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High-resolution thermal micro-imaging using Europium chelate luminescent coatings

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Abstract:	Micro-electronic devices often undergo significant self-heating when biased to their typical operating conditions. This paper describes a convenient optical micro-imaging technique which can be used to map and quantify such behavior. Europium thenoyltrifluoroacetonate (EuTFC) has a 612 nm luminescence line whose activation efficiency drops strongly with increasing temperature, due to T-dependent interactions between the Eu3+ ion and the organic chelating compound. This material may be readily coated on to a sample surface by thermal sublimation in vacuum. When the coating is excited with ultraviolet light (lambda <= 337 nm) an optical micro-image of the 612 nm luminescent response can be converted directly into a map of the sample surface temperature. This technique offers spatial resolution limited only by the

	microscope optics (about 1 micron) and time resolution limited by the speed of the camera employed. It offers the additional advantages of only requiring comparatively simple and non-specialized equipment, and giving a quantitative probe of sample temperature.
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Re: Submitted Manuscript

Dear Sir or Madam,

On behalf of myself and my co-authors, I would like to submit the accompanying manuscript, entitled "**High-resolution thermal micro-imaging using Europium chelate fluorescent coatings**" for publication in the Journal of Visualized Experiments.

It describes a technique by which self-heating of microelectronic devices can be directly and quantitatively imaged, using only comparatively simple optical microscopy equipment. We therefore feel that it will be of strong interest to the scientific community, and that JoVE's video format would be ideal for conveying how to implement this technique in practice, and correctly dealing with some of the subtleties involved. We would like to thank Jessica Haggett and Molly Cummings at JoVE for their help with submitting this manuscript.

Author contributions to this work were as follows:

Timothy Benseman & Ulrich Welp: Fabrication & testing of microdevices studied to demonstrate this technique.
Vitalii Vlasko-Vlasov & Wai-Kwong Kwok: Setup of thermal imaging microscope system.
Alexei Koshelev: Theoretical support.
Yang Hao & Courtney Keiser: Collection of thermal image data.
Ralu Divan: Microlithography support.
Chiharu Watanabe & Kazuo Kadowaki: Growth of single $Bi_2Sr_2CaCu_2O_8$ crystal from which the studied device was constructed.

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Yours Sincerely,

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TITLE:
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optical microscopy; fluorescence; semiconductor; cryogenics; high-temperature superconductivity; self-heating; europium chelate

SHORT ABSTRACT:
Europium thenoyltrifluoroacentonate (EuTFC) has an optical luminescence line at 612 nm, whose activation efficiency decreases strongly with temperature. If a sample coated with a thin film of this material is micro-imaged, the 612 nm luminescent response intensity may be converted into a direct map of sample surface temperature.

LONG ABSTRACT:
Micro-electronic devices often undergo significant self-heating when biased to their typical operating conditions. This paper describes a convenient optical micro-imaging technique which can be used to map and quantify such behavior. Europium thenoyltrifluoroacetonate (EuTFC) has a 612 nm luminescence line whose activation efficiency drops strongly with increasing temperature, due to *T*-dependent interactions between the Eu^{3+} ion and the organic chelating compound. This material may be readily coated on to a sample surface by thermal sublimation in vacuum. When the coating is excited with ultraviolet light ($\lambda \leq 337$ nm) an optical micro-image of the 612 nm luminescent response can be converted directly into a map of the sample surface temperature. This technique offers spatial resolution limited only by the microscope optics (about 1 micron) and time resolution limited by the speed of the camera employed. It offers the additional advantages of only requiring comparatively simple and non-specialized equipment, and giving a quantitative probe of sample temperature.

INTRODUCTION:
Many electronic devices undergo strong self-heating when electrically biased to their normal operating conditions. This is usually due to a combination of low thermal conductivity (such as in semiconductors) and high power dissipation density. Furthermore, in devices with a semiconducting-like electrical resistivity (*i.e.* with $\partial\rho/\partial T < 0$) it has long been known that there exists the possibility of localized thermal runaway under certain biasing conditions [1, 2], in which the bias current flows not uniformly through the device, but rather in narrow filaments which are associated with highly localized self-heating, typically on a scale of microns.

Understanding such self-heating physics may in some cases be essential for optimizing the design of a particular device, meaning that techniques for imaging temperature on micron scales are very useful. There has been a recent resurgence of interest in such techniques from two areas of technology development. The first of these is for imaging quench processes in high-temperature superconducting tapes in which thermal micro-imaging allows quench nucleation sites to be identified and studied [3, 4]. The second application is for understanding self-heating in stacked intrinsic Josephson junction terahertz sources, which are fabricated from $Bi_2Sr_2CaCu_2O_8$. These have the combination of low thermal conductivity and semiconductor-like electrical conductivity along the relevant direction of current flow (i.e. their crystalline *c*-axis) described above. Not only do they experimentally show complex inhomogeneous self-heating behavior [5 – 11] it has been theoretically predicted that this may be beneficial for THz power

emission [12, 13].

A number of techniques exist for imaging the temperature of a sample at microscopic length scales. The thermoluminescent technique described here was originally employed for semiconducting devices near room temperature [14 – 16] but has more recently been applied at cryogenic bath temperatures to the superconducting tapes and THz sources described above [3, 4, 10, 11]. Improvements in the resolution and signal-to-noise performance of CCD cameras have enabled considerable performance improvements in this technique over the last few decades. The Eu-coordination complex europium thenoyltrifluoroacetonate (EuTFC) has an optical luminescence which is strongly temperature dependent. The organic ligands in this complex effectively absorb UV light in a broad band around 345 nm. The energy is transferred radiation-less via intra-molecular excitations to the Eu^{3+} ion, which returns the complex to its ground state through the emission of a luminescence photon at 612 nm. The strong temperature dependence arises from the energy transfer process [17] making for a sensitive thermal probe of an object coated with this material. When the coating is excited with a near-ultraviolet source – such as an Hg short-arc lamp – regions with lower luminescence intensity correspond to higher local temperature. The resulting images are limited in spatial resolution by the resolution of the microscope optics and the wavelength of the luminescence (in practice, to around 1 micron). Depending on the signal-to-noise ratio required, time resolution is limited only by the shutter speed of the camera, and more fundamentally by the decay time of the luminescence (no more than 500 μsec) [15]. These characteristics make the technique a very fast probe of device temperature, which yields direct temperature measurements, using comparatively simple and economical equipment.

Variations of this technique published in the past by other groups have employed small concentrations of Eu-chelates dissolved in polymer films and spin-coated on to the sample surface [3, 4]. This results in a coating which is highly uniform locally, but which has significant thickness variations at steps in the sample topography – such as commonly occur in microdevices – resulting in strong spatial variations in the luminescent response which can give artifacts in the images. The technique variation which we describe here employs thermal sublimation in vacuum. Not only does this avoid the macroscopic film thickness variation problem, but the higher EuTFC concentration achieved per unit area significantly improves the sensitivity and reduces the image acquisition time. A related technique employs a coating of SiC granules on the surface instead of the EuTFC [7, 8, 9]. SiC offers temperature sensitivity comparable to the EuTFC coatings described here, but the size of the granules limits the smoothness and resolution of the resulting images.

