# In situ transmission electron microscopy studies enabled by microelectromechanical system technology

M. Zhang,[a] E.A. Olson, R.D. Twesten, J.G. Wen, L.H. Allen, I.M. Robertson,[b] and I. Petrov[c]
*Frederick Seitz Materials Research Laboratory and Department of Materials Science, University of Illinois, Urbana, Illinois 61801*

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We have designed and fabricated a standardized specimen holder that allows the operation of a microelectromechanical system (MEMS) device inside a transmission electron microscope (TEM). The details of the design and fabrication processes of the holder are presented. The sample loading mechanism is simple and allows reliable electrical contact to eight signal lines on the device. Using a MEMS-based, nanojoule calorimeter, we performed rapid-heating experiments on Bi nanoparticles to demonstrate the functionality of the holder. We show that the heat capacity can be measured simultaneously with TEM observations. The size-dependent melting of Bi nanoparticles was observed simultaneously by nanocalorimetry and selected area diffraction measurements. We believe this approach will open up new experimental pathways to researchers, combining the speed and resolution of transmission electron microscopy with the flexibility, precision, and compactness of MEMS-based sensors and actuators.

## I. INTRODUCTION

There has been a growing tendency to use the transmission electron microscope (TEM) as a dynamic research tool to study atomic-scale mechanisms of complex materials processes. The main obstacle toward wider use of the technique is the limited space available in the objective lens pole-piece for incorporating an experimental laboratory. A new approach to in situ microscopy is to fabricate the experimental laboratory by employing micro- and nano-lithographic techniques and to use microelectromechanical systems to add functionality. This strategy for miniaturizing the laboratories makes judicious use of the available space and offers enhanced capabilities to stimulate and probe the system response compared with conventional approaches. Recent advances in microelectromechanical system (MEMS) fabrication allow for high-precision electrical, thermal, and mechanical manipulation of individual nanostructures, while simultaneously measuring their properties and carrying out real-time, atomic-resolution imaging, electron diffraction, and spectroscopy. There are currently only a few functional devices, but they illustrate the potential of this approach.[1,2]

These experimental laboratories require a power source and must have the ability to return a control or measurement signal. This capability necessitates establishing a simple, reliable, and robust method for making the electrical connections between the experimental laboratory and the external controllers. This is different from conventional holders with electrical connections, as the connections are generally permanent and are not remade by the user.

In this paper, we describe a methodology for creating up to eight electrical signals with an experimental laboratory on a standard holder base. To demonstrate the utility of the approach, we present data on the operation of a MEMS nanocalorimeter allowing in situ, rapid-heating experiments to investigate the dependence of the melting temperature on the size of bismuth nanoparticles while simultaneously observing the accompanying structural changes.

## II. FABRICATION AND ASSEMBLY OF THE MEMS TEM HOLDER

Using the proposed method for creating miniaturized versions of functional experimental laboratories requires establishing electrical connections to deliver power and

[a]Present address: Micron Technology, Inc., Boise, ID 83707.
[b]This author was an editor of this focus issue during the review and decision stage. For the *JMR* policy on review and publication of manuscripts authored by editors, please refer to http://www.mrs.org/publications/jmr/policy.html.
[c]Address all correspondence to this author
e-mail: petrov@mrl.uiuc.edu
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to probe the response of the system. As each device is intended for single use, a necessary requirement is that they can be changed by the user. A schematic illustration showing the front end of an eight-contact holder to which a MEMS device can be attached is presented in Fig. 1. Part (a) is a 1-mm-thick Al baseplate.[3] In the JEOL 2010, the underside of this plate actually faces the electron beam and, therefore, this surface is left conductive to prevent charging. The electrical contacts [Part (b)] are chemically etched from a CuBe alloy and then plated with Au for corrosion resistance and low contact resistance. The resistance between the individual contacts and between the contacts and the stage body is 100 MΩ. Each set of contacts is fabricated as a single piece to aid alignment and assembly. This approach was used in preference to wire bonding as the latter method restricted sample exchange and the protruding wire would occupy additional space. The contacts are sandwiched between the baseplate and a 0.5-mm-thick Al frame [part (c)] to hold the contacts in place and help align the MEMS sensor [part (d)]. The contact pads on part (d) are shown on the top face of the sensor for clarity. Part (e) is a pair of spring clips used to hold the device in position. Part (f) is also made of Al and has a number of grooves machined into its underside. These grooves help align the electrical contacts and clamp them in place along with wires (not shown). These wires run the length of the holder body to an electrical connector. Part (g) attaches the entire assembly to the holder body. The parts are held together by nonmagnetic screws (not shown). Parts (a), (c), and (f) are all anodized Al, where the aluminum oxide coating provides electrical insulation. The overall dimensions of the setup are completely compatible with a regular JEOL 2010 TEM holder.

