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Electron-Beam Deposition of Superconducting Molybdenum Thin Films for the Development of Mo/Au TES X-ray Microcalorimeter

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***Abstract*—We are exploring the properties of electron-beam evaporated molybdenum thin films on silicon nitride coated silicon wafers at substrate temperatures between room temperature and 650 °C. The temperature dependence of film stress, transition temperature, and electrical properties are presented. X-ray diffraction measurements are performed to gain information on molybdenum crystallite size and growth. Results show the dominant influence of the crystallite size on the intrinsic properties of our films. Wafer-scale uniformity, wafer yield, and optimal thermal bias regime for TES fabrication are discussed.**

***Index Terms*—Molybdenum, superconducting, transition temperature, thin film, X-ray microcalorimeter.**

I. Introduction

SUPERCONDUCTING/Normal proximity bilayers have been the preferred choice of material for producing transition-edge sensors (TES) for a large variety of detector applications [1]. Within an established fabrication process, the transition temperature (T_C) of a TES can be tailored by simply adjusting the film thickness of each individual layer [2]. In general, quality and characteristics of such sensors, and their reproducibility through fabrication hinge strongly on understanding of the material properties of the superconducting component. Molybdenum-based bilayers (Mo/Au, or Mo/Cu) are commonly used as a reliable, thermally and electrochemically robust material combination, mainly deposited by sputtering and/or thermal deposition via electron-beam evaporation [3]. Where sputtered molybdenum films tend to have near-zero film stress on unheated substrates, our electron-beam deposition process produces low-stress superconducting films only at highly elevated substrate temperatures [4]. The latter process, however, offers an improvement in film quality of our bilayers and a potential enhancement of performance and versatility of our TES devices. In conjunction with an ultra-high vacuum environment, the control of substrate temperature adds an important process parameter through which a different regime of film growth can be explored that an unheated process does not permit.

Here we will focus on our results on the substrate temperature dependent properties of electron-beam evaporated molybdenum thin films. Thin film uniformity on a wafer scale, wafer yield and the optimal thermal bias regime for TES fabrication will be discussed.

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II. Fabrication Details

We have been using two separate ultra-high vacuum systems to exclusively deposit molybdenum and gold thin films on 4-inch silicon wafers by electron-beam evaporation. One system is a single-wafer load-lock system reaching a base pressure $\leq 2 \cdot 10^{-9}$ Torr. Its single-wafer stage offers sample rotation and heating. The heating element made out of pyrolytic graphite coated graphite (PgG) is irradiating the backside of the wafer and heating it up to maximal substrate temperature of nearly 650 °C. The other evaporation system is a multi-wafer batch system with easy front-door chamber access reaching a base pressure of $2 \cdot 10^{-8}$ Torr. Its planetary heater assembly consists of an 8-wafer carousel, which is enclosed in a stationary heater chamber. Twelve IR quartz lamps at the bottom of this chamber irradiate the front side of the wafers when passing by, heating them up to a maximum temperature of 550 °C. An opening in the heater chamber above the electron-beam source defines the deposition zone.

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In both systems the molybdenum films are deposited onto silicon wafers coated with a 0.5 to 1 μm thick low-stress LPCVD silicon nitride (SiN) in order to produce membrane suspended devices. The electron-beam target material is a 99.999% pure vacuum-melted molybdenum disc placed in a molybdenum crucible liner. The power, position and sweep of the electron-beam are adjusted to uniformly melt the molybdenum disc and to reach an optimized deposition rate of 2 Å/sec. "Floating" thermocouples in close proximity to the wafers control the wafer temperature. In addition, a band-edge thermometer (kSA, BandiT) can be used to remotely scan across the whole wafer surface generating a complete map of the wafer temperature in real-time. Our goal is to limit the substrate temperature gradient across a wafer to 10 °C, or less, by using a thermally engineered wafer holder assembly. After reaching a stable target wafer temperature, the wafers are coated and then left in the chamber to passively cool down to room temperature with the heater being off. This cooling step lasts more than 12 hours and is usually taken overnight.

