Atomic Resolution in Photon Emission Induced by a Scanning Tunneling Microscope

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A low-temperature ultrahigh-vacuum scanning tunneling microscope (STM) is used to excite photon emission from Au(110) surfaces. In the detected photon intensity the (1 × 2) reconstruction of the Au surface is clearly resolved. This first observation of atomic resolution in STM-induced photon emission is interpreted in terms of local variations of the electromagnetic interaction of tip and sample occurring at constant tunneling current. Similar effects are expected to affect other scanning probe methods, in particular those involving photons.

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Since the work of Abbe, the resolution of optical microscopy with light of wavelength λ is known to be limited to $\approx\lambda/2$. Higher resolution may be obtained by using radiation of shorter wavelength, e.g., x rays or electrons. Alternatively, various scanning probe methods (SPM)—a well-known example is STM—aim at superior resolution by probing the near-field properties of a sample. An important motivation to develop SPM involving visible or infrared light is that many excitations of solids and molecules fall into this energy range. This may permit investigations of these excitations with high lateral resolution or, in turn, the identification of, e.g., single molecules on surfaces by their excitations. One probe to the electromagnetic near field is scanning near-field optical microscopy (SNOM) where resolution of up to several tens of nm has been achieved with visible light [1]. Even higher resolution of 3 nm has been deduced from the sharpness of edges of topographic features for a combined STM and SNOM [2].

In this Letter, we report STM-stimulated photon emission from periodic atomic-scale gratings comprised of one-dimensional Au rows separated by 8.16 Å on Au(110) surfaces (cf. Fig. 1). For the first time such a grating is clearly resolved in a photon emission experiment. Less intense photon emission is detected when the tip is on top of atomic rows. This correlation between the atomic structure and an electrodynamic phenomenon is interpreted in terms of local variations in the electromagnetic interaction of the tip and sample.

SPM take advantage of various interactions of a probe with a sample to achieve two tasks: first, to control the proximity of probe and sample in a constant-interaction mode of operation, and second, to perform a measurement under this condition [3]. Different interactions are used for control and measurement in many of these methods, an example being STM-stimulated photon emission. In this case the tunneling current enables the proximity of tip and sample to be controlled to a picometer level while their electromagnetic coupling, which determines the photon emission, is being monitored [4,5]. The underlying reasons for the atomic contrasts reported here are the different length scales of the interactions used for control and measurement. With the tunneling current as a control signal, the proximity of tip and sample is maintained constant with angstrom lateral and subangstrom vertical resolution. The measured photon emission is due to localized, tip-induced plasmon (TIP) modes which result from the electromagnetic coupling [5–7]. This coupling reflects sample properties on a nanometer lateral scale. It is sensitive, however, to the vertical dimension of the cavity formed by tip and sample [5]. Consequently, the high lateral resolution of the control interaction enforces variations in the measurement signal. Similar effects will have to be considered in other SPM with mixed interactions such as SNOM [1], laser-driven STM [8], and plasmon-induced current experiments [9].

The experiments were performed with a custom-built low-temperature ultrahigh-vacuum (UHV) STM employing temperatures of 5 and 50 K [10]. The Au(110) surface was prepared under UHV conditions by standard proce-

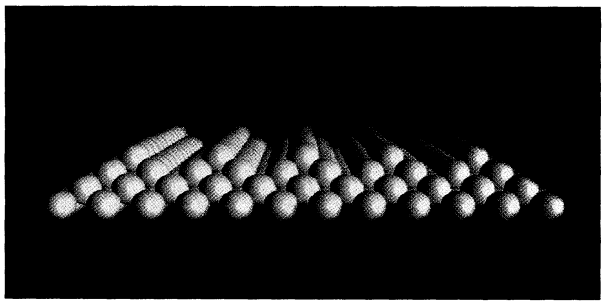

FIG. 1. A hard-sphere model of the (1 × 2) reconstructed Au(110) surface. Au rows running along [1$\bar{1}$0] are used as an atomic grating with a 8.16 Å periodicity to study photon emission.

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dures consisting of Ne ion bombardment and annealing cycles. Electrochemically etched W tips were prepared in UHV by heating and Ne ion bombardment. During the experiment, photons emitted from the tunneling gap are collected by a condenser lens (acceptance angle ≈ 0.1 sr) placed at an angle of 30° with respect to the surface and transmitted through a sapphire viewport. Outside the vacuum chamber these photons are focused onto a cooled multialkali photomultiplier which is operated in a pulse counting mode at a dark rate of 2 counts per second (cps). The photon emission from Au surfaces peaks at a wavelength of ≈ 600 nm, which is easily detected with this photomultiplier [5]. The count rate, which is measured with a ratemeter, is recorded by the STM electronics quasisimultaneously with the acquisition of constant-current topographs for each image pixel. By this procedure a *photon map* of the surface is obtained.

