Low-temperature scanning probe microscopy of surface and subsurface charges

Markus Vogel, Bernhard Stein, Håkan Pettersson, and Khaled Karrai[a)]
Center for NanoScience, Sektion Physik der Ludwig-Maximilians, Universitat München, Geschwister-Scholl-Platz 1, 80539, München, Germany

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The operation of a cryogenic scanning force microscope is demonstrated with a sensitivity of about $50\,\mathrm{fN}/\sqrt{\mathrm{Hz}}$ at 5 kHz modulation. This microscope is used as an electrometer in noncontact mode in order to map the local electrostatic forces and capacitance of several nanostructures at 4.2 K. Capacitance imaging of nanostructured surfaces with subatto-Farad resolution is demonstrated.
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Thanks to the extraordinary advances in scanning probe microscopy,[1] it is now possible to map single electronic surface charges with a spatial resolution of the order of 100 nm.[2–4] Recently, such techniques were extended to the detection of subsurface charge accumulation deeper inside a semiconductor heterostructure.[5] Room-temperature scanning probe electrometers are based on atomic-force microscopy techniques using differential interferometery[6] monitoring lever deflection. The complex optical arrangement and its fine tuning makes this microscope unsuitable for operation in an enclosed environment such as in a cryostat or a vacuum chamber where remote adjustment of the interferometer alignment would be required. Alternatively, a very sensitive electrometer based on scanning a single electron transistor over a sample surface was demonstrated with an unprecedented combination of spatial and charge resolution.[3] So far, this microscope remains unique since its operation requires a very critical preparation of the probe itself and necessitates liquid-helium temperatures for the operation of the single electron transistor probe. Tessmer *et al.*[5] developed a related version of a cryogenic charge-sensitive scanning probe microscope. There the probe is a metallic tip connected to the gate of a high electron mobility transistor (HEMT) with extremely high sensitivity to charges. Although the principle of its operation is very simple and elegant, its performance depends critically on the operating temperature, the proper contacting of the tip onto the HEMT's gate, and its shielding from spurious electrical field sources such as sample contacts. In order to remedy some of these shortcomings, we report in this letter on the development of a simple and robust optical-fiber-based scanning electrostatic and capacitance microscope with a high sensitivity to electrical charges (0.01 electrons/$\sqrt{(\mathrm{Hz})}$ at the probe apex) for operation ranging from liquid-helium temperatures to ambient conditions.

The scanning probe force detection scheme in our microscope design, which is shown in Fig. 1(a), is based on detecting the deflection of a lever-holding tip using a miniature-fiber-based Fabry–Pérot interferometer.[7–9] Its cavity is formed by the metallized back of a commercial Si cantilever and the polished gold-coated end of a monomode optical fiber. The cavity length of the interferometer is typically of the order of 30 μm. The particularity of our design is that the cavity length is electrically tunable. The 20 nm semitransparent gold coating on the flat end of the fiber has two functions: that of increasing the finesse of the Fabry–Pérot miniature cavity, but also and more importantly, to insure the electrical fine tuning of its length at all temperatures. The cavity length tuning is based on the fact that the Si soft lever [spring constant $K=0.04$ N/m (Ref. 10)] can be pulled electrostatically toward the gold-coated fiber end when a voltage U_F is applied between both mirror ends. As seen in Fig. 1(b), a tuning of the order of several wavelengths ($\lambda=670$ nm) can be achieved with voltages U_F ranging from 0 to 40 V. Stiffer cantilevers with K up to 0.5 N/m were successfully used but with a reduced range of cavity length adjustment. A laser beam is launched into the other end of the optical fiber. The signal reflected back from this miniature Fabry–Pérot cavity is detected outside the cryostat. The coherence length of our laser diode is short so that no spurious interference within the cavity formed in the 2-m-long monomode optical fiber will affect the lever signal. The power of the laser is adjusted so that less than about 0.5 μW of heat is absorbed at low temperatures. Maximum sensitivity to lever deflection is obtained by setting U_F on one of the maximum slope of interference fringes such as seen in Fig. 1(b). Since the diameter of the optical fiber and the lever dimensions are in the 0.1 mm range, the whole interferometer is made very compact. Its sub-mm size and *in situ* electrical tunability makes it ideal for operation in cryostats, high magnetic fields, or high vacuum chambers. This compact interferometer is found particularly stable since we observed no measurable drift from its optimal working point during the typical time scale of an experiment (i.e., hours). Pelekhov, Becker, and Nunes[9] re-

