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PHOTON EMISSION INDUCED BY THE SCANNING TUNNELING MICROSCOPE

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Abstract

By using the tip of a scanning tunneling microscope (STM) as a local source for electrons (or holes) light emission can be excited from metals, semiconductors and molecules. Using this technique, it is possible to combine the high spatial resolution of STM with optical techniques. We review results obtained using a variety of modes of measurements including fluorescence spectroscopy, isochromat spectroscopy and simultaneous mapping of photon emission and surface topography. In spatial maps of the photon emission, clear contrasts are observed with lateral resolutions below 1 nm which are related to the geometric and electronic structure of the sample and the tip. In particular, recent results on atomic resolution of Au(110) are discussed to highlight the important role of the electromagnetic interaction of the tip and the sample for the observed photon emission.

Key Words: Scanning tunneling microscopy, inelastic tunneling, luminescence, Au(110).

Introduction

The resolution of optical microscopy is limited by the wavelength of the light being used. One possibility to increase the resolution is to apply radiation of shorter wavelength. A different approach is taken by the scanning probe microscopies: by using an excitation source (or a detector) of sub-wavelength dimensions, these techniques probe the near-field of a sample. In the case of photon emission from the scanning tunneling microscope (STM), the scanning tip serves as a local excitation source and the stimulated photon emission is detected in the far-field by conventional optics. The motivations to undertake photon emission experiments with the STM are numerous. Many excitations of solids and molecules lead to the emission of visible or infrared light. The possibility of studying these excitations with high spatial resolution or, in turn, to use them for identification of e.g., individual molecules, is a goal that justifies substantial effort. A combination of STM and photons offers distinct advantages. First, by detecting photons emitted from a STM, sensitive optical techniques may be used for detection and analysis. Photons thus represent a particularly rich, additional channel of information. Their intensity, spectral distribution, angular emission pattern, polarization status, and time correlation represent unique probes to the tunneling region which are related to various sample properties. Second, by exciting the photon emission with a STM, a well-defined source is used. Its lateral extension is of atomic dimensions and its distance from the sample, a crucial factor in any near-field technique, may be controlled with picometer precision. Third, photon emission provides direct information on inelastic processes, such as, inelastic tunneling (IET) and hot-electron thermalization. These intriguing processes [25, 48] are difficult to detect in STM by conventional tunneling spectroscopy due to their small contribution to the total tunneling current.

STM induced photon emission has been investigated from metals, semiconductors and molecules by a number of research groups. Before discussing recent advances for metallic systems, we will briefly review their work.

Experimental Techniques

Experimental aspects of photon emission from the STM are addressed by Berndt *et al.* [9]. In the majority of experiments, the emission of photons in the energy range 1-4 eV from the tip-sample region has been detected above the sample using lenses or ellipsoidal mirrors. From thin metal films, however, a weaker photon signal can also be measured from the backside of these films [38, 39, 41]. Various modes of measurement have been employed to characterize the photon emission, including, fluorescence spectra, isochromat spectra, simultaneous tunneling spectroscopy and optical spectroscopy, and angular distributions. The detected photon intensities also permit spatial mapping of the photon emission intensity ("photon maps") simultaneously with conventional constant-current imaging.

Metal Surfaces

Scientific context

Photon emission from metals in the STM is related to previous investigations of light emission from metal-oxide-metal tunnel junctions which was first discovered by Lambe and McCarthy in 1976 [30]. Theoretically, the emission was described with an inelastic tunneling model where a tunneling electron loses energy in the barrier region to a plasmon [36]. Unsolved discrepancies between experiment and theory led Kirtley *et al.* [29] to propose an alternative hot-electron excitation mechanism in which the electron tunnels elastically and then thermalizes inside the metal, generating plasmons which finally radiate. The question as to which of these two possible processes dominates is still not entirely answered.

An early proposal to detect light and secondary electrons generated by electrons from a field emission tip is due to Young [50]. The first observation of photon emission from silver films in the STM was reported by Gimzewski *et al.* in 1988 [17, 23] who proposed that inelastic tunneling and local plasmons were responsible for the observed emission. With the STM, the freedom to vary experimental parameters on well-characterized surfaces has permitted detailed investigations of the processes involved, and several theoretical models have been invoked to describe the emission spectra.

