NANOSTRUCTURES and METHODOLOGIES

Reversible diffusion of aluminium and silicon in a nanowire

ABSTRACT

MRS

Diffusion is a phenomenon whereby matter is transported from a region with **high concentration** to a region with **lower concentration**. We all know its effect: If I open the window on a cold day, the inside and outside temperatures will slowly converge since fast and hot gas molecules from inside mingle with the slower and colder gas molecules from outside. We also know that we cannot reverse this effect and "unmix" the hot and cold molecules, we cannot command the hot molecules to come back inside! But in a specific solid-state system, we can do exactly this.

This **reversible** diffusion phenomenon was an accidental observation during research on making electrical contacts on semiconductor nanowires. A nanowire is a thin, wire-like structure, with a diameter of several nanometres to tens of nanometres. Nanowires fabricated from semiconducting materials can be used in devices such as field-effect transistors, light-emitting diodes and solar cells. **Nanowires** can incorporate insertions of different materials and, in devices, they require high-quality electrical contacts. With colleagues at the Institute for Interdisciplinary Research of Grenoble we have been fabricating **aluminium contacts onto Silicon-Germanium** (Si-Ge) alloy nanowires positioned on a thin **SiN membrane** transparent to the very fast electrons of a Scanning Transmission Electron Microscope (STEM). These structures were then **heated** inside the TEM, to **image the solid-state reactions** with nanometre resolution. At 580°C, a solid-state reaction is started where the aluminium from the metal contact pad diffuses progressively into the nanowire, replacing germanium and silicon atoms. The **Ge and Si atoms diffuse in the opposite direction, along the nanowire surface**, exit the nanowire, and **incorporate** at surfaces and grain boundaries in the **aluminium** pad. Thus, a near perfect **monocrystalline aluminium** nanowire section is formed, with a **perfectly abrupt interface** to the SiGe nanowire, and making an excellent electrical contact with it, see Fig. 1 (top). Surprisingly, if the heating temperature is lowered slowly, the Al contacts can no longer contain all the silicon atoms they had stored. The Si atoms (and also, but much less, the heavier Ge atoms) **diffuse back** along the nanowire and nucleate at the Al/SiGe interface, progressively forming a region of almost pure Si, with a low Ge content. This region grows in the reverse direction as the Al atoms return to the metal pad (Fig. 1, bottom). We found that the aluminium could move in and out repeatedly. To the best of our knowledge, this work demonstrates a **reversible diffusion mechanism** for the first time, and absolutely unambiguously, thanks to the in-situ Electron Microscopy experiments. The reversible diffusion arises due to the nanowire geometry and crystallinity. This concept may be extended to other material systems. In our own work, the different diffusion properties of Si and Ge have allowed us to fabricate and contact complex and abrupt Al metal/Si-rich/SiGe alloy heterostructures with sharp interfaces in a single processing step, all compatible with current Si/Ge technology, see Fig. 2.

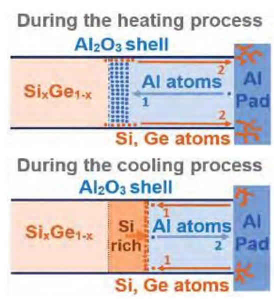


Fig. 1: At top: Diffusion of aluminium into the Si_xGe_{1-x} alloy nanowire, and diffusion of silicon and germanium out into the Al contact pad. Below: The reverse diffusion process for Al and Si; the lesser reversibility for Ge diffusion produces a Si-rich region [1].

