Majorana Multipole Response of Topological Superconductors

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In contrast to elementary Majorana particles, emergent Majorana fermions (MFs) in condensed-matter systems may have electromagnetic multipoles. We developed a general theory of magnetic multipoles for helical MFs on time-reversal-invariant superconductors. The results show that the multipole response is governed by crystal symmetry, and that a one-to-one correspondence exists between the symmetry of Cooper pairs and the representation of magnetic multipoles under crystal symmetry. The latter property provides a way to identify unconventional pairing symmetry via the magnetic response of helical MFs. We also find that most helical MFs exhibit a magnetic-dipole response, but those on superconductors with spin-$3/2$ electrons may display a magnetic-octupole response in leading order, which uniquely characterizes high-spin superconductors. Detection of such an octupole response provides direct evidence of high-spin superconductivity, such as in half-Heusler superconductors.

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Introduction.—The emergence of Majorana fermions (MFs) in electron systems has led to intense interest in searching for such exotic new excitations in condensed-matter physics. Particularly, recent developments have shown that emergent MFs appear as gapless Andreev bound states in topological superconductors (TSCs) [1–14], which provide a potential candidate for fault-tolerant qubits for topological quantum computation [15]. The increased interest in topological materials has led to a proposal of versatile three-dimensional (3D) time-reversal-invariant (TR-invariant) TSCs, such as superconducting doped topological insulators (TIs) [16–23] and Dirac semimetals [24–30], which commonly host helical MFs forming Kramers pairs on their surfaces.

Emergent MFs share some properties with elementary Majorana particles [31,32]: Both of them obey Dirac equations with charge-conjugation symmetry. Furthermore, a pair of MF zero modes are required to define the fermionic creation and annihilation operators, from which zero modes exhibit non-Abelian anyon statistics. However, compared with elementary Majorana particles, emergent MFs respond very differently to electric and magnetic fields. On the one hand, neither electric nor magnetic multipoles are possible for elementary MFs [33–35]: $\mathcal{CPT}$ invariance, where $\mathcal{C}$ is charge conjugation, $\mathcal{P}$ is space inversion, and $\mathcal{T}$ is time reversal, is a fundamental symmetry that any relativistic elementary particles are expected to respect. This symmetry forbids intrinsic electric and magnetic multipoles for elementary Majorana particles because they are their own antiparticles under $\mathcal{CPT}$. On the other hand, in superconductors, fundamental symmetry is just charge conjugation (namely, particle-hole (PH) symmetry), and the emergent MFs are self-conjugate under $\mathcal{C}$. Therefore, MFs in condensed-matter physics are not subject to such a strong constraint, and no systematic study on their electromagnetic multipoles has yet been attempted.

In this Letter, we develop a theory describing the electric and magnetic response of MFs in superconductors. For clarity, we focus here on surface helical MFs on 3D TR-invariant TSCs. A key ingredient specific to emergent MFs is crystalline symmetry. In analogy with $\mathcal{CPT}$ invariance for elementary MFs, crystal symmetry provides additional symmetry constraints on electromagnetic structures of emergent MFs. Considering the constraints, we establish a response theory for helical MFs in a low-energy limit, in which the problem reduces to the selection rule for crystal-symmetry groups. Applying our theory to possible crystal-symmetry groups, we find that helical MFs can host magnetic-multipole structures of dipole or octupole orders as the leading contribution. Additionally, the results predict a one-to-one correspondence between irreducible representations (IRs) of Cooper pairs and magnetic multipoles, which helps to determine the pairing symmetry experimentally through the magnetic response of MFs.

Particularly, the proposed theory provides a unique way to identify topological superconductivity of spin-$3/2$ electrons. Although research interest has recently focused on high-spin topological superconductivity [29,36–51], little is known about distinguishing TSCs of spin-$3/2$ electrons from those of spin-$1/2$ electrons. We clarify here that magnetic responses of helical MFs can unambiguously distinguish between these two types of SCs because the magnetic-octupole response is unique to high-spin TSCs.

To illustrate this, we apply the proposed theory to superconducting TIs of ordinary spin-$1/2$ electrons [17] and parity-mixed half-Heusler superconductors of spin-$3/2$ electrons [41,43]. The results of both numerical and analytical analyses show that only the latter exhibits the octupole response under the same crystalline symmetry.