Theoretical modelling

On the theoretical side, the role of inelastic tunneling as the predominant excitation mechanism (as compared to hot-electron injection) was established by Berndt *et al.* [7] based on the theory of Persson and Baratoff [33]. Within the model of Johansson, Monreal, and Apell [27, 28] photon emission spectra were calculated which showed qualitative similarities with experimental results. Recently, it was demonstrated that photon spectra obtained from Cu, Au and Ag surfaces using W, Ag and Au tips can be modelled fairly well [8, 13]. In these models, the photon emission is attributed to radiative decay of localized, tip-induced plasmon (TIP) modes which are excited predominantly by inelastic tunneling (IET) processes. Subsequent model calculations have confirmed this interpretation. Tsukada *et al.* [42] addressed the role of high frequency components of the tunneling current spectrum which drive the TIP modes. Photon emission from thin metallic films was calculated by Uehara *et al.* [43]. Their model concerns the respective characteristics of direct emission from TIP modes and emission from surface plasmon-polaritons from the backside of a film.

Experimental results

Spectroscopic results Photon emission has been found to occur in two distinct regimes [8]. At elevated tip voltages ($V_t > 100$ V), which correspond to distances s between tip and sample larger than the wavelength λ of the emitted light, fluorescence spectra indicate that the emission is due to transition radiation and emission from surface plasmons which are excited by the impinging electrons. In the proximity field-emission ($V_t < 40$ V) and tunneling ($V_t < 5$ V) regimes, where $\lambda >> s$, the photon emission is red-shifted with an enhanced quantum efficiency of 10^{-4} photon per tunneling electron for W tips on Ag surfaces. Similar results were obtained on other noble metal [8, 45] and transition metal surfaces [10]. The intense emission in the tunneling regime has been interpreted as being due to TIP modes excited by inelastic tunneling for the following reasons [7, 8, 17, 23, 28, 33]. The high intensities and the lack of a significant bias dependence suggest a resonant process. Model calculations, which reproduce the fluorescence spectra measured with transition-metal tips and the experimental photon intensities, clearly favor the above interpretation. Furthermore, higher intensities and new spectral features found using noble-metal tips [13], as well as the angular distribution of the photon intensity, are in qualitative agreement with these models. Finally, field emission resonances detected in isochromat spectra have been used as a clear indication that the inelastic processes occur in the gap between tip and sample as expected for IET [5, 17].

Measurements of total photon intensity as a function of tunneling voltage conducted in air have been reported for several combinations of tip and sample materials [39, 40, 46]. These results differ from results obtained in ultra-high vacuum with isochromat spectroscopy, which was performed over a wide range of photon energies and bias voltages [5, 17]. We attribute these differences of

measurements in air, most notably, to the effects of contamination and the resulting tunneling characteristics, to material transfer between tip and sample, as well as to electrical breakdown at elevated voltages.

Photon mapping The interpretation of results of photon mapping is a formidable task. For experiments carried out under ambient conditions, there are problems with contamination and electrical breakdown at the voltages used to excited the emission of visible light. Nevertheless, several reports have shown that photon mapping and spectroscopy are possible on a variety of noble metal surfaces [21, 37, 38, 41, 45, 46]. A framework for the interpretation of photon maps from metal surfaces was presented in Berndt and Gimzewski [6]. The local photon emission properties of surface defects, adsorbates and structures produced with the STM itself have been investigated to explore the potential of the technique for chemical mapping. Contrasts in photon maps on a scale of some tens of nanometers were attributed to local variations in the field strength of tip-induced plasmon modes which are determined by the geometry of the tunneling junction and its dielectric properties [6, 7, 21, 31, 37]. On a (sub)nanometer scale, a second contrast mechanism was observed to occur consistent with geometry-induced variations in the matrix element for inelastic tunneling. A comparison of electron spectroscopic data with bias-dependent photon maps indicated that contrasts on a sub-nanometer scale are further mediated by local modifications of the density of final states positioned one quantum of energy ($\hbar\nu$) below the bottom of the elastic tunneling channel.

Molecules

More recently, photon emission has been studied from C_{60} monolayers on Au [11, 12, 14]. It has been demonstrated that individual molecules can be resolved within these densely packed monolayers which implies a lateral resolution better that 1 nm. The photon emission has been tentatively attributed to molecular fluorescence enhanced by the cavity which is comprised of the tip and the sample. Photon emission from squeezable tunneling junctions, with embedded molecules, has also been discussed in terms of molecular fluorescence [19]. These investigations indicate that it may be feasible to identify individual adsorbed chromophores with sub-nm lateral resolution by their photon emission in the STM.

