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Authors

Baldasseroni, C
Queen, DR
Cooke, David W
et al.

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Heat transfer simulation and thermal measurements of microfabricated x-ray transparent heater stages

C. Baldasseroni,^{1,a)} D. R. Queen,² David W. Cooke,² K. Maize,³ A. Shakouri,³ and F. Hellman²

¹*Department of Materials Science and Engineering, University of California Berkeley, Berkeley, California 94720, USA*

²*Department of Physics, University of California Berkeley, Berkeley, California 94720, USA*

³*Department of Electrical Engineering, University of California Santa Cruz, Santa Cruz, California 95064, USA*

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A microfabricated amorphous silicon nitride membrane-based nanocalorimeter is proposed to be suitable for an x-ray transparent sample platform with low power heating and built-in temperature sensing. In this work, thermal characterization in both air and vacuum are analyzed experimentally and via simulation. Infrared microscopy and thermoreflectance microscopy are used for thermal imaging of the sample area in air. While a reasonably large isothermal area is found on the sample area, the temperature homogeneity of the entire sample area is low, limiting use of the device as a heater stage in air or other gases. A simulation model that includes conduction, as well as radiation and convection heat loss, is presented with radiation and convection parameters determined experimentally. Simulated temperature distributions show that the homogeneity can be improved by using a thicker thermal conduction layer or reducing the pressure of the gas in the environment but neither are good solutions for the proposed use. A new simple design that has improved temperature homogeneity and a larger isothermal area while maintaining a thin thermal conduction layer is proposed and fabricated. This new design enables applications in transmission x-ray microscopes and spectroscopy setups at atmospheric pressure. © 2011 American Institute of Physics. [doi:10.1063/1.3640407]

I. INTRODUCTION

The advancement of x-ray optics has allowed spatially resolved x-ray spectromicroscopies using synchrotron light. These powerful analytical techniques provide information such as chemical state, phase identification and structure determination at a local level in thin films, and other nanomaterials. Temperature dependent *in situ* measurements allow chemical, phase, and structural analysis in real time. Microfabricated heaters have been used for *in situ* experiments in a transmission electron microscope¹ and for *in situ* catalysis experiments in a scanning transmission x-ray microscope,² but most *in situ* temperature dependent experiments still rely on traditional massive heater stages that require large amounts of power and are slow to stabilize.

Microfabricated membrane-based microcalorimeters have been used for well over a decade to study the thermodynamic properties of thin films and small samples.³ Our group recently reported a scaled down design: a nanocalorimeter with an amorphous silicon nitride membrane as thin as 30 nm.⁴ Figure 1 shows a top view micrograph of a typical nanocalorimeter. It consists of a silicon frame with a thin amorphous silicon nitride membrane, a platinum resistor heater, and an ensemble of platinum and niobium-silicon alloy resistor thermometers on the top. The central part of the membrane constitutes the sample area where, for example, a thin film can be deposited on the backside. The Pt resistor

heater has heater line-width = 12 μm and spacing between heater lines = 68 μm .

The thinness of the membrane makes our nanocalorimeter, a good candidate for a portable x-ray transparent platform with low power heating, rapid thermal response, and built-in temperature sensing capabilities. This new device offers a range of applications to temperature dependent spectromicroscopy studies in sensitive equipment that do not support macroscale heating of a sample platform. Applications include measurement of structure and magnetic domain changes at a phase transition and real time imaging of annealing effects on microstructure for materials ranging from magnetic films to polymers and photovoltaic thin films. For these applications, the sample area can be cycled between room temperature and hundreds of degrees in less than a second with mW of power while the frame of the device remains at room temperature. The temperature of the sample area can be measured either by the resistance of the heater itself or by one of the thermometers, allowing feedback control of the temperature.

The method used for calorimetry is based on small increases of sample area temperature above the base temperature of the frame T_0 (the so-called small- ΔT relaxation method, performed in high vacuum⁵). Many of the temperature-dependent spectromicroscopy applications require large ΔT . In addition, the typical environment of an x-ray microscope ranges from high vacuum to atmospheric pressure. We have previously performed simulation work to understand the thermal uniformity and heat transfer behavior of our device in vacuum and in the small ΔT regime for both thin films⁶ and small bulk samples.⁷ For spectromicroscopic

^{a)}Electronic mail: cbaldasseroni@berkeley.edu.

FIG. 1. (Color online) Micrograph of the nanocalorimeter. The Si-N membrane (2 mm $\times$ 2 mm) and the sample area (1 mm $\times$ 1 mm) are indicated. The arrows point to the Pt heater and thermometer. The other features on the sample area are additional thermometers and the features on the Si frame are matching thermometers to allow differential temperature measurements. The full device can be cleaved to fit on a 5 mm $\times$ 5 mm chip but the standard 10 mm $\times$ 10 mm chip is usually used.

applications, the high temperature and non-vacuum environment require that heat loss by radiation and by conduction and convection through the gaseous environment be taken into account and the consequences on the temperature uniformity of the sample area be investigated.

In this article, we first show experimental characterization of the thermal behavior of our device in both vacuum and air with large ΔT . Then, we present the numerical model that is used to simulate the steady-state temperature distribution in which heat loss contributions from conduction, radiation, and convection are analyzed. Small ΔT and large ΔT , and effects of radiation, convection, and gas pressure will be discussed. Results of simulation will be discussed and used to suggest a new design. The new design has been fabricated and thermally characterized; results will be discussed.

