PHYSICAL REVIEW B **102**, 014422 (2020)

Editors' Suggestion   FeState Key Laboratory

(Received 24 March 2020; accepted 28 May 2020; published 15 July 2020)

The energy vs crystal momentum ( ) diagram for a solid (band structure) constitutes the road map for navigating its optical, magnetic, and transport properties. By selecting crystals with specific atom types, composition, and symmetries, one could design a target band structure and thus desired properties. A particularly

be treated just as SOC-induced splitting in nonmagnetic (NM) materials [1,2], through the usual Thomas term [4], for example, allowing for antiferromagnetism in calculations on $BiCoO_3$ [20] having SOC manifests but a small change in its spin splitting; furthermore, if SOC is deliberately removed from the Hamiltonian, the predicted spin splitting vanishes in the whole Brillouin zone (BZ). Also, the field-free magnetic mechanism discussed in the present paper differs from the anomalous spin-orbit coupling in antiferromagnets induced by applying external magnetic field, discussed in Refs. [21,22].

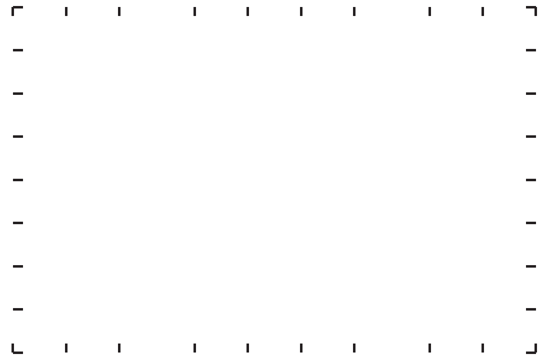
A phenomenological theory of magnetic spin splitting was proposed in 1964 by Pekar and Rashba [23], suggesting that the presence in magnetic compounds of a spatially dependent intrinsic magnetic field $\mathbf{h}(\mathbf{r})$, periodic with the crystal period, can lead to coupling of Pauli matrices $\boldsymbol{\sigma}$ to this $\mathbf{h}(\mathbf{r})$. This would result in a magnetic mechanism of $\boldsymbol{k}$-dependent spin splitting, suggestive of a new type of spin-orbit coupling. Because the $\boldsymbol{k} \cdot$ formalism used in Ref. [23] did not afford an atomistic definition of $\mathbf{h}(\mathbf{r})$ and its ensuing spin splitting, nor did it provide for guiding principles to select a target material for investigating such effects, examination of these 1964 ideas remained dormant for a long time.

In the present paper, inspired by Ref. [23], we demonstrate an AFM mechanism that creates $\boldsymbol{k}$-dependent spin splitting $(\boldsymbol{k})$ even in centrosymmetric, low- compounds, persists even at time-reversal invariant wave vectors, and has an unusual quadratic scaling on momentum $\boldsymbol{k}$. The coupling of spin to lattice degrees of freedom via the periodic spatial dependent intrinsic magnetic field $\mathbf{h}(\mathbf{r})$ is analogous to a new form of spin-orbit ; the fact that spin splitting can, however, exist even in absence of the electrical mechanism of the spin-orbit in the Hamiltonian is noteworthy. We formulate the general magnetic space group conditions ("design principles") for spin splitting in different AFM prototypes, either with or without SOC, and illustrate via detailed first-principles calculations a case of purely AFM-induced spin splitting.

II. MAGNETIC SYMMETRY CONDITIONS FOR AFM-INDUCED SPIN SPLITTING

A. Symmetries that enforce spin degeneracy

To select a compound for direct magnetic $\boldsymbol{k}$-dependent spin splitting we inspect the underlying symmetry requirements. We first list the symmetries that spin degeneracy, preventing spin splitting (SS), then discuss how to violate those symmetries. (i) As is known [24], the combination θ of time reversal θ and spatial inversion symmetries ensures double degeneracy for arbitrary wave vector $\boldsymbol{k}$. Likewise, (ii) when SOC is turned off, the spin and spatial degrees of freedom are decoupled, so there could exist pure spin rotation , a spinor symmetry, that reverses the spin but keeps momentum invariance, thus preserving spin degeneracy for all wave vectors. The spin rotation does not exist in AFM when the alternating magnetic moments reside on different



nor electric SOC induces any splitting; (2) the point shows zero spin splitting when $\lambda_{SOC} = 0$ and linear dependence of λ_{SOC}, illustrating a cooperation of both magnetic and SOC mechanisms; notice that despite being a TRIM point, it shows spin splitting, unlike the case of purely SOC-induced effects; (3) the nontrivial case of purely magnetic-induced spin splitting occurs along the - (as well as *A*-) line, where nonzero spin splitting is present even at $\lambda_{SOC} = 0$ and is almost independent of λ_{SOC}. The appearance of such distinct spin splitting behaviors at different wave vectors in a single compound would be advocated for multifunctional spintronic applications.