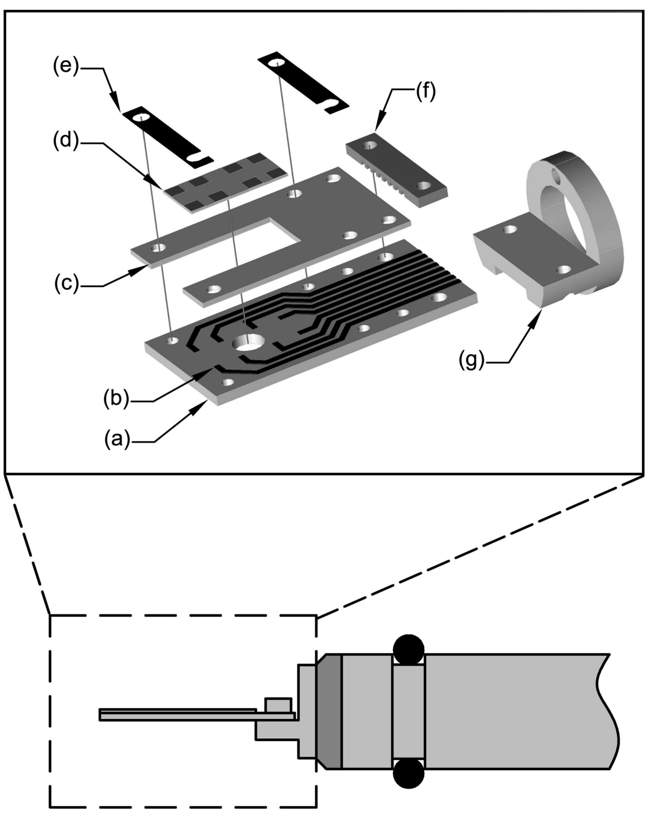


FIG. 1. Schematic illustration of the MEMS sample holder. The bottom section shows a side view of the assembled holder. The top section shows the individual components: (a) baseplate, (b) electrical contacts, (c) alignment frame, (d) MEMS device, (e) spring clips to hold the device in place, (f) clamp to align and hold the contacts, and (g) connector to the holder body.

The TEM holder body is machined from a single piece of phosphor bronze alloy. To easily make an ultrahigh-vacuum-quality seal, a standard 1.33 in., Varian-ConFlat-compatible flange is cemented onto the end with a vacuum compatible sealant (TorrSeal; Varian, Inc., Palo Alto, CA). A standard electrical feedthrough is connected to the flange using a copper gasket.

While the active area for TEM observation of a MEMS device is several square millimeters, similar to traditional samples, the platform should accept devices that are large enough to allow for the integration of sensors and actuators and that are also robust enough for manipulation. It is also necessary to have surface area available for contact pads. These pads should be approximately $1 \times 1$ mm$^2$ to allow reliable contact alignment during mounting. In the standard design of a JEOL 2010 TEM single-tilt holder, the sample sits at the end of a rod in an assembly approximately 26 mm long, 13 mm wide, and 2 mm thick. The space available for the MEMS device plus the contacts is therefore fixed by these dimensions. We have chosen a standard device size of $5 \times 10$ mm for this work. This choice allows the use of all the standard features of the microscope including tilting of the stage and insertion of the objective aperture.

