Controlling $T_c$ of Iridium films using interfacial proximity effects

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High precision calorimetry using superconducting transition edge sensors requires the use of superconducting films with a suitable $T_c$, depending on the application. To advance high-precision macrocalorimetry, we require low-$T_c$ films that are easy to fabricate. A simple and effective way to suppress $T_c$ of superconducting Iridium through the proximity effect is demonstrated by using Ir/Pt bilayers as well as Au/Ir/Au trilayers. While Ir/Au films fabricated by applying heat to the substrate during Ir deposition have been used in the past for superconducting sensors, we present results of $T_c$ suppression on Iridium by deposition at room temperature in Au/Ir/Au trilayers and Ir/Pt bilayers in the range of ~20-100 mK. Measurements of the relative impedance between the Ir/Pt bilayers and Au/Ir/Au trilayers fabricated show factor of ~10 higher values in the Ir/Pt case. These new films could play a key role in the development of scalable superconducting transition edge sensors that require low-$T_c$ films to minimize heat capacity and maximize energy resolution, while keeping high-yield fabrication methods.

I. MOTIVATION AND BACKGROUND

The first glimpse of physics beyond the Standard Model of particle physics came from the observation of neutrino oscillations, which imply that neutrinos have a non-zero, albeit small, mass.5,6 This discovery has given emphasis onto the study of the Dirac or Majorana particle nature of the neutrino.7 With two leptons in the final state and none in the initial, existence of neutrinoless double-beta decay would imply lepton flavor violation by two units and provide a strong clue for theories beyond the Standard Model of particle physics.6,8 An observation of neutrinoless double-beta decay would be a direct proof that neutrinos are their own anti-particles and therefore Majorana fermions.6 Distinction of the energy from the two electrons produced in a neutrinoless double-beta decay, from other ordinary processes (backgrounds) requires the state of the art particle detectors that can provide excellent energy resolution and timing.

In the context of improving superconducting Transition Edge Sensors (TES) for applications such as next generation searches for neutrinoless double-beta decay,10,11 we are investigating how to fabricate superconducting films with a low critical temperature ($T_c$). TES technology has been widely used in experiments searching for Dark Matter12 in measuring the Cosmic Microwave Background10,11 and in the detection of x-rays, infrared and optical photons12. One of the main features of a TES is its excellent energy resolution. When voltage-biased13, the energy resolution of the detector is proportional to $\sqrt{T^2 C_{tot} T + 1/\alpha}$ where $\alpha = T \frac{dR}{dT}$, and $\beta = T \frac{dR}{dT}$. The total heat capacity can be partitioned as $C_{tot} = C_{bolo}(T^3) + C_{TES}(T) + C_{other}$, where $C_{bolo}$ is the heat capacity corresponding to the absorber (Debye model), $C_{TES}$ is the term associated with the TES material content and $C_{other}$ corresponds to other heat capacity contributions, such as those of magnetic impurities in the crystal. There is a clear dependence of the energy resolution on the temperature hence the need for low-$T_c$ TES in order to maximize the detector sensitivity.

The CUORE Upgrade with Particle IDentification, or CUPID, is a proposed next generation double-beta decay experiment that requires a low-threshold optical photon light detector. The use of a secondary bolometer as a light detector is being investigated since $\alpha$-particles (the expected main background in CUORE14,15) could be tagged by comparing the heat and light signals for each event, given that the two betas from a neutrinoless double beta event would produce both heat and light (Cherenkov light in the case of TeO$_2$) while $\alpha$ events would produce only heat in the absorber. A baseline resolution of at least 20 eV is required15 in order to achieve a rejection factor of 99.9% of the $\alpha$ background while keeping >90% of the signal events of interest. Furthermore, improve-

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ments in timing resolution could also be achieved using TES technology.

In order to achieve high production yield of low-$T_c$ TES, we are investigating how to effectively suppress $T_c$ using simple fabrication techniques at room temperature. We have studied $T_c$ suppression in superconducting Iridium by means of the proximity effect, in which a normal metal is deposited on top (or bottom) of an Iridium film. Suppression of $T_c$ in the superconducting metal near the superconductor-normal metal interface arises as part of the proximity effect. The Cooper pairs from the superconductor penetrate into the normal metal and induce superconductivity in the metal near the interface (proximity effect) while the normal electrons from the metal diffuse into the superconductor causing suppression of $T_c$ in the superconductor (inverse proximity effect). This phenomenon can be applied to superconducting thin films to tailor their $T_c$ in various applications, including TES first used by Nagel et al.\cite{14,13} and by G. Angloher et al.\cite{13} in the context of microcalorimeters for x-ray detection. We present two room temperature multilayer Iridium depositions, Ir/Pt and Au/Ir/Au. We believe these films could be useful in the development of high production yield low-$T_c$ TES in the context of next generation rare-event searches in which large arrays of detectors are needed. Furthermore, room temperature deposited films may allow application of TES technology not only in semiconductor Si or Ge wafers but also directly on to the bulk of large crystals such as TeO$_2$ or Li$_2$MoO$_4$, to name a few examples.

