



# Atomic force microscopy with qPlus sensors

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Atomic force microscopy is one of the most important tools in nanoscience. It employs an atomic probe that can resolve surfaces with atomic and subatomic spatial resolution and manipulate atoms. The qPlus sensor is a quartz-based self-sensing cantilever with a high stiffness that, in contrast to Si cantilevers, allows to oscillate at atomic radius amplitudes in the proximity of reactive surfaces and thus provides a high spatial resolution. This article reports on the development of this sensor and discusses applications in materials research.

## Introduction

“Some of the best innovations come from combining two previous innovations,” as Walter Isaacson noted on page 61 of his recent biography of Elon Musk.<sup>1</sup> The introduction of atomic force microscopy (AFM) in 1986 by Binnig, Quate, and Gerber<sup>2</sup> was an innovation that, together with scanning tunneling microscopy (STM), embodies the most important tools of the burgeoning field of nanoscience. AFM is an extension of the STM, invented by Gerd Binnig and Heinrich Rohrer in 1981. The invention of STM was quickly rewarded with the Nobel Prize in Physics in 1986, shared with Ernst Ruska who had invented electron microscopy 50 years earlier. In AFM, the STM tip is replaced by a cantilever with a sharp spike sitting on its end that allows to measure the forces between tip and sample. The cantilever and its tip with their detection electronics embody the core of the atomic force microscope, it determines its spatial- and force resolution as well as the imaging speed and the other functions of AFM (i.e., performing atomic manipulation or chemical bond breaking or making).

The choice of silicon microfabrication as the best way to create the core of the atomic force microscope (AFM), the cantilever, seems to be straightforward. Here, it is argued that combining the AFM innovation with the innovation of the quartz revolution brought to the watch industry in the 1970s creates valuable outcomes.

## True atomic resolution by AFM

Shortly after AFM was introduced, atomic lattice images were produced. However, these images did not show any atomic defects or atomic steps. Therefore, it had to be concluded that the tip exposed a relatively large contact area with many atomic contacts. I started my PhD thesis with G. Binnig at the IBM Physics Group Munich, hosted by T. Hänsch in the Ludwig-Maximilians-Universität in downtown Munich. At that time, Binnig had three research goals which he offered me as a PhD topic: (a) to build a tunneling detector sensitive enough to measure gravity waves; (b) to sequence DNA by STM; and (c) to improve AFM to obtain true atomic resolution. I chose (c). The thinking was that if atomic resolution by AFM is possible, it should be in the cleanest environment possible: ultrahigh vacuum and liquid helium temperatures (4 K). Binnig and C. Gerber had already designed an ultrahigh vacuum (UHV) system with an elevator for a liquid helium dewar that allowed to quickly submerge a conflat 35 vacuum tube into a helium bath and thus cool it to 4 K. My job was to design the microscope that went into the tube.

**Figure 1** a shows the force detector that was commonly based on a tunneling junction in the early years of AFM. It consisted of a micromachined  $\text{Si}_3\text{N}_4$  cantilever that was coated with Cr and Au for electrical conductivity. The cantilever was held onto a wedged cantilever support plate by a spring clamp, allowing

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lateral adjustment to meet the tunneling tip. The tunneling tip was Pt coated to ensure good conductivity and mounted onto a piezo tube that allowed fine adjustment. The wedged support hinged around two spring-loaded screws, held by a rod that connected to a coarse adjust beam that again hinged around a pivot to provide mechanical connection to a rod that connected to a linear UHV feedthrough to adjust the distance by stepper motors such that the range of the z-piezo was sufficient to bring the tip within tunneling distance to the cantilever.

The lateral adjustment of the cantilever with respect to the tunneling tip had to be done on the lab bench under an optical microscope. This adjustment had to withstand the bakeout procedure of the microscope (about 130°C for 24 h) as well as the cooldown procedure to liquid helium. In fact, the device only worked properly if the tip position after bakeout and cooling came close to the root of the cantilever (green spots in Figure 1b); otherwise the van der Waals (vdW) forces would pull the soft cantilever into uncontrolled contact with the tunneling tip. This instability is widely known as the jump-to-contact problem; it occurs in the static operation mode if the gradient of the tip-sample force is negative and its magnitude is greater than the stiffness of the cantilever. The cantilever used here was relatively soft with  $k = 0.37$  N/m. In the dynamic operation modes, the jump-to-contact problem can be avoided for large enough oscillation amplitudes (more is discussed next).

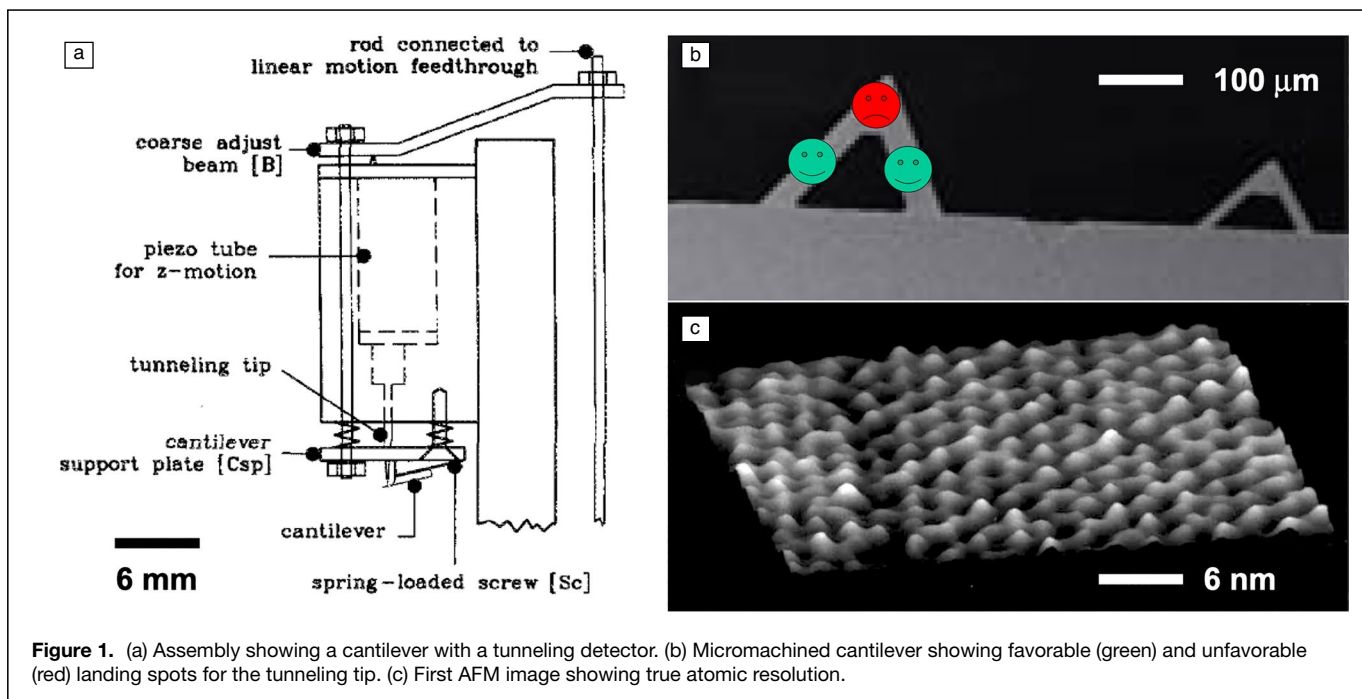
Although this microscope was cumbersome to operate and the success rate in getting good images was low, we did achieve true atomic resolution on KBr(001) with it as shown in Figure 1c.<sup>3,4</sup> However, a special technique was required to obtain atomic resolution. Upon approaching the surface, jump-to-contact occurred, leading to an uncontrolled hard