II. EXPERIMENTAL MEASUREMENT OF POWER AND TEMPERATURE

A. Power versus temperature measurement

First, we characterize how the power dissipation is affected by the environment (e.g., vacuum or non-vacuum) for large ΔT . The temperature of the sample area ($T_0 + \Delta T$) here is measured by the resistance of the heater itself. The first step is to calibrate the resistance versus temperature, $R(T)$, of the Pt heater. This is done in a vacuum cryostat with the Si frame attached to a sample block which has an external heater and calibrated thermometer. The sample block temperature, T_0 , is raised incrementally, and the resistance of the thin-film Pt heater is measured by a 4-wire measurement. The current I used in this $R(T)$ calibration measurement is kept small to prevent self-heating. Next, keeping the sample block and Si

FIG. 2. (Color online) Measured temperature rise ΔT of heater from $R(T)$ calibration versus power P for a nanocalorimeter in vacuum and in air. As shown in Sec. II A, in vacuum, this is the sample area average temperature; in air, because of sample area thermal inhomogeneities, this is the average heater temperature $\Delta T_{av,h}$. Inset: same data on an expanded scale to show more clearly, the data in vacuum. Sample block temperatures: $T_0 = 295$ K, 323 K, 338 K, and 363 K. Additional temperatures in vacuum: $T_0 = 200$ K and 250 K.

frame's temperature constant at some T_0 , another 4-wire measurement is performed where the current I to the heater is incrementally increased, raising the temperature of the device Pt heater while the voltage V across the Pt heater is monitored. The temperature of the sample heater ($T_0 + \Delta T$) is then determined from $R = V/I$ using the calibration curve. The power P dissipated by the sample heater is calculated from $P = VI$.

Figure 2 shows a measurement of the change in heater temperature ΔT as a function of power for different T_0 and in both air and vacuum. The curves labeled "vacuum" were taken in a high vacuum cryostat with a pressure of 4×10^{-7} Torr with the sample block held at different base temperatures T_0 ranging from 200 K to 363 K. ΔT decreases with increasing T_0 due to increasing thermal conductivities of the Si-N membrane and Pt leads with temperature. The curves labeled "air" were taken in air at atmospheric pressure with the sample block held at room temperature or on a hotplate with T_0 ranging from 323 to 363 K. These ΔT data also decrease with increasing T_0 .

A large difference in power is observed between the thermal behavior of the device in vacuum and in air. As will be later detailed, this dramatic difference is due to heat transfer through the air. In vacuum, 0.4 mW is sufficient to reach $\Delta T = 100$ K at $T_0 = 295$ K. In air, the device reaches the same $\Delta T = 100$ K with 15 mW of power.

In this experiment, the temperature measured is the average heater temperature $\Delta T_{av,h}$. For small ΔT in vacuum, we have previously shown that the sample area is isothermal to 2% of ΔT and the sample area average temperature is then the same as $\Delta T_{av,h}$. But, in situations as described in this paper (large ΔT in air), non-uniformities in temperature in the sample area are found, resulting in $\Delta T_{av,h}$ being different from the sample area average temperature, and this measurement lacks information about the spatial temperature distribution.

Microscale thermography provides a complete spatial temperature distribution of the sample area which can then be compared to 2D simulation results presented in Sec. III. However, limitations of the thermography techniques used, which are inherent to the specifics of our device, were discovered and will be discussed.

Two complementary microscale thermography techniques were used to perform thermal imaging of a nanocalorimeter in air: infrared microscopy and thermoreflectance microscopy.⁸ Materials that are good reflectors can be easily imaged by thermoreflectance microscopy, while materials that are good emitters can be easily imaged by infrared microscopy. The membrane, consisting of thin highly transparent Si-N, is not suitable for either of these imaging techniques. Because, we are unable to directly measure its temperature the Si-N outside of the sample area is not shown. Metal films (Pt heater and thermometer on the front, or Au film on the back, commonly used in our group to provide a thermal equilibration of the sample area) allow us to use these techniques to measure the sample area temperature distribution. Thermal images of the sample area of the nanocalorimeter with a 50 nm Si-N membrane and a 45 nm Au layer on the back using both techniques are shown in Figure 3 and discussed in Secs. II B and II C.

B. Infrared microscopy

Infrared microscopy measures the infrared thermal radiation (mid-wavelength infrared radiation 2–5.5 μm) emitted from the different areas of the device with a spatial resolution of 2–4 μm and a thermal sensitivity better than 0.1 K.⁹ In addition, it provides experimental emissivity parameters needed in the simulation. In particular, this measurement shows that the Si-N has a low emissivity, contrary to what was assumed in previous work. Figure 3(a) shows a thermal map obtained by infrared microscopy. The measurement was performed at an elevated base temperature $T_0 = 90^\circ\text{C}$ to increase detection efficiency and image quality.

The first step is to calibrate the image by determining the materials' emissivity. This is done for each pixel by comparing the measured radiance to a blackbody reference. This calibration step assumes that the transmitted component is negligible. This assumption is not valid for the transparent Si-N membrane and even the metal layers are partially transparent at these wavelengths due to their thinness (40–50 nm). As a result, the infrared calibration measurement overestimates the emissivities of the Pt and Au areas of the device and, therefore, underestimates the temperature.

