Tuning spin excitations in magnetic films by confinement

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Spin excitations of magnetic thin films are the founding element for magnetic devices in general. While spin dynamics have been extensively studied in bulk materials, the behaviour in mesoscopic films is less known due to experimental limitations. Here, we employ resonant inelastic X-ray scattering to investigate the spectrum of spin excitations in mesoscopic Fe films, from bulk-like films down to three unit cells. In bulk samples, we find isotropic, dispersive ferromagnons consistent with previous neutron scattering results for bulk single crystals. As the thickness is reduced, these ferromagnetic spin excitations renormalize to lower energies along the out-of-plane direction while retaining their dispersion in the in-plane direction. This thickness dependence is captured by simple Heisenberg model calculations accounting for the confinement in the out-of-plane direction through the loss of Fe bonds. Our findings highlight the effects of mesoscopic scaling on spin dynamics and identify thickness as a knob for fine tuning and controlling magnetic properties.

The never-ending demand for energy efficient devices, as well as recent advances in material deposition and nanoscale fabrication techniques, has set the basis for novel branches of electronics—magnonics. One of these is magnonics in which spin waves replace charge as the means of signal transport with substantial technological advantages, the most crucial being the reduction of heat dissipation. However, establishing spin-wave-based transport as a viable solution for the next generation of electronics poses several challenges that need to be resolved to enable logic operations (for example, effective spin wave signal generation, propagation, control and detection). At a fundamental level, there are still many outstanding questions concerning how magnetic properties and excitations evolve and emerge in low-dimensional systems, such as thin films. The interplay between confinement, dimensionality, interface/surface hybridization, strain and exchange bias can provide effective tuning of magnetism in such thin films, ultimately leading to exquisite control of spin dynamics. Understanding how such spin dynamics unfold in low-dimensional magnetic systems is thus an essential step towards gaining control of spin-wave-based transport phenomena.

From an experimental standpoint, the investigation of how spin dynamics evolve from a bulk-like system down to the nanoscale—the so-called mesoscale range, where at least one dimension is comparable to the electron mean free path—has been limited so far by the lack of suitable probes. While inelastic neutron scattering (INS) is an established technique to study spin dynamics in bulk magnetic materials, the low cross-section of neutrons makes difficult its application to mesoscaled systems. Alternatively, spin-polarized electron energy loss spectroscopy has been employed in the past to detect spin excitations in few-monolayer-thick films of 3d metals but provides insights mostly on the two-dimensional spin dynamics confined to the surface (due to the small electron escape depth), failing to access the properties of the whole film. Other techniques such as Brillouin light scattering, spin wave tomography and ferromagnetic resonance spectroscopy have been employed to study spin excitations but are limited to momentum transfer or to magnetostatic spin waves at long wavelength. Recently, resonant inelastic X-ray scattering (RIXS) has emerged as an alternative tool for the detection of spin excitations and their dispersion in magnetic systems. Owing to the large cross-section and RIXS resonant character, elementary excitations in mesoscaled materials can be efficiently studied.

Here, we use RIXS to investigate the spin dynamics in thin films of ferromagnetic Fe grown using molecular beam epitaxy. By mapping out the evolution of the spin excitations over almost two orders of magnitude of thickness across the mesoscale range, we establish confinement as an effective variable to manipulate the spin dynamics. For bulk-like Fe films, RIXS is used to detect ferromagnetic spin excitations and their isotropic three-dimensional dispersion relation. By progressively reducing the film thickness, we observe spin excitations surviving down to the ultra-thin limit. Furthermore, we uncover the evolution of the spin dispersion as it deviates from bulk behaviour and develops anisotropically between the in-plane and out-of-plane directions: renormalizing to lower energies along the [0,0,L] out-of-plane direction while being unchanged along the [H,0,0] in-plane direction. This anisotropic modification is captured by an isotropic Heisenberg-like model that predicts that the out-of-plane dispersion changes when the film thickness approaches the length scale of the Fe magnetic interaction. Our work therefore sets the basis for selectively gaining control of the spin excitation energy through confinement, enabling the design of optimized materials for future applications.

