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**Author:** Heeres, Erwin  
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A compact, two-stage nanomanipulator was designed and built for use inside a scanning electron microscope (SEM). It consists of a fine stage employing piezo-stacks that provide a 15 micrometer range in 3 dimensions and a coarse stage based on stick-slip motors, commercially available from Attocube. Besides the fabrication of enhanced probes for scanning probe microscopy and the enhancement of electron field emitters, other novel manipulation processes were developed, such as locating, picking up and positioning small nanostructures with an accuracy of ~10 nm. In combination with in situ $I-V$ experiments, welding and etching, this results in a multi-purpose nano-factory, enabling a range of new experiments.

This chapter is based on the following publication:
3.1 Introduction
Nanomanipulation inside an electron microscope can give control on a very fine scale while providing real time feedback on the object being manipulated. A nanomanipulator extends the applicability of the electron microscope far beyond that of an imaging tool, much like other available SEM add-ons, like GIS, EDX or a variable temperature stage. Sample fabrication processes often include characterization and localization of features of interest using optical microscopy, AFM or SEM and a subsequent design of actuation, measurement or control structures, often by lithographic processes. Drawback of AFM and STM manipulation is that the process cannot be imaged, only the result, as the object which is used to manipulate with, is also used to obtain the image. With a manipulator inside an SEM however, the feature of interest can be accurately positioned in situ, immediately after localization onto another predefined structure.

In this chapter we first discuss the constraints to our design set by our electron microscope, then we discuss the design considerations that improve the user friendliness of the manipulator, the properties of the manipulator and finally we give some examples of fabricated structures.

3.2 Design considerations
To image and manipulate even the smallest nano-objects, like as-grown single-walled carbon nanotubes (SWNTs) lying on a Si substrate, or protruding from the edge of such substrates, we employ a 30 kV SEM (FEI, Nova NanoSEM), which is equipped with a field emission source and a magnetic immersion lens system and has a measured resolution of 1 nm. To reduce the deposition of amorphous carbon during SEM imaging, a plasma-cleaner is installed and used to regularly clean the SEM chamber. Because of this, it is also necessary to use exclusively UHV compatible materials inside. Such a high-resolution microscope also imposes a number of restrictions on the design of a nanomanipulator to be used inside. Because of its magnetic immersion lens, all materials used in the manipulator have to be non-magnetic. Due to the size of the chamber, a compact design with a height of less than 57 mm is needed, such that the manipulator fits in the limited space
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Figure 3.1 (a) 3D image of the nanomanipulator with the following numbered parts: (1) fine stage piezo-actuator, (2) IV-connector, (3) flat substrate holding objects to be manipulated (e.g. nanotubes, nanowires, diamond nanocrystals), (4) flat substrate slider on coarse stage, (5) AFM chip onto or by which objects are manipulated, (6) AFM chip slider on fine stage, (7) fine stage. (b) CCD image of the nanomanipulator installed in the SEM. The total available height underneath the polepiece is 62 mm. To be able to work at eucentric height, a working distance of 5 mm is required. The entire manipulator (total height: 52 mm) fits underneath the final lens and is screwed onto the default SEM stage. An additional adapter block facilitates installation and removal of wiring. (c) Schematic diagram of the nanomanipulator setup. (d) Two additional sliders. Above: field emission gun (FEG) source holder. Below: probing tip holder, e.g. to hold an etched tungsten tip.
underneath the final lens and experiments can be performed at the SEM eucentric working distance of 5 mm (see also Figure 3.1). With a total height of 52 mm for the entire manipulator we can thus work at a maximum working distance of 10 mm down to the smallest allowable working distance. The entire manipulator can be positioned within the chamber by moving the SEM stage.

To allow a wide variety of experiments, sliders were made that allow manipulation of different types of objects: sharp tips, e.g. etched metallic wires, AFM chips or field emission sources, but also flat samples, see Figure 3.1a and d. By using such sliders, the time needed to create a functionalized probe is reduced, because it allows the rapid exchange of the tip and/or the sample that contains the objects that are to be mounted. For this too, the SEM is more convenient than a TEM, where sample sizes are restricted to a few millimeters and waiting times are often longer. The sliding system has been designed in such a way that different holders – each designed for a specific tip – slide onto the manipulator base. A guiding rail and spring clamping assembly enable a stable but movable connection. Furthermore, the detached slider enables simple installation and positioning of a tip or substrate outside the confined environment of the SEM chamber.

