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Preparing a celadonite electron source and estimating its brightness

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Corresponding Author:	Evelyne Salancon Aix-Marseille Universite Marseille, FRANCE
Corresponding Author's Institution:	Aix-Marseille Universite
Corresponding Author E-Mail:	salancon@cinam.univ-mrs.fr
Order of Authors:	Evelyne Salancon
	Alain Degiovanni
	Laurent Lapena
	Roger Morin
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TITLE:

Preparing a Celadonite Electron Source and Estimating its Brightness

AUTHORS & AFFILIATIONS:

Evelyne Salançon, Alain Degiovanni, Laurent Lapena, Roger Morin

CINaM, Aix-Marseille University, CNRS, UMR 7325, Marseille, France

Alain Degiovanni: degiovanni@cinam.univ-mrs.fr
Laurent Lapena: lapena@cinam.univ-mrs.fr
Roger Morin: morin@cinam.univ-mrs.fr

CORRESPONDING AUTHOR:

Evelyne Salançon: salancon@cinam.univ-mrs.fr

KEYWORDS:

Unique crystal deposition; low-energy electron point-source; field emission; brightness estimation; electron holography; electron microscopy

SUMMARY:

The article presents a protocol to prepare a celadonite source and estimate its brightness for use in a long-range imaging low-energy electron point-source projection microscope.

ABSTRACT:

The electron celadonite source described here performs well in a low-energy electron point-source projection microscope in long-range imaging. It presents major advantages compared to sharp metal tips. Its robustness affords a lifetime of months and it can be used under relatively high pressure. The celadonite crystal is deposited at the apex of a carbon fiber, maintained itself in a coaxial structure ensuring a spherical beam shape and easy mechanical positioning to align the source, the object and the electron-optical system axis. There is a single crystal deposition via generation of celadonite-containing water droplets with a micropipette. Scanning electron microscopy observation can be performed to verify the deposition. However, this adds steps and therefore increases the risk of damaging the source. Thus, after preparation, the source is usually inserted directly under vacuum in the projection microscope. A first high voltage supply provides the kick-off needed to start the electron emission. The field emission process involved is then measured: it has already been observed for dozens of electron sources prepared in this way. The brightness is under-estimated through an over-estimation of source size, intensity at one energy and cone angle measured in a projection system.

INTRODUCTION:

Metal/insulator structures used for electron emission have been studied for almost 20 years due to their low macroscopic field[1]. The electric field involved is only of the order of some V/µm[2,3,4], in contrast to the V/nm required for classic field emission with sharp metal tips[5,6,7].

This probably explains the starting plasma discharges that are so useful in electron source technologies. Some years ago, we sought to explore this low field emission by depositing films of natural insulators on electron transmission carbon layers[8]. Celadonite, an insulator mineral found in the basalt of the Parana Traps in the mines of Ametista di Sul in Brazil, was chosen.

When celadonite is ground, the crystal shape is a rectangular slab with micrometric dimensions and a thickness of less than 100 nm (typically: 1,000 nm x 500 nm x 50 nm). It is perfectly flat and recognizable in scanning electron microscopy (**Figure 1**). The film is formed by deposition of a celadonite-containing water droplet on the carbon layer. As applied voltage increases, it emits electrons following a Fowler-Nordheim regime with intensity saturation for the highest voltages. A study using a diaphragm in a projection system showed that one emitter is a point-like source[9]. However, using this large film with a diaphragm to select the source did not exploit the potential of the point-source. For example, the point-sources commonly used in low-energy electron point-source projection microscopy allow a source-to-object distance of about 100 nm. However, such a source-to-object distance would be out of the question with a film. Finding a way to isolate one crystal so as to be able to move something towards this electron source was a challenge. Our solution was first, to use a 10 µm carbon fiber: depositing the droplet at the apex of the fiber necessarily limits the number of celadonite crystals. Second, we decided to limit droplet size: a micropipette with a tip end of about 5 µm is filled with celadonite-containing water and pressure is applied at the entrance of the micropipette to create a small drop to wet the apex of the fiber. The protocol details the full source preparation process.

The resulting source is a coaxial point-source allowing good alignment between the source, the object and the electron optical system[10]. Because its 10 µm diameter is still wider than ultra-sharp tips, the source-to-object distance is limited to some tens of micrometers. However, we recently showed that the celadonite source emitter combined with an Einzel lens performs comparably to a classic point-source projection microscope. The long-range imaging thus made accessible even limits the charge effect[11] on the object and the image distortions involved[12,13]. The celadonite source also presents major advantages compared to sharp metal tips. It is robust: the point-source is under the crystal and thus protected against sputtering. The source can operate under relatively high pressure: it was tested at 10^{-2} mbar during some minutes. Yet its lifetime and its stability remain dependent on the right vacuum conditions. We usually employ the celadonite source at 10^{-8} mbar and obtain a lifetime of months.

This article is intended to help all those wishing to use the celadonite source to produce a coherent electron beam.

PROTOCOL:

1. Preparation of the source

NOTE: In our microscope, the source-support is composed of a machinable glass ceramic plate from which emerges 1 cm of a stainless-steel tube of 90 µm internal diameter with an electrical connection on the plate.

1.1. Preparation of the fiber

1.1.1. Fix the source support under an optical microscope.

1.1.2. Insert the 10 µm carbon fiber into the stainless-steel tube. Glue the carbon fiber to the tube with silver lacquer.

1.1.3. Cut the fiber with a cutting tweezers (under a binocular microscope) so that between 100 µm and 3 mm are left outside the stainless-steel tube.

NOTE: The carbon fiber is brittle; leaving more than 1 cm outside the tube will increase the chance of the structure breaking during manipulation.

1.2. Celadonite-containing water preparation

1.2.1. Grind the celadonite with a mortar and pestle.

1.2.2. Weigh 0.2 mg of celadonite powder and dilute in 10 mL of deionized water.

1.2.3. Use an ultrasound tip directly in the 10 mL of celadonite-containing water to break the aggregates. Typically, use an ultrasonic frequency of 30 kHz for a power of 50 W over 30 s.

1.3. Preparation of the deposition environment

1.3.1. Connect a capillary holder to a pressure controller.

1.3.2. Maintain the capillary holder under an optical microscope with a multidirectional micro-manipulator.

1.3.3. Place the support with the carbon fiber facing the capillary holder under the optical microscope.

1.4. Celadonite deposition

1.4.1. Pull a micropipette with an internal-end diameter of 2-10 µm to allow the dispersed celadonite to flow without obstruction.

