Parity-Violating Antiferromagnetic Order Leading to Asymmetric Electronic Band Structure in LaMnSi

T. Iwata[A], K. Shiraishi[A] , T. Aoyama[A], D. Senba[A], T. Takeda[A, C], Y. Fujisawa[D], M. Nurmamat[A] , K. Nakanishi[A], K. Yamagami[E] , Y. Yanagi[F], T. Yamada[F], A. Kimura[A,B,G, H], H. Tanida[F], and K. Kuroda[A,B,H]

[A] *Graduate School of Advanced Science and Engineering, Hiroshima University, 1-3-1 Kagamiyama, Higashi-Hiroshima 739-8526, Japan*
[B] *International Institute for Sustainability with Knotted Chiral Meta Matter (WPI-SKCM²), Hiroshima University, 2-313 kagamiyama, Higashi-Hiroshima, 739-0046, Japan*
[C] *Department of Electrical Engineering and Information Systems, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan*
[D] *Research Institute for Synchrotron Radiation Science, Hiroshima University, 2-313 Kagamiyama, Higashi-Hiroshima 739-0046, Japan*
[E] *Japan Synchrotron Radiation Research Institute (JASRI), Sayo, Hyogo 679-5198, Japan*
[F] *Liberal Arts and Sciences, Toyama Prefectural University, 5180 Kurokawa, Imizu, Toyama 939-0398, Japan*
[G] *Synchrotron Radiation Research Center, National Institutes for Quantum Science and Technology (QST), 1-1-1 Kouto, Sayo-cho, Sayo-gun, Hyogo 679-5148, Japan*
[H] *Research Institute for Semiconductor Engineering (RISE), Hiroshima University, 1-4-2 Kagamiyama, Higashi-Hiroshima 739-8527, Japan*

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Spontaneous symmetry breaking modulates electronic states and serves as the microscopic origin of functional properties. For example, spin-split bands in systems with broken time reversal (*T*) or spatial inversion (*P*) symmetry provide a mechanism to manipulate spin degrees of freedom (Fig.1 **a**, **b**). Extending this concept to antiferromagnets, the interplay between crystal and magnetic structures induces specific symmetry breakings [1]. This allows for the emergence of functional properties that can be manipulated via antiferromagnetic order, offering a unique platform for next-generation spintronics [2].

Among these systems, antiferromagnets that preserve combined parity-time (*PT*) symmetry while breaking both *P* and *T* symmetries are attracting particular attention [3]. In these materials, the specific symmetry breaking induces momentum-space asymmetric band structures while retaining spin degeneracy (Fig.1 **c**). Crucially, this electronic modulation permits the emergence of time-non-invariant nonlinear susceptibilities. These susceptibilities are the origin of exotic phenomena such as non-reciprocal transport and nonlinear optical effects. Unlike systems where inversion symmetry is broken merely by the crystal structure, the magnetic symmetry breaking here is intrinsically coupled to the antiferromagnetic order, offering superior controllability of these functional properties through magnetic domain manipulation.

LaMnSi serves as an ideal candidate to realize such a state [4]. It crystallizes in the non-symmorphic space group *P4/nmm*, where two Mn sublattices are related by the *P* operation [red and blue tetrahedron in Fig.2 **a**]. Below the Néel temperature $T_N = 293$ K, the Mn moments form a collinear antiferromagnetic order along the *c*-axis. In this magnetic structure, *P* symmetry is broken because the inversion operation exchanges the sublattices while leaving the axial spin vectors invariant. Nevertheless, the combined *PT*-symmetry is preserved, as the original configuration is recovered by a subsequent *T* operation (Fig.2 **b**). Consequently, LaMnSi is established as a parity-violating antiferromagnet, providing fertile ground to investigate the functional properties driven by the magnetic symmetry breaking.

In this study, we investigated the electronic structure and symmetry breaking in LaMnSi. We performed soft X-ray angle-resolved photoemission spectroscopy (SX-ARPES) at SPring-8 BL25SU [5]. The observed three-dimensional electronic structures showed good agreement with density-functional theory calculations for the antiferromagnetic phase (Fig.2 **d**, **e**). Crucially, this result demonstrates that Mn 3d electrons play a dual role, governing both the magnetic ordering and the itinerant electrical conduction.