landing of the cantilever on the surface that resulted in a local damage of the surface. The attractive van der Waals force was on the order of 10 nN, and this load is too high for a single atomic contact to counter. Therefore, once jump-to-contact had occurred, the load of the strong vdW forces needed to be reduced by gently withdrawing the sample a few nanometers while the tip was still in contact to the sample. When the repulsive force on the front atom of the tip was thus reduced to a value on the order of 1 nN, the sample could be cautiously moved laterally to an undestroyed surface area.

Shortly after, Binnig and his second graduate student F. Ohnesorge even achieved true atomic resolution in ambient conditions on calcite<sup>5</sup> using a Si cantilever with optical beam deflection detection. In the beam deflection method, the bending of the cantilever is measured by shining a focused laser beam on the end of the cantilever and collecting the reflected light with a split photo diode.<sup>6,7</sup> This method is widely used in commercial AFMs for ambient conditions.

The holy grail though, atomic resolution of Si(111)-(7×7), had to wait two more years as the challenges imposed by the jump-to-contact problem were so high that even Binnig, the very optimistic inventor of AFM, thought it would be highly nontrivial.

Perhaps, it is noteworthy that in those early days of AFM, we not only built every part of the microscope ourselves and thus knew the machine inside out, we also employed as many senses as possible when performing experiments. Wearing a headset with an amplified audiosignal derived from the tunneling current was a must in addition to the visual monitoring of the current on the display of the oscilloscope. Therewith, we could verify the health of the machine, a slight slap on the vacuum chamber would result in a metallic ping that showed that the tunneling junction was clean and fully sensitive and



**Figure 1.** (a) Assembly showing a cantilever with a tunneling detector. (b) Micromachined cantilever showing favorable (green) and unfavorable (red) landing spots for the tunneling tip. (c) First AFM image showing true atomic resolution.

any external noise sources could be quickly noticed by the highly sensitive auditorial sense. Even the tactile sense was employed - a slight touch of the vacuum chamber with the cusps of a few fingers would indicate if any line frequency related humming made its undesired way into the experiment.

### Resolving Si(111)-(7×7) by AFM

Resolving Si(111)-(7×7) by AFM was an obvious goal for many groups around the world at that time for historical and technical reasons. The beautiful images that STM delivered from Si(111)-(7×7) convinced the scientific community that atomic resolution was real. Therefore, repeating this feat by AFM was important. While atomic resolution by AFM on KBr and calcite had been demonstrated, imaging Si(111)-(7×7) posed extra challenges because of its chemical reactivity and the strong bonds it can form with the front atom of the AFM tip.

During my time as a PhD student with Binnig in Munich (1988–1991), we obtained micromachined silicon cantilevers from the Quate group in Stanford. S.-i Park, a graduate from Quate who designed one of the first STMs that successfully imaged Si(111)-(7×7) as his PhD project, had founded Park Scientific Instruments in Mountain View, Calif., initially based on the fruits of his dissertation. Customers demand though quickly turned toward an AFM. Park offered me a job to design an AFM that operates in ultrahigh vacuum in Sunnyvale, Calif. Ever since the start of my studies I devoured the *Feynman Lectures on Physics* and I badly wanted to study and work in California. What an opportunity to move to silicon valley, work on a fun and important project, train for and take part in triathlons year round, learn to fly an airplane, and explore the Santa Cruz mountains by motorcycle and mountain bike.

B. Trafas and I designed a microscope for UHV at room temperature that was based on S.-i Park’s Stanford STM SU-2.<sup>9</sup> The SU-2 already had a preparation stage for Si(111)-(7×7) and a tip carousel that allowed to exchange STM tips *in situ*. We upgraded the microscope with an inertial stage for coarse motion in  $x$ - (lateral) and  $z$ - (vertical) sample motion and we exchanged the single electrode spring-loaded STM tip holder with a magnetic holder with three electrical contacts. In addition, the control electronics was switched from an HP workstation to an IBM compatible personal computer based on Microsoft Windows 3.11 and the analog feedback control was upgraded to a Motorola digital signal processor board.

The core of this new AFM “AutoProbe VP,” the force sensor, was a piezoresistive Si cantilever (“piezolever”) developed by Tortonesi et al. in Quate’s group.<sup>11</sup> The piezolevers allowed for a simple electrical deflection detection, a deflection of 1 pm caused a small but measureable relative resistance change of  $3 \times 10^{-9}$ . The resistance of those cantilevers was about 2 k $\Omega$ , and making them part of a Wheatstone bridge (Figure 3 in Reference 9) allowed to transform deflection to voltage. The voltage we used across the bridge was generated by a precision voltage source of either 2.50 V or 10.0 V.<sup>9</sup> The sizable power dissipation of  $P_{loss} = U^2/R$ , amounting to  $P_{loss} = 12.5$  mW for a half

bridge voltage of  $U = 5$  V lead to a significant heating of the piezolever, limiting their use in low temperature microscopes. The piezolevers had a relatively large  $1/f$ -noise, meaning that constant deflections could only be measured with limited accuracy and a sizable drift.