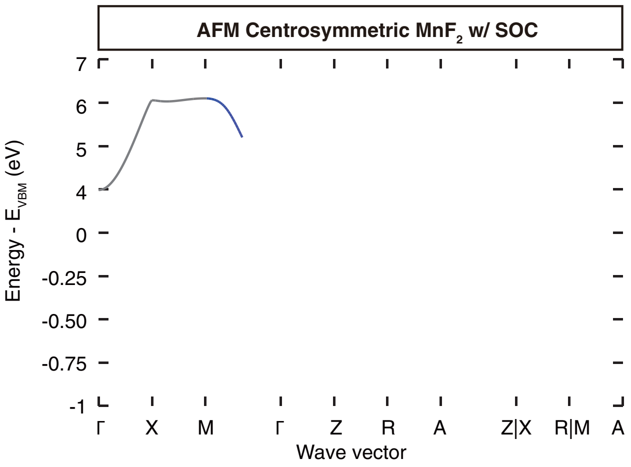
IV. DISCUSSION

This study uncovers the design principles of spin splitting in AFM compounds based on magnetic symmetry analysis

composed of , 1, 2, 3, ′, 1′, 2′, 3′ and their combination with a rotation of 2π ( ): $_D$ = { , $\overline{C_2}$, , $\overline{\sigma}$ }, $1_D = \{C_2|0\}$ $_D$, $2_D = \{C_2|\boldsymbol{\tau}\}$ $_D$, $3_D = \{C_4|\boldsymbol{\tau}\}$ $_D$, and $'_D$, $1'_D$, $2'_D$, $3'_D$

TABLE VIII. The transformation properties of symmetrized matrix and irreducible tensor under symmetry operations of the little point group at with SOC.

Symmetrized matrix	Irreducible tensor	$\{C_2 \mid \boldsymbol{\tau}\}$	$\{C_2$


AFM Centrosymmetric MnF2 w/ SOC
Energy - EVBM (eV)
7
6
5
4
0
-0.25
-0.50
-0.75
-1
Γ X M Γ Z R A Z|X R|M A
Wave vector

[26] S. V. Gallego, J. M. Perez-Mato, L. Elcoro, E. S. Tasci, R. M. Hanson, K. Momma, M. I. Aroyo, and G. Madariaga, J. Appl. Crystallogr. **49**, 1750 (2016).
[27] R. A. Erickson, Phys. Rev. **90**, 779 (1953).
[28] J. Rodriguez-Carvajal, M. Fernandez-Diaz, and J. Martinez, J. Phys.: Condens. Matter **3**, 3215 (1991).
[29] M.-R. Li, U. Adem, S. R. C. McMitchell, Z. Xu, C. I. Thomas, J. E. Warren, D. V. Giap, H. Niu, X. Wan, R. G. Palgrave, F. Schiffmann, F. Cora, B. Slater, T. L. Burnett, M. G. Cain, A. M. Abakumov, G. van Tendeloo, M. F. Thomas, M. J. Rosseinsky, and J. B. Claridge, J. Am. Chem. Soc. **134**, 3737 (2012).
[30] I. Tomeno, H. N. Fuke, H. Iwasaki, M. Sahashi, and Y. Tsunoda, J. Appl. Phys. **86**, 3853 (1999).
[31] P. Wadley, V. Hills, M. R. Shahedkhah, K. W. Edmonds, R. P. Campion, V. Novák, B. Ouladdiaf, D. Khalyavin, S. Langridge, V. Saidl, P. Nemec ., Sci. Rep. **5**, 17079 (2015).
[32] E. Ressouche, N. Kernavanois, L.-P. Regnault, and J.-Y. Henry, Phys. B (Amsterdam, Neth.) **385-386**, 394 (2006).
[33] A. A. Belik ., Chem. Mater. **18**, 798 (2006).
[34] S. Hayami, Y. Yanagi, and H. Kusunose, J. Phys. Soc. Jpn. **88**, 123702 (2019).
[35] M. Naka, S. Hayami, H. Kusunose, Y. Yanagi, Y. Motome, and H. Seo, Nat. Commun. **10**, 4305 (2019).
[36] J. W. Stout and H. E. Adams, J. Am. Chem. Soc. **64**, 1535 (1942).
[37] W. H. Baur and A. A. Khan, Acta Crystallogr., Sect. B: Struct. Crystallogr. Cryst. Chem. **27**, 2133 (1971).
[38] J. O. Dimmock and R. G. Wheeler, Phys. Rev. **127**, 391 (1962).
[39] J. P. Perdew, J. A. Chevary, S. H. Vosko, K. A. Jackson, M. R. Pederson, D. J. Singh, and C. Fiolhais, Phys. Rev. B **46**, 6671 (1992).
[40] J. Varignon, M. Bibes, and A. Zunger, Nat. Commun. **10**, 1658 (2019).
[41] G. Trimarchi, Z. Wang, and A. Zunger, Phys. Rev. B **97**, 035107 (2018).
[42] J. W. Stout, J. Chem. Phys. **31**, 709 (1959).
[43] K. Ishizaka, M. S. Bahramy, H. Murakawa, M. Sakano, T. Shimojima, T. Sonobe, K. Koizumi, S. Shin, H. Miyahara, A. Kimura, K. Miyamoto ., Nat. Mater. **10**, 521 (2011).
[44] D. D. Sante, P. Barone, R. Bertacco, and S. Picozzi, Adv. Mater. **25**, 509 (2012).