Field and temperature tuning of magnetic diode in permalloy honeycomb lattice

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The observation of magnetic diode behavior with ultra-low forward voltage renders new venue for energetically efficient spintronic device research in the unconventional system of two-dimensional permalloy honeycomb lattice. However, detailed understanding of temperature and magnetic field tuning of diode behaviors are imperative to any practical application. In this report, our study unveils many important properties of magnetic diode that not only pave the way for practical applications, but also underlines the role of emergent phenomena of magnetic charge correlation on honeycomb vertices. We find that magnetic diode behavior persists across a broad temperature range. In a surprising observation, magnetic field application tends to induce a peculiar reentrant characteristic where diode behavior is suppressed in remnant field but reappears after warming to room temperature. Analysis of $I$-$V$ data suggests a modest energy gap, $\sim$ 0.03 - 0.1 eV, which is comparable to magnetic Coulomb's interaction energy between emergent magnetic charges on honeycomb vertices in the reverse biased state. It affirms the role of magnetic charge correlation in unidirectional conduction in 2D honeycomb lattice. The experimental results are expected to spur the utilization of magnetic diode in next generation spintronic device applications.

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1. Introduction

Spin or magnetic diode is a critical component to the spintronic device design for information processing and logic operations [1–4]. Therefore, a lot of emphasis is made to develop a practical prototype of magnetic diode, which could function at room temperature. Previously, researchers have used the concept of spin-charge interaction mechanism to develop a magnetic diode [5–8]. Several magnetic materials and systems are intensively investigated in the past that can be artificially tailored to depict the asymmetric spin population. It includes multilayered magnetic devices that exhibit a very high level of unidirectional spin polarization and the design of bipolar and unipolar spin diodes across a magnetic domain wall or magnetic tunnel junction [9–14]. However, application of magnetic field is universally required. For the first time, we have demonstrated the magnetic diode effect at room temperature in an unconventional system of two-dimensional geometrically frustrated permalloy honeycomb lattice that can function without magnetic field application [15,16].

An artificial magnetic honeycomb is an archetypal two dimensional geometrically frustrated magnet. In 2D permalloy (Ni$_{0.81}$Fe$_{0.19}$) honeycomb, magnetic moments align along the length of constituting element due to large shape anisotropy [17]. Consequently, two-types of local moment arrangements of two-in & one-out (or vice-versa) or, all-in or all-out states, arise on honeycomb vertices [18,19]. Under the Dumbbell formalism, a magnetic moment is considered to be made up of a pair of ‘+q’ and ‘-q’ charges, also called magnetic charges, that interact via magnetic Coulomb's interaction [20]. Here $q$ is given by $M/L$, $M$ being the net magnetic moment along the length $L$ of honeycomb element. The algebraic summation of magnetic charges on a given vertex results in the two integer multiplicities of $|Q|$ and $|3Q|$ units for the local magnetic configurations of two-in & one-out (or vice-versa) or, all-in or all-out, respectively [21]. 3Q charges are highly energetic, thus unstable [22]. In general, the magnetic charges are highly mobile in
Detailed study unveils several new properties that not only sets it apart from a conventional semiconductor diode [26], but also sheds new light on the underlying weak semiconducting characteristic of the magnetic lattice. It includes the demonstration of ultra-small forward threshold voltage ($V_{F}$ range) with diminishing magnitude as a function of temperature, an unusual freezing of magnetic charges in remnant state and an unequivocal connection between the electrical gap energy (mathematically equivalent to the Arrhenius energy) and the magnetic Coulomb interaction between emergent magnetic charges in the reverse biased state.

