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**DOI**

[10.1073/pnas.1718931115](https://doi.org/10.1073/pnas.1718931115)

**Publication date**

2018

**Document Version**

Final published version

**Published in**

Proceedings of the National Academy of Sciences of the United States of America

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[Link to publication](#)

**Citation for published version (APA):**

Gao, S., Flicker, F., Sankar, R., Zhao, H., Ren, Z., Rachmilowitz, B., Balachandar, S., Chou, F., Burch, K. S., Wang, Z., van Wezel, J., & Zeljkovic, I. (2018). Atomic-scale strain manipulation of a charge density wave. *Proceedings of the National Academy of Sciences of the United States of America*, 115(27), 6986-6990. <https://doi.org/10.1073/pnas.1718931115>

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# Atomic-scale strain manipulation of a charge density wave

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Edited by J. C. Séamus Davis, Cornell University, Ithaca, NY, and approved May 18, 2018 (received for review October 30, 2017)

**A charge density wave (CDW) is one of the fundamental instabilities of the Fermi surface occurring in a wide range of quantum materials. In dimensions higher than one, where Fermi surface nesting can play only a limited role, the selection of the particular wavevector and geometry of an emerging CDW should in principle be susceptible to controllable manipulation. In this work, we implement a simple method for straining materials compatible with low-temperature scanning tunneling microscopy/spectroscopy (STM/S), and use it to strain-engineer CDWs in 2H-NbSe<sub>2</sub>. Our STM/S measurements, combined with theory, reveal how small strain-induced changes in the electronic band structure and phonon dispersion lead to dramatic changes in the CDW ordering wavevector and geometry. Our work unveils the microscopic mechanism of a CDW formation in this system, and can serve as a general tool compatible with a range of spectroscopic techniques to engineer electronic states in any material where local strain or lattice symmetry breaking plays a role.**

scanning tunneling microscopy | charge density waves | strain | NbSe<sub>2</sub>

Strain is one of few experimental handles available that can in principle be used to controllably and reversibly tune electronic and optical properties of materials, ranging from bulk (1–3) to reduced dimension materials (4–7). However, achieving sufficient strain to generate novel behavior and simultaneously detecting the resulting emergent phenomena can be highly nontrivial. In thin films, strain has been successfully generated by utilizing the lattice mismatch between the film and the substrate, but the film growth on lattice mismatched substrates can often be challenging. In bulk single crystals, strain can be applied by attaching materials to piezoelectric substrates (1, 2, 8), but applicability to a wide range of characterization techniques has been limited by the necessity of independently controlling one or more piezoelectric stacks. Moreover, in real, imperfect materials, the strain may not transmit uniformly through the bulk to the top surface studied, so there is a pressing need for concomitant nanoscale structural and electronic characterization.

Transition-metal dichalcogenides (TMDs) are an emerging family of extremely elastic quasi-2D materials able to withstand large amounts of in-plane strain (>10%), thus providing the ideal playground for bandgap engineering, the design of new topological phases, and the manipulation of many-body ground states (4, 5). A charge density wave (CDW) is one of the emergent states occurring in a range of TMDs (4), often accompanied by other, possibly competing, phases. A prototypical example is 2H-NbSe<sub>2</sub>, which exhibits both superconductivity ( $T_c \sim 7.2$  K) and a triangular (3Q) CDW phase ( $T_{CDW} \sim 33$  K) (9) that has intrigued the community for decades (10–22). CDW formation can in principle arise from Fermi surface nesting, electron–electron interactions, or electron–phonon interactions (23). Inspection of the Fermi surface of NbSe<sub>2</sub> shows little propensity to nesting (12), and alternative mechanisms have been sought since the earliest studies (24, 25). Although there is a growing consensus that electron–phonon coupling might play a role

(15, 19, 20, 26), a fundamental question remains as to what drives the choice of a particular CDW wavevector and geometry in this and other quasi-2D TMDs, and how these phases could be manipulated.