Film synthesis and experimental details

High-quality, crystalline films of Fe with varying thickness were prepared by molecular beam epitaxy. The Fe[001]-oriented films were grown epitaxially on MgO[001] with a 45° rotation around the [001] axis at a substrate temperature of 300°C. The thicknesses of the films, extracted from X-ray reflectivity (Supplementary Note 1B), were 105 unit cells (u.c.), 54 u.c., 29 u.c., 17 u.c., 6 u.c., and 3 u.c. (30 to 0.87 nm, respectively). All films retained a body centred...
The Fe saturation magnetic moment measured by SQUID magnetometry was ~2 Bohr magnetons ($\mu_B$) for all the films (Supplementary Note 1C). Furthermore, considering the relatively large lattice mismatch of ~3.8% for bulk $\alpha$-Fe (bcc, with lattice parameter $a = 2.87\, \text{Å}$) grown on top of bulk MgO (diagonal axis, 2.98 Å), we observed strain relaxation as we increased the thickness of the Fe films. In particular, the 29 u.c. film was fully relaxed, while the 3 u.c. film was fully strained to the substrate. This relaxation was evident in the evolution of the measured in-plane lattice parameters:

- $a_{29\text{u.c.}} = 2.89 \pm 0.01\, \text{Å}$,
- $a_{6\text{u.c.}} = 2.97 \pm 0.01\, \text{Å}$, and
- $a_{3\text{u.c.}} = 2.98 \pm 0.01\, \text{Å}$

(Supplementary Note 1D for the strain study).

The oxidation state of the films was measured using X-ray absorption spectroscopy (XAS). To achieve XAS with high sensitivity, we measured partial fluorescence yield using the RIXS spectrometer. The spectra in Fig. 1a display a broad main peak around $E_{\text{max}} \approx 708.4\, \text{eV}$ for all films, due to the transition from localized $2p_{3/2}$ core states to the delocalized $3d$ valence states, consistent with metallic Fe (refs. 38,39). The absence of additional peaks, characteristic of iron oxides (FeO, Fe$_2$O$_3$, and Fe$_3$O$_4$), demonstrates a negligible oxidation of Fe.

High-resolution RIXS experiments were performed at the SIX 2-ID beamline of the National Synchrotron Light Source II (NSLS-II) with an energy resolution of $\Delta E = 23\, \text{meV}$ (full-width at half-maximum) at the Fe L$_3$ edge; $\pi$-polarized light was used to minimize the elastic scattering signal. All the data were measured at 100 K. The films were aligned with both the surface normal [001] and the [100] axis lying in the scattering plane as depicted in Fig. 1b. The spectra were normalized to the integrated fluorescence to have comparable intensities for all films.

**Isotropic spin excitations in bulk-like Fe film**

We first show that RIXS probes the spin excitations in bulk-like Fe. We initially collected an incident energy dependence of the RIXS signal across the Fe L$_3$ edge on a bulk-like film. Figure 1c displays the RIXS map for the 105 u.c. film: the most apparent features of the spectra consist of a broad fluorescence line due to the incoherent $3d \rightarrow 2p$ emission and a low-energy ($<100\, \text{meV}$) Raman-like peak close to the elastic line. We achieve the best contrast between this Raman-like peak and the background by selecting an incident photon energy detuned by $-0.4\, \text{eV}$ below the maximum of the XAS.

