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ULTRA-LOW THERMAL CONDUCTIVITIES IN LARGE-AREA $Si_{0.8}Ge_{0.2}$ NANOMESHES GROWN BY DC-SPUTTERING FOR THERMOELECTRIC APPLICATIONS

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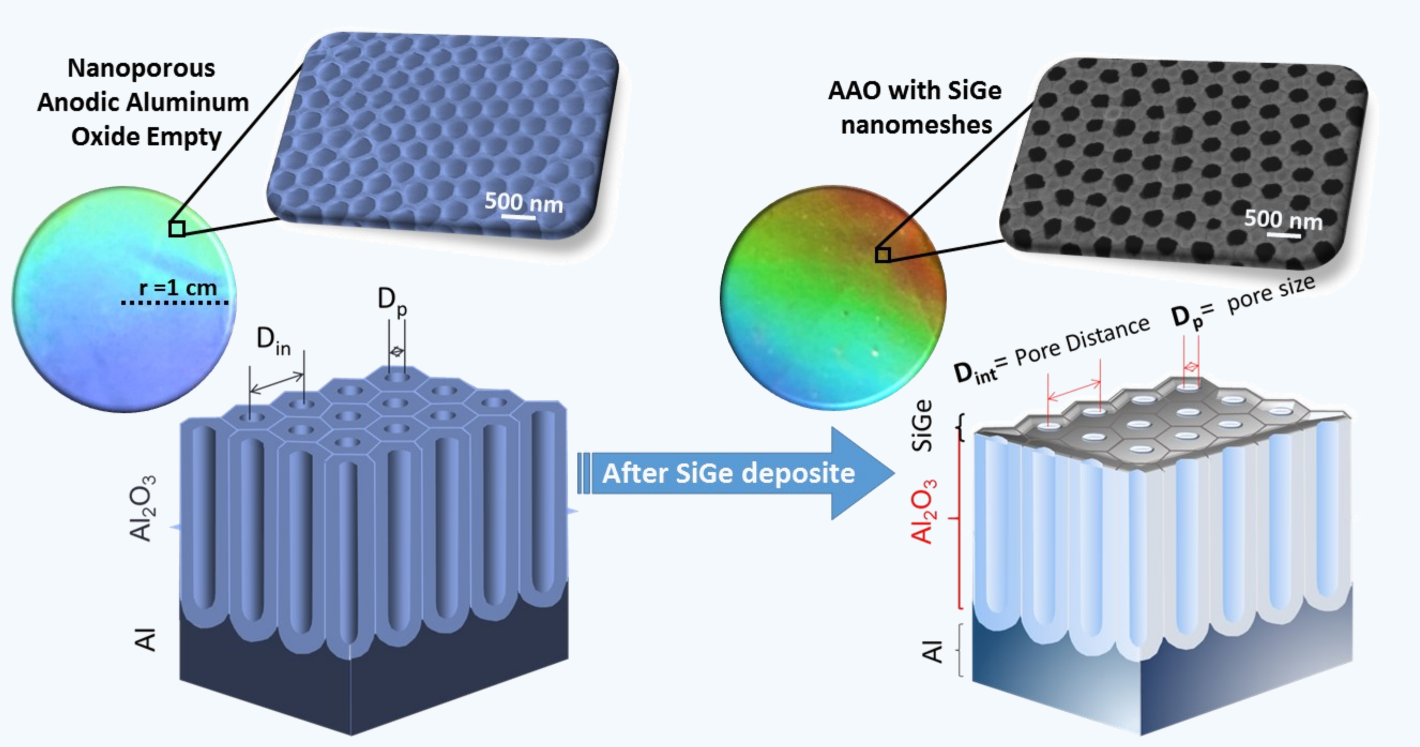
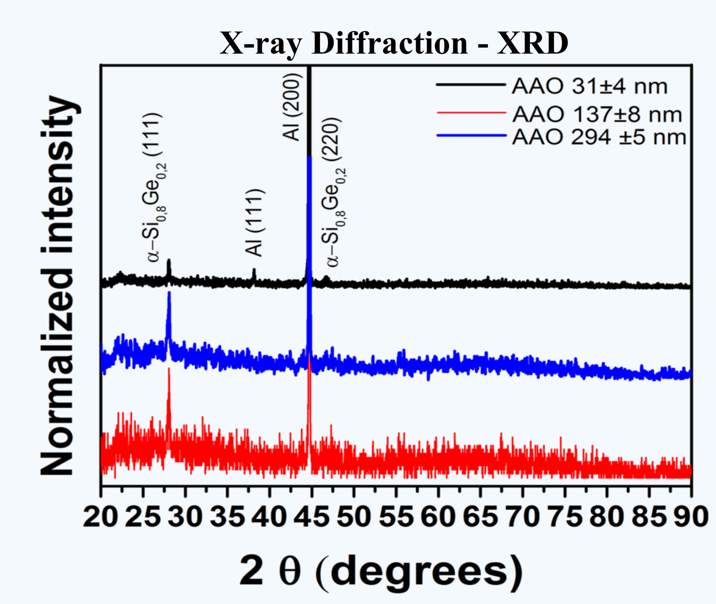
In this work, we measure the structural, morphological, compositional, thermal and thermoelectric properties of large-area $Si_{0.8}Ge_{0.2}$ nanomeshed films manufactured by DC-sputtering of $Si_{0.8}Ge_{0.2}$ on highly ordered porous alumina matrices [1]. The $Si_{0.8}Ge_{0.2}$ film replicated the porous alumina structure resulting in nano-meshed films. Very good control of the nanomesh geometrical features (*pore diameter, pitch, neck*) was achieved through the alumina template, with pore diameters ranging from 294 ± 5nm down to 31 ± 4 nm. The method that we developed is able to provide large areas of nano-meshes in a simple and reproducible way, being easily scalable for industrial applications. Most importantly, the thermal conductivity of the films was reduced as the diameter of the porous became smaller to values that varied from $\kappa = 1.54 \pm 0.27$ W $K^{-1}m^{-1}$, down to the ultra-low $\kappa = 0.55 \pm 0.10$ W $K^{-1}m^{-1}$ value. The latter is well below the amorphous limit, while the Seebeck coefficient and electrical conductivity of the material were retained. These properties, together with our large area fabrication approach, can provide an important route towards achieving high conversion efficiency, large area, and high scalable thermoelectric materials. Using this approach, it is possible to control thermal transport of these films through nano-engineering.

