Several prominent mechanisms for reduction in thermal conductivity have been shown in recent years to improve the figure of merit for thermoelectric materials. Such a mechanism is a hierarchical all-length-scale architecturing that recognizes the role of all microstructure elements, from atomic to nano to microscales, in reducing (lattice) thermal conductivity. In this context, there have been recent claims of remarkably low (lattice) thermal conductivity in Bi$_{0.5}$Sb$_{1.5}$Te$_3$ that are attributed to seemingly ordinary grain boundary dislocation networks. These high densities of dislocation networks in Bi$_{0.5}$Sb$_{1.5}$Te$_3$ were generated via unconventional materials processing with excess Te (which formed liquid phase, thereby facilitating sintering), followed by spark plasma sintering under pressure to squeeze out the liquid. We reproduced a practically identical microstructure, following practically identical processing strategies, but with noticeably different (higher) thermal conductivity than that claimed before. We show that the resultant microstructure is anisotropic, with notable difference of thermal and charge transport properties across and along two orthonormal directions, analogous to anisotropic crystals. Thus, we believe that grain boundary dislocation networks are not the primary cause of enhanced ZT through reduction in thermal conductivity. Instead, we can reproduce the purported high ZT through a favorable but impractical and incorrect combination of thermal conductivity measured along the pressing direction of anisotropy while charge transport measured in the direction perpendicular to the anisotropic direction. We believe that our work underscores the need for consistency in charge and thermal transport measurements for unified and verifiable measurements of thermoelectric (and related) properties and phenomena.
The impact of extra Te on the orientation of grains, the microstructure, and eventually the thermoelectric properties remains substantially obscure. In addition, there are doubts whether dense dislocation scattering can be as effective in lowering the lattice thermal conductivity to these low levels as claimed given that the well-known Bi$_{0.5}$Sb$_{1.5}$Te$_3$ structure already has a very short phonon mean-free path. Therefore, it is critical to investigate in detail the influence of the excess Te content on the microstructure and composition of melt-spun Bi$_{0.5}$Sb$_{1.5}$Te$_3$ ribbons and the impact of Te melting and its expulsion during the SPS process on the composition, microstructure, and thermoelectric properties of sintered samples. The results show that the extra Te distributes homogeneously at the boundaries of melt-spun ribbons of Bi$_{0.5}$Sb$_{1.5}$Te$_3$. Subsequent melting and extraction of Te during SPS promotes recrystallization and directional realignment of grains to improve the orientation factor $F$ and the electrical conductivity perpendicular to the pressure direction (18). The obtained samples have a dense array of dislocations in the grain boundaries, as described by Kim et al. (16). However, the lattice thermal conductivity of these samples at 300 K, despite the large amount of dislocations and defects generated during the SPS process, shows a similar value of 0.65 W m$^{-1}$ K$^{-1}$ (17) and is much higher than 0.33 W m$^{-1}$ K$^{-1}$ reported by Kim et al. (16). As a result, the highest ZT of our samples is comparable to that measured by Jo et al. (17) but well below the value reported by Kim et al. (16). The discrepancy arises despite us following as closely as possible the procedures and techniques reported by Kim et al. (16), and our structural characterization indicates that we have obtained the samples as described by Kim et al. (16).

RESULTS AND DISCUSSION
Phase and structure characterization
Powder x-ray diffraction (XRD) patterns of melt-spun ribbons of Bi$_{0.5}$Sb$_{1.5}$Te$_3$ with $x$ wt % excess of Te ($x$ = 5, 10, 15, 20, 25) are presented in fig. S1A. The expanded view of XRD in the range of 20° to 35° in fig. S1B shows that the main Bragg peak of elemental Te is detected at 27° to 28° when $x \geq 5$. The intensity of this peak increases notably with increasing content of Te. This indicates that Te is present in its elemental form and segregates during the rapid solidification of the melt-spun ribbons. The crystal lattice of Bi$_{0.5}$Sb$_{1.5}$Te$_3$ does not accommodate any overstoichiometric amount of Te because it is essentially a line compound. To further analyze the form and distribution of elemental Te in the ribbons, their microstructure containing various different excess amounts of Te was characterized in detail. The contact surface (the ribbon in direct contact with the rotating copper drum as the molten charge rapidly solidifies) and the free surface of ribbons containing 0 and 20 wt % of Te are shown in fig. S2A. The cross section of a ribbon with the thickness of 5 μm is shown in fig. S2B. The grain size increases gradually from the contact surface to the free surface because of the cooling rate gradient, where the contact surface experiences a much faster rate of cooling than the free surface. Consequently, the grain growth on the contact surface is significantly inhibited.