With this approach to fabricating the experimental laboratory, it is necessary to remove a portion of the Si substrate to create the electron transparent region. Flat, continuous surfaces can be incorporated into a MEMS device by the use of a membrane, usually made of low-residual-stress silicon nitride ($SiN_x$).[4] Membranes can be large, a few millimeters on a side, but can be as thin as 30 nm. Supports such as these, in the traditional 3-mm form factor, are even commercially available (e.g., SPI Supplies, Inc., West Chester, PA) with $SiN_x$ thicknesses between 30 and 100 nm. Thin films or individual nanostructures, e.g. nanotube, nanowires, etc., may be spanned across gaps in the silicon substrates and biased electrically and/or mechanically while performing TEM imaging and analysis.

An existing MEMS-based nanocalorimeter[5,6] was modified to the required dimensions described here. The nanocalorimeter is fabricated with a thin (30 ± 0.4 nm) $SiN_x$ membrane. $SiN_x$ is transparent to the electron beam, and provides a uniform, low-noise support suitable for the imaging of small features. A metal (we used both Al and Au) strip (50 nm thick and 500 μm wide) is patterned on top of the $SiN_x$ to act as a precision thermometer.

These electron-transparent MEMS devices, like conventional TEM samples, are very fragile. It is critical that the device can be mounted and removed from the holder gently and reliably. To that end, so-called "cleave lines" can be incorporated into the MEMS design process to accurately control the outside dimensions of the device without the need to mechanically cut the individual devices, keeping breakage to a minimum. Better control of the size of the finished device also allows it to be aligned accurately in the TEM holder. These lines can be fabricated by standard dry or wet etching techniques and can be included in an existing MEMS fabrication process, thus requiring no additional process steps. An example of this procedure is shown in Fig. 2. If an anisotropic, wet etching method is used (e.g., 30% KOH, 80 °C), narrow lines would etch partway through the wafer and wide ones would etch all the way through.

Photographs of the eight-contact MEMS TEM holder along with a MEMS-based nanocalorimeter are shown in Figs. 3(a) and 3(b), respectively.

## III. EXPERIMENT

A MEMS-based nanocalorimeter was mounted on the TEM holder. Bi films of varying thickness were deposited on the sensors beforehand, by thermal evaporation in a vacuum of ~$5 \times 10^{-8}$ Torr.

The voltage drop $V$ across the metal strip of the MEMS calorimeter and the current $I$ through it are measured during a dc electrical pulse. Current is determined by measuring the voltage drop across a series resistor of known value. The power $P$ dissipated in the nanocalorimeter as a function of time $t$ is simply given by

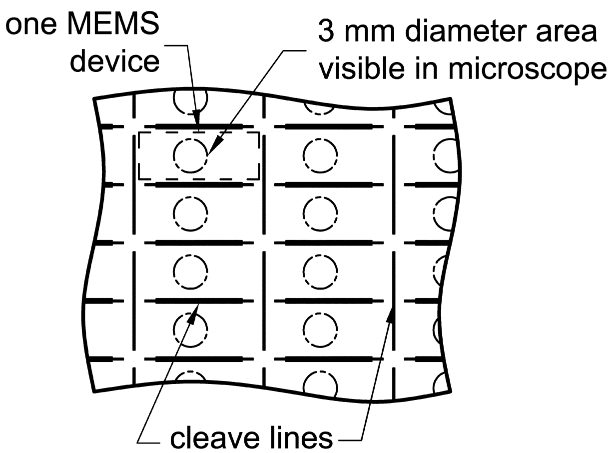


FIG. 2. Cleave lines used to reliably break a wafer into individual devices. When using anisotropic etching techniques, the narrow lines etch partway through the wafer, and the wide lines etch all the way through.