Semiconductors

In scanning electron microscopy (SEM), the nanometer-sized electron beam can be used to locally stimulate cathodoluminescence (CL) from semiconductors. This technique is widely applied to characterize optical and electronic properties of semiconductors [49]. The lateral resolution, which can be obtained in CL, is often determined by the thermalization volume of the high-energy electrons used in SEM which usually is significantly larger than the diameter of the electron beam. In a STM, the electrons tunneling from or to the tip provide a bright and extremely localized source of electrons (or holes). The low energies of these electrons, which may even be below the vacuum level, permits nanometer lateral resolution to be achieved.

Photon emission from a STM was first reported for Si(111) surfaces where ultraviolet photons were detected [22]. The data exhibited similarities with conventional inverse photon emission. Emission due to optical transitions between the band edges was observed on direct band-gap materials analogous to conventional cathodoluminescence [1]. Spectral resolution of semiconductor luminescence was reported for CdS surfaces [4]. The spectra revealed contributions from band-edge transitions and from deep traps in the band gap. The luminescence can be excited either by injecting minority carriers or by impact ionization [1, 4]. Applications of this scanning tunneling induced luminescence have included mapping of quantum well [3] and quantum wire [34] hetero-structures and the profiling of local band-offsets [35]. Quantum size effects in porous Si have also been addressed [18, 24]. Luminescence from GaAs under ambient conditions has been studied from passivated [47] and non-passivated surfaces [16]. The first attempts at low-temperature STM studies of luminescence from InP have been made by Montelius *et al.* [32]. The polarization of photons emitted from GaAs has been interpreted in terms of spin-polarized tunneling [2]. Polarization effects have also been addressed for Si(111) surfaces [44].

Atomic Resolution in Photon Mapping

Here, we discuss experiments performed to explore the resolution capability of STM-induced photon emission from metal surfaces. The (1 × 2) reconstructed Au(110) surface, which is comprised of atomic rows, is used as an atomic-scale line grating. The experiments were performed with a custom-built ultra-high-vacuum STM employing low temperatures (5K and 50K) [20]. Photons emitted from the tunneling gap were detected with a photomultiplier and recorded by the STM electronics quasi-simultaneously with constant-current topographs for each image pixel.

The STM image in Figure 1a displays an area of such a gold surface. The surface consists of terraces of close-packed gold rows separated by 0.81 nm and monatomic steps (height 0.14 nm). Rows with the same atomic periodicity are observed in the photon intensity