2. Results and discussion

The fabrication process of large throughput nanoscopic honeycomb lattice involves multiple steps that include the development of nanoporous polymer template on top of a silicon substrate [27], followed by the reactive ion etching to transfer the template to the underlying silicon substrate and permalloy material deposition on the patterned substrate in near parallel configuration (see Experimental Methods for detail). In Fig. 1a, we show the atomic force micrograph of a typical artificial honeycomb lattice resulting from the nanofabrication process, which confirms the high quality of the sample. Typical element size of the honeycomb lattice is 11 nm (length) $\times$ 4 nm (width $\times$ 8.5 nm (thickness)). The large specimen size allows for the bulk properties investigation using various macroscopic probes, such as electrical and magnetic measurements (see Experimental Methods). In Fig. 1b, we show the plots of electrical resistance as a function of temperature at different magnetic fields. Electrical measurements manifest several peculiarities. First we notice that the resistivity of permalloy honeycomb lattice at room temperature (0.29 $\Omega$ cm) is higher compared to a typical metal where the resistivity is several orders of magnitude smaller (for example, tin has a resistivity of $\sim 1.8 \mu\Omega$ cm). Such high resistivity is typically found in semiconductors or in materials that are on the verge of being a semiconductor [26]. Second, the normalized resistance ($R(T)/R(300 \text{ K})$) depicts complex temperature dependence in both zero and applied magnetic field. As temperature decreases, electrical resistance decreases and develops a plateau around $T = 150 \text{ K}$. For further decrease in temperature, the ratio $R(T)/R(300 \text{ K})$ reduces at a faster rate, indicating a phase transition. Similar behavior, albeit more resistive, is also detected in a thinner, 6 nm, permalloy honeycomb lattice [28]. Interestingly, magnetic field application of $\mu_0H = 0.5 \text{ T}$ (applied in-plane to the sample) diminishes the sharp downturn in resistance, thus altering the electrical characteristic. Finally, the trend is reversed at low temperature by registering a modest increase in resistance as temperature decreases further.

Since a diode is well defined by the unidirectional electrical transport, $I$–$V$ measurement can yield meaningful information about this characteristic at different temperatures and magnetic fields. We have performed $I$–$V$ measurements at different temperatures in zero and applied magnetic field in standard four-probe configuration. Four probe measurement tends to remove the artifact due to contact resistance. We show the $I$–$V$ plots at different temperatures in Fig. 1c. Several features, resembling diode-type behavior, are immediately noticed. It includes the observation of unidirectional conduction at ultra-low threshold voltage, $\sim \text{mV}$ and its persistence across a broad temperature range. Interestingly, the forward voltage seems to be reducing as temperature decreases. $I$–$V$ plots are obtained at modest current application of 10–300 $\mu\text{A}$, with corresponding current density of $\sim 10^6$–$10^7 \text{ A/m}^2$. At this current density, a Joule heating of less than 1 K can develop in the material [29]. Therefore, it cannot affect the experimental results. To quantify the forward voltage's dependence on temperature, we have plotted the representative forward voltage, $V_F$ at several current values as a function of decreasing temperature in Fig. 1d. It clearly shows that $V_F$ decreases from 4 $\text{mV}$ at $T = 300 \text{ K}$ to 0.6 $\text{mV}$ at $T = 150 \text{ K}$. Although the depicted behavior in forward voltage is consistent with $R$ vs $T$ measurement, shown in Fig. 1b, where electrical resistance is found to be decreasing as a function of decreasing temperature, it is in strong contrast to the conventional semiconductor-based diode where the forward voltage increases as a function of decreasing temperature [29]. It suggests an impending new mechanism behind the unidirectional conduction in permalloy honeycomb lattice.

