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Imaging the three-dimensional morphology of granular superconductors with energy-filtered TEM tomography

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Background incl. aims

This work aims to resolve the structure - property relationship in granular Aluminum (grAl) thin films. GrAl is currently applied in microwave kinetic inductance detectors, parametric amplifiers, fluxonium qubits, resonators and filters. The granular structure and crystallite size distribution of the Aluminum particles have been known for a long time through dark field TEM imaging. The disadvantage of dark-field TEM is that it shows a 2D-projection of a 3D-network. In order to resolve the interconnection between the Al particles, 3D-STEM tomography was applied to resolve the morphology of the grAl. Combination with accurate models for simulations will advance the understanding of granular superconductors and their properties.

GrAl is produced by physical vapor deposition of Aluminum in presence of partial Oxygen pressure. It is a so-called 'dirty superconductor'. The emerging granular Aluminum nanostructure that is embedded in amorphous Aluminum oxide features tunable non linearity, kinetic inductance and low AC losses in its superconducting state. The Aluminum grains are assumed to form Josephson junctions between each other. A phase shift is induced on the superconducting current while tunneling through a small insulating Aluminum oxide gap between grains. Tunability of the non-linear properties is achieved by adjusting the room temperature resistivity of the film and varying the number of junctions in the network by changing the volume of a grAl component.

The critical temperature for superconductivity TC over the room temperature resistivity of the films reveals a dome shaped relationship with its maximum of 3 K at around 500 µΩcm while TC for pure aluminum is at 1.2 K. Depositing the films on cold substrates decreases Aluminum crystallite size and raises TC even beyond 3 K.

Methods

Two samples are investigated with 120 µΩcm and 25000 µΩcm resistivity. The films of 20 nm thickness are deposited on lacey Carbon TEM grids for easy nanowire templating. Gold fiducial markers are drop cast on the grid. The coated Carbon laces are investigated by energy-filtered TEM (EFTEM) on a Thermo Fisher Scientific Titan Themis Z at 80 kV with a Gatan image filter. EFTEM tomography was applied to reveal changes in al plasmon peak. The tilt series is aligned with the help of fiducial marker positions and is reconstructed using the Discrete Algebraic Reconstruction Technique (DART). Contrast levels are being estimated from an initial Simultaneous Iterative Reconstruction Technique (SIRT) reconstruction.

Results

STEM and TEM imaging show minimal contrast variations between Aluminum Oxide and metallic Aluminum due to similar atomic masses. Image corrected HRTEM shows partially polycrystalline grains in projection. EFTEM at 14 to 16 eV shows good contrast of the Aluminum volume plasmon which is confined to the Aluminum particles. Aluminum Oxide and Carbon give minimal intensity contributions. The low resistivity sample with 120 μΩcm is reconstructed as a network of interconnected 5 nm Aluminum particles with a surface oxide cover of 3 nm. An oxide barrier separating close grains cannot be resolved. The non-linear inductive behavior can be explained with constrictions in the conductive path through the grains instead of oxide barriers as initially assumed. The high resistivity sample with 25000 μΩcm is reconstructed with 3 nm grains a notably bigger surface oxide layer of 12 nm. Many grains are still interconnected but there are oxide barriers separating clusters of grains from each other. The interconnection of grains at high resistivity is either a reconstruction artefact or shows that already a small amount of oxide barriers is sufficient for high non-linearity.

Conclusions
This study provides a novel insight into the 3D morphology of granular Aluminum (grAl) thin films, challenging previous assumptions about the structure-property relationship in these materials. The advanced electron microscopy techniques have revealed that the non-linear properties of grAl are more likely due to constrictions in the conductive path through the grains rather than oxide barriers. Furthermore, the study has shown that even a small number of oxide barriers can result in high non-linearity, particularly in high resistivity samples. These findings pave the way for more accurate simulations and could potentially enhance the performance of devices that utilize grAl. Further research is needed to fully understand the implications of these results and to explore the influence of substrate temperature during deposition on the structure of these films.

Keywords:

EFTEM Tomography Granular-Superconductor

Reference:

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