Several other techniques exist, which offer different combinations of advantages and disadvantages. Direct infrared imaging of blackbody radiation from the sample is simple and has spatial resolution of a few microns, but is only effective when the sample is significantly above room temperature. Scanning probe thermal microscopy techniques (such as scanning thermocouple microscopy or Kelvin probe microscopy) offer excellent sensitivity and spatial resolution, but have slow image acquisition times, necessarily limited by the scanning speed of the tip, as well as requiring highly complex equipment. Scanning laser or scanning electron

beam thermal microscopy measures the voltage perturbation when a modulated beam is rastered across the surface of a current-biased device [6, 7, 18]. This offers excellent sensitivity, and is somewhat faster than scanning probe techniques, but once again requires highly complex equipment, and also gives an indirect, qualitative map of the sample temperature.

PROTOCOL:

1. Preparation of sample for coating

NOTE: If possible, remove all organic contamination from the surface of the sample to be thermally imaged. Any such contamination may react with the deposited EuTFC film and alter its luminescent response, causing position-dependent artifacts in the resulting thermal images. This is of particular importance with samples with Au surface electrodes, which tend to attract organic contamination from the atmosphere. Remove any particles or dust sitting on the sample surface at the same time, since these may result in artifacts also. The authors recommend the following procedure:

1.1) Make current and voltage connections to the devices on the sample such as superconducting bridges or resistive devices (*e.g.* wire bonds, painted-on connections using conductive epoxy, *etc.*) *before* cleaning it in preparation for thin film coating, as these steps may introduce contamination which should be removed before coating. Use Au wires if possible, since this will make it easier to connect the sample to the cryostat after the film has been deposited. (See step 4.6 below.)

1.2) Clean the sample in 100% acetone in an ultrasonic bath for 15 sec.

1.3) Without allowing the sample to dry, clean it in 100% isopropyl alcohol in an ultrasonic bath for 5 sec.

1.4) Blow the sample dry using a nitrogen gun.

1.5) If possible, clean any remaining organic residues off the sample surface using oxygen plasma ashing. To do this, use plasma power of 100 W, O_2 flow rate of 22 cm^3/sec, and gas pressure of 160 mTorr, for 60 sec. To avoid re-contamination of the sample, deposit the EuTFC coating as soon as possible after this step.

2. Preparation of coating system for EuTFC deposition

2.1) Use a sublimation source consisting of a purpose-built boat 20 × 10 × 10 mm^3 in size (l × w × h) made from stainless steel foil, enclosing a 10 Ω coil of manganin resistance wire, for operation at approximately 100 – 200 °C. Dissolve any melted-residues of EuTFC from the boat by soaking in acetone, as these will adversely affect the properties of the new film.

2.2) Rinse the boat in isopropyl alcohol.

2.3) Allow the boat to dry completely in air before proceeding to load EuTFC into it.

2.4) Protect EuTFC powder from water vapor and light while it is being stored. Thoroughly grind EuTFC powder using an agate mortar and pestle to remove any visible lumps.

NOTE: Even when the powder is protected from water vapor, it may still crystallize into large lumps of 100 microns diameter or more. These must be removed as they will result in a grossly non-uniform film when sublimated, causing artifacts in the thermal images.

2.5) Install the sample holder and sublimation source in the vacuum coating system such that the sample sits approximately 10 mm directly above the source boat (suitably oriented crystal thickness sensor to monitor the deposition rate). Connect the source boat heater leads to their associated vacuum feedthroughs.

2.6) Fill the source boat approximately 2/3 full with approximately 0.2 g of ground EuTFC powder.

2.7) Mount the sample upside-down directly above the source boat (to ensure uniformity of the deposited film), preferably using double-sided tape or sticky dots, rather than vacuum grease which may contaminate the film.

2.8) To minimize exposure of the sample surface and the EuTFC powder to the atmosphere (especially water vapor) begin evacuation of the deposition chamber using a rotary pump as soon as possible.

3. Deposition of EuTFC thin film by thermal sublimation

3.1) Pump the deposition chamber to 3×10^{-5} mbar or less, preferably using a turbo-molecular pump.

3.2) Program the crystal thickness monitor to read for a film density of 1.50 g/cm^3.

3.3) Apply 0.5 W of power to the source boat heater, to gently warm the source until the EuTFC begins to sublimate. It will take 2 - 3 minutes for the thickness monitor to begin reading an appreciable deposition rate.

3.4) Adjust the heater power to maintain a deposition rate of 6 - 7 nm/minute. Make only small, slow adjustments, as the deposition rate typically takes 1 – 2 minutes to respond to changes in power input.

NOTE: Boat temperatures sufficient to deposit more than 10 nm/minute in this configuration may cause the powder to melt in the boat, drastically reducing its surface area and thus the sublimation rate. More importantly, excessive boat temperatures may chemically alter the EuTFC and thus strongly reduce the thermal sensitivity of its luminescence.

3.5) After 200 nm (read by the thickness monitor) of film deposition, turn off the power to the source. ($\pm$ 20 nm is acceptable here, although thicknesses significantly outside this range will

result in lower film sensitivity.)

3.6) After the reading on the thickness monitor reaches zero, vent the chamber, with dry nitrogen gas. After removal, protect the sample from light and water vapor as soon as possible, by storage in a light-proof container in a vacuum dessicator.
NOTE: This will respectively prevent bleaching and chemical degradation of the EuTFC thin film.

4. Installation of sample in measurement cryostat
4.1) Place a blob of vacuum grease on the center cryostat sample stage approximately 1-2 mm in diameter. Use a sample stage comprising a copper cold finger with a circular top surface 15 mm in diameter.

NOTE: This is a sufficient size to ensure strong thermal contact between the stage and the sample when the sample is pressed down flat on top of it.

4.2) If the sample substrate is electrically conducting, isolate it from the stage by placing a 10 micron sheet of mylar on top of the grease, and a second similarly sized blob on top of the mylar.

NOTE: The authors find that it is better to use grease with a comparatively high viscosity (e.g. silicone-based high vacuum grease) than specialized heat-sinking compounds, as the latter typically contain low-viscosity components which may flow on to the top surface of the sample and contaminate its EuTFC coating.

4.3) Press the sample down on top of the grease using tweezers to apply force to two diagonally opposite corners simultaneously, and then clamp in place at least two corners, using brass screws and BeCu clamps.

NOTE: If the sample is not securely held in position, then it may drift significantly relative to the microscope when power is applied to it, making the resulting images difficult to analyze.

4.4) Make any necessary electrical connections such as for current and voltage leads from the sample to the cryostat wiring, taking care not to allow contamination (*e.g.* droplets of solder flux) to land on the EuTFC film.

NOTE: Do this by using only the smallest amount of flux which will do the job, and preferably avoid using flux for this step at all. Flux should not be necessary if Au wires are used for the connections to the sample.

4.5) Mount the sample cryostat on its xyz translation stage underneath the microscope, install its heat shield and optical window, and evacuate its sample space with a turbomolecular pump.

4.6) Cover the optical window of the cryostat with a piece of aluminum foil (or similar) to prevent bleaching of the EuTFC by ambient lighting in the room. Take care not to damage or

contaminate the microscope lens when doing this.

4.7) Cool the cryostat to the bath temperature of interest. For the samples described in this paper, this is typically between 5 K and 100 K.