An example of such simultaneous imaging is presented in Fig. 2. The STM image [Fig. 2(a)] displays a (1×2) reconstructed area consisting of terraces of close-packed gold rows extending along $[1\bar{1}0]$ and monatomic steps. The rows which are separated by 8.16 Å are clearly resolved. In the corresponding photon map [Fig. 2(b)] a similar pattern is observed: rows along $[1\bar{1}0]$ with identical periodicity. The overall modulation of the photon intensity is $\approx \pm 25\%$ of the average intensity [11]. Closer inspection reveals that in the photon map the contrast is reversed as compared to the topograph. This contrast is unchanged upon reversal of the bias voltage polarity and also exhibited no pronounced variation within the bias range from 2 to 4 V. In the present experiment the STM tip is used as a local electron source (or collector). By scanning the tip laterally, the point of injection is moved with atomic-scale precision. Consequently, the observations of Fig. 2 show that less photons are emitted when the electron source is placed on top of an atomic row.

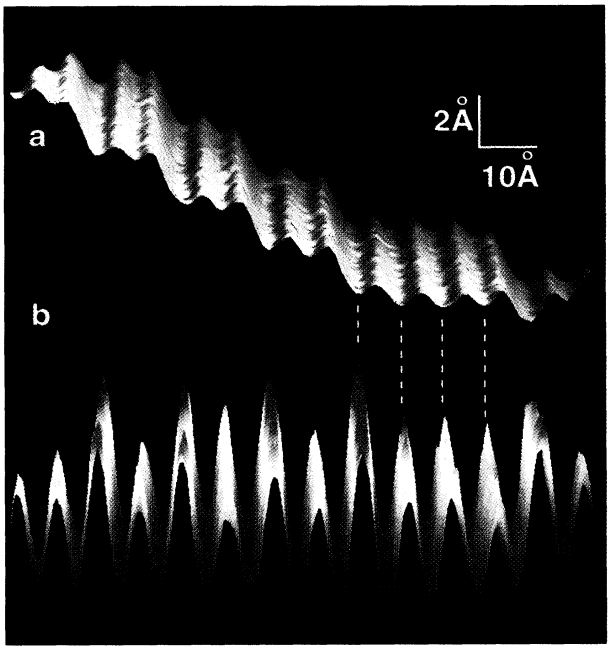

FIG. 2. Constant-current STM image (a) and photon map (b) of a stepped area of a (1×2) reconstructed Au(110) surface measured simultaneously. In (b) the photon intensity is represented as height and false color. Close-packed gold rows along the $[1\bar{1}0]$ direction, which are separated by 8.16 Å, are resolved in the topograph. An identical but phase shifted periodicity is found in the photon map. Lines serve to guide the eye. Note the increased photon intensity maxima near steps. $T = 50$ K, $I_t = 4.4$ nA, and $V_t = 3.0$ V.

A test of the phase shift between STM image and photon map was performed from an evaluation of data recorded during the forward and the backward scanning of the tip. It was further verified by simultaneous acquisition of the actual tunneling current that the observed photon contrasts were not caused by deviations of the actual current from the set value. We also exclude the possibilities that variations of either the angular distribution of the emitted light or its spectral composition are mediating the observed effect. For the subangstrom modulations in tip-sample distance that occur in the present experiment, the angular distribution of the emitted light exhibits no significant variation [12]. The spectral distribution of the emission from Au surfaces is only weakly dependent on the tunneling conditions, and the wide spectral range detected by the photomultiplier further diminishes such an effect to be of negligible importance [5].

There are several possible mechanisms for explaining the observed atomic resolution in photon maps. These mechanisms may affect the probability of inelastic tunneling which excites photon emission: first, via their effect on the electromagnetic coupling of tip and sample, second, by modulating the density of states, and third, by varying the tunneling barrier for inelastic excitation [13]. On Au(110), a possible variation of the apparent tunneling barrier between Au rows and troughs has been suggested [14]. However, the apparent barrier heights deduced for the elastic and inelastic tunneling channels were virtually identical, rendering no evidence for a causal relation [5]. Another factor found to be important for the photon emission from metallic samples is the density of final states (DOS) available for inelastic tunneling processes. In a STM experiment this DOS can be selected directly by varying the voltage. No contrast reversal is observed in conventional STM over a wide range of biases on Au(110), thus excluding this possibility. Moreover, no clear variation of the photon maps upon reversal of the bias polarity was observed which permits us to discard this possibility.