II. EXPERIMENTAL SETUP

A. Film fabrication

We fabricated superconducting multi-layers by sputtering deposition on high-resistivity (>10000 Ω-cm) silicon wafers, 280±10μm thick, at Argonne National Laboratory. The base pressure of the sputtering chamber was 10$^{-7}$ mbar previous to the insertion of the argon gas. The sputtering was performed with Ar gas at 3 mbar, allowing for the ignition of the plasma. For each metal target in the chamber, we kept track of the position of the target, and all depositions for the same metal were done with the same pressure, conserving the same deposition rate. There were two distinct types of sputtering conditions, one in which heat was applied during deposition of the iridium layer (followed by deposition of the normal metal at room temperature) and the rest in which the iridium base and all the sputter depositions were made at room temperature. Deposition rates of about 2.6 Å/sec for Ir, 2.9 Å/sec for Au and 2.1 Å/sec for Pt films were used. In the case of Au/Ir/Au trilayers, a 3 nm thick iridium layer was deposited prior to the trilayer in order to help the Au/Ir/Au trilayer stick to the silicon wafer. After all film layers were deposited, the wafers were diced into squares of 3 mm per side. Subsequently, the chips were attached to a copper plate using GE-varnish and wire bonded in 4-wire measurement configuration for a resistance measurement.

B. Resistance measurement and thermometry

A 4-wire resistance measurement is made using the Lakeshore model 370 AC resistance bridge\cite{21}. A 13.7 Hz excitation current is injected on one pair of leads while the voltage is measured on the other lead pair. The resistance bridge model 370 applies a phase sensitive detection technique that is used in lock-in amplifiers. The uncertainty on the resistance measurement due to the precision of the 370 AC resistance bridge is between 0.05-0.1%\cite{21}. The superconducting multilayers where installed on the mixing chamber plate of an Oxford Triton 400 dilution refrigerator unit.

The temperature measurements in the range of 50-200 mK were made using a calibrated ruthenium oxide thermometer from Lakeshore cryotronics.\cite{21} Between 30-50 mK this thermometer was calibrated against a nuclear demagnetization $^{60}$Co thermometer mounted at the center of the mixing chamber plate. Between 8-30 mK we utilized either a $^{60}$Co decay anisotropy thermometer or a Johnson noise thermometer from Magnicon\cite{22} that we calibrated against the $^{60}$Co. Systematic uncertainty between 30-200 mK is less than 1% and in the range between 8-30 mK is less than 0.5 mK.

The critical temperature of the superconducting multilayer film samples is determined by a chi-square fit of a function expressed as:

$$R = D \frac{e^{(AT+B)}}{1 + e^{(AT+B)}} + C$$

(1)
However, for the case of Ir/Au bilayer we observed that the impedance of the films should reduce proximity effect, an increase in normal metal thickness due to the difference in temperature between the mixing chamber plate and the superconducting sample during temperature scans across $T_c$.

The dominant systematic error on the $T_c$ measurement is due to the difference in temperature between the mixing chamber plate and the superconducting sample during temperature scans across $T_c$.

FIG. 2. Measurements of $T_c$ of Ir/Pt bilayers (black triangles) and Ir/Au bilayers (green triangles) in which 80 nm iridium base was deposited applying heat to the silicon wafer and then let cool until room temperature before deposition of a 20 nm platinum layer and 160 nm gold layer respectively.

where $R$ is the resistance, $T$ is the temperature, $D$ is fixed as the maximum resistance and $C$ is a nonzero parasitic resistance. The critical temperature is determined by:

$$T_c = -\frac{B}{A} \quad (2)$$

The conditions of the sputtering chamber for Pt deposition were varied and in which the thickness of Ir/Au and Ir/Pt bilayers in which both the iridium (100 nm) and platinum (20-70 nm) layers were deposited at room temperature. A bias current of 3.16 $\mu$A was used and the uncertainty in the $T_c$ measurement includes the difference between scans increasing or decreasing in temperature. The critical temperature is determined by:

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III. IRIDIUM $T_c$ SUPPRESSION RESULTS

A. Ir/Au and Ir/Pt bilayers

We investigated $T_c$ suppression on Ir/Au and Ir/Pt sputtered at room temperature. In principle, due to the proximity effect, an increase in normal metal thickness should reduce $T_c$. We found this to be true for Ir/Pt all the way to our lowest cryostat temperature ($\sim$8mK). However, for the case of Ir/Au bilayer we observed that after a Au thickness of $\sim$200 nm, adding more Au would instead increase the $T_c$ of the the Ir/Au bilayer. This effect could be associated with the way the films were being fabricated. We moved on to produce Au/Ir/Au trilayers at room temperature in which $T_c$ could be suppressed more effectively on both sides of the iridium, as shown on Section III B.