NOTE: Do not allow the sample stage to sit for prolonged periods of time at temperatures between 125 K and 175 K, since in this range the EuTFC film will eventually crystallize into a polygranular state with inhomogeneous luminescence properties which can also drift over time. Cooling through this temperature range at 2 K/minute or faster will ensure that this problem does not occur. If the cryostat is accidentally left in this temperature range for too long, the EuTFC film may be reproducibly 'reset' by simply warming the cryostat to at least 190 K for 5 minutes.

5. Collection of thermal image data

5.1) Install a short-pass filter with 500 nm cut-off wavelength in the illumination optics path.

5.2) Install a band-pass filter with passband center wavelength = 610 nm, and FWHM = 10 nm, in the collection optics path.

NOTE: A narrow passband is advantageous here, since it minimizes collection of background light which contributes to the noise, but not to the signal. The filters must also be chosen to minimize spectral crosstalk between them.

5.3) Allow the light source to warm up and stabilize at its steady-state operating temperature, and allow the camera to cool to its equilibrium operating temperature. This should take around 30 minutes in both cases.

5.4) With all optical filters in place (since the focus position is wavelength-dependent) illuminate the sample and align and focus the microscope to the region of interest.

NOTE: While the sample is not being imaged, use a shutter or similar to avoid unnecessary illumination of the sample and resultant bleaching of the EuTFC film.

5.5) Collect a reference image with zero current applied to the sample. When collecting each image, make a correction for dark counts, which can vary strongly from pixel to pixel, as well as giving a significant offset to the true image counts from the luminescent signal.

NOTE: The exposure conditions used will depend on the requirements of the experiment (see discussion) but it is important to choose the exposure conditions such that the image contains no saturated pixels. The reference image is needed since the collected luminescent intensity will typically vary strongly depending on the surface reflectivity of the sample, even when its temperature is entirely uniform.

5.6) Apply electrical bias to the sample, collect an image under the same exposure conditions as

the reference, and compute the intensity ratio of these. NOTE: The level of electrical bias required depends strongly on the combination of device and self-heating behavior which are being studied. The examples presented here typically result from sample bias currents of the order of tens of mA, resulting in a few volts of bias across the device.

NOTE: If the sample has moved significantly relative to the reference image, then the pixel data should be shifted to compensate. (However, depending on the performance of the camera, this shift may introduce noise from pixel-to-pixel variations in its sensitivity to light, which is why movement of the sample should be minimized if at all possible.) If high absolute accuracy in temperature measurements is required, small drifts in the lamp intensity may be corrected for by normalizing the image-to-reference ratio to be 1 in a suitable region of the sample (*i.e*. one which is sufficiently far from the self-heated device as to be unaffected by it).

5.7) Repeat step 5.6 for all bias conditions of interest, while keeping the bath temperature constant.

5.8) Repeat steps 5.4 through to 5.7 for all bath temperatures of interest.

NOTE: Depending on the cryostat, the sample may need to be realigned and refocused at each new bath temperature.

6. Calibration of results

6.1) Collect zero-applied-current reference images sufficient to cover the entire temperature range of interest. 3 to 4 images at each temperature will suffice to establish reproducibility, while 20 K spacing will give enough data points to generate an accurate calibration curve. (See Figure 1b)

6.2) From this curve, convert the normalized intensity images into temperature maps. While the absolute luminescent intensity depends strongly on the local surface reflectivity of the sample, its normalized behavior with respect to temperature is only very weakly affected by this.

7. Sample storage and film re-use

7.1) As always, keep the film protected from bleaching by ambient light. NOTE: If necessary, the EuTFC coating on a sample can withstand repeated thermal cycling, and its properties will remain stable over a period of 2-3 weeks when kept in high vacuum.

NOTE: However, even when stored in high vacuum at room temperature, the film will degrade over 2-3 months. (Discoloration and roughening of the film can be easily seen under an optical microscope.) If this occurs on a sample which requires additional thermal images, then clean the film off and replace it as per steps 1 to 3.

REPRESENTATIVE RESULTS:

An example of a typical measurement configuration for conducting this experiment at cryogenic

bath temperatures is shown in Figure 1a, while a typical curve of 612 nm luminescent response intensity versus temperature is plotted in Figure 1b.

Figure 2 shows an example of typical thermal images of self heating in a $Bi_2Sr_2CaCu_2O_8$ THz source, which consists of a 'mesa' of stacked 'intrinsic' Josephson junctions with dimensions 300 $\times$ 60 $\times$ 0.83 microns, fabricated on the surface of a single crystal, and having a superconducting T_c of 86 K.

In such a device, the current flow is along the *c*-axis direction (*i.e.* into the plane of the page as shown in the images) due to the extremely anisotropic electrical resistivity of this material. As shown in Figure 2a, $\rho_c(T)$ for $Bi_2Sr_2CaCu_2O_8$ falls strongly with increasing temperature, allowing the possibility of thermal instabilities and localized thermal runaway under certain biasing conditions. Thermal images of the device are shown in Figure 2d, which were collected as described in the text under 160$\times$ magnification, using summed exposures of 4 $\times$ 2 sec on a 1024 $\times$ 1024 pixel CCD camera with 16-bit resolution, Peltier-cooled to -50 °C. The sample was illuminated with a short-arc Hg lamp using a 500 nm short-pass filter, and net intensity of approximately 1 W/cm^2. To avoid the requirement of normalizing the images by an un-self-heated area as described in section 5.6, the lamp was operated using a variable iris with closed-loop feedback to keep illumination intensity constant over time.

The images reveal a localized hotspot, where local self-heating gives rise to self-sustaining filament of current flowing through the device in the *c*-axis direction. In this filament, the current density is over 5 times higher than in the rest of the mesa. The current-voltage characteristic for the mesa at T_{bath} = 25 K is shown in Figure 2b. This contains hysteretic jumps associated with the nucleation/annihilation of the hotspot at around I_{bias} = 11 mA, and with the jumping of the hotspot from the electrode end of the mesa to the opposite end between 40 and 60 mA. Figure 2c shows longitudinal cross-sections of the mesa surface temperature under different bias conditions. For the camera and imaging conditions used here, the temperature noise is around $\pm$ 0.2 K, when smoothed over a diameter of 4 microns, corresponding to a 5 $\times$ 5 pixel region at this magnification. The lines visible in Figure 2d at the edges of the mesa and of the electrode are artifacts due to reflection off near-vertical sidewall surfaces.

Figure 3 shows raw image examples of situations which should be avoided as described in the protocol. Figure 3a shows a 612 nm luminescent image in which the film was sublimated using EuTFC in which mm-sized lumps were present. (See step 2.4.) These sublimated violently when heated, depositing particles of EuTFC several microns in diameter on to the sample. Figure 3b shows a sample whose EuTFC coating has crystallized into domains after 16 hours at 150 K, resulting in uneven and noisy luminescent response. (See step 4.6.)

FIGURE LEGENDS:

Figure 1: Thermal Imaging setup and typical calibration curve.

(a) Configuration of Microscope, UV light source, and cryostat with optical window, modified from reference 10. (b) Response curve normalized to 10 K for 200 nm sublimated EuTFC film.