1.4.1.1. Fix a glass capillary in the puller jaw. Ensure the right puller parameters according to patch pipette size (**Table 1**). Fill the micropipette with the celadonite-containing water.

1.4.2. Mount the micropipette on the capillary holder under the microscope. Align the micropipette and the carbon fiber under the optical microscope.

1.4.3. Approach the micropipette, to a distance of 2-10 µm from the apex of the carbon fiber.

1.4.4. Apply progressive pressure on the wide entry to the micropipette. Typically, apply 100 mbar so that a drop forms at the tip but does not fall. This drop wets the apex of the carbon fiber

1.4.5. Retract the micropipette.

2. Kicking-off the source

NOTE: In our microscope, the source-support is fixed on a manual rotating flange also carrying the piezo-electric actuator that moves (100 nm resolution, 25 mm range), with an electrical command, the object relative to the source (See **Figure 2**). This object plays the role of an electrical anode for electron emission; it is generally electrically grounded and placed in front of the source. In our experiment, voltages are hand controlled with different power supplies.

2.1. Install the source holder under vacuum.

2.2. Connect the carbon fiber and the object to two high-voltage electrical feedthroughs.

2.3. Check electrical continuity of contacts everywhere: anode-object, lens and screen; turn on the vacuum pumping.

2.4. Connect a nano-ammeter of a caliber in the µA range between the object and the electrical ground.

2.5. Increase the negative bias voltage applied to the source slowly, at approximately 1 V/s. If the anode is 1 mm away from the source, the kick-off takes place at about 2 kV. Intensity suddenly increases.

2.6. Decrease the voltage to stabilize the intensity at some hundred nA. At the beginning, intensity can fluctuate over several orders of magnitude.

2.7. Leave the system fluctuating for several hours, until fluctuations decrease. Cut off the voltage when fluctuations are lower than 10%.

3. Source characterization

NOTE: We present a way to probe the source characteristics. To estimate the source brightness, two projection microscopes are used. In these setups, the shadow of an object is observed on a fluorescent screen placed farther away (**Figure 2**). The source (cathode) and the object (anode) are mounted on a micro-manipulation flange and can rotate together in the projection plane. A simple short projection setup with a fluorescent screen allows for low magnification projection.

The second setup involves an electrostatic lens and a dual microchannel-plate/fluorescent screen assembly for the strongest magnifications[12]. Information available on each projection image is used to under-estimate the brightness: the smallest detail in the record[13]. This smallest visible detail depends on the apparent source-size, that includes the source-size geometrical blur, the vibrations between the object and the source, and the detector resolution.

3.1. Measurement of the cone angle

3.1.1. Turn the source towards the simple projection setup, with the rotating flange, to observe the electron beam.

3.1.2. Decrease the source-to-screen distance, with the manual micro-manipulator, to obtain the entire spot on the screen; then, measure the source-to-screen distance, *D*.

3.1.3. Take pictures of the screen by changing the angle between the electron beam and the normal to the screen, with the rotating flange.

3.1.4. Plot the gray-level intensity profile along one axis and determine the emission radius, *R* at a given source-to-screen distance, *D* (**Figure 3**).

3.1.5. Calculate the cone angle: $\Omega = \frac{\pi R^2}{D^2}$ with *R*, the emission radius at a given source-to-screen distance, *D*.

3.2. Measurement of the Fowler-Nordheim plot

3.2.1. Measure the emission intensity versus the voltage applied to the source: *I(V)* with *I* the intensity measured at the anode and *V* the voltage applied at the carbon fiber.

3.2.2. Plot $\ln\left(\frac{I}{V^2}\right) = f(\frac{1}{V})$. The curve shows a decreasing straight line with saturation for highest voltage. An example is given in **Figure 4**. The longest straight line is the signature of the field emission process.

3.3. Measurement of the source size

3.3.1. Turn the source towards the electrostatic lens, with the rotating flange.

3.3.2. Produce a projection image containing a huge Fresnel diffraction pattern along an edge of an object: magnification of about 20,000x is required. In our microscope, this is possible with a source-to-object distance of some 100 µm, fixed with the piezo-actuators, and an Einzel electrostatic lens.

3.3.3. Measure the sharpest visible detail on the image on the screen (**Figure 5**).

NOTE: The sharpest fringe-to-fringe distance, δ, is used.

3.3.4. Calculate the source size: $s = \frac{\delta}{2G}$.

REPRESENTATIVE RESULTS:

Several scanning electron micrographies of carbon fibers prepared as detailed in protocol were obtained in an SEM at 15 kV. Sources exhibit one, sometimes two, crystals at their apex (**Figure 1**). However, the use of the SEM involves another support for the carbon fiber, which is hard to mount and demount without breaking. It is safer to attempt direct electron emission. Tested in a projection microscope (**Figure 2**), every source prepared this way emitted. The kick-off is required only once. With old sources, sometimes, a kick-off can be used for another source.

Most of these sources show one single point source (**Figure 3**): the emission profile indicates only one continuing image without any other spot. The beam has a cone angle of about 1srd.

The Fowler-Nordheim plot exhibits 10 orders of magnitude straight and saturation at higher voltage (**Figure 4**). The saturation regime obtained for a given voltage depends on the structure, but the slope decreases systematically for higher current intensities from about 10 µA.

Energy distribution is not measured here, because the energy resolution is not good enough to obtain better accuracy than a few eV by simply biasing the entrance of the detector. Another point is that highly structured fringe patterns can be observed in some holograms rejecting a large energy distribution that would blur such patterns. Since the process involved is the Fowler-Nordheim regime, energy distribution close to 250 meV is expected[14].

The source size is estimated by measuring the smallest detail on the image produced. This image is the Fresnel diffraction pattern of the object. Here, loss of interference fringes is attributed to the size of the source (**Figure 5**); this is a way to over-estimate this measurement. In this case, the source radius is smaller than 4nm ($s = \frac{\delta}{2G} = \frac{245\mu m}{2\times 18000} = 6.8nm$). Finally, the brightness of the source is obtained, $B = \frac{I(E\pm\Delta E)}{\Omega.S} = \frac{10\mu A}{1sr\times\pi\times 12\times 10^{-14}cm^2} = 3.10^7 A.cm^{-2}.sr^{-1}$. The method presented here under-estimates the brightness because the source size is necessarily smaller.

Figure 1: Carbon fiber with celadonite deposited on it (green arrow), observed with a scanning electron microscope. Inset: Typical close-up of a celadonite crystal.