We also investigated Ir/Au and Ir/Pt films in which heating was applied to the silicon wafer during deposition of the iridium base film. Once the iridium deposition is done, we turn off the heater and let the sample cool off to room temperature, at which point we proceed to sputter the Au or Ir film. We fabricated a set of samples in which only the applied heat during iridium deposition was varied and in which the thickness of Ir/Au and Ir/Pt was fixed. Results from this set of samples are shown in Figure 2 in which a factor of 8mK. In Figure 2, the critical temperature of the Ir/Au bilayer was obtained. The conditions of the sputtering chamber for Pt deposition were varied and in which the thickness of Ir/Au and Ir/Pt bilayers in which both the iridium (100 nm) and platinum (20-70 nm) layers were deposited at room temperature. A bias current of 3.16 $\mu$A was used and the uncertainty in the $T_c$ measurement includes the difference between scans increasing or decreasing in temperature.

The critical temperature of the Ir/Au bilayer was obtained. The conditions of the sputtering chamber for Pt deposition were varied and in which the thickness of Ir/Au and Ir/Pt bilayers in which both the iridium (100 nm) and platinum (20-70 nm) layers were deposited at room temperature. A bias current of 3.16 $\mu$A was used and the uncertainty in the $T_c$ measurement includes the difference between scans increasing or decreasing in temperature.
sitions were the same as for the Ir/Au bilayers, except now a Pt target was used. All layers were deposited at room temperature and had a 100 nm iridium thickness. The only difference between each film sample was the thickness of the Pt layer deposited on top. Results of the $T_c$ measurements are shown in Figure 3. We observe suppression of $T_c$ from $\sim$110 mK to $\sim$20 mK for a Pt thickness of 20 nm and 80 nm respectively. Figure 3 bottom shows the temperature vs resistance curves of the Ir/Pt data shown on top. A bias current of 3.16 $\mu$A was used for all the Ir/Pt data. From the above results it is clear that Pt has a much stronger pair-breaking effect than Au on the Ir superconductor. The suppression of the superconducting temperature in S-N bilayers is affected by the density of states near the Fermi energy of the two metals and the transparency of the interference barrier. Pt has ten times higher density of electron states at Fermi level than gold, which, assuming the same microstructure of Iridium films and barrier transparency, results in approximately eight times higher effectiveness of $T_c$ suppression by Pt than by Au as shown in Figure 2. Therefore, Ir/Pt is a promising room temperature bilayer candidate for low-$T_c$ TES fabrication.

B. Au/Ir/Au trilayers

Motivated by the goal to have a room temperature fabrication using iridium and gold for low-$T_c$ films, we studied $T_c$ suppression using gold above and below a 100 nm iridium film. Using the same sputter deposition method, we investigated depositing a thinner gold layer on top and bottom of an iridium film. We fabricated Au/Ir/Au trilayers at room temperature, all of them with an iridium thickness of 100 nm. The gold thickness on top and bottom of the iridium was different for each trilayer. The results of the $T_c$ measurements of these trilayers are shown in Figure 4. $T_c$ suppression between $\sim$85 mK and $\sim$20 mK is achieved for a gold thickness of 100 nm and 400 nm respectively (top+bottom layers) for a bias current of $\sim$31.6 $\mu$A.

Temperature vs resistance data is shown on Figure 4 bottom. Ir/Pt bilayers show a normal resistance $\sim$10 times higher than the Au/Ir/Au trilayers for a similar $T_c$.

IV. CONCLUSION

We present $T_c$ suppression between $\sim$ 20 – 100 mK of Ir/Pt bilayers and Au/Ir/Au trilayers fabricated by sputtering deposition without heating the substrate. The fact that these films can be deposited at room temperature allows for the possibility to sputter the TES directly on the bulk of the crystals being used as a macrocalorimeter. We also show results of $T_c$ suppression in Ir/Au and Ir/Pt bilayers while applying heat to the substrate during deposition of the iridium. We believe these measurements to be key in triggering the development of superconducting low-$T_c$ TES sensors for large macrocalorimeter arrays such as those utilized in experiments searching for Dark Matter or neutrinoless double beta decay.

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FIG. 4. Top) Measured $T_c$ vs gold thickness on top (or bottom) for Au/Ir/Au trilayers in which all three layers were deposited at room temperature. The iridium thickness is 100 nm for all samples. The same amount of gold was deposited on top and bottom of the iridium. Bottom) Resistance vs temperature data for each Au/Ir/Au trilayer. A bias current of 31.6 $\mu$A was used.
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