Nanocantilever Signal Transduction by Electron Transfer

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Microfabricated cantilever beams promise to bring about a revolution in the field of chemical, physical, and biological sensor development. The resonance frequency of a microfabricated cantilever shifts sensitively because of mass loading from molecular adsorption. The minimum detectable adsorbed mass on a cantilever sensor can be increased by orders of magnitude by changing the dimensions of the device; smaller and thicker cantilevers offer higher resonance frequency and therefore better mass detection sensitivity. Here we describe micromachined silicon cantilevers that are 0.5 to 4 μm in length, fabricated with the use of a focused ion beam (FIB). In addition, we demonstrate a technique for detection of the cantilever resonance frequency that is based on electron transfer.

Keywords: Nanocantilevers, Resonance Frequency, Focused Ion Beam, Micromachining, Electron Transfer.

1. INTRODUCTION

Microfabricated cantilever beams are a focus of recent interest because of their potential use as physical, chemical, and biological sensors.[1–3] Microcantilevers are the simplest micro-electro-mechanical systems (MEMS) that can easily be micromachined and mass produced. A plethora of physical, chemical, and biological sensors based on the microcantilever sensor platform have been demonstrated.[1–3] Cantilever-based sensors can be based on adsorption-induced resonance frequency shifts or cantilever bending. The resonance frequency of a microcantilever varies sensitively as a function of mass loading due to molecular adsorption.[4] In addition, if the cantilever surfaces are chemically different the cantilever undergoes bending due to adsorption-induced changes in surface stress. Although many cantilever sensors take advantage of adsorption-induced bending as the transduction method, an approach based on resonance frequency shifts can potentially provide ultimate sensitivity for the detection of a single molecule. The resonance frequency shifts and bending of a cantilever can be measured with high precision with different readout techniques, such as optical beam deflection, variations in piezoresistivity, capacitance, and piezoelectric properties. One great advantage of cantilever-based sensors is that four resonance response parameters (resonance frequency, phase, amplitude, and Q-factor) can be measured simultaneously. Moreover, cantilever sensors can operate under liquid environments. Therefore, microfabricated cantilevers can provide the basis for a universal platform for real-time, in situ measurement and determination of physical, chemical, and biochemical properties.

The sensitivity of the cantilever bending increases as the spring constant of the cantilever is reduced. Therefore, longer cantilevers with very small spring constants are attractive for use with the adsorption-bending method. However, thermal motion of the cantilever severely limits the extent to which the spring constant of the cantilever can be reduced. On the other hand, the sensitivity of the resonance frequency shifts-based approach increases as a function of frequency of operation. Therefore, shorter, higher frequency cantilevers are more suitable for increasing the detection limit of an approach that takes advantage of frequency shifts. Therefore, cantilever sensors with extremely high sensitivity can be fabricated by simply reducing the cantilever dimensions. These cantilevers with reduced sizes belong to a class known as nano-electro-mechanical systems (NEMS).[4–6] Reducing the dimension increases energy efficiency, time response, and sensitivity. However, decreasing the cantilever size results in increased difficulties in fabrication as well as in monitoring the cantilever response.[7–10]

In this work we report on our demonstration of nanocantilevers fabricated by a focused ion beam (FIB) technique.[11] We also point out that conventional detection techniques such as optical beam deflection are not suitable for signal transduction when cantilever length is reduced below tens of micrometers. To overcome this challenge we have developed a novel transduction method based

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on electron transfer that is ideally suited for the detection of nanocantilever motion. As an electrically floating cantilever vibrates between two biased electrodes, a current flows through the system because of charge transfer between the electrodes through the floating cantilever. The basic concept of charge transport is similar to electrostatic charge shuttle demonstrated by Tuominen et al.[12]

