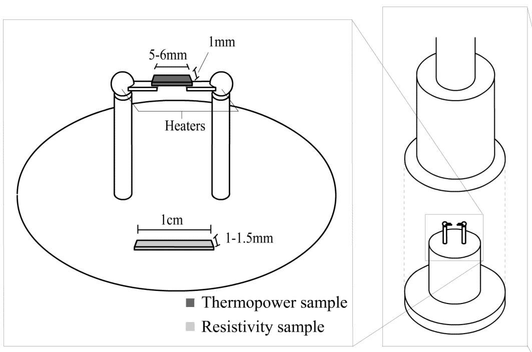
Greg Nelson traveled to the Institute of Physics in Zagreb, Croatia, for a seven week period in the summer of 2005. His research plan was to measure the thermopower and resistivity at low temperatures of Tantalum deficient Tantalum Nitride, which has the potential to be used as a barrier material for high-speed digital electronics based on superconducting Josephson junctions. It also is a material that has an interesting metal-insulator transition as a function of the Tantalum vacancies that does not occur near particle-hole symmetry.[1] Both Georgetown and Arizona State University do not have the capability of making these measurements in house, so the collaboration was beneficial to all parties, and necessary for completion of the project.

Mr. Nelson took with him six thin film samples of Tantalum deficient Tantalum Nitride grown on Silicon Dioxide at ASU by Dr. Lei Yu, a recent Ph.D. from Prof. Nate Newman's group. The samples corresponded to Ta_xN with x=.5, .6, .7, .8, 1. When in Croatia, he worked directly with Dr. Miroslav Ocko by preparing the samples for measurement of low-temperature transport properties. The preparation of the samples was much more complicated compared to crystal-grown samples, which are the type Dr. Ocko usually works with. The samples from ASU were circular wafers measuring about 4 inches in diameter; the sample sizes needed for measuring the resistivity and thermopower using his apparatus were 1cm x 1-1.5mm and 5-6mm x 1.5mm, as shown in the diagram. Four samples suitable for measurement were made from each wafer of TaN. There were no specialized tools available for making cuts of the accuracy needed.

Mr. Nelson set up a clamp to hold a Dremel-type handheld hobby saw, and, with the assistance of a stereoscopic microscope, cut the samples after gluing them with GE varnish onto a flat sheet of aluminum which was fed through the blade by hand. With this setup and much practice, Mr. Nelson prepared the samples by slowly lowering the blade onto the middle of the sample, as they would usually fracture irregularly otherwise. Great care was also taken not to damage the surface of the finished samples in any way, so that the electrical contacts later made on the surfaces would not be affected. Following the sample preparation, the samples were measured with the complex apparatus Dr. Ocko had assembled, modeled in fig. 1 and pictured below.



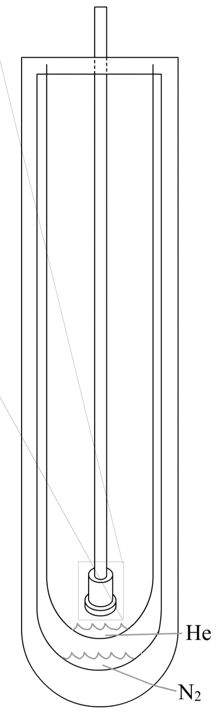
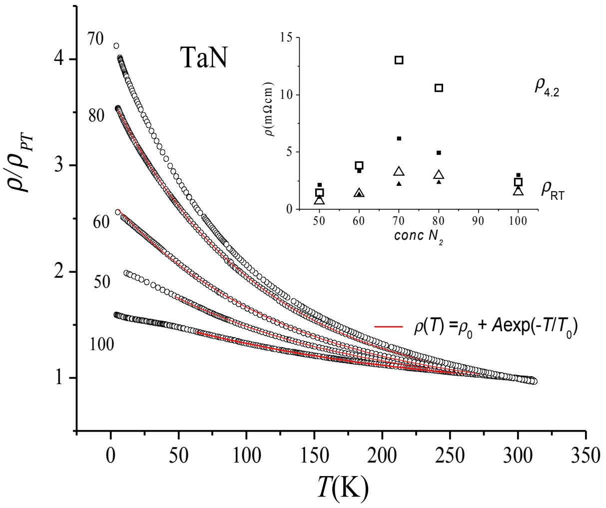