Figure 2: $Bi_2Sr_2CaCu_2O_8$ mesa THz source: I-V characteristics & thermal images.
(a) (Main) Plot of device resistance against temperature. Blue squares plotted below T_c are values extrapolated from I-V curves shown in inset. (b) I-V characteristic showing hysteretic switching of Josephson junctions in device at T_{bath} = 25 K, for current-biased mesa. Insets (i) and (ii) show jumps in mesa resistance associated with hotspot nucleation and relocation respectively. (c) Longitudinal temperature cross-sections of mesa. (d) Thermal images at T_{bath} = 25 K, modified from reference 11, with conventional optical micrograph of mesa shown at left.

Figure 3: Examples of problems to avoid with EuTFC film.
(a) Film sublimated without removing large crystallized lumps from EuTFC powder, resulting in lumps deposited on sample. (b) Film (deposited on a different mesa) which has undergone local crystallization after 16 hours in cryostat at 150 K, showing uneven luminescent response.

DISCUSSION:
As demonstrated by our results, the technique described in this article yield high-resolution thermal images of microdevices, with good sensitivity and using only simple optical microscopy equipment. The illumination and exposure parameters which should be used, and the resulting signal-to-noise, depend on the requirements of the experiment. Here we discuss some of the considerations which limit the performance of the technique.

There are four main possible contributions to the noise in this experiment, namely photon shot noise, microscopic variation in the luminescent response of the film, variations in camera pixel sensitivity, and camera dark count shot noise. Where I is the excitation illuminance (in incident photons per unit of pixel-equivalent sample area), $F(T)$ is the T-dependent overall luminescent conversion efficiency for each pixel-equivalent area of the film (which is affected by the local film thickness), S is the CCD count yield from a pixel per incident photon (at λ = 612 nm), and D is the number of dark counts collected over exposure time t, then when averaged over P pixels, these parameters will be approximately normally distributed as follows:

$$I \sim N\left(\bar{I}, \sqrt{\bar{I}/P}\right)$$
$$F(T) \sim N\left(\bar{F}(T), \ \sigma_F(T)/\sqrt{P}\right)$$
$$S \sim N\left(\bar{S}, \ \sigma_S/\sqrt{P}\right)$$
$$D \sim N\left(\bar{D}Pt, \ \sigma_D\sqrt{Pt}\right)$$

$\sigma_F(T)$ depends on the uniformity of the EuTFC coating, while the standard deviation σ_S in pixel-to-pixel light sensitivity and dark count rate standard deviation σ_D depend on the performance of the camera. The counts collected over P pixels for time t therefore have mean:

$$\langle Counts \rangle = \bar{I}\bar{F}(T)\bar{S}Pt + \bar{D}Pt$$

where the last term corresponds to the dark count contribution, and variance:

$$\sigma^2_{Counts} = \begin{bmatrix} \bar{I}^2\bar{F}(T)^2\sigma_s^2/P + \bar{I}^2\bar{S}^2\sigma_F^2(T)/P + \bar{I}\bar{F}(T)^2\bar{S}^2/P \\ + \bar{I}^2\sigma_F^2(T)\sigma_s^2/P^2 + \bar{I}\bar{F}(T)^2\sigma_s^2/P^2 + \bar{I}\bar{S}^2\sigma_F^2(T)/P^2 \\ + \bar{I}\sigma_F^2(T)\sigma_s^2/P^3 \end{bmatrix} P^2t + \sigma_D^2Pt$$

Therefore the standard error in the measured temperature when averaged over *P* pixels with total exposure time *t* is given by:

$$\sigma_T = \frac{\sigma_{Counts}}{\langle Counts\rangle - \langle Dark\ Counts\rangle} \times \frac{1}{\bar{F}(T)} \frac{d\bar{F}(T)}{dT}$$

$$= \frac{\sqrt{\left[\begin{array}{l} \bar{I}^2 \bar{F}(T)^2 \sigma_s^2 / P + \bar{I}^2 \bar{S}^2 \sigma_F^2(T)/P + \bar{I}\bar{F}(T)^2 \bar{S}^2 / P \\ + \bar{I}^2 \sigma_F^2(T) \sigma_s^2 / P^2 + \bar{I}\bar{F}(T)^2 \sigma_s^2 / P^2 + \bar{I}\bar{S}^2 \sigma_F^2(T)/P^2 \\ + \bar{I}\sigma_F^2(T)\sigma_s^2 / P^3 \end{array}\right] P^2 t + \sigma_D^2 Pt}}{\bar{I}\bar{F}(T)\bar{S}Pt} \times \frac{d}{dT}\left[\ln \bar{F}(T)\right]$$

For a highly uniform film and a CCD with low pixel response non-uniformity , the terms in $\sigma_F(T)$ and σ_S respectively may usually be neglected. The temperature error thus simplifies to:

$$\sigma_T = \frac{\sqrt{\left[\bar{I}\bar{F}(T)^2 \bar{S}^2 / P\right] P^2 t + \sigma_D^2 Pt}}{\bar{I}\bar{F}(T)\bar{S}Pt} \times \frac{d}{dT}\left[\ln \bar{F}(T)\right]$$

$$= \frac{\sqrt{\bar{I}\bar{F}(T)^2 \bar{S}^2 + \sigma_D^2}}{\bar{I}\bar{F}(T)\bar{S}\sqrt{Pt}} \times \frac{d}{dT}\left[\ln \bar{F}(T)\right]$$

For the conditions normally employed in this technique, the rate of luminescent photon collection $IF(T)$ is of the order of 5000 photons per pixel per second. For a modern cooled CCD camera, the rate of dark counts and thus σ_D is significantly less than this, meaning that σ_T is usually limited by photon shot noise [19]. If σ_D can be neglected, then the temperature error simplifies further to:

$$\sigma_T = \frac{1}{\sqrt{\bar{I}Pt}} \times \frac{d}{dT}\left[\ln \bar{F}(T)\right]$$

Increasing the illumination intensity thus reduces the exposure time required for any given σ_T, especially in exceptional cases where the luminescent yield is low (*e.g.* at temperatures close to 300 K), and where dark counts are in fact significant. However, intense UV illumination may photodope carriers into semiconducting samples, and break Cooper pairs in superconducting ones, thereby perturbing the properties of the device being studied. In samples whose surfaces have a weak thermal path to the cold bath, strong illumination may also introduce a heat load which causes a significant rise in the sample temperature.

All of these considerations may sometimes necessitate low illumination intensities and longer exposure times. As a modification, shorter exposures may be required to image fast phenomena such as current filament oscillation or breathing modes [20], or the millisecond timescales of quench development in superconductors. Where high signal-to-noise ratios in absolute temperature measurements are required, then longer total exposure times are called for. This may require summation of multiple exposures, depending on the bit resolution of the CCD electronics. Image-intensified cameras have close to single-photon detection efficiency, and offer a more attractive trade-off between image noise, illumination intensity, averaging area, and exposure speed, albeit at higher system cost.

In summary, the thermoluminescent imaging technique which we describe here offers a direct quantitative measure of sample surface temperature, with high temporal and spatial resolution. It is also effective at a wide range of temperatures, from 5 K to over 300 K. As described in the Introduction, alternative techniques exist, but each of these offers a combination of advantages and disadvantages.

Scanning probe techniques offer excellent sensitivity, at the cost of long measurement times and highly specialized equipment. A recently-published pyro-magneto-optical technique also offers excellent sensitivity [21]. However, this technique relies on a ferrimagnetic garnet indicator crystal placed on top of the sample, which limits spatial resolution, especially where the sample is not topographically flat. At temperatures above 300 K, the luminescent yield from EuTFC becomes low, and direct imaging of infrared blackbody radiation from the sample becomes a more effective technique.

