Electrical transport properties of single-crystal Al nanowires

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Abstract
Single-crystal Al nanowires (NWs) were fabricated by thermally induced substitution of vapor-liquid-solid grown Ge NWs by Al. The resistivity of the crystalline Al (c-Al) NWs was determined to be \( \rho = (131 \pm 27) \times 10^{-9} \Omega \text{m} \), i.e. approximately five times higher than for bulk Al, but they withstand remarkably high current densities of up to \( 1.78 \times 10^{12} \text{A m}^{-2} \) before they ultimately melt due to Joule heating. The maximum current density before failure correlates with the NW diameter, with thinner NWs tolerating significantly higher current densities due to efficient heat dissipation and the reduced lattice heating in structures smaller than the electron–phonon scattering length. The outstanding current-carrying capacity of the c-Al NWs clearly exceeds those of common conductors and surpasses requirements for metallization of future high-performance devices. The linear temperature coefficient of the resistance of c-Al NWs appeared to be lower than for bulk Al and a transition to a superconducting state in c-Al NWs was observed at a temperature of 1.46 K.

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(Some figures may appear in colour only in the online journal)
become the primary choice for modern ICs due to its higher conductivity and in particular its higher resistance to electromigration (EM), which increases the on-state resistance and can eventually lead to complete failure.

Although Cu wires and interconnects can withstand approximately five times higher current densities than Al [10], EM effects still govern the reliability of the devices. Grain boundary diffusion is the main migration process in Al metallization [10] and the most common cause of failure at high current densities. This can be effectively eliminated in single-crystal NWs, which may significantly improve their overall stability against high current densities leading to Al NWs being a high-potential metallization material for future devices.

The recently reported approach to fabricate single-crystal Al NWs by thermally induced substitution of a Ge NW by Al [11] enables the synthesis of c-Al NWs with well-controlled geometries. These c-Al NWs present a versatile model system for studying the electrical properties of quasi-1D metallic nanostructures. The confinement of charge carriers and the crystalline nature may drastically alter the electrical and thermal transport properties of metallic NWs [12, 13] as well as their resistance to electrical stress [14]. Thus, optimized c-Al NWs may allow for high performance interconnects [15] or e.g. electrodes for field-effect transistors [14], where stability against high current and long-term electrical stress are key requirements. Moreover, metallic NWs exhibiting a transition to a superconducting state may pave the way for single-photon detectors [16] or Josephson junctions based on NW heterostructures [17, 18].

2. Experimental details

To obtain c-Al NWs, single-crystal Ge NWs were grown heteroepitaxially on silicon substrates in a low-pressure chemical vapor deposition system by use of the gold-assisted VLS process. The (111) oriented Ge NWs with diameters ranging from 25 to 160 nm and lengths of several micrometers were dispersed onto a piece of a highly p-doped Si wafer with a 100 nm thick layer of SiO₂. Al pads contacting individual NWs and tracks connecting them to prepatterned Ti–Au bonding pads were structured by electron beam lithography, Al sputter deposition preceded by a HI dip and lift-off techniques. Subsequently, the diffusion of Al into the Ge NWs was induced by rapid thermal annealing at 623 K. The significantly different diffusion coefficients in the Al–Ge system result in the substitution of the (111) oriented Ge NWs by Al and thus to axial growth of crystalline Al sections intruding into the NWs from the Al pads [11], which can even be monitored in situ by scanning electron microscopy (SEM) or transmission electron microscopy (TEM). For prolonged annealing, Al substitutes Ge over the entire length of the NWs and continuous single-crystal Al NWs can be obtained. Details of the NW formation process are given in [11].

To confirm the morphology and composition of the NWs after the thermally induced substitution, TEM and energy dispersive x-ray analysis (EDX) were performed in situ directly after the propagation of the Al phase inside the Ge NWs. The NWs appeared to be pure Al within the detection limit of the EDX system. In the TEM image and inserted diffraction pattern (figure 1(a)), the NW appears to be single-crystalline with the atomic planes visible in the high resolution TEM image along the whole NW with a spacing of 0.236 nm, which is in good agreement with the theoretical interplanar spacing of 0.234 nm for (111) planes in face centered cubic Al [11]. Therefore, the analysis of the crystal structure in the reacted part as well as the elemental analysis confirm that Ge is entirely replaced by Al while retaining the crystalline structure of the original NW.

To eliminate the parasitic contact resistance during electrical characterizations, c-Al NWs were integrated in four-terminal test devices as shown in figure 1(b).

Resistivity measurements were conducted by linearly increasing the current forced through individual NWs while recording the voltage drop across two electrodes.
Subsequently, the resistance was obtained from a linear fit of the voltage-over-current (V–I) plot.