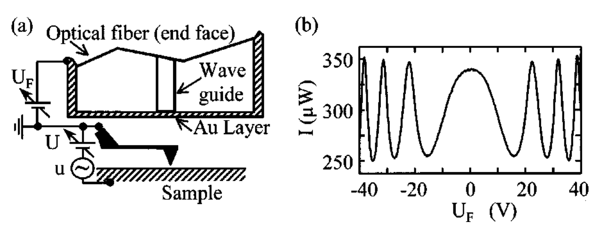

FIG. 1. (a) Schematic of the lever deflection detection. (b) Reflected photosignal as the microcavity is electrostatically tuned.

[a)]Electronic mail: khaled.karrai@physik.uni-muenchen.de

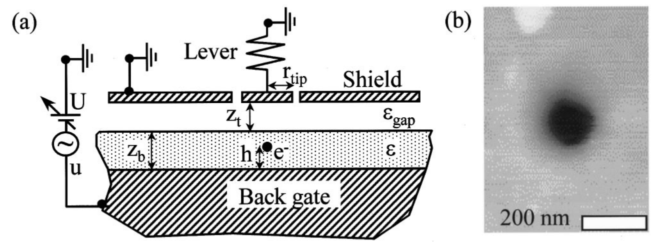

FIG. 2. (a) Schematics of the flat tip electrostatic model with the electrical connections. (b) Aperture topography measured in contact mode at 4.2 K.

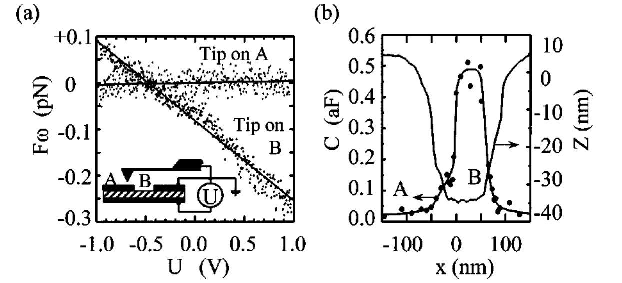

FIG. 3. (a) Force F_ω experienced by the tip over the metal screen (position A) and over the 150 nm aperture (position B). Inset: electrical schematics. (b) Corresponding tip–sample capacitance and topography across the aperture. The measurements were performed at 4.2 K.

ported on a cryogenic interferometer scanning contact force microscope operating on a similar principle, the tuning of their cavity length is achieved with a long piezoelectrical ceramic in order to achieve sufficient length of cavity tuning. Such a long mechanical length between the two mirror ends of the cavity leads to difficult control of the total optical length after cooling and increased sensitivity to mechanical noise. In order to operate such a scanning probe microscope as an electrostatic force microscope, a voltage U is dropped across a conducting tip and a back contact behind the sample. The commercial Si tip/lever is boron doped at $10^{20}\,\mathrm{cm}^{-3}$ so that it remains metallic at all temperatures. Nevertheless, we coated the whole lever with a 10-nm-thin platinum film in order to ensure the shortest possible depletion layer thickness at the tip apex. In order to screen the sample from the fields originating from the metal-coated fiber end at a potential U_F, the tip must be maintained at ground potential. A small modulation $u=\sqrt{2}u_{\mathrm{rms}}\sin(\omega t)$ is added to U in order to separate the local forces acting on the tip from nonelectrostatic background forces. The electrostatic force acting on the tip[11,12] is given by $F=(1/2)\times(\partial C/\partial z)(U+\phi+u)^2$. where C is the probe to back contact capacitance, ϕ is a sample potential that includes the tip–sample contact potential ϕ_0 and the surface or subsurface localized charges, and z is the tip–sample distance. Such an expression can be decomposed in the sum $F=F_{\mathrm{dc}}+F_\omega+F_{2\omega}$, where $F_\omega=\sqrt{2}(\partial C/\partial z)(U+\phi)u_{\mathrm{rms}}\sin(\omega t)$, and $F_{2\omega}=\frac{1}{2}(\partial C/\partial z)u_{\mathrm{rms}}^2\sin(2\omega t-\pi/2)$. Hence, ϕ and $\partial C/\partial z$ can be obtained from a measurement of both the ω and 2ω forces. In order to obtain the capacitance C, one would need to compute it from the measured z dependence of $F_{2\omega}(z)$. We will assume instead that $\partial C/\partial z$ has the functional form obtained from a parallel-plate capacitor formed by a flat tip and a sample. This rough approximation is, in principle, reasonable as long as the tip–sample distance remains much smaller than the tip radius.