The underlying mechanism behind magnetic diode effect is dictated by static and dynamic properties of magnetic charges. It can be broken into two parts: the unidirectional conduction and the ultra-high conductivity in the forward biased state. We first discuss the origin of unidirectional conduction. As shown in Fig. 2a, local 2-
in & 1-out’ or vice-versa and ‘all-in or all-out’ magnetic moment configurations on honeycomb vertices yield $\pm Q$ and $\pm 3Q$ charges. But only $\pm Q$ charge manifests a net magnetization. $\pm 3Q$ charges are accompanied by zero net magnetization. Magnetic measurements of permalloy honeycomb lattice in current-biased states reveal distinct net magnetization at room temperature, see Fig. 2b and c. The forward biased state has much larger magnetization than the reverse biased state, which basically corresponds to the higher population density of $\pm Q$,($\pm 3Q$) charges in the forward (reverse) biased state. The schematic distribution of magnetic charge pattern in the unbiased and the electrically biased states, shown in Fig. 2d, are consistent with magnetic measurements. Direct confirmation to the charge distribution patterns in the current biased states were recently obtained from the spin polarized neutron reflectivity measurements and the ensuing modeling of experimental data (see Fig. S1 in Supporting Information) [16]. The $\pm 3Q$ charges play an important role in the unidirectional conduction. Theoretical calculations under the reasonable assumption of drift diffusion formalism [30] unequivocally showed that the kagome network of $\pm 3Q$ charges has much larger resistance than that of $\pm Q$ charges (see Fig. S4 and Table 1 in the Supporting Information). Consequently, the high multiplicity $\pm 3Q$ charges inhibit electrical transport process by creating an energy barrier, while the low multiplicity charges ($\pm Q$) facilitate conduction [31]. After all, $\pm 3Q$ charges have much larger Coulomb’s interaction energy than $\pm Q$ charges due to the large disparity in magnitude.

Basicallly, the symmetry defying property of honeycomb lattice in the current biased states is responsible for the unidirectional conduction in the thermally tunable permalloy honeycomb lattice. However, the honeycomb element must have modest demagnetization energy. The concave element shape in our honeycomb lattice facilitates this process. Detailed micromagnetic simulations of concave element shape honeycomb lattice (as in our honeycomb lattice) and the standard rectangular element shape honeycomb lattice in current biased states reveal drastically different behaviors. While in the former case, we observe lower energy and higher magnetization in the forward biased state (consistent with experimental results), both the energy and magnetization are symmetric in the latter case (see Figs. S2–S3 in Supporting Information). The discrepancy in the current responses are mainly attributed to the demagnetization term. Due to the concave element shape, our honeycomb lattice has lower demagnetization energy. That makes the current induced tuning of magnetization feasible, but that is not the case in honeycomb lattice made of rectangular element where the demagnetization energy is quite robust.

Now, the ultra-high conductivity in permalloy honeycomb lattice arises as a result of the fast relaxation of magnetic charge defect, $2Q$, between the vertices. In recent studies, it was shown that the charge defect’s dynamics or magnetic charge fluctuation on honeycomb vertices generate transverse fluctuations in local magnetic field $B(k)$ that couple to conduction electron’s spin $\sigma$ [32]. Since magnetic charge defect relaxes at very fast rate of $T_m \sim 50$ ps, it can cause a net positive drag on electric charge carriers motion. The interaction is found to spur magnetic charge propelled electrical conduction in nanoscopic permalloy honeycomb lattice [33]. In the rest of the manuscript, we focus on the important practical aspects of magnetic diode e.g. temperature and magnetic field tuning that can pave way for practical applications in the spintronic device designs.

The underlying physics in a conventional semiconductor-based diode is dictated by the band gap between valence and conduction bands, which can be tuned by appropriate chemical doping [26]. On the other hand, the semiconducting property in magnetic honeycomb lattice is governed by the magnetic charge correlation on honeycomb vertices. Magnetic field application tends to polarize magnetic moment towards the field directions, thus alters the charge pattern by removing the energetic $3Q$ charges. The implication of field induced magnetic charge rearrangement on unidirectional electrical conduction is not clear. In principle, the absence of $3Q$ charges on honeycomb vertices would nullify the magnetic diode behavior. However, we observe a more interesting trend in electrical properties in applied magnetic field, see Fig. 3. We find that field application of $\mu_0H = 0.5$ T at $T = 200$ K does not affect the...
diode behavior (magnetic field was applied along the forward current application direction). But when the field is set to zero and temperature is reduced to $T = 150$ K in the remnant state, the unidirectional tendency in electrical transport property disappears. Instead of the diode behavior, we observe symmetrical conduction. In addition to the temperature, $T$ field measurement at multiple temperatures in succession: $T = 200$ K, 150 K, 300 K and back to 150 K. As shown in Fig. 3, magnetic diode behavior is recovered when the sample is warmed to room temperature. Consequent measurements at lower temperature, $T = 150$ K, in zero field do not seem to affect the diode behavior. In addition to the field measurement at $T = 200$ K and the subsequent cooling to $T = 150$ K in the remnant field, successive measurements in applied field and the subsequent cooling to lower temperature in remnant field were also performed at $T = 250$ K to $T = 200$ K. Similar observations were made in the latter case also (see Fig. S5).