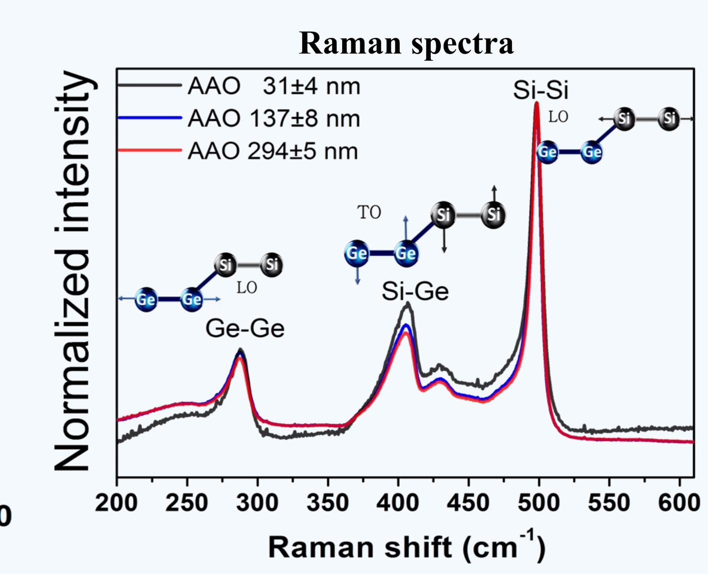
Sketch of the manufacturing process on a porous alumina template