Here we implement a simple method that can achieve strain at the surface of a bulk material, while simultaneously allowing the measurement of electronic properties with atomic-scale precision. Our strain method exploits the mismatch in the thermal expansion coefficient (TEC) of materials to generate strain (Fig. 1A and *Methods*). Specifically, we glue a material of interest to a substrate with a vastly different TEC and cool it down from room temperature to  $\sim 4$  K to induce strain. The striking simplicity of this method makes it suitable for rigid spatial constraints of spectroscopic imaging scanning tunneling microscopy (SI-STM) employed here, and it can also be easily extended to other low-temperature techniques. Although STM experiments have occasionally observed induced strain upon cooling down the sample (21, 27), we note that our STM experiment utilizes the sample-substrate TEC mismatch for intentional strain application. Applying this method to 2H-NbSe<sub>2</sub>, we discover a remarkable emergence of two unexpected charge-ordered phases, which we study to unveil the distinct roles of phonons and electrons in determining the ordering wavevector and geometry of a CDW.

## Results

STM topographs of the surface of unstrained NbSe<sub>2</sub> reveal a hexagonal lattice of Se atoms with a characteristic triangular

### Significance

Charge density waves (CDWs) are simple periodic reorganizations of charge in a crystal, and yet they are still poorly understood and continue to bear surprises. External perturbations, such as strain or pressure, can in principle push a CDW phase into a different ordering geometry. However, engineering this type of quantum criticality has been experimentally challenging. Here, we implement a simple method for straining bulk materials. By applying it to 2H-NbSe<sub>2</sub>, a prototypical CDW system studied for decades, we discover two dramatic strain-induced CDW phase transitions. Our atomic-scale spectroscopic imaging measurements, combined with theory, reveal the distinct roles of electrons and phonons in forming these emergent states, thus opening a window into the rich phenomenology of CDWs.

Author contributions: S.G., F.F., Z.W., J.v.W., and I.Z. designed research; S.G., F.F., R.S., H.Z., Z.R., B.R., and J.v.W. performed research; S.G., F.F., H.Z., S.B., and I.Z. analyzed data; and S.G., F.F., F.C., K.S.B., J.v.W., and I.Z. wrote the paper.

The authors declare no conflict of interest.

This article is a PNAS Direct Submission.

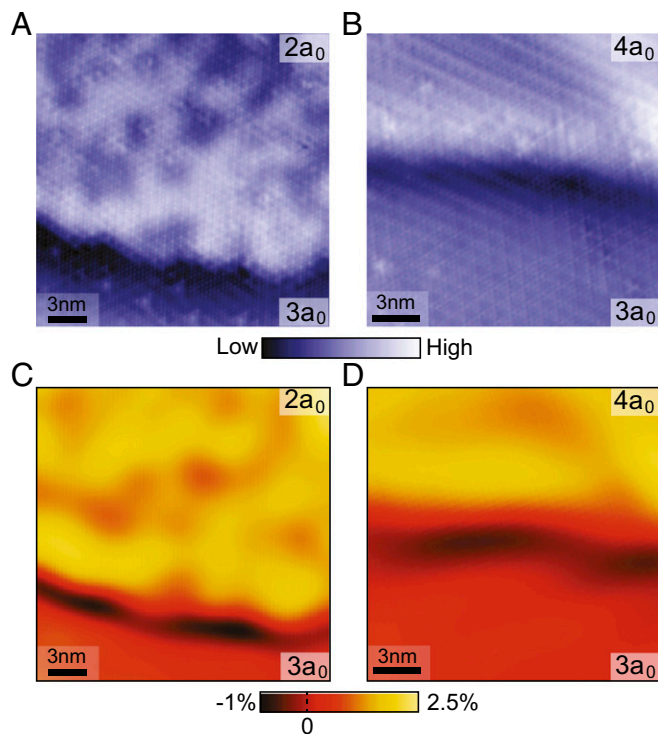
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This article contains supporting information online at [www.pnas.org/lookup/suppl/doi:10.1073/pnas.1718931115/-DCSupplemental](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1718931115/-DCSupplemental).

Published online June 18, 2018.