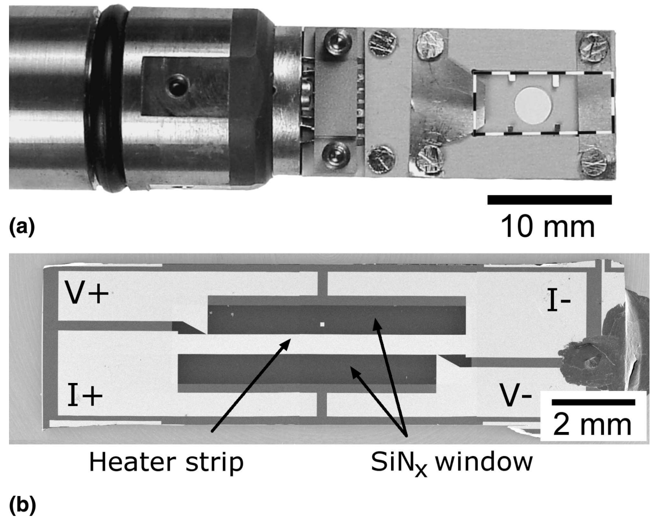


FIG. 3. Photograph of the eight-contact MEMS TEM holder and the MEMS device used in this work. (a) The sample holder; the dashed line indicates the location of the MEMS device when installed. (b) The MEMS nanocalorimeter; current is passed through the heater strip from $I^+$ to $I^-$ while the resistance of the metal strip (and hence its temperature) is determined from the voltage difference ($V^+ - V^-$).

$$P(t) = V(t)I(t) \quad . \tag{1}$$

Resistance $R$ is a function of the temperature $T$ of the sensor, and is calibrated for each sensor before the experiment. $R$ is measured by the four-point method and can be calculated by

$$R(t) = \frac{V(t)}{I(t)} \quad . \tag{2}$$

The heat capacity $C_P$ can then be determined from

$$C_P(t) = P(t)\left(\frac{dT}{dt}\right)^{-1} \quad . \tag{3}$$

$C_P(t)$ is transformed to $C_P(T)$ using $T(t)$. Details of the calculations, including corrections for heat loss, are given in Ref. 7.

The heating rates achievable with this device are very high, in the range from $3 \times 10^4$ °C/s up to $10^6$ °C/s. The heating cycle lasts for only about 10 ms. Fast heating reduces heat loss, and makes the measurement conditions close to adiabatic. By controlling $I(t)$, the heating rate can be adjusted. A constant, small $I$ can be used to heat the sensor for very long periods of time, essentially turning this calorimeter into a heating stage. The cooling rate is also quite high, ~$3 \times 10^3$ °C/s, due to the low thermal mass of the system. Thus, operating this MEMS-based calorimeter in the TEM offers a range of heating rate experiments unachievable with traditional heating stages.

We have performed in situ melting experiments using bismuth particles with diameters in the range of 5–50 nm.

A Bi film with a nominal thickness of 4 nm was thermally evaporated onto the $SiN_x$ membrane; in situ annealing causes the film to de-wet from the substrate and form spherical particles. In these experiments, a direct current (dc) electric current pulse is applied to the nanocalorimeter, rapidly increasing the temperature of the sensor wire and surrounding area. Heat capacity data for this sample is shown in Fig. 4(a). These particles had an average radius of ~10 nm. Because of the size-dependent melting that occurs in this size regime, the melting temperature in Fig. 4 was measured to be 233 °C, well below the bulk melting temperature of Bi, 271 °C.[8]

In addition to direct imaging of the Bi particles, the structure of the particles can be characterized by selected-area electron diffraction (SAED). Near room temperature, the particles are crystalline, and diffraction spots are clearly visible [Fig. 4(b)]. During the calorimetric scans, the temperature increases above the Bi melting point, and the diffraction spots disappear indicating melting of the particles [Fig. 4(c)]. Upon cooling, the diffraction spots reappear in random positions along the original Bi diffraction rings indicating crystallization from a molten state.