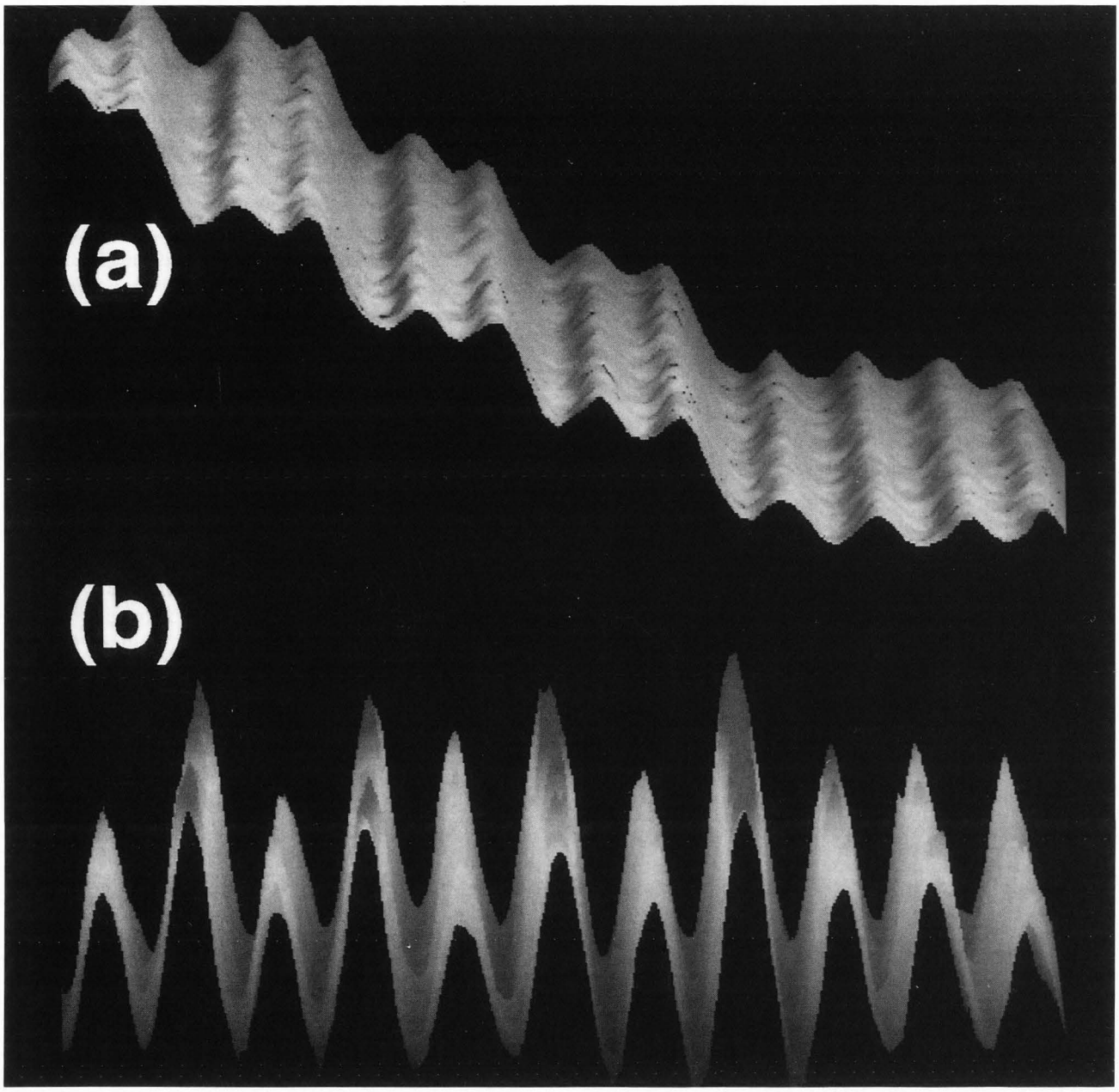

Figure 1. Constant current STM image (**a**) and photon map (**b**) of a stepped area of a (1 × 2) reconstructed Au(110) surface measured simultaneously. In (b) the photon intensity is represented by height and grey level. The overall modulation of the photon intensity is ≈ ±25% of the average intensity. Close-packed gold rows along the [110] direction which are separated by 0.81 nm are resolved in the topograph. An identical, but phase shifted, periodicity is found in the photon map. Note the increased photon intensity maxima near steps. Tip voltage 3.0 V, tunneling current 4.4 nA. The distance between the maximas in Figures 1a and 1b corresponds to 0.81 nm. (From reference [15]).

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map recorded simultaneously (Fig. 1b). Closer inspection reveals that less photons are emitted when the tip is placed on top of an atomic row.

Several origins of contrasts in photon maps have been investigated previously [6]. As discussed by Berndt *et al.* [15], we can exclude local variations of the density of final states or the tunneling barrier as possible sources of the atomic resolution.

A coherent description of the experimental data can be obtained from an analysis of the electromagnetic field strength of the TIP modes. As discussed by Berndt *et al.* [13], this strength depends sensitively on the distance

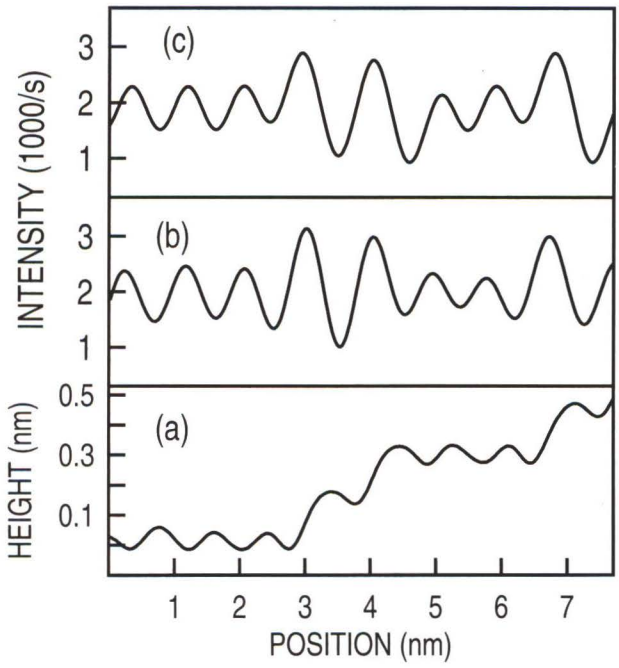

Figure 2. Cross-sections through a stepped area of a gold surface and results of a model calculation. **(a)** Section of a STM topograph perpendicular to the Au rows. Atomic rows and several small terraces separated by monatomic steps are resolved. **(b)** Corresponding section of the photon map. The atomic periodicity is resolved and phase shifted with respect to the topograph. A significantly stronger photon intensity maximum occurs when the STM tip is placed next to a step on the side of the lower terrace. **(c)** Calculated photon intensity.