In 1991, Albrecht, Grütter, Horne, and Rugar introduced the frequency modulation (FM) mode in AFM and demonstrated in impressive magnetic force microscopy experiments that FM-AFM could provide much lower noise than amplitude modulation.<sup>8</sup> Albrecht even designed an analogue FM detector box that was utilized in the ambient magnetic force microscopes of Park Scientific Instruments. We then adapted this box to the AutoProbe VP and utilized the FM mode with piezolevers in vacuum. This frequency detector used an LC bridge and a phase comparator to generate the frequency shift output, it was notorious for drift - approaching a hand from far away would cause several Hz of apparent frequency shift.

First results on KBr(001) were quite promising (Figure 5 in Reference 9) and atomic rows on Si(111)-(7×7) appeared in late 1993 using an oscillation amplitude of  $A = 3.3$  nm (Figure 11 in Reference 10).

The oscillation amplitude is noteworthy here because in the frequency modulation mode, the frequency shift  $\Delta f$  is the physical observable. The frequency shift  $\Delta f$  is independent from amplitude in the gradient approximation

$$\Delta f(z) = \frac{f_0}{2k} k_{ts}(z), \quad 1$$

where  $f_0$  is the eigenfrequency of the cantilever,  $k$  is its stiffness, and  $k_{ts}(z)$  is the tip-sample force gradient. In practice, chemical bonding forces typically decay exponentially with distance with a decay length on the order of  $\lambda \approx 50$  pm. A precise calculation shows that the force gradient  $k_{ts}$  needs to be replaced by an averaged force gradient  $\langle k_{ts} \rangle(z, A)$ <sup>12</sup> that is obtained by weighting the force gradient with a semicircular weight function over the vertical distance interval from  $-A$  to  $+A$ :

$$\langle k_{ts} \rangle(z, A) = \frac{2}{\pi} \int_{-1}^1 k_{ts}(z + \zeta A) \sqrt{1 - \zeta^2} d\zeta. \quad 2$$

When keeping the lower turnaround point of the oscillating cantilever at a constant height, for amplitudes of  $A \ll \lambda$ , the frequency shift remains indeed constant. However, once  $A \approx \lambda$ , the frequency shift starts to roll off proportional to  $A^{-1.5}$ ;<sup>13</sup> therefore, it is important to use small amplitudes.

On the experimental side, it is key to hone skills and to watch closely for seemingly ancillary data. Binnig taught me the habit of listening to the physical observables on an audio channel. In those experiments where an amplitude of  $A = 3.3$  nm was used, the frequency shift channel was plugged into the input of my audio headset amplifier. The signal sounded noisy and scratchy upon approaching the tip, as if the amplitude controller had a hard time to maintain a constant amplitude. To remedy this instability, I increased the oscillation amplitude set point and heard on the audio channel that

the oscillation was much more stable. Finally, I got a nice and stable frequency shift signal although the amplitude had to be increased to the giant value of  $A = 34$  nm. A later calculation<sup>13</sup> showed that for a tip-sample force with range  $\lambda$ , optimal signal-to-noise ratio would be observed for  $A \approx \lambda$ . This apparent paradox is solved if one considers that a primary condition needed to be fulfilled: stable oscillation, that is, avoidance of jump-to-contact. Once the oscillation amplitude remained stable at this large value and the frequency shift signal had only little noise, one could hear a low frequency signal on the frequency shift channel that was synchronized with the fast ( $x$ ) scanning motion and sounding like “wush-wush” similar to the sound of a windshield wiper on a car. When reducing the frequency shift set point to an even more negative value, a faint ringing similar to the bell of a bicycle could be heard. This ringing was the effect of the surface atoms on the frequency shift, and soon, those atoms also appeared on the computer screen as well. With more fine-tuning of the imaging parameters, the quality of the images improved and the data shown in **Figure 2** and published in Reference 14 convinced us and our colleagues that a method that resolves reactive surfaces by AFM with atomic resolution had been found.

It did not take long before similar results could be reproduced in Japan by the AFM manufacturer JEOL<sup>15</sup> and by S. Morita’s group in Osaka.<sup>16</sup> Those experiments used similar cantilever stiffnesses of  $k \approx 20$  N/m and oscillation amplitudes of  $A \approx 20$  nm, showing that this *soft cantilever-large amplitude* method was viable. Morita then launched an “International Conference on Noncontact Atomic Force Microscopy” that first took place in 1998 in Osaka and that is still held annually today in various places around the globe.

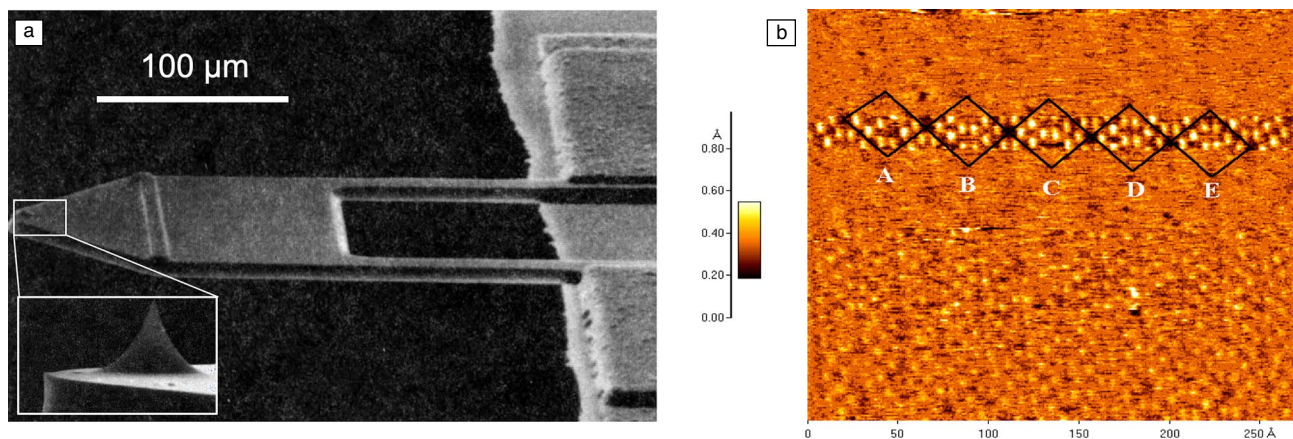
When the silicon 7×7 challenge was solved with our commercial AutoProbe VP AFM, I was promoted to director of vacuum products. Deep down, I thought the hard problems are solved and it is time to leave the field for good and venture off into something new. Our microscope had solved such a hard

problem, but I wondered why it did not right away sell in quantities of many hundreds. My expectation was that if you solve a tough scientific problem, this solution potentially provides a great benefit to mankind that should be reflected in large sales numbers. Clearly, I needed to learn a little more about how to make sure that a good idea is adapted out in the world. I thought that joining a management consulting firm could help me gain this knowledge in the shortest possible time.

