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.*