Figure 2: Experimental setup. The projection electron microscope using a celadonite on carbon source and an electrostatic lens; and the simple projection setup.

Figure 3: Measurement of the cone angle. (**a**) Schematic setup with the projection-distance D = 5 cm and, α, the angle between the carbon fiber and the normal of the screen; α is manually

changed to observe the emission pattern (**c**) and to measure the emission profile, along the blue dashed line, obtained on the screen for α = 0° (**b**). Note that the projection of the grid appears in the profile as a null intensity but clearly, the profile intensity is Gaussian with an extension of about 5 cm.

Figure 4: Fowler-Nordheim plot of a celadonite source

Figure 5: Measurement of the sharpest detail in the image to over-estimate the source size. The profile (**a**) is plotted along the white line in the image (**b**). (**c**) is a detail of (**b**).

Table 1: Pulling parameters to obtain an internal-end diameter of 2-10 µm

DISCUSSION:

This protocol is not critical because the geometry of the source at a microscopic scale changes from one source to another one. The difficulty is that since a carbon fiber is brittle, its cutting can lead to an inappropriate length. An adequate length is about 500 µm; the microscopic shape of the cut is not crucial. The critical step is to have a very small number of crystals (ideally one) deposited on the apex of a conductive wire. Adapting the crystal concentration with the deposited volume is the most important point. If too many crystals aggregate, emission is damped. Here, we describe a way to manage this. Due to the kick-off procedure, if a small number of crystals are deposited, only one of them is finally responsible for the emission. Another requirement is to build a protruding structure in order to approach the anode and to obtain a directive emission. This cannot be achieved if celadonite-crystals were deposited on a carbon film as in previous studies.

The electron celadonite source is now regularly used in a low-energy electron point-source projection microscope, associated with an Einzel lens system. Because of the high brightness of the source, at this large working distance of 600 µm, a resolution of about 30 nm is generally obtained[12]. In point-projection microscopes, working at such a large source-object distance is comfortable and is advantageous. Moreover, such large working distances avoid any field effects on the object. The high emission intensities provided by this source enable image acquisition at a video rate of about 500 images/s, and the robustness of the source is a practical advantage over classic field emission metal tips. Except in our microscope, this recently developed source has not yet been used in another microscope. The emission instabilities previously measured could be problematic for a scanning microscope. Although these instabilities are observable during point-projection imaging, the emission location is stable, making image averaging possible. Compared to classic metal-tip sources for identical magnification, holograms obtained with the present source are identical but obtained for a much larger working distance. Ultimate spatial resolution is presently an open experimental problem.

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DISCLOSURES

The authors have no competing financial interests.

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Figure 1

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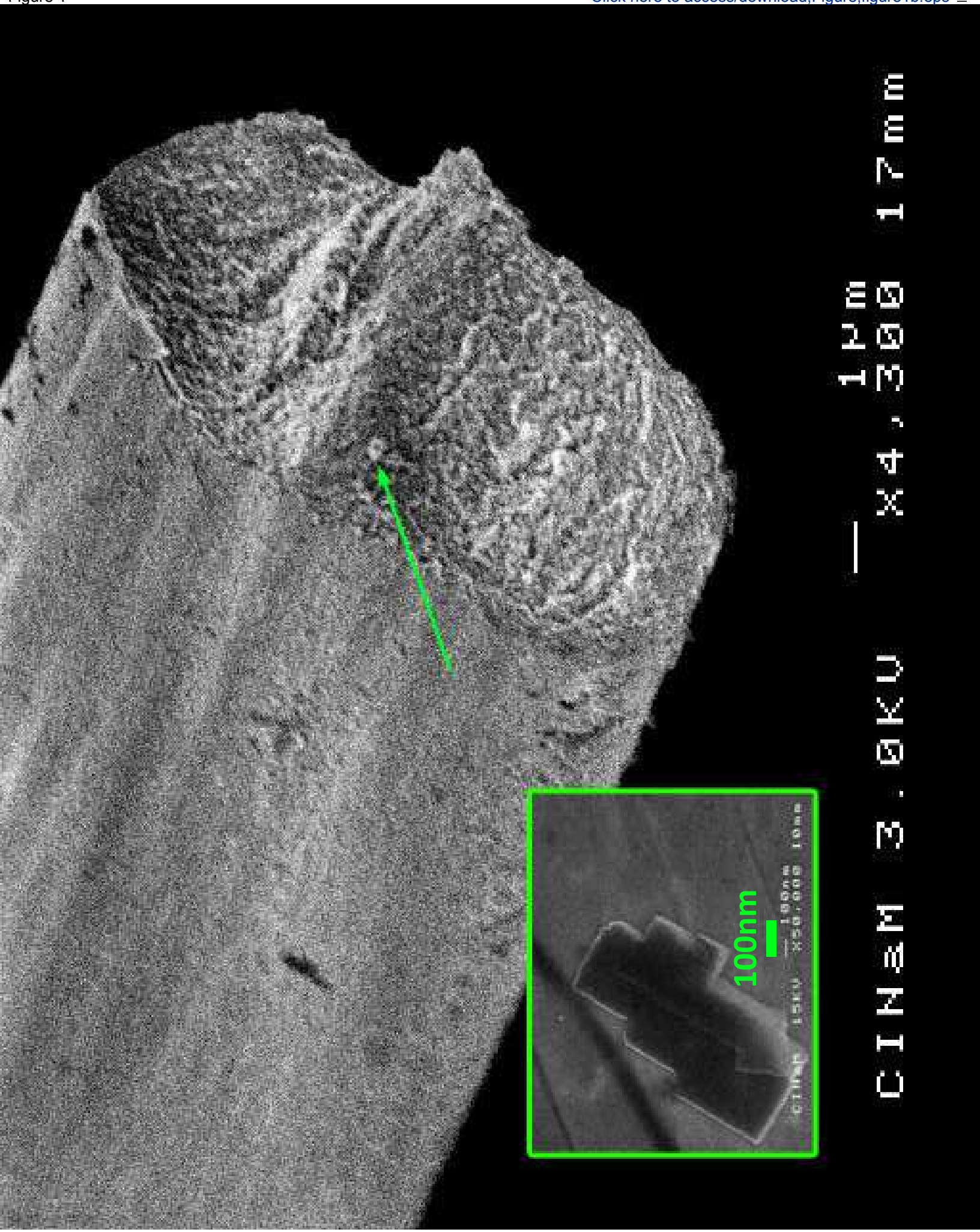