At that time, I often played tennis in Mountain View with M. Sakharov-Liberman, granddaughter of a famous physicist and human rights activist, the Nobel Peace Prize laureate Andrei Sakharov. Like her grandfather, Sakharov-Liberman was a physics graduate of Moscow State University and, at that time, was studying for her MBA degree at Stanford University.

Upon receiving a rejection letter from the German office of McKinsey, a leading management consultancy, I showed my initial application to Sakharov-Liberman. I had written it, brimming with self-confidence of someone who had just single authored his first *Science* article. Sakharov-Liberman threw her hands over her head in disbelief how I could send off such a poorly written letter. She helped me to revise it, and I sent it to McKinsey again. Of course, they were aware of my earlier unsuccessful application, but they let me know that persistence is a very important trait in a management consultant, in particular when paired with a sophisticated new approach as a response to initial failure. This time, I got an invitation to a few rounds of interviews with their recruiting team of consultants and senior partners.

Shortly after, I received a very attractive offer as a senior associate with the consulting firm I had dreamed of working for. The new calling was exciting and rewarding with highly intelligent and worldly colleagues - some of them became friends for life. The downside was a stress level that I had not known before. I was used to working long and very long hours in the lab, but the psychological



**Figure 2.** (a) Piezoresistive cantilever. (b) First AFM image showing atomic resolution of Si(111)-(7×7). Recorded in the topographic mode (constant frequency shift) with  $f_0 = 114$  kHz,  $k = 17$  N/m,  $\Delta f = -70$  Hz, and  $A = 34$  nm.

challenge of entering a new field of expertise and the rightful expectation of the client that one delivers profound value within a few weeks was new. I often thought, that's what a crossing of the Sahara must be like, beautiful sights around the clock, but all is at the brink of survival. After a while, a gnawing thought grew stronger and stronger: I had been given the unique gift of a PhD training by *the* pioneer of the nanotech revolution. Binnig has only trained two graduate students in his life, F. Ohnesorge and myself. F. Ohnesorge was no longer working in science, so I felt I should go back and carry that torch from one of the founding fathers of nanoscience further rather than doing consulting work that could be done even better by many other people.

Our clients usually engaged consultants when they sought help in identifying and curing the source of a lack in profit. McKinsey liked to hire physicists, as we are trained to create mathematical models of complex situations and often quickly identify the levers that turn the outcome in the intended direction. Once the specific function in the client organization that was most responsible for poor financial performance was identified, we often compared this function to the best in class worldwide, we benchmarked it, to see how we can improve.

### Introduction of the qPlus sensor

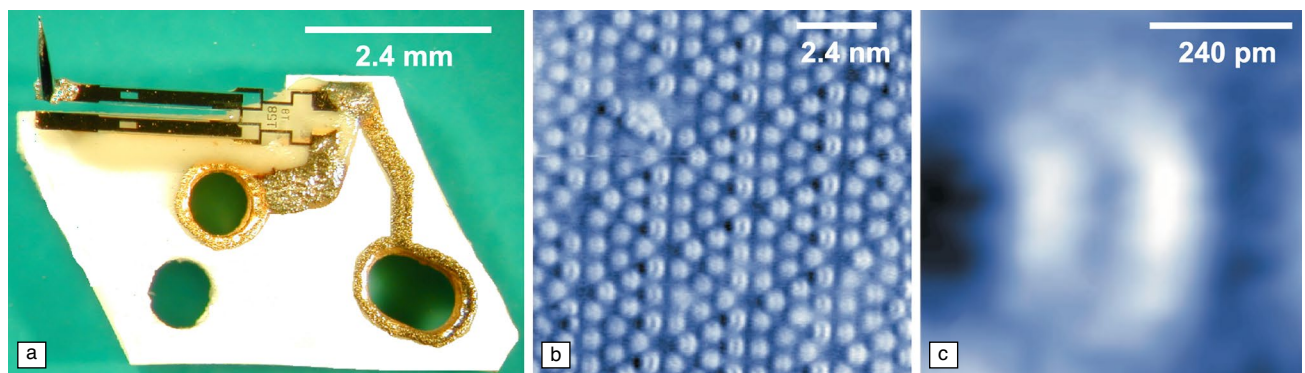
Triggered by the benchmark studies, I started to wonder whether building the force sensing cantilevers from silicon was really the best choice. After all, the averaged force gradient that acts between tip and sample alters the oscillation frequency of the cantilever, and exact frequency measurements would lead to precise force measurements. A benchmarking mindset would immediately ask: what method is best to measure frequency or its inverse, time? The watchmakers had been working on this question for centuries and the 1970s brought the quartz watch revolution. Could one turn the quartz tuning forks into force sensors? Most of the tuning forks for watch applications came at a frequency of  $f_0 = 32768$  Hz because dividing this 15 times by 2 yields a beat of one per second. Tuning forks come in different sizes though and it turned out

that the type used in Swatch wristwatches had an almost ideal stiffness of  $k = 1800$  N/m, allowing for stable oscillation with sub-Angstrom amplitudes even in the force field of strong covalent interactions.

I told my supervisors at McKinsey about this idea and was granted paid and unpaid vacation to pursue this idea in my quickly established home laboratory. Two questions sprang up immediately concerning the replacement of silicon cantilevers by quartz tuning forks:

1. Is it possible to obtain very high quality factors when immobilizing one of the tines of the fork?
2. Is the electrical deflection signal still measurable when its amplitude amounts to the radius of a hydrogen atom rather than 200 000 times larger as prevailing in quartz watches?

The experiments in my home laboratory resulted in a positive answer to both questions, leading to a patent application and the drive to find a place where I could pursue this idea full time. In that first patent application, I already named the device QPLUS or qPlus sensor to reflect the difference to tuning fork sensors that had been employed in scanning near-field acoustic microscopy by Dransfeld et al.<sup>17</sup> and scanning near-field optical microscopy by Karrai and Grober.<sup>18</sup> A tuning fork is a system of two coupled oscillators, and its high  $Q$ -value is a consequence of the perfect antisymmetric oscillation mode. Breaking this symmetry by mounting a tip on one prong leads to a dramatic loss in  $Q$ . Putting a counter mass on the other prong would not cure the asymmetry once the tip interacts with the surface: a single covalent bond between two Si atoms has a stiffness of 340 N/m, so one prong would have an unchanged stiffness of 1800 N/m, the effective stiffness of the other would amount to 2140 N/m. The innovation of the qPlus sensor was that the prong without tip was immobilized as shown in **Figure 3a**, rendering it into a self-sensing quartz cantilever. This allowed for a high  $Q$ -value even when using massive tips or when the tip of the sensor was subject to a large force gradient.



