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Degradation of Superconducting Nb/NbN Films by Atmospheric Oxidation

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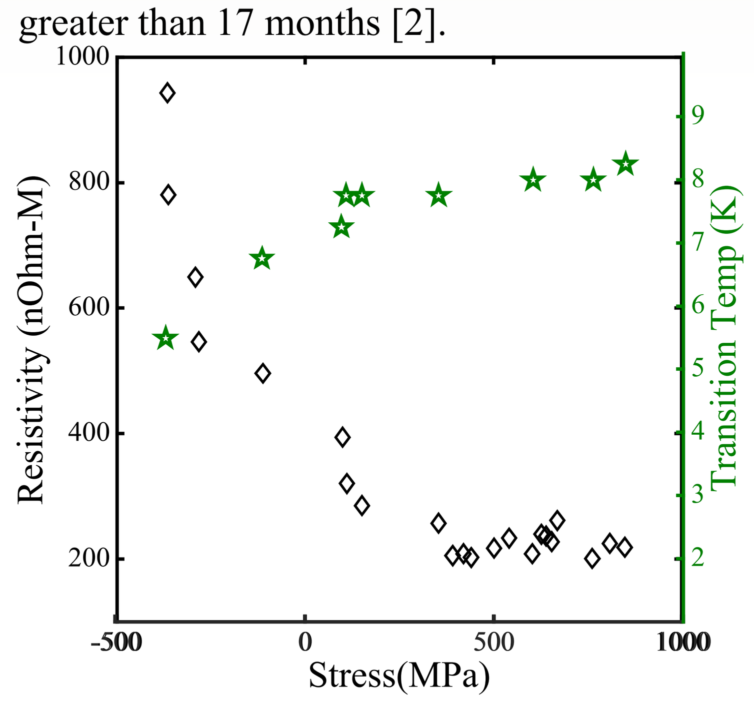
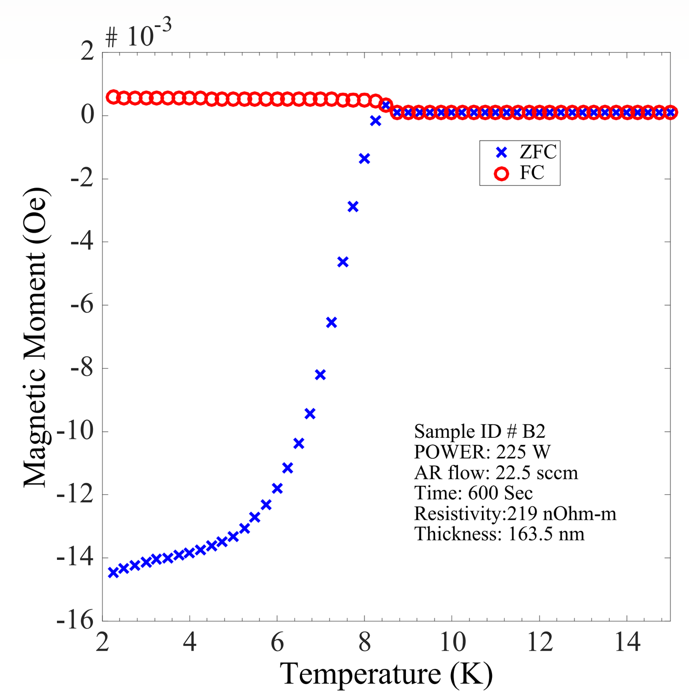
Abstract

