Superconducting-Insulating Quantum Phase Transition in Homogenous Thin Films

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ABSTRACT

Superconductivity can break down in one of two ways. Either the amplitude of the order parameter $\Psi$ diminishes to zero, or fluctuations in the phase of the order parameter destroy the coherence of the condensate, turning it into either a normal metal, or an insulator. Recent experiments have shown evidence that, in certain materials, the fluctuations in the phase that normally would extinguish the supercurrent can be suppressed, and superconducting behavior ensues. For this experiment, two-dimensional amorphous Molybdenum-Germanium (MoGe) films were chosen as the superconducting material. By covering this film with a layer of Gold-Palladium (AuPd), we seek for a suppression of the fluctuations and an enhancement of superconductivity. This effect was not observed to take place in a 3nm film covered with AuPd at temperatures of 2.0 Kelvin and above.

I. Background

When electrons in a superconducting material pair to form Cooper pairs, they no longer behave like fermions, as normal electrons do. The pairing of electrons creates a boson of charge $2e$, and these bosons can occupy a single quantum state. The current flowing through a superconducting material can thus be treated with a macroscopic wave function. This function is called the order parameter, and is given by $\Psi = |\Psi| e^{i\phi}$, where $|\Psi|$ is the magnitude and $\phi$ is the phase [1].

In two-dimensional films at zero magnetic field, the breakdown of coherence can be attributed to the suppression of the amplitude of the current wave. This happens due to a decrease in the thickness of a sample, increasing its sheet resistance ($R$) beyond the resistance quantum for cooper pairs, $R_Q = h/4e^2 \approx 6.5 \text{ k}\Omega$. It is hypothesized that when the resistance of a sample is near this critical limit, quantum fluctuations in the phase state of the supercurrent wave take over. This allows Cooper pairs to tunnel from one phase to another, causing an uncertainty in the phase of the order parameter, and consequently destroying the global coherence of the supercurrent. The point at which this happens is called the superconductor-insulator transition.

In the sense of superconductivity, a two dimensional material is one whose thickness, $d$, is less than its coherence length, $\xi$. The dynamics of superconducting behavior in this limit are particularly interesting, and have been studied at length recently, specifically the behavior around the superconducting-insulating transition. A variety of materials have been tested in this limit, including amorphous and granular films, as well as arrays of Josephson junctions [2]. Such materials are on the boundary of where quantum effects that affect superconductivity can become important, such as fluctuations in the phase of the current wave.

When a normal electron is placed in close proximity to a sample exhibiting this behavior, the electron becomes bound the phase of the condensate due to a voltage induced by the fluctuation. This coupling state drastically decreases the probability that the cooper pair will tunnel between different phase states, thus suppressing the effect of phase fluctuations on the supercurrent. Rimberg, et al. [2] showed this
effect by creating a normally insulating 40 x 40 array of Josephson junctions, and then placing it ~100nm away from a two dimensional electron gas, whose density they varied by an applied voltage. By changing the density of this electron gas, they were able to change the sample’s behavior from insulating to superconducting.

The purpose of this project was to recreate this effect, using amorphous films of Molybdenum-Germanium (MoGe) in place of the Josephson junction array. In place of the two-dimensional electron gas, a film of normal metal (AuPd) is placed atop the MoGe, separated by a thin (~10 nm) insulating layer of Silicon. A problem presents itself when working with thin films, however. Though it is possible to fabricate films with sheet resistances very close to $R_Q$, it is difficult. Sheet resistance increases as the thickness of the film decreases. Graybeal [3] showed that even down to film thicknesses less than one nanometer, the sheet resistance was still far less than $R_Q$. For a film 9 Angstroms thick, he measured a sheet resistance of 2.85 kΩ, which is significantly less than $R_Q$.

Yazdani and Kapitulnik [4] demonstrated a way to drive an S-I transition by placing a film in a perpendicular magnetic field. When the superconductor-insulator phase transition is driven in this way, the requirement for square resistance becomes solely a zero-field limit. Samples with square resistances equal to or above 6.5 kΩ are insulating no matter what field is applied. Samples which normally superconduct, with square resistances less than 6.5 kΩ, become insulating at a certain critical field.

MoGe, being a type-II superconductor, has two critical fields. At $H_{c1}$, the perfect diamagnetic property superconductors exhibit gives way to quantized lines of magnetic flux within the superconductor. These lines of flux, called vortices, create disturbances in the phase of the order parameter. When stationary, the Cooper pairs simply rotate around them, thus shifting the phase by $2\pi$. But vortices are mobile at temperatures greater than absolute zero, and this mobility creates fluctuations in the phase.