Magnetic field application leads to anisotropic magnetoresistance (AMR) in artificial spin ice systems [34–38]. To understand the role of anisotropic magnetoresistance in the unusual field response of magnetic diode in artificial magnetic honeycomb lattice, we have performed magnetoresistance (MR) measurements at several temperatures. We show the plots of MR as a function of magnetic field at characteristic temperatures in Fig. 4. As we can see in this figure, the MR data exhibit asymmetric behaviors, characterized by a dip in resistance at $\mu_0 H = 0.003$ T followed by a peak shape structure, in magnetic field sweeps. At higher field of $\mu_0 H > 0.05$ T, both positive and negative field sweep curves merge on top of each other and register gradual decrement as a function of magnetic field. Similar anisotropic MR behavior were previously reported in field measurements on magnetic honeycomb of large element size lattice, albeit at varying field values [34,35]. Although the AMR behavior is persistent throughout the measurement temperature range, the MR percentage is less than 1%.

The unusual field effect on unidirectional conduction in permalloy honeycomb lattice is intriguing. If the AMR effects were the main mechanism behind the unusual field response of magnetic diode behavior, then we would expect to see the field induced disappearance of unidirectional conduction at any temperature. Instead of that, the diode behavior disappears only when it is cooled to a lower temperature in the remnant field. It suggests that the magnetic charge freezing on honeycomb vertices plays important role. We note that the magnitude of inplane applied magnetic field, $\mu_0 H = 0.5$ T, is much larger than the coercivity, $H_c \approx 0.1$ T, of the sample [16]. As reported previously, net magnetization of permalloy honeycomb saturates above $\mu_0 H_c \approx 0.1$ T, indicating the strength of magnetic coercivity of the system. Thus, the applied field of $\mu_0 H = 0.5$ T is sufficient to polarize magnetic moment in honeycomb element along the field application direction. Accordingly, the population density of high multiplicity charges on honeycomb vertices will be reduced. It is schematically described in Fig. 5. Yet, the magnetic diode behavior remains unperturbed to magnetic field application. A possible explanation to this unusual effect, perhaps, lies in understanding the role of thermal fluctuation to moment and magnetic charge fluctuations. Thermal fluctuation is an important factor in the occurrence of magnetic diode behavior, as current is a soft tuning parameter and cannot reverse the moment on its own. That’s why we don’t see diode behavior at low temperature as thermal fluctuation is not strong enough to assist the current-biased transformation of magnetic charges on honeycomb vertices. We think that thermal fluctuation at high temperature keeps reverting the moment at very high rate, such that the current-induced asymmetric charge distribution on honeycomb vertices dominates the field induced effect. But when the sample is cooled in remnant field, then the field-induced magnetic charge configuration (primarily comprised of $\pm Q$ charges) freezes as thermal fluctuation becomes weaker. We note that the dip in MR is observed at very low field of $\mu_0 H = 0.003$ T, which is comparable to the remnant field. So, the system does not exhibit the diode behavior at lower temperature. However, when the sample is heated to room temperature, then thermal fluctuation prevails and recovers the current-induced modification of magnetic charge.
configuration (primarily responsible for magnetic diode effect). The large thermal energy at high temperature overweighs Zeeman’s term, thus inhibits the system from attending the low energy configuration. At low temperature where thermal fluctuation is weaker, field effect would clearly dominate the charge arrangement.