Majorana multipole.—Helical MFs are a superconducting analogue of surface Dirac fermions of TIs and can be realized in 3D TR-invariant TSCs. From the bulk-boundary correspondence, the existence of helical MFs is ensured by the so-called 3D winding number [4,5,52,53]. Whereas the 3D winding number is defined only for fully gapped TSCs, its parity is well defined even for nodal superconductors [18]. Provided TR symmetry is maintained, these invariants are well defined and protect surface helical MFs for both nodal and nodeless superconductors.

We consider the quantum response of helical MFs when exposed to external electric or magnetic fields. First, we notice that electric fields only give moderate responses from helical MFs: Since electric fields keep TR symmetry, helical MFs remain gapless so they cannot respond so much. Conversely, magnetic fields may substantially affect them. Magnetic fields break TR symmetry, so the 3D winding number and its parity become invalid. However, this does not mean that helical MFs are not immune to any magnetic fields because actual TSCs have their own crystalline symmetry. Depending on the direction of the applied magnetic field, TR symmetry may be partially preserved by combining it with crystalline symmetry. Such magnetic crystalline symmetry determines the stability of helical MFs under magnetic fields [54].

As relevant point group operations, we consider rotations and mirror reflections that are compatible with the surface. The rotation axis and the mirror plane should be normal to the surface (see Fig. 1). We consider any surface-preserving point group G formed by them: $G = C_2, C_3, C_4, C_6, C_s, C_{2v}, C_{3v}, C_{4v}, C_{6v}$, in addition to TR symmetry $\mathcal{T}$. (G contains only the unbroken part of crystalline symmetry, if Cooper pairs spontaneously break a part of it.) Under a magnetic field, we retain magnetic twofold rotation or magnetic mirror reflection. Note that the retained magnetic symmetry is selected by the direction of an applied magnetic field. Only for a magnetic field normal (parallel) to the rotation axis (mirror plane), magnetic twofold rotation (magnetic mirror reflection) is preserved: The above magnetic field is easily seen to flip under TR, but it points back to the original when we simultaneously do a twofold rotation (mirror reflection).

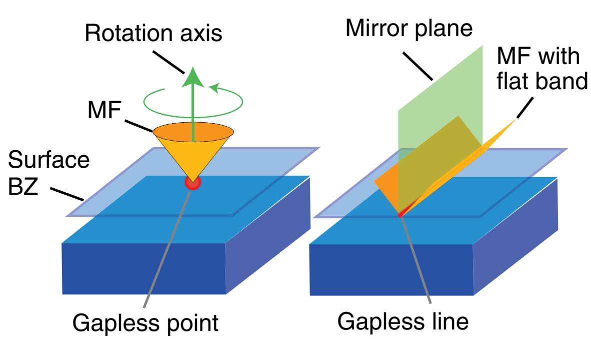

FIG. 1. MFs with gapless point and line in the surface Brillouin zone (BZ) that are protected by twofold rotation and mirror reflection, respectively.

The retained magnetic symmetry enables TSCs to host an additional topological number that is valid even when the TSC is exposed to a magnetic field: Let $g_0 \in G$ be twofold rotation or mirror reflection that defines the magnetic symmetry. Combining the magnetic symmetry with charge conjugation $\mathcal{C}$, one can introduce the magnetic chiral operator $\Gamma_{\mathrm{M}} \propto \tilde{g}_0 \mathcal{T}\mathcal{C}$, which involves the magnetic one-dimensional (1D) winding number w_{M1D} [55–59]. If w_{M1D} for magnetic twofold rotation (magnetic mirror reflection) is nonzero in the absence of magnetic fields, then helical MFs remain gapless even under a magnetic field normal (parallel) to the rotation axis (mirror plane), provided the system maintains the bulk gap. On the other hand, helical MFs do not necessarily remain gapless under other magnetic fields. This direction dependence results in an anisotropic magnetic response of helical MFs. Note that w_{M1D} for magnetic twofold rotation (magnetic mirror reflection) is defined on the symmetric axis (plane), so it protects the gapless point (line) of helical MFs at the symmetry axis (plane) in the surface Brillouin zone (see Fig. 1).

The gapless points or lines are obtained as zero modes $|u_0^{(a)}\rangle$ of the Bogoliubov–de Gennes (BdG) equation. ($a = 1, 2$ labels the Kramers degeneracy.) The index theorem for w_{M1D} [60] implies that the stable zero modes $|u_0^{(a)}\rangle$ have a common eigenvalue of Γ_{M}, say

$$\Gamma_{\mathrm{M}} |u_0^{(a)}\rangle = |u_0^{(a)}\rangle. \tag{1}$$

since zero modes with opposite eigenvalues are easily gapped in pairs. (This property is rigorously proven for generic lattice systems [59].) Moreover, from crystalline symmetry, the zero modes $|u_0^{(a)}\rangle$ should transform as a (double-valued) representation under the action of G.