Next, a mechanism proposed by Tsukada *et al.* [15] for a related experiment is considered. They theoretically study conventional surface plasmons excited by a STM on Ag films. These plasmons are detected by their photon emission from the backside of the film through a prism. The atomic structure of the sample was proposed to give rise to local variations in the frequency spectrum of the tunneling current. The photon emission is predicted to be

most intense when the tip is placed above a topographic maximum. However, in the present experiment on corrugated Au(110) surfaces the opposite effect is observed: a 180° shift between the STM image and the photon map contradicts such a mechanism.

We propose that local variations in the strength of TIP modes affect the coupling of tunneling electrons to these modes and hence the photon intensity. These variations result from the subangstrom vertical motion of the tip as it scans the corrugated Au surface. Conceptually this may be described as follows: The lateral extent of the TIP modes has been estimated theoretically to be approximately 50 Å [16]. Therefore, *the strength of the TIP modes is sensitive to the tip-sample distance averaged over a surface area of similar lateral dimension.* The tunneling current, on the other hand, is localized laterally to within a few angstroms [17]. Given that the electromagnetic interaction of the tip and sample depends sensitively on this distance [5], we conclude that the variation of the distance between the tip and the height-averaged surface imposed by the atomic corrugation leads to the observed modulations of the photon intensity.

The size of the effect invoked may be estimated as the product of the measured corrugation (0.7 Å) and the intensity variation with tip-sample distance (40%/Å [18]). One obtains an intensity variation of ≈30% which is comparable to the experimental value. This mechanism explains the contrast reversal between the STM image and the photon map. The tip-sample distance averaged over the lateral extent of the plasmon is larger on top of a Au row than in a trough. This larger distance corresponds to a decrease in plasmon field strength and consequently less photon emission in agreement with the experiment.

Further evaluation of this contrast mechanism is obtained from an analysis of steps (cf. Fig. 2) which in the photon map show distinct intensity variations. Figure 3 displays cross sections of another stepped area along [001], i.e., perpendicular to the gold rows. In the topographic cross section [Fig. 3(a)], atomic rows and several small terraces are resolved. The apparent corrugation of the rows is typically 0.4–0.8 Å. The terraces are separated by monatomic steps (height 1.45 Å). In the corresponding section of the photon map [Fig. 3(b)] one sees again the atomic resolution of the photon signal and the contrast reversal with respect to the STM image. Moreover, a distinct variation of the modulation of the photon signal is found in the vicinity of steps. A significantly stronger photon intensity maximum occurs when the STM tip is placed next to a step on the side of the lower terrace. In addition, the modulation of the photon signal is slightly smaller on narrow terraces than on larger ones. Virtually identical observations were made in several experimental runs.

In order to understand these peculiarities observed at step structures, model calculations were performed based on the above outlined concept. The photon emission intensity from a corrugated surface $h(x)$, which is observed

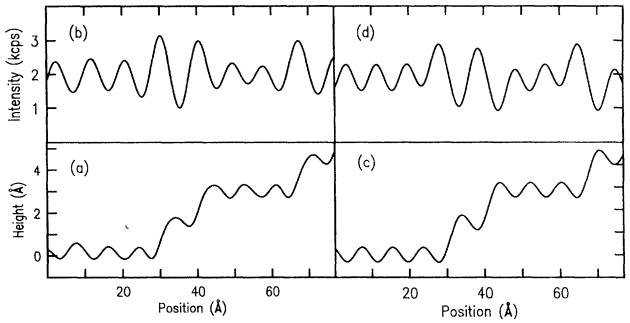

FIG. 3. Cross sections through a stepped area of a Au surface and results of a model calculation. (a) Section of a STM topograph, (b) corresponding section of the photon map, (c) surface corrugation in the simulation, and (d) calculated photon intensity. Section (b) has been low-pass filtered to decrease the photon shot noise.

in a STM topograph, is estimated by defining an average surface height $\bar{h}(x)$ from a convolution of the topographic surface with the lateral extent of the TIP mode:

$$\bar{h}(x) = \int_{-\infty}^{\infty} h(\tilde{x}) g(x - \tilde{x})\, d\tilde{x}\,,$$

$$g(x) = \frac{1}{c} \exp\left[-\frac{1}{2}\left(\frac{x}{\sigma}\right)^2 \right].$$

$g(x)$ is a weighting function representing an approximately Gaussian variation of the electrical field strength with lateral distance from the tip axis. c is a normalization factor [19]. For a given distance between the tip and this averaged surface, the emitted photon intensity is p_0. Previous experimental and theoretical results have shown that small tip displacements from this distance result in a proportional variation of the photon intensity [5]. Thus, the photon emission $p(x)$ from a corrugated surface is estimated to be

$$p(x) = p_0 + \alpha\,[\bar{h}(x) - h(x)]\,,$$

where α is a constant.