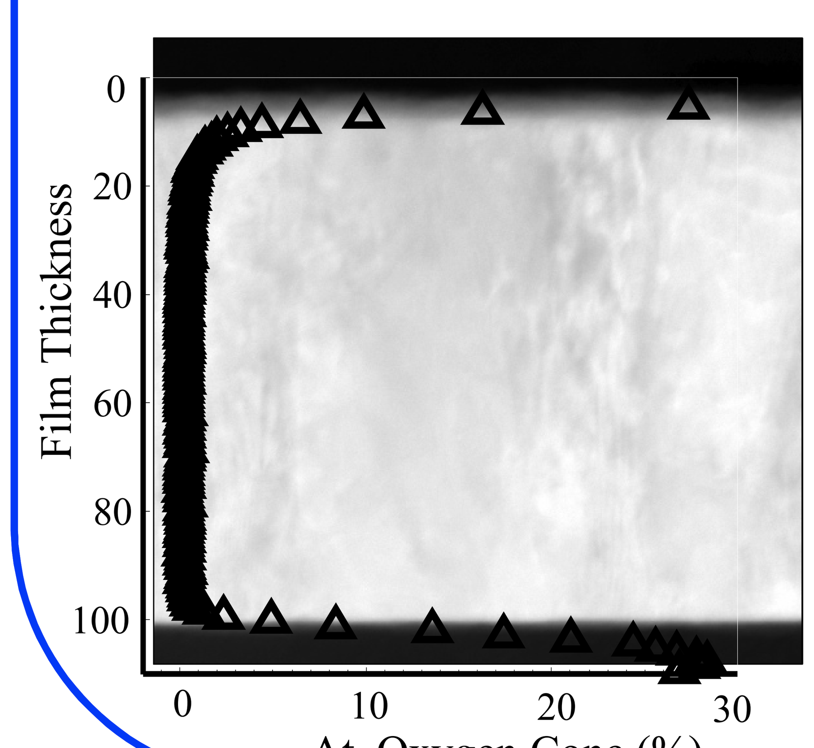
Niobium and niobium nitride thin films are transitioning from research novelty toward wafer scale manufacturing with technology drivers that include superconducting circuits and electronics (SCE), optical single photon detectors (SPD), logic, and memory. Successful microfabrication requires precise control over the properties of sputtered superconducting films, including oxidation, with good within-wafer and wafer-to-wafer uniformity. Previous work has demonstrated the mechanism in oxidation of Nb [1] and how film structure could have deleterious effects upon the superconducting properties [2]. This work continues with the examination of oxidation of NbN films. By examination of the RT sheet resistance of NbN bulk oxidation can be identified and confirmed by SIMS. Meissner magnetic measurements confirm the bulk oxidation not observed with simple cryogenic resistivity measurements.

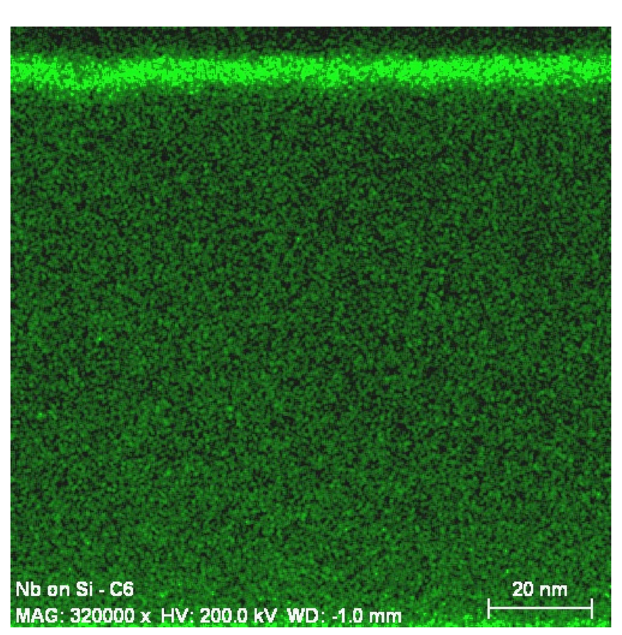
Mechanism of Oxidation in Nb

Grain structure which is not compacted adequately (compressive) can lead to deep grooves within the grain structure to absorb oxygen. This mechanism was described in detail for Nb by Halbritter [1]. Any oxygen absorption generates NbO_x up to a terminal Nb_2O_5 compound reducing the transition temperature (NbO_x) to the insulating phase (Nb_2O_5) which has the ability to continue oxygen mobility. With compressive Nb films the oxidation undergoes a limiting Mott-Cabrera surface oxidation resulting in a protective thin film. This thin film has demonstrated protecting the bulk superconductor for periods greater than 17 months [2].