To systematically study the magnetic response of MFs, we examine possible contributions of MFs to a local operator $\hat{O}(x) = \hat{c}_\sigma^\dagger(x) O_{\sigma,\sigma'} \hat{c}_{\sigma'}(x)$ of electrons, where $\hat{c}_\sigma^\dagger(x)$ and $\hat{c}_\sigma(x)$ are the electron operators with internal degrees of freedom σ such as spin and orbital, and $O_{\sigma,\sigma'}$ is a Hermitian matrix. The MFs have a nonzero response to external fields through such a local operator. For instance, if MFs make a nonzero contribution to the electron-spin operator $\hat{S}_i(x) = \hat{c}_\sigma^\dagger(x)[s_i/2]_{\sigma,\sigma'} \hat{c}_{\sigma'}(x)$ with the Pauli matrix s_i, then the MFs show a nonzero magnetic response through the Zeeman term of electrons.

The contribution of MFs is evaluated as follows: In the Nambu space with $\hat{\Psi}^\dagger(x) = [\hat{c}_\sigma^\dagger(x), \hat{c}_\sigma(x)]$, $\hat{O}(x)$ is recast

TABLE I. Magnetic multipole for Kramers pair of MFs. From left to right, each column shows two-dimensional point groups G, IRs of Δ with $w_{\mathrm{M1D}} \neq 0$, the basis of Δ, $\tilde{g}_0$ associated with Γ_{M}, IRs of $\mathcal{O}$, and the basis of $\mathcal{O}$. Here, J_i are the spin matrices, "–" means the absence of IRs, and $\mathcal{O}$ is the leading order of the magnetic multipole. We omit $e^{-i\pi J_y}$ in the basis of Δ.

G	IR of Δ	Basis of Δ	$\tilde{g}_0$	IR of $\mathcal{O}$	Basis of $\mathcal{O}$
C_2, C_4, C_6	A	$\boldsymbol{k}\cdot\boldsymbol{J}$	C_2	A	J_z
C_3	–	–	–	–	–
C_s	A	$k_xJ_z, k_xJ_y, k_yJ_x, k_zJ_x$	$\sigma_v(yz)$	A	J_x
C_{2v}	A_2	k_zJ_z	C_2	A_2	J_z
	B_1	k_xJ_z, k_zJ_x	$\sigma_v(yz)$	B_1	J_x
	B_2	k_yJ_z, k_zJ_y	$\sigma_v(xz)$	B_2	J_y
C_{3v}	A_1	$k_z(J_x^3 - J_xJ_yJ_y - J_yJ_xJ_y - J_yJ_yJ_x)$	$\sigma_v(yz)$	A_1	$J_x^3 - J_xJ_yJ_y - J_yJ_xJ_y - J_yJ_yJ_x$
C_{4v}	A_2	k_zJ_z	C_2	A_2	J_z
C_{6v}	A_2	k_zJ_z	C_2	A_2	J_z
	B_1	$k_z(J_x^3 - J_xJ_yJ_y - J_yJ_xJ_y - J_yJ_yJ_x)$	$\sigma_v(yz)$	B_1	$J_x^3 - J_xJ_yJ_y - J_yJ_xJ_y - J_yJ_yJ_x$
	B_2	$k_z(J_y^3 - J_yJ_xJ_x - J_xJ_yJ_x - J_xJ_xJ_y)$	$\sigma_d(xz)$	B_2	$J_y^3 - J_yJ_xJ_x - J_xJ_yJ_x - J_xJ_xJ_y$

into $\hat{O}(x) = (1/2)\hat{\Psi}^\dagger(x)\mathcal{O}\hat{\Psi}(x)$, with $\mathcal{O} = \mathrm{diag}(O, -O^T) = \mathrm{diag}(O, -O^*)$, where we have used the Hermiticity of O. Then, by performing the mode expansion of the quantum field $\hat{\Psi}(x) = \sum_{a=1,2}\hat{\gamma}^{(a)}|u_0^{(a)}\rangle + (\text{nonzero modes})$ and using the PH symmetry $\mathcal{C}\hat{\Psi} = \hat{\Psi}$, we obtain the coupling between $\hat{O}(x)$ and the MFs $\hat{\gamma}^{(a)}$ in the low-energy limit,

