaxation mechanisms that can take place after the excitation.

Risks and Mitigation Measures

The sample preparation will be performed at INSP using pulsed laser deposition (PLD). Note that our team has great expertise in using PLD for epitaxial thin film and heterostructure growth, especially in perovskite systems ($SrTiO_3$, $BaTiO_3$, $SrRuO_3$...) and strain tuning of physical properties[11, 12]. The project team members are embedded into a research "ecosystem" that will ensure a fast and systematic progress of the sample characterization. Transmission electron microscopy and x-ray diffraction will both be performed in-house to assess the structural properties of the thin films. These techniques can easily be complemented, if necessary, by other tools that provide additional information on strain and vacancies (Raman spectroscopy), on the stoichiometry (XPS) and surface morphology (AFM). Consequently, these first steps will have very little risk associated.

RIXS synchrotron measurements will be performed in France (ESRF – beamline ID32), UK (Diamond – beamline I20), and the USA (NSLS-II – beamline SIX) on 3 beamlines having 2 calls/year each. These beamlines have the best energy resolution which is required to measure the magnons. There is some risk of not getting all required beamtimes, however this risk is not high as the PI is an experienced synchrotron user and gets routinely ~10 weeks of beamtime/year. Her proposals are ranked among the best for example her past RIXS proposal submitted to NSLS-II (holds the best energy resolution in the world) scored 1.5 (1.0 being the best) where proposals graded up to 2.38 were granted beamtime.

The main uncertainty relies on the proposals' acceptance at XFELs facilities. However, we do not see this as a major risk since both our research groups are users of these facilities and have realized several XFELs experiments including the 1st time-resolved RIXS experiment on a solid system[13].

Bibliography (denotes our own publication)*

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2. Team Skills and Consistency

The PhD student will be supervised by Hebatalla Elnaggar (CNRS chargée recherche, IMPMC), Marcel Hennes (maître de conférence, INSP) and Amélie Juhin (CNRS directrice de recherche HDR, IMPMC). Our team has the specificity that all three members have great expertise in all aspects of the project: sample growth and characterization, X-rays spectroscopy at synchrotrons and XFELs, data analysis and theoretical interpretation. As a result, the PhD student will always find the help needed.

Nonetheless, each team member will be responsible for specific parts of the project. Marcel Hennes will be in charge of the sample growth and characterization. The sample will be grown by pulsed laser deposition in the deposition chamber of INSP on which Marcel Hennes has worked for several years. The sample will be characterized statically mostly at INSP by different techniques (X-ray diffraction and reflectivity, magnetic force microscopy, magneto-optical Kerr effect, etc...). Hebatalla Elnaggar will be responsible for the X-ray spectroscopy experiments done at the synchrotrons and XFELs. Amélie Juhin will formally supervise the PhD student and as an expert in theory of X-ray spectroscopy will oversee the progress with the theoretical calculations and the interpretation. We will interact with other members in both institutes as with Benjamin Lenz (IMPMC, Sorbonne University) who will provide theoretical support using cluster DMFT calculations to simulate the X-ray scattering experiments and Franck Vidal (INSP, Sorbonne University) who will provide support with the time resolved experiments.

B. Research plan with forecast schedule

The project will start with the production of the samples. In the first half of the first year, the PhD student will learn how to use the pulsed laser deposition and produce their first sample sets. In parallel, the PhD student will characterize statically those samples and will participate in beamtime already granted to familiarize themselves with large facilities know-about which will help them to write their own proposal. In the second half of the first year, synchrotron experiments will take place followed by data analysis and theoretical interpretation of the data.

In the second year, we will perform the fs time-resolved experiments at XFELs. Beamtimes may continue in the 3rd year on this specific project but they will not be the primary responsibility of the PhD student. We expect that the PhD work will result in 3 papers which are: (1) sample growth and characterization of the chemical and strain tuned magnetic perovskites, (2) synchrotron results regarding tuning the spin non-conserving interactions on the properties of multi-magnons, (3) XFEL results regarding the dynamics of single- versus multi-magnons.

Tasks	1st year		2nd year		3rd year	
	1st Term	2nd Term	1st Term	2nd Term	1st Term	2nd Term
Thin-film growth/characterization						
Proposal writing/preparation						
X-ray beamtimes						
Data analysis and theory						
Articles writing						
Thesis writing						