Figure 2

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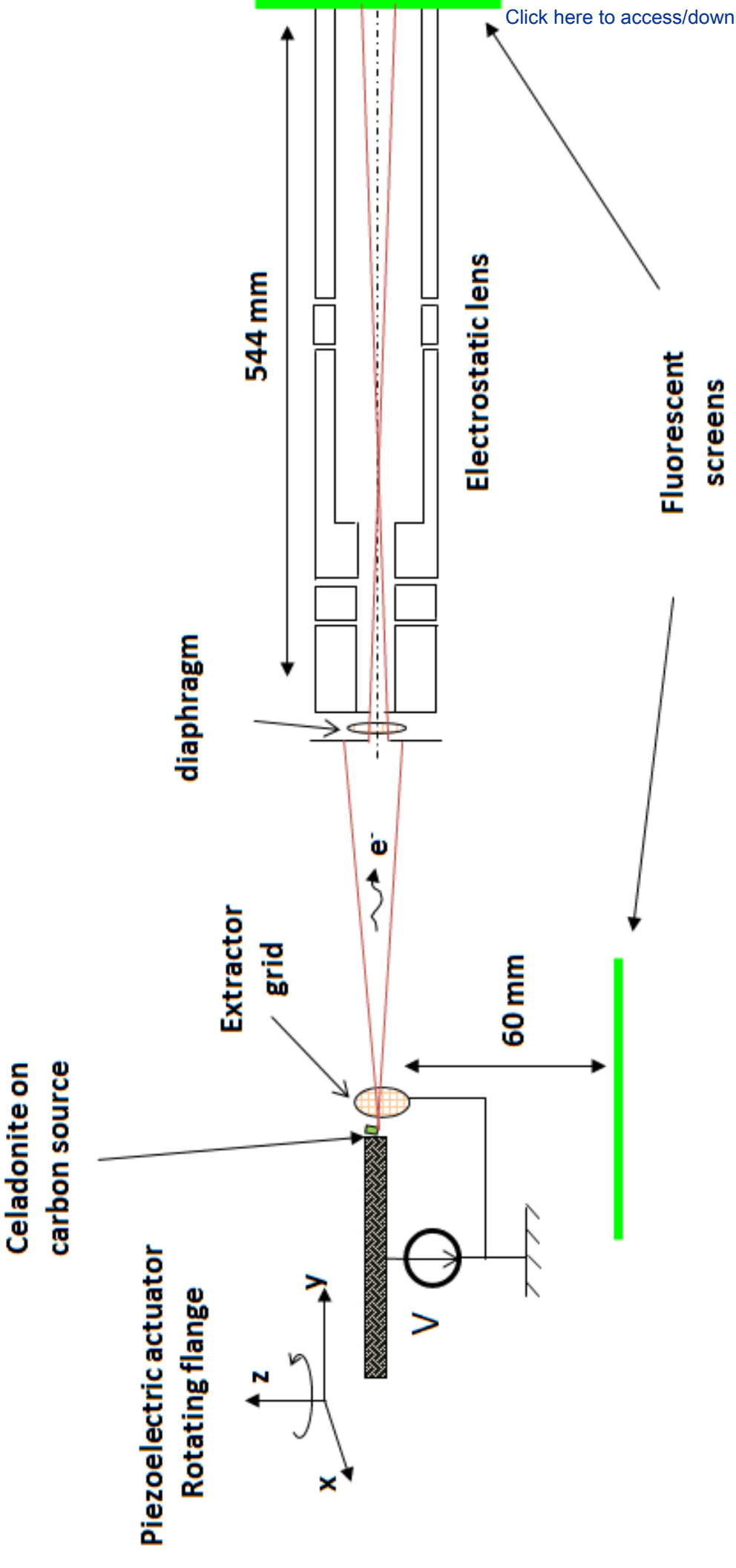

