Unusual magnetoresistance in cubic B20 Fe$_{0.85}$Co$_{0.15}$Si chiral magnets

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Abstract

The B20 chiral magnets with broken inversion symmetry and C$_4$ rotation symmetry have attracted much attention. The broken inversion symmetry leads to the Dzyaloshinskii–Moriya that gives rise to the helical and Skyrmion states. We report the unusual magnetoresistance (MR) of B20 chiral magnet Fe$_{0.85}$Co$_{0.15}$Si that directly reveals the broken C$_4$ rotation symmetry and shows the anisotropic scattering by Skyrmions with respect to the current directions. The intimacy between unusual MR and broken symmetry is well confirmed by theoretically studying an effective Hamiltonian with spin–orbit coupling. The unusual MR serves as a transport signature for the Skyrmion phase.

1. Introduction

Broken symmetry is a fundamental concept prevailing in many branches of physics. The physical properties of a crystalline solid are intimately linked to its symmetry. Any broken symmetry in the solid is likewise consequential. This is evident in the cubic non-centrosymmetric B20 chiral magnets (MnSi, FeGe, Fe$_{1-x}$Co$_x$Si etc), which have attracted much attention because they harbor the helical and the Skyrmion states$^{[1-4]}$ due to the broken inversion symmetry. The B20 magnets have a simple cubic unit cell, thus with the highest symmetry. However, the basis within the cubic unit cell breaks the inversion symmetry and the four-fold (C$_4$) rotation symmetry, both have consequences. The broken inversion symmetry leads to the Dzyaloshinskii–Moriya (DM) interaction$^{[5, 6]}$, in addition to the Heisenberg exchange interaction. The D–M interaction with energy $DM \cdot (\nabla \times M)$, where $D$ is the Dzyaloshinskii–Moriya constant and $M$ is the magnetization, favors perpendicular spin alignment, as opposed to the collinear spin alignment demanded by the Heisenberg interaction with energy $A(\nabla M)^2$, where $A$ is the exchange stiffness constant. The competition between the Heisenberg and the D–M interactions leads to a spin helix ground state$^{[7, 8]}$ with zero net magnetization, instead of the usual ferromagnetic ground state. More interestingly, at temperatures close to but below the Curie temperature ($T_C$), and under a magnetic field, an exotic magnetic Skyrmion state$^{[2, 3]}$ emerges with a non-trivial topology. The spin helix and Skyrmion state in the B20 magnets, first revealed by neutron diffraction and Lorentz transmission electron microscopy$^{[1, 3, 4]}$, have captured much attention for its intriguing physics such as the Skyrmion lattice, the topological Hall effect$^{[9-11]}$, the emergent electromagnetic field, and unique prospects for applications$^{[12-14]}$. However, there has been no observation of the consequential properties due to the broken C$_4$ rotation symmetry in the B20 magnets. We report in this work the observation of the unusual magnetoresistance (MR) in (Fe–Co)Si, a prototype B20 magnet, with characteristics different from those of their counterparts in cubic centrosymmetric ferromagnets, such as Fe, Ni, and permalloy. Equally important, the experimental observation of the unusual MR also places constraints on the Hamiltonian for describing the physics of B20 materials. Our Hamiltonian with two spin–orbit terms fulfills all the symmetry requirements and can account for the experimental results.
Fe$_{0.85}$Co$_{0.15}$Si and Ni single crystal samples were oriented by x-ray diffraction and cut into rectangular cuboid

**Methods**

Fe$_{0.85}$Co$_{0.15}$Si and Ni single crystal samples were oriented by x-ray diffraction and cut into rectangular cuboid shapes with square cross section with edges parallel to the x[001], y[010], and z[001] directions for magnetic and transport measurements (figure 2(a)). The Fe$_{0.85}$Co$_{0.15}$Si single crystal sample has dimension of about 0.45 mm × 0.45 mm × 5 mm. In the resistance (R) and MR measurements, the current and the voltage leads are made along the long direction (x axis) of the specimen. Magnetic fields are applied along the x, y, and z axes, or scanned in the xyy, xzz, and yz planes. The voltage leads for longitudinal resistivity measurements are made by wire bonder with 20 μm aluminum wires and carefully aligned along the current direction within 50 μm accuracy, to avoid contamination from the transverse Hall voltage. This is verified by the symmetric MR loop ($\rho(H) = \rho(-H)$) when magnetic field is along the z direction.

