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Meeting-report

Unveiling a Large Supermodulation Underlying Electronic Anisotropy in Uranium Chalcogenide

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Uranium compounds often exhibit complex, strongly correlated electron behaviors (e.g., spin-triplet superconductor UTe_2 [1], hidden order compound URu_2Si_2 [2], ferromagnetic superconductor UCoGe [3] and many more). Here we show a new order parameter in a layered uranium chalcogenide: supermodulation (Fig. 1a). The supermodulation reported herein is a large periodic structural distortion that forms corrugated layers of crystals that resembles supermodulation of $Bi_2Sr_2CaCu_2O_8$ [4, 5]. Electronic anisotropy accompanies this structural distortion; the crystal is semiconducting along the supermodulation and metallic in orthogonal direction. In addition, in situ heating reveals the superlattice is robust at high temperatures up to 700°C (Fig. 1b i–iv). Determining the precise atomic displacement pattern of this supermodulation is key to understanding the electronic structure.

Plan-view high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM, Fig. 1c) shows sinusoidal intensity modulation along [100] direction. In selected area electron diffraction (SAED, Fig. 1d), the superlattice diffracts swift electrons as satellite peaks (yellow) decorating Bragg peaks (blue). Intensity distribution of the superlattice peaks indicates a structural ordering such as periodic lattice distortions (PLDs) in charge ordered systems [6–8].

Surprisingly, the cross-sectional investigations (Fig. 1e) reveal the superlattice order have strong out-of-plane modulation and forms corrugated structure. This is consistent with so-called 'supermodulation' found in cuprate superconductors [4, 5]. The strong interlayer coupling accompanies out-of-plane corrugation—apparent from antiphase out-of-plane order where sinusoidal modulation shows 180° phase shifts every layer. Cross-sectional SAED (Fig. 1f) further confirms strong long-range interlayer antiphase order from absence of in-plane fundamental superlattice peak at (q,0,0) (Fig. 1e, red dotted circle) and strong and sharp presence of out-of-plane superlattice peak at (q,0,½) (Fig. 1e, purple circle).

In addition, we report out-of-plane modulation amplitude of 35 pm and in-plane amplitude up to 15 pm with 90° phase shift between the in-plane and out-of-plane modulation. Supermodulation quantification is performed by nonlinear least squares fitting 2D gaussians on to each atomic column (Fig. 2e). Atomic displacement quantification requires a reference lattice free of distortions [9]. Here, we generate a synthetic reference image by Fourier demodulation [7, 10], where structural distortions are de-modulated by suppressing superlattice peak amplitudes in Fourier space (Fig. 2a–d). De-modulated reference image (Fig. 2d) clearly shows corrugation has been flattened out. The supermodulation amplitude reported herein is colossal compared to sub-10 pm distortions typically found in charge ordered materials [7, 10].

In summary, we revealed a strong supermodulation with extreme thermal resilience in layered uranium compound. The precise structure of the supermodulation was mapped using high resolution electron microscopy with advanced image analysis techniques [11].