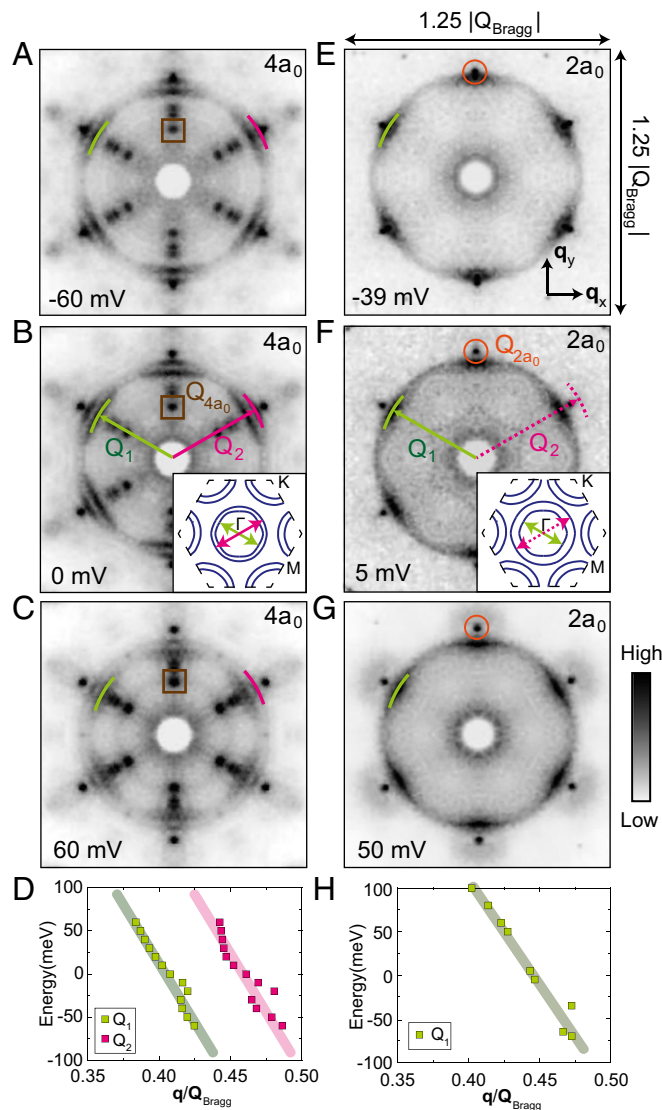


**Fig. 2.** Local strain mapping. (A and B) STM topographs and (C and D) biaxial (isotropic) strain maps of the atomically smooth boundaries between regions hosting different CDW phases. The biaxial strain maps have been calculated from the derivatives of the strain fields as  $(s_{xx} + s_{yy})/2$ , using the procedure described in *SI Appendix, section II*. The algorithm assumes that strain is zero in the CDW- $3a_0$  area, and calculates the relative strain with respect to it. Larger positive values represent tensile strain (stretching of the lattice). As can be seen, both CDW- $2a_0$  and CDW- $4a_0$  regions are characterized by tensile strain relative to the CDW- $3a_0$  area. STM setup conditions were (A)  $I_{set} = 350$  pA and  $V_{sample} = -70$  mV; (B)  $I_{set} = 200$  pA and  $V_{sample} = -100$  mV.

direct proof that in-plane tensile strain plays an important role in driving the observed charge ordering transitions.

To gain insight into the effects of strain on local electronic band structure in each region of the sample, we use QPI imaging, a method that applies 2D Fourier transforms (FTs) to the STM  $dI/dV$  maps to extract the electronic band dispersion. First, we focus on a large region of the sample hosting exclusively CDW- $4a_0$ , in which the FTs of the  $dI/dV$  maps show a circular QPI morphology (Fig. 3 A–C) with the strongest intensity along the  $\Gamma$ -M direction. Higher momentum-space resolution of our data compared with previous experiments on NbSe<sub>2</sub> hosting a CDW- $3a_0$  (10) allows us to disentangle two distinct QPI peaks  $Q_1$  and  $Q_2$  (Fig. 3B), which arise from backscattering within the two Fermi surface pockets concentric around  $\Gamma$  (Fig. 3B, *Inset* and *SI Appendix, section III*). By measuring the positions of these peaks as a function of energy, we can map the two bands crossing the Fermi level along the  $\Gamma$ -M direction (Fig. 3D). Interestingly, the electronic band structure is only slightly different compared with that of the well-characterized unstrained material (10) (*SI Appendix, section IV*), despite the dramatic changes in both the observed CDW wavelength and its geometry.