For a rectangular cantilever, the spring constant for vertical deflection is given by[13]

$$K = \frac{Ewt^3}{4L^3} \tag{1}$$

where E is the modulus of elasticity for the composing material and w, t, and L are the width, thickness, and length of the beam, respectively. Assuming the contribution from variation in the spring constant is small, a mass dependence of the fundamental frequency can be written as

$$f = \frac{1}{2\pi}\sqrt{\frac{K}{m^*}} = \frac{t}{2\pi(0.98)L^2}\sqrt{\frac{E}{\rho}} \tag{2}$$

where ρ is the density of the cantilever material and m^* is the effective mass of the cantilever. The effective mass can be related to the mass of the beam, m_b, through the relation $m^* = nm_b$, where n is a unitless geometric parameter. For a rectangular cantilever, n has a typical value of 0.24. The mass of the adsorbed material can be determined from the initial and final resonance frequency and the initial mass of the cantilever as

$$\frac{(f_1^2 - f_2^2)}{f_1^2} = \frac{\delta m}{m} \tag{3}$$

where f_1 and f_2 are the initial and final frequencies, respectively, and δm and m are the adsorbed mass and initial mass of the cantilever, respectively. If the adsorption is confined to the free end of the cantilever, Eq. (3) needs to be modified appropriately to take into account the effective mass of the cantilever.

The mass sensitivity of a cantilever sensor can be written as

$$S_m = \lim_{\Delta m \to 0} \frac{1}{f}\frac{\Delta f}{\Delta m} = \frac{1}{f}\frac{df}{dm} \tag{4}$$

where Δm and dm are normalized to the active sensor area of the device ($\Delta m = \delta m/A$, where A is the area of the cantilever). As can be seen from the expression in Eq. (4), the sensitivity is the fractional change of the resonant frequency of the structure with the addition of mass to the sensor. When this definition is applied to the case of the cantilever sensor, the sensitivity can be expressed as

$$S_m = \frac{1}{\rho t} \tag{5}$$

where ρ and t are the density and the thickness of the cantilever, respectively. As mass is added uniformly to a cantilever, its resonance frequency decreases as a function of adsorbed mass. Note that the sensitivity of a cantilever sensor depends only on its thickness and material density.

Another characterization parameter of a cantilever sensor is its minimum detection mass density. The minimum detectable mass density can be obtained by rearranging Eq. (4) and Eq. (5) as

$$\Delta m_{\min} = \frac{1}{S_m}\frac{\Delta f_{\min}}{f} \tag{6}$$

where $\Delta m_{\min}$, $\Delta f_{\min}$ are the minimum detectable mass density and minimum detectable frequency change, respectively. Typically, minimum detectable mass density values are experimentally quoted results due to specifics of the sensor as well as the frequency detection limitations determining $\Delta f_{\min}$. Therefore, by changing the physical dimension of a cantilever one can increase its detection limits by many orders of magnitude.

2. EXPERIMENTAL DETAILS

2.1. Nanocantilever Fabrication

We have fabricated cantilevers that are 0.8 to 2 μm in length, 50 to 500 nm in width, and 25 to 100 nm in thickness, using an FIB technique. The array shown in Figure 1 has cantilevers 1000 nm in length, 250 nm in width, and 50 nm in thickness. The FIB (model FIB 200 manufactured by FEI) used for fabricating cantilevers delivers a Ga ion beam at 30 keV. The starting surfaces for fabricating cantilevers were single-crystal Si membranes with an initial thickness of 10 μm. Prior to cantilever fabrication we used the FIB to reduce the thickness of the Si membrane (down to a few micrometers), which provided us a region in which to fabricate cantilevers with small dimensions. We also used commercially available

