Atomic programming of crystalline order into a crystal at the mesoscale

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Controlling the arrangement of individual atoms with lasers, ion traps, and scanning probe techniques has enabled quantum simulation and computing platforms that transcend naturally occurring configurations of matter. Yet achieving comparable atomic control within a solid and at scale remains a foundational challenge that could revolutionize the design of artificial matter.Here, we demonstrate deterministic atomic engineering inside a scanning transmission electron microscope, enabling control over atomic motion within a three-dimensional crystal [1]. Using the layered magnetic semiconductor CrSBr, an air-stable material with strong excitonic excitations and correlated magneto-electronic physics [2-5], we observe that exposure to 200 keV broad electron beam irradiation induces Cr atom displacements that generate defect complexes [6]. These include the δ_2 Cr interstitial-vacancy complex, which calculations predict to be electronically localized impurity states and optically active [7].To realize controlled displacement of Cr atoms and deterministically create these and other defect complexes, we developed several automated beam-control approaches. First, we realized a rapid, low-dose, in situ beam-control approach to position the electron beam with sub-20-picometer accuracy onto an atom or atomic column of interest [8]. We demonstrate this technique, termed “atomic lock-on,” by revealing single-atom events such as displacement, recapture, and random telegraph noise in monolayer WS_2. Second, we developed a highly dose-efficient positioning algorithm, termed “stay locked-on,” that allows the electron beam to be walked between user-defined lattice sites [9]. This algorithm is essential for the creation of defects in precisely lattice-ordered arrangements. Third, we developed an approach for high-frequency directional beam motion between two or more atomic columns, termed “atom steering” [1]. This enables simultaneous control and monitoring over the direction and final position of atomic displacement, which is essential for deliberately creating user-defined defect complexes.These results establish atomic engineering within the electron microscope as a practical reality. This capability opens opportunities to design quantum defects and many-body phases with tunable charge and spin interactions and to control host lattice excitations by arranging atoms in patterns that are commensurate or incommensurate with the underlying crystal order over mesoscopic and potentially even macroscopic length scales.

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