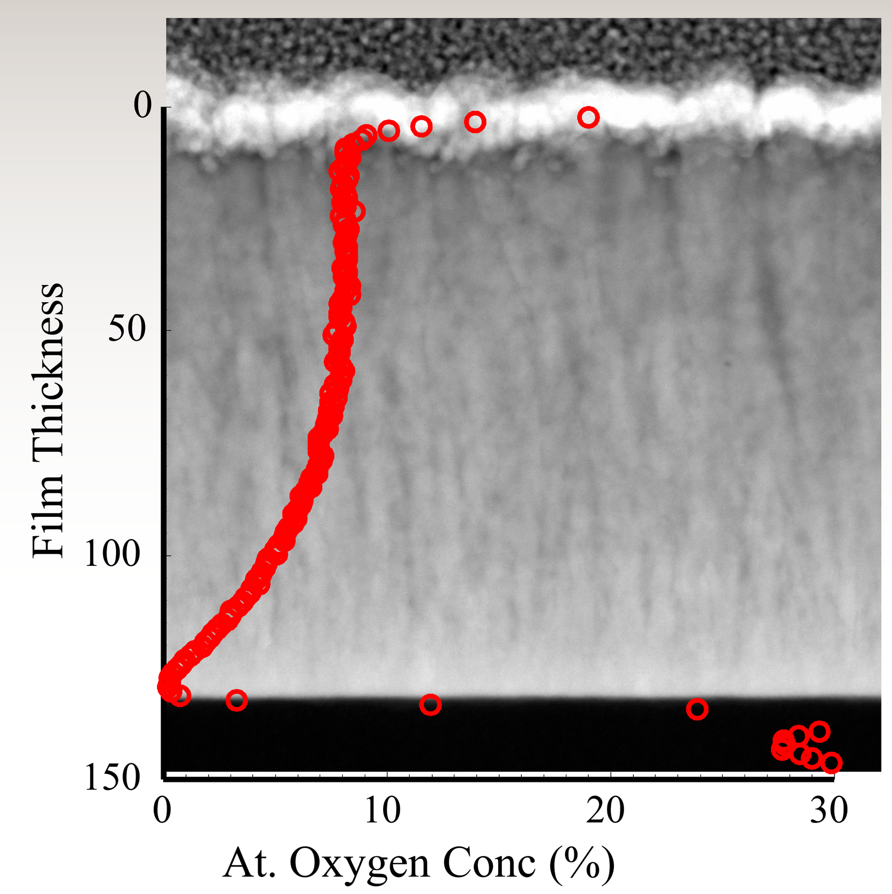


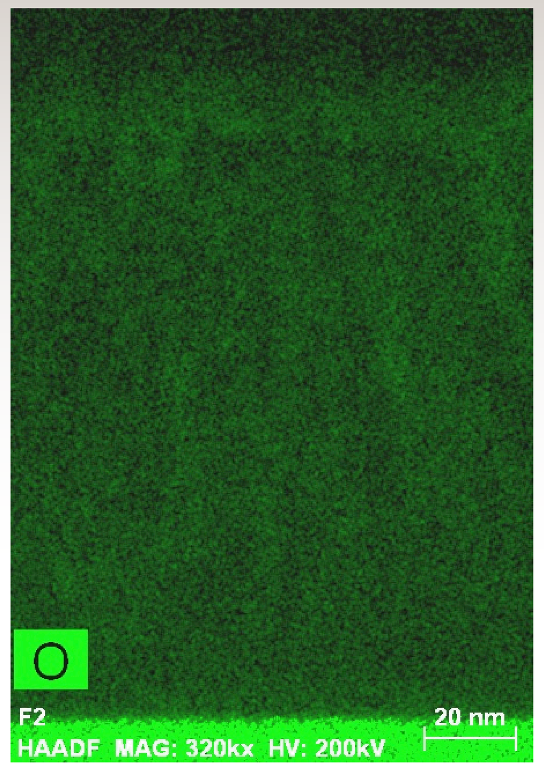


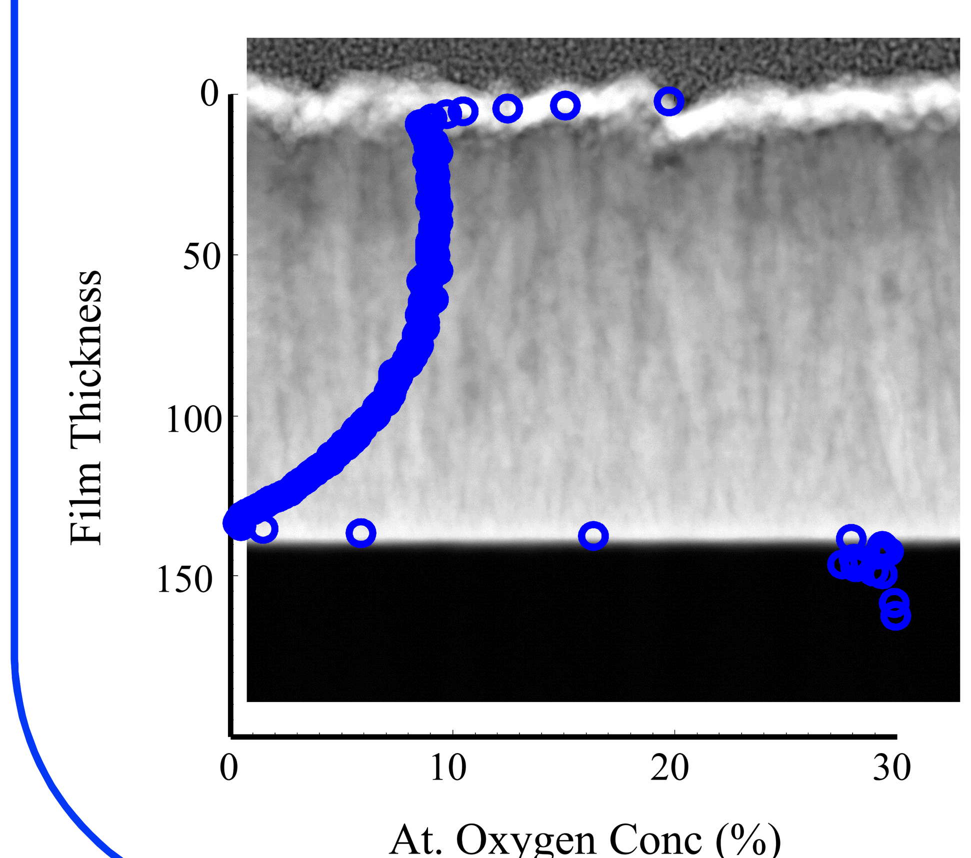


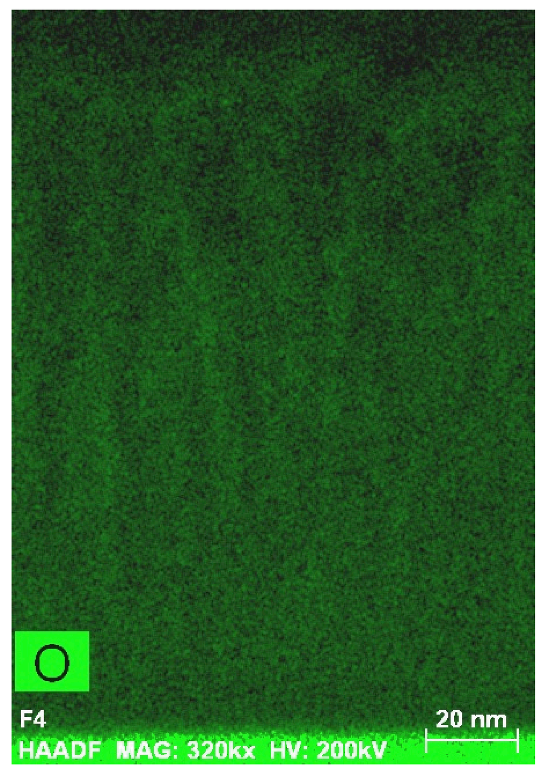
Oxidation of NbN

The oxidation process of NbN is understood based on Nb oxidation fundamentals and progress in much the same fashion. NbN also oxidizes and creates a similar surface oxide terminating as Nb_2O_5. In this work, NbN was sputter deposited at room temperature in a Denton Discovery 550 with Ar flow set to 15 sccm. The top film (red) was deposited 147 nm thick using N_2 flow of 1 sccm at 125 W DC power. The bottom film (blue) was deposited 150 nm thick at 0.4 sccm and 75 W. The left figures are TEM bright field images with SIMS oxygen measurements performed (calibrated using XPS). The right figures are oxygen EDS scans using the KA transitions. Both films indicated a bulk oxidation, particularly along the NbN grain structure, until the grains begin to compact at the SiO_2 interface and oxygen transport into the film is reduced.