Field-emission scanning electron microscopy (FESEM) images of the contact surface of ribbons with 0 and 25 wt % excess of Te are shown in fig. S2 (C and D, respectively). In both cases, the grains are fine with the size of about 100 to 300 nm, but it is difficult to tell the difference between the two samples because of the limit of detection. FESEM images of the free surface of ribbons with 0, 5, 15, and 25 wt % excess of Te are shown in Fig. 1 (A to D, respectively). The free surface has a typical dendritic structure with the size of dendrites of about 100 to 200 nm in width and 2 μm in length. Grain boundaries in ribbons having no Te excess are smooth and clear and with no sign of the presence of any secondary phases. In contrast, grain boundaries of ribbons that have some excess of Te are blurred, and the color tone near the boundaries turns deeper than that of the matrix, as marked by red arrows. The difference in contrast increases with the increasing Te content. The composition analysis shows that grain boundaries of ribbons containing excess Te are Te-rich zones. Results of energy-dispersive spectroscopy performed on the free surface of a ribbon with 25 wt % excess of Te are shown in fig. S3. Here, spots 1 and 2 represent grains of the matrix having the main component of Bi$_{0.5}$Sb$_{1.5}$Te$_3$,
spots 3 and 4 represent a typical $\text{Bi}_0.5\text{Sb}_1.5\text{Te}_3$-Te eutectic structure, and spots 5 and 6 depict pure elemental Te. This confirms that any Te excess has segregated at the boundaries of the $\text{Bi}_0.5\text{Sb}_1.5\text{Te}_3$ matrix during melt spinning, in accordance with observations described in the literature (16).

Displacement motions of a puncher die during the SPS process of samples having different content of Te are shown graphically in Fig. 1E. Two large displacements are observed: The first one occurs at 260°C and corresponds to a vanishing of big pores between the grains and densification of the grains. The other one at 410°C reflects melting of the excess Te ($T_m = 449°C$ for Te) and its ejection from the interior of the sample. This temperature corresponds to the eutectic point of BiSbTe and Te mixture, which is slightly lower than the melting temperature of Te. In addition, the real temperature of the BiSbTe sample is usually higher than the temperature detected by the thermocouple inserted in the die. The above puncher die motion is to be contrasted with only one notable puncher die displacement observed for a pure sample of $\text{Bi}_0.5\text{Sb}_1.5\text{Te}_3$ at 260°C that corresponds to the densification of grains in the structure. Moreover, in samples containing excess of Te, the absolute size of the displacement and its slope notably increase with the increasing Te excess.

Powder XRD patterns of melt-spun ribbons, sintered bulk, and ejected material during the SPS process are shown in Fig. 1F. Before SPS, both $\text{Bi}_0.5\text{Sb}_1.5\text{Te}_3$ and elemental Te were detected in ribbons containing excess amounts of Te. After SPS, only $\text{Bi}_0.5\text{Sb}_1.5\text{Te}_3$ is detected in the sintered ingot because all excess Te was “squeezed out” during the SPS process, as described by Kim et al. (16). XRD of this squeezed-out material indicates that it is mostly Te with a small amount of $\text{Bi}_0.5\text{Sb}_1.5\text{Te}_3$, as confirmed also by images of back-scattered electrons. The actual graphite dies containing samples with 5, 15, and 25 wt % of excess Te after SPS are shown in Fig. 1 (G to I). The amount of ejected material increases with the increasing content of excess Te. More characterization of the resulting bulk $\text{Bi}_0.5\text{Sb}_1.5\text{Te}_3$ and Te samples obtained after the squeeze-out process is given in the Supplementary Materials.