**Fig. 1 | Fe L$_3$ XAS of Fe films versus thickness and representative RIXS measurement from Fe bulk-like film.**

a, Thickness dependence of the Fe L$_3$ XAS measured in partial fluorescence yield mode. The blue arrow and dashed line mark the incident photon energy used for the RIXS spectra. This energy is defined as $-0.4\, \text{eV}$ with respect to the Fe L$_3$ $E_{\text{max}}$ (dotted line). Top right corner, sketch of the bcc unit cell of Fe (ref. 50). b, Experimental configuration used for the RIXS measurements: the shaded scattering plane, containing the incoming and outgoing X-ray wave vectors $k_i$ and $k_{\text{out}}$, is perpendicular to [010]. The momentum transfer $q$ is instead parallel to [001]. c, Incident energy dependence of the RIXS signal across the Fe L$_3$ edge, for Fe 105 u.c., representative of a bulk-like sample. The blue arrow and dashed line mark the selected photon energy for the study of the spin excitations. d, RIXS spectrum at $E_i = 708.0\, \text{eV}$ and [0,0,0.23] for Fe 105 u.c. film.
To characterize the dispersion of the spin excitations in three dimensions, we recorded measurements for $q$ mainly along the $[H,0,0]$ in-plane direction. Due to kinematical constraints, $q$ cannot be transferred solely along $[H,0,0]$, but a small $L$ component will also be present ($L \approx 0.13$). In Fig. 3a–c and Supplementary Fig. 5 we exhibit the spectra at $[0,0,0.3]$ and $[0.29,0,0.13]$, which have degenerate energy and comparable line shape. This degeneracy further corroborates the assignment of the low-energy peaks to genuine $\alpha$-Fe spin excitations and demonstrates the three-dimensional isotropy of the spin excitations in the 54-u.c.-thick film.

**Evolution of spin excitations in thin films**

We now extend our data presentation to thinner films, using the same experimental conditions of the 54 u.c. film. The results are summarized in Fig. 3 for all the samples. We observe spin excitations down to the thinnest film. It is now important to separately discuss spectra for in-plane and out-of-plane momentum transfer. Along the out-of-plane direction $[0,0,L]$ (Fig. 3a–c), a clear peak in the $\sim 50$–$120$ meV range is present at all thicknesses down to 17 u.c. Remarkably, we uncover a gradual down renormalization of the spin excitations while decreasing the thickness. The spin excitation energy is the same for the 105 u.c. and 54 u.c. films (with bulk-like dispersion). For 29 u.c. and 17 u.c., however, the spin excitation peak deviates from bulk-like behaviour to lower energies with an overall $\sim 25\%$ reduction at $q=0.00.23$ and $[0.0,0.3]$ r.l.u. and an $\sim 15\%$ reduction at $q=0.0,0.32$ r.l.u. At $q=0.0,0.23$, where the spin excitation peak is well defined, we notice a slight decrease of the peak width for films of thicknesses between 105 u.c. and 17 u.c.,
indicating a lower damping (we limit our analysis to films thicker than 17 u.c.; see Supplementary Figs. 7 and 8 for details). At these thicknesses, a mesoscopic behaviour clearly emerges as a renormalization of the spin excitation dispersion.

For the 6 u.c. and 3 u.c. films, the spin excitation dispersion in the out-of-plane direction cannot be similarly estimated, as the peak is no longer well defined, and we find an ~2–3 times larger peak width. This broader peak width seems unrelated to spin excitations as they might be hidden underneath the elastic line (below 20–30 meV) or completely washed out. This peak is likely originated by residual particle–hole excitations dominating this energy range in the absence of spin excitations. Because of this, we do not further discuss the out-of-plane spin excitations for 6 u.c. and 3 u.c., and leave them out of the summary presented in Fig. 4a.

The in-plane RIXS spectra for the 54 u.c., 17 u.c. and 3 u.c. films at [0.29, 0, 0.13] are reported in Fig. 3d. No renormalization is observed for this direction; instead, a peak with similar energy position, intensity and width (refer also to Fig. 8b) is present for all film thicknesses (down to 3 u.c.). The position around ~100 meV matches the bulk-like dispersion of the spin excitations in α-Fe. Therefore, this result shows that bulk-like in-plane spin excitations persist even in a few layers of Fe.

In the field of spintronics, an important property of the spin excitations is their lifetime or damping. In all our data, the spin excitations are not resolution limited and damped, with the damping increasing at higher momentum transfer (Supplementary Note 2 for details). This agrees with the common knowledge that high-$$q$$ spin excitations couple more with the Stoner continuum at high energy, as observed by INS$^{19,44}$. As previously discussed, from our bulk-like sample we extract damping values similar to INS data (Supplementary Fig. 8 and the literature$^{19,20}$). As for the energy, the in-plane damping of the spin excitations appears unaffected by film thickness as displayed in Fig. 3d and Supplementary Fig. 8. Along [0,0,L], instead, we observe a decrease of the peak width of the spin excitations as the thickness is decreased. The reason behind this decrease can be twofold: the lower energy of the spin excitations leads to a reduced interaction with the continuum, or the electronic continuum itself becomes narrower, quenching the coupling. The latter is unlikely as the Stoner continuum is not altered by thickness (at least above the 29 u.c. film), lending support to the conclusion that the softening of the spin excitations 'trivially' decreases the damping of the spin flips. Such reduced damping or increased lifetime of the spin excitations along [0,0,L] as the thickness is decreased may result in a positive impact for applications demanding tuning of the lifetime of the spin excitations.

