SPIN SPIRALS IN MULTIFERROIC BISMUTH FERRITE AND AT METAL SURFACE: FROM FULLY FIRST PRINCIPLES

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EXECUTIVE SUMMARY

This work explores the long-ranged noncollinear magnetic textures, called spin spirals, in the multiferroic material $BiFeO_3$, as well as the $BiFeO_3$/metal interface. These spin spirals are of prime importance for technological applications owing to their potential for spintronics and low-power magnetoelectric devices. The presence of spin spirals is induced by the Dzyaloshinskii–Moriya interaction (DMI), which originates from spin-orbit coupling. This research investigates the magnetic interaction, and especially the DMI, in multiferroic materials and the interface between multiferroics and metals. With the computational resources of Blue Waters, the research team carried out fully first-principles or *ab initio* calculations in $BiFeO_3$ to reveal the stability of spin cycloids (the precession of spins along a propagation direction with a perpendicular rotational axis) under various strain conditions and to obtain the accurate magnetic interaction parameters that will be used to construct an effective Hamiltonian for combining with Monte–Carlo simulations to study finite-temperature properties.

RESEARCH CHALLENGE

$BiFeO_3$ (BFO) is a multiferroic magnetoelectric material at room temperature. This means that a magnetic field can change its polarization and an electric field can change its magnetization. This property is very rare in nature, which explains the strong interest in BFO and why it is called the "holy grail of multiferroic physics." BFO is also a noncollinear antiferromagnetic (AFM) material, exhibiting a magnetic cycloid at room temperature. Although this spin spiral is well characterized experimentally, several questions remain, such as the stabilization mechanism(s) of the different spin spirals and their interplay with structural distortion.

For instance, the type of cycloid in bulk BFO in the R3c phase is well characterized as being propagated along one of the [1–10] directions. However, a few recent experimental studies proposed that a type-II cycloid with a propagation direction along [11–2] may be favored in BFO thin films that are moderately strained, but the measurements could also be explained as a mixture of different type-I cycloids. Further, when BFO is deposited on a substrate with a large compressive strain (more than 4.5% in magnitude), it undergoes a phase transition toward the so-called super tetragonal phase, which is of great interest because of its very large polarization. This phase may host exotic noncollinear magnetic structure.

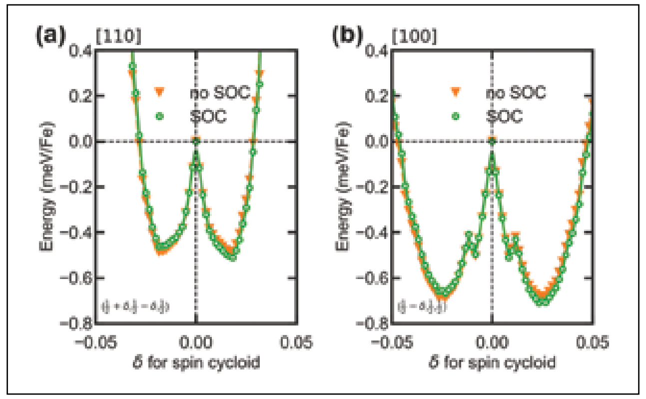

Figure 1: Spin spiral energy dispersion curves near the G-type antiferromagnetic state of BFO in the P4mm or super-tetragonal phase. (a) Cycloid propagation along the [1–10] direction; (b) cycloid propagation along the [100] direction.

In addition, one of the challenges in spintronics is to understand the interfacial DMI in sputtered films, which are commonly used in industry. In that case, the difficulty is to model the polycrystalline of the sample. Experimentally, the DMI can only be measured with a few techniques such as bubble expansion or via Brillouin light scattering (BLS). In the latter, only the averaged DMI over the entire sample can be measured. It is therefore of prime importance to understand the origin of the different contributions to the DMI. To that end, the research team has explored the magnetic exchange interaction and the DMI in $Fe_{1-x}Co_x$/Pt in collaboration with experimentalists.

METHODS & CODES

The research team used first-principles calculations based on density functional theory (DFT) in order to study the magnetic spin spirals in BFO. They applied the full potential linearized augmented plane wave (FLAPW) method [1,2] as implemented in the FLEUR code (www.flapw.de). This method is one of the most accurate implementations within DFT, owing to the self-consistent treatment of all the involved electrons (core and valence) and the precise and "natural" expansion of the wave functions (*i.e.*, in spherical harmonics around the nuclei and in plane waves in the interstitial region). The choice of an accurate all-electron full-potential code was motivated by the fact that this project requires the treatment of noncollinear magnetism and spin-orbit coupling on the same footing as well as a highly accurate description of eigenfunctions and eigenenergies (leading to the need for an extremely good basis set). Therefore, the simulations of complex magnetic states in the presence of spin orbit coupling (SOC) represent a cutting-edge problem in supercomputing. FLEUR is to date the only existing code containing all of the above-mentioned features and that is, therefore, capable of calculating SOC effects in noncollinear systems fully *ab initio*.

Further, the team computed the total energies at various reciprocal-space points based on the generalized Bloch's theorem. The ground-state magnetic configuration was determined and compared with experiments. Remarkably, this approach allowed the team to extract the exchange and DMI interaction coefficients by considering neighboring interactions over a long range.

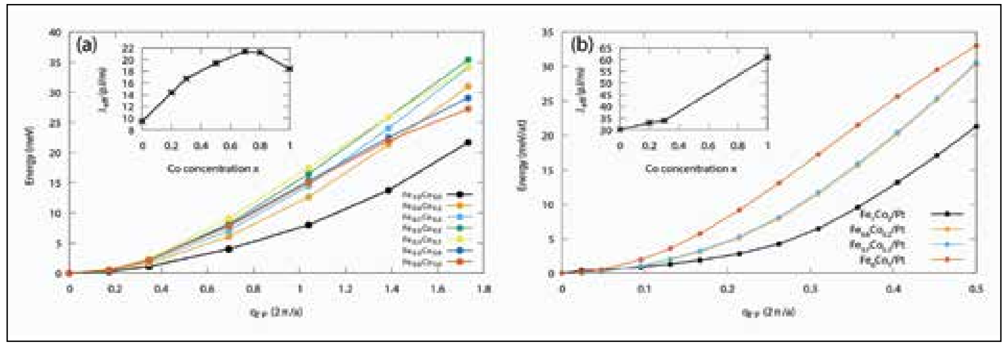

Figure 2: Comparison between the magnetic exchange interaction of $Fe_{1-x}Co_x$/Pt without the Pt substrate (panel a) and with the Pt substrate (panel b). The magnetic exchange interaction of an ultrathin $Fe_{1-x}Co_x$ depends significantly on the substrate hybridization.

RESULTS & IMPACT

The research team explored for the first time different spin spirals in BFO at an *ab initio* level. The study revealed that the competition between the first- and second-nearest neighbor exchange interactions leads to a flat energy landscape near the G-type antiferromagnetic state, slightly favoring a spin cycloid propagating in the [1–10] direction. Its stability is enhanced by the polarization but suppressed by the antiphase oxygen octahedral tiltings. Surprisingly, a similar trend is found for the DMI energy, which further lowers the energy of the cycloidal state and reduces the cycloidal period. The predicted period is in good agreement with experiment.

For the super-tetragonal phase of BFO, the calculated energy dispersion in the vicinity of the G–AFM state suggests that cycloid formation is strongly favored. As illustrated in Fig. 1, the cycloid is likely to propagate along the [100] direction, as it has a noticeably lower energy than that of propagation along [1–10]. To our knowledge, this is the first work that predicts the existence and type of cycloid in T–BFO, which can then be verified by future experiment and may lead to promising spintronic applications.

To shed light on the open issue of slightly strained BFO, the research team performed a systematic study in the Cc phase with strain from compressive (–2%) to tensile (+2%). Comparing the energetics of cycloids with [1–10] and [11–2] propagation directions, the researchers found that type-II cycloid propagating in the [11–2] direction is not favored over type-I cycloid propagating in the [1–10] direction, implying that the experimentally observed type-2 cycloid is likely to be a mixture of two type-1 domains.

In parallel, the team explored the magnetic exchange and the DMI in $Fe_{1-x}Co_x$/Pt as shown in Fig. 2. Counterintuitively, the magnetic exchange interaction changes dramatically as a function of the concentration. The magnetic exchange shows a maximum for Co-rich $Fe_{1-x}Co_x$/Pt. The value of the magnetic exchange is, however, much smaller than the measurements realized by BLS on $Fe_{60}Co_{20}B_{20}$/Pt (30 pJ/m). These findings suggest that the hybridization of the $Fe_{1-x}Co_x$ with the Pt plays a major role in both the magnetic exchange and the DMI, as shown in Fig. 2(b). Taking into account the presence of the Pt substrate increases the exchange at 30 pJ/m, in agreement with the experimental finding. The calculations of the DMI are now under consideration. In the next period, the team will be able to study the effect of an electric field on $Fe_{1-x}Co_x$/Pt created by a ferroelectric layer owing to proximity effect.

WHY BLUE WATERS

A challenge in this study was to obtain the full magnetic phase diagram of the systems with strain, temperature, and electric field. Owing to the very long period of the spin spiral in BFO (approximately 60 nm), an extremely dense k-grid is necessary to resolve the q-point corresponding to the ground-state period. Given the superior stability of the FLEUR code with a large number of cores (beyond 10,000), the study has greatly benefited from using the Blue Waters supercomputer.

PUBLICATIONS & DATA SETS

X. Changsong, B. Xu, B. Dupé, and L. Bellaiche, "Magnetic interactions in $BiFeO_3$: A first-principles study," *Phys. Rev. B*, vol. 99, p. 104420, 2019.

B. Xu, B. Dupé, and L. Bellaiche, "Exploring the origin of non-collinear magnetism in $BiFeO_3$," in preparation, 2019.

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J. Seidel *et al.*, "Exchange splitting of a hybrid surface state and ferromagnetic order in a 2D surface alloy," submitted, 2019.