Fiber optic light collection system for scanning-tunneling-microscope-induced light emission

Neil J. Watkins, a) James P. Long, Zakya H. Kafafi, and Antti J. Mäkinen

Naval Research Laboratory, Washington, DC 20375

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We report a compact light collection scheme suitable for retrofitting a scanning tunneling microscope (STM) for STM-induced light emission experiments. The approach uses a pair of optical fibers with large core diameters and high numerical apertures to maximize light collection efficiency and to moderate the mechanical precision required for alignment. Bench tests indicate that efficiency reduction is almost entirely due to reflective losses at the fiber ends, while losses due to fiber misalignment have virtually been eliminated. Photon-map imaging with nanometer features is demonstrated on a stepped Au(111) surface with signal rates exceeding $10^4$ counts/s.

I. INTRODUCTION

Scanning-tunneling-microscope-induced light emission$^{1-4}$ (STM-LE) is potentially capable of correlating atomic-scale surface features with light emission induced by the tunneling current. In recent years, STM-LE with sub- and nanometer spatial resolution has been achieved at metal,$^{1,5}$ semiconductor,$^{6-8}$ and absorbate-covered$^{9,10}$ surfaces.

A primary technical challenge in STM-LE measurements is the low yield of photons produced by the tunneling current. Typical maximum quantum efficiencies, defined as photons generated per tunneling electron, are between $10^{-5}$ and $10^{-4}$ for all known mechanisms, for example, charge injection into semiconductors,$^{11}$ or inelastic tunneling at metal surfaces. Since sample surfaces will not tolerate more than a few nanoamperes of current in STM measurements, the only way to maximize the signal for STM-LE is by optimizing the light collection. In practice, this means using optics with suitably large numerical apertures (NA) ($\sim 0.5$), enabling collection of light over a large solid angle. Ideally, light collection optics would be incorporated into the original design of STM instrumentation, as has been demonstrated by a number of groups who have integrated lenses,$^{12}$ mirrors,$^{11,13}$ and fiber optics$^{14}$ into STM systems. However, it can be an attractive option to add an STM-LE capability to an existing STM setup; the use of multimode optical fibers as described here provides a compact, flexible, and inexpensive approach.

In retrofitting existing STM instrumentation with light collection optics, difficulties arise from designing the optics to accommodate the nonideal geometry of the STM while providing sufficient light collection efficiency. The use of optical fibers as collection elements can solve some of the geometry-related issues because of their inherent flexibility and because they can be easily integrated with mechanical manipulators. Fiber-based collection optics for a commercial ultrahigh vacuum (UHV) STM was recently reported by Arafune et al.,$^{15}$ who used four 0.2 numerical aperture (NA) optical fibers positioned radially about the tip-sample gap. The authors found that the effective solid angle of light collection for the fiber system was 0.053 (2$\pi$ sr) which represented $\sim 63\%$ of the total solid angle defined by the four fibers. The smaller effective solid angle of the fiber system was attributed to inaccuracies in assembling the four fibers.

In this article, we report a simple light collection approach using only two optical fibers that permits photon-map imaging with a commercial room-temperature UHV STM (Multimode UHV scanning probe microscope by Omicron Associates). We find that the combination of a large core diameter and a high numerical aperture of the fibers facilitates in situ alignment of the fibers with modest (submillimeter) precision to attain a collection efficiency close to that predicted by the solid angle subtended by the fiber pair. This was verified by measuring the collection efficiency of the two-fiber system using a point source with a known angular distribution of emission. The collection efficiency was measured to be 91% of the maximum possible, which is comparable to values recently reported for other lens- and fiber-based optical collection system of STM-LE instrumentation.$^{12}$

II. OPTICAL DESIGN AND ACQUISITION ELECTRONICS

The collection optics comprised two 1 mm diameter silica-core multimode fibers with a 0.39 numerical aperture (Thorlabs). The fibers were supported by a custom-made vertical support attached to a vacuum x-y-z manipulator [Fig. 1(a)]. The manipulator permitted the fibers to be retracted for tip and sample exchange and to be subsequently realigned by optimizing the signal. Based on a widely predicted skewed-dipole pattern for STM-LE,$^5$ the ideal orientation for the fibers is at a 60° angle from the sample normal, but due to physical constraints imposed by the STM system...
fibers to a single photomultiplier tube within a tight enclosure. The detector comprised optics coupling the pickup and were coupled to the detector housed in a light-tight enclosure. The tip-sample gap and oriented toward the gap. The two fibers were individually coupled to atmosphere via an aspheric lens configured to transfer the full emission cone from the fiber while providing sufficient space between the lens and PMT body to allow for an insertion of interference filters for a spectroscopic analysis.