To interpret our results on the evolution of the energy of the spin excitations, we point out that the renormalization of the spin excitations in the out-of-plane direction cannot be ascribed to strain. In the 3 u.c. film, the strain along the out-of-plane direction is smaller (~2%) than along the in-plane direction (~3.8%), for which the measured spin excitations are unaffected by thickness. Considering that the larger in-plane strain is unable to affect the energy of the spin excitations (Fig. 3d), we can exclude the idea that the softening of the out-of-plane spin excitations as a function of thickness is primarily caused by strain. Similar arguments can in fact be extended to all other thicker films, where additionally the strain effect is less relevant. We can also rule out effects such as substrate hybridization and changes in magnetic moment. The hybridization of the Fe bands with the MgO substrate is unlikely to explain the spin excitation renormalization observed in the relatively thick 29 u.c. film. Moreover, MgO is an insulator and lacks bands close to the Fermi level where the hybridization should affect the spin properties, making this scenario improbable. Our SQUID measurements of the Fe saturation magnetic moment confirm its independence on the thickness, hence eliminating this possibility. Finally, we can disregard surface effects because the RIXS penetration depth is larger.
actions at the Fe model systems46,47, effectively modelling the itinerant
was previously used in theoretical works to describe the spin inter-
mines the effective length scale of the interaction. This formulation

\[ J_{ij} = J_0 \left( \frac{a}{R_{ij}} \right)^p, \]

where \( R_{ij} \) is the distance between the two sites, \( a \) is the lattice con-
tant, \( J_0 \) sets the strength of the exchange interaction and \( p \) deter-
mines the effective length scale of the interaction. This formulation
was previously used in theoretical works to describe the spin inter-
actions in Fe model systems46,47, effectively modelling the itinerant
electron behaviour46.

Figures 4a,b show as solid lines the out-of-plane and in-plane
spin excitation dispersions obtained from evaluating the model with
a short-range \( J_{ij} \) interaction (\( p = 8 \); see ref. 48 and Supplementary
Note 3). Interestingly, this simple, isotropic exchange model is able
to qualitatively reproduce the anisotropic thickness-dependent dis-
persion observed in our experiments (solid markers in Fig. 4a,b).
In particular, the out-of-plane renormalization is due to the reduc-
tion of the near-neighbour spins at the film surfaces. The isotropic
exchange interaction together with the loss of Fe magnetic inter-
actions at the Fe film surfaces produces an apparent anisotropic reduction of \( J \); the calculations reveal that this effect impacts the
energies of the out-of-plane modes far more than the in-plane ones.
By contrast, when using a long-range \( J_{ij} \) interaction \( (p = 4) \), we find
that the in-plane modes scale down similarly to the out-of-plane ones (see Supplementary Note 3 and Supplementary Fig. 11), in
contradiction with the experimental trends.

While this simple model misses a complete one-to-one corre-
spendence with the data, it correctly predicts the qualitative behav-
ior as a function of thickness and reciprocal space direction. The
outcome of this theory is thus twofold. First, it proves that the
confinement effect, associated with the geometrical loss of Fe spins
due to the limited thickness, causes a renormalization of the spin
excitations. Second, it supports the short-range nature of the mag-
netic interactions in Fe, responsible for the anisotropic spin excita-
tion evolution between out-of-plane and in-plane directions. Our
direct measurement of the length scale of spin interactions \( J_{ij} \) is
consistent with INS experiments that fitted the dispersion using a
nearest-neighbour Heisenberg model19,20.