**Figure 3.** (a) First generation qPlus sensor. (b) AFM image of Si(111)-(7×7) with the qPlus sensor shown in (a). Parameters:  $f_0 = 16860$  Hz,  $\Delta f = -160$  Hz,  $k = 1800$  N/m,  $A = 800$  pm. (c) Enlarged view of a single adatom.

I needed a new position with a well-equipped laboratory to enable me to pursue these ideas on a professional basis. Gerber introduced me to former IBMer J. Mannhart who had just gotten an offer for a Chair at the University of Augsburg and who was looking for an AFM expert. After an on-site interview in Augsburg, Mannhart offered me a permanent position at his Chair and he was enthusiastic about supporting ideas about a new force sensor, as this sensor would also facilitate low temperature AFM. We started in January 1997 in a brand new building with empty labs and purchased a used AutoProbe VP system including UHV chamber and Si preparation tools.

In the first year in Augsburg, while we were establishing the labs, I finished a theoretical study that supported the feasibility of a force sensor that was one hundred times stiffer than what was used then, mainly because it would allow it to operate at very small oscillation amplitudes without suffering from jump-to-contact.<sup>19</sup> Also, we published first experimental results on using the qPlus sensor for imaging in ambient conditions.<sup>20</sup> We were very grateful for having obtained a prototype of a quartz controlled, digital phase-locked-loop detector from Nanosurf AG that showed no noticeable drift and was later known as the easyPLL. Once we got ready to operate the sensor in UHV, a big surprise waited for us.

**Figure 3b** shows a much nicer image of the Si(111)-(7×7) surface than the data recorded with a piezolever in **Figure 2**.<sup>21</sup> There is a defect in the upper left part of the image, and most notably, every surface atom appears to be split in two halves. The origin of the two halves in the image is, according to our understanding, the presence of two tip orbitals originating in a single front atom of the tip that are imaged by the spatially highly localized  $sp^3$  orbitals of the Si adatoms image (**Figure 4A** in Reference 21). We modeled these data successfully with the Stillinger–Weber potential,<sup>22</sup> a seven-parameter model potential that describes the bonding character of Si very well.

This result came to a surprise to the community. For more than a decade, it was accepted as a fact that the spatial resolution of AFM was inferior to STM, and here we claim that AFM can even obtain subatomic spatial resolution. The

**Table I. Comparison between size, stiffness, typical operational amplitude, material, and detection mechanism between Si cantilever and qPlus sensor.**

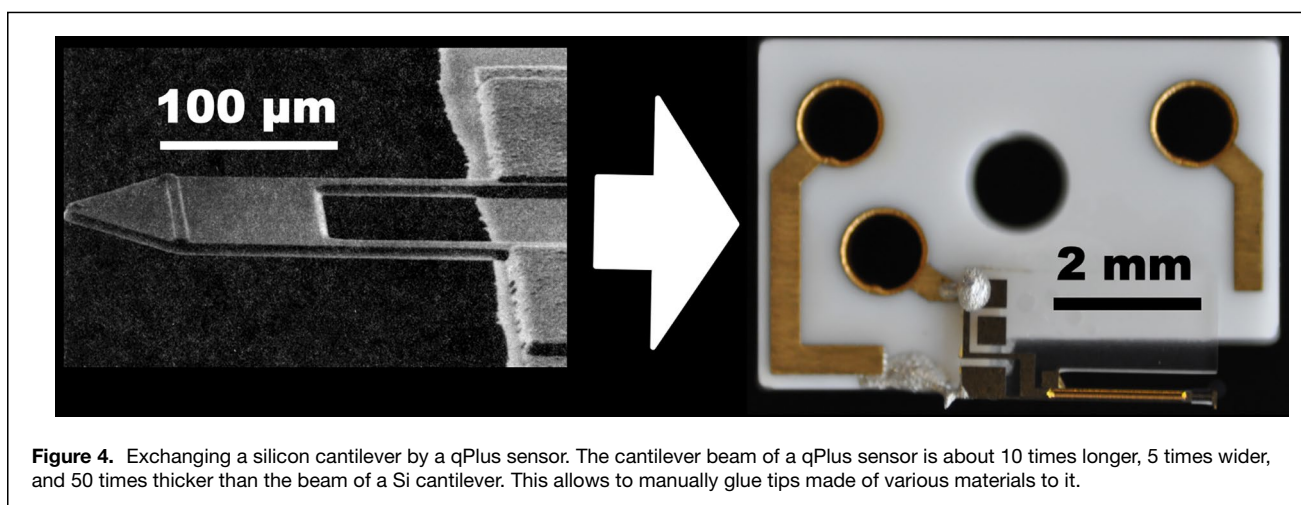
Sensor Type	Si Cantilever	qPlus Sensor
Length	200 $\mu\text{m}$	2 mm
Stiffness	20 N/m	2000 N/m
Amplitude	30 nm	30 pm
Material	Si	SiO <sub>2</sub>
Detection mechanism	Optical, piezoresistive (hot)	Piezoelectric (cold)

scientific community was divided over this claim. Some were excited and shared our enthusiasm, others were more skeptical. A technical comment<sup>23</sup> claimed that feedback oscillations could also cause such data, and it was also criticized that the Stillinger–Weber potential was not state-of-the-art, density functional (DFT) calculations must follow. Our reply ruled out the presence of feedback oscillations<sup>24</sup> and F. Liu's group performed those DFT calculations a few years later<sup>25</sup> that fully vindicated the Stillinger–Weber calculations, that is, they found the same two signature lobes that were present in the experiment and in the previous calculations.

Although this transition from soft cantilevers with large amplitudes to stiff ones with small amplitudes (see **Table I**) revealed the potential of AFM to provide subatomic spatial resolution, two weak spots remained in this study that waited for improvement:

1. It was not possible to prepare the tip state with two dangling bonds at will; the tip state was rather a random product of its preparation.
2. The known sample was imaging an unknown tip, while an ideal microscope would probe a sample with a perfectly known tip.