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DISCLOSURES:
The authors have nothing to disclose.

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Figure 1

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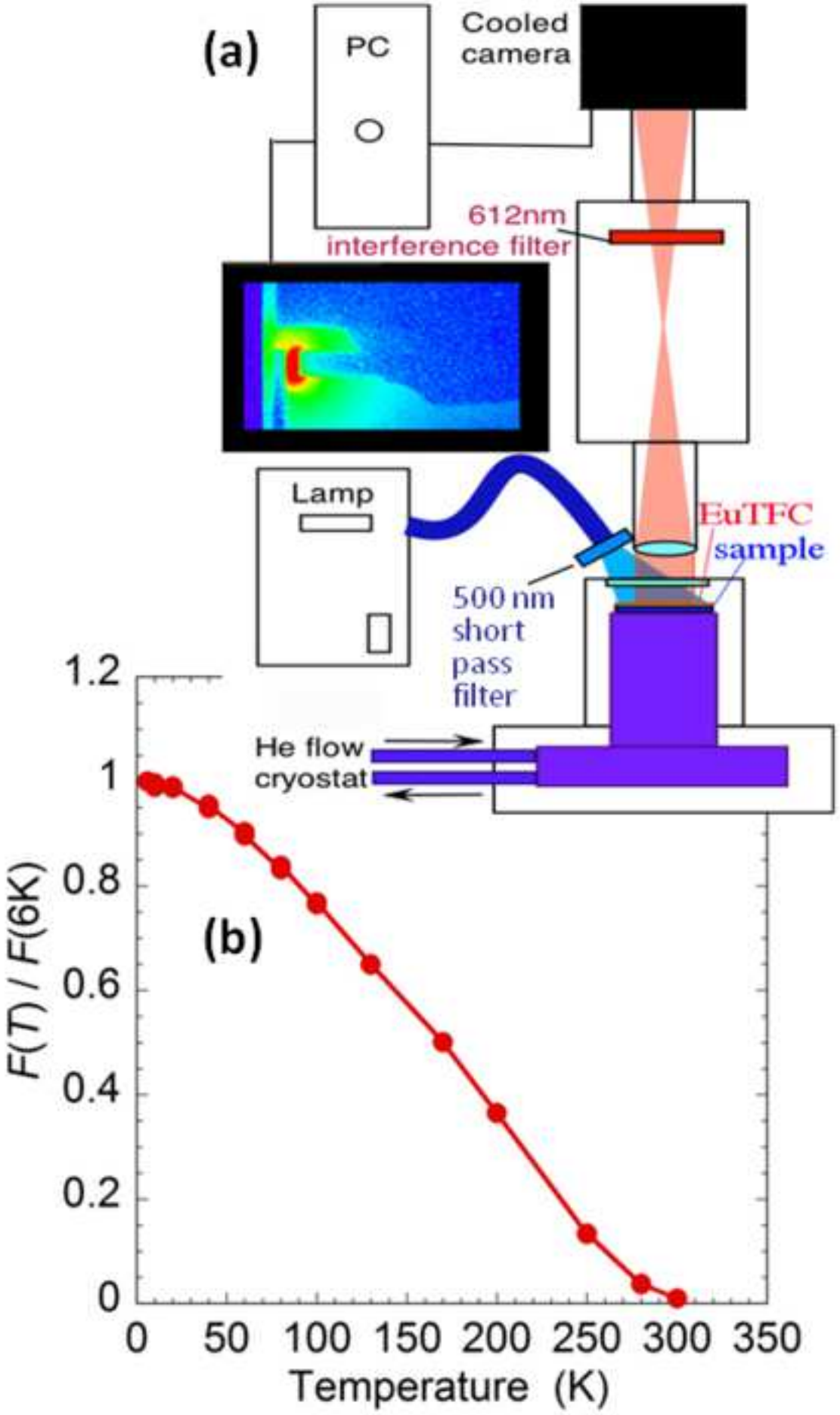

Figure 2

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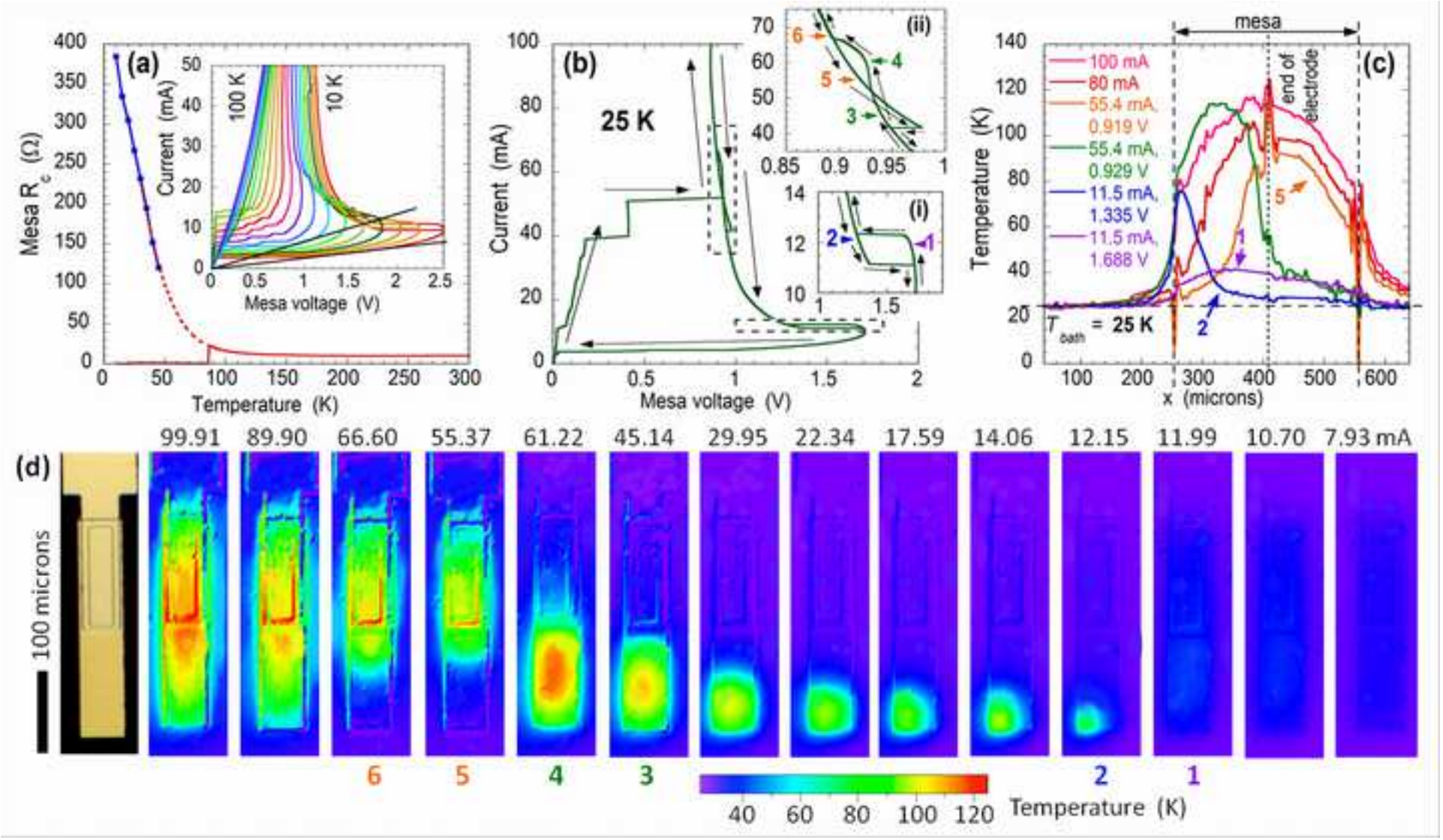