III. Sample Testing

Global film stress measurements are taken before the deposited films are photolithographically patterned. A laser-based stress gauge (Frontier Semiconductor, FSM 128) is used to measure the wafer deflection before and after the film deposition. Based on mechanical properties of the wafer, the uniform thickness of the wafer and the film, the Stoney formula is applied to determine the in-plane component of the film stress across the entire film/substrate system [5], [6].

Data on transition temperature and electrical thin film properties between room temperature and 50 mK are gained by testing patterned film samples in a variety of cold platforms such as a LHe-4 dipstick and a He-3/He-4 dilution refrigerator. Resistance measurements are carried out with a low-noise 4-wire AC resistance bridge (Linear Research, LR-700) capable of sensing resistance values down to the sub-mΩ range.

In order to investigate the crystal structure of our films, we use an x-ray powder diffractometer (Inel, Equinox 100) assuming a polycrystalline structure for our films. A diffraction pattern is obtained at a fixed angle of x-ray incidence for which the x-ray penetration depth exceeds the film thickness. The film composition, its crystal phase, local intrinsic strain and mean crystallite size can be determined by measuring the position, intensity and the width of the diffraction peaks. Since the azimuth angle is fixed in x-ray powder diffractometry, conclusions on the preference of specific crystal orientations (texturing) are omitted.

IV. Thin Film Properties

The results on the characteristics of molybdenum films deposited on a heated SiN coated silicon wafer is best presented using T_C and film stress as the most prominent film parameters regarding TES fabrication. Fig. 1 displays these two parameters as they change with wafer temperature ranging from room temperature (unheated) to 650 °C. All measurements shown were taken of 50–60 nm thick film samples deposited in our single-wafer system. The wafer temperature is measured before the start of the deposition process.

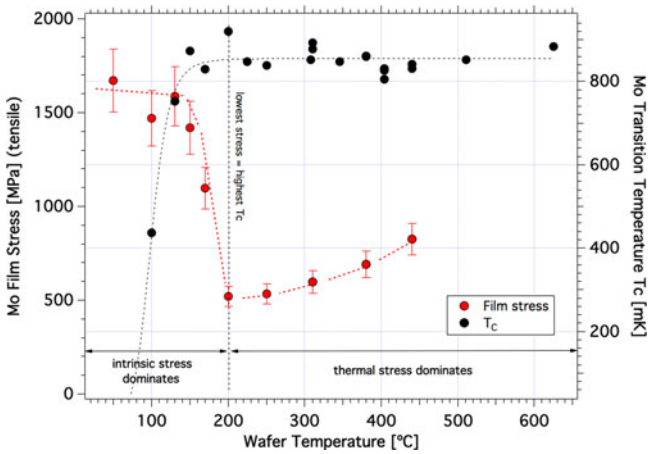

Fig. 1. Wafer temperature dependence of film stress and transition temperature for 50–60-nm-thick molybdenum films deposited by electron-beam evaporation (dotted lines are guides for the eye).

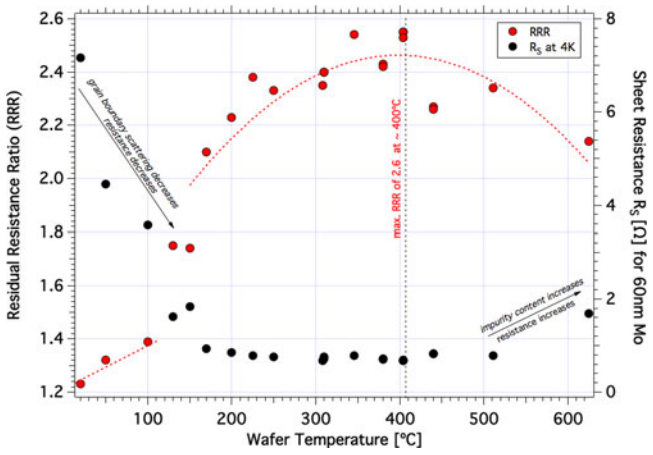

Fig. 2. Sheet resistance and residual resistance ratio of 60-nm-thick molybdenum films (dotted lines are guides for the eye).

