Optical and electrical characterization at nanoscale with transparent probe of a scanning tunnelling microscope

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Abstract. A new type of a scanning probe microscope, combining features of the scanning tunnelling microscope (STM), the scanning tunnelling luminescence microscope (STML) with a transparent probe and the aperture scanning near-field optical microscope (aperture-SNOM) is described. Proof-of-the-concept experiments were performed under ultra high vacuum (UHV) conditions at varying temperature on GaAs/AlAs heterostructures.

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1. Introduction

Optical and electrical properties of nanostructures can be addressed using electromagnetic radiation or electrical current as a probe. The probed properties, on the other hand, are dependent on the excitation conditions being that of electrical or optical nature. The parameters of the excitation, such as a pumping wavelength for the optical probing or an applied bias for the electrical characterization, pre-define measurement results.

In general, a near-field type of electromagnetic interaction is necessary for an optical probe to enter nanoscale regime. In this case the optical diffraction limit ($\sim 1 \mu\text{m}$) for spatial resolution can be overcome. A typical aperture scanning near-field optical microscope (aperture-SNOM) provides such an opportunity both for the excitation and collection of light [1]. However, this instrument employs a dielectric fiber tip as an aperture, which makes it unsuitable for electrical measurements. An apertureless SNOM, on the contrary, has a metallic, needle-like opaque tip. The local field enhancement at the tip apex due to the plasmon effect enhances signal in the light scattering experiments [2]. But, unlike the case of aperture-SNOM, the far-field optics alignment is necessary and the scattered light from the tip shaft can distort the measurements. In addition, the aperture-SNOM can operate in the combination with a scanning tunnelling microscope (STM) and it was demonstrated that such a combination can yield superior resolution for SNOM imaging compared to the conventional shear-force approaching method [3].

On the other hand, the STM, which is capable of atomic-resolution measurements using electrical current, can also cause an optical response in materials [4]. The collection of light in ordinary STM machines with opaque metal tips is realized by far-field optics [5]. When the carriers are tunnelled

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to the sample it is the excitation (tunnelling) area that limits the spatial resolution. Therefore the sub-10 nm resolution can be achieved with such an instrument [6]. The collection efficiency of the emitted light can be improved by employing a transparent STM tip, where the light is coupled directly to the tip rather than to a far-field detector [7]. However, only the electrical probing of nanostructures was proven feasible in such a scanning tunnelling microscope luminescence (STML) experiment [8].

Here we expand the STML approach to show an ultimate STM-based configuration, where both optical and electrical excitation is available. The tips can be exchanged preserving UHV conditions, which makes possible probing the same sample using different microscopy regimes. In addition, optical and electrical characterization is demonstrated with the same tip, thus targeting the same area on the sample.