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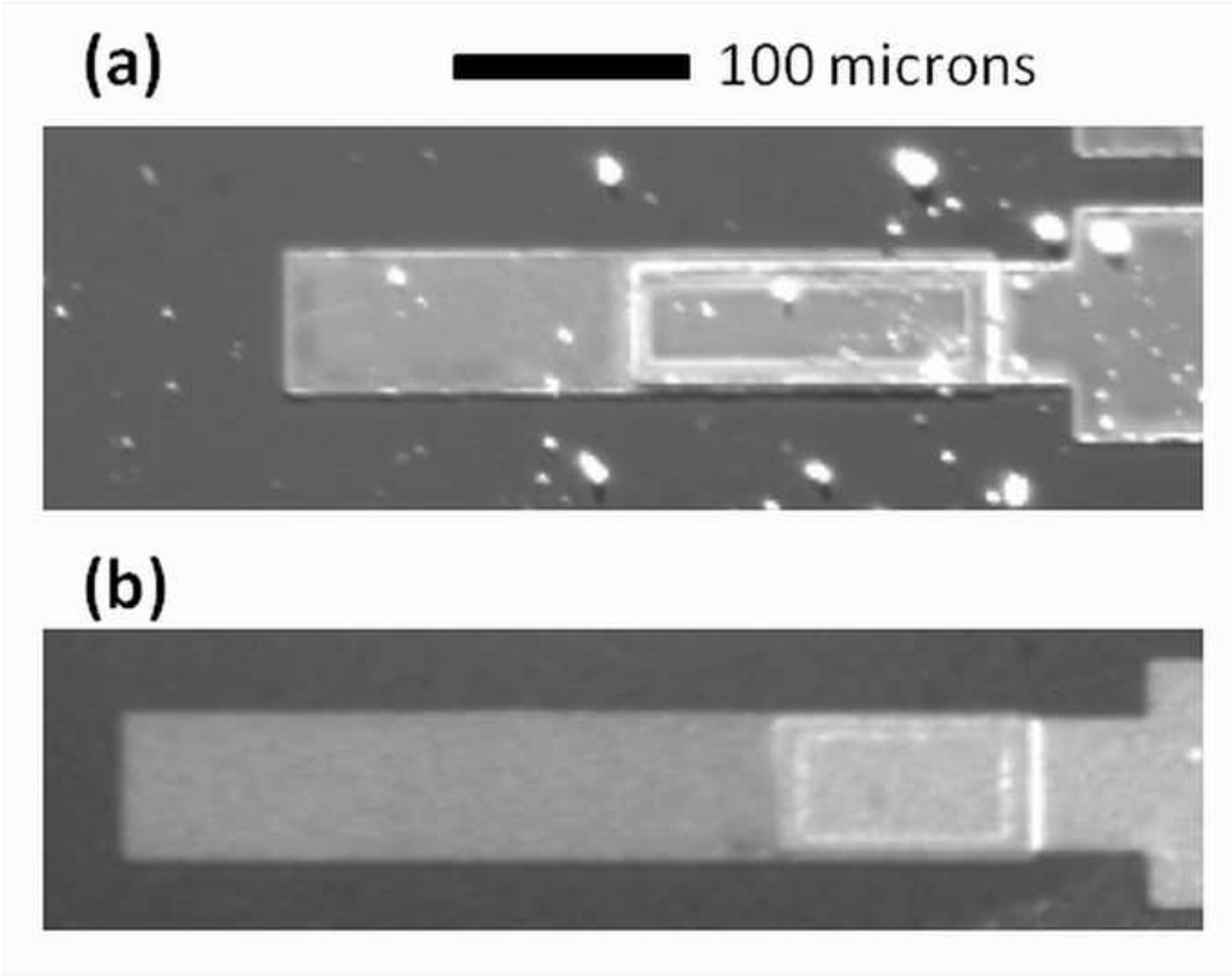

Name of Material/ Equipment	Company	Catalog Number	Comments/Description
Europium thenoyltrifluoroacetonate powder	Sigma-Aldrich	176494-1G	Also known as Europium tris[3-(trifluoromethylhydroxymethylene)-(+
Mercury short-arc lamp with flexible light guide	Lumen Dynamics	X-Cite Exacte	Light source includes internal iris and photosensor for output intensit
Peltier-cooled CCD camera	Princeton Instruments	PIXIS 1024	1024 x 1024 pixels, 16-bit resolution
610 nm band-pass filter	Edmund Optics	65-164	Passband has CWL 610 nm, FWHM 10 nm
500 nm short-pass filter	Edmund Optics	84-706	OD4 in stopband
Helium flow cryostat with optical window	Oxford Instruments	MicrostatHe2	
high vacuum grease	Dow Corning		
Digital Current source	Keithley	Model 2400	Computer-controllable current & voltage source
Digital Voltmeter	Hewlett-Packard	Model 34420A	Digital Nanovoltmeter now available as Agilent Model 34420A

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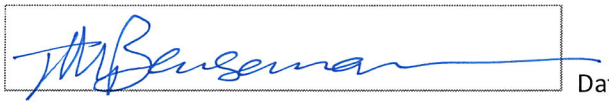
Signature:  Date: 07/15/15

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Replies to the editors comment, describing the changes we have made, are shown below in italics.

1. Please adjust the formatting of your protocol section so that all text is aligned to the left margin with no indentations.
Altered as requested - see updated text.

2. Please add a one line space between each step and sub-steps of your protocol section.
Altered as requested - see updated text.

3. Please re-write steps of your protocol section in imperative tense, as if you are telling someone how to do the technique (i.e. "Do this", "Measure that" etc.), “Use a sublimation source consisting of ….”, etc. “
Altered as requested - see updated text.

4. In step 1.1 please describe or give details of what electrical connections are made? What is the sample used? please provide specific description.
Altered as requested - see updated text.

5. Is the sample dried in between the acetone and isopropyl alcohol washes? What strength of organic solvents are used?
Altered as requested - see updated text.

6. In step 2.4 “EuTFC powder should be protected from water vapor and light while it is being stored. Even when this precaution is taken, it may still crystallize into large lumps of 100 microns diameter or more. These must be removed as they will result in a grossly non-uniform film when sublimated, causing artifacts in the thermal images” can be mentioned as a NOTE. Please mention step 2.4 as “Thoroughly grind EuTFC powder using an agate mortar and pestle to remove any visible lumps.”.
Sections requested to be mentioned as NOTEs have been identified as such in the revised protocol text. If these need to be relocated to some other section of the protocol then please do so as appropriate.