The measurement works best for the Pt layer (corresponding to the heater and one thermometer) as it is the thickest layer on the membrane and the least transparent. The back-side Au layer is too thin to give a good infrared image and so it appears much colder (i.e., closer to 90°C of the stage). The relative temperatures of the Pt regions are accurate to 0.4 K. In order to measure absolute temperatures, we must correct for the overestimation of the Pt emissivity and the consequent underestimation of the absolute temperature. The emissivity of Pt was measured to be $\epsilon^{\text{Pt}} = 0.155$ and is corrected to $\epsilon^{\text{Pt}} = 0.12$, as explained in Sec. III A 2.

FIG. 3. (Color online) Experimental thermal maps showing ΔT of the sample area of a nanocalorimeter with a 45 nm Au thermal conduction underlayer, a 50 nm Si-N membrane, and 50 nm Pt heaters and thermometers, obtained in air by (a) infrared thermography with heater power of 12.4 mW and an elevated base temperature $T_0 = 90^\circ\text{C}$ to improve infrared emission, (b) thermoreflectance microscopy with heater power of 11.5 mW and a base temperature of $T_0 = 23^\circ\text{C}$. Figure 3(a) shows ΔT for Pt only due to low emissivity of Au and Figure 3(b) shows ΔT for Au only due to low thermoreflectance coefficient of Pt. ΔT color scale is shown on the right of each image. (c) Composite image of the Pt area from image (a) and the Au area from image (b), as explained in text.

C. Thermoreflectance microscopy

Thermoreflectance microscopy enables thermal mapping with a sub-micron spatial resolution and a thermal sensitivity as low as 0.01 K.¹⁰ It measures a relative change of reflectivity $\mathfrak{R}$ as a function of temperature. This change is small for most materials, but it is larger for Au than Pt by over an

order of magnitude. It, therefore, provides a good thermal map of the Au thermal conduction layer and is shown in Figure 3(b) which displays a non-uniform temperature distribution of Au. Since the image is not calibrated pixel by pixel, but rather with one single value corresponding to the Au, both the Pt and Si-N appear cold due to much smaller change in $\mathcal{R}$. In our measurement, the change of reflectivity of the Pt and Si-N layers is lost in noise.

The relative temperatures of the Au regions in Figure 3(b) are accurate to 0.5 K. In order to put an absolute temperature scale onto the Au data in Figure 3(b), we need the thermorefectance coefficient $d\mathcal{R}/dT$. In addition to being material-dependent, the thermorefectance coefficient $d\mathcal{R}/dT$ is also wavelength dependent and is strongly affected by interferences with covering layers. The literature value of $d\mathcal{R}/dT = -2.2 \times 10^{-4} \text{ K}^{-1}$ for bare Au. This value and the non-monotonic effect of overlayers is discussed in Ref. 11. Au covered with the 30 nm Si-N membrane was found in our experiment to have $|d\mathcal{R}/dT| = 8.5 \times 10^{-4} \text{ K}^{-1}$ at a wavelength of 470 nm.¹² Figure 3(b) was taken with a 50 nm membrane. To get the value of $d\mathcal{R}/dT$ for this sample, we use the heater $R(T)$ calibration, from which we calculate $\Delta T_{av,h}$ and from this and the data in Figure 3(b), we derive $d\mathcal{R}/dT = -3.6 \times 10^{-4} \text{ K}^{-1}$, which is within the expected range.

Figure 3(a), therefore, provides the temperature distribution of the Pt heater and thermometer with $T_0 = 90^\circ\text{C}$ and Figure 3(b) provides the temperature distribution of the Au thermal conduction layer with $T_0 = 23^\circ\text{C}$. Both data sets are taken in air and show that the central sample area has a reasonably uniform temperature. A similar $\Delta T = 100 \text{ K}$ is achieved for both measurements. The temperature distributions are similar, with small differences due to differences in base temperature T_0 , heater power and environment between the two measurements. A composite image is constructed by combining the Pt area from the infrared microscopy experimental map with the Au area from the thermorefectance microscopy experimental map in Figure 3(c). Two complementary techniques were combined to obtain the complete thermal characterization of the nanocalorimeter.

III. SIMULATION

A. Parameters determination

In order to model the heat transfer behavior of our nanocalorimeter used as a heater stage, we must take into account conduction heat transfer and two other significant contributions to heat loss not included in the 2D numerical simulation model previously reported in Refs. 6, 7: radiation and convection/conduction through the gas. The first conduction term represents the heat lost by conduction through the different layers of materials that constitute the device and is modeled by the linear Fourier's law. The heat lost by radiation is non-negligible above 100 K and is modeled based on the Stefan-Boltzmann law of blackbody radiation $P = A\sigma\epsilon T^4$ making it a non-linear term in the equations. Finally, the heat lost by convection and conduction through the gas can be ap-

proximated by a term that is linear in ΔT , using a coefficient h determined by empirical correlations.¹³