Figure 1: Low temperature part of apparatus. Vacuum insulation not shown. Current lines to heaters are independent and allow creation of temperature gradient from either side. GE varnish was used to secure the samples and provide electrical isolation from surface. Contacts to copper wires were made with silver paint on opposite ends of each sample (30μm dia., not shown) and run up to instruments through central pipe for measurement. Temperature difference across the sample for thermopower were measured with AuFeO.07%-chromel thermocouples (80μm dia., not shown). Cooling from room temperature to ~87K provided by liquid N_2. A very small amount of liquid He is then added in the bottom of the main chamber where the sample holder is housed, which provides further cooling down to ~2K.

Mr. Nelson was in charge of manually adjusting the level of liquid nitrogen in the apparatus as well as the current through the heating elements in order to maintain a relatively constant temperature as the resistance of the sample under an applied voltage was measured. The thermopower was calculated by measuring the resulting voltage across the sample under the application of a small temperature gradient created by varying the power to each of the heaters and dividing that voltage by the temperature gradient. The temperature gradient was controlled by a personal computer, as was the data processing. These were all new laboratory techniques for Mr. Nelson. The laboratory phase of the project was successfully completed with the thermopower (the ratio of the potential difference and the temperature gradient) and resistivity having been measured for the six samples.



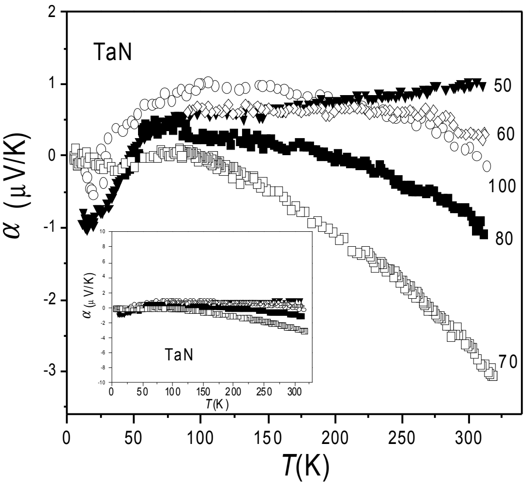


Figure 3 (left): Resistivity/(Resistivity at room temperature) graph. Inset shows room temperature resistivity(triangles) and resistivity at 4.2 K(squares), with open symbols showing measurements from Croatia with TaN grown on SiO_2 and closed symbols for those from ASU for TaN on sapphire. Graph by Dr. Ocko.
Figure 4: Thermopower graph. Inset is a version of the larger graph scaled to show the y-axis from -10 to 10. Graph by Dr. Ocko.

The results were curious for two reasons. First, the thermopower appeared to be much lower in magnitude than expected for many of the samples, and it was often below the sensitivity of the instruments at low temperature (similar measurements made on a sapphire substrate by another group showed significantly larger thermopower values). Second, the resistivity decreased exponentially in temperature as T was raised, which is likely a disorder effect, since it has also been observed in Dr. Ocko's lab for some disordered intermetallic alloys, such as $Zr_{36}Ni_{64}$ (for a conventional insulator, the resistivity should decrease exponentially in 1/T not T).

Upon returning to Georgetown in the beginning of July, Mr. Nelson worked with Dr. Tahvildar-Zadeh and Prof. Freericks to try to model the measured transport via the Falicov-Kimball model, the simplest quantum model of a solid that features electron-electron interactions.[2] At a basic level it considers itinerant (conduction) and localized (scattering) particles on a lattice, the Coulomb interactions between the two types when they occupy the same lattice site (the electron-electron interactions that cause scattering), and the effects of different densities of the two types on the lattice. One can obtain the energy-dependent relaxation time from the solutions of the model with different values of the Coulomb interaction and density of itinerant electrons and scattering electrons. The numeric values for the relaxation time function were obtained with a computer program written by Dr. Tahvildar-Zadeh.[3] Using the following equations