XRD patterns collected on planes of samples perpendicular to the pressing direction during SPS are presented in Fig. 2A. The intensity of peaks corresponding to planes of (006), (009), and (0015) increases with the increasing Te content, and this indicates an enhanced orientation. The relationship between the orientation factor $F$ and the content of extra Te is shown in Fig. 2B. Excess Te in the structure seems to enhance texturing (orientation) in these samples. The values of $F$ for samples with 0 and 25 wt % excess of Te are 0 (no orientation) and 0.17 (high orientation), respectively.

To assess the influence of excess Te on the orientation and microstructure of sintered samples, two typical samples were selected for detailed microstructural characterization. FESEM images of a freshly fractured surface of a sample having no Te excess are presented in Fig. 2 (C and D). Figure 2 (E and F) shows similar images for a sample having 25 wt % excess of Te. The orientation of the surfaces with respect to the pressing direction during SPS is shown in the upper right corners of the above figures. Lamellar structures with a typical size of about 10 to 50 μm are apparent. The grains of samples having no excess Te are randomly arranged in all directions, as depicted in Fig. 2 (C and D). The morphology, however, quite different in samples that have some excess of Te. For instance, the sample with the excess amount of 25 wt % of Te, shown in Fig. 2E, has a distinctly layered structure with blocks of about 30 μm in thickness that extend along the direction perpendicular to the pressure applied during SPS. The microstructure of this sample observed in the direction parallel to the pressure is shown in Fig. 2F. Thus, on the basis of the different microstructures that develop in the perpendicular and parallel directions, it appears that liquid Te during the sintering process promotes plane slipping and recrystallization, making a-β planes extend perpendicularly and stack parallel to the pressing direction. The resulting much enhanced orientation is quantified by an increase in the orientation factor $F$ that rises from 0 (no excess Te) to 0.17 (25 wt % Te excess). This is a key finding because it affects the directions of thermal transport measurements, which will be discussed below.

High-resolution transmission electron microscopy (HRTEM) was carried out to look more closely at the microstructure of the sample made from melt-spin ribbons containing 25 wt % excess of Te. A low-magnification TEM image, including an inset displaying selected-area electron diffraction (SAED) corresponding to the $\text{Bi}_0.5\text{Sb}_1.5\text{Te}_3$ matrix, is shown in Fig. 3A. Numerous pleated structures (overlaps of two grains in the boundaries) with the width of 40 nm are embedded at grain boundaries (marked with red lines). Images of fast Fourier transforms (FFTs) of the numbered regions in Fig. 3B reveal that the grain boundaries tend to be polycrystalline. Figure 3C shows a Moiré pattern of two adjacent crystalline grains. A high density of dislocations is found in this region, as shown in the inverse FFT (IFFT) in Fig. 3D. Rearrangement of grains that takes place during the application

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of high pressure expedites the creation of dislocations at the grain boundaries. In general, we observe that the microstructure of our sample is very similar to that reported by Kim et al. (16).

Thermoelectric properties

Now that we have established that oriented samples with a high density of dislocations at the grain boundaries have been obtained, we can examine in detail their electrical and thermal transport properties. Figure 4 (A and B) presents the temperature dependence of the total thermal conductivity measured perpendicular ($k_{||}$) and parallel ($k_{\perp}$) to the pressing direction for samples sintered from melt-spun ribbons of composition $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ with various excess content of Te. The perpendicular thermal conductivity $k_{\perp}$ increases slightly with increasing excess of Te in the whole temperature range. The minimum value of $k_{\perp}$ is about 0.94 and 1.03 Wm$^{-1}$K$^{-1}$ for samples with 0 and 25 wt % excess of Te, respectively, and both values were reached at 350 K. In samples prepared from melt-spun ribbons having excess content of Te, the perpendicular thermal conductivity is enhanced on two counts. The increased perpendicular electrical conductivity contributes a larger charge carrier thermal conductivity term according to the Wiedemann-Franz relation, and the enhanced a-b plane orientation leads to an increased lattice thermal conductivity because the phonon transport is more facile along the a-b plane than along the c axis. Therefore, our measurements of the thermal conductivity along the perpendicular SPS pressing direction on samples having a comparable Te excess are much higher than the 0.65 Wm$^{-1}$K$^{-1}$, the value reported by Kim et al. (16) (about) on the sample with a comparable Te excess. In contrast, the parallel thermal conductivity $k_{||}$, shown in Fig. 4B, is greatly reduced as the excess amount of Te increases. For the sample with 25 wt % excess of Te, the value of about 0.74 Wm$^{-1}$K$^{-1}$ is reached, close to the value reported by Kim et al. (16), where the sample was measured perpendicular to the pressing direction of SPS. As the temperature increases, the thermal conductivity of all samples, and for both perpendicular and parallel orientations, first decreases, reaches minimum near 350 K, and then increases. The turn-about point coincides with the temperature where intrinsic excitations set in, giving rise to the well-known bipolar diffusion. Figure 4D gives a relationship between the measured anisotropy ratio $k_{||}/k_{\perp}$ and the orientation factor $F$ for the sample with $x$ wt % excess of Te. The symbols represent the data of this paper, and the line stands for the data of Shen et al. (18). Our data fall satisfactorily on the line.