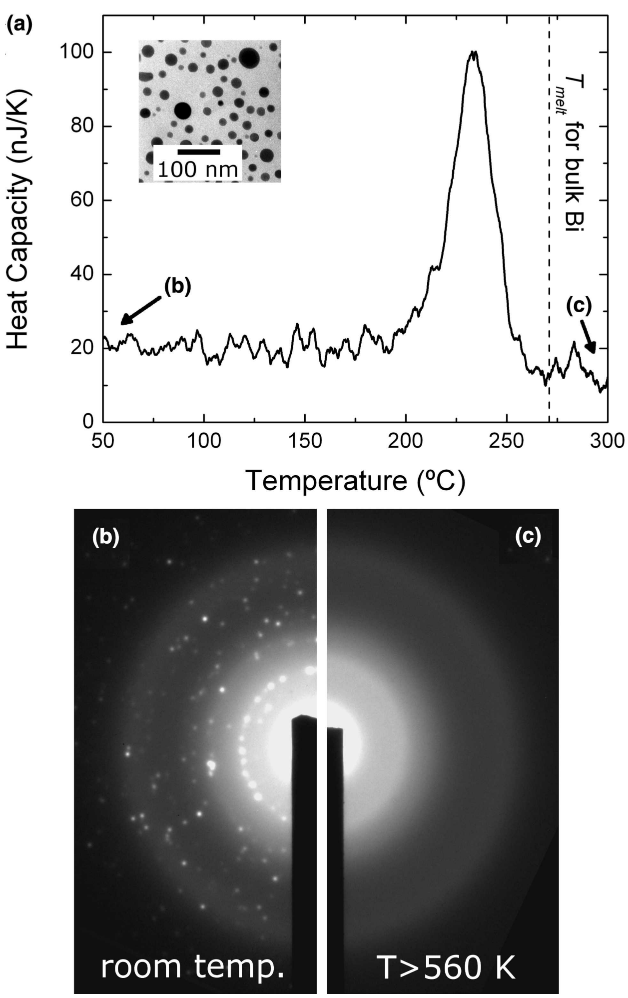


FIG. 4. In situ monitoring of the melting of Bi nanoparticles. (a) Heat-capacity measurements on a Bi nanoparticle sample; a bright-field TEM micrograph of the particles is shown in the inset. (b) Selected-area diffraction of the Bi particles at room temperature; diffraction spots are evident, reflecting the crystalline nature of the particles. (c) Selected-area diffraction of the same sample taken at elevated temperature (>271 °C). The diffraction spots disappear upon heating, showing the loss of crystalline order during melting.

By using smaller currents, this calorimeter can be turned into a precision microheater. In this case, the power applied to the device is ramped until the desired temperature is reached. The temperature can be held for any length of time. An example of this mode of operation is shown in Fig. 5, where the temperature was increased from ambient to 280 °C and held for 10 s. The heating and cooling rates were both ~120 °C/s, but can be adjusted to nearly any rate by the user.

This mode of operation is different from other TEM heating stages in that only a small area is increasing in temperature. This mode can also make use of the high heating and cooling rates possible with the MEMS device. It would, for example, be possible to heat a sample, cool it rapidly (up to 3000 °C/s),[9] and examine a quenched-in microstructure.

Using this mode of operation we directly observe the size-dependent melting effect. Figure 6 shows dark-field TEM images captured from video during the heating of Bi nanoparticles at a rate of ~5 °C/s. In real-time, dark-field image observation, one can monitor the melting of nanoparticles frame-by-frame using the loss of illumination from the selected diffracted electron beams. In Fig. 6(a), we observe three bismuth nanoparticles of different sizes; their strong contrast indicates crystalline diffraction and hence their solid nature. As the temperature

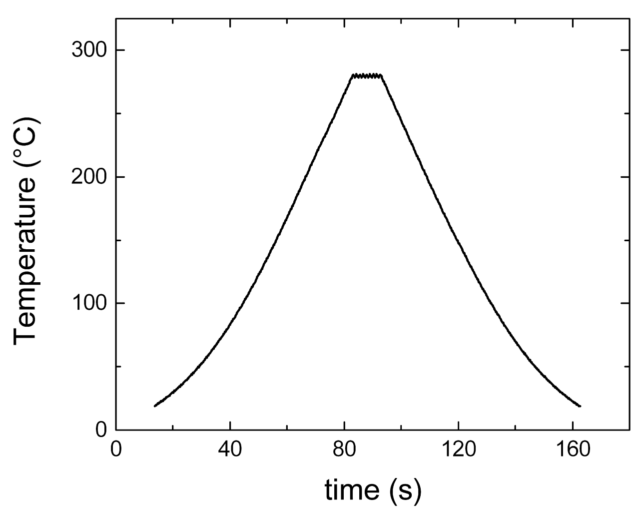


FIG. 5. Using the nanocalorimeter as a microheater. The current through the device is kept low and gradually increased until the desired temperature is reached. It can be held for an arbitrary length of time. Here, the temperature was increased from ambient to 280 °C at ~120 °C/s and held for 10 s before cooling.