The main idea of this experiment, then, was to drive an S-I transition in a MoGe film, using magnetic field. Once this was done, a film of MoGe would be covered by a normal metal, and tested to see if the normal metal would reduce the phase fluctuations induced by the magnetic field.

II. Methods

Many techniques were utilized in the fabrication of the MoGe thin films. First, 3” silicon wafers were diced into quarters and cleaned in preparation for film deposition. Molybdenum-germanium films of composition $\text{Mo}_{29}\text{Ge}_{21}$ were deposited via sputtering onto the diced silicon substrate. A 10 nm layer of silicon was then sputtered on top, and served a dual purpose. First, it protected the film from oxidation due to exposure to moisture in the air. Second, it served as a barrier between the normal metal and the molybdenum-germanium films. By rotating the samples while sputtering, these films were uniformly deposited on the quarter-wafers so that samples taken from different parts of the deposited film would have approximately the same properties. A diagram of these samples is shown in Figure 1. Samples of thicknesses ranging from 1 to 15 nm were prepared in this way, and their basic properties were tested.

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**Figure 1**: A diagram of the film layers.

Portions of the films were then diced further, to a size of 5mm by 5mm, and
optical lithography was used on them to create a pattern for simple sheet resistance measurements. During this process, photoresist, a chemical highly resistant to most etching procedures, was applied to the samples, and then soft-baked at a temperature of 100 degrees Celsius for 40 seconds, so that it could easily be removed by a simple acetone wash. An optical mask was then placed over the surface and the samples were exposed to intense ultraviolet light for 5.5 seconds. The resist was then developed, allowing the exposed resist to be removed easily. Reactive ion etching using sulfur hexafluoride was then performed to remove the developed resist, and all nonessential MoGe and Si. What was left then was the film in the shape of the optical mask.

This simplified the measurement of sheet resistance for the film created. The samples were then mounted, using carbon tape, onto plastic chips. Indium dots formed the contacts holding gold wires to the surface, and the wires were soldered onto the chip. The chip was then placed into a four probe measurement system which was used to determine the resistance of the sample across the bridge (figure 2). Sheet resistance was then calculated by adding up the number of squares between the voltage contacts and dividing the measured resistance by the number of squares.

Samples were then connected to a four probe measurement that took resistance data for the portion of the film called the bridge. This was done by hooking the current and voltage leads on the sample chip up to a Keithley 2000 multimeter. The sample was connected to the Keithley through a box containing pi filters, and current and voltage signals were sent to separate PAR 113 preamps to filter the signal. The bridge in this case was a rectangular area 1000µm by 100µm, and thus it contained 10 squares. The measured resistance was then divided by 10 to acquire the sheet resistance, or resistance per square.

These measurements were used to get an idea about whether the samples would become superconducting or insulating at low temperatures, to determine which sample thickness would be the best to do magnetic measurements on. In all cases, the films were well below the critical sheet resistance for the zero field S-I transition. Critical temperature data was also taken to determine which samples could readily be made to superconduct at liquid helium temperatures.
After these initial measurements, new samples were diced into rectangular portions for measurement. Photolithography was not performed on these samples. Some samples were covered with a thin film of Gold-Palladium about 20 nm thick, which was to be the normal metal in our measurements. Each sample was mounted on a dipstick, covered in a copper canister to reduce noise, and then submerged in a bath of liquid nitrogen to cool it to approximately 77 K. When this was complete, the dipstick was removed from the nitrogen and transferred to a dewar filled with liquid helium, to cool the sample down to 4.2 K.

All resistance versus temperature data was taken using computers running LabVIEW 6.0, connected to Keithley 2000’s by a General Purpose Interface Bus (GPIB). The temperature was monitored via a four probe resistance measurement on a Cernox resistor. The high dependence of the resistance of the Cernox thermometer on temperature allows the measured four probe resistance to be converted into temperature. During the cooling process, the resistance was also closely monitored. Both temperature and resistance were recorded and plotted using a specially created program in LabVIEW. To obtain precision Resistance vs. Temperature (R-T) curves at low temperature, a high resistance wire was wound around the copper canister. A GPIB controlled voltage source was used to raise the voltage across the wire at precise increments. The wire’s resistance causes it to heat the copper canister containing the thermometer and sample resulting in a high precision R-T curve. This heating system could also be used to hold a sample at a fixed temperature by applying a constant voltage and allowing the system to come to equilibrium.