### 3. Results and discussion

The resistivity of more than 20 of such fabricated c-Al NWs with diameters ranging from 25 to 50 nm was determined at room temperature with four-terminal measurements to be $\rho_{\text{Al NW}} = (131 \pm 27) \times 10^{-9} \Omega \text{m}$, which is approximately five times the resistivity of bulk Al [19]. NWs are known to exhibit increased resistivity compared to bulk material due to an increased significance of grain boundary and surface scattering in nanostructures with dimensions approaching the mean free path of charge carriers [20]. For the actual case of single-crystal Al NWs, grain boundary scattering can be excluded, but surface scattering may increase the resistivity. Furthermore, we assume that residual Ge atoms below the EDX detection limit may also act as scattering centers in the NWs. This hypothesis is supported by the observation that for longer annealing durations the resistivity of the fabricated c-Al NWs decreases (see supporting information). Dissolved Ge atoms, which remained in the c-Al NWs during an incomplete exchange reaction, may diffuse into the extended Al contact pads during prolonged annealing. Therefore, a longer annealing duration leads to further ‘purification’ of the c-Al NWs and thus a decrease in resistivity of about 10%.

To investigate the temperature dependence of the resistivity, the samples were introduced into a vacuum cryostat and the current-over-voltage ($I$-$V$) characteristics of c-Al NWs were determined as a function of temperature. The plot in figure 2(a) shows the resistivity of a 2.95 $\mu$m long c-Al NW with a diameter of 33 nm as a function of the absolute temperature between 5 K and room temperature and unambiguously displays the metallic character of the c-Al NW.

In the temperature regime below 50 K, where the current transport is dominated by electron scattering at defects or at the surface of the NW, the resistance appears to be widely independent of the temperature. In the adjacent temperature regime up to 150 K the resistance increases almost linearly with temperature due to phonon scattering leading to a linear temperature coefficient of the resistance of $\alpha_{\text{low}} T = (1.32 \pm 0.03) \times 10^{-3} \text{K}^{-1}$. In the regime from room temperature to about 370 K (see supporting information), we determined a higher value of $\alpha_{\text{high}} T = (1.80 \pm 0.78) \times 10^{-3} \text{K}^{-1}$ for NWs with different diameters, which is still approximately half of the temperature coefficient of the resistance of bulk Al of $\alpha_{\text{bulk}} = 3.69 \times 10^{-3} \text{K}^{-1}$ [21]. This is in agreement with previous reports of decreasing temperature coefficients with decreasing diameters in Ag and Cu NWs, which is attributed to a modified phonon scattering behavior and enhanced diffuse surface scattering in thin NWs [22, 23].

The current-carrying capacity of c-Al NWs was investigated by gradually increasing the current forced through the NWs in a two-terminal configuration and monitoring the voltage across the NWs. Figure 3(a) shows exemplary $V$–$I$ curves recorded for five NWs with diameters ranging from 25 to 50 nm. The steep increase in the voltage indicates failure of the NW. For all NWs, the slope in the $V$–$I$ curve, i.e. the resistance, increased right before the point of failure, which is an indication of effective Joule heating ultimately leading to melting induced failure. SEM images taken after failure (see inset in figure 3(a)) clearly show signs of melting for the Al NWs as reported also for Au NWs by Aherne et al [24].

As expected, thicker NWs were observed to tolerate higher absolute currents (see figure 3(a)). However, calculating current densities leads to the remarkable result, that the failure current density of thinner NWs is significantly higher than that of thicker ones (see figure 3(b)). Overall, more than 50 NWs with different geometries were tested and notably high failure current densities of $J_{\text{fail}} = 2.24 \times 10^{11} \text{A m}^{-2}$ to $J_{\text{fail}} = 1.78 \times 10^{12} \text{A m}^{-2}$ were measured. Such exceptional ampicity of thin metallic NWs was also observed for Au NWs [24, 25] and is attributed to the combination of efficient heat dissipation due to the high surface-to-volume ratio of
thin NWs and the reduced lattice heating in structures smaller than the electron–phonon scattering length [25].

With respect to the abovementioned EM issues, reliability tests of single-crystal Al NWs under electrical stress were performed in order to investigate the long-term stability of c-Al NWs at high current densities. The resistance of the c-Al NWs remained stable over 15 h at current densities of about $10^{12} \text{A}\cdot\text{m}^{-2}$, which is about four orders of magnitude above the failure current density of bulk Al. At slightly higher current densities, NWs appeared to gradually deteriorate and experienced failure after extended electrical stress (see supporting information).

Previous studies reported extraordinarily high failure current densities for Au NWs [25] with a maximum of $3.5 \times 10^{12} \text{A}\cdot\text{m}^{-2}$ [26] and for NiSi NWs with a maximum of $3 \times 10^{12} \text{A}\cdot\text{m}^{-2}$ for a NW with a diameter of 29 nm [14]. Although not exceeding the highest reported failure current density of Au NWs, a maximum failure current density of $J_{\text{fail}} = 1.78 \times 10^{12} \text{A}\cdot\text{m}^{-2}$ and the performance of c-Al NWs in long-term tests make them again considerable for device interconnects. As shown in figure 4, the remarkably high failure current density of single-crystal Al NWs exceeds the level recommended by the International Technology Roadmap for Semiconductors (ITRS) [27, 28] by orders of magnitude and therefore makes c-Al NWs an attractive candidate for interconnects or high-performance electrodes in future device applications.