The global film stress of our molybdenum films is exclusively tensile. The film stress curve experiences a sharp drop between 100 °C and 200 °C, where intrinsic stress declines and extrinsic stress, here thermal, becomes the dominant part. The transition temperature seems to follow an inverse trend, peaking at about 200 °C at the T_C value of bulk Mo and indicating a firm relationship between film stress and T_C. Above 200 °C the transition temperature becomes much less dependent on the wafer temperature with a slight slope to lower values up to 450 °C. The width in the transition temperature also improves steadily to less than 2 mK. Samples deposited at wafer temperatures below 70 °C are not superconducting at or above our lowest operational bath temperature of about 50 mK.

The temperature dependence of electrical sheet resistance R_S at 4 K and residual resistance ratio RRR($RRR = R_{300K}/R_{4K}$) is shown in Fig. 2. Both reveal a large change at the same wafer temperature, where Tc and film stress show their transition-like behavior. Film quality reaches a maximum near 400 °C (i.e. high RRR and low R_S) and starts to decline above

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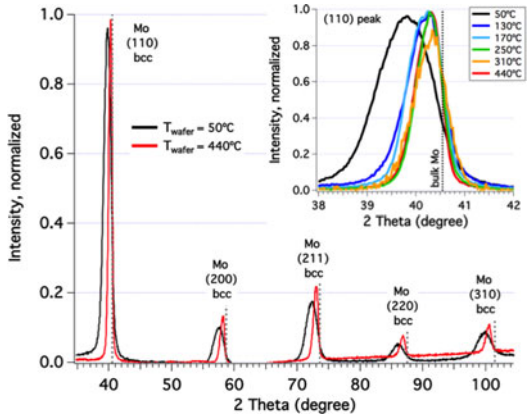

Fig. 3. X-ray diffraction traces of 50-nm-thick Mo films deposited at a wafer temperature of 50 °C and 440 °C. The inset presents the thermal evolution of the (110) peak. Dotted vertical lines indicate the peak positions of bulk Mo.

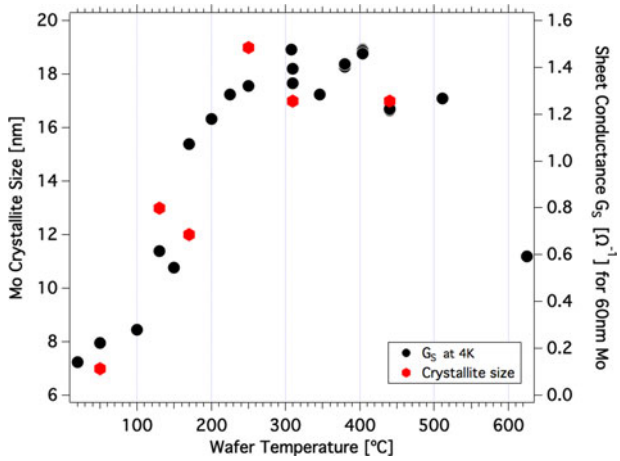

Fig. 4. Growth in Mo crystallites and sheet conductance showing conformal behavior with wafer temperature. The Warren-Averbach method was used to determine the mean crystallite size through peak shape analysis of x-ray diffraction traces.

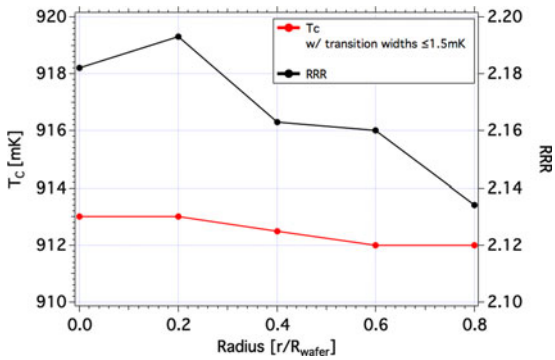

Fig. 5. Measured gradients in molybdenum transition temperature $T_{C,Mo}$ and RRR across a 4-inch wafer from center to edge. The 40-nm-thick molybdenum film was deposited at 300 °C.

this mark due to increased incorporation of impurities such as hydrogen and hydrocarbons into the growing film caused by increased chamber outgassing at higher temperatures. The negative resistance slope seen before that change (below $\sim$ 100 °C) can be attributed to a decrease in grain boundary scattering due to the increase of grain or crystallite size with wafer temperature.

