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# Imaging Single Dopant Atoms and Nanoclusters in Highly n-type Bulk Si

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We report the imaging of individual Sb atoms still bonded to their crystalline Si host using atomic-resolution ADF-STEM. Samples with an Sb concentration of $9.35\times10^{20}$ cm$^{-3}$, measured by RBS, were prepared by low-temperature molecular beam epitaxy [1]. Figure 1 is an ADF-STEM image of a $\langle 110\rangle$ cross section of the sample. In the substrate region, where no Sb was deposited, all of the atomic columns are similar in intensity. In the region doped with Sb, some atomic columns are much brighter than the surrounding columns. These columns contain at least one Sb atom.

The intensity histograms of the images, shown in Figure 2, demonstrate that we have single Sb sensitivity. For random substitution of Sb atoms in the Si lattice, *s*, the number of Sb atoms in a column of *n* atoms is given by the binomial distribution,

$$P_{n,c}(s) = \frac{n!}{s!(n-s)!} c^{s} (1-c)^{n-s}, \qquad (1)$$

where $c$ is the Sb concentration. The sample thickness $n$ was determined by comparing the ratio of the $\langle 111\rangle$ to zero beam intensities in a CBED pattern to multislice simulations [2]. The intensity distribution of unoccupied columns is well-fit by a Gaussian, the width of which measures the residual effects of sample surface roughness. The occupied column distribution is fit by two Gaussians, the second at twice the excess intensity of the first. We identify the peaks as singly and doubly Sb-occupied columns, respectively. This is confirmed by the excellent agreement of the areas under the peaks with the predictions of Eq. 1, as shown in Table I. Our results are consistent with a random distribution of Sb on Si sites, imaged with single Sb sensitivity and ~100 % Sb detection efficiency. While single atoms have been previously imaged on surfaces [3], we believe this is the first time single atom sensitivity in the bulk has been quantitatively demonstrated. The spread of intensities associated with a single Sb atom in a column is due to dynamical probe channeling effects, as shown in Figure 3.

The key to single-atom detection is good sample preparation. We prepared cross-sections by double-wedge mechanical polishing using an Allied High Tech Inc. TechPrep/MultiPrep polishing wheel and diamond coated plastic lapping paper, finishing with a 0.02 μm colloidal silica polish. This is similar to tripod polishing [4], but the wheel provides more even pressure than polishing by hand, and the final silica polish is as much chemical as mechanical. The polishing results in samples with a highly damaged surface layer, which is quickly oxidized in air. The oxide is stripped off using a weak HF dip (30 s in 100:1 HF), leaving a sample with regions that are <25 A thick, have <1 A rms roughness, and no amorphized surface layer. Ion milling must be avoided entirely, as it produces a damage layer which is not removed by the etch [5].

References

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[5] We thank Partha Mitra and David Wittman for help with the image analysis, and Earl Kirkland for sharing unpublished image simulation codes.

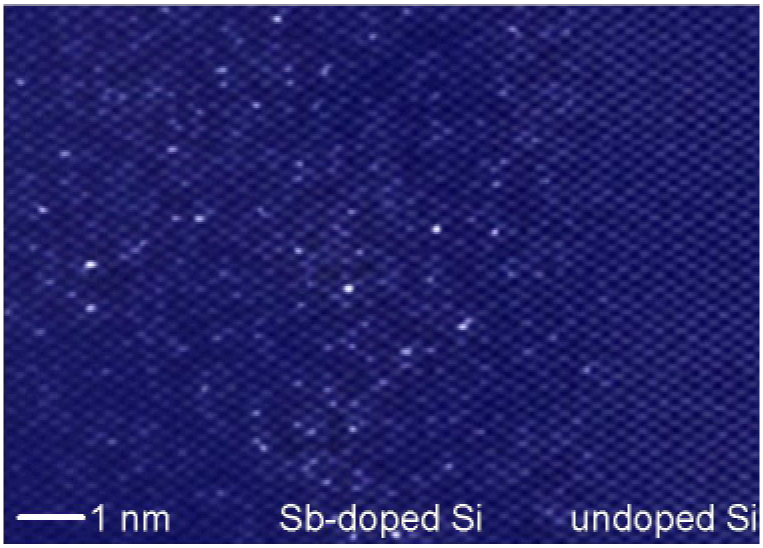

FIG. 1. ADF-STEM image of Sb-doped Si. The undoped region shows columns of uniform intensity. The brightest columns in the doped region contain at least one Sb atom. The image is smoothed and background subtracted.

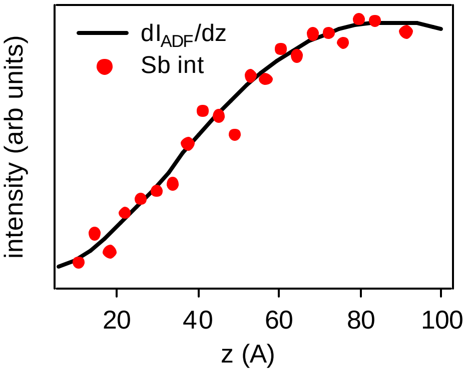

FIG. 3. Simulated effects of probe channeling on the intensity of one Sb in an Si column. The STEM intensity of a column as a function of Sb depth tracks the differential contribution to the image intensity from that plane of atoms, which varies with depth due to channeling.

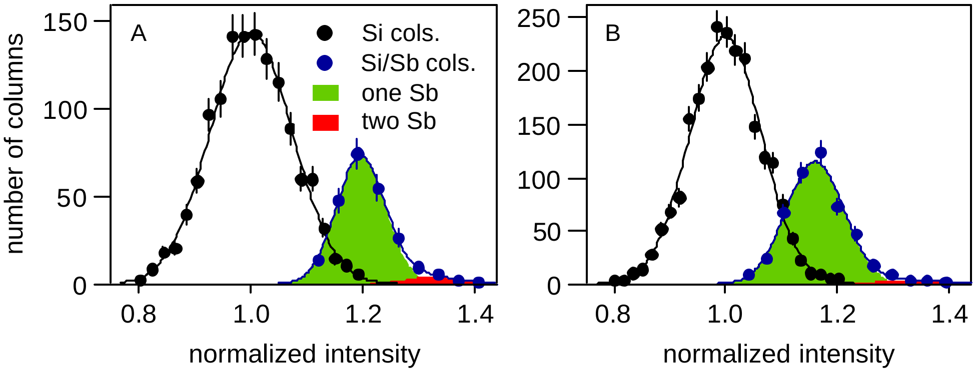

FIG. 2. Intensity histograms from two images. Intensities are normalized to the average unoccupied column intensity. Images A and B are 15 and 23 Å thick, respectively.

TABLE 1. Predicted and observed column occupancies. Predictions are from Equation 1. Observed values are the areas of the Gaussians in Figure 2. The agreement is excellent, indicating that the images show single-atom sensitivity.

| | Image A | | Image B | |
|---|---|---|---|---|
| # of Sb atoms | predicted | observed | predicted | observed |
| 0 | 1300 | 1300 (50) | 2234 | 2240 (70) |
| 1 | 223 | 230 (30) | 468 | 470 (40) |
| 2 | 17 | 15 (15) | 45 | 20 (20) |