The lattice thermal conductivities are calculated by the Wiedemann-Franz formula $\kappa_l = \kappa_e - L\sigma T$, where $\sigma$ is the electrical conductivity, $L$ is the Lorenz number, and $T$ is the absolute temperature (see Materials and Methods for details). The obtained lattice thermal conductivity in the perpendicular direction is shown in Fig. 4C. The inset shows data of Kim et al. (16). The lattice thermal conductivity varies little with the increasing excess amount of Te, and the minimum value reaches $\approx$0.65 Wm$^{-1}$K$^{-1}$ for the sample with 25 wt % excess of Te. This lattice thermal conductivity value is almost twice as large as the value reported by Kim et al. (16) for the sample prepared with a comparable excess of Te.

Figure 5 depicts the temperature-dependent electrical conductivity, Seebeck coefficient, and power factor of all samples with different excess Te measured perpendicular to (designated with “$\perp$”) and parallel to (designated with “$||$”) the pressing direction to document the effect of texturing. Figure 5A shows clearly that the perpendicular electrical conductivity on samples having a comparable Te excess are much higher than the 0.65 Wm$^{-1}$K$^{-1}$, the value reported by Kim et al. (16) (about) on the sample with a comparable Te excess. In contrast, the parallel thermal conductivity $k_{||}$, shown in Fig. 4B, is greatly reduced as the excess amount of Te increases. For the sample with 25 wt % excess of Te, the value of about 0.74 Wm$^{-1}$K$^{-1}$ is reached, close to the value reported by Kim et al. (16), where the sample was measured perpendicular to the pressing direction of SPS. As the temperature increases, the thermal conductivity of all samples, and for both perpendicular and parallel orientations, first decreases, reaches minimum near 350 K, and then increases. The turn-about point coincides with the temperature where intrinsic excitations set in, giving rise to the well-known bipolar diffusion. Figure 4D gives a relationship between the measured anisotropy ratio $k_{||}/k_{\perp}$ and the orientation factor $F$ for the sample with $x$ wt % excess of Te. The symbols represent the data of this paper, and the line stands for the data of Shen et al. (18). Our data fall satisfactorily on the line.

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conductivity rises with increasing Te content, whereas the parallel electrical conductivity decreases with the increasing Te content. At room temperature, the electrical conductivity for a sample with 25 wt% excess of Te measured in the direction perpendicular to the pressing direction is distinctly increased. Liquid Te here apparently plays a role of an efficient lubrication agent (for example, lubricant and flux) facilitating the directional alignment that increases with the increasing excess Te. Because of the enhanced electrical conductivity and the only mildly decreased Seebeck coefficient, the power factor PF in the perpendicular direction increases with the increasing excess Te and reaches 4.2 mW m⁻¹ K⁻¹ at room temperature for the sample with 25 wt% excess of Te, some 20% higher than for Bi₀.₅Sb₁.₅Te₃ itself. In contrast, the more significant decrease in electrical conductivity along the parallel direction with increasing excess Te, despite the increase of Seebeck coefficient, leads to reduction of the power factor PF from 3.7 mW m⁻¹ K⁻¹ for pure Bi₀.₅Sb₁.₅Te₃ to 3.1 mW m⁻¹ K⁻¹ for the sample with 25 wt% excess of Te.

