Supporting information

Section 1. Fluctuation electron microscopy

Fluctuation electron microscopy experiments were designed to assess the presence of medium range order in thin lamellae extracted using the conventional focused ion beam (FIB) lift-out method from samples that were either as-deposited or after they had been annealed at 500 °C in air. The samples were thinned down until a thickness of 35 nm (as-deposited) and 45 nm (annealed at 500 °C) using a final gallium beam voltage of 5 kV. To ensure that gallium irradiation during sample preparation did not result in surface amorphisation and hence in decrease in a decrease of the fluctuation electron microscopy signal, 40 nm of ZTO was deposited onto 30 nm Si_3N_4 thin films. In a probe-Cs corrected FEI TITAN microscope operated at 300 kV, hundreds of nanobeam electron diffraction patterns (up to 1200 individual patterns per condition acquired per group of 100 patterns at different position of the sample) were recorded using different probe sizes, namely 1.3 nm (convergence semi-angle of 0.5 mrad), 2 nm (0.3 mrad) and 3 nm (0.2 mrad). The nanobeam diffraction patterns were filtered in energy using a slit width of 10 eV. Following the methodology presented in Ref.[1], a Mathematica code[2] was developed to i) find the center of each individual nanobeam electron diffraction pattern using a semi-automatic procedure (by maximizing the maximum intensity of the rotationally averaged diffraction intensities) to then ii) measure the corresponding rotationally averaged diffraction intensity (shown in Fig. 4e and f), before iii) computing the normalized variance for groups of 100 nanobeam diffraction patterns. The mean signal of the 12 variance curves is then reported with one standard deviation to the mean error bars.

The results, which are presented in Supp FIG *1*, demonstrate the absence of well-defined peaks in the variance and hence the absence of significant medium range order after deposition and after annealing at 500 °C in air, at least within the detection limit of the technique. The FIB sample preparation does not appear to modify significantly the variance measurements, as a similar signal is obtained when depositing ZTO directly onto Si_3N_4.
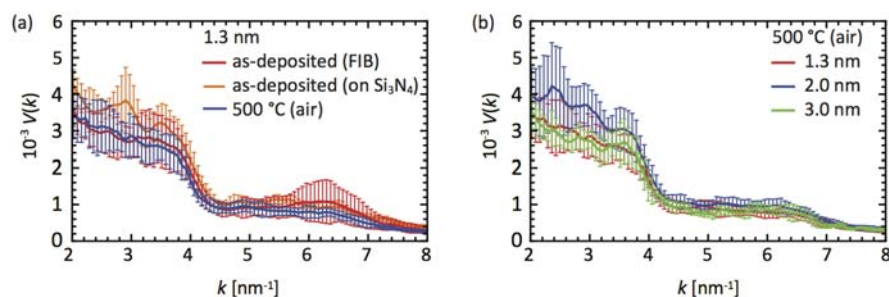

Supp FIG 1. (a) Variance of the diffracted intensities of the samples after deposition (prepared either by FIB or by directly depositing ZTO onto Si3N4 thin supports) and after annealing at 500 °C in air. This data set was acquired using a probe size of 1.3 nm and a convergence semi-angle of 0.5 mrad. (b) Variable resolution variance data of the FIB-prepared sample annealed at 500 °C acquired using various probe sizes, ranging from 1.3 to 3 nm.

Section 2. Temperature dependent Hall effect

According to Matthiessens Rule [3], the resistivity in presence of different scattering mechanisms is the sum of the resistivities that would affect the electrical behavior of the system if each alone was present, in terms of mobility this can be described as:

. To decouple the scattering mechanisms that depend of temperature, Hall effect measurements were performed in-situ while increasing the temperature from -190 °C to 45 °C. Measurements were done to as-deposited and samples annealed in the different atmospheres using a HMS-5000 equipment. The results provide an idea of which is the

dominant defect that limits mobility of the material. As shown if Supp FIG *2*, all samples show independence of μ and N_e with temperature, hence the scattering mechanism that limits mobility is independent of temperature in this range.
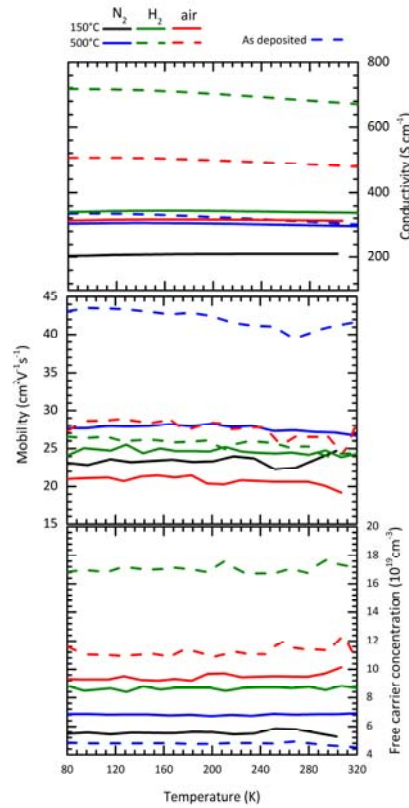

Supp FIG 2. Temperature dependence of μ and N_e for a-ZTO annealed in air, H2 and N2 independently. Samples were annealed at 150 °C and 500 °C for 30 min.

This results, summed to the lack of grain boundaries suggest that ionized and neutral impurity scattering are the dominant mechanisms limiting the mobility of a-ZTO.

Section 3. Fourier Transform infrared spectroscopy

FTIR experiments were done to calculate the value of the effective mass of electrons in the different samples. A Bruker Vertex 80 FTIR spectrometer was used to measure the reflectance in the 1-25 um region. The data resulting from this measurements is fitted to adjust to Drude model electron conductivity. Using as input the electron (optical) mobility, free carrier concentration and thickness it is possible to estimate the effective mass m*.
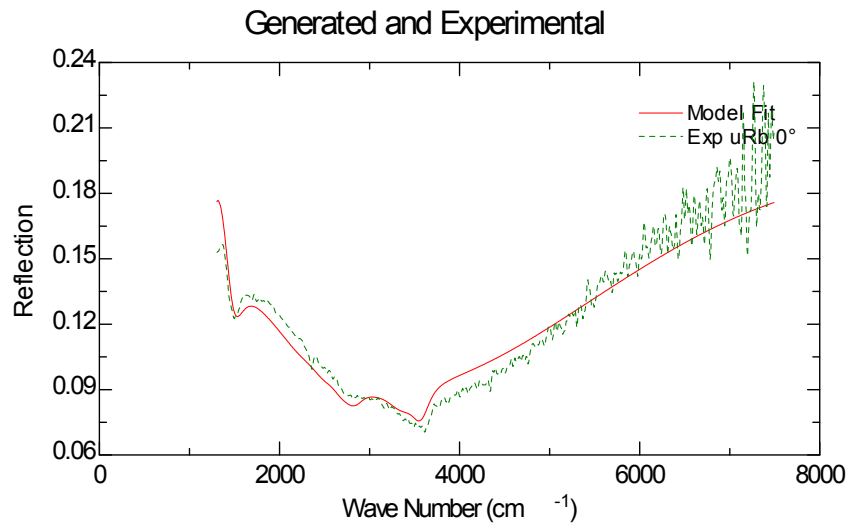
Supp FIG. 3. FTIR reflectance of as deposited ZTO. Experimental data in green and fitting of the Drude model in red.

As an example, in Supp FIG. 3 we show the FTIR reflectance of as deposited a-ZTO and the Drude model-fitting. For this particular sample, the calculated effective mass was 0.3 m_e.

[1] P.M. Voyles and D.A. Muller, Ultramicroscopy **93**, 147 (2002).
[2] Wolfram-Research-Inc, (2016).
[3] D.M. Neil Ashcroft, *Solid State Physics*, Internatio (Saunders College, Philadelphia, 1976).