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Observation of ice-rule violation and monopole dynamics via edge nucleation of domain walls in artificial spin ice lattice

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In a patterned Co honeycomb spin ice structure, we show that violation in the ice-rule or magnetic monopoles, can be observed during a magnetization reversal process in 430 Oe $\le H \le$ 760 Oe magnetic field (H) range. The monopoles are shown to originate from the nucleation of domain walls at the edges, and they hop towards the other edge via the propagation of magnetic domain walls. The paths that the domain walls traveled or the Dirac strings, are shown to increase in length with magnetic fields increment and no random flipping of the bars are observed in the structure.

1. Introduction

Emergent quasiparticles with similar properties as Dirac monopoles [1] have recently been observed in Pyrochlore frustrated magnetic materials such as $Dy_2Ti_2O_7$ [2–4]. In such materials, the quasiparticles, or the monopoles, are observed at the ends of a set of overturned dipoles, which propagated into opposite directions under external perturbations at low temperatures [2,4]. These observations have then inspired the investigations of finding isolated magnetic particles at room temperature. Detection and control of these magnetic charges might then lead to the realization of high-density memory and logic devices [5,6]. One approach to observe the monopole is by making use of patterned ferromagnetic nanoislands to represent the dipoles [7]. The nanoislands structures are also termed as artificial spin ice systems [8–12] because they share similar properties with frustrated magnetic compounds [2–4] as well as with water ice, such as the ice-rule and the degenerate ground energy states [13,14]. In the water ice, the ice-rule states that for every Oxygen (O) atom, there are two Hydrogen (H) atoms placed close to it and other two H atoms placed farther from it [15]. Each O–H bond can then be represented by an arrow that points either away or towards the O atom. Similarly, in artificial spin ice systems, the spins around each vertex are pointed either away or towards the vertex, as stated by the ice-rule [13,16]. The quasiparticles, *i.e.* the monopoles, are then formed when the ice-rule is broken via random spins flip in both connected [17] and disconnected [18] honeycomb/kagome spin ice structures [19,20]. These monopoles have been shown to propagate into the lattice under the influence of external magnetic field [17,18,21,22]. In the honeycomb spin ice structures, the random nucleation and motion of the monopoles were explained either by the variations in the interaction strengths at each vertex or by the variations in the coercivity of the individual bars [17,18,23]. These two variations arise due to the roughness and the small non-uniformity in the bar dimensions during the lithography process, which promotes the generation of magnetic monopoles at random positions [17,18]. In comparison, the direct observation of the controlled formation of magnetic monopoles still needs to be investigated.

In this work, we show that it is possible to control the formation of magnetic monopoles at room temperature in connected honeycomb artificial spin ice structure via magnetic domain wall (DW) propagation from the edge of the structure. The monopoles are trapped at the vertices of the artificial spin ice structure similar to frustrated magnetic compounds [2–4,14]. Under the application of external magnetic field, the monopoles are shown to propagate in the nanostructure via continuous propagation of the DWs. Magnetic force microscopy measurements are then employed to detect the monopoles and to image the path imprinted by the DWs during their propagation through the spin ice structures. With the support of micromagnetic simulations, clear evidence of the formations of isolated magnetic monopoles and Dirac strings are presented.

2. Experimental details

A thin film stack of Ta(5 nm)/Co(15 nm)/Ta(5 nm) is deposited using ultra high vacuum magnetron sputtering. The spin ice