Next, we try to understand the mechanism behind the semiconducting characteristic in permalloy honeycomb lattice. This exercise can provide quantitative insight in the energy crossing barrier or gap energy in the reverse biased state. For this purpose, we analyze the I/C0V traces to extract the energy gap, analogous to the band gap in a conventional semiconductor. The typical diode equation, describing the relation between current and voltage, is given by

$$I = I_0 \exp \left( \frac{eV}{kT} \right)$$

where $I$ is the current through the diode, $I_0$ is the maximum current for a large reverse bias voltage (in this case 24 V), $V$ is the voltage across the diode and $T$ is sample temperature. It is the quantity $I_0$, which is directly dependent on the gap energy $E_g$.

Following the research work by Precker et al. [29], we directly write down the formula describing the dependence of $I_0$ on $E_g$ as

$$I_0 = A T^{\gamma} \exp \left( \frac{E_g}{k_B T} \right)$$

Where $A$ is a constant, and $\gamma$ is a phenomenological exponent (arguably varying between 1/2 and 3) [29]. It is worth noting that a small variation in $\gamma$ does not affect $E_g$ at a given temperature. Combining equations (1) and (2) gives us

$$I = A T^{\gamma} \exp \left( \frac{E_g}{k_B T} \right) \exp \left( \frac{eV}{kT} \right) - 1$$

We fit experimental data using equation (3) for different values of $\gamma$. For fitting purposes, we plot $I/T^{\gamma+2}$ as a function of $(\exp[eV/k_BT] - 1)$ at different temperatures. Basically, the equation predicts a straight line dependence of y-ordinate on the x-axis variable, the slope of which yields $E_g$, see Fig. 6a and b. Fig. 6c shows the resulting plot of $E_g$ vs temperature for different $\gamma$ values. Clearly, $E_g$ at a given temperature does not seem to be much affected by the variation in $\gamma$. The estimated value of $E_g$ increases modestly from ~0.03 eV to ~0.1 eV as temperature increases. Apparently, the gap energy exhibits opposite trend to conventional semiconductor where $\Delta$ increases as temperature decreases. Nevertheless, the gap energy is of similar magnitude as the magnetic Coulomb's
interaction between magnetic charges on honeycomb vertices in the reverse biased state \[16, \langle \mu/p(4\pi) \rangle q(3Q)^2/r = 0.03 \text{ eV} \]. The prevalence of high multiplicity charges (3Q) in the reverse biased state impedes electrical conduction in magnetic diode \[16, 31\]. In other words, the magnetic Coulomb’s interaction energy between 3Q charges creates a barrier for the conduction electrons \[39\]. This barrier acts as the gap energy between low energy magnetic charge state (mainly comprised of ±Q charges) and the energetic metastable state of ±3Q charges. This new analysis further invokes the role of magnetic charge physics in magnetic diode phenomena in permalloy honeycomb lattice.

3. Conclusion

Finally, we summarize the findings. Magnetic diode provides one of the very few prototypes of room temperature spintronic diode, which operates at an ultra-low forward voltage ~mV. The comprehensive study has not only elucidated the magnetic diode, which operates at an ultra-low forward voltage ~mV. The one of the very few prototypes of room temperature spintronic devices. It establishes the unidirectional conduction. It suggests that magnetic honeycomb lattice of ultra-small element size. Atomic force micrograph of the artificial magnetic honeycomb lattice, used in this study, is shown in Fig. 1a.

Electrical measurements: Electrical measurements were performed on a 7 mm × 4 mm sample using the four-probe configuration, see inset in Fig. 1b. Electrical contacts were made using silver paste. All four contacts were in linear configuration. Electrical data at \( T = 300 \text{ K} \) was also verified using a floating four-probe contact, made by hanging steel wires. Measurements at low temperature were performed on samples with silver paste contacts only, due to their robustness, in a cryogen-free 9 T magnet with a base temperature of ~5 K. While the R vs. T measurements in zero field effect to lower temperature. The R vs. T measurements in zero field effect to lower temperature. The R vs. T measurements in zero field effect to lower temperature. The R vs. T measurements in zero field effect to lower temperature. The R vs. T measurements in zero field effect to lower temperature. The R vs. T measurements in zero field effect to lower temperature. The R vs. T measurements in zero field effect to lower temperature.