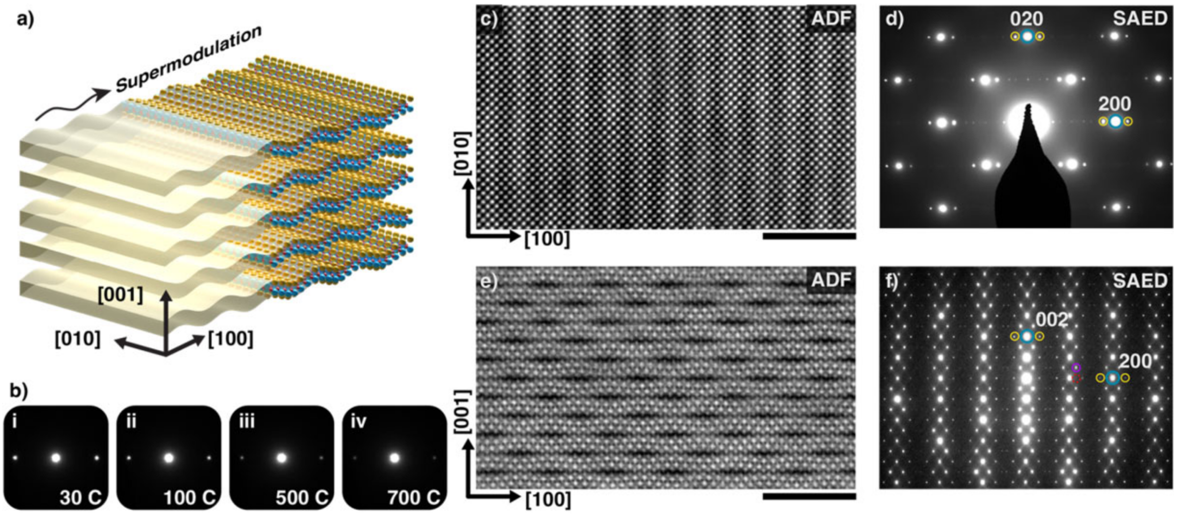

Fig. 1. Large 1D Supermodulation. a) Schematic illustration of layered, supermodulated uranium chalcogenide with out-of-plane antiphase ordering. b) In-situ SAED (i) 30°C, ii) 100°C, iii) 500°C, iv) 700°C) shows the supermodulation peaks remains sharp even at high temperatures. c) Plan-view ADF-STEM shows sinusoidal intensity modulation, further confirmed by SAED (d). e, f) Cross-sectional investigation reveals the intensity modulation in (c) is due to corrugated superstructure with out-of-plane antiphase ordering. Scale bars are 4 nm.

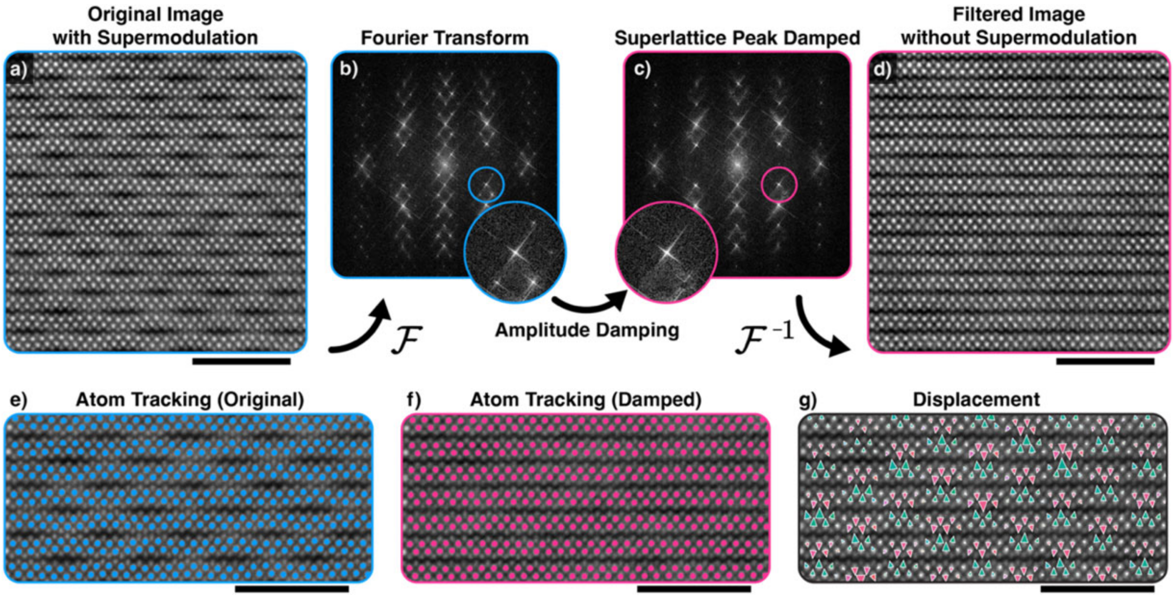

Fig. 2. Supermodulation quantification and de-modulation using Fourier damping. a) Original atomic resolution ADF-STEM shows strong supermodulation. b) Fourier transform renders supermodulation into superlattice peaks. c) Superlattice peak amplitude is suppressed to noise level without disturbing the complex phase. d) Inverse Fourier transform of c) reveals de-modulated crystal lattice. e) Fitting 2D gaussian to atomic columns to (e) original and (f) de-modulated image reveals the (g) quantitative displacement pattern. Scale bars are 4 nm.

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