The range of motion of the manipulator should be large enough to be able to preposition the samples manually without the need of an optical microscope. As nanomaterials are often grown onto substrates of several cm², a range of several millimeters is desirable such that cleaving of the sample is not necessarily needed and a large area can be searched to find a suitable nano-object to be mounted. The sizes of samples that can be accommodated onto our manipulator range up to 30 mm by 30 mm with a maximum height of 10 mm.

A drawback of a system with a very large range often is its poor positioning accuracy. To take advantage of a large range and a high positioning accuracy, a coarse positioning system used for the approach was combined with a separate fine positioning system. For the coarse stage, a system consisting of three stackable positioners was used (Attocube, ANP 50 series, ANC 150 step controller), all non-magnetic and UHV compatible. The fine stage, a flexure hinge design, is operated
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using piezo stacks (PI, PICMA™, P-883.50, -20 V to +120 V) with a continuous range of motion. The piezo stacks are integrated into the flexure hinges in a way that limits shear stress on the piezo stacks, such that these stacks do not break if forces act laterally on them. Due to this design, coupling between the two mutually orthogonal directions of motion is avoided as x and y motions are integrated into the same body of material. Furthermore, this design enables easy installation of an actuator if replacement would be needed.

Separation of fine and coarse motion on two different stages allows accurate imaging of the tip and overcomes problems arising from unwanted motion during coarse positioning, such as hysteresis and vibrations of the stick-slip motor, which are discussed in detail in the next section. When changing the step direction of a coarse stage actuator, it needs several step actuations before it is running properly in the desired direction. In between, a combination of two unwanted effects is observed: motion in the opposite direction (which accumulates to a total of ~300 nm) and motion in the orthogonal directions (which accumulates to a total of ~700 nm). We attribute these effects to the reorientation of the rod-shaped piezo inside its housing after a step-direction change has been given, as the housing is clamped mechanically using springs onto the piezo over which it runs forwards and backwards. We find that it is not possible to use the coarse stage to perform accurate positioning processes. However, the magnitude of these effects is an order of magnitude smaller than the range of the fine stage and simple approach and retract operations can repeatedly be performed without any tip crashes, as the fine stage is designed to be robust and incorporating a large range.

3.3 Stage movement and stability

Figure 3.2 shows the (x,y)-motion of the coarse stage as its motors step in x, y and z. For each panel in the figure a motor is given ten single step actuations in one direction followed by ten steps back, after which this sequence is repeated one more time. Arrows indicate after which points the step direction is reversed. As can be seen from the figures, the coarse stage shows hysteresis; unwanted motion in the opposite direction as well as an unwanted motion perpendicular to
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The total movement in the opposite direction accumulates to \( \sim 300 \text{ nm} \) for the x-actuator and \( \sim 25 \text{ nm} \) for the y-actuator during 10 steps. The motion in directions perpendicular to the intended motion of the x, y and z actuators, is measured to be roughly 700 nm, 300 nm, and 300 nm respectively. Due to imaging in the x-y plane, motion in the z-direction cannot be observed directly. However, as measured on the z-actuator, unwanted motion in both orthogonal directions are present, hence due to design similarities this is also

Figure 3.2 Measured movement of the coarse stage in the XY-plane after applying individual step actuations to each of the three actuators. Before the measurements, each actuator has been pre-conditioned by actuating steps (>10) in the starting direction, ensuring proper linear movement. From each starting point, 10 step actuations were given in the starting direction, after which the direction of the steps was reversed. The point after which this change is performed has been indicated with arrows. In the opposite step-direction, another 10 steps were actuated, after which the entire sequence is repeated again, without pre-conditioning. The data points were obtained by in situ SEM imaging and have been determined with an accuracy of about 10 nm. The grid lines have a separation distance of 100 nm. Actuation in the (a) y-direction, (b) x-direction and (c) z-direction.
expected to be the case for the x- and y-actuator. Although the x and y actuators are of similar types, a large difference in behavior is observed. The x actuator needs more than 10 steps for linear motion after a direction change, whereas the y actuator needs 4.