$$\hat{O}_{\mathrm{MF}} = \frac{1}{2}\sum_{a,b=1,2}\hat{\gamma}^{(b)}\hat{\gamma}^{(a)}\langle\mathcal{C}u_0^{(b)}|\mathcal{O}|u_0^{(a)}\rangle = \frac{1}{2}\hat{\gamma}^{(2)}\hat{\gamma}^{(1)}\mathrm{tr}[\mathcal{O}\rho^{(12)}], \tag{2}$$

where $\rho^{(ab)} \equiv |u_0^{(a)}\rangle\langle\mathcal{C}u_0^{(b)}| - |u_0^{(b)}\rangle\langle\mathcal{C}u_0^{(a)}|$ and an irrelevant constant term has been omitted in the last line. A nonzero $\hat{O}_{\mathrm{MF}}$ requires the following two conditions: (i) There should be zero modes $|u_0^{(a)}\rangle$ that satisfy Eq. (1). (ii) $\mathcal{O}$ should share the same IR with $\rho^{(12)}$ under the action of G. As discussed below, the former condition determines possible pairing symmetry of Cooper pairs, and the latter decides possible magnetic responses.

We outline here how the condition (i) specifies the pairing symmetry of Cooper pairs. First, we note that the symmetry of Cooper pairs determines the commutation relations between charge-conjugation and point group operations. Let us consider the BdG Hamiltonian

$$\mathcal{H}(\boldsymbol{k}) = \begin{pmatrix} \mathcal{E}(\boldsymbol{k}) & \Delta(\boldsymbol{k}) \\ \Delta^*(\boldsymbol{k}) & -\mathcal{E}^T(-\boldsymbol{k}) \end{pmatrix}, \tag{3}$$

where $\mathcal{E}(\boldsymbol{k})$ is the Hamiltonian of the normal state and $\Delta(\boldsymbol{k})$ is the gap function of the superconducting state. A point group G implies that $g\mathcal{E}(\boldsymbol{k})g^{-1} = \mathcal{E}(g\boldsymbol{k})$ with $g \in G$. The BdG Hamiltonian retains G if the Cooper pairs have symmetry $g\Delta(\boldsymbol{k})g^T = e^{i\theta_g}\Delta(g\boldsymbol{k})$ ($e^{i\theta_g}$ is a phase factor) as it holds that $\tilde{g}\mathcal{H}(\boldsymbol{k})\tilde{g}^{-1} = \mathcal{H}(g\boldsymbol{k})$, where $\tilde{g} \equiv \mathrm{diag}[g, e^{-i\theta_g}g^*]$ is the point group operator in the Nambu space. Then, the charge conjugation $\mathcal{C} \equiv \tau_x K$ and $\tilde{g}$ obeys the relation $\tilde{g}\mathcal{C} = e^{i\theta_g}\mathcal{C}\tilde{g}$, where τ_i is the Pauli matrix in the Nambu space and K is the complex-conjugation operator. Note that $\mathcal{T}$ always commutes with $\tilde{g}$ (and $\mathcal{C}$), irrespective of the pairing symmetry. For TR-invariant TSCs, $e^{i\theta_g}$ must be real, so $e^{i\theta_g} = \pm 1$. Thus, the gap function should be either even or odd under $g \in G$, which leads to $[\mathcal{C}, \tilde{g}] = 0$ ($\{\mathcal{C}, \tilde{g}\} = 0$) for g-even (odd) pairing symmetry.

As we discussed before, all zero modes, if they exist, have a common eigenvalue of Γ_{M}. Combining this with the result in the above, we can determine the pairing symmetry that is consistent with the presence of helical MFs: Since a zero mode $|u_0^{(1)}\rangle$ and its Kramers partner $|u_0^{(2)}\rangle \equiv \mathcal{T}|u_0^{(1)}\rangle$ have the same eigenvalue of Γ_{M}, we should have $[\mathcal{T}, \Gamma_{\mathrm{M}}] = 0$. This relation determines the phase ambiguity of the magnetic operator as $\Gamma_{\mathrm{M}} = \tilde{g}_0\mathcal{C}\mathcal{T}$. Then, using $\mathcal{C}^2 = 1$ and $\tilde{g}_0^2 = \mathcal{T}^2 = -1$, we obtain $[\tilde{g}_0, \mathcal{C}] = 0$ so as to be consistent with $\Gamma_{\mathrm{M}}^2 = 1$. This yields that the gap function should be even under g_0. Moreover, any other $\tilde{g} \in G$ should not anticommute with Γ_{M} for the same reason, so by considering the multiplication law between $\tilde{g}$ and $\tilde{g}_0$ as well, we can establish the commutation or anticommutation relation between $\mathcal{C}$ and $\tilde{g}$ [61]. The obtained set of commutation or anticommutation relations specifies the IR of the gap function under G, namely the pairing symmetry of Cooper pairs. We summarize it for given G and $\tilde{g}_0$ in Table I.