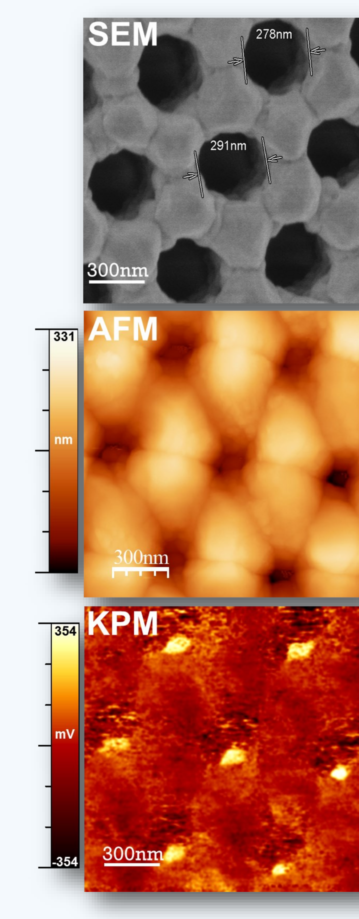


The sputtered $Si_{0.8}Ge_{0.2}$ films replicated the porous alumina matrices (AAO) highly oriented structure, resulting in the nano-meshed films with different pore sizes. This fabrication process allows growing large areas of $Si_{0.8}Ge_{0.2}$ nano-meshed films in a simple and reliable way, and can be easily industrially scalable.