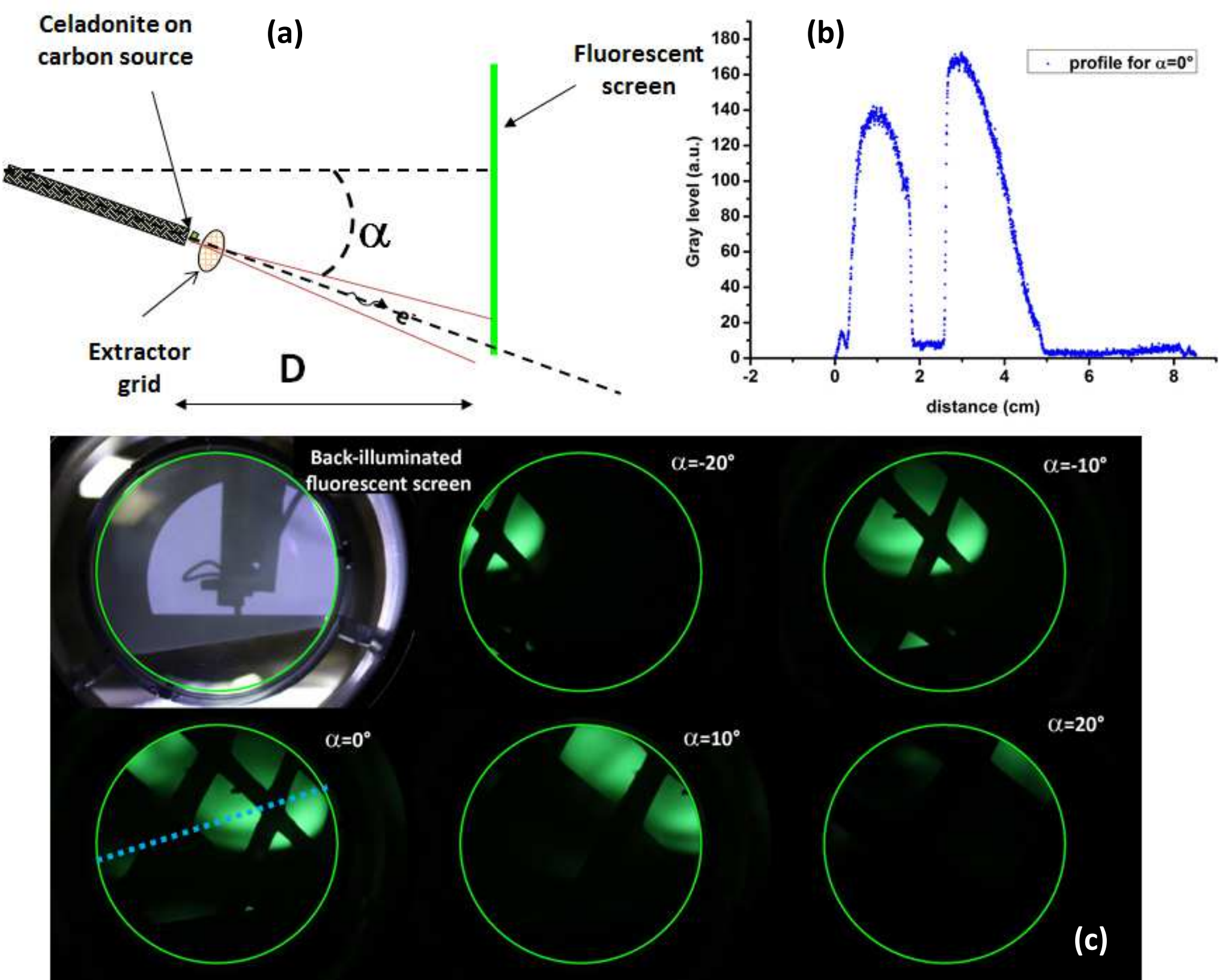

Figure 3

Figure 4

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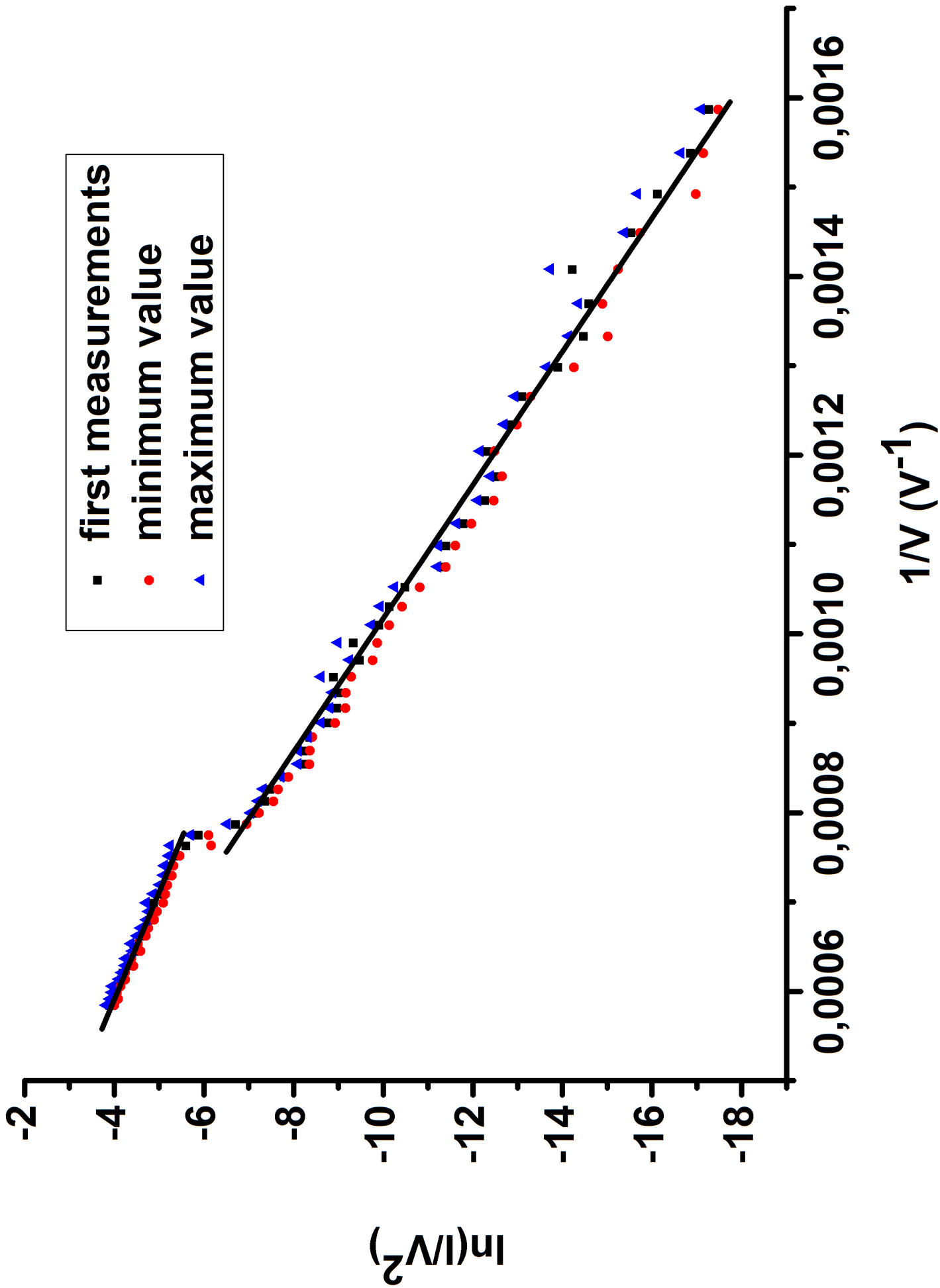