We tested the microscope on a nanostructure of known geometry. A circular nanoaperture 150 nm in diameter is defined in a 50-nm-thick gold film deposited onto a GaAs modulation-doped structure. The GaAs sample is made of a 200 nm insulating thin film grown on a highly *n*-doped GaAs back contact. The gold layer and the tip are placed at the same potential while the GaAs back contact is biased at U. This sample geometry with a flat tip is sketched in Fig. 2(a). The electrostatic force measurements were performed in a vibration-isolated liquid-helium bath magnet cryostat operating at 4.2 K. The microscope which is located in a vacuum-tight thin-walled stainless steel tube insert is plunged in the He bath and cooled by 10 mbar of He exchange gas. The tip is first brought into soft contact (~1 nN) with the sample so that a topographical image of the nanoaperture in the gold metal can be obtained. Such a topographical image is shown in Fig. 2(b). The tip is then pulled away at about 30 nm above the surface at a safe distance away from jump to contact instabilities. The Fabry–Pérot interferometer is here very useful since its signal gives the absolute tip–sample surface distance z_t. In this configuration, the oscillating F_ω and $F_{2\omega}$ forces are obtained from the interferometrically measured lever deflection assuming a spring constant of $K=0.04\,\mathrm{N/m}$. In Fig. 3(a) we show the magnitude of F_ω for the tip positioned inside the hole aperture and outside of it as a function of the back-gate voltage U. Clearly, when the tip is over the gold layer away from the nanoaperture, the modulated voltage u is screened and the resulting oscillating forces are vanishing. In contrast, when the tip is positioned over the hole aperture, it experiences a modulated force F_ω measured to be proportional to U in agreement with the expression given above. For this measurement we have set $u_{\mathrm{rms}}=20\,\mathrm{mV}$, $\omega=2\pi\times5$ kHz, and a lock-in integration time constant of 1 s. During that integration time, the typical rms z noise in the lever–fiber distance was $0.5\times10^{-12}\,\mathrm{m}$, corresponding to $20\times10^{-15}\,\mathrm{N}$. The situation for which F_ω vanishes is reached when the applied potential U cancels the contact potential ϕ_0 at $U\approx-0.45\,\mathrm{V}$, as seen in Fig. 3(a). In order to quantify the sensitivity of this electrometer, it is useful to relate the measured force to the fraction of electron charge driven in and out of the tip. We assume for now that the tip can be approximated by a small plate parallel to the back contact positioned at z_b below the sample surface, as shown in Fig. 2(a). Within such an idealized electrostatic model, the force experienced by the lever is given by $F_\omega=(1/\epsilon_{\mathrm{gap}})C_0(U+\phi_0)u/(z_t+z_b)[\alpha/\epsilon+(1-\alpha)/\epsilon_{\mathrm{gap}}]^{-2}$, where C_0 is the equivalent tip-to-back-contact vacuum capacitance and $\alpha=z_b/(z_t+z_b)$ is the fraction of the tip-to-back-contact volume filled with the sample material. In our measurements $z_b=200\,\mathrm{nm}$, $\epsilon\approx13$, $\epsilon_{\mathrm{gap}}=1$, and the tip–sample z_t distance is typically set such as $\alpha=0.9$. Within this model, the slope $\partial F_\omega/\partial U=C\alpha^2(u/z_b)/(\epsilon\epsilon_{\mathrm{gap}})[\alpha/\epsilon+(1-\alpha)/\epsilon_{\mathrm{gap}}]^{-2}$ is proportional to $C=C_0\epsilon/\alpha$, the capacitance between the back gate and the surface (i.e., the local sample capacitance). Similarly, the tip-to-sample surface capacitance is given by $C_t=C_0\epsilon_{\mathrm{gap}}/(1-\alpha)$ and $\partial F_\omega/\partial U=C_t\alpha(1-\alpha)(u/z_b)\times(1/\epsilon_{\mathrm{gap}}^2)[\alpha/\epsilon+(1-\alpha)/\epsilon_{\mathrm{gap}}]^{-2}$. A profile of the local sample capacitance C is obtained by measuring the slope $\partial F_\omega/\partial U$ spectra along the tip position across the hole aperture. The capacitance profile of C is plotted in Fig. 3(b). Given a conservative estimate for the noise floor of $\delta F_\omega \approx 50\times 10^{-15}\,\mathrm{N}/\sqrt{\mathrm{Hz}}$, we conclude that the Fabry–Pérot measurement of the lever deflection is sensitive to a *modulated charge induced in the tip apex* of $\delta Q = \delta C_t u/[1+\alpha/(1-\alpha)(\epsilon_{\mathrm{gap}}/\epsilon)] \approx 0.012$ electrons/$\sqrt{\mathrm{Hz}}$. This figure is comparable to the HEMT-based scanning electrometer of Tessmer *et al.*[5] The main motivation behind this work is to reach a force sensitivity to single localized subsurface electrons when charging is induced near the back gate at a distance h above it. In this case, and in the limit of $h \ll z_b$ and $z_t > 0$, which apply to subsurface localized charges, the parallel-plate capacitor model of Fig. 2 shows that the tip experiences an excess force due to a single charge of $F_{(1e)} = F^0_{(1e)}(f_- - f_+)$, where $F^0_{(1e)} = -eE/2 = -(1/2)e(U+\phi_0+u)/(z_t+z_b)$ is half the force experienced by a single electron placed in a homogeneous electrical field E. Both dimensionless factors f_- and f_+ are due, respectively, to the single electron charge and its image in the back gate acting on a disk-shaped tip of radius r_{tip}, and include the dielectric constants as well as the sample and tip geometrical information. They are given by:

$$f_\pm = 2[\alpha/\epsilon + (1-\alpha)/\epsilon_{\mathrm{gap}}]^{-1}[1+\epsilon/\epsilon_{\mathrm{gap}}]^{-1} \times \{1-[r^2_{\mathrm{tip}}/(z_t+z_b\pm h)^2+1]^{-1/2}\}.$$

The term $(f_- - f_+)$ is positive and always smaller than unity. This shows that while $F^0_{(1e)} \approx 0.4\,\mathrm{pN}$ for $U+\phi_0+u \approx 1\,\mathrm{V}$, the correction factor $(f_- - f_+)$ is for our sample and tip geometry of the order only of 0.015. In estimating this geometrical correction factor we have assumed $h = 25\,\mathrm{nm}$, $z_t = 15\,\mathrm{nm}$, $z_b = 200\,\mathrm{nm}$, and $r_{\mathrm{tip}} = 50\,\mathrm{nm}$. A measurement of the ω component of $F_{(1e)}$ with $u \approx 10\,\mathrm{mV}$ would then lead to $F_{(1e)\omega} \approx 0.06\times 10^{-15}\,\mathrm{N}$, which is well bellow the noise level. The detection of such a deep single subsurface charge would be, in this case, hopeless without the possibility of modulating the charge itself. This can be done in charge-tunable semiconductor heterostructures[13] via tunneling of the electron from the back gate into a quantum well located at distance h away. As an example for a realistic GaAs/InAs heterostructure, we have estimated that $F_{(1e)} \approx 0.238\,\mathrm{pN}$ can be induced when tunneling occurs at 1 V potential for $z_b = 50\,\mathrm{nm}$, $h = 25\,\mathrm{nm}$, $z_t = 10\,\mathrm{nm}$, $\epsilon_{\mathrm{gap}} = 1$, and $\epsilon = 13$. Such a force is typically an order of magnitude larger than that of the noise level.