The measured movement of the fine stage over its entire range is presented in Figure 3.3. Data points were obtained by analyzing the frames from in situ recorded SEM movies. The stage was moved forward and backward once over its entire range of 15 μm; the change of direction is indicated with an arrow. From a linear fit, the angle between x and y directions was found to be $\left(90.9 \pm 0.1\right)^\circ$. Actuation in the (a) y-direction and (b) x-direction. The grid lines have a separation distance of 1 μm; data points have been determined with an accuracy of about 60 nm. (c) Actuation in z-direction with a grid line separation distance of 100 nm. This figure shows how much the fine stage moves in x and y when the actuator is moved in z by 15 μm and back. Due to the SEM’s limited depth of focus, the error bars increase when the tip is moving out of focus.

**Figure 3.3** Measured movement of the fine stage in the XY-plane over its entire range. The data points were obtained by analyzing the position of a sharp tip on the fine stage from in situ recorded SEM movies. The stage was moved forward and backward once over its entire range of 15 μm; the change of direction is indicated with an arrow. From a linear fit, the angle between x and y directions was found to be $\left(90.9 \pm 0.1\right)^\circ$. Actuation in the (a) y-direction and (b) x-direction. The grid lines have a separation distance of 1 μm; data points have been determined with an accuracy of about 60 nm. (c) Actuation in z-direction with a grid line separation distance of 100 nm. This figure shows how much the fine stage moves in x and y when the actuator is moved in z by 15 μm and back. Due to the SEM’s limited depth of focus, the error bars increase when the tip is moving out of focus.
SEM movies and measuring the position of a sharp tip that was mounted onto the stage. Because the fine stage motion is continuous, one out of every 200 frames was analyzed. The angle between x and y directions was obtained by a linear fit and equals $(90.9 \pm 0.1)$°, which shows the two directions are orthogonal within one degree. The observed diagonal motion is caused by a slight difference between the electron beam scan line direction and the positioning of the stage inside the SEM. Figure 3.3c shows the motion in x-y direction during actuation of the z-direction. A shift of about 450 nm in y direction is observed, which can be compensated for by programming the piezo control software to move the y-stage in opposite direction.

In Figure 3.4 we show the vibrations of the fine and coarse stage while obtaining an image, with a dwell time of 24 μs per pixel and 24.6 ms per line. These are the residual vibrations after engaging the active vibration isolation that the SEM is equipped with. As can be concluded from the amount and magnitude of flags and spikes, see Figure 3.4b, the coarse stage suffers much more from instabilities. These vibrations are inherent to the design of the piezo-electric actuators.

In addition, vibrations are present during actuation of the coarse stage. In Figures 3.4c-e three subsequent images obtained from a movie are depicted, that show the coarse stage shaking with an amplitude of approximately 500 nm, while it is being operated in single-step mode. The fine stage moves with an amplitude roughly three times smaller. After a few frames the vibrations are damped out.