Magnetic hysteresis loops were measured in Quantum Design SQUID (superconducting quantum interference device) magnetometer. AC susceptibility measurement was made by the ACMS (AC/DC magnetometry system) option of Quantum Design PPMS (physical property measurement system). Transport...
measurements were made by the DC resistivity option with sample rotator in Quantum Design PPMS, or by the Keithley 220 current source and 2182A nanovoltmeter in Quantum Design SQUID.

3. Results

3.1. MR due to broken C₄ symmetry

The Fe₀.₈₅Co₀.₁₅Si sample has $T_C \approx 23$ K as revealed by the AC susceptibility measurement (inset of figure 2(b)). The hallmarks of common ferromagnets are the $M$–$H$ curves showing distinct magnetic anisotropy, magnetic hysteresis and finite remanence at zero field. In contrast, the $M$–$H$ curves of Fe₀.₈₅Co₀.₁₅Si show no hysteresis and zero remanence due to the helical ground state with zero net magnetization. The $M$–$H$ curve is quasi linear in field until the saturation field ($H_S$) due to the formation of the conical phase under a magnetic field. The value of $H_S$ is the sum of the demagnetization field due to shape anisotropy and $H_D = D^2M/2A$, the critical field of the conical phase transforming into ferromagnetic alignment, where $D$ is the Dzyaloshinskii–Moriya coefficient and $A$ is the exchange stiffness mentioned above. Specifically, the $M$–$H$ curves are the same for fields along [010] and [001] directions, as shown in figure 2(b) even though the $C_4$ symmetry is broken. Aside from a small difference in the demagnetization field, the same result has also been observed for the [100] direction. Thus, the B20 magnet Fe₀.₈₅Co₀.₁₅Si shows complete magnetic isotropy with the same characteristics with fields along the three crystalline axes of [100], [010], and [001]. The reason is that the ferromagnetic state is a long-
range order with a characteristic length scale much larger than that of the lattice constant. Thus, the broken C4 symmetry within the unit cells of B20 is not revealed in the magnetic properties.

In contrast, broken C4 symmetry may be seen in the transport measurements since the conduction electrons, with shorter characteristic length scales comparable to the lattice constant, can probe inside the unit cells. In the transport measurements, we apply a current along the field direction, weakly dependent on temperature (at T < T_C) and exists even above T_C [17, 18], suggesting a non-ferromagnetic origin. Despite of the same M-H curves, the field dependence of MR is completely different for fields along the y(010), and z(001) directions, as shown in figure 2(c).

To recognize the unusual MR results in the B20 magnets, it is useful to first describe the well-known AMR behavior in centrosymmetric ferromagnets such as Ni and permalloy (Py) [19, 20]. In polycrystalline ferromagnetic metals, the resistivity $\rho$ depends only on the angle ($\varphi$) between the directions of the electric current (I) direction (defined as x axis) and magnetization (M) with an axial symmetry of