The capability to fabricate single-crystal Al NWs with well-controlled geometries may allow for novel devices with minimized heat dissipation using effects such as superconductivity. Therefore, we investigated if the basic property of a superconductor, i.e. dissipationless electric current, is preserved at the reduced scales of the actual NWs.

As shown in figure 2(b), temperature dependent $I$–$V$ measurements in an adiabatic demagnetization refrigerator (ADR) revealed a clear transition to a superconducting state for the quasi-1D c-Al NW with a length of 2.95 μm and a diameter of 33 nm. Although the resistance in the superconducting state should ideally be zero, a finite residual resistance below the transition was observed, which is attributed to non-superconducting wires and feedthroughs used in the ADR setup due to the two-terminal configuration. Nevertheless, the steep decrease of the resistance between 1.81 and 1.10 K is a clear indication of the c-Al NW

Figure 3. (a) $V$–$I$ characteristics of c-Al NWs with different diameters in a two-point configuration. A sudden increase in the voltage denotes failure. Inset: SEM image of NW displaying melting induced failure due to Joule heating under excessive current stress. (b) Failure current density of c-Al NWs as a function of NW diameter.

Figure 4. Comparison of failure current density (ampacity) and conductivity of common metals, nanocarbons, carbon nanotube—copper composites and c-Al NWs. Both carbon nanotube—copper composites and c-Al NWs clearly exceed the ampacity recommended for conductors by the ITRS. Figure adapted with permission from Subramaniam et al [27] copyright 2013.
transitioning to a superconducting state. The midpoint of a linear fit of the transition was used to obtain a critical temperature of \( T_c = 1.46 \) K, which is significantly higher than for bulk Al \( (T_{c,\text{bulk}} = 1.19 \) K \( )^{[29]} \). Such an enhancement of the critical temperature in Al has previously been reported in thin films \( ^{[30]} \) and NWs \( ^{[31]} \) and is attributed to the so-called shape-resonance effect \( ^{[31]} \) and to a modification of the phonon spectrum by surfaces \( ^{[32]} \). The study of the size-dependent enhancement of superconductivity in Al NWs by Shanenko et al \( ^{[31]} \) revealed a significant increase of \( T_c \) for thinner Al NWs, which is in line with the presented experimental data.

A common figure of merit for the purity of superconductors is the residual resistance ratio (RRR), which is typically defined as the ratio of the resistance \( R_{273} \) at the melting point of ice, i.e. at 273 K, and the resistance \( R_{4.2} \) at the boiling point of He, i.e. at 4.2 K \( ^{[33]} \). The RRR measures the contribution of electron scattering at impurities or lattice defects to the total normal state resistance of a superconductor, with high values indicating high-purity materials. A RRR of 1.45 was calculated for the actual Al NW indicating a high number of impurities, possibly due to dissolved Ge, as mentioned above. However in contrast to bulk superconductors, the significant contribution of surface scattering to the total resistance of quasi-1D NWs may lead to a large residual resistance at 4.2 K and thus a lower RRR although according to HRTEM investigations the number of lattice defects is small.

4. Conclusion

Single-crystal Al NWs with various geometries were fabricated by thermally activated substitution of VLS-grown Ge NWs by Al. The resistivity of such Al NWs was determined to be \( (131 \pm 27) \times 10^{-5} \Omega \text{m} \) and thus significantly higher than that of bulk Al, which is mainly attributed to enhanced surface scattering of electrons due to the high surface-to-volume ratio of rod-like NWs.

The linear temperature coefficient of the resistance was observed to be lower than for bulk Al, which was attributed to the modified phonon scattering behavior and increased diffuse surface scattering in thinner NWs. Single-crystal Al NWs were shown to withstand remarkably high current densities. A maximum failure current density of \( 1.78 \times 10^{12} \text{A m}^{-2} \) was observed and a correlation between the NW diameter and their current-carrying capacity was established. The NWs exhibit good long-term stability at extremely high current densities without indications of EM. The extraordinarily high current-carrying capacity of c-Al NWs, which exceeds the level recommended for conductors by the ITRS \( ^{[28]} \) by orders of magnitude is comparable to that of carbon nanotube—copper composites \( ^{[27]} \) and suggests that c-Al NWs may be considered again as high-performance interconnects in future devices.

The presented transition to a superconducting state in c-Al NWs at a critical temperature of 1.46 K may prove important for studying effects such as the superconducting proximity effect or Josephson currents in quasi-1D systems.

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