A simulated emission pattern for the step structure of Fig. 3(c) is displayed in 3(d). It shows an encouraging similarity with the observed patterns, in particular the intensity variations near steps. Two parameters were used in the calculation. The amplitude of the modulation of the photon intensity is controlled by the parameter α. To reproduce the experimental data $\alpha \approx 80\%/\text{Å}$ was used, which is larger than a theoretical prediction [18]. This deviation is not surprising given the simplicity of that theoretical model: The tip was simulated as an isolated sphere and tunneling was modeled essentially one dimensionally [6]. The predominant effect of the second parameter σ is to determine the width of the region near steps that differs from the flat surface. $\sigma = 10$ Å results in a width of ≈20 Å of the weight function g. This value is slightly smaller than the width predicted by the model of Ref. [6]. A conical form would furnish a more realistic description of this width [20] and would also affect α.

Finally we would like to propose that steps may reduce the extent of the TIP mode as compared to defect-free regions.

When molecules are inserted between tip and sample, new phenomena which require additional consideration may occur. These include molecular excitations, the interaction of the molecular states with the metal surfaces, and the quantum electrodynamical modifications of molecular emission within a microscopic cavity. Indeed, molecurlar resolution in photon emission was achieved from C_{60} monolayers on Au(110) and tentatively interpreted in terms of a molecular photon source [21].

In summary, the first experimental observation of atomic resolution in STM-induced photon emission has been reported. The contrasts observed at 50 K on (1×2) reconstructed Au(110) surfaces have been interpreted in terms of local variations of the electromagnetic coupling between tip and sample. These variations are introduced by the subangstrom vertical motion of the STM tip as it scans the sample in a constant-current mode of operation. These effects are expected to play an important role in SPM involving mixed interactions for control and measurement.

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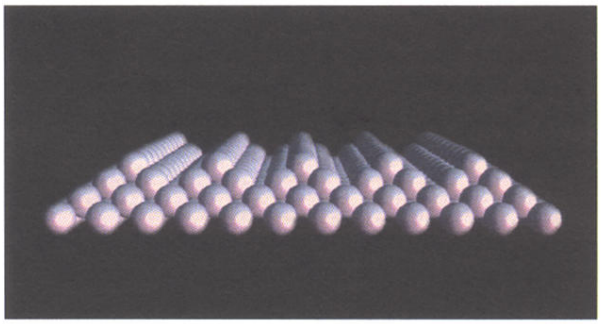

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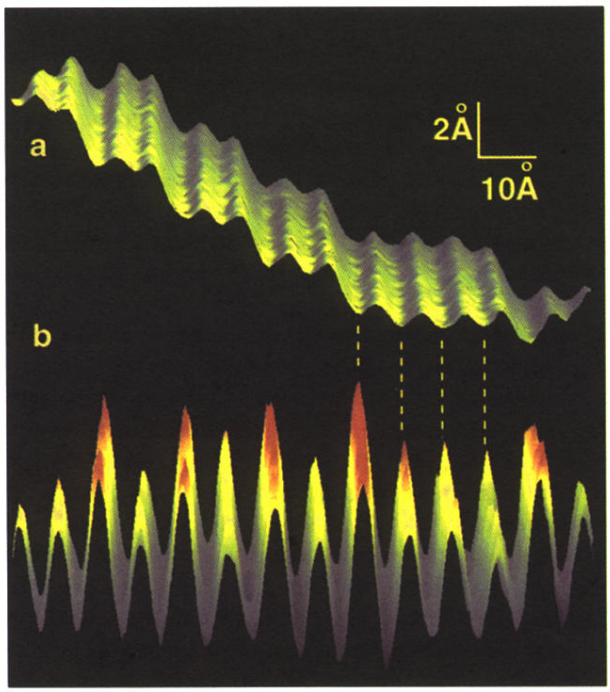

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