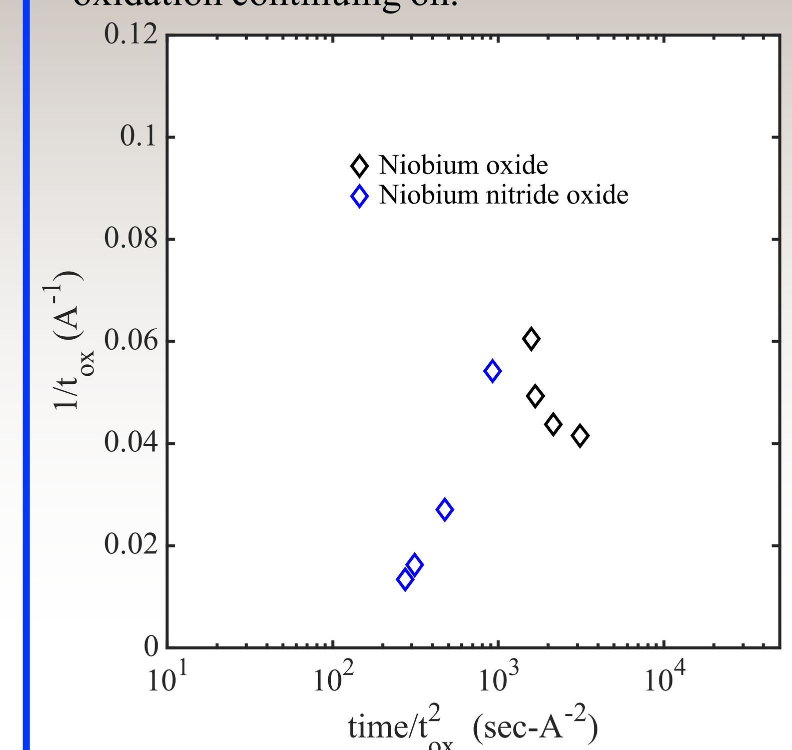


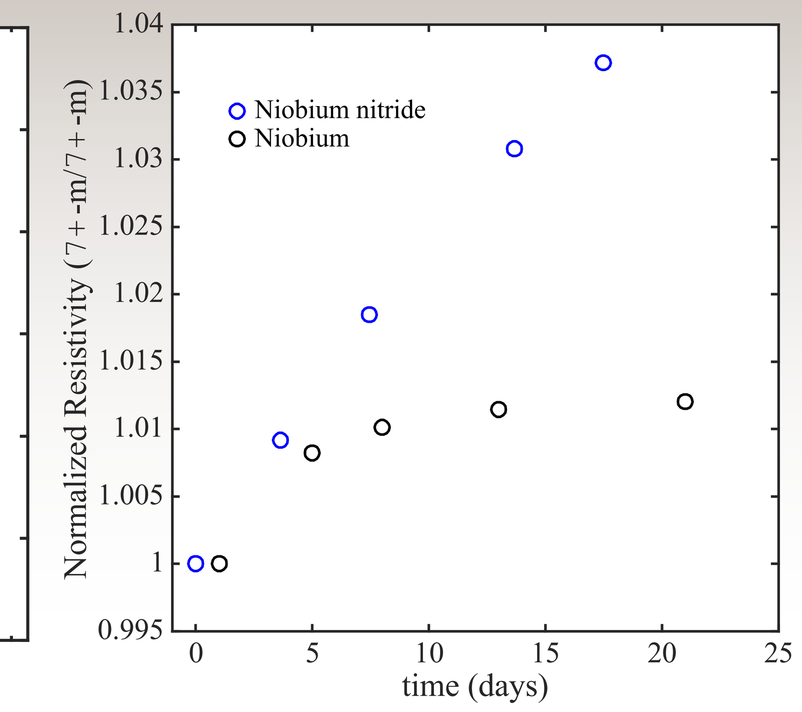




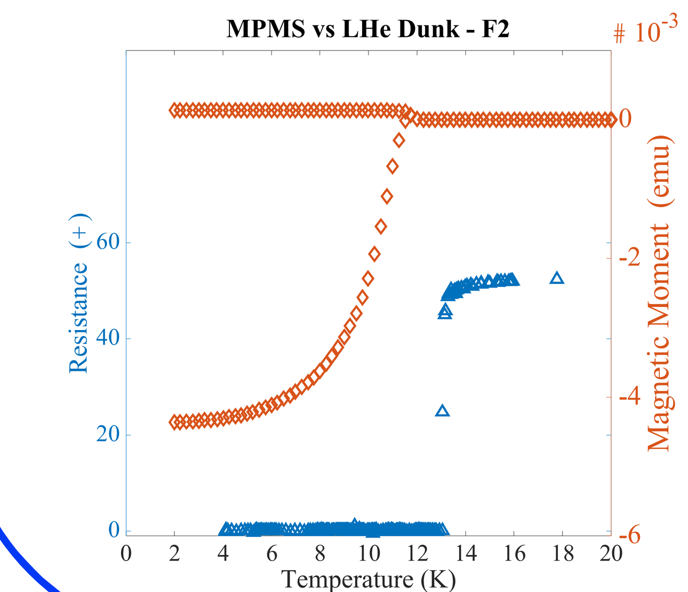
NbN vs Nb Electrical Performance

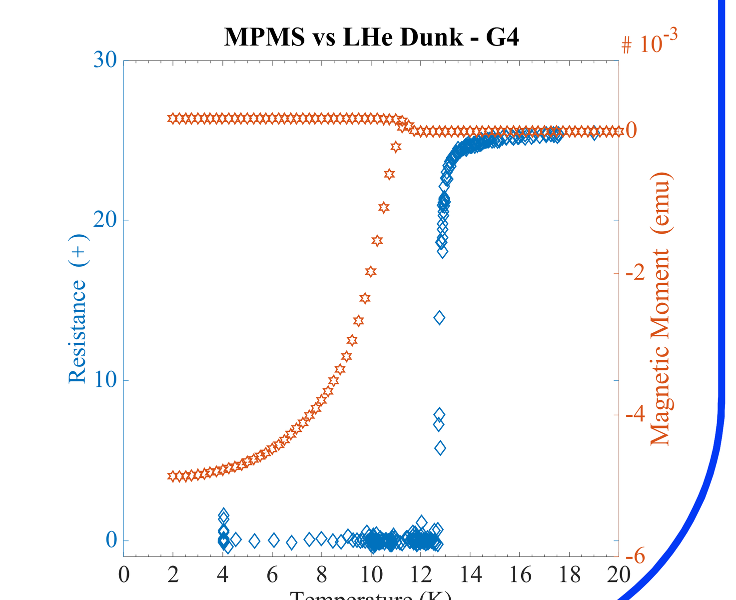
A Mott-Cabrera oxidation mechanism is summarized as where the oxidation reduction becomes limited due to a reduced electron tunneling probability across the formed oxide barrier and the oxidation rate dramatically slows. For Nb, this means a 5-7 nm terminal oxide. The Nb (**o**) is measured to create a limiting oxide which continues for about 2.4 nm whereas the NbN (o) is continuing growth well past 7.4 nm after 17 days. Ghez has shown that plotting this in the form of 1/x vs (time/x^2) should yield a straight line with negative slope if this type of oxidation occurs [3]. Ghez simple Mott-Cabrera test confirms the theory for Nb with a 5 nm oxide predicted after 550 days. NbN fails this test with strong oxidation continuing on.





MPMS (both ZFC and FC) measurements (red) yield a bulk transition temperatures in NbN of about 9-10 K whereas cryogenic (LHe dunked) resistivity measurements (blue) show transitions closer to 13 K. This suggests that the thin NbN grain close to the SiO_2 interface, which was measured as low in at. oxygen, dominates the superconductive response but fails to describe the oxidation of the full film.





References

[1] J. Halbritter, "On the oxidation and on the superconductivity of niobium", Appl. Phys. A 43, 1 (1987).

[2] M.D. Henry, S. Wolfley, T. Monson, B.G. Clark, E. Shaner, R. Jarecki, "Stress dependent oxidation of sputtered niobium and effects on superconductivity," *Journal of Applied Physics*, vol. 115, 2014.

[3] R. Ghez, "On the Mott-Cabrera oxidation rate equation and the inverse-logarithmic law", J. Chem. Phys. A 58, 5 (1973).

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National Nuclear Security Administration

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