Fig. 6. Estimation of gap energy \( E_g \). (a–b) Plot of \( 1/T^2 = \) vs. \( \langle \exp(\text{eV}/k_BT) \rangle - 1 \) at a given temperature \( T \). Equation (3) yields \( E_g \) via the fitted slope of the curve for different gamma.

(c) In this plot, we show the temperature dependence of gap energy \( E_g \). Interestingly, \( E_g \) decreases as a function of decreasing temperature. Also noticeable is a very weak dependence of \( E_g \) on temperature exponent gamma.

4. Experimental methods

Nanofabrication of Artificial Magnetic Honeycomb Lattice - The fabrication process of large throughput nanoscopic honeycomb lattice involves multiple steps that include the development of nanoporous polymer template on top of a silicon substrate \[27\], followed by the reactive ion etching to transfer the template to the underlying silicon substrate and permalloy material deposition on the patterned substrate in near parallel configuration. The template fabrication process utilizes diblock copolymer polystyrene(PS)-b-poly-4-vinyl pyridine (P4VP) of molecular weight 29 k Dalton with the volume fraction of 70% PS and 30% P4VP. The diblock copolymer tends to self-assemble, under right condition, in a hexagonal cylindrical structure of P4VP in the matrix of polystyrene. Submerging the samples in ethanol for 20 min releases the P4VP cylinders, yielding a porous hexagonal template. Reactive ion etching with \( CF_4 \) gas was performed to transfer the hexagonal pattern, including the concave shape of the connecting element, to the underlying silicon substrate. RIE was performed at 50 W power at 100 mTorr \( CF_4 \) gas pressure for 20 s. The top layer of the substrate resembles a honeycomb pattern. This topographical property is exploited to create magnetic honeycomb lattice by depositing ~8 nm thick permalloy on top of the uniformly rotating substrate in near parallel configuration. It produces the desired magnetic honeycomb lattice of ultra-small element size. Atomic force micrograph of the artificial honeycomb lattice, used in this study, is shown in Fig. 1a.

Electrical measurements: Electrical measurements were performed on a 7 mm × 4 mm sample using the four-probe configuration, see inset in Fig. 1b. Electrical contacts were made using silver paste. All four contacts were in linear configuration. Electrical data at \( T = 300 \text{ K} \) was also verified using a floating four-probe contact, made by hanging steel wires. Measurements at low temperature were performed on samples with silver paste contacts only, due to their robustness, in a cryogen-free 9 T magnet with a base temperature of ~5 K. While the R vs. T measurements in zero field effect to lower temperature. The R vs. T measurements in zero field effect to lower temperature. The R vs. T measurements in zero field effect to lower temperature. The R vs. T measurements in zero field effect to lower temperature. The R vs. T measurements in zero field effect to lower temperature. The R vs. T measurements in zero field effect to lower temperature.

Since the element size of the honeycomb lattice is in the classical limit, with typical size much larger than the mean free path, we can get a rough idea about the resistivity (\( \rho \)) by using the standard formula \( R = \rho l/A \) where \( A \) and \( l \) are the area of cross-section and distance between the voltage probes, respectively.
However, unlike the thin film, honeycomb lattice has only 30% coverage. So, the area of cross-section in the direction of current flow (given by $A = w \cdot t$ where $w$ and $t$ are the width (4 mm) and thickness (8.5 nm) of the measured sample) is multiplied by a factor of 0.3. Also, given the ultra-small element size (~ 11 nm) of hon-

**Credit authors statement**

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**Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

**Data availability**

Data will be made available on request.

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**Appendix A. Supplementary data**

Supplementary data to this article can be found online at https://doi.org/10.1016/j.mtadv.2023.100386.

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