**Perspective**

These results demonstrate that the confinement effect can be
used as a knob to tune the spin excitations in mesoscaled systems.
This tuning can be parameterized by noting that the calculated
dispersion curves for each thickness in Fig. 4a can be scaled by a
\( q \)-independent factor, which we call the renormalization factor in
Fig. 4c. Such a renormalization factor follows a power law behaviour
as a function of thickness. By knowing the exchange coupling of the
bulk material, this factor can be used to predict the thickness depen-
dence of the spin excitations in cubic systems. Being based only on
geometrical arguments, the scaled dispersion can be extrapolated to
low-energy spin excitations (on the order of gigahertz), which are
more commonly employed in magnonics, extending the applicabil-
ity range of our investigation. More generally, in applications where
magnetic media are used to propagate the signal, our study dem-
onstrates the use of confinement to select the appropriate energy of
spin excitations and their lifetimes to be employed for specific pur-
poses. Our approach is analogous to the modification of refractive
index in optics, where the film thickness acts to tune the ‘magnetic’
refractive index of the medium in magnonics.

In summary, we combine molecular beam epitaxy synthesis and
RIXS experiments to unveil the thickness dependence of the spin
excitations in Fe films covering almost two orders of magnitude in
thickness. In bulk-like films, RIXS is used to measure ferromag-
etic spin excitations consistently with INS. From this demonstra-
tion, we extend our study to mesoscopic thin films and uncover a
remarkable renormalization of the out-of-plane spin excitations as
the film thickness is decreased down to a few unit cells. By contrast,
in-plane spin excitations are thickness independent, preserving a
bulk dispersion down to 3 u.c. These trends can be explained using a
Heisenberg-like formalism based on a short-range exchange isotro-
pic interaction and accounting for confinement through the loss of
Fe spin interactions at the film surface. Using this theory, we are able
to predict the dependence of the spin excitations as a function of
thickness. Our predictions can be applied in other systems to real-
ize tunable spin excitations for which confinement is the dominant
effect. From an application perspective, our study shows a method
to tailor the spin excitation energies and their lifetimes with impact
on spintronics and magnonics by providing an effective way to tune

![Fig. 4](image_url)
the refractive index in magnetic materials, equivalent to manipulating the dielectric function in photonics.

Online content
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Methods
Sample preparation. High-quality, crystalline films of Fe with varying thickness were prepared by molecular beam epitaxy. The Fe [001](110)-oriented films were grown epitaxially on MgO[001](100) substrates with a 45° rotation around the [001] axis at a substrate temperature of 300 °C. During the growth, the films were monitored using in situ reflection high-energy electron diffraction to assess the crystalline quality (see Supplementary Note 1 for details on the growth and characterization of the films). An ~5 nm protective layer of MgO was deposited to avoid oxidation. More details of the sample growth and characterization are reported in Supplementary Note 1.

XAS and RIXS measurements. XAS was measured at the SIX 2-ID beamline of NSLS-II with a resolution of ~50 meV. XAS at the Fe L3 edge (~707 eV) was measured in partial fluorescence yield integrated over an energy window of 670–720 eV and acquired using the RIXS spectrometer.

High-resolution RIXS experiments were performed at the SIX 2-ID beamline of NSLS-II using the Centurion RIXS spectrometer. The combined energy resolution at the Fe L3 edge (~708 eV) was $\Delta E = 23$ meV (full-width at half-maximum) throughout the experiment; π-polarized light was used to reduce the elastic scattering signal.

All XAS and RIXS data were measured at 100 K.

For the detailed analysis of the RIXS data, see Supplementary Note 2.

Calculations. Details of the calculations are reported in Supplementary Note 3.

Data availability
Data that support the findings of this study are available upon reasonable request from the corresponding authors.

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Author contributions
V.B. conceived the project. J.P., S.L., J.L., Y.G., A.B., I.J. and V.B. performed the RIXS experiments. S.L., C.H.A. and F.J.W. prepared the Fe/MgO thin films and characterized them using SQUID and X-ray reflectometry. V.B. analysed and interpreted the data, with the help of J.P.; K.G. performed the theory calculations. J.P. and V.B. wrote the manuscript with input from all the authors.

Competing interests
The authors declare no competing interests.

Additional information
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