Figure 5

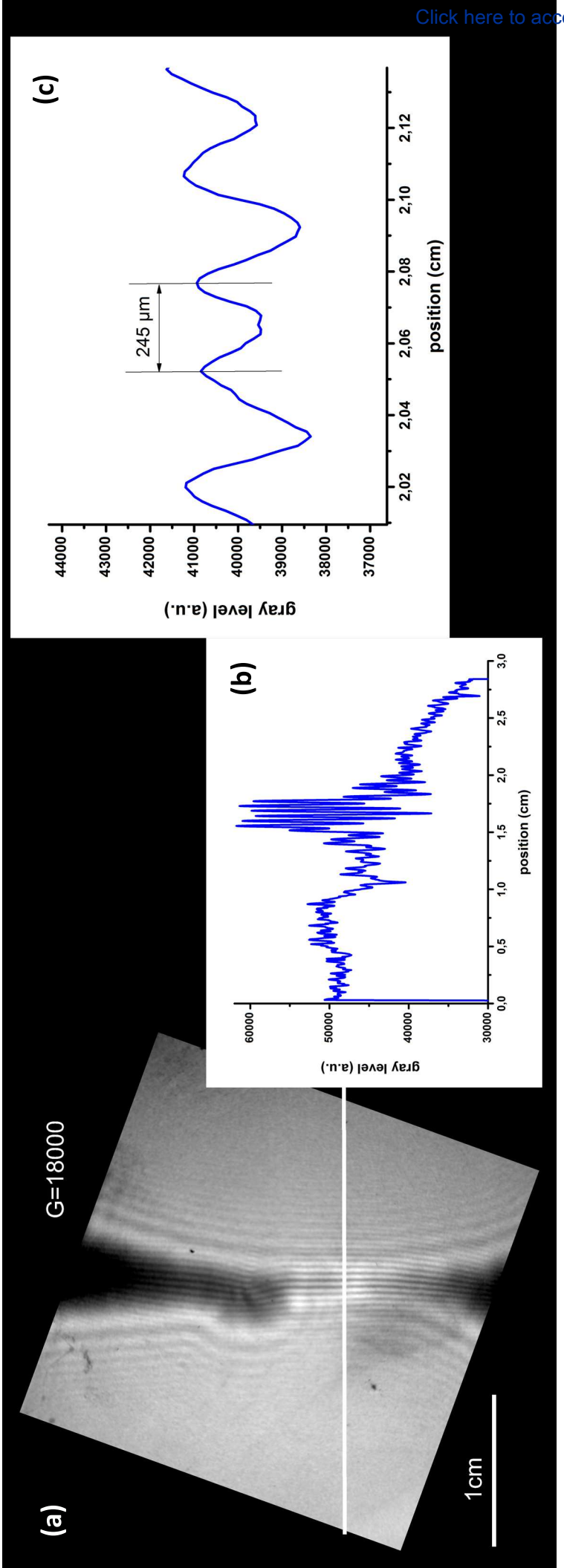

Table 1

heat	filament	velocity	delay	pull
450	3	5	200	120
350	4	40	200	0

Table of Materials

Click here to access/download;Table of Materials;table_of_material.xlsx

Item	Company	Catalog Number
Carbon fiber filament	Goodfellow	C 005711
Carbon fiber filament	Mitsubishi Chemical	DIALEAD
Carbon fiber filament	Solvay	THORNEL P25
Carbon fiber filament	Zoltek	PX35 Continuous Tow
Celadonite		
Dual-stage microchannel plate and fluorescent screen assembly	Hamamatsu	F2225-21S
Flow controller	Elveflow	OB1
Machinable glass ceramic	Macor	
Micropipette Puller	Sutter Instruments	P2000
Piezo-electric actuators	Mechonics	MS30
Quartz capillary	Sutter Instrument	B100-75-15
Silver Lacquer	DODUCO GmbH	AUROMAL 38
Ultrasonic processor	Hielscher / sonotrode MS3	UP50H

Notes

Verona Green earth / pigment

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CORRESPONDING AUTHOR

Name:	Salançon Evelyne
Department:	CINaM - UMR 7325
Institution:	AMU - CNRS
Title:	Preparing a celadonite-electro source and measuring its brightness
Signature:	Salançon
Date:	10/05/19

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Dear Editor,

We thank you for the work you made on our paper. We hope we made the changes required. Here are the different answers with respect to your remarks. In the following, you will find answers to the reviewers.

Reviewers 3 and 4 have read the first version of the paper and their remarks are not adapted to this new paper. I do not know if they are the same reviewers for the second version. I don't think so. Then, I have prepared some answers if you want to interest them to the new version.

With best regards,
Evelyne Salançon.

Editorial comments:

General:

1. Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammar issues.

Our paper is read and corrected by an English editor. We apologize if there is still some language issue, it should not.

2. Please ensure that the manuscript is formatted according to JoVE guidelines–letter (8.5" x 11") page size, 1-inch margins, 12 pt Calibri font throughout, all text aligned to the left margin, single spacing within paragraphs, and spaces between all paragraphs and protocol steps/substeps.

Here, also, we have tried to respect your format. If there are some mistakes, we apologize in advance.

3. Please include email addresses for all authors in the manuscript.
4. Please include at least 6 key words or phrases.
5. Please do not include citations in the abstract.

OK, all were made.

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For example: Macor, Sutter Instrument

I have indicated the commercial language and references in "table of materials".

Protocol:

1. There is a 10 page limit for the Protocol, but there is a 2.75 page limit for filmable content. If revisions cause the highlighted portion to be more than 2.75 pages, please highlight 2.75 pages or less of the Protocol (including headers and spacing) that identifies the essential steps of the protocol for the video, i.e., the steps that should be visualized to tell the most cohesive story of the Protocol.

2. Please add more details to your protocol steps. Please ensure you answer the "how" question, i.e., how is the step performed? Alternatively, add references to published material specifying how to perform the protocol action. If revisions cause a step to have more than 2-3 actions and 4 sentences per step, please split into separate steps or substeps.

Specific Protocol steps:
1. 1.1.4: What do you cut with?

"Cut the fiber with a cutting pliers (under a binocular microscope) so that between 100µm and 3mm are left outside the stainless steel tube."

2. 1.2.1: What is used to grind?

"Grind the celadonite with a mortar and pestle"

3. 1.2.4: Please include ultrasound parameters.

"Use an ultrasonic probe directly in the 10ml celadonite-containing water to break the aggregates. We use a typical ultrasonic frequency of 30kHz for a power of 50W during 30s"

4. 2,3: Please include more information on operation of these systems-e.g., do you use a control panel? Software? How are voltages, distances, and angles adjusted?

"In our microscope, the source-support is fixed on a manual rotating flange also carrying the piezo-electric actuator that moves (100nm resolution, 25mm range), with an electrical command, the object relative to the source (See Figure 2). This object plays the role of an electrical anode for electron emission; it is generally electrically grounded and placed in front of the source. In our experiment, voltages are hand controlled with different power supplies."

Figures, Tables, and Figure Legends:
1. Please remove the embedded figures from the manuscript.
2. Please remove the embedded table from the manuscript. All tables should be uploaded separately to your Editorial Manager account in the form of an .xls or .xlsx file. Each table must be accompanied by a title and a description after the Representative Results of the manuscript text.
Sorry, we were convinced that figures were included at the end of the document, we apologize for this mistake. We made a specific file with "tables, figures and legends".