Therefore, the steady-state heat equation has the form

$$-\nabla \cdot (k_{2D} \nabla u) + hu + \sigma\epsilon((u + u_0)^4 - u_0^4) = P_{2D}, \quad (1)$$

where $u = \Delta T$, k_{2D} is the two dimensional thermal conductivity in W/K (thermal conductivity multiplied by thickness of film), h is the convection heat transfer coefficient in $\text{W K}^{-1} \text{cm}^{-2}$ (including conduction through the gas), σ is Stefan-Boltzmann constant, ϵ is the emissivity, $u_0 = T_0$ is the base temperature (for example room temperature) and P_{2D} is the two dimensional power dissipated in the heater in W/cm^2 . In this equation all parameters depend on x, y coordinates which represent the different elements of the nanocalorimeter. Two dimensional thermal conductivities are assumed to add in parallel; for example, where Au is on top of Si-N $k_{2D}^{\text{total}} = k_{2D}^{\text{Au}} + k_{2D}^{\text{Si-N}}$.

This two-dimensional non-linear partial differential equation is solved to give $u(x, y)$ with a finite element method using the MATLAB partial differential equation toolbox function `pdenonlin`. The following approximations from the previous simulation work are still assumed to be valid: (1) the 2D approximation (because the out of plane dimension is orders of magnitude smaller than all in plane dimensions) and (2) uniform dissipation of power at all x, y points in the Pt heater. In the simulation, the tolerance on the size of the residual at termination, which controls the accuracy of the computed solution, had to be increased to 10^{-12} in order for the non-linear solution to converge. Each contribution to Eq. (1) is modeled by one or several parameters. First, we discuss how each parameter is determined, and then we will turn to the simulation results.

1. Conduction

In order to determine the appropriate $k_{2D}(x, y)$ parameters, we turn to small- ΔT measurements in vacuum with different base temperatures. Figure 4 shows the thermal link of the nanocalorimeter $\kappa = P/\Delta T$ plotted as a function of its average temperature. In the limit of an isothermal sample area for small ΔT ($\Delta T \leq 0.01 T_0$), the average temperature of the sample area (Au layer) is $T_0 + \Delta T$, and the average temperature of the thermal link (Si-N membrane and Pt leads) is $T_0 + \Delta T/2$.

In vacuum, for small ΔT there are three different contributions to the thermal link between the sample area and the environment: conduction through the Si-N membrane, conduction through the Pt leads to the heater and thermometers, and the first order contribution to radiation, $A\sigma\epsilon T_0^3$. The contribution of conduction from Pt is calculated from a resistivity measurement of the Pt heater or thermometer using the Wiedemann-Franz law. The contribution from radiation is calculated from the cubic term of $\kappa(T)$. Both radiation and Pt contributions are subtracted from the measured κ to give $\kappa^{\text{Si-N}}$ as a function of temperature. A detailed description of this analysis can be found in Ref. 14.

The simulation parameter $k_{2D}^{\text{Si-N}}$ is adjusted until the simulated membrane contribution fits the experimental

FIG. 4. (Color online) Thermal link of nanocalorimeter in vacuum $\kappa = P/\Delta T$ versus average temperature of the thermal link $T_0 + \Delta T/2$, comparison of experimental data taken with small- ΔT measurement (continuous lines) and simulation data using thermal conductivities and emissivities as explained in the text (star symbols).

curve. The following relationship between the experimental thermal conductance and the two dimensional thermal conductivity used in the simulation is then determined: $\kappa^{\text{Si-N}} = 10.2 \times k_{2D}^{\text{Si-N}}$, where 10.2 is a geometric factor characteristic of the membrane, in good agreement with previous work.⁶ The two dimensional thermal conductivity of the thermal conduction layer (in our case gold) is calculated by $k_{2D}^{\text{Au}} = k^{\text{Au}} \times t$ with k^{Au} is the thermal conductivity value reported in the literature,^{15,16} and t is the thickness of the film.

For the simulations to be used in this paper, a constant value of k_{2D} is assigned to each material layer. Including a temperature-dependent conductivity would vastly increase the computational time needed to iterate to a solution and for the limits examined in this work is not relevant. In vacuum for small ΔT ($\Delta T \leq 0.01T_0$) at all T_0 , k is to first order linear in T , so taking k as a constant value corresponding to the average temperature is correct. For example, the average temperature of the membrane is $T_0 + \Delta T/2$ so the value assigned for Si-N is $k_{2D}^{\text{Si-N}}(T_0 + \Delta T/2)$. In air at temperatures above room temperature the k_{2D} of all layers will be set to their value at T_0 ; this is a good approximation because temperature-dependent deviations in thermal conduction are negligible compared to the contributions from radiation and convection. This constant approximation would *not* be true for large ΔT at low temperatures, even in vacuum, and a more complete treatment of the thermal conductivity in a simulation would then be necessary.¹⁴

2. Radiation

From the calibration done for the infrared thermography measurements shown in Sec. II C we determined the emissivity of each material on the device. This revealed that the Si-N membrane is a low emitter compared to the metals due

to its high transparency and thinness. The emissivity values of the metal layers were measured by infrared microscopy to be $\epsilon^{\text{Au}} = 0.085$ and $\epsilon^{\text{Pt}} = 0.155$, with the $\epsilon^{\text{Si-N}}$ much smaller than either. We, therefore, take the emissivity of Si-N to be zero and assume that only the metal layers on the device have a non-zero emissivity and radiate through the transparent Si-N. As discussed in Sec. II, emissivities determined by infrared microscopy were overestimated due to the partial transparency of all the materials. Not surprisingly, it is found that values smaller than the measured values are needed to obtain a good match between simulation and experimental data in Figure 4. The best match is achieved for $\epsilon^{\text{Au}} = 0.06$ and $\epsilon^{\text{Pt}} = 0.12$. These values are also in good agreement with values reported in the literature¹⁷ and will be used in all the simulation results presented here.

