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Near-field thermal imaging of nanostructured surfaces

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We show that a near-field scanning thermal microscope, which essentially detects the local density of states of the thermally excited electromagnetic modes at nanometer distances from some material, can be employed for nanoscale imaging of structures on that material's surface. This finding is explained theoretically by an approach which treats the surface structure perturbatively. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3025140]

It is well known that a dielectric body kept at constant temperature is surrounded by the fluctuating thermal electromagnetic field generated by fluctuations inside the material. These fluctuating fields can be divided into propagating and evanescent modes, i.e., radiating and nonradiating modes. The familiar radiating modes can be described by means of the Kirchhoff–Planck law of thermal radiation. Furthermore, the evanescent modes can also contribute to the radiative heat transfer due to photon tunneling, if a second dielectric body with a different temperature is brought close to the first one. This additional contribution can exceed the radiative heat flux between two blackbodies by several orders of magnitudes if the distance between them is much smaller than the dominant thermal wavelength.

A theoretical description within the framework of the fluctuational electrodynamics of Rytov *et al.*[1] of the radiative heat transfer between two closely spaced semi-infinite bodies with parallel surfaces was given by Polder and van Hove.[2] Early experimental observations of the additional contribution of the evanescent modes in a geometry of two parallel plates were made by Hargreaves[3] and by Domoto *et al.*[4] In a quite recent experiment by Hu *et al.* with precision glass optical flats,[5] good agreement with the prediction of Polder and van Hove has been found for a distance of 1.6 μm. It is rather difficult to reach shorter distances in such experiments with plates, because these plates have to be kept perfectly parallel. Moreover, the achievable minimum distances are limited by the surface roughness. These restrictions are overcome by a near-field scanning thermal microscope (NSThM).[6,7] The scanning tunneling microscope (STM) tip of such an instrument also serves as a thermocouple, so that the radiative heat transfer between a cooled sample and this probe can be measured directly for distances ranging from 1 nm up to the resolution limit of the apparatus, which currently lies in the range of some 10 nm.[8] Hence, the NSThM acts in a regime of probe-sample distances in which simple local macroscopic models for the radiative near-field heat transfer[9–13] based on the theory of Rytov *et al.* may become questionable.[14,15] In this letter, we demonstrate that a NSThM provides images of a nanoscale structured surface. Despite the above objection, the measured signals can at least qualitatively be understood within a simple local approach, describing the sensor tip by means of a dipole model and the sample's surface by perturbation theory.[16]

The experiments were conducted with a NSThM developed[6,7] and refined[8] in our group. It is based on a commercial variable temperature ultrahigh vacuum STM. In order to measure the near-field contribution to the heat flux, the STM tip has been functionalized to act as a microscopic thermometer. This has been achieved by implementing a gold-platinum thermocouple in coaxial configuration at the very end of the tip. The fabrication of these thermocouple tips is described in detail in Ref. 8. At the foremost end, the tip consists of a platinum wire with a tip radius of less than 50 nm, protruding about 500 nm from a glass capillary and covered by a thin gold film (few nanometers thick), as sketched in Fig. 1(a). A micrograph taken with a scanning electron microscope (SEM) is shown in Fig. 1(b).

During a measurement the sample is cooled down to about 110 K, whereas the tip holder remains at ambient temperature (about 293 K). Due to near-field heat transfer, the very end of the tip becomes slightly colder than its back side. This small temperature difference is the source of the detected thermovoltage V_{th}. Knowing both the tip's thermal resistance and the Seebeck coefficient of the thermocouple, even an absolute measurement of the near-field heat current between probe and sample is possible.[8] Here we merely consider the thermovoltage itself, which is proportional to the heat flux.

Figure 2(a) displays a STM micrograph of a gold surface fabricated on mica, which exhibits some structure. During the scan performed at constant distance (constant-current mode: I_t=2 nA, V_t=0.5 V), the thermovoltage V_{th} has been detected at each point. The resulting distribution of the thermovoltage is shown in Fig. 2(b). Obviously, the heat flux across the vacuum gap between probe and sample reflects the topography, even though the probe-sample distance is kept constant. However, it does not give quite the same information as the topographic image; in particular, it tends to em-

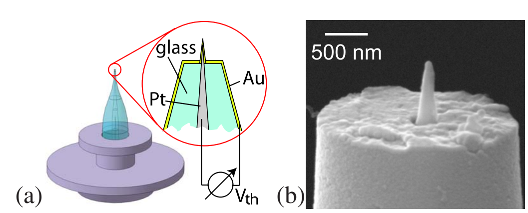

FIG. 1. (Color online) (a) Schematic drawing of the tip in its holder, with blown-up cross section of the tip's very end, displaying the glass capillary, the platinum wire, and the gold coating which form the thermocouple. (b) SEM image of a typical NSThM tip. The platinum wire protrudes from the center of the glass mantle. Both are covered by a gold film.