7. In step 2.4, how much of EuTFC powder is used?
About 0.2 grams (although the exact amount is not critical) - see updated text.

8. In step 2.5 how is the vacuum coating system configured? What are the settings used?
'Configuration' as used here referred to the positioning of the sample holder and source boat. This sentence has been altered to make the wording clearer.

9. In step 2.6 is the EuTFC powder just added in the boat?

Yes. Do we also need to specify the use of a suitably sized spatula?

10. In step 2.7 what is mounted upside-down? How is this mounted?
The word "sample" had been mistakenly deleted from the sentence. This error has now been corrected.

11. In step 2.8 how is the deposition chamber as evacuated?
This has been clarified in the text.

12. In step 3.1 how is the deposition chamber pumped? Using what?
Use of a turbomolecular pump (in our case the pumps are an integral part of the vacuum deposition system) has been clarified in the text.

13. In step 3.2 how is the crystal thickness monitor set?
Details of how to set this are highly specific to each individual vacuum deposition system, so we are unable to provide any further information beyond specifying which knob to turn on our particular system. Such information is of limited use to the reader.

14. In step 3.4, please mention "Boat temperatures…thermal sensitivity of its fluorescence.", as a NOTE.
Altered as requested.

15. In step 3.5 how does one know if the deposited film thickness reaches 200 nm?
Text has been clarified to describe measuring this using the crystal thickness monitor.

16. In step 3.6, how is the chamber vented? How is the sample protected from light and water vapor?
See clarifications in updated text.

17. In step 4.1, what are the dimensions of the cryostat sample stage? Please do not mention "sufficient size" please provide all the details and specifics.
Specific dimensions have been added.

18. In step 4.1, please mention "The authors find …sample and contaminate its EuTFC coating" as a NOTE.
Altered as requested.

19. Please split step 4.1 into two steps.
Altered as requested.

20. In step 4.2 how is the sample pressed down? Using what?
Use of tweezers for this step has been specified - see updated text.

21. In step 4.3, what electrical connections are made? Please specify.
Specification for current and voltage leads has been added.

22. How is step 4.4. carried out?
Details for our particular cryostat mounting arrangement have been added.

23. In step 4.6 please specify the temperature. Please mention "Do not allow…" as a NOTE.
Temperatures added as requested, but this will in general depend on the requirements of the individual experiment.

24. Using the comments above as a reference, please add details to section 5, 6 and 7. Please ask yourself the "how" question to obtain more details.
Details beyond those which we have now added are entirely specific to the particular microscope and optical cryostat system being used, and are thus of little use to readers.

25. JoVE is unable to publish manuscripts containing commercial sounding language, including trademark or registered trademark symbols (TM/R) and the mention of company brand names before an instrument or reagent. Please remove all commercial sounding language from your manuscript and replace it with a more generic term as much as possible throughout the entire manuscript. All commercial products should be sufficiently referenced in the table of materials/reagents. Examples of commercial sounding language in your manuscript are Dow Corning, etc.
This reference has been removed from the protocol, and the materials list has been updated to include the vacuum grease used, as well as the details of the current source and digital voltmeter used for this experiment.

26. After you have made all of the recommended changes to your protocol (listed above), please re-evaluate the length of your protocol section. There is a 10 page limit for the protocol text, but there is a 3 pages limit for filmable content. If your protocol is longer than 3 pages, please highlight (in yellow) 2.75 pages (or less) of text to identify which portions of the protocol are most important to include in the video; i.e. which steps should be visualized to tell the most cohesive story of your protocol steps. Please see JoVEs instructions for authors for more clarification. Remember that the non-highlighted protocol steps will remain in the manuscript and therefore will still be available to the reader.
Highlighted sections have been updated in our modified text.

27. If your figures and tables are original and not published previously, please ignore this comment. For figures and tables that have been published before, please include phrases such as “Re-print with permission from (reference#)” or “Modified from..” etc. And please send a copy of the re-print permission for JoVE’s record keeping purposes.
We are still waiting to receive approval for re-use of parts of Figures 1 and 2 from the journals in which they were originally published, and will forward this to JoVE as soon as we receive it.

28. Please make sure that the “Discussion” section covers the following points.
a. Critical steps within the protocol.
b. Modifications and troubleshooting.
c. Limitations of the technique.
d. Significance of the technique with respect to existing/alternative methods.
e. Future applications or directions after mastering this technique.
Existing/alternative methods have been discussed in detail in the introduction, and critical steps and appropriate troubleshooting have been explained at the relevant steps in the protocol. Please let us know if this material should be moved to or duplicated in the discussion section.

29. DOI’s missing in some references. Please make sure that your references comply with JoVE instructions for authors. In-text formatting: corresponding reference numbers should appear as superscripts after the appropriate statement(s) in the text of the manuscript. Citation formatting should appear as follows: (For 6 authors or less list all authors. For more than 6 authors, list only the first author then *et al.*): [Lastname, F.I., LastName, F.I., LastName, F.I. Article Title. *Source*. **Volume** (Issue), FirstPage – LastPage, doi:DOI, (YEAR).]

Reference 2 has no DOI, as it was published in 1935, and is unavailable online since it is in a journal which ceased publication in 1945.

30. Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammar issues. The JoVE editor will not copy-edit your manuscript and any errors in the submitted revision may be present in the published version.

List of changes, 10/29/15

Our responses to the referees' and editor's comments are shown in italics.

Editorial comments:

• Please download this version of the Microsoft word document (File name: 53948_R1_080315) for any subsequent changes

•Grammar: 2.4 – Please use imperative tense.

Altered as requested.

•Additional detail is required:

-4.5 – Please specify what "in position relative to the microscope" means.

Text has been altered to clarify that the sample cryostat is mounted on an xyz translation stage to allow focusing and alignment.

-How much electricity is applied to the sample?

An extra sentence has been added to specify this.

•Discussion: Please discuss the significance of the technique with respect to other methods, the critical steps of the protocol, and future applications.

Extra sentences have been added to the discussion section, in which this technique is compared to other available thermal imaging techniques.

Further comment: We have altered the as-edited wording of the first sentence of protocol step 7.1. Writing this sentence in imperative tense would obliterate its meaning, since it refers to process steps which an experimenter _can_ decide to follow if necessary.

Reviewers' comments:

Reviewer #1:

Manuscript Summary:

Joule heating is an important issue in electronic devices. This paper describes in details an optical micro-imaging technique which can be used conveniently to map and quantify temperature distribution in various devices including intrinsic Josephson junction emitters. This technique offers spatial resolution limited only by the microscope optics (about 1 micron) and time resolution limited by the speed of the camera employed. The system requires rather simple and non-specialized equipment. Based on the

detailed description of material preparation of the sensing material, Europium thenoyltrifluoroacetonate (EuTFC), system construction, imaging process, and temperature calibration, one can easily apply this technique to different circles.
The article is well written and organized. With minor revision, I hope this article can be published in JoVE.

Major Concerns:
N/A

Minor Concerns:
There are several typos.
1) Line 109, "due" should be "due to";
2) Line 414, "and used" should be "used";
3) Line 483, "signal-to noise" should be "signal-to-noise".

These typos have now been corrected.

Additional Comments to Authors:
N/A

Reviewer #2:
Manuscript Summary:
I have no concerns regarding this paper. The paper is well written and the technique is sufficiently explained. Furthermore, potential pitfalls are sufficiently indicated.