### 3.4 Nanomanipulator operation

The coarse stage has a range of motion of 4 mm in x and y and 2.5 mm in z. The step sizes of the coarse stage are controlled by varying the driver signal amplitude and are specified to range from 25 nm to 500 nm. The step size depends on the clamping force which is set by the manufacturer as well as on the mass that is being moved and the state of the sliding surfaces. Hence the step size as a function of driving signal amplitude will vary for each actuator. When operating the coarse stage at a 10 V actuation amplitude, which represents a compromise between minimal step size and reasonable reliability, this yields an average step size of approximately
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Figure 3.4 Stability of the nanomanipulator as observed by SEM imaging. (a)-(b) Stability during image acquisition. Each image was obtained with a dwell time of 24 μs per pixel, and a linetime of 24.6 ms. Flags in each image are present due to vibrations of the stages. The length of the scalebar equals 100 nm. (a) Fine stage with a sharp AFM tip. Few spikes are observed with a maximum of ~10 nm. (b) Coarse stage with a sample of MWNTs protruding from a support sample. Many flags and spikes with a maximum up to ~50 nm are visible. (c)-(e) Actuation stability during single-step coarse stage movement. Images obtained from a movie which was recorded with a frame rate of 40 Hz. The scalebar has a length of 500 nm. (c) Immediately after step actuation. (d) After 25 ms. (e) After 50 ms. Due to conversion of the raw SEM images to an avi movie, the frame rate is automatically converted to 100 fps. In such a movie, multiple frames show the exact same SEM capture, so this is not the true frame rate which is 40 Hz. Judging from the avi movie, the second frame follows after 13 ms, whereas the third frame follows 30 ms after frame 1. However, with a SEM imaging rate of 40 Hz, this should be 25 ms and 50 ms respectively.
Although the step sizes of the coarse stage actuators are not constant, as was discussed above, this does not interfere with any of the experiments we perform due to the fine stage design.

The fine stage shows continuous motion within a range of 15 μm in x, y and z. It is operated by home-built piezo-drivers which receive an input signal from a DAC card inside the PC. Motion in x and y are orthogonal within one degree, as

Figure 3.5 The process of pulling a MWNT from its as-grown material by using a sharply etched tungsten tip mounted onto the fine stage. After approaching and attaching the MWNT (not shown) the tip is carefully retracted; movement is performed only by operating the fine stage. The substrate with the MWNT material is mounted onto the coarse (approach) stage. Four still images show the process at 4 s, 40 s, 76 s, and 112 s (movie available online). (Scale bars: 300 nm, 300 nm, 300 nm, and 3 μm, respectively)
was shown before. The z-motion is not completely decoupled from the y-motion, probably due to the use of two piezo-actuators that are not completely balanced. Over the entire range of motion of the fine stage z-piezo (15 \( \mu \)m) the stage moves by 450 nm (3 %) in the perpendicular directions, which can be compensated for by the piezo control software. When changing the direction of movement, the fine stage does not show overshoot in the wrong direction. An example of the fine stage operation during the process of mounting a MWNT is presented in a movie which can be viewed online, see Figure 3.5.7

Using Labview, a user-interface was created that can be controlled using a three axes joystick system (Saitek, X52). The speed with which either the coarse or the fine stage moves, is determined by the joystick and can be adjusted to be more or less sensitive on the joystick motion. The joysticks are also used to switch between coarse and fine positioning and to apply single step actuations, voltage pulses, etc.

To perform \( I-V \) measurements and in situ field emission tests of mounted carbon nanotubes, the sample and tip stages were electrically isolated from the base of the manipulator and wired to high-voltage connectors and feedthroughs. All wires can be disconnected from the SEM stage after which the manipulator can be removed within a few minutes for normal SEM imaging. In Figure 3.1c a schematic diagram of the manipulator system is depicted. To prevent damaging the piezo actuators during venting or evacuating the SEM, an interlock system was designed. Hence the manipulator can be operated only at ambient pressure and pressures below \( 10^{-4} \) mbar.

### 3.5 Applications

As is shown by some examples below, our design will work for a large variety of applications. To fabricate novel electron field emitters, both single-walled and multi-walled carbon nanotubes (MWNTs) and semiconductor nanowires have been mounted.\(^8\)\(^9\) Closed MWNTs were mounted by pulling them from a sample with agglomerates of carbon nanotubes, see Figure 3.5. We have managed to repeatedly mount single MWNTs with their as-grown cap by pulling the entire MWNT without
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breaking it from its as-grown material. In these experiments a large fine stage range is needed to be able to handle flexible nanotubes or nanowires. With the fine stage it is possible to manipulate micrometer-sized as-grown nanotubes and -wires, without running out of range. Not only field emitters, but also high-aspect ratio AFM-tips with carbon nanotubes have been created, which enable the studies of rough surfaces in liquid. In such mounting processes, nanotubes were cut using a voltage pulse, or by electron beam etching which was facilitated by introducing water vapor into the chamber. Using a gas injection system (GIS) attached to our SEM, fixation of the nanotube or -wire was improved by electron beam induced deposition (EBID) of platinum at the position of overlap, see Figure 3.6a. Novel nanometer-sized electrochemistry electrodes consisting of an insulating AFM tip and mounted carbon nanotube were also created to study the electrochemistry properties of substances on a very small scale. A combination of the techniques mentioned before, yields a very sensitive MRFM resonator, see Figure 3.6b.

