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Previous theoretical efforts have predicted a type of unconventional antiferromagnet characterized by a crystal symmetry that connects antiferromagnetic sublattices in real space and simultaneously couples spin and momentum in reciprocal space. This results in a unique crystal-symmetry-paired spin-valley locking and related properties including piezomagnetism and noncollinear spin current even without spin-orbit coupling. However, most known unconventional antiferromagnets do not meet the necessary symmetry requirements for nonrelativistic spin current, and this limits applications in spintronic devices. Here, we demonstrate crystal-symmetry-paired spin-valley locking in a layered room temperature antiferromagnetic compound, Rb1-δV2Te2O. Spin-resolved photoemission measurements directly show the opposite spin splitting between crystal-symmetry-paired valleys. Quasi-particle interference patterns show the suppression of inter-valley scattering due to the spin selection rules that are a direct consequence of the spin-valley locking. These results suggest that Rb1- δ V2Te2O is a potential room-temperature altermagnet candidate. Our observations highlight a methodology that enables both the advantages of layered materials and possible control through crystal symmetry manipulation for advancements in magnetism, electronics, and information technology.

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