Incorporating the photon detection electronics with the rest of the STM control instrumentation, as shown in Fig. 1(d), was facilitated by the use of a commercial universal counting board (UCB) module, available for the Omicron SCALA data acquisition system. The UCB has four input channels for transistor-transistor logic (TTL) signals that the SCALA system can utilize as standard data channels. This permitted a simultaneous acquisition of topography and photon-map images.

III. CALIBRATION OF FIBER OPTIC COLLECTION

Before installing the fiber optic collector onto the STM system, its efficiency was measured by using a point source with known emission characteristics. A point source was created by placing an incandescent lamp behind a 0.006 in. (152.4 μm) pinhole covered with a piece of Teflon tape to act as a diffuser. The resulting intensity distribution was measured with a Si photodiode that was placed on a rotatable arm, with the center of rotation fixed at the pinhole. The intensity distribution of the point source is compared in Fig. 2 with that of a Lambertian source and the theoretical tip-induced skewed-dipole pattern. A GaAsP photocathode was selected for high quantum efficiency at visible wavelengths (37% maximum). The PMT was operated at high gain for single-photon counting and was thermoelectrically cooled for a low dark-count rate. The end of each fiber was imaged onto the full 5 mm diameter of the photocathode with a separate aspheric lens (Thorlabs, 8 mm focal length, 0.5 numerical aperture) configured to transfer the full emission cone from the fiber while providing sufficient space between the lens and PMT body to allow for an insertion of interference filters for a spectroscopic analysis.

To determine the collection efficiency, we measured the total power that was transmitted through the pinhole and compared it to the power that was collected and transmitted through each optical fiber by successively pressing the Si photodiode against the pinhole and each fiber end. We found that each fiber collected a little less than 5% of the light, 4.9% for one fiber, and 4.5% for the other, which together amounted to 9.4% of the total emitted light. This compares to a theoretical value of 10.3%, obtained by numerically integrating the experimental point-source intensity distribution over the total solid angle subtended by the two fibers. Therefore, the measured fiber collection efficiency was 91% of the maximum possible. It is worth noting that the 9% efficiency itself, the optical fibers were fixed at a 65° angle. In use, the cleaved fiber faces were manipulated to within ~1 mm of the tip-sample gap and oriented toward the gap. At this close approach, the solid angle of collection does not depend on the distance between the fiber end and the tip-sample gap, and hence collection efficiency is tolerant of a slight misalignment of the fiber end.

The two fibers were individually coupled to atmosphere through custom vacuum feedthroughs, comprising a Swage-lock fitting with a Teflon ferrule that is sealed against the cladding with no discernible loss of vacuum integrity (low 10^-10 torr, as monitored by an ion gauge and by an ion current in a commercial 300 l/min ion pump). In air, the fibers were sheathed in opaque sleeving to eliminate stray-light pickup and were coupled to the detector housed in a light-tight enclosure. The detector comprised optics coupling the fibers to a single photomultiplier tube (PMT) (Hamamasu H7422-40). The PMT was operated at high gain for single-photon counting and was thermoelectrically cooled for a low dark-count rate. The end of each fiber was imaged onto the full 5 mm diameter of the photocathode with a separate aspheric lens (Thorlabs, 8 mm focal length, 0.5 numerical aperture) configured to transfer the full emission cone from the fiber while providing sufficient space between the lens and PMT body to allow for an insertion of interference filters for a spectroscopic analysis.