3. Convection and conduction through the gas

There are three different regimes for heat loss through a gaseous medium for this geometry: a turbulent regime known as classical free convection, a laminar regime where conduction through the gas occurs very much like conduction through a solid, and a molecular flow regime where the mean free path of the molecules exceeds the characteristic system size so that heat is transferred by direct molecular impingement.¹⁸ Using empirical correlations based on dimensionless parameters, the transition pressures between the different regimes were determined based on the characteristic length of our system (silicon frame thickness of 475 μm or Pt heater dimensions in the range of 300–500 μm).

The transition between laminar and turbulent regimes is determined by the Rayleigh number.^{19,20} For our system, because of the small lengthscale, the turbulent regime is estimated to occur well above 760 Torr so it is not accessible in our experimental conditions. The transition between laminar fluid flow and molecular flow is determined by the Knudsen number.^{19,20} For nitrogen, the transition is reached for $p \approx 140$ mTorr, calculated for a characteristic length $d = 475 \mu\text{m}$. Variation of the characteristic length between 300 and 500 μm gives a transition pressure ranging from 230 mTorr to 130 mTorr.

In the laminar regime the convection coefficient h can be estimated by $h = 2 \times \frac{k}{d}$ with k being the thermal conductivity of the gas and d being the characteristic length of the system (factor 2 accounts for heat loss on top and bottom of the membrane). The thermal conductivity of gases is independent of pressure in this laminar regime. If we approximate air by its main constituent nitrogen ($k = 0.026 \text{ WK}^{-1}\text{m}^{-1}$)¹⁵ we find $h = 0.0109 \text{ WK}^{-1}\text{cm}^{-2}$ at room temperature, calculated for $d = 475 \mu\text{m}$. Variation of the characteristic length between 300 and 500 μm gives a laminar regime convection coefficient h ranging from 0.02 $\text{WK}^{-1}\text{cm}^{-2}$ to 0.01 $\text{WK}^{-1}\text{cm}^{-2}$.

In the molecular regime, the coefficient becomes highly pressure-dependent and decreases linearly with pressure. It is calculated from the molecular impingement flux $J(p)$ by $h = 2 \times J \times \frac{C_V}{N_A}$ with C_V being the molar heat capacity of the

gas and N_A being the Avogadro constant.^{18,19} For example, for a pressure of 10 mTorr, $h = 3.74 \times 10^{-4} \text{ WK}^{-1}\text{cm}^{-2}$.

We vary h to get the simulated $\Delta T_{av,h}$ to match the experimental $\Delta T_{av,h}$ calculated from the thermal maps. We obtain for (a) infrared microscopy $h = 0.0151 \pm 0.0007 \text{ WK}^{-1}\text{cm}^{-2}$ and for (b) thermoreflectance microscopy $h = 0.0137 \pm 0.0006 \text{ WK}^{-1}\text{cm}^{-2}$. With the above values well within the range of calculated laminar convection coefficient (0.01 to 0.02), experimental thermal maps shown in Figure 3 are well matched (further discussed in Sec. IV).

B. Simulation results

After determining the parameters of the model, we simulate selected sets of conditions that cover the full range of applications of the device from small ΔT in high vacuum to large ΔT at atmospheric pressure. The isothermal contours of the calculated temperature solutions are shown in Figure 5. The frame T_0 is set to 295 K and 1% isothermal contours are shown. For each set of conditions a small $\Delta T_{av,h}$ of 1 K and a large $\Delta T_{av,h}$ of about 100 K are simulated. The heater power is adjusted to reach this $\Delta T_{av,h}$.

Figures 5(a) and 5(b) were simulated using our previous model that ignored radiation (appropriate for the lower T_0 discussed there).⁶ The sample is shown to be 3% isothermal (meaning that the 3% contour line is the highest contour that lies on corners of the sample area), slightly higher than the 2% previously reported at 20 K because the ratio of thermal conductivities $k_{2D}^{\text{Au}}/k_{2D}^{\text{Si-N}}$ is lower at 295 K than 20 K. No difference in thermal contour lines between the two different powers (corresponding to small ΔT and large ΔT) is observed in this linear model (no radiation). Note: this is observed because the thermal conductivities k are assumed independent of T in the simulation; differences are expected in large ΔT when including $k(T)$.

Figures 5(c) and 5(d) show that by adding the radiation term the power required to reach $\Delta T = 1 \text{ K}$ and 100 K increases somewhat and is non-linear. The sample is now only 4% isothermal, and the contour lines slightly shrink toward the center with increased power, i.e., the value of the heater power now affects the thermal contour lines. These changes in power needed to reach $\Delta T = 1 \text{ K}$ and 100 K were seen experimentally in Figure 2.