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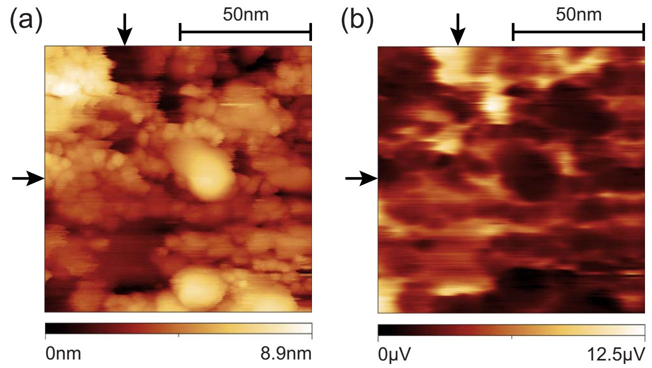

FIG. 2. (Color online) (a) STM topography of a gold surface on mica. (b) Spatial distribution of the local thermovoltage measured by the thermocouple. The temperature of the sample is 110 K, that of the probe 293 K.

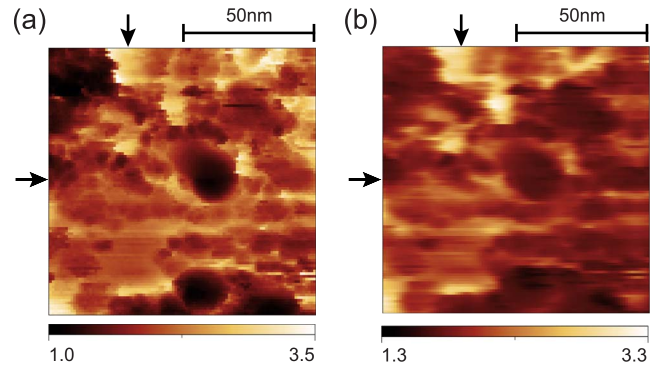

FIG. 3. (Color online) (a) Numerically calculated LDOS in 10^6 m^{-3} s at a constant distance of 9 nm above the two-dimensional topography directly extracted from the STM data in Fig. 2. (b) A plot of the thermovoltage data (in arbitrary units) from Fig. 2 rescaled as described in the text.

phasize the structure's edges. This is in itself an important finding: The NSThM provides nanoscale thermal surface images.

In what follows we explain this finding by comparing the measured signals with results obtained from a theoretical model describing the probe's tip as a metallic sphere, and considering its electric and magnetic dipole moments. Such a model leads to a general expression for the radiative heat transfer between the sensor tip and an arbitrary body.[17,18] The geometry and material properties of that body are incorporated via its electric and magnetic local density of states[19] (LDOS) evaluated at the point of measurement, i.e., at the point inside the tip where the dipole moments are located. The LDOS above a surface with a given structure has recently been calculated by means of a perturbative approach up to first order in the surface profile.[16] It has been shown that the signal detected by a spherical probe when scanning a surface with some topographical structure coincides reasonably well with the sample's LDOS, evaluated at the dominant thermal frequency. For metals such as gold, the magnetic part of the LDOS dominates in the distance regime of interest.[18] Therefore, in order to qualitatively compare the experimental data with the theory, it suffices to calculate the magnetic LDOS at the dominant thermal frequency. While a full-fledged theoretical treatment would require significantly more effort, this coarse approach can be expected to capture the significant features.

As detailed in Ref. 16, this description is subject to several restrictions. First, the dipole approximation is strictly valid only for probe-sample distances much greater than the tip radius (about 50 nm), whereas for smaller distances higher moments have to be considered. Second, the perturbative approach provides a viable approximation for observation distances much greater than the maximum height variation of the surface profile (about 5 nm). Third, at distances much smaller than the mean free path (about 20 nm) of the conduction electrons, nonlocal effects have to be taken into account. All three issues are relevant for our experiment. Nonetheless, we show that the signals obtained with the NSThM can at least qualitatively be understood on the basis of our simple approach, thus clarifying the physical principles underlying near-field thermal imaging.