The values of ZT are shown in Fig. 5D. Fabricating Bi₀.₅Sb₁.₅Te₃ samples from melt-spinning ribbons with excess of Te seems to slightly benefit the thermoelectric performance as documented by a ZT value of 1.24 achieved at 350 K with a sample containing 20 wt% excess of Te (in the perpendicular direction). This is about a 15% improvement over the sample processed by the exact same route but with no excess of Te. Nevertheless, this value falls far short of ZT = 1.86 reported in the literature (16). However, if one would naively assume that, following the SPS processing, the samples are isotropic structures and combine thermal conductivity measurements parallel to the pressing direction with electrical conductivity and the Seebeck coefficient measurements obtained perpendicular to the pressing direction, then ZT values of 1.7 at 350 K with our samples can be obtained, as shown by open triangles and open hexagons in Fig. 5D.

**CONCLUSION**

To conclude, introducing excess amounts of Te to a stoichiometric charge of Bi₀.₅Sb₁.₅Te₃ during the melt-spinning process has considerable impact on the microstructure, composition, and transport properties of SPS structures. Our results show that the excess Te distributes homogeneously at the boundaries of Bi₀.₅Sb₁.₅Te₃ grains following rapid solidification of the melt-spinning ribbons. Melting and squeezing out the excess Te during the SPS process promotes recrystallization and reorientation of grains because they are forced to fill empty spaces left by the expelled Te liquid. This gives rise to dense dislocation arrays embedded in grain boundaries, as reported by Kim et al. (16), as well as to a partial directional alignment of Bi₀.₅Sb₁.₅Te₃ grains in all samples prepared with excess amounts of Te. It seems as if liquid Te plays a role of an efficient lubrication agent (for example, lubricant and flux) facilitating the directional alignment that increases with the increasing content of excess Te. Whereas the SPS-processed samples prepared from melt-spinning ribbons without excess of Te turned out to be essentially isotropic structures with a minimal degree of transport anisotropy, the structures sintered from ribbons containing excess of Te acquired distinct structural anisotropy reflected in the anisotropy of their transport parameters. The directional alignment makes it imperative to take into careful account the direction in which the
transport properties are measured with respect to the pressing direction applied during the SPS process. Power factors obtained for the direction perpendicular to the pressing direction for our samples that contained some excess of Te are very high and comparable to the power factor reported by Kim et al. (16). With the Seebeck coefficients showing a rather small directional effect, the enhanced perpendicular electrical conductivity as the excess of Te increases results in progressively increasing power factors. The induced grain alignment following SPS has also a significant impact on heat transport. Unfortunately, here, the effect is less favorable from the perspective of thermoelectricity. The perpendicular lattice thermal conductivity, although rather small at 0.65 W m⁻¹ K⁻¹ for a sample with 25 wt % excess of Te, remains significantly higher than claimed by Kim et al. (16). The anisotropy in the heat transport of structures fabricated from melt-spun ribbons that contained large excess of Te is substantial. Measured in the parallel orientation, the thermal conductivity is much lower (0.74 W m⁻¹ K⁻¹ for the sample with 25 wt % excess of Te) because it is more difficult for phonons to propagate across the van der Waals gaps of the crystal structure because the grains of Bi₀.₅Sb₁.₅Te₃ are partly c axis–oriented. The maximum value of ZT that we have measured reached 1.24 at 350 K for samples with 20 and 25 wt % excess of Te, representing about 15% increase over the figure of merit of identically prepared Bi₀.₅Sb₁.₅Te₃ having no excess of Te.

MATERIALS AND METHODS
Sample preparation and measurement
High-purity Bi, Sb, and Te granules were weighed according to the stoichiometric composition Bi₀.₅Sb₁.₅Te₃ and loaded into a vacuum-sealed quartz tube with the diameter of 15 mm. The charge was melted and homogeneously mixed in a rocking furnace for 10 hours at 1073 K and then furnace–cooled down to room temperature. The obtained ingots were ground into a fine powder, sieved through a 200 size mesh, mixed with the excess amount of Bi powder (0, 5, 10, 15, 20, and 25 wt %), and finally cold-pressed into pellets under a pressure of 10 MPa.