Furthermore, we performed optical second-harmonic generation (SHG) measurements to probe the parity-violating antiferromagnetic order. We resolved the time-noninvariant nonlinear optical susceptibility components and tracked their temperature dependence by employing polarization-dependent SHG. This analysis clarified the behavior of susceptibility components specifically derived from the parity-violating antiferromagnetic order. Moreover, scanning SHG microscopy visualized antiferromagnetic domains on the scale of several hundred micrometers. These results establish LaMnSi as a promising platform for exploring functional properties driven by the parity-violating antiferromagnetic order.

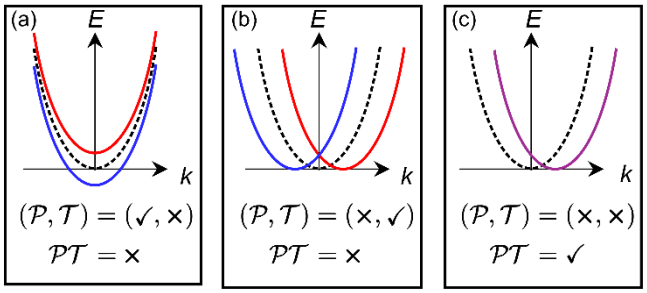

FIGURE 1. Relationship between band structure, symmetry breaking, and nonlinear optical response. (**a**) the symmetric spin splitting, (**b**) the asymmetric spin splitting, and (**c**) the asymmetric band with spin-degeneracy. Red/blue and purple lines denote spin-split and spin-degenerate states, respectively, while dashed curves indicate reference bands with both P and T preserved. The presence (✓) or absence (×) of symmetries are listed at the bottom of each panel.

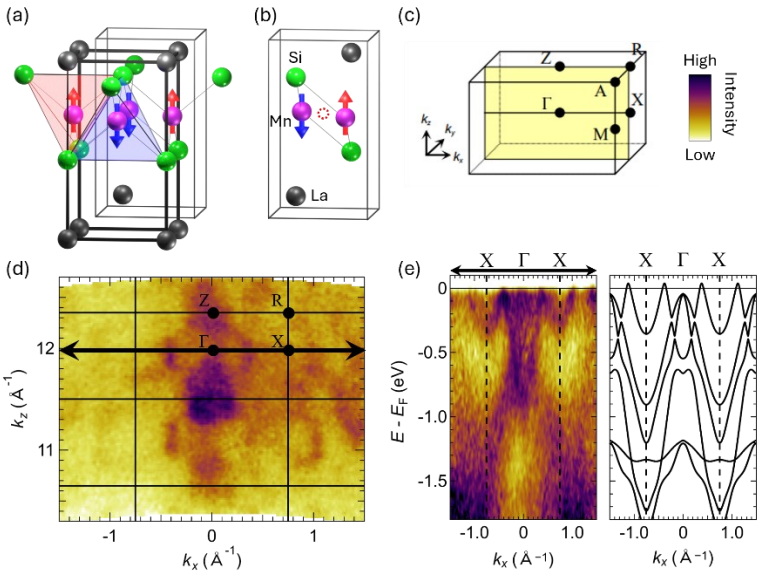

FIGURE 2. Magnetic and electronic structures of LaMnSi. (**a**) Magnetic structure of LaMnSi. Mn moments form a $q = 0$ antiferromagnet with moments aligned along [001] (*c* axis), as indicated by red and blue arrows. Thick lines denote the primitive unit cell. (**b**) The structure is redrawn using the alternative unit cell [thin line in **a**], where the dotted circle highlights the crystallographic inversion symmetry broken by the antiferromagnetic order. (**c**) Brillouin zone with high-symmetry points (**d**) Fermi surface map on the momentum plane denoted by the yellow plane in **c**, which is obtained by varying hu from 400 to 600 eV in steps of 2 eV. (**e**) Left: ARPES image along with Γ-X line indicated the solid arrow in (d). Right: calculated band structures along with Γ-X line for the antiferromagnetic phase.

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