We calculated the zeroth- and first-order magnetic LDOS in a constant distance of 9 nm above the surface of the sample via a discrete Fourier transform of the sample profile directly taken from the STM data shown in Fig. 2 without any smoothing. According to Ref. 16, the LDOS has to be evaluated at the dominant thermal frequency of the tip $\omega_{th} = 10^{14}$ rad s^{-1}, as corresponding to 293 K. The result of the calculation is shown in Fig. 3(a). In order to account for the proportionality between thermovoltage and heat flux on the one hand, and for the approximate proportionality between LDOS and heat flux on the other, we fitted the thermovoltage data to the LDOS performing a least-square fit. Scaling and shifting the thermovoltage data in Fig. 3(b) accordingly, the overall agreement of their main features with

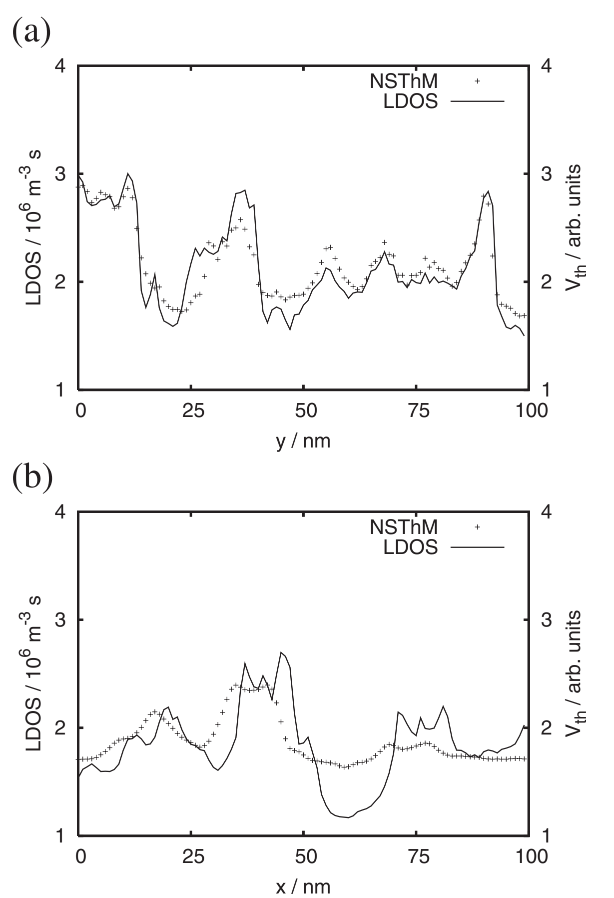

FIG. 4. (a) LDOS at a constant distance of 9 nm above the surface profile in comparision to the thermovoltage data, for the line scan indicated by the vertical arrow in Fig. 3. (b) LDOS at a constant distance of 9 nm above the profile in comparision to the thermovoltage data, for the line scan indicated by the horizontal arrow in Fig. 3.

the LDOS becomes obvious. We have to emphasize that it is not clear *a priori* which distance should be used for the calculation of the LDOS, since the radius of the foremost part of the probe's interaction region is unknown. Our choice of 9 nm appears plausible, and yields good agreement between the LDOS and the NSThM signal.

There is no strict one-to-one coincidence between the NSThM data and the LDOS. In view of the many simplifications underlying the theoretical description, some deviations have to be expected. However, the model actually does reproduce the main experimental trend: The NSThM-signal obtained in the constant-current mode roughly corresponds to the inverted profile function. In Fig. 4 we show two line scans taken from the scaled thermovoltage data, and from the theoretically obtained LDOS in Fig. 3, for the horizontal and vertical line indicated by the arrows. In both cases a good qualitative agreement between the experimental and the theoretical data can be observed. However, for the horizontal line scan the LDOS and the thermovoltage data appear to be shifted against each other in the x-direction, suggesting that the centers of interaction for the STM and the NSThM are shifted in that direction. This finding might be explained by the shape of the foremost part of the tip, which is not perfectly axially symmetric.

To conclude, we have demonstrated that a NSThM operated at a constant distance provides a thermal near-field image exhibiting nanoscale features of isothermal, structured surfaces. The measured signal qualitatively follows the local density of states of the thermal electromagnetic field above the surface, obtained by first-order perturbation theory in the profile function and evaluated at the dominant thermal frequency, neglecting nonlocal effects. More detailed, quantitative comparisons with the predictions of a refined model are desirable for further exploring the possibilities opened up by near-field thermal imaging.

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