Now we discuss the condition (ii). Since $|u_0^{(a)}\rangle$ and $|\mathcal{C}u_0^{(a)}\rangle$ ($a = 1, 2$) are double-valued representations of G, $\rho^{(12)}$ is their product representation. Thus, using the standard group theory, we can decompose $\rho^{(12)}$ into IRs under G, which determines the representation of $\mathcal{O}$ with a nonzero $\hat{O}_{\rm M}$. (See discussions in S2 of the Supplemental Material [61].) We find that $\mathcal{O}$ consists of a single IR, which is summarized in Table I. Remarkably, the IR for $\mathcal{O}$ coincides with the paring symmetry of Cooper pairs. In other words, helical MFs respond to magnetic fields in accord with the IR of Cooper pairs. This notable property allows us to determine the pairing symmetry through the magnetic response of MFs.

We note an additional constraint $\Gamma_M \mathcal{O} \Gamma_M^\dagger = \mathcal{O}$ as Eq. (1) yields $\Gamma_{\rm M}\rho^{(12)}\Gamma_{\rm M}^\dagger = \rho^{(12)}$. This constraint implies $\mathcal{T}\mathcal{O}\mathcal{T}^{-1} = -\mathcal{O}$ as it holds that $\mathcal{C}\mathcal{O}\mathcal{C}^{-1} = -\mathcal{O}$ and $\tilde{g}_0 \mathcal{O} \tilde{g}_0^{-1} = \mathcal{O}$. Therefore, $\mathcal{O}$ should be a magnetic operator, as we expected. In Table I, we provide a representative set of bases for the gap function and the magnetic operator $\mathcal{O}$.

Majorana octupole in spin-3/2 superconductors.— Table I reveals that MFs show a magnetic-octupole response $\propto J^3$ when the surface has C_{3v} or C_{6v} symmetry. This unique behavior is intrinsic to high-spin TSCs of spin-3/2 electrons: In fact, the relevant basis vanishes, $J_x^3 - J_xJ_yJ_y - J_yJ_xJ_y - J_yJ_yJ_x = J_y^3 - J_yJ_xJ_x - J_xJ_yJ_x - J_xJ_xJ_y = 0$ for spin-1/2 electrons ($J_i = \sigma_i/2$).

To confirm this result, we calculate the magnetic response of MFs in half-Heusler superconductors. In these compounds [36–41], a strong spin-orbit interaction (SOI) and high crystal symmetry provide a fourfold degenerate band at the Γ point, which is well described by spin-3/2 fermions [43]. Additionally, recent experiments have suggested the existence of parity-mixed superconductivity with line nodes [40,41]. We show here that the parity-mixed superconductor exhibits a magnetic-octupole response. Consider the low-energy model with T_d symmetry [43]:

$$H_{\rm LK}(\boldsymbol{k}) = \alpha \boldsymbol{k}^2 + \beta \sum_i k_i^2 J_i^2 + \gamma \sum_{i\neq j} k_i k_j J_i J_j + \delta \sum_i k_i (J_{i+1} J_i J_{i+1} - J_{i+2} J_i J_{i+2}), \quad (4)$$

where $i = x, y, z$ and $i+1 = y$ if $i = x$, etc., and J_i are the 4×4 spin matrices of spin-3/2 fermions. Because inversion symmetry is absent, the Hamiltonian includes the antisymmetric SOI, which is proportional to δ and causes spin splitting at the Fermi surface. In their superconducting states, Cooper pairs form between spin-3/2 electrons, which allows quintet and septet parings in addition to the conventional singlet and triplet pairings [43,64,65]. Furthermore, the antisymmetric SOI generally mixes the parity of the gap function, so the even- and odd-parity components coexist in the gap function [62,66–69] and the odd-parity component is aligned with the antisymmetric SOI [67], providing the spin-septet pairing. Based on this insight, the gap function is given by the mixture of spin-singlet and spin-septet components, $\Delta(\boldsymbol{k}) = \Delta/\sqrt{1+\eta^2}[\eta 1_4 + \sum_i k_i (J_{i+1}J_iJ_{i+1} - J_{i+2}J_iJ_{i+2})]e^{-i\pi J_y}$, even when we choose the conventional A_1 state of T_d, where