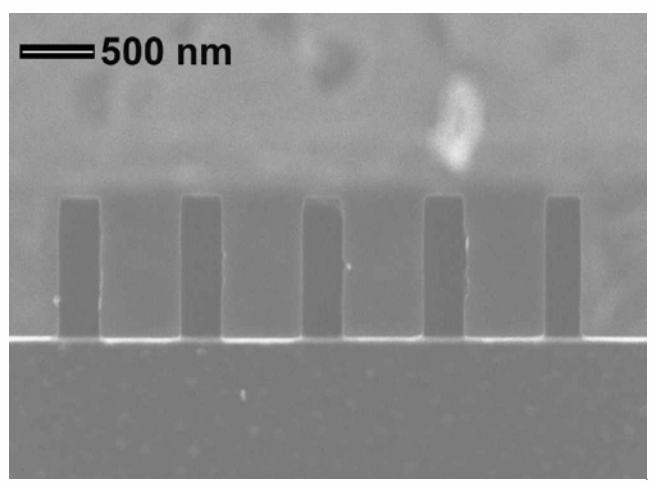

Fig. 1. An ion micrograph of an array of nanocantilevers fabricated from commercially available microcantilevers with the use of a focused ion beam.

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V-shaped silicon microcantilevers (Thermomicroscopes, Sunnyvale, CA) as a starting substrate for nanocantilever fabrication. Typical dimensions of the commercial cantilevers were 100-μm length, 20-μm width, and 1-μm thickness. Examples of such nanocantilever arrays fabricated from commercially available microcantilevers are shown in Figure 1. The calculated resonance frequency of a rectangular Si cantilever with these dimensions is 265 MHz.[13]

2.2. Nanocantilever Measurements

Optical detection of cantilever motion of nanocantilevers and nanocantilever arrays at their resonance frequency was found to be difficult because of the reduced reflection (scattered) signal from the cantilever surface. To overcome the problems associated with resonance frequency detection we have developed a novel detection method using electron transfer between cantilever and the two fixed electrode structures. With the use of an FIB this technique can be used to nanofabricate devices from different materials and is not limited to Si.[14] The cantilever and the associated electrode system were fabricated monolithically with the use of an FIB. A schematic and a scanning micrograph of the monolithic cantilever and detection system are shown in Figures 2 and 3. The length of the cantilever was 4.5 μm and the thickness of the cantilever was 0.5 μm. This type of geometry allowed in-plane motion of the cantilever. The motion of the cantilever was in the plane of the image. The micrograph shows the thickness of the cantilever. Two electrodes (labeled as source and drain) were micromachined in such a way that the cantilever can oscillate between these two structures. The source and drain electrodes were equidistant from the cantilever beam. The separation distance between the cantilever and the source and drain was approximately 50 nm. All three components, cantilever (gate), source, and drain, were electrically isolated

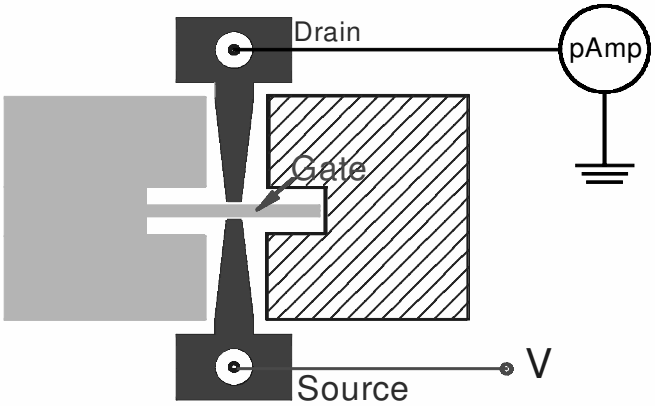

Fig. 2. Schematic of an electron transfer readout technique for cantilever motion response detection. A bias voltage is applied between the source and drain electrode. A nanocantilever acting as a gate is used to shuttle electrons between the source and drain.