Discussion:
1. Discussion: As we are a methods journal, please revise the Discussion to explicitly cover the following in detail in 3–6 paragraphs with citations:
a) Critical steps within the protocol
b) Any modifications and troubleshooting of the technique
c) Any limitations of the technique

These 3 points are treated in this new paragraph:

“This protocol is not critical because the geometry of a source at a microscopic scale changes from one source to another one. Difficulty is that since a carbon fiber is brittle, its cutting can lead to inappropriate length. Adequate length is about 500µm, the microscopic shape of the cut is not crucial. The critical step is to have a very small number of crystals (ideally one) deposited on the apex of a conductive wire. Adapting the crystal concentration with the deposited volume is the most important point. If too many crystals aggregate, emission is damped. Here we describe a way to manage this. Due to the kick-off procedure, if a small number of crystals is deposited only one of them is finally responsible for the emission. Another requirement is to build a protruding structure in order to approach the anode and to obtain a directive emission. This cannot be achieved if celadonite-crystals were deposited on a carbon film as in previous studies.”

d) The significance with respect to existing methods
e) Any future applications of the technique

These 2 points are treated in this part:

“Except in our microscope this recently developed source has not yet be used in another microscope. The emission instabilities previously measured could be problematic for a scanning microscope. Although these instabilities are observable during point-projection imaging, the emission location is stable, making image averaging possible. Compared to classic metal-tip sources for identical magnification, holograms obtained with present source are identical but obtained for a much larger working distance. Ultimate spatial resolution is presently an open experimental problem.”

Acknowledgment and Disclosures:
1. Please include a Disclosures section, providing information regarding the authors’ competing financial interests or other conflicts of interest. If authors have no competing financial interests, then a statement indicating no competing financial interests must be included.

OK

References:
1. Please include the reference section in the main text (i.e., not as an endnote).
2. Reference 15- has this been accepted somewhere? If so, please include more information (see below); if not, please do not include this reference.
3. Please ensure that the references appear as the following: [Lastname, F.I., LastName, F.I., LastName, F.I. Article Title. Source. Volume (Issue), FirstPage – LastPage (YEAR).] For more than 6 authors, list only the first author then et al. Please do not abbreviate journal titles.

OK

Table of Materials:
1. Please ensure the Table of Materials has information on all materials and equipment used,

especially those mentioned in the Protocol (e.g., source support, carbon fiber, celadonite).

A lot of things are fabricated in the laboratory or accessible from former experiment. For example, carbon fibers are available from many sources (7µm diameter carbon fibers are for instance available from Goodfellows) our carbon fibers were likely coming from Carbone Lorraine but are not anymore available.

Celadonite is a rock coming from talcum quarry in Brasil given by a geophysicist of our laboratory (celadonite is also called Verona green earth and is known as a pigment). We found an advantage using it because of reproductive shape of the microcrystals. However, our previous studies on emission from various mineral [8] show that many candidates are possible.

The source support is fabricated from a Macor® plate. I hope that the new table of material is comprehensive enough. The source support is fabricated from a Macor® plate. I hope that the new table of material is comprehensive enough.

We would like to thank the reviewers for the work they made on our manuscript. The different answers are given in the following.

Reviewers' comments:
Reviewer #1:
Manuscript Summary:
All good.

Major Concerns:
None.

Minor Concerns:
1. Fig. 3. The right panel: is the x-axis "Distance D (cm)"? or what distance is it? Why the plot exhibits two maxima and minima at distance 2-2.5 cm, is such distribution typical?

You are right, the legend was not clear enough, the new legend of this figure should be clearer: "Measurement of the cone angle. (a) Schematic setup with the projection-distance D=5cm and, α, the angle between the carbon fiber and the normal of the screen; α is manually changed to observe the emission pattern (c) and to measure the emission profile, along the blue dashed line, obtained on the screen for α=0° (b). Note that the projection of the grid appears in the profile as a null intensity but clearly, the profile intensity is Gaussian with an extension of about 5 cm."

The plotted distance is along the blue dashed line, on the screen, and the two minima come from the shadow of the grid.

2. Section 3.3.4 What is "G"? It has not been defined.

Thank you! In section 3.3.2, we have added: "magnification of about G=x20,000 is required".

3. "The source size is estimated by measuring the smallest detail on the image produced" - this statements is not obvious, a reference should be added.

This question is interesting and the key point of the brightness determination. Estimating an apparent source size is not as critical as determining the real source size. We should use the real source size to determine the brightness and I discuss this point just after. Here, we know that the source size is necessarily smaller than the one measured this way: We use the fact that the smallest detail available in a record corresponds to the smallest visible distance in the object plane (see ref. RSI 89, 2018). In practice, this smallest visible distance depends on the apparent source-size, including the geometrical blur due to the source-size, vibrations between the object and the source, and detector resolution limits.

We use this method regularly to under-estimate the brightness of our sources. This is widely discussed in the answer to the second reviewer.

Introduction of "Source characterization": "In fact, information available on each projection image is used to under-estimate the brightness: the smallest detail in the record [13]. This

smallest visible detail depends on the apparent source-size, that includes the source-size geometrical blur, the vibrations between the object and the source, and detector resolution limits."

In reality, using an interferometry experiment [15, 16], we have measured a smaller source size, about 1nm. Here, vibrations and detector resolution prevent from confirming this result.

[15] – Morin, R., Degiovanni, A. Interferometry with low-energy electrons, *Journal of Vacuum Science & Technology B* 13(2), 407 (1995)

[16] – Degiovanni, A., Laï, W., Morin, R. Electron sources of atomic size, Symposium on Advanced Surface Analytical Technique, JSPS, Kyoto, Japan (1996)

Representative results:

"Finally, the brightness of the source is obtained, $B = \frac{I(E \pm \Delta E)}{\Omega . S} = \frac{10 \mu A}{1sr \times \pi \times 12 \times 10^{-14} cm^2} = 3.10^7 A.cm^{-2}.sr^{-1}$. The method presented here under-estimates the brightness because the source size is necessarily smaller."

Reviewer #2:

Manuscript Summary:

The authors present a method to produce and characterize isolated electron point sources made out of small celadonite crystals.

Major Concerns:

The preparation is well described. Regarding the characterization, the authors made an approximation regarding the source size that appears to be too simple. They use the fringe distance as a measure for the resolution and consequently also for the determination of the source size. However, the fringes are an interference effect from different spherical waves and not a real space image of the sample. Consequently, this can not be used to determine the resolution in the sample plane. However, the fringes can be used to determine the effective source size, see for example:

Spence, J. C. H., Qian, W. & Silverman, M. P. Electron source brightness and degeneracy from Fresnel fringes in field emission point projection microscopy. Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films 12, 542-547 (1994).

Please do a more careful analysis of the source size and only claim a certain spatial resolution after numerical reconstruction of the hologram.