Structural and compositional characterization

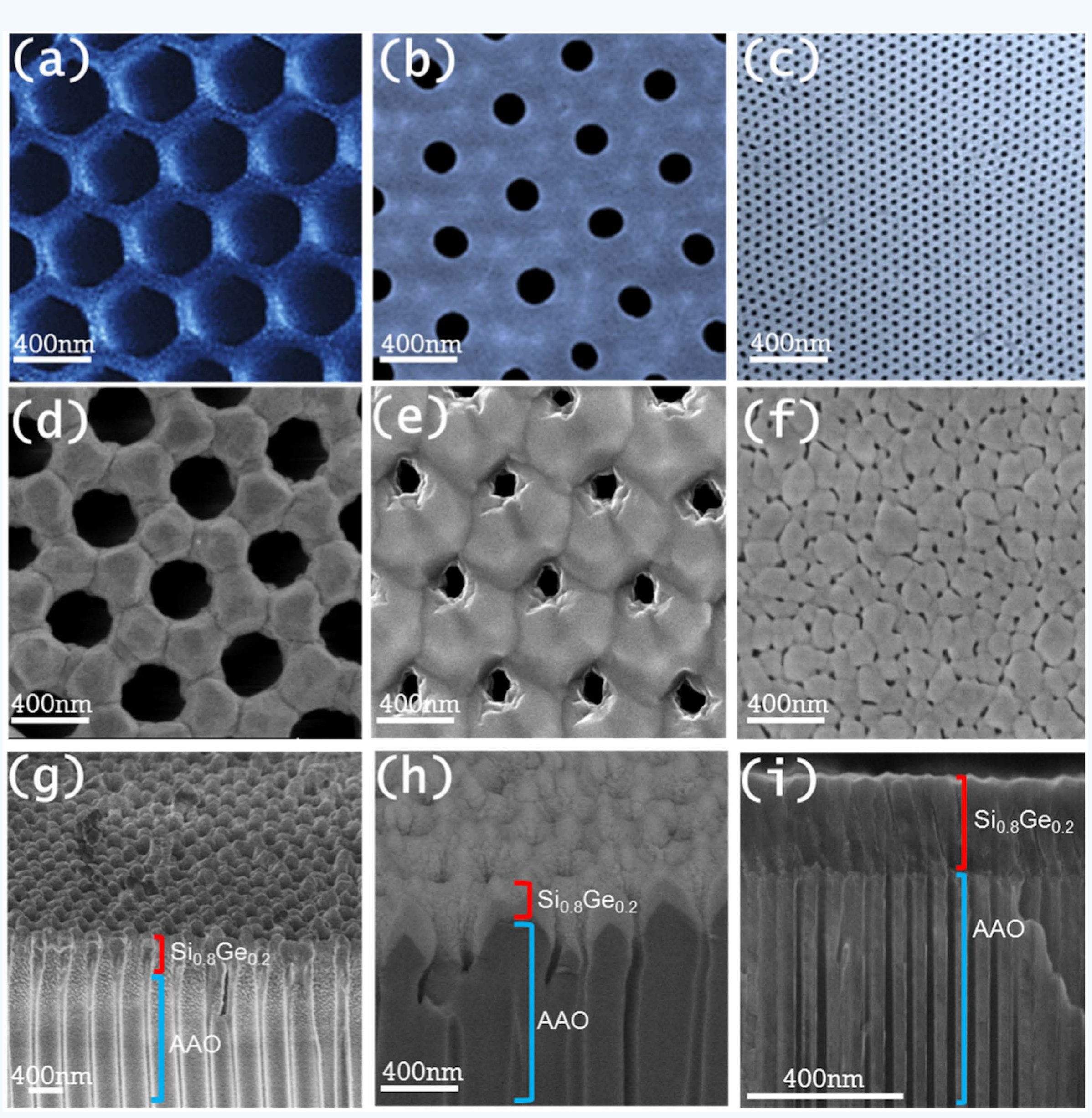






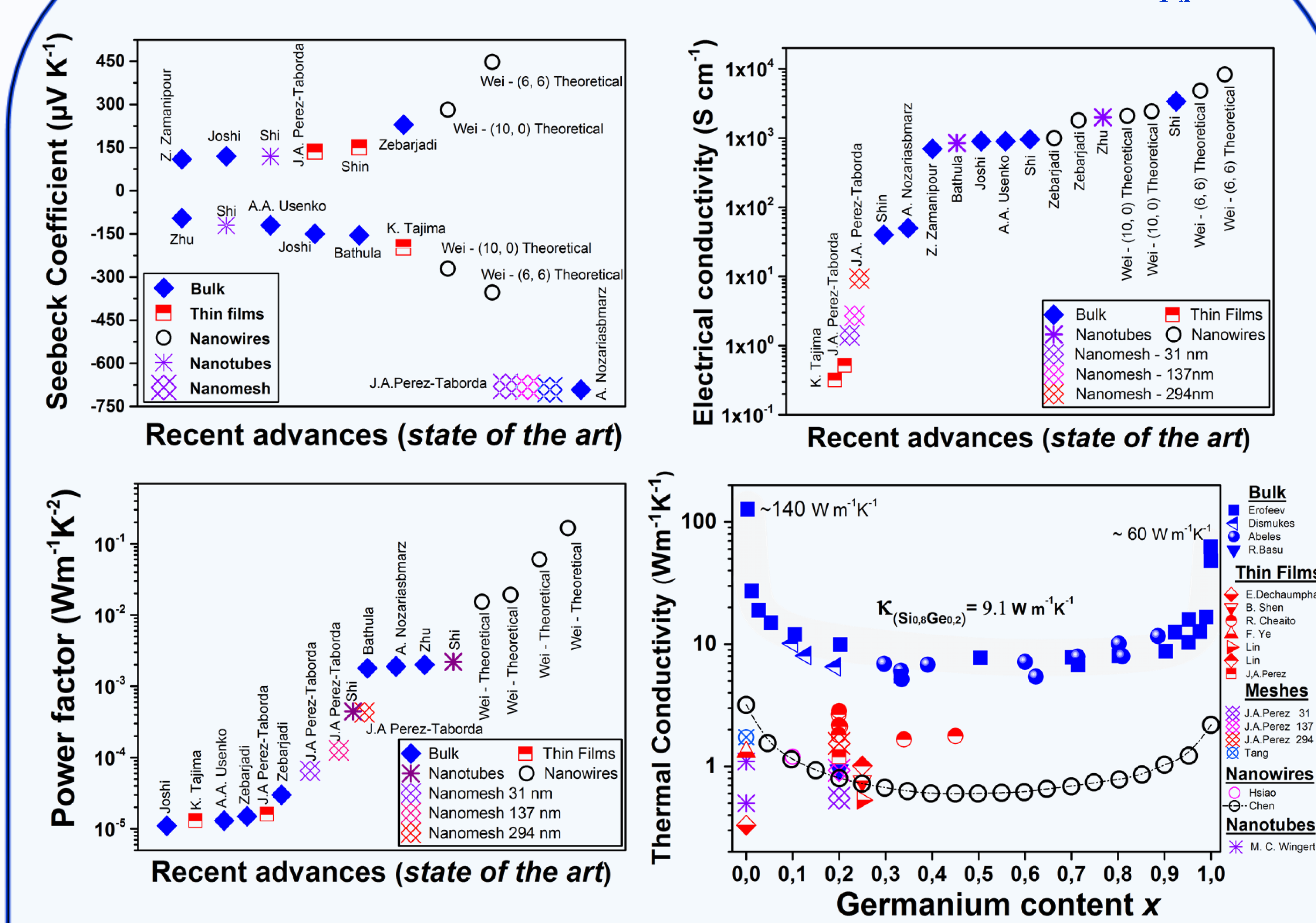
All nano-meshed $Si_{0.8}Ge_{0.2}$ polycrystalline films were oriented along the [111] direction, as revealed from **XRD** measurements and **Raman spectra** showed the three characteristic vibrational modes Ge-Ge, Si-Ge and Si-Si. A vertical right is shown a **Scanning Electron Microscopy - SEM** image of a $Si_{0.8}Ge_{0.2}$ nano-meshed film of ~294 nm porous size. Topography by Atomic Force Microscope - **AFM** and surface potential image by KPFM it is shown just below. The uniformity in the contrast of the **Kelvin probe force microscopy - KPFM** image reveals homogeneity in the surface potential of the film.