Scientific methods:

- Silicon nitride membranes: electron beam lithography contacting of nanowires [5].
- In-situ heating / electrical TEM experiments [1-6]
- Fabrication of abrupt metal – semiconductor interfaces [1-5]
- Mapping electrical fields [6,7]

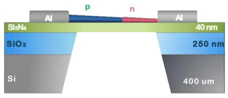
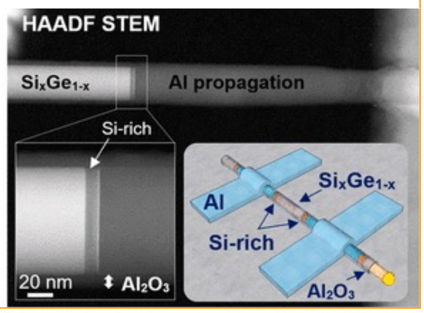
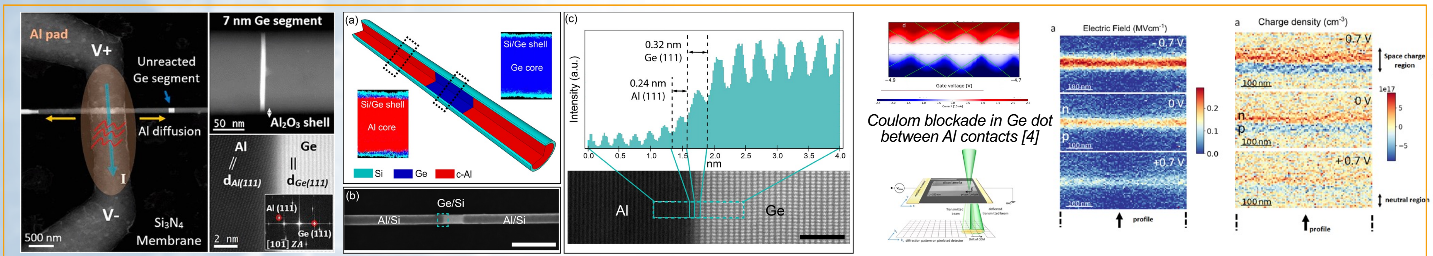


Fig. 2: High angle annular dark field (HAADF) scanning transmission electron microscopy (STEM image of the original Si_xGe_{1-x} nanowire (NW) on the left, the intermediate Si rich region with a zoom below, and the transformed Al region on the right, and the Al contact just visible on the far right. The inset shows the schematic of the structure[1].



Related Studies



HAADF images showing the in-situ fabrication of a 7 nm Ge quantum dot [2], the Al/Ge interface without and with a si shell [1,3], 4D STEM on a pn junction with electric field and charge density maps [6,7]

Collaborations & FURTHER READING

Collaborations: Quanteca, Nanofab, POM, CEA Grenoble: IRIG (LEMMA, NPSC, SINAPS), LETI, TU Vienna.
Projects: ERC stg e-See, PEPR METSA-SETDIA, LANEF

[1] M.A. Luong, et al, "*Reversible Al Propagation in Si_xGe_{1-x} Nanowires: Implications for Electrical Contact Formation*", ACS Appl. Nano Mater. **3**, 10, 10427 (2020).

[2] M.A. Luong, et al, "*In-Situ Transmission Electron Microscopy Imaging of Aluminum Diffusion in Germanium Nanowires*", for the Fabrication of Sub-10 nm Ge Quantum Disks, ACS Applied Nano Materials **3**, 2, 1891–1899 (2020)

[3] M. Sistani, et al, "*Highly Transparent Contacts to the 1D Hole Gas in Ultrascaled Ge/Si Core/Shell Nanowires*", ACS Nano **104**, 102102 (2019)

[4] M. Sistani, et al, "*Coulomb blockade in monolithic and monocrystalline Al-Ge-Al nanowire heterostructures*", Applied Physics Letters 116, 013105 (2020)

[5] M. Spies, et al, "*Correlated and in-situ electrical transmission electron microscopy studies and related membrane fabrication*", Nanotechnology **31**, 472001 (2020)

[6] B.C. da Silva, et al. "*Assessment of Active Dopants and p–n Junction Abruptness Using In Situ Biased 4D-STEM*", Nano Letters **22**,23, 9544–9550 (2022)

[7] A. Wartelle, et al, "*Sub-microradian angular detection limits for field mapping by Lorentz 4D scanning transmission electron microscopy on a Si p-n junction*", JAP **138**, 105703 (2025)