In order to illustrate the operation of the scanning electrometer, we prepared and imaged a sample made of a $z_b = 25$-nm-thick Al_2O_3 film lithographically patterned in 1-μm-wide stripes placed on a gold back electrode. Figure 4(a) shows the sample surface topography. We measured, simultaneously, the F_ω and $F_{2\omega}$ electrostatic forces induced by the potential $u_{\mathrm{rms}} = 0.2\,\mathrm{V}$, which was superimposed to the dc potential $U = +2.5\,\mathrm{V}$ applied on the gold back electrode. The tip-to-back-gate distance $z_b + z_t$ was kept at 42 nm. Given the above parameters, we estimated that the force due to a localized charge on the surface of the Al_2O_3 stripes was about 0.06 pN per electron. All measurements were performed at 77 K using two lock-ins integrating the signals with a time constant of 30 ms. The $F_{1\omega}$ image plotted in Fig. 4(b) shows structures that correlate neither to the capacitance

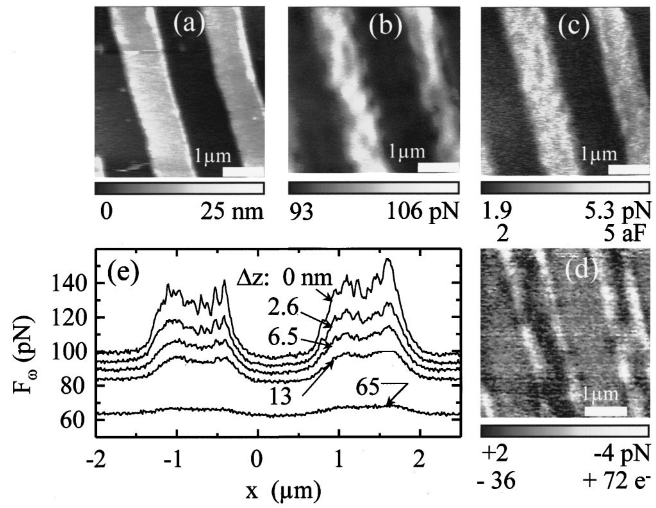

FIG. 4. (a) Topography of the Al_2O_3 stripes (bright) on a gold back contact (dark). (b) and (c) Corresponding F_ω and $F_{2\omega}$ forces. The resulting local charge image is shown in (d). The line scans of F_ω are shown in (e) for decreasing tip-to-gold-contact distances ($\Delta z = 0$ corresponds to $z_b + z_t = 42\,\mathrm{nm}$). The noise-like features in (e) over the Al_2O_3 stripes are fully reproducible.

[Fig. 4(c)] nor to the topography image [Fig. 4(a)]. Figure 4(d) shows the components of the $F_{1\omega}$ image that do not correlate to the $F_{2\omega}$ image, revealing in this way a map of localized charges. The number of localized charges indicated in Fig. 4(d) is estimated by dividing this force image by 0.06 pN, the elementary force due to a single charge estimated above. The noise level in the present measurement for this sample structure limits our detection to about ten electrons. In Fig. 4(e), several $F_{1\omega}$ line scans are plotted for different tip height. The noise-like features are fully reproducible and correspond to the local charges on the Al_2O_3 stripes.

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