A very pleasant side effect was that G. Richter, one of the most influential visual artists of our time, spotted a newspaper



**Figure 4.** Exchanging a silicon cantilever by a qPlus sensor. The cantilever beam of a qPlus sensor is about 10 times longer, 5 times wider, and 50 times thicker than the beam of a Si cantilever. This allows to manually glue tips made of various materials to it.

report in *Frankfurter Allgemeine Zeitung* about our paper with an image similar to Figure 3c and used it to create an offset print *First View* from it. This led to further projects and an ongoing exchange, enriching for me in particular for seeing how an artist moves into uncharted territory and deals with uncertainty.<sup>26</sup>

Figure 4 compares a Si cantilever with a qPlus sensor.

### Expanding the use of the qPlus sensor

In the year 2000, the *soft cantilever-large amplitude* method was well established. Many groups were using this technique, commercial microscopes employing it were available, and almost all contributions to the annual International Conference on Noncontact AFM used it. Given that it takes a lot of investment in time and equipment to master the *soft cantilever-large amplitude* method, I was not surprised that initially, there was little interest from this community.\* However, three areas where the qPlus sensor could potentially have an impact appeared worthwhile to pursue. Based on my personal experience as a microscope designer and a graduate from an IBM lab, I contacted AFM manufacturers as well as the IBM Almaden and Rüsçhlikon labs to see if they would be interested in trying out the qPlus sensor.

### Improving performance, simplifying production and operation, reducing cost of commercial AFMs

When I worked as a microscope design engineer at Park Scientific Instruments in the 1990s, I very much appreciated the simplicity that the piezolever provided over a cantilever with tunneling detection. Purely electrical detection as in the qPlus sensor brings a profound simplification over optical detection measurements. Foremost, the performance enhancement, enabled by the large stiffness and consequently, the usability of sub-Angstrom amplitudes as well as the frequency stability is an important benefit of the qPlus sensor. In addition, the cost of building a qPlus AFM is much less than building an optical one and it is easier to use. Therefore, Omicron Nanotechnology, RHK Technology, CreaTec Fischer, Sigma Surface Science (later merged with Omicron), Unisoku, and Nanosurf now build qPlus microscopes, creating a wide offering of a large variety of instruments for many purposes.

### Measuring the forces that act in atomic manipulation at IBM Almaden

D. Eigler had started to build a low temperature STM around the same time we built our low temperature STM/AFM in Munich. Our machine was located on the third floor in downtown Munich, where vibrations from traffic had to be mitigated by a very small and compact microscope design without the possibility of preparing and exchanging samples according to state-of-the-art surface science. Eigler, in contrast, had

\* Over the years, this has changed. Now, the majority of contributions at the NCAFM make use of the qPlus sensor.

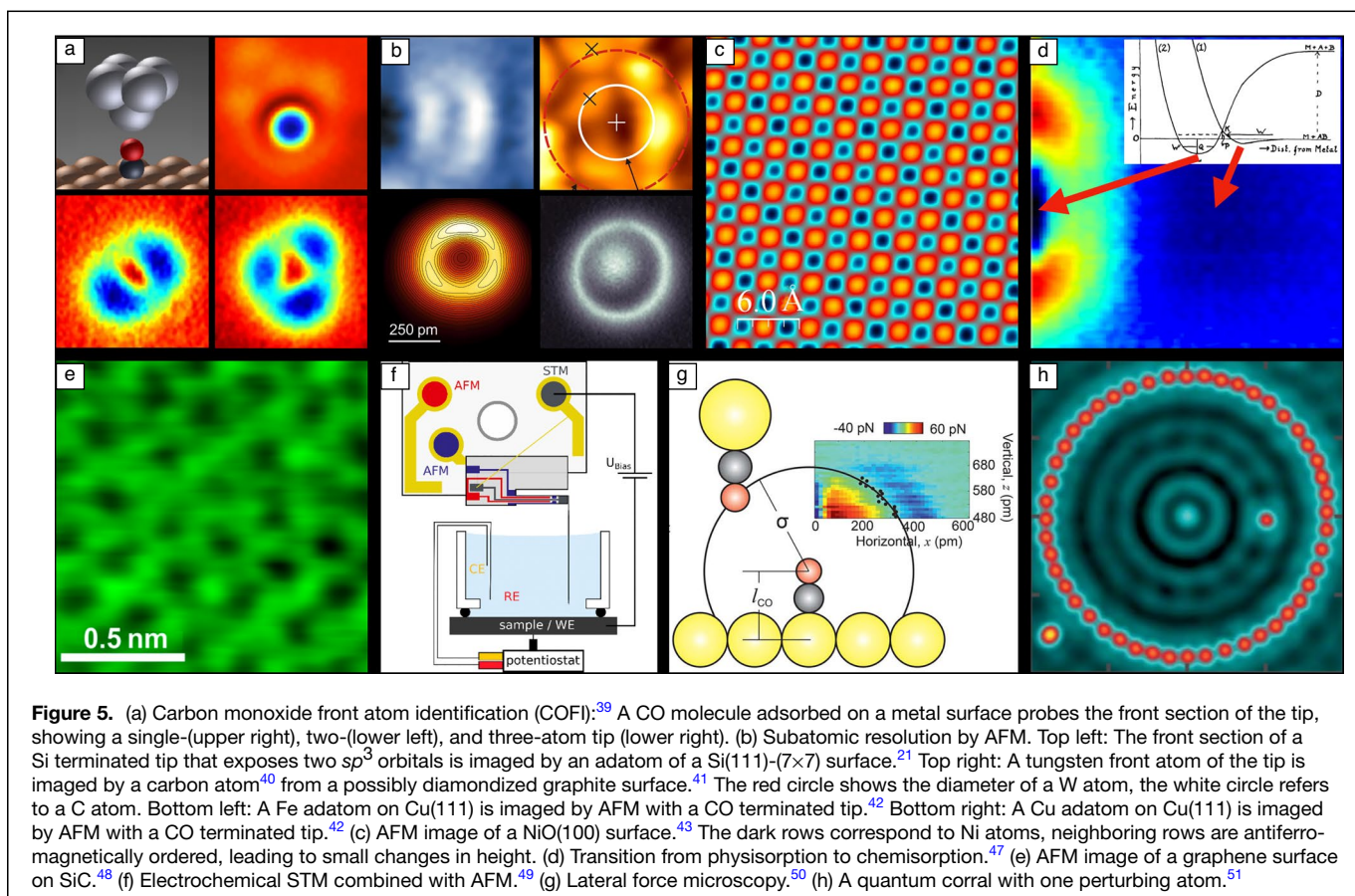
built an STM that was located in a sound-proofed laboratory in the IBM Almaden Research Center. A whole string of groundbreaking science was done there, starting with atomic manipulation<sup>27</sup> to building quantum corrals<sup>28</sup> and many other feats. A. Heinrich, who had initially been a Postdoc in Eigler's group and was later promoted to manage the group, invited me with my whole family to stay with him, his wife Michelle and their kids in their beautiful house at Lake Anderson in Morgan Hill for the summer of 2005, and to install the qPlus sensor in Eigler's original STM. Working with this extremely low-noise machine was a revelation to me. I was so excited that several times, I woke up at 3 am and, instead of tossing and turning sleeplessly in bed, put on my running shoes and jogged the 24 km under a sparkling night sky and the sounds of wild animals up to the IBM Almaden lab to continue the work on the microscope at 7 am after a large cup of coffee. After about 5 weeks, we had first data and knew it would work. In addition to A. Heinrich, M. Ternes, C. Hirjibehedin, and C.P. Lutz<sup>†</sup> continued to work on two questions at the core of atomic manipulation and the relation between STM and AFM:

1. How much force does it take to move an atom on a surface? The force depends both on the atom or molecule that it moves as well as on the substrate.<sup>29</sup>
2. How does the interaction length of chemical bonds between two metal atoms relate to the one of the tunneling current? These lengths were very similar - both about 50 pm.<sup>30</sup>

### Bringing the qPlus sensor to IBM Rüsçhlikon, the birthplace of STM

Gerhard Meyer is one of the inventors of the optical beam deflection technique that measures the force that acts on an AFM cantilever,<sup>6,7,31</sup> and he was nevertheless very interested to implement the qPlus sensor in his low temperature STM. He was a permanent research staff member at IBM Rüsçhlikon, the birthplace of STM, at that time. His team member J. Repp et al. had previously performed STM experiments of single Au atoms on NaCl and found that applying voltage pulses to them leads to a switch in their apparent height, and the calculations of the DFT collaborators in their study<sup>32</sup> found the adatoms were permanently charged by the pulses. Meyer was interested in checking the predicted charging of the Au adatom with a direct AFM Kelvin probe measurement, verifying the hypothesis.<sup>33</sup> In the same year, the experimental team with L. Gross et al. made a groundbreaking discovery: picking up a CO molecule to the tip led to a dramatic increase in

<sup>†</sup> Christopher P. Lutz holds a master's degree in computer science and taught himself the intricacies of quantum science through books, articles, and discussions. He was awarded an honorary PhD by the Physics Department of the Universität Regensburg in April 2019, based on his seminal contributions to low temperature scanning probe microscopy and the impact he left in our group in the further development of low temperature AFM.



spatial resolution of a pentacene molecule.<sup>34</sup> This discovery opened a new field of imaging molecules, graphene ribbons, metal clusters, and other samples with unprecedented resolution.

### Present and future developments

As of the end of 2022, 425 commercial AFMs based on the qPlus sensor were installed.<sup>††</sup> At the University of Regensburg alone, we have about 10 homebuilt systems so we estimate there are at least 500 qPlus based microscopes installed worldwide. Given this large number, it is impossible to cover the output of these here.

To provide an overview of applications of the qPlus sensor, we briefly list some of the projects that we pursued in our group and a selection of data from other groups.

#### Projects in our group

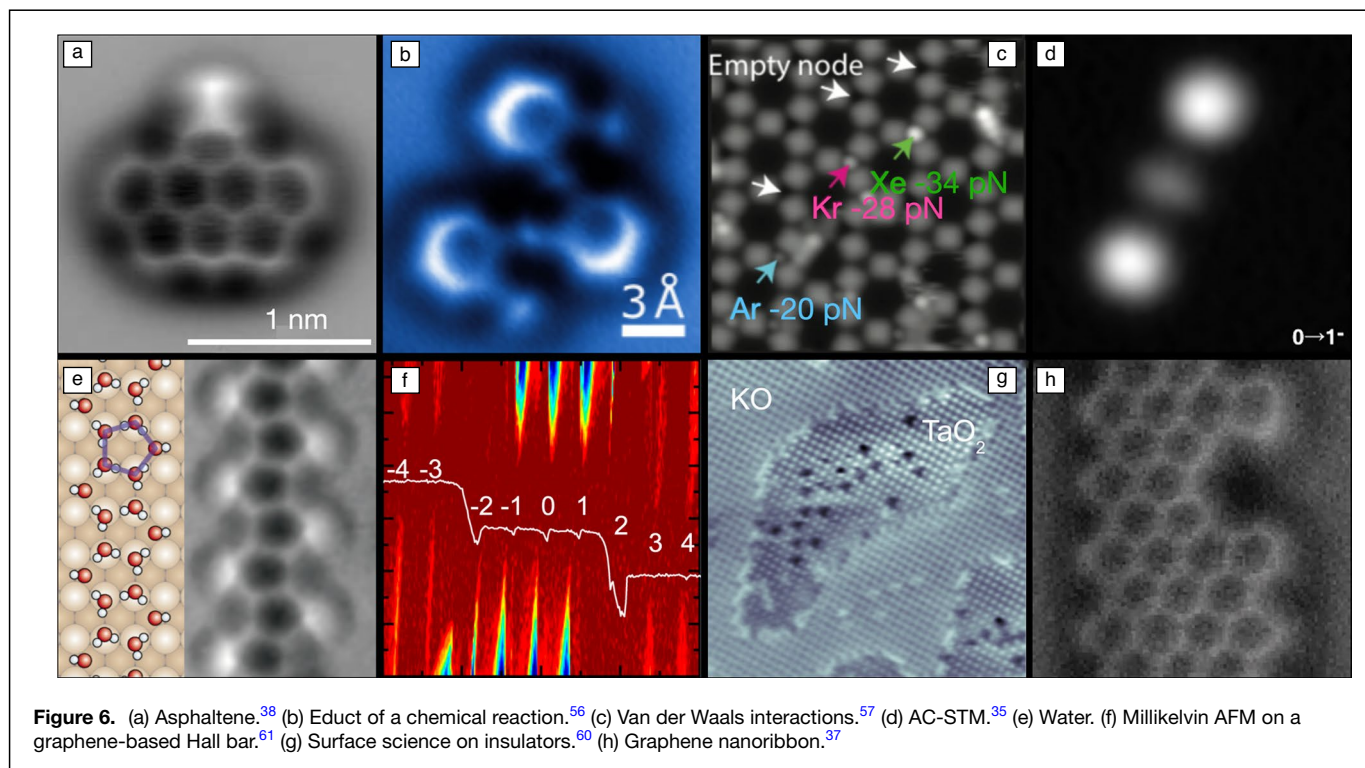
1. Tip characterization by carbon monoxide front atom identification (COFI).<sup>39</sup> The tip in AFM is perhaps more important than in STM. Therefore, a method to probe the tip apex is important. Note that our initial interpretation

of the tip images shown in **Figure 5a** was wrong: we assumed that all three tips shown here were single atom tips, while it turned out later (see Reference 42) that the tips were single-, dimer- and trimer-tips.