FIG. 2. Polar plots of intensity distributions for experimental point source (solid circles), theoretical Lambertian source (solid line), and expected skewed-dipole source (dashed line). Each intensity distribution is normalized to its peak intensity value.
n reduction is comparable to typical reflective losses at fiber ends, implying that losses due to fiber misalignment have virtually been eliminated.

Similarly, the maximum possible collection efficiency for the skewed-dipole distribution of STM-LE was calculated to be 18.2%.\(^4\) Assuming the same ratio between the observed and the maximum possible efficiencies as in the case of the point source (0.91), the fiber system should collect 16.6% of the total STM-LE intensity. Given the maximum quantum efficiency of the PMT, 37%, and assuming no loss in the relay optics coupling the fibers to the PMT, the overall efficiency of the light collection and detection system was estimated at 6.1%.

IV. PERFORMANCE

Both tungsten and Au tips were used in STM-LE measurements. Tungsten STM tips were prepared by electrochemical etching in a 0.1M NaOH solution, and gold STM tips were simply cut from a 0.5 mm diameter Au wire. After placing the tips in vacuum, they were Ar ion sputtered (0.5 keV) for 20 min. As reported by others, the use of a gold tip increased light emission by over an order of magnitude compared to a tungsten tip, indicative of plasmonically assisted light emission.\(^10\)

A single-crystal Au substrate (Accumat Materials Co.) was prepared by repeated cycles of Ar ion sputtering (0.5 keV) and annealing (680–720 K) before performing STM-LE experiments. Figure 3 compares the simultaneously recorded topographic and photon-map images of a 50 \(\times 50 \text{ nm}^2\) Au(111) surface. The topography of the Au surface comprises a series of monoatomic steps (height \(-0.23 \text{ nm}\), resulting from a small miscut (2.3°–3.5°) defined by a sample normal tipped from [111] toward [110].\(^19\)

The location of these steps in the topographic image correlates well with the troughs observed in the photon intensity map. The STM-LE intensity level in these troughs (2800 counts/s) is significantly less than that of the flat terraces (14400 counts/s). A similar contrast in intensity has previously been observed over monoatomic steps on Au(111),\(^20\) Ag(111),\(^5\) and Cu(111)\(^21\) surfaces. Based on detailed STM scans over monoatomic steps, the light intensity modulation is believed to result from different spatial distributions of the local density of final states in the elastic and inelastic tunneling channels.\(^5\)

The data in Fig. 3 provide a means to assess the performance of the collection system. The minimum signal levels (2800 counts/s) far exceed the dark-count level of the PMT (35–40 counts/s) and provide sufficient photon intensity to perform imaging with reasonable pixel integration times at room temperature. The sample data in Fig. 3 was recorded in constant current mode with a positive sample bias \(V_S=2.1 \text{ V}\), tunneling current \(I_T=1.2 \text{ nA}\), and 5 ms/pixel integration time. For a 200x200 pixel image, this translates to a 200 s recording time. Accounting for the overall system detection efficiency of 6.1% given above, we estimate the quantum efficiency of STM-LE on Au at \(-3 \times 10^{-5}\) photons/electron, which is within the range of previously reported values.\(^1\) Note that this value represents a lower limit because it assumes the maximum PMT efficiency over the spectral width of the STM-LE emission.

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\(^12\)G. Hoffmann, J. Kröger, and R. Berndt, Rev. Sci. Instrum. 73, 305 (2002).
\(^16\)Certain vendors and commercial instruments are identified to adequately specify the experimental procedure. In no case does such identification imply an endorsement by the Naval Research Laboratory.

FIG. 3. Simultaneously recorded (a) topographic and (b) photon maps of a stepped Au(111) surface (50x50 nm\(^2\)) (\(V_S=2.1 \text{ V}\), \(I_T=1.2 \text{ nA}\)).
The solid angle of collection is independent of distance $L$ between the point source and the fiber end when $L = L_0 = d/2 \tan[a \sin(NA)]$, where $d$ is the fiber diameter and NA is the numerical aperture of the fiber. The maximum solid angle of collection $\phi$ for the fiber is given by $2\pi[1 - \cos(a \sin(NA))]$. For the fibers used in this work, $L_0 = 1.18$ mm and $\phi = 0.079(2\pi)$ sr.