In the CDW- $2a_0$  region, we observe only the  $Q_1$  vector, while  $Q_2$  is notably absent in our measurable momentum range, in contrast to the CDW- $4a_0$  area (Fig. 3 E–G). This suggests a more prominent change in the band structure. Our strain measurements in Fig. 2 reveal that this region of the sample is under tensile strain, which would lead to a larger momentum-space separation of the pockets around  $\Gamma$  (Fig. 3F, *Inset*), owing to



**Fig. 3.** Electronic band-structure mapping using QPI imaging. FTs of  $dI/dV$  maps acquired at (A)  $-60$  mV, (B)  $0$  mV, and (C)  $60$  mV over a CDW- $4a_0$  region of the sample. (B, *Inset*) Schematic of the Fermi surface within the first Brillouin zone. (D) The dispersion of the QPI peaks as a function of energy along the  $\Gamma$ -M direction in the CDW- $4a_0$  region. FTs of  $dI/dV$  maps acquired at (E)  $-39$  mV, (F)  $5$  mV, and (G)  $50$  mV over the CDW- $2a_0$  region of the sample. (F, *Inset*) Schematic of the Fermi surface under small tensile strain, which is expected to move the Fermi surface pockets around  $\Gamma$  further apart. Only  $Q_1$  vector in E–G can be seen, while  $Q_2$  is notably absent. (H) The dispersion of the QPI peak as a function of energy along the  $\Gamma$ -M direction in the CDW- $2a_0$  region. QPI peak positions in D and H are determined using Gaussian peak fitting to a one-dimensional curve extracted along a line connecting the center of the FT and the atomic Bragg peak. QPI peaks and CDW peaks are denoted by the guides for the eye in panels A–C and E–G:  $Q_1$  (green line),  $Q_2$  (pink line),  $Q_{2a_0}$  (orange circle), and  $Q_{4a_0}$  (brown square). The center of all FTs has been artificially suppressed to emphasize other features. All FTs have been sixfold symmetrized to enhance signal to noise, and cropped to the same  $1.25|Q_{Bragg}|$  square size window. The region of the sample where the data in A–C were taken contains domains of CDW- $4a_0$  along only two lattice directions (*SI Appendix, Fig. S2A*). As CDW- $4a_0$  is intrinsically a unidirectional order, the sixfold symmetry of the  $Q_{4a_0}$  peak in A–C is an artifact of the symmetrization process. STM setup conditions: (A–C)  $I_{set} = 320$  pA,  $V_{sample} = -60$  mV, and  $V_{exc} = 10$  mV (zero-to-peak); (E)  $I_{set} = 200$  pA,  $V_{sample} = -39$  mV, and  $V_{exc} = 1$  mV; (F)  $I_{set} = 20$  pA,  $V_{sample} = 5$  mV, and  $V_{exc} = 1.5$  mV; (G)  $I_{set} = 300$  pA,  $V_{sample} = 50$  mV, and  $V_{exc} = 10$  mV.



technique can be applied to a range of other materials. For example,  $1T\text{-TiSe}_2$  could be strained to induce superconductivity (39) or novel CDW wavevectors and geometries in analogy to what we observe in  $2H\text{-NbSe}_2$ . Similarly, Fe-based superconductors could be strained, potentially using substrates with a TEC along a preferred direction (3), to create a rich playground to study the interplay of nematic order and superconductivity (40) within a single material using SI-STM.