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between tip and sample. From model calculations, the lateral extent of the TIP modes is estimated to be of the order of several nm [26]. Therefore, the distance between the tip and the sample, which is relevant for the photon emission, represents a lateral average over a surface area of similar dimensions. On the other hand, the tunneling current, which is used to control the proximity of tip and sample, is localized laterally to within a few Angstroms. As a consequence, the atomic corrugation in the constant-current scan imposes a variation of the distance between the tip and the height-averaged surface which entails a modulation of the photon intensity. The distance between the tip and the average surface is smaller between Au rows leading to a stronger TIP mode and hence more intense photon emission as observed in the experiment.

Further tests of the interpretation outlined above may be performed from an analysis of intensity variation at steps. In cross-sections of stepped areas (Fig. 2), a distinct variation of the modulation of the photon signal is observed in the vicinity of steps. In order to understand these observations, model calculations were performed based on the above outlined concept. The photon emission intensity from a corrugated surface $h(x)$, which is observed in a STM topograph, is estimated by defining an average surface height from a convolution of the topographic surface with the lateral extent of the TIP mode. For a small change of the distance between the tip and the averaged surface a linear intensity variation is expected from previous results [13]. Thus, the photon emission $p(x)$ from a corrugated surface is estimated to be:

$$p(x) = p_0 - \alpha \{h(x) - \int_{-\infty}^{\infty} h(\bar{x})\, g(x - \bar{x})\, d\bar{x}\}$$

where p_0 is the average photon intensity and α is a constant. $g(x)$ is a weight function representing an approximately Gaussian variation of the electrical field strength with lateral distance from the tip axis.

Simulated emission patterns (Fig. 2c) for step structures show an encouraging similarity with the observed patterns, in particular, the intensity variations near steps. This suggests that despite its simplicity, this model describes the essential physics underlying the observed atomic resolution. In summary, an overview of experiments and theories for photon emission from the STM has been presented. The recent observation of atomic resolution in photon emission has been analyzed to highlight some aspects of the intriguing physics of photon emission from metal surfaces.

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Discussion with Reviewers

M.S. Isaacson: Could the author explain, in greater detail, how spatial resolution and bias voltage on the probe are related?

Author: If one neglects tip and sample interactions, the effect of the bias voltage on the tunneling current is two-fold. First different voltages (and polarities) will change the weight of the occupied and unoccupied states of the tip and the sample that contribute to the tunneling current. Second, the distance between the tip and sample will change with the bias voltage (if one assumes an experiment performed at constant tunneling current). Therefore, the spatial positions where the respective densities of states are probed change as well. The latter effect often causes a reduced lateral resolution at large bias voltages due to an increased distance of tip and sample. The former effect can modify the STM image more drastically. If there is a strong variation of the density of states of the sample (or the tip) over the relevant bias range, the apparent shape of the imaged object may change, e.g., from an apparent hole to a protrusion. At larger voltages exceeding the work functions of tip and sample, electron standing waves occur in the tunneling gap and may lead to an enhanced lateral resolution as was first observed by Coombs *et al.* [17]. The situation also becomes more involved at low bias voltages where tip-sample interactions may no longer be neglected. Finally, images of, e.g., organic molecules may involve the interference of tunneling current components from different molecular orbitals. The interferences may depend critically on small changes of the bias voltage {Fisher AJ, Bloechl PE (1993) Adsorption and scanning-tunneling-microscope imaging of benzene on graphite and MoS_2. Phys. Rev. Lett. **70**: 3263-3266}.

R.J. Warmack: The characteristics of light emitted from such a confined region as a STM tip present us with a remarkably detailed source of information. Will you comment upon the potential limitations in optically resolving adjacent molecules because of energy transfer, surface plasmons, etc.?

Author: On the basis of the experimental data available at present, I prefer not to speculate on this topic. The mechanism involved in the molecular emission is not yet fully understood, although the experimental data presented above indicates that both the molecules themselves and the localized plasmons of the tip/sample cavity play a role. I feel that more experimental results are required as a solid basis for such a discussion.