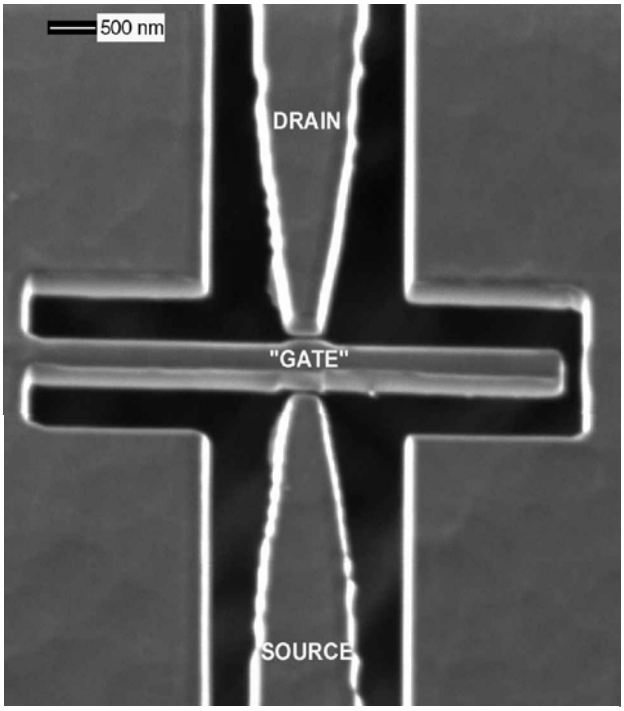

Fig. 3. An ion micrograph of a nanocantilever with a readout system fabricated from single-crystal silicon with an FIB. The cantilever "gate" was actuated acoustically and had a resonance frequency of 5.2 MHz.

from each other. The nanocantilever was excited into resonance with the use of an acoustic excitation source. The source and drain electrodes were electrically biased, and the current flowing through the system was measured with a picoammeter. All measurements were performed under ambient conditions.

3. RESULTS AND DISCUSSION

We studied the motion of the nanofabricated cantilevers with the experimental set-up described above (see Fig. 2). When a bias voltage was applied between the source and drain electrodes, no electron current flowed through the system under d.c. voltage because of the air gap between the cantilever and the two electrodes. It is possible that when the system is biased with a d.c. the cantilever can bend because of applied electrostatic force. However, such cantilever bending due to electrostatic force was too small to cause the cantilever to bend enough and make contact with the electrodes. Moreover, since d.c. bias voltage is used, no displacement current flowed through the system. As the cantilever was excited into resonance (using an acoustic source), the oscillatory motion caused the cantilever to make electrical contact with the source and the drain electrode. Electron charge was transferred to the electrically floating cantilever "gate" when the cantilever made contact with the source electrode. The electron charge on the cantilever was subsequently transferred to the drain electrode as the cantilever oscillated in the

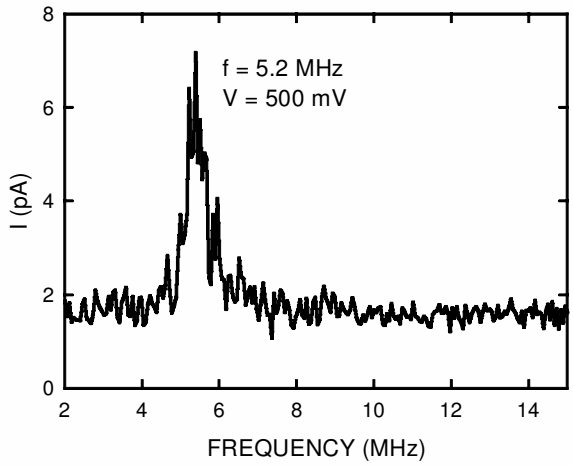

Fig. 4. Electron current measured as a function of frequency. The nanocantilever gate was actuated acoustically around its first harmonic resonance, and the effective electron transport was monitored for an applied bias of 500 mV.

opposite direction and made contact with the drain electrode. As the cantilever made intermittent alternating contact with the source and the drain electrodes, electrical charge was transferred from the source to the drain electrode, causing a net electron current to flow through the system.