This point is crucial and one of the most important to announce a brightness for the source. We understand your point of view and clearly, we under-estimate the brightness with this method. We have changed the word « measure » with « estimate » in order to be right and we have also explained this in introduction of the paragraph concerning the characterization of the source.

Source characterization:

« In fact, information available on each projection image is used to under-estimate the brightness: the smallest detail in the record [13]. This smallest visible detail depends on the apparent source-

size, that includes the source-size geometrical blur, the vibrations between the object and the source, and detector resolution limits. »

This paper is intended to help all those wishing to make up a celadonite source to produce a coherent electron beam. We expose a method to over-estimate the size of the source. We could obtain a better estimation of the source-size with the method that you cite or with the method that we used, with an interferometry method published elsewhere ([15],[16]). Using this method, R. Morin and A. Degiovanni measured a source size of 0.16nm for tungsten electron emission tips. With the celadonite source, we have measured 1nm. We have decided to not mention this, here. This is not the most important point of this article.

[15] – Morin, R., Degiovanni, A. Interferometry with low-energy electrons, *Journal of Vacuum Science & Technology B* 13(2), 407 (1995)

[16] – Degiovanni, A., Laï, W., Morin, R. Electron sources of atomic size, Symposium on Advanced Surface Analytical Technique, JSPS, Kyoto, Japan (1996)

Representative results:

"Finally, the brightness of the source is obtained, $B = \frac{I(E \pm \Delta E)}{\Omega . S} = \frac{10 \mu A}{1sr \times \pi \times 12 \times 10^{-14} cm^2} = 3.10^7 A.cm^{-2}.sr^{-1}$. The method presented here under-estimates the brightness because the source size is necessarily smaller."

Reviewer #3:

Manuscript Summary:

The authors describe a method to fabricate electron sources for point-projection microscopy that need lower extraction voltages, provide larger emission angles and operate at lower vacuum requirements.

We think you had the previous version of this paper that is now published elsewhere (ultramicroscopy, 2019). Most of your concerns were corrected except some of them. I try to answer them, here behind.

Major Concerns:

- I could not understand one of the most crucial steps of the emitter formation: How do you get the crystal to stick to the nanotube?

Deposited this way, the celadonite sticks on the carbon without any glue, likely resulting from surface tension and from small weight of a microcrystal.

- It would be useful to discuss the applicability of those sources for other types of electron microscopy.

  It is now discussed.
  "Except in our microscope this recently developed source has not yet be used in another microscope. The emission instabilities previously measured could be problematic for a scanning microscope. Although these instabilities are observable during point-projection imaging, the emission location is stable, making image averaging possible. Compared to

classic metal-tip sources for identical magnification, holograms obtained with present source are identical but obtained for a much larger working distance. Ultimate spatial resolution is presently an open experimental problem."

- What is the energy spread of the extracted electron beam?

It is now discussed.

« Energy distribution is not measured here, because the energy resolution is not good enough to obtain better accuracy than a few eV by simple biasing the entrance of the detector. Another point is that highly structured fringe patterns can be observed in some holograms rejecting a large energy distribution that would blur such patterns. Since the process involved is the Fowler-Nordheim regime, energy distribution close to 250meV is expected. »

- Can you quantify how robust the gun tip is to higher pressures? E.g., how long can you run it at HV? How does Fig. 5 look like for different pressures?

A part of the answer is now discussed there:

« The source can operate under relatively high pressure: it was tested several times at 10^{-2}mbar for half an hour, never more time. Yet its lifetime and its stability remain dependent on the right vacuum conditions. We usually employ the celadonite source at 10^{-8}mbar and obtain a lifetime of months. »

Clearly, the discussion around the blur due to the pressure is not the point here. We will look at this, in a later work.

Minor Concerns:

- Could you add where D = 66 cm and 12 cm are measured in Fig. 2?

It is now correct.

- Which silver lacquer is used for the glueing and how is it cured?

References are given in « Table of materials ». The lacquer is not cured but well dried.

- The larger emission angles also increase spherical aberrations. Could you comment on the spherical aberration coefficient of your illumination and imaging system and deduce an optimal emission angle?

We have not yet measured this interesting point.

- Figure 5 deserves a more detailed discussion. In particular the fits are not described at all. What are the extracted parameters if you fit a full Fowler-Nordheim equation to the data?

This concerns the previous article; however, there is no real possibility to compare the shape of the emitter to metal tips parameters usually intended. Here, the field responsible for the emission is due to a reinforcement of the field [10].

- You chose for a design with two beam cross-overs (Fig. 7). What is the influence of electron-electron interaction in these points? Could your resolution benefit from a design without cross-overs?

This concerns the previous submission; however, this scheme was an optical construction of two lenses. We use only one beam in the system.

- - What is the 'periodic' dot pattern in Fig 10? Is this structure on the sample or an imaging/reconstruction artifact?

This concerns the previous submission; however, this periodic dot pattern corresponds to the channels distribution on the detector itself. This is widely discussed in the paper [13].

Reviewer #4:

Manuscript Summary:

In this work, the authors describe their construction of a low-energy electron point-projection microscope that uses an insulator crystal as the electron source. In addition to the source, there are two unique and interesting points. First, the microscope includes an electron optical lens to achieve a higher magnification. Second, a counting imaging method is utilized to improve its spatial resolution. I would recommend publication of this work on JOVE with minor revision.

We think you had the previous version of this paper that is now published elsewhere (ultramicroscopy, 2019). Most of your concerns were corrected except some of them. I try to answer them, here behind.

Major Concerns:

None.

Minor Concerns:

1. I have a concern on "the usual near-field source-object distance" in the last sentence of the abstract. The word "near-field" typically refers to a distance smaller than several times of the wavelength. A typical low-energy electron point-projection microscope operates at a distance of 100 nm or larger, which should be considered as "far field". I have never seen any point-projection microscope that operates at near field. I suggest the authors can rewrite this part of the sentence.

Yes, we first completely missed this understanding. It was quickly corrected!

2. For the counting imaging, I hope one to several images containing micrometric stains can be provided. I would like to see whether the center of mass in each stain can be determined easily.

This is widely discussed in the paper [13].

3. For the counting imaging, the authors state that the maximum dose is about 10,000 hits per frame. How is this number determined? For the experimental result shown in Fig. 10b, the authors state 80 hits per frame. Is there any reason why the dose cannot be increased to several thousand hits per frame, which can reduce the entire imaging time by two orders of magnitude.

This is not the maximum dose but a classical « in run » recording. We could increase the dose and this is now widely discussed in [12].