Adding a convection term greatly increases the power required to reach $\Delta T = 1 \text{ K}$ and 100 K and greatly increases the thermal non-uniformity of the sample area, as can be seen in Figures 5(e) and 5(f). In a N_2 pressure of 10 mTorr, the sample area is now only 8% isothermal. The relative radiation loss contribution is diminished compared to the convection loss contribution; as a result the power needed to reach $\Delta T = 1 \text{ K}$ and 100 K is almost linear and only small changes are seen between small ΔT and large ΔT .

Finally, the power needed to reach $\Delta T = 1 \text{ K}$ and 100 K is 50 times greater than in vacuum. Figures 5(g) and 5(h) show that in N_2 at 760 Torr, the convection heat loss is dominant. Most of the temperature gradient occurs on the sample area and the entire membrane is close to T_0 . The power needed to reach $\Delta T = 1 \text{ K}$ and 100 K is linear and no difference

FIG. 5. Comparison of simulations showing 1% isothermal contours for different operating conditions of the nanocalorimeter. Each layer of the modeled nanocalorimeter (Si-N membrane, Au thermal conduction layer and Pt heater and thermometers) is 50 nm thick. The base temperature $T_0 = 295 \text{ K}$ and the thermal conductivities input in the model are for 295 K for simplicity ($k_{2D}^{\text{Si-N}} = 1.434 \times 10^{-7} \text{ W/K}$, $k_{2D}^{\text{Pt}} = 5.824 \times 10^{-7} \text{ W/K}$ and $k_{2D}^{\text{Au}} = 1.586 \times 10^{-5} \text{ W/K}$). The heater power ranges from $2.5 \mu\text{W}$ to 10.6 mW , chosen in each case to give $\Delta T_{av,h} = 1 \text{ K}$ ("small ΔT ": a, c, e, g) or 100 K ("large ΔT ": b, d, f, h). Emissivities $\epsilon^{\text{Au}} = 0.06$ and $\epsilon^{\text{Pt}} = 0.12$ except for (a) and (b) where radiation is ignored. Parameter values used in the simulation: convection heat transfer coefficient h in $\text{WK}^{-1}\text{cm}^{-2}$ and two dimensional power dissipated in the heater P_{2D} in W/cm^2 . (a) and (b) vacuum $h = 0$, radiation ignored $\epsilon^{\text{Au}} = \epsilon^{\text{Pt}} = 0$, $P_{2D} =$ (a) 0.0056, (b) 0.56; (c) and (d) vacuum $h = 0$, $P_{2D} =$ (c) 0.0080, (d) 0.90; (e) and (f) 10 mTorr $h = 0.000374$, $P_{2D} =$ (e) 0.021, (f) 2.1; (g) and (h) air, 760 Torr, $h = 0.0109$, $P_{2D} =$ (g) 0.25, (h) 25.

can be seen between small and large ΔT regimes because at atmospheric pressure, the (linear) convection loss dominates the radiation loss.

IV. DISCUSSION

A. Vacuum

In vacuum, ignoring radiation and for small ΔT , we have previously shown that keeping the sample area better than 2% isothermal (a condition necessary for the relaxation method of heat capacity measurement to be valid) is achieved by using a thermal conduction layer of approximately the same thickness as the membrane.⁶ Here, we investigate if this still holds in the large ΔT regime, with and without radiation.

Figure 5(d) shows that at room temperature, a nanocalorimeter with a Au thermal conduction layer as thick as the membrane is 4% isothermal. This is due to decreased $k_{2D}^{\text{Au}}/k_{2D}^{\text{Si-N}}$ at 295 K compared to earlier work at low T as mentioned in Sec. III B and to radiation heat loss. Increasing the thickness of the thermal conduction layer is sufficient to recover the 2% isothermal criterion. For example, 150 nm of Au is sufficient at a base temperature of 295 K for a large $\Delta T = 200$ K for a 50 nm Si-N membrane. We note, however, that 2% of 200 K represents a temperature gradient across the sample area of 4 K, which for some purposes is too large. At room temperature using a 150 nm Au thermal conduction layer the largest ΔT where the sample area will be 1 K isothermal is 50 K. The Au thickness must be increased further to maintain a 1 K accuracy at larger ΔT .

B. Atmospheric and other non high vacuum environment

In air the sample area becomes so non-isothermal that increasing the thickness of the thermal conduction layer is not a practical solution. Even for small ΔT , use of the current design as a heater stage requires an isothermal area large enough for x-ray spectromicroscopy and the ability to extract the temperature of that particular area. Simulated temperature profiles allow us to locate areas of interest on the current nanocalorimeter design and calculate their temperature. As explained earlier, the temperature measured experimentally is $\Delta T_{av,h}$. We calculate the equivalent in the simulation by averaging 176 points over the heater.

Figure 6(a) shows a thermal map simulated in conditions corresponding to the thermoreflectance measurement (Figure 3(b)). These conditions are similar to the conditions used in the thermal distribution simulated in Figure 5(h) (large ΔT in air at 760 Torr). Using a coefficient $h = 0.0137 \text{ WK}^{-1} \text{ cm}^{-2}$, a good match is achieved for $\Delta T_{av,h}$ between the experimental measurement and the simulation ($\Delta T_{av,h} = 90$ K). Comparison between experimental and simulated temperature profiles along the horizontal direction of the nanocalorimeter is shown in Figure 6(b). Matching between the simulation and the experiment can be improved by increasing the coefficient h and decreasing the $d\mathcal{R}/dT$ used in the thermoreflectance calibration in an iterative manner. Results from the first iteration are qualitatively good enough for the purposes of this work and are shown here.