To strengthen this assumption and to determine the crystal structure of our films, we examined our molybdenum samples by x-ray powder diffractometry yielding diffraction traces as shown in Fig. 3. Diffraction peaks for all wafer temperatures are strong and reproducible, indicating polycrystalline molybdenum films of mostly body-centered cubic crystal structure. The peaks shift with temperature to higher diffraction angles toward their bulk values due to the release of intrinsic strain in the films. This self-annealing process seems to be mostly completed at about 130 °C. Peak widths decrease with temperature due to the growth in crystallite size which is determined through peak shape analysis and applying the Warren-Averbach method [7]. This method permits to make the difference between broadening due to crystallite size and broadening due to microstrains. Compared to atomic force microscopy, x-ray diffractometry allows a more accurate grain size analysis of sub-micron grain structures on intrinsically rough substrate surfaces such as those of our SiN films.

Fig. 4 presents the growth in mean crystallite size together with the electrical sheet conductance. Both quantities develop conformally with wafer temperature, and reach a maximum value between around 300 °C and 400 °C. Between room temperature and 400 °C, the crystallite size has increase by 180% from about 70 Å to nearly 200 Å. In fact, when one estimates the electronic mean free path using the Sommerfeld theory of metals [8], this quantity is nearly identical to the crystallite size obtained from x-ray diffraction measurements. Furthermore, it can be concluded that the thermal evolution of the intrinsic stress in our Mo films and transition temperature, appears to be strongly influenced by Mo crystallite growth.

V. Uniformity and Yield

Our molybdenum films deposited between 200 °C and 500 °C show high wafer-scale uniformity in their thin film characteristics, an important prerequisite for successfully building very large TES detector arrays. For most of our devices, molybdenum transition temperature $T_{C,Mo}$ and molybdenum RRR values vary along a shallow gradient of $\leq \pm$ 3mK, respectively $\leq$ 3%. The latest test runs in our multi-wafer batch system yielded a $T_{C,Mo}$ gradient of only $\pm$ 1mK across a 4-inch wafer (Fig. 5). Together with a film thickness uniformity of less than $\pm$ 2% (wafer center to edge), we are able to produce Mo/Au bilayers with a T_C-gradient within $\pm$1 mK across a whole wafer.

The wafer yield strongly depends on a reproducible conditioning process of the Mo/Au interface and on the scattering of the measured $T_{C,Mo}$ values at a given target wafer temperature. Even for wafers, which are temperature biased above 200 °C the scatter in $T_{C,Mo}$ can exceed 30 mK, missing the usual target range in bilayer T_C by 10 mK. Our new multi-wafer batch system has improved our wafer yield significantly by limiting $T_{C,Mo}$ scatter to 10 mK, or less, within a wafer batch. In addition, its capability of applying an ion source cleaning process to the molybdenum top surface prior to the gold deposition, ensures a more stable and reproducible Mo/Au interface. Generally, the gold is deposited onto the bottom molybdenum layer at room temperature without breaking the ultra-high vacuum of the chamber.

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Taking into account all the presented results, we can determine an operational thermal bias regime for our films, which stretches between 200 °C and 500 °C. For best film quality of our Mo films and consequently bilayers, the sub-range of 300 °C to 400 °C is normally preferred for TES device fabrication.

VI. Conclusion

Since the introduction of molybdenum-based TES by our group almost two decades ago [3], we have developed and continuously improved the fabrication process of electron-beam deposited molybdenum films for Mo/Au TES x-ray microcalorimeters. The quality of our bilayers highly depends on the thin film quality and uniformity of the underlying molybdenum film. Molybdenum films prepared at wafer temperatures between 300 °C and 400 °C got the advantage of combining highest film quality with wafer-scale uniformity.

Furthermore, the intrinsically lower and wider T_C target range of our electron-beam evaporated molybdenum films ($T_{C,Mo} \sim$ 0.75 K to 0.92 K) compared to that of our standard sputtered films ($T_{C,Mo} \sim$ 1.0 K) lead to a wider and lower range in gold layer thickness needed in order to yield the same bilayer T_C. Hence, the normal resistance value of a TES, which is dominated by its top gold layer, can be more easily adjusted to better accommodate specific detector readout requirements without changing the lateral dimensions of the TES.

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