Major Concerns:
N/A

Minor Concerns:
N/A

Additional Comments to Authors:
N/A

Reviewer #3:
Manuscript Summary:
In the paper "High-resolution thermal micro-imaging using Europium chelate fluorescent coatings" by T. Benseman et al. are describing in detail a way of imaging temperature distributions using EuTFC films.

The method is based on the temperature dependent luminescence of EuTFC. Publications of the authors on that subject exist. Here they describe, and this is exactly the scope of the JoVE, their "cooking recipe" in order to quantify temperature distributions at low temperatures. The paper not only includes the imaging technique itself, but also deals with the deposition of the detection layer and the obstacles to avoid during preparation.
The paper is well written and the details given in the description seem to be nicely describing the whole process in detail. Given that, I can highly recommend the publication of the paper in JoVE.

Major Concerns:
N/A

Minor Concerns:
Only a small comment: The authors briefly discuss other optical methods, including their advantages and disadvantages. There are a few alternative methods, one recently published in Adv. Mater. 2015, DOI: 10.1002/adma.201501859.

We have added a sentence to the discussion section which briefly covers the advantages and disadvantages of this technique, and added a reference to it.

Additional Comments to Authors:
N/A

Reviewer #4:
Manuscript Summary:
In this article, a conventional optical micro-imaging technique is described by using of the temperature-sensitive luminescence of Europium thenoyltrifluoroacetonate (EuTFC). This technique offers micro-sized spatial resolution and fast response time, and only requiring comparatively simple and non-specialized equipment. Considering the quality of the paper, it is recommended to be published in the journal except for a minor revision-the term of "fluorescence" should be replaced with "luminescence", as it originates from Eu3+ ion.

The title of the manuscript has been altered as per this request, and 'fluorescence' has been replaced with 'luminescence' throughout the manuscript.

Major Concerns:
N/A

Minor Concerns:
N/A

Additional Comments to Authors:
N/A

Reviewer #5:
Manuscript Summary:
In this work, the authors describe a method of thermal imaging using Europium chelate fluorescent coatings. The active layer is coated using thermal sublimation in vacuum, which increases the homogeneity of the active layer thickness and improves the accuracy of the imaging technique.
I have several questions and suggestions related to the manuscript.

Major Concerns:
1) What is the electrical conductivity of the Europium chelate coatings that you use in your work?

As far as our measurements have been able to determine, these coatings are very good insulators for the purposes of this experiment, since depositing them on a sample surface makes no measureable difference to the resistance of the devices on it.

2) You mention the improved homogeneity of the coating thickness compared to other methods. Do you have any data on the typical thickness variation in your films?

Profilometry measurements on our deposited films show less than +/- 5 nm of surface roughness over a length scale of half a micron. This is obviously higher than the level of surface roughness which can be achieved if the film is spin-coated, as other authors have done. The advantage of thermal sublimation over spin coating is not to optimize microscopic film homogeneity, but to optimize macroscopic film homogeneity, i.e. to avoid the significant variation of thickness which occurs in spin-coated films at step edges, which are usually present in most microdevices for which this technique is of interest.

3) Can you comment on the reason why the 200 nm thickness gives you the best sensitivity (lines 256-258)? What is the dependence of the sensitivity on the film thickness?

The luminescent response scales approximately linearly with film thickness up to and beyond 200 nm. However, we find that for films substantially thicker than 200 nm, film stresses introduced upon cooling the sample to cryogenic temperatures may result in a spatially non-uniform luminescent response, thus negating any signal-to-noise benefits of a higher luminescent response due to increased film thickness. Also, as the film must be deposited slowly, sublimating a very thick film can take an impractically long time.

4) In point 7 of the protocol (lines 382-383), you mention that the film can be cleaned off. Please describe the best way to do it.

At present, the manuscript refers the reader to the steps specified in 1.2 - 1.5, namely ultrasonic cleaning in acetone followed by IPA, after which the sample should be cleaned with oxygen plasma.

Have you observed any signs of surface degradation of the studied samples due to the coating? Can you say that the technique is non-invasive, in the way that the studied samples can be reused?
Can you actually reuse the film? Line 375 reads 'film re-use'. Did you mean 'sample re-use'?

Yes, the film can actually be reused, subject to the caveats regarding film & sample storage mentioned in Section 7 of the protocol. We have observed no reaction of the sample with the coating, which is not surprising, given the completely inorganic composition of the device. We agree that for samples containing organic molecules, then the coating may react with these, and the technique could no longer be strictly considered non-invasive.

5) Figure 1 (bottom) shows that the fluorescent response approaches zero at about 300 K. Is there a way to extend the range so that the same method could be used above room temperature?

The technique can be used up to about 350 K if necessary, since while the luminescent response itself is low in this range, d ln[F(T)]/dT is still comparatively large. However, above 350 K, the near-infrared blackbody radiation from the sample is sufficiently strong that it can be readily imaged directly, obviating the need for a luminescent technique such as this.

6) On page 11 where you discuss different contributions to the noise, you ignore photon shot noise (line 453) and conclude that the experimental noise is dominated by dark counts.
However, Fellers and Davidson in their work that can be found on the Hamamatsu web site http://hamamatsu.magnet.fsu.edu/articles/ccdsnr.html say 'Cooling the CCD reduces the dark current dramatically, and in practice, high-performance cameras are usually cooled to a temperature at which dark current is negligible over a typical exposure interval.'
The photon noise however is equal to the square root of the number of photons incident on the CCD. So the more photons you get the higher the photon noise (although the signal-to-noise ratio gets also higher). It is then possible that at some level of the signal, the photon noise will become higher than the dark noise (the latter should be independent of photon-induced signal).
Can you support you conclusion with a reference or, otherwise, elaborate on this question further?

We would like to thank the referee for this helpful and carefully-considered comment. For temperatures close to 300 K where the luminescent yield is comparatively low, then for this technique the dark counts are the largest contribution to the noise, unless very powerful illumination is employed to increase the rate of photon generation. (Please note that the luminescent response drops by almost three orders of magnitude between 5 K and 300 K.) However, we agree with the referee that under most conditions, photon shot noise will be the dominant contribution to the error levels obtained, even when these are of the order of 1 part in 1000. We have therefore modified our error analysis in the discussion section to take photon shot noise into account, and have included a reference to the webpage mentioned by the

referee.

Minor Concerns:
Line 132: References can be useful after '...THz sources described above' [10, 11?]

References have been added as requested.

Line 148: I would avoid using the phrase 'comparatively very simple'.

This sentence has been altered to read ‘comparatively simple’.

Lines 151-153 'Variations of this technique published in the past...': In my opinion, specific references can be useful here as well, even if they were already given before.

References have been added as requested.

Line 335: '...zero power applied...': The phrase does not sound right to me. You can apply voltage but not power, which is the rate of changing energy.

We have altered this to read ‘zero current applied’, since in this experiment the devices studied are current-biased.

Line 364: I think that point 6 (Calibration) in the protocol should be highlighted, as it is of high importance

Highlighted as requested.

Additional Comments to Authors:
In addition, please proofread the text. There are several typos and missing words, sometimes a figure is referred as 'Figure', sometimes as 'Fig.', etc.

We have proofread the text, and altered it such that the abbreviation ‘Fig.’ is no longer used.