Figure 7 shows simulated isothermal contours (same simulation as Figure 6). Two symmetric maximums in the temperature distribution are located along the heater. We can distinguish a large 1% isothermal area located between two

FIG. 6. (Color online)(a) Simulated thermal map with the following parameters chosen to match the thermoreflectance measurements of Figure 3: $T_0 = 295$ K, $k_{2D}^{\text{Si-N}} = 1.434 \times 10^{-7}$ W/K, $k_{2D}^{\text{Pt}} = 5.824 \times 10^{-7}$ W/K and $k_{2D}^{\text{Au}} = 1.427 \times 10^{-5}$ W/K, $P_{2D} = 27.1$ W/cm², $\epsilon^{\text{Au}} = 0.06$ and $\epsilon^{\text{Pt}} = 0.12$, $h = 0.0137 \text{ WK}^{-1} \text{ cm}^{-2}$ (large ΔT in air, similar to Figure 5(h)). (b) Comparison of experimental and simulated temperature profiles along the horizontal profile cut indicated in (a). The experimental data points (blue circles) are from Au areas along the same horizontal profile cut of the experimental thermal map shown in Figure 3(b).

FIG. 7. (Color online) Isothermal contours simulated with the same parameters as Figure 6 (experimental conditions of thermoreflectance measurement). 5% contours are shown over the entire nanocalorimeter and 1% contours are shown in the expanded inset with the largest isothermal area highlighted.

thermometer segments, almost at the center of the device. Its temperature is 7% to 8% below the maximum temperature in the simulation, and 4% higher than $\Delta T_{av,h}$. A disk of diameter $50\text{ }\mu\text{m}$ fits in this area.

Since convection is the dominant term of heat loss, reducing it can result in substantial improvements in power consumption and uniformity of temperature distribution of the sample area. In some cases, this can be done by experimentally achievable intermediate pressures. Our simulation shows, for example, that the size of the largest 1% isothermal area increases by 50% when the pressure is decreased to 100 mTorr (not pictured), and by more than a factor of 10 when the pressure is reduced to 10 mTorr, where the molecular flow convection regime is reached (Figure 5(f)).

C. Improved design

Sections IV A and IV B show that the isothermal character of the calorimeter's sample area can often be improved by decreasing the background pressure and/or increasing the thickness of the thermal conduction layer, but in many cases these experimental conditions cannot be altered. Therefore, the intrinsic design of the heater stage needs to be improved for use as a versatile x-ray stage.

There are several current characteristics of the device that we would like to maintain as they fit the purpose of a portable x-ray transparent heater stage best. First, to maintain good x-ray transparency, it is best to keep a similar design of a thin wire-like heater so that an x-ray beam can be focused on a small sample area without any additional metal layer. We maintain the same central square sample area so that both our nanocalorimeter and our heater stage are compatible with the same deposition mask used for the sample and a metallic thermal conduction layer.⁴ The heater can serve the dual purpose of heater and thermometer. Finally, Pt is the ideal resistance temperature detector due to its linear behavior in the temperature range of interest and its excellent chemical stability at high temperatures, so we keep Pt as our heater and thermometer material.

We use experimental measurements and simulation to characterize the temperature distribution of an alternate heater geometry that consists of a double spiral Pt heater. The Pt heater line-width is increased and the spacing between Pt heater lines is reduced, compared to the nanocalorimeter geometry ($12\text{ }\mu\text{m}$ heater line-width and $68\text{ }\mu\text{m}$ between heater lines). The geometry is shown in Figure 8. The double Pt spiral has a heater line-width = $40\text{ }\mu\text{m}$ and a heater line spacing = $20\text{ }\mu\text{m}$. A device was fabricated with a 30 nm Si-N membrane and 20 nm thick Pt heater giving a room temperature resistance $R \approx 6\text{ k}\Omega$. The temperature distribution of a 30 nm thick Au layer on the back of this device was measured experimentally by thermoreflectance microscopy.

Figure 8 shows the temperature contours of the improved design simulated in air with 15.5 mW of power dissipated in the heater, providing a ΔT of about 100 K. The new heater design results in only one maximum located at the center of the sample area instead of the previous two maximums observed on the nanocalorimeter. By reducing the Pt heater

FIG. 8. (Color online) Improved design with simulated isothermal contours for $\Delta T = 108\text{ K}$ in air. The first five 1% isothermal contours are shown, which cover much of the sample area, and 5% contours are shown after that. The parameters used in the simulation are as follows: $T_0 = 295\text{ K}$, $k_{2D}^{\text{Si-N}} = 8.604 \times 10^{-8}\text{ W/K}$, $k_{2D}^{\text{Pt}} = 2.330 \times 10^{-7}\text{ W/K}$ and $k_{2D}^{\text{Au}} = 7.1348 \times 10^{-6}\text{ W/K}$, $P_{2D} = 2.33\text{ W/cm}^2$, $\epsilon^{\text{Au}} = 0.06$ and $\epsilon^{\text{Pt}} = 0.12$, $h = 0.0137\text{ WK}^{-1}\text{ cm}^{-2}$, $\Delta T = 108\text{ K}$. The double Pt spiral has a heater line-width = $40\text{ }\mu\text{m}$ and a heater line spacing = $20\text{ }\mu\text{m}$.

line spacings from $68\text{ }\mu\text{m}$ of the original nanocalorimeter to $20\text{ }\mu\text{m}$, the isothermal area is increased by a factor of more than 4 from the nanocalorimeter, resulting in an area that fits a disk of diameter $240\text{ }\mu\text{m}$.