$$\rho = \rho_\parallel + (\rho_\parallel - \rho_\perp) \cos^2 \varphi,$$

where $\rho_\parallel$ and $\rho_\perp$ are the resistivities with M parallel and perpendicular to I respectively. Most notably, the yz scan, with M perpendicular to I, shows no variation at all (figure 3(a)). In single crystalline materials, however, AMR reflects the crystal symmetry [20, 21]. For centrosymmetric FMs with C4 symmetry, such as FCC Ni, with I in the x(100) direction, the resistivities with field along the y(010) and z(001) are the same, i.e., $\rho_y = \rho_z$. However, the yz scan is not constant but shows a four-fold symmetry. This is indeed observed in a single crystal Ni as shown in figure 3(a), where there are 4 maxima at [0, ±1,0] and [0,0, ±1] separated by 4 minima at [0, ±1, ±1].
In contrast, for MR of Fe_{0.85}Co_{0.15}Si (open symbols, figure 3(b)), all three resistivities $\rho_x$, $\rho_y$, and $\rho_z$ are different with $\rho_y < \rho_x < \rho_z$. Equally unusual, all three $xy$, $xz$, and $yz$ field scans show a two-fold symmetry (solid lines, figure 3(b)). Thus, the MR of this B20 magnet reveals the broken $C_4$ symmetry and only $C_2$ prevails. Note that, above $T_C$ (inset of figure 3(c)), MR curves of Fe_{0.85}Co_{0.15}Si for field along $x$, $y$, and $z$ directions are the same within 0.02% accuracy (MR curve for $H_z$ is slightly different from others due to the geometric effect of ordinary MR due to the Lorentz force). The magnitude of MR of Fe_{0.85}Co_{0.15}Si, as represented by $\rho_x-\rho_y$, decreases with increasing temperature from 2.1 $\mu$Ω cm at 5 K and vanishes at and above $T_C$, as shown in figure 3(c), unequivocally showing the magnetic origin of this unusual MR. It should be noted that this unusual MR is also observed in other compositions of B20 Fe_{1-x}Co_xSi and is a generic behavior inherent to the broken $C_4$ symmetry. The noncentrosymmetric cubic B20 magnets lack the $C_4$ symmetry but only $C_2$ symmetry is observed experimentally. This is the first report of the experimental signature revealing directly the broken $C_4$ rotation symmetry.

The discrepancy of symmetries in magnetometry and transport measurements arises from different dominating length scales. In the magnetometry measurements, the ordering length, both for the helical and the ferromagnetic phases, are much larger than the lattice constant of Fe_{0.85}Co_{0.15}Si. Therefore, this measurement reveals only the symmetry of the Bravais lattice, which is cubic with the $C_4$ rotational symmetry. However, in the transport experiment, the relevant length scale is the mean free path of the electrons, on the order of the atomic scale in Fe_{0.85}Co_{0.15}Si, as indicated by its very large resistivity. As a result, electrons sense the intricate structure inside the unit cell. The filling of the Fe, Co, and Si atoms brings down the symmetry from $O_h$ group of cubic to $T$ group of B20. Transport measurement thus senses the $T$ group symmetry where $C_4$ is lost.

To theoretically account for the experimental results, one needs a Hamiltonian that preserves the symmetries of $C_2$, $C_3$, and time reversal ($T$), but breaks the inversion ($I$) and $C_2$ symmetries. The microscopic D–M interaction between two neighboring spins $S_i$ and $S_j$ located at $r_i$ and $r_j$ respectively has the form of $D_{ij} \cdot (S_i \times S_j)$. This well-known interaction favors $S_i$ and $S_j$ to be perpendicular to each other and be situated in a plane perpendicular to $D_{ij}$, which is parallel to along the line joining the two spins $r_i - r_j$. When an electron with spin $\sigma$ moves from $r_i$ to $r_j$, its spin must rotate as if under a magnetic field along the momentum direction. Thus an electron effectively experiences a magnetic field along its trajectory. This leads to an interaction term of $\mathbf{k} \cdot \sigma$, where $\mathbf{k}$ is the electron momentum and $\sigma$ measures the strength of this special spin–orbit coupling imposed by the D–M interaction. The $(k \cdot \sigma)$ term clearly preserves the time reversal ($k \Rightarrow -k$, $\sigma \Rightarrow -\sigma$) symmetry but breaks the inversion ($k \Rightarrow -k$, $\sigma \Rightarrow \sigma$) symmetry. It is simple to see that the $(k \cdot \sigma)$ also preserve $C_2$ and $C_3$ symmetry, but unfortunately also preserves $C_4$ symmetry. For example, under a $C_4$ rotation about $z[001]$, the operation of $(k_x, k_y, k_z) \Rightarrow (k_y, -k_x, k_z)$ and $(\sigma_z, \sigma_y, \sigma_x) \Rightarrow (-\sigma_z, -\sigma_y, \sigma_x)$ leaves $(k \cdot \sigma)$ intact, thus unacceptable. In fact, the $(k \cdot \sigma)$ term, with the full rotation symmetry, generates only a constant MR with no directional dependence. Spin-orbital terms with quadratic momentum do break the time reversal symmetry, and are thus also unacceptable. Cubic terms in momentum are thus required for the unusual transport properties as well as other response properties.