## Methods

Single crystals of  $2H\text{-NbSe}_2$  were grown using vapor transport growth technique with iodine ( $I_2$ ) as the transport agent, and exhibit superconducting transition temperature  $T_c \sim 7$  K based on the onset of diamagnetic signal due to the Meissner effect in magnetization measurements (SI Appendix, section IX). Superconducting transition temperature remained approximately the same with  $T_c \sim 7$  K after the samples were strained and remeasured. Typical size of the single crystals used was  $\sim 2$  mm  $\times$  2 mm, with  $\sim 0.1$ -mm thickness before cleaving and  $\sim 0.01$ -mm to  $\sim 0.1$ -mm thickness postcleaving. Instead of attaching the  $2H\text{-NbSe}_2$  crystals directly to a metallic holder with TEC comparable to that of  $NbSe_2$ , as typically used in most STM experiments, we use conducting epoxy (EPO-TEK H20E) to glue the bottom of  $NbSe_2$  to silica ( $SiO_2$ ), a material with a vastly different TEC (Fig. 1A). Then, the  $NbSe_2$ /silica structure is attached to the STM sample holder and cooled down to  $\sim 4.5$  K (more information in SI Appendix, section IX). Based on the difference between TECs of  $NbSe_2$  and silica,  $NbSe_2$  is expected to stretch isotropically in-plane by  $\sim 0.15\%$ . As we demonstrate from STM topographs, the actual induced strain at the sample surface can be spatially inhomogeneous. To create a clean surface necessary for STM measurements, the samples were cleaved in ultra-high vacuum (UHV), and inserted into the STM head within minutes. We studied four different  $NbSe_2$  crystals glued on silica (five different surfaces as one sample was recleaved for the second approach). For each of these five, we approached the tip on several different points on the sample, which are typically tens of micrometers away from one another, and searched for different types of CDWs. We observed: all three

CDWs on two surfaces, just CDW- $2a_0$  and CDW- $4a_0$  on two other surfaces, and just CDW- $3a_0$  on one surface.

STM data were acquired using a Unisoku USM1300 STM at the base temperature of  $\sim 4.5$  K. All spectroscopic measurements have been taken using a standard lock-in technique at 915-Hz frequency and varying bias excitation as detailed in the figure legends. The STM tips used were homemade, chemically etched W tips annealed to bright-orange color in UHV. Tip quality has been evaluated on the surface of single-crystal  $Cu(111)$  before performing the measurements presented in this paper. The  $Cu(111)$  surface was cleaned by repeated cycles of heating and argon sputtering in UHV before it was inserted into the STM head.

To construct a model which captures experimental observations, we employ a tight-binding fit to the ARPES data for the two bands crossing the Fermi level (described in detail in refs. 26 and 35). The model assumes the two bands to be bonding and antibonding combinations of the two Nb  $d_{3z^2-r^2}$  orbitals. We include both biaxial and uniaxial in-plane strain by modifying the hopping integrals based on the assumption that overlap integrals are linearly dependent on displacement, with an equal prefactor for all overlaps. In modeling uniaxial strain, we assume that a tensile strain in one direction leads to a compressive strain in the perpendicular in-plane direction, conserving the volume of the unit cell. Then, we employ the Random Phase Approximation to calculate the phonon softening as seen in resonant inelastic X-ray scattering (20, 36). The CDW wavevector is identified as the first wavevector to soften to zero. By including nonlinear terms in a Landau free-energy expression we are able to reveal whether the CDW geometry consists of stripes (1Q) or triangles (3Q) (see SI Appendix, section VI for more details).

**ACKNOWLEDGMENTS.** We thank Peter Littlewood and Vidya Madhavan for helpful conversations. F.F. acknowledges support from a Lindemann Trust Fellowship of the English-Speaking Union, and the Astor Junior Research Fellowship of New College, Oxford. J.v.W. acknowledges support from a Vidi grant financed by the Netherlands Organisation for Scientific Research. K.S.B. appreciates support from the National Science Foundation under Grant NSF-DMR-1709987. Z.W. is supported by the Department of Energy Grant DE-FG02-99ER45747. I.Z. gratefully acknowledges the support from the National Science Foundation under Grant NSF-DMR-1654041 for the partial support of S.G., H.Z., B.R., and Z.R.

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