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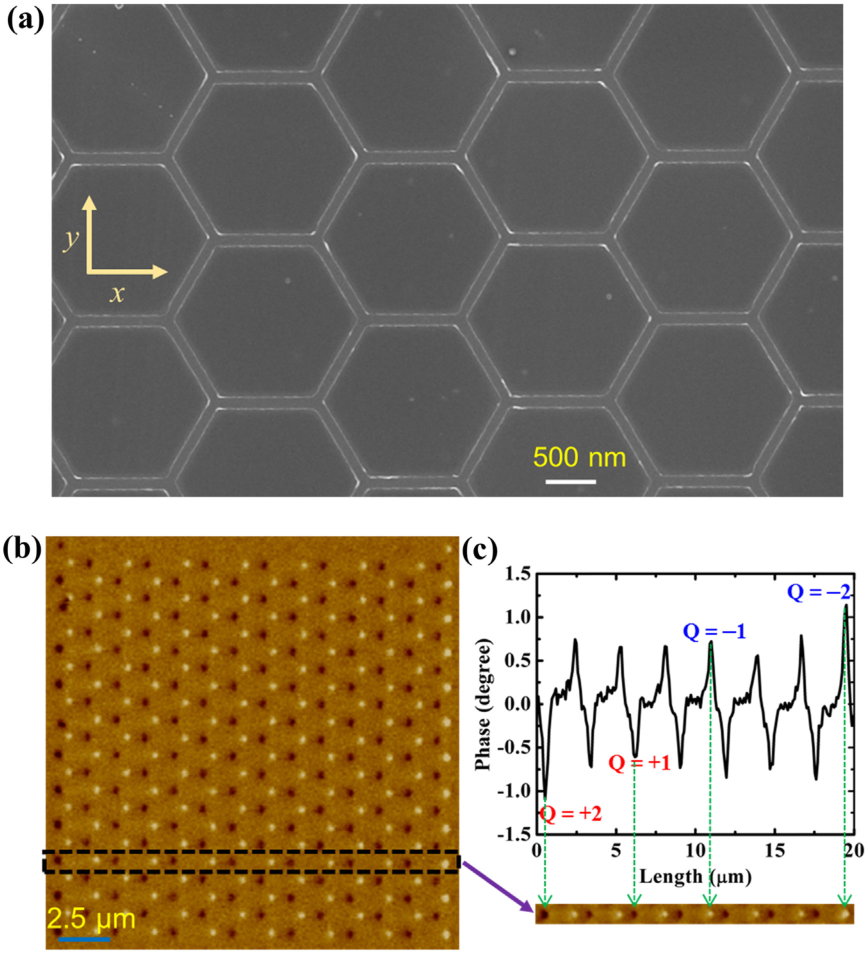

Fig. 1. (a) SEM image of connected Co honeycomb spin ice structure. (b) MFM image of artificial connected honeycomb spin ice structure saturated along negative *x*-direction with applied magnetic field of -2 kOe. The black and white contrasts at the vertices correspond to the attraction and repulsion between the MFM tip and the magnetic charges. (c) The phase shift reading of the MFM image within the enclosed area. The relative magnitude and sign of the corresponding magnetic charges are included respectively.

structure was fabricated on Si/SiO_2 (300 nm) substrates. The stack was coated with a negative resist prior to the patterning of the honeycomb spin ice structure by means of e-beam lithography. The pattern transfer was completed using Ar ion milling and wet chemical stripping techniques. The scanning electron microscopy (SEM) image of the patterned honeycomb spin ice structure is shown in Fig. 1(a). The length and width of each bar of the honeycomb structure are chosen as 1 μm and 100 nm respectively, to ensure the magnetization within each bar to form a single domain due to shape anisotropy. The spin ice array was initially saturated along $-x$ direction with a high magnetic field of $H_x = -2$ kOe. The magnetization direction of each bar is mapped using magnetic force microscopy (MFM) system at the remanence. The measurements were performed using an AppNano commercial tip with approximate magnetic moment of 10,313 emu, at lift-scan height of 140 nm to obtain an optimised magnetic topography signal. For our measurements the MFM tip was initially magnetized to have a south pole induced at its apex. In the honeycomb or kagome spin ice, three spins interact at each vertex and follow 2-in/1-out or 1-in/2-out pseudo ice-rule state [19]. The 2-in/1-out or 1-in/2-out state favor to carry a net magnetic charge at each vertex as shown in the Fig. 1(b). The vertex is said to have a net magnetic charge $Q = -1(+1)$ when the resultant magnetic flux was diverging *i.e.* 1-in/2-out state (converging *i.e.* 2-in/1-out state). More details of the charge convention are discussed in the Supplementary 1. The bright (magnetic charge, $Q = -1$) and dark ($Q = +1$) contrasts at the vertices in the Fig. 1(b), are correspond to the repulsive (south) and attractive (north) interactions between the MFM tip and the net magnetic charge at the vertices, respectively. The alternating order of contrasts at the honeycomb structure shows that each bar possesses a single domain at remanence. The MFM phase shift of the dotted area is plotted in Fig. 1(c). The magnitude and the sign of the magnetic charges of the corresponding area are also included in the figure, which can be inferred from the relative peak height of the MFM phase shift. The results show that the structure has $Q = -2$ at the right end and $Q = +2$ at the left end of the structures due to the edge effects and alternating $Q = \pm 1$ elsewhere.

3. Results and discussion

To create the magnetic excitations within the structure, the applied magnetic field is switched in the opposite direction *i.e.* along the $+x$ direction. The MFM images of the structure were captured during the stepwise reversal. Shown in Fig. 2(a), is the MFM image of the structure in quasi-remanent state after a magnetic field of $H_x = +430$ Oe was applied. The results show that disorders in the magnetic charge distribution are observed during the magnetization reversal process. The magnetic charge disorders can be seen as charges with similar contrasts being shown next to each other in the marked areas (dashed ellipses) of Fig. 2(a). Here,