Figure 3.6 Two examples of probes fabricated with the nanomanipulator. (a) InAs nanowire (1) after mounting on a sharply etched tungsten tip (2). Using EBID a layer of Pt (3) was deposited maskless at two positions to ensure a proper fixation of the nanowire onto the tip. The inset shows the tungsten tip with the nanowire before deposition. (Scale bars: 1 μm) (b) SiC nanowire (1) mounted on an AFM chip (2). After fixing the nanowire by EBID (3), a small magnetic (NdFeB) particle (4) was added to the very end of the nanowire tip, also by EBID. In this way novel, very sensitive MRFM cantilevers can be constructed. (Scale bar: 30 μm)
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Besides mounting, the nanomanipulator is used as a positioning tool. It has been used to pick up nano-objects and put them down somewhere else, as has been demonstrated for diamond nanocrystals placed accurately inside a photonic crystal.\textsuperscript{17,18} The initial placement of a nanometer sized object can be performed with an accuracy of about 20 nm. After placement, the positioning accuracy can be enhanced by pushing the object with the tip. This final positioning is limited only by SEM imaging resolution of about one nanometer as the fine stage has a continuous range. The procedure to position these crystals is shown in Figure 3.7. The tungsten tip was etched in such a way as to yield a somewhat blunt, stiff and strong tip for the sole purpose to select and pick up the nanocrystals. Using a tip that was etched too sharply, resulted in deformation of the tip, as some nanocrystals

\textbf{Figure 3.7} Positioning of a diamond nanocrystal. In this process a nanocrystal is picked up from a substrate onto which many were dispersed and positioned onto a different substrate containing markers located a few millimeters away. The insets in the lower left corners show a schematic representation of the position of the tip, substrate and diamond. (a) Demagnified view of both substrates and the etched tungsten tip. Substrates are tilted to facilitate picking up and positioning of nanocrystals. (Scale bar: 500 μm) (b)-(c) Picking up the nanocrystal from the substrate. (Scale bars: 500 nm) (d) and (e) Positioning near a marker on the other substrate. (Scale bars: 1 μm) (f) Demagnified view of (e), showing the positioned nanocrystal in the vicinity of a reference marker. (Scale bar: 2 μm)
were stuck to the sample very tightly. To determine whether or not the tip touches the surface, a bias voltage was set between tip and sample and the current was measured using a picoammeter (Keithley). In order to reposition nanometer sized objects it is important to create a situation in which the adhesion of the object to the tip – with which it was picked up – is smaller than the adhesion to the surface onto which the object will be put down. This can be achieved by a combination of the following strategies. We try to keep the contact area between the object that is to be repositioned and the tip as small as possible. When the object is put down one can try to roll the object, in effect wiping it off the tip. It is also possible to put the object against another object and scrape it off the tip. Finally, we can use electron induced deposition to fixate the manipulated object to the surface onto which it is to be deposited.

Another strategy to facilitate manipulation is to apply a voltage difference across the sample and the tip. The detection of a current makes it easier to navigate the tip towards the surface. Additionally, the current through the manipulated object can be used to ‘weld’ it to the tip or the surface onto which it is to be repositioned. Subsequent $I-V$ measurements can be used to characterize the quality of the electrical connections, which can be useful, e.g. for subsequent field emission, STM or electrochemistry experiments.

### 3.6 Conclusions and discussion

In conclusion, we have presented a stable and compact nanomanipulator consisting of a coarse stage with a range in $x$, $y$ and $z$ of several millimeters and a fine stage with a continuous range of $15 \, \mu m$ in all three dimensions. Its use has been demonstrated already in a wide range of experiments. Vibrations are limited to approximately 10 nm except during coarse stage actuation. Further improvements on a new coarse stage design are in progress.
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