The right minimal Hamiltonian breaking the $C_4$ symmetry while preserving the right direction of the effective magnetic field is given by:

$$H = \frac{\hbar k^2}{2m} - \alpha (k_x \sigma_x + k_z \sigma_z + 2 k_3 \sigma_3) + \beta [k_x \sigma_x (k_x^2 - k_z^2) + k_y \sigma_y (k_y^2 - k_z^2) + k_z \sigma_z (k_z^2 - k_z^2)].$$

It was derived in [22] by some of us based on the theory of invariants [23] as motivated by this experiment. As a support of the physical picture above, the relation between resistivity and the mean free path is theoretically studied in this work. By employing the standard Kubo formula technique, $\rho_x$ and $\rho_z$ as a function of the mean free path $l$ have been calculated as shown in figure 3(d). It indeed shows the discrepancy between $\rho_x$ and $\rho_z$ and the difference diminishes when $l$ increases, in which circumstance, the symmetry inside the unit cell becomes less important.

### 3.2. Anisotropic MR by the scattering of Skyrmions

At temperature close to $T_C$, exotic Skyrmion state with non-trivial topological spin textures emerges. When electrons traversing through a Skyrmion, topological Hall effect due to the transverse scattering by Berry phase in real space has been observed [9–11]. However, a closely related effect, the longitudinal scattering due to Skyrmion (i.e., Skyrmion resistance) is less studied. In fact, resistances due to scattering by Skyrmion for electrons traversing in, or perpendicular to, the Skyrmion plane are not known. As shown in figure 4, Skyrmion spin textures indeed contribute to resistance. In all three cases the current is in the $x$-direction, while the magnetic field is in the $x$, $y$, and $z$ directions with field ranging from 200 to 600 Oe at 20 K. These features correlate with magnetization measurements, which show distinctive features of the derivative $(dM/dH)$ when the Skyrmion phase emerges. Because the Skyrmion spin structure forms perpendicular to the magnetic field, the current $I$ has a specific direction with respect to the orientation of the Skyrmion as shown in the top row of
The additional resistivity due to Skyrmion can be positive or negative and includes contributions from the atomic AMR and collective spin scattering. It is about 0.05 $\mu\Omega \text{cm}$, $-0.1 \mu\Omega \text{cm}$ and 0.1 $\mu\Omega \text{cm}$ for field along x, y and z directions, respectively. Determination of intrinsic resistance due to Skyrmion spin textures is much more complicated depending on the detailed Skyrmion spin structure. Nevertheless, the MR measurement provides a simple way to detect the presence of the Skyrmion phase without resorting to neutron scattering or Lorentz TEM. Indeed, electrical detection of orientation and dynamics of Skyrmions are essential if Skyrmions are to be exploited for new technological applications.

4. Conclusions

The fascinating properties of the cubic B20 symmetry are due to the broken inversion and broken four-fold ($C_4$) rotation symmetry. We have observed a direct consequence of the broken $C_4$ rotation symmetry via the MR measurements. This observation not only deepens our understanding between symmetry and spin-dependent transport, but also constraints the minimal Hamiltonian consistent with the symmetry. The observed MR results are well reproduced by an effective Hamiltonian with two spin–orbit coupling terms that satisfies symmetry requirements. It has been well known that spin–orbit coupling is the origin of asymmetric spin interactions in B20 chiral magnets, which host the exotic Skyrmion states. Our result shows that spin–orbit coupling is equally essential for electron transports in these materials. This effective Hamiltonian can be broadly used in future studies of transports in B20 compounds. In addition, we have observed anisotropic MR by Skyrmions which can be used to detect the Skyrmions phase and the orientation of Skyrmions.

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