Morphological characterization: variation of pore size



The sputtered $Si_{0.8}Ge_{0.2}$ films replicated the porous structure of the alumina, resulting in the nano-meshed films with different pore sizes. In the figure **(a–c)** are SEM images of porous alumina templates with 436 ± 16 nm, 162 ± 11 nm and 31 ± 4 nm diameters, respectively, that were used as substrates in the sputtering process. **(d–f)** show SEM images of the sputtered $Si_{0.8}Ge_{0.2}$ nano-meshed films grown on the previous templates, which have replicated the porous alumina. **(g–i)** are SEM images of the lateral of these samples, where the $Si_{0.8}Ge_{0.2}$ films and the alumina matrix can be observed.

Thermoelectric characterization: Current state of the art for $Si_{1-x}Ge$



A summary of the latest reported measurements of different structures of $Si_{1-x}Ge$ including our measurements designated Nanomesh for 31, 137 and 294 nm pore size is shown. From left to right, shows *Seebeck coefficient*, *electrical conductivity*, and *power factor* reported for bulk, thin films, nanomeshes, nanowires, and nanotubes. The *thermal conductivity* for different $Si_{1-x}Ge$ nanostructures and bulk samples as a function of the alloy composition. This figure is adapted from Ref. [1].

Conclusions:

- A novel approach to grow large area $Si_{0.8}Ge_{0.2}$ nano-meshed films with different porous sizes through DC-Sputtering processes is shown. Additionally Raman spectra and XRD diffractograms show good crystallinity and reproducibility of the samples regardless of pore size.
- The thermal conductivity reduction of the $Si_{0.8}Ge_{0.2}$ nano-meshed films varies from **1.54 ± 0.27 W $K^{-1}m^{-1}$** for the 294 ± 5 nm nanopore film, **0.93±0.15W $K^{-1}m^{-1}$** for the 137±8 nm of the porous size and **0.55 ± 0.10 W $K^{-1}m^{-1}$** for the 31 ± 4 nm nanopore film.
- The power factors of the $Si_{0.8}Ge_{0.2}$ nano-meshed films varied from ~445 μ W m^{-1} K^{-2} to ~65 μ W m^{-1} K^{-2} at room temperature for the largest diameter pore nano-mesh (294 ± 5 nm) and the smallest one (31 ± 4 nm), respectively.
- The ***zT*max = 0.9** was obtained at room temperature. Nevertheless, it is remarkable that the thermal conductivity in the small pore structures can be reduced to such low values while still retain reasonable power factors, which opens the door for higher efficiency thermoelectric applications for this alloy once it is further optimized to improve its electrical conductivity.

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References:

[1] Perez-Taborda, J. A., Rojo, M. M., Maiz, J., Neophytou, N., & Martin-Gonzalez, M. Ultra-low thermal conductivities in large-area Si-Ge nanomeshes for thermoelectric applications. Scientific Reports, 6, 32778 (2016).