In this optimized design, the Pt heater covers the entire sample area, therefore, the average sample area temperature is relatively well measured by the heater. $\Delta T_{av,h}$ is found to be 102 K which differs from the 1% isothermal central area temperature (between 108 and 107 K) by only 5%, thus allowing use of the direct experimental measurement of the heater resistance. If the thickness of the Au layer is doubled to 60 nm, $\Delta T_{av,h}$ is only 2% below the 1% isothermal central area temperature.

Figure 9(a) shows the experimental thermal map of the improved design measured by thermoreflectance and Figure 9(b) shows the experimental and simulated temperature profiles, plotted along the central vertical cut of the sample area, as indicated on the thermal map. The thermoreflectance measurement is calibrated for the Au layer with $|d\mathcal{R}/dT| = 8.7 \times 10^{-4}\text{ K}^{-1}$ (see Ref. 21). The Pt appears cold due to a smaller $|d\mathcal{R}/dT|$. The experimental vertical profile data are pixels corresponding to the Au layer (in between Pt segments). It is in good agreement with the vertical profile data simulated using our model (line). The results are in good agreement and show a much improved central thermal homogeneity compared to the nanocalorimeter (Figure 6(b)). The temperature gradient along the profile cut is reduced from 28 K for the nanocalorimeter to 8 K for the new device; more importantly the temperature gradient in the central 0.5 mm is reduced from 15 K to 2 K.

FIG. 9. (Color online) (a) Experimental thermal map of the new design measured by thermoreflectance microscopy and calibrated for Au.²¹ The device has a 30 nm Si-N membrane, a 30 nm Au thermal conduction underlayer, and a 20 nm thick Pt heater. (b) Temperature profile along the central vertical cut of the sample area of the improved design shown in (a). Circles are experimental temperatures from the thermoreflectance Au thermal map and the solid line is the temperature simulated in the experimental conditions of the thermoreflectance measurement. The same parameters as in Figure 8 were used in the simulation.

V. CONCLUSION

By improving our previous simulation model, we evaluated the applicability of our nanocalorimeter as an x-ray transparent heater stage at room temperature or above in both air and vacuum where the sample heater is used to produce a large temperature difference between the frame and the sample area.

Taking into account radiation, the nanocalorimeter with a thermal conduction layer of the same thickness as the membrane is 3%–4% isothermal at room temperature in vacuum and becomes increasingly less isothermal with either increasing T_0 or ΔT . While the isothermal criterion can easily be recovered in the small ΔT range by increasing the thickness of the thermal conduction layer, the thermal homogeneity rapidly decreases, when ΔT is increased.

In the use of the device as a heater stage in air we identified convection as the dominant heat loss term which leads to a very large temperature non-uniformity with the current design. At 10 mTorr, the sample area is 10% isothermal, with 90% of the ΔT occurring across the membrane border, but by 760 Torr, the sample area is extremely non-isothermal with nearly the entire membrane remaining at T_0 , even for large ΔT . Successful use of our device in non vacuum environment, therefore, requires re-design of the heater.

A simple double spiral design that achieves a relatively large isothermal sample area was fabricated. Thermal measurements and simulations show greatly increased homogeneous areas (up to 240 μm diameter at 1% homogeneity). This heater stage will enable x-ray transmission microscopy

and spectroscopy studies with easy, fast, and precise temperature control, in various environments.

The nanocalorimeter and the double spiral heater stage have been successfully used in soft x-ray transmission experiments. Changes in the spectra of alkane crystals upon melting was studied with a scanning transmission x-ray microscope where the non-homogeneous temperature of the nanocalorimeter was first observed. Then in a proof of principle experiment using the nanocalorimeter, the disappearance of magnetic domains in a Ni thin film heated above its Curie temperature (358 °C) was observed with a full field transmission x-ray microscope. The double spiral design is currently used in investigating the temperature driven magnetic transition in FeRh thin films. Results are being analyzed and will be presented in future publications.

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laminar to turbulent regime occurs at $Ra > 10^9$. At 760 Torr of nitrogen, based on the microscale dimension of our system $Ra = 0.85 \ll 10^9$ so we are far from the turbulent transition. The Knudsen number is the ratio of gas molecule mean free path to the system's characteristic length: $Kn = \ell/L$ with the mean free path given by $\ell = RT/\sqrt{2}\pi d^2 N_A p$ where d is the diameter of the gas molecules, p is the pressure of the gas, R is the universal gas constant, and N_A is the Avogadro number. In the molecular regime, the molecular impingement flux is defined by $J = N_A P/\sqrt{2\pi MRT}$ with M being the molecular weight of the gas.

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