The applied bias voltage between the source and the drain electrode was 500 mV. When the cantilever was acoustically excited the current through the circuit exhibited a peak at 5.2 MHz (see Fig. 4). At resonance the acoustic frequency used for cantilever excitation matches the natural frequency of the cantilever. In Figure 4 we show the electron current through the system, measured with a picoammeter, as a function of the acoustic frequency that excited the cantilever into motion. From the data in Figure 4 it appears that at resonance the electron current increases to 7 pA against the noise floor of 1.7 pA. The mechanical Q-factor of the cantilever was calculated to be around 10 when the device was operated in air. The Q-factor can be increased by an order of magnitude in a He gas environment; when the cantilever oscillates in a low-viscosity medium such as He, oscillation damping is reduced. At the resonance frequency the measured electron current was approximately 7 pA.

The flow of electron current between the source and drain electrodes facilitated by the cantilever motion can be understood as follows. As the cantilever comes in contact with the energized source electrode, the cantilever will pick up charge q, either by electron tunneling or by physical contact. This charge will then be transferred to the drain electrode as the cantilever oscillates and makes contact with the drain electrode. The total current due to electron transfer can be expressed as

$$i = 2nef \tag{7}$$

where n is the number of charge quanta (electrons) transferred to the cantilever, e is the electronic charge, and f is the excitation frequency. From the data presented in Figure 4 we obtain a value of $n = 5$ at resonance and $n = 1$ at frequencies far from resonance. These values correspond to 1.7 pA noise current and 7 pA current at resonance, respectively.

From the physical dimensions of the Si nanocantilever, a cantilever mass of 10^{-15} g can be calculated. If the resonance frequency can be measured with a resolution of 100 Hz, the minimum detection limit for adsorbed mass can be calculated as 10^{-19} g at a resonance frequency of 5.2 MHz. This detection limit is ample to detect changes in adsorbed mass due to a single biomolecule of a large protein or a DNA strand of 50-nm length. The detection limit can be further improved by increasing the Q-factor of the cantilever. The Q-factor can possibly be amplified by a feedback system, which was demonstrated for optical signal transduction.[15, 16]

Compared to currently used signal transduction methods, electron transfer by a cantilever system is ideally suited for NEMS nanocantilevers. Conventional techniques of measuring the resonance frequency, such as optical beam deflection, fall short when applied to micromachined nanocantilevers. In optical beam deflection the cantilever motion is measured by reflecting a laser diode off the free end of a cantilever into a position-sensitive detector. The shortcomings of optical techniques are due simply to lack of sufficient reflected (or scattered) optical signal from the cantilever beam. Optical beam deflection is extremely sensitive when used for cantilevers that are 50 μm to a few hundreds of micrometers long, whereas electron transfer signal transduction is extremely sensitive for cantilevers that are a few *hundred* nanometers to a few microns in length.

The applicability of electron transfer signal transduction for aqueous environments is extremely challenging. The presence of electro-active ions in the water can cause a large Faraday leakage current that can overwhelm the electron transfer signal. The leakage current, however, can be significantly reduced with proper insulation, reduced bias voltage, and reduced number of charge carriers in the solution. However, this technique may be applicable for biosensors that can be operated in a humid atmosphere.

4. CONCLUSIONS

In conclusion, we have described an integrated nanocantilever and a cantilever response detection system fabricated from single-crystal silicon wafers with the use of a FIB. The signal transduction is based on charge transport from a source to a drain electrode facilitated by an acoustically driven, electrically floating nanocantilever. The electron transfer signal transduction is simple and compatible with MEMS/NEMS fabrication. Optimized

nanocantilevers can be used as chemical and biological sensors with single-molecule sensitivity. Moreover, quantized electron transport with a nanocantilever offers a novel path for the development of a new sensor paradigm based on electron transport. Furthermore, electron transfer current as a function of bias voltage may provide a wealth of information on quantized charge transport.

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