Data for measurements that took place in magnetic fields was also acquired in a similar way. The temperature and resistance data were monitored via four probe measurements as the samples were then immersed into a liquid helium dewar which contained a superconducting solenoid. When current is applied across the solenoid, a magnetic field is created at its center. This field can be held constant, or swept at a continuous rate. The solenoid used in this experiment could attain field strengths from 0 to 11.16 Tesla. To attain the higher fields, the dewar must be pumped on with a vacuum pump to decrease the temperature of the liquid inside. If this is not done, the solenoid could quench, converting all of the energy stored in the magnet into thermal energy, boiling off the helium in the dewar, and damaging the setup. A quench could also cause an explosion, if the pressure created inside the dewar by the boiling helium is great enough.

For the first type of measurement, the current in the magnet was held at constant values while the temperature, while the voltage across the heater wire was incremented to trace out R-T curves. For the second type of measurement, the temperature in the sample was held constant, while the magnitude of current through the solenoid was varied, thus varying the strength of the magnetic field. The first measurement was used to determine the existence of the S-I transition in this field range, and the second to pinpoint the exact field at which it occurred.

### III. Results

Critical temperature and sheet resistance data for the various films is shown below (Table 1). After analysis of this data, the 3nm film was chosen to take magnetic field data on. This is because its zero field critical temperature of 4.55 Kelvin is easily attainable with liquid helium.

<table>
<thead>
<tr>
<th>Thickness (nm)</th>
<th>R_\text{c} (\Omega/\square)</th>
<th>T_\text{c} (K)</th>
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<tbody>
<tr>
<td>1</td>
<td>3500.0</td>
<td>&lt;1.5</td>
</tr>
<tr>
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<td>3</td>
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</tr>
<tr>
<td>15</td>
<td>104.1</td>
<td>6.95</td>
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</tbody>
</table>

**Table 1.** R_\text{c} and T_\text{c} values for each thickness of MoGe film.
Portions of the 3nm film were then placed into the magnetic dewar. In the plain Molybdenum-Germanium film, we were able to drive an S-I transition by applying a current of 46 Amperes or greater to the solenoid. This corresponds to a field strength of 10.26 Tesla (see figure 3). At this field strength, the R-T graph was approximately linear, though it has a few fluctuations. The initial noise on the measurement was +/- 1 Ω. This was reduced during analysis by using averaging techniques to create the curves in the graph.

The field was measured to be close to 10.1 Tesla in this fashion.

AuPd covered MoGe films were then tested similarly. The S-I transition was again found to be around 10.26 Tesla (figure 5). This transition seemed to be more stable, though this was most likely due to a decrease in the amount of noise present in the measurement.

The field was then swept again to determine the critical field for these samples. A value of 10.16 Tesla was measured as the convergence point of these sweeps (figure 6).

Figure 3. Resistance vs. Temperature curves corresponding to different applied magnetic fields of 8.93 T, 9.37 T, 9.81 T, 10.26 T, 10.71 T, 11.16 T onto a plain 3nm MoGe sample with 10nm Si overlayer. The top three have are characteristic of insulating behavior, while the lower three show resistance beginning to decrease toward zero, characteristic of superconducting behavior.

Figure 4. Magnetic field sweep curves at 2.0 K, 2.5 K, and 3.0 K temperatures for the same sample as in Figure 3. The three curves all cross each other at around 10.1 K, though not in the exact same point.

To pinpoint the field strength at which supercurrent coherence breaks down, field sweeps were done at various temperatures from 2 to 3 K. The point where all of the sweep curves cross gives the zero temperature critical magnetic field. At this field strength, sample resistance is hypothesized to be independent of temperature [4], though in this case some fluctuations were observed. These curves are shown in Figure 4. The critical magnetic
Figure 5. Resistance vs. Temperature curves corresponding to different applied magnetic fields of 8.93 T, 9.37 T, 10.26 T, 10.71 T, 11.16 T onto a 3nm MoGe sample with 10nm insulating Si overlayer, and 20 nm AuPd overlayer.

Figure 6. Magnetic Field Sweep curves at 2.0 K, 2.5 K, and 3.0 K for the same sample as in Figure 5. This graph shows a clear convergence of the three sweep plots at 10.16 Tesla.

IV. Conclusions

More precise experimentation at lower temperatures may provide evidence for quantum phase suppression in these samples, but the supercurrent stabilizing effect we had hoped to measure was not evidenced by this experiment. However, our results do indicate that high magnetic fields can be used to drive an S-I transition in samples with sheet resistances lower than $R_Q$, a result that could prove useful in future projects.

V. Acknowledgements

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VI. References