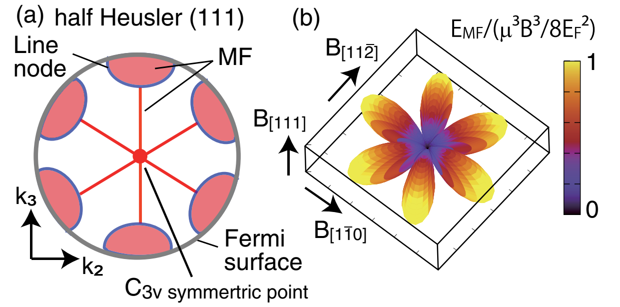

FIG. 2. (a) Surface states of the half-Heusler superconductor in (111) plane. The red line and red areas indicate helical MFs with flat dispersion and the line-node-induced Majorana flat bands. $k_1 = (1/\sqrt{3})(k_x + k_y + k_z)$, $k_2 = (1/\sqrt{2})(k_x - k_y)$, and $k_3 = (1/\sqrt{6})(k_x + k_y - 2k_z)$. (b) Energy gap of helical MF at $k_2 = k_3 = 0$ as a function of $\boldsymbol{B}$ under the Zeeman magnetic field $\mu \boldsymbol{B}\cdot\boldsymbol{J}$.

η parametrizes the mixing between the spin-singlet and spin-septet components and 1_n is the $n\times n$ identity matrix.

The superconducting state hosts six line nodes encircling the k_x, k_y, and k_z axis, in analogy with other parity-mixed superconductors [60,70–75]. Here, we focus on the (111) surface because the magnetic-octupole response requires C_{3v} symmetry. To verify the existence of helical MFs, we numerically diagonalize the BdG Hamiltonian with the surface normal to the [111] direction and find a helical MF with three flat dispersions (see Fig. S2 [61]), as schematically depicted in Fig. 2(a). Each flat dispersion lies on the mirror planes with mirror-reflection symmetries, σ, $C_3^\dagger \sigma C_3$, and $(C_3^\dagger)^2 \sigma (C_3)^2$, where σ is the mirror reflection with respect to the $(1\bar{1}0)$ plane and C_3 is threefold rotation around the [111] direction. Using these mirror reflections, we obtain three $\Gamma_{\rm M}$ and the associated three $w_{\rm M1D}$, which protects zero modes on each flat dispersion. In particular, the three flat dispersions meet at a C_3 invariant point, at which the zero modes become simultaneous eigenstates of $\Gamma_{\rm M}$ and C_3. To demonstrate magnetic response, we add a Zeeman magnetic term $\mu \boldsymbol{B}\cdot\boldsymbol{J}$ in Eq. (4), which leads to an anisotropic response with C_{3v} symmetry in Fig. 2(b). The Zeeman magnetic term contributes to the energy gap of the MFs on the order of $3\sqrt{2}\mu^3 B^3/32E_F^2$ (See Fig. S3 [61]), where μ is a coefficient of Zeeman term and E_F the Fermi energy, implying a magnetic-octupole response.

Several remarks are in order. (i) We also examine the magnetic response of a spin-1/2 TSC with $C_{3\rm v}$ symmetry. In sharp contrast to the spin-3/2 case, the helical MFs only show the dipole response (see the magnetic response of superconducting TIs in Fig. S1 [61]). (ii) The octupole response also appears in orbital magnetic effects since the orbital magnetic terms [76–81] also should be the same IR. (iii) Another high-spin superconductor of spin-3/2 electrons was recently proposed for antiperovskite materials with O_h group [28,29]. We obtain a similar magnetic-octupole response of MFs on the (111) surface when its pairing symmetry is A_{2u} of O_h.

Conclusions.—In this Letter, we develop a theory of Majorana multipoles for 3D TR-invariant TSCs, which provide novel experimental means to identify bulk pairing symmetry and high-spin superconductivity. The Majorana multipoles may be observed through spin-sensitive measurements such as spatially resolved NMR measurements [82] or the surface tunneling spectroscopy under magnetic fields [76–80,83].

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