$$\rho(T) = \frac{ha}{e^2}\left[\frac{1}{k_B T}\int_{-\infty}^{+\infty}\frac{t(\varepsilon)\,d\varepsilon}{(\exp(\beta\varepsilon)+1)(\exp(-\beta\varepsilon)+1)}\right]^{-1}$$

$$S(T) = -\frac{k_B}{|e|\,T}\times\left(\frac{\int_{-\infty}^{+\infty}\frac{t(\varepsilon)\,\varepsilon\,d\varepsilon}{(\exp(\beta\varepsilon)+1)(1+\exp(-\beta\varepsilon))}}{\int_{-\infty}^{+\infty}\frac{t(\varepsilon)\,d\varepsilon}{(\exp(\beta\varepsilon)+1)(1+\exp(-\beta\varepsilon))}}\right)$$

where n = number of conduction electrons per unit volume, $\beta = 1/k_bT$, a = lattice spacing, one can obtain the resistivity (ρ) and thermopower (S) as functions of temperature from the model using the relaxation time $t(\varepsilon)$.[4]

It was thought that by tuning the concentration of the conduction electrons and the concentration of scattering centers (in the model) to the corresponding concentration of Ta vacancies in Tantalum Nitride, theoretical results that matched the experimental observations could be produced. Using this interpretation, the hopping between lattice sites remains the same, the Coulomb interaction between conduction electrons and the Tantalum vacancies remains the same, and the concentration of electrons is tied to the concentration of Tantalum vacancies, which is the variable adjusted to fit the data. This modeling procedure was unable to fit this data, due to an inability to model the strange T dependence of the resistivity. Some fits for the resistivity could be generated, but only by varying the hopping constant (which reflects the energy at which the particles can move to different lattice sites) and the strength of the Coulomb interaction for the scattering sites *for each sample*, even though in the model, the hopping constant should be the same for different amounts of Tantalum vacancies. Another sign that the model did not correspond to the physics of the samples was that varying the concentration of the scattering and conduction electrons was not required to make these fits – in fact, it almost always made it impossible to generate fits to match the data once the scatterer and electron concentrations were moved away from about half-filling, where they are equal, and it is easy to generate an insulating phase that comes close to the exponential behavior of the resistivity. The Tantalum vacancies should change the average number of conduction electrons and concentration of scattering sites in the Falicov-Kimball model applied to Ta_xN, and the disagreement with the experimental data showed that the FK model does not capture the physics of the material when grown on SiO_2. The thermopower measurements also were in total disagreement with the predictions generated by the model; there were disagreements in sign and monotonicity (the theoretical predications gave a monotonic thermopower curve).

Currently, it is our general consensus that the problems with the data stem from the fact that the films involve disordered grains of materials and transport along the films, as measured in Croatia, measures how well the grains are interconnected. What is of interest for applications is, however, the transport in the perpendicular direction, since that is the direction of current flow in tri-layered devices such as a Josephson junction, made from a sandwich of a superconductor on top of the barrier (made from Ta_xN) on top of another superconductor, which have the potential to be used as components of high performance digital electronics. Unfortunately, due to this issue of connectivity and disorder of the grains, one cannot infer the intrinsic properties of the perpendicular transport by measuring the transverse transport. Recent work from the ASU group appears to agree with such a picture.[5]

During his time in Croatia, Mr. Nelson was often surprised with the level of quality and resourcefulness of the experimental scientists who worked and lived with a comparatively limited funding base. He has come to appreciate the ingenuity required in experimental work; although it is obvious that some scientist designed the once novel setups that are used in experiments, meeting and working with Dr. Ocko showed him the effectiveness of executing simple solutions to problems rigorously. Not having a large amount of money to buy new equipment that may solve unforeseen difficulties necessitates methods that do not hinge on expensive instruments that do a large amount of work for the researcher, as evidenced by the low-tech solution to sample preparation that nonetheless worked very well. Mr. Nelson's experience also highlighted the fundamental role that the collaboration between theorists and experimentalists holds in physics research.

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