2. Subatomic spatial resolution. The whole idea of microscopy is to resolve the structure of a sample, and to our knowledge, AFM is the only tool that allows to see structure within an atom. While the first data in the upper row of Figure 5b show subatomically resolved images of a single atom Si tip<sup>21</sup> and a W tip,<sup>40</sup> the bottom row demonstrates subatomic resolution of a Fe and a Cu adatom on Cu(111) observed with a CO terminated tip.<sup>42</sup>
3. Spin resolution of an antiferromagnetic NiO(100) surface<sup>43</sup> is shown in Figure 5c.
4. Atomic manipulation was studied in the collaboration with A. Heinrich<sup>29</sup> and is still an active field of interest in our group.<sup>44,45</sup>
5. In 1932, Lennard–Jones proposed the existence of two different adsorption states of an atom or molecule on a surface: weak physisorption due to van der Waals forces and stronger chemisorption due to covalent bonds<sup>46</sup> as shown in the inset of Figure 5d. Ninety years later, Huber et al.<sup>47</sup> directly observed this transition from physisorption to chemisorption with an AFM when studying the force evolution of a CO tip versus a Fe adatom on Cu(111) (see Figure 5d).

†† Source: Cretec Fischer GmbH (D), Nanosurf AG (CH), RHK Technology (USA), ScientaOmicron (D), Sigma Surface Science (D) - merged with ScientaOmicron, Unisoku Ltd. (JP).





**Figure 6.** (a) Asphaltene.<sup>38</sup> (b) Educt of a chemical reaction.<sup>56</sup> (c) Van der Waals interactions.<sup>57</sup> (d) AC-STM.<sup>35</sup> (e) Water. (f) Millikelvin AFM on a graphene-based Hall bar.<sup>61</sup> (g) Surface science on insulators.<sup>60</sup> (h) Graphene nanoribbon.<sup>37</sup>

6. Atomic resolution in ambient conditions is facilitated with qPlus sensors. Figure 5e shows an image of graphene on SiC.<sup>48</sup>
7. Andrea Auer has obtained her PhD degree at the University of Innsbruck in electrochemical STM. She joined our group as a Humboldt Fellow and implemented simultaneous electrochemical STM and AFM as shown in Figure 5f.<sup>49</sup>
8. Lateral force microscopy is a focus of Jay Weymouth, permanent staff member in our group. Figure 5g shows a key experiment<sup>50</sup> where the lateral interaction of two CO molecules was studied.
9. The quantum corral introduced in 1993 by Crommie, Lutz, and Eigler<sup>28</sup> can be viewed as an artificial atom. We studied the very weak bonds of the eigenstates of the corral (down to the 10 fN scale) with the AFM tip<sup>51</sup> and found that placing atoms inside the corral affects the occupation of the various angular momentum states as shown in Figure 5h.
10. AFM provides interesting images of the surface of topological insulators. Pielmeier et al.<sup>52</sup> showed that AFM reveals that the TlBiSe<sub>2</sub> surface cleaves on the Tl planes, leaving about 50% of Tl on either cleavage plane. Liebig et al.<sup>53</sup> characterized defects in the topological insulator Bi<sub>2</sub>Se<sub>3</sub> at the picometer scale.
11. A combination of inelastic electron tunneling spectroscopy and AFM allows to directly measure the impact of an external force field of the tip to the bonding forces and the oscillation frequencies of an adsorbed species.<sup>54</sup>
12. Finally, we note a mathematical detail regarding the derivation of forces from the frequency shift data. While the derivation of the frequency shift from the force versus distance curve is straightforward as given by equation 2, the inverse is not true. This deconvolution of forces from frequency shift spectra is a mathematical inverse problem that carries its own challenges, and a criterion as to when this inversion is correct has been found in collaboration with J. Sader.<sup>55</sup>

### Selection of projects of other groups

1. The introduction of the CO terminated tips by Gross et al.<sup>34</sup> has opened a new horizon in the atomic study of molecules. Figure 6a shows an AFM image of asphaltene, a component found in crude oil.<sup>38</sup>
2. Figure 6b depicts an educt of a chemical reaction on a surface. The products, induced by thermal annealing, could also be identified by AFM.<sup>56</sup>
3. Van der Waals interactions are weak, and the vdW interaction between individual noble gas atoms is difficult to measure. S. Kawai et al.<sup>57</sup> have succeeded in this measurement by placing Ar, Kr, and Xe in a molecular matrix and measuring its interaction with a Xe terminated tip. The maximal attractive forces between the Xe tip and Ar, Kr, and Xe atoms are shown in Figure 6c.
4. L. Patera et al.<sup>35</sup> have found a method they called AC-STM that measures the damping of a qPlus sensor to identify an electron transfer to molecular states that can be far from the Fermi level. Figure 6d shows such a transition.

5. The structure of water on surfaces can be studied by AFM as well.<sup>58,59</sup>
6. The group of J. Stroscio at NIST in Gaithersburg, Md., has built a combined STM/AFM that works at millikelvin temperatures and high magnetic fields. Figure 6f shows damping data on a graphene quantum Hall device.<sup>61</sup>
7. The group of U. Diebold at TU Vienna has done surface science on insulators for decades. AFM is now a very important tool for this group. Figure 6g shows an oxide surface as imaged by AFM.<sup>60</sup>
8. Figure 6h shows a defect in a graphene nanoribbon;<sup>37</sup> AFM became an important tool here for the study of graphene devices.

Finally, we note that the qPlus sensor provides a versatile platform for many new experiments. L. Sellies et al. have recently shown that measuring electron spin resonance with AFM leads to much longer spin coherence times compared to STM.<sup>36</sup> Going back to the starting statement “Some of the best innovations come from combining two previous innovations,” we also note the potential of combining AFM with ultrafast optics (see, e.g., Reference 62), a road we are pursuing in the newly established Regensburg Center for Ultrafast Nanoscopy (RUN).

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## Data availability

Included in cited original research papers.

## Code availability

Included in cited original research papers.

## Conflict of interest

The author owns patents as well as a registered trademark to the qPlus sensor.

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