Each obtained pellet was put into a quartz tube with a 0.3-mm-diameter nozzle at the bottom, and the tube was mounted on a melt-spinning apparatus. The chamber was filled with Ar (0.04 MPa) to protect the ingot from oxidation while the ingot was melted by radio frequency heating. The melt was ejected under a pressure pulse of 40 kPa of Ar onto a copper roller rotating with a linear speed of 45 m s⁻¹. Thin ribbons (5 to 10 μm in thickness, 2 mm in width, and 5 to 10 mm in length) were obtained through this rapid solidification process. The melt-spun ribbons were hand-ground into fine powders and compacted using SPS at 480°C for 3 min under 70 MPa. During the SPS process, the liquefied excess Te was expelled from the structure into a graphite die. The fully condensed bulks with the relative density over 99% were obtained in sizes of 12.7 mm × 13 mm. The entire Synthesis process, including melt spinning and SPS processing, is identical to that used by Kim et al. (16).

Phase composition of bulk samples was identified by powder XRD analysis (PANalytical-Empyrean; Cu Kα). Composition and morphology of the bulk samples was investigated by electron probe microanalysis (EPMA) with wavelength-dispersive spectroscopy (JXA-8230, JEOL), FESEM (Hitachi SU-8020), and HRTEM (JEM-2100F, JEOL) equipped with an energy-dispersive spectroscope to determine the actual composition.

The electrical conductivity σ and the Seebeck coefficient α were measured simultaneously by a standard four-probe method in the range of 300 to 473 K using an Ulvac-Riko ZEM-3 system. The total thermal conductivity was calculated from $κ = DC_Bρ$, where $D$ is the thermal diffusivity obtained by the laser flash method (LFA-457, Netzsch), $C_B$ is the specific heat measured by a differential scanning calorimeter (DSC Q20, TA Instruments), and $ρ$ is the density determined by the method of Archimedes. The room-temperature carrier concentration and the Hall mobility were measured using a Physical Property Measurement System (PPMS, Quantum Design). All tests were performed perpendicular to the pressing direction. The orientation factor $F$ was calculated according to the following equations (18, 28):
where $e$ is the elemental charge and $\alpha$ is the Seebeck coefficient. The calculated Lorenz numbers at temperatures of measurement vary within the range from $1.5 \times 10^{-8}$ to $1.6 \times 10^{-8}$ $\text{W} \text{ohms}^{-1} \text{K}^{-2}$ for all samples with different excess content of Te.

**Density-of-states effective mass calculation**

The effective mass was calculated assuming a single parabolic band model and the dominance of acoustic phonon scattering. The effective mass is calculated from (1, 20)

$$
\alpha = \frac{\hbar}{e} \left[ n_F - \frac{(r + 3/2)F_{r+1/2}(\eta_F)}{(r + 5/2)F_{r+3/2}(\eta_F)} \right]
$$

$$
F_i(\eta_F) = \int_0^\infty \frac{x \sin x}{1 + \exp(x - \eta_F)}
$$

$$
\eta_F = E_i / (k_B T)
$$

$$
\eta_F = 4\pi^2(2k_B T m^*)^{3/2} F_{1/2}(\eta_F)
$$

where $F_i(\eta_F)$ is the Fermi-Dirac integral, $\eta_F$ is the reduced Fermi level, $k_B$ is the Boltzmann constant, $h$ is the Planck constant, and $r$ is the scattering ($r = -3/2$ for acoustic scattering).

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/4/6/eaar5606/DC1

fig. S1. Powder XRD patterns of melt-spun ribbons.

fig. S2. FESEM images of two surfaces of ribbons.

fig. S3. EDS of the free surface of ribbon containing 25 wt % extra Te.

fig. S4. BSE images of SPS samples and ejection material.

fig. S5. Carrier concentration and mobility of samples.

table S1. Room-temperature physical parameters of samples.

**REFERENCES AND NOTES**


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Data and materials availability: All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Materials. Additional data related to this paper may be requested from the authors.

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Thermal conductivity in $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_{3+x}$ and the role of dense dislocation arrays at grain boundaries

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