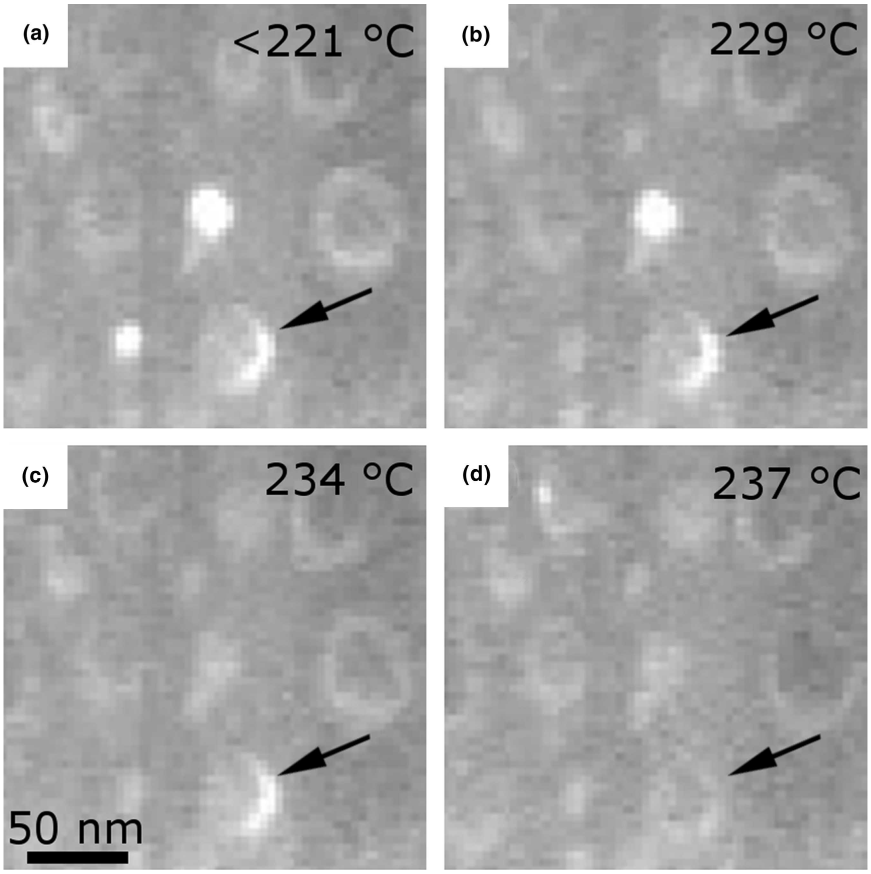


FIG. 6. Dark-field TEM micrographs of Bi particles being heated. Bi particles were slowly heated from room temperature to above the bulk melting temperature. Shown are four frames from a video, taken ~1 s apart. There is a sharp contrast change upon melting for three particles, one of which is marked with an arrow in each image. The particles show size-dependent melting temperatures, with smaller particles melting at lower temperatures than their larger counterparts. The approximate temperature for each image is (a) <221 °C, (b) 229 °C, (c) 234 °C, and (d) 237 °C. The three particles have radii of about 9, 11, and 20 nm.

increases, the 17 nm particle melts first [Fig. 6(b)], followed by the 22-nm nanoparticle [Fig. 6(c)], and then the 41 nm particle [Fig. 6(d)]. The four images were taken at <221, 229, 234, and 237 °C.

## IV. SUMMARY

In summary, we have designed a specimen holder to allow the use of MEMS sensors and actuators inside a TEM, allowing the versatility of MEMS devices to be exploited while maintaining the full capabilities of the microscope. The sample loading mechanism is simple and reliable. We successfully operated in situ a MEMS-based nanocalorimeter capable of achieving heating rates in the range from $3 \times 10^4$ °C/s up to $10^6$ °C/s and performed size-dependent melting experiments on bismuth nanoparticles simultaneously with TEM imaging and diffraction, demonstrating that the MEMS device functions reliably during TEM observations. We have demonstrated that operating a MEMS-based device in the TEM offers a range of heating experiments unachievable with traditional heating stages. The platform for interchangeable MEMS-based modules is available at the Center for Microanalysis of Materials (a user facility), for scientists to develop and use their own miniaturized laboratories for dynamic TEM studies.

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