Noncollinear magnetic structure in tetragonal Heusler materials

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Magnetic Heuslers with multiple magnetic sublattices provide a perfect platform for the design of anisotropic and acentric room-temperature magnets with flexible magnetic configurations. Based on these principles we have designed several new Heusler materials with extraordinary properties. Here, we show that the acentric Heusler materials Mn_2RhSn and Mn-Pt-Sn show noncollinear magnetic structures. With the help of Lorentz transmission electron microscopy and topological Hall effect, we have demonstrated the existence of helical and skyrmion phases in these materials.

Efficient control of magnetic degrees of freedom has led to a broad range of applications in the rapidly developing field of spintronics. So far, magnetic materials with collinear spin alignments have been dominated in most of the practical applications. It was recently proposed that the back and forth motion of an atomically engineered chiral domain wall (noncollinear spin alignment) can provide a basis for novel high density solid-state storage memory devices - Racetrack Memories [1]. However, the threshold current to move such domain walls is much higher than that expected theoretically. In contrast, much lower threshold currents have been found for the motion of recently discovered *skyrmions*, which are topologically stable vortex-like objects with a twisted spin configuration found in acentric-type magnets [2-4]. However, , skyrmions are mostly found only at low temperatures and in significant magnetic fields [2-4].

Heusler compounds, X_2YZ (where X, Y are transition metals and Z is a main-group element), are well known for their multifunctional properties. The magnetic Heuslers with multiple magnetic sublattices are excellent candidates to provide a perfect platform for the design of anisotropic and acentric room-temperature magnets [5]. In particular, Mn_2YZ-based tetragonal Heusler materials are perfect candidates for finding skyrmions. In a very recent work, we have experimentally shown that the acentric magnet Mn_2RhSn exhibits a noncollinear spin structure in its tetragonal crystal structure [6]. Based on theoretical predictions, this compound could show free skyrmion excitations or field-driven skyrmion lattices [6]. Owing to the polycrystalline nature, we failed to characterize this material experimentally by a skyrmion perspective.

We have prepared thin films of Mn_2RhSn by dc-magnetron sputtering to find out skyrmions in this system. Like the bulk compound, the thin films also show a tetragonal crystal structure with a Curie temperature (T_C) of around 300 K. It can be mentioned here that the materials showing skyrmions exhibit an extra component to the Hall effect called *topological* Hall effect [7,8]. Therefore, we have measured Hall effect in the Mn_2RhSn thin films to characterize the skyrmion phase.

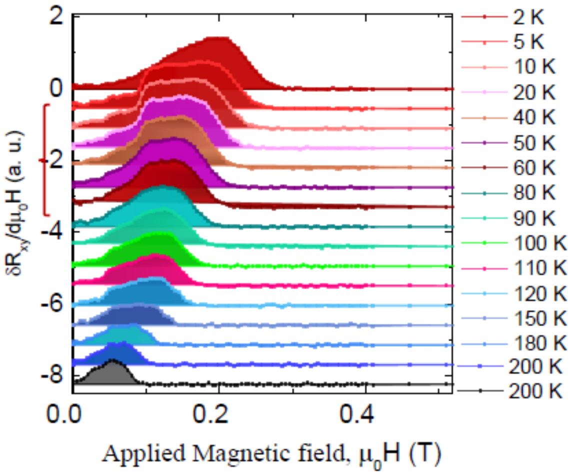

FIG.1 First derivative of the virgin curve of Hall resistance, R_{xy}, versus magnetic field measured at different temperatures. The shaded area corresponds to the topological Hall contribution.

From the measurements showing Hall resistance, R_{xy}, versus magnetic field, we observed that a hump exists in the virgin curve. Therefore, we take the first derivative R_{xy} to clearly see the extra component of the Hall measurement as shown in Fig.1. The deviation of the Hall resistance from the electronic and anomalous components corresponds to the topological Hall effect, which arises when the electron passes through the skyrmion spin texture. The shaded areas at different temperatures in Fig. 1 correspond to the contribution from the topological Hall effect. This indicates that there is indeed a special spin texture in the Mn_2RhSn thin films.

We found that in case of Mn_2YZ Heusler compounds with a nonmagnetic heavy element Y , the magnetic ground state can be noncollinear owing to the competition between two different AFM interactions among different magnetic sublattices. Therefore, to further tune the magnetic anisotropy we have taken another nonmagnetic heavy element Pt to prepare the Heusler compound $Mn_{1.4}PtSn$. The magnetic measurements show that $Mn_{1.4}PtSn$ exhibits a T_C around 400 K followed by a spin-reorientation transition around 160 K. The ac-susceptibility measurement displays a dip between 130 K and 160 K. The dip in the ac-susceptibility curve is typical found in the skyrmion phase [9,10]. Therefore, we have performed Lorentz transmission electron microscopy (TEM) at various temperatures and magnetic fields.

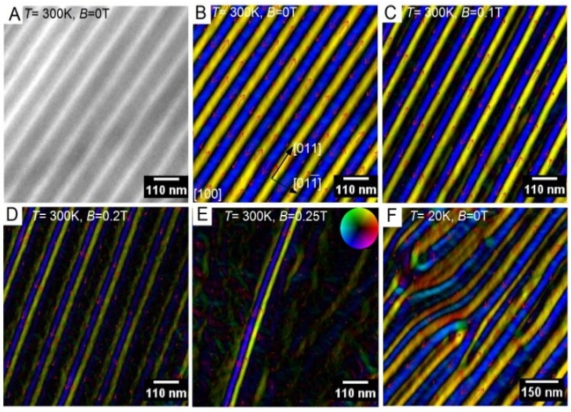

FIG. 2. A. Room temperature Lorentz TEM image (under-focused image) showing stripe modulations of the magnetic structure. (B-E) Helimagnetic stripe structures at various magnetic fields indicated in the figures. (F) Complex stripe magnetic structure at 20 K.

The Lorentz TEM images taken in zero magnetic field displays a modulation of contrast in one direction (Fig. 2A). These magnetic stripes are consistent with a helimagnetic structure, where the helix has a period of 110 nm that propagates along the [011] direction. The transport-of-intensity equation analysis of the Lorentz TEM images taken at 300 K and various magnetic fields are shown in Fig. 2B-2E. The stripes with two different colors correspond to opposite directions of the in-plane magnetization as shown by the color wheel. With application of magnetic fields, the helimagnetic structures slowly develop into a soliton lattice that disappears for 0.25 T. At 20 K, the stripes are irregular with varying periodicities and contain large number of dislocations (Fig. 2F).

Since the ac-susceptibility measurements show the signature of a skyrmion phase around 130–160 K, we have focused the Lorentz TEM measurements around this temperature. Figure 3 shows the Lorentz TEM image taken at 140 K in a zero magnetic field. As seen from the figure the helical magnetic stripes modify to a bubble-like magnetic structure with a size of approximately 50 nm. These magnetic bubble-like structures with rotating magnetic states correspond to skyrmions. Most importantly, these skyrmions exist at a zero applied field. The mechanism of formation of skyrmions in the Heusler class of materials can be distinguished from previous material classes. In the present case, the skyrmions have been formed owing to the presence of multiple magnetic sublattices. This provides a novel route to the formation and stabilization of new arrangements of skyrmions even in zero magnetic fields. Because of the wide composition range of Heuslers we anticipate that their magnetic properties can be tuned to achieve spontaneous skyrmion states even at room temperature.

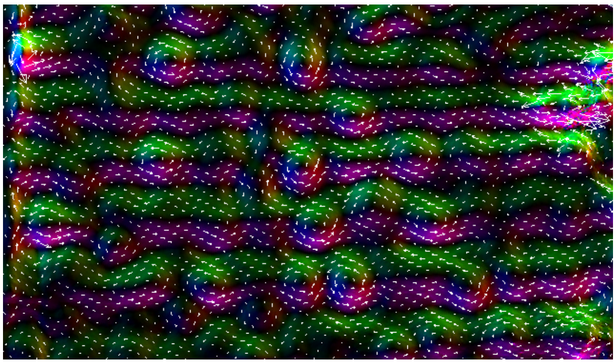

FIG. 3. Lorentz TEM image around 140 K showing modification of helical magnetic structure to skyrmion in $Mn_{1.4}PtSn$.

Since the motion of skyrmions can be accompanied in much lower threshold current density, their implementation in racetrack is one of the most important future research plans. A material with skyrmion phase will be deposited with Pt underlayer. The role of Pt is to generate a spin current that applies a torque on the spins in a skyrmion layer. This will help to move the skyrmions even at lower current densities and higher velocities. Another aim of using Pt in skyrmion-based racetrack is to utilize the extra DMI at the interface of the skyrmion and Pt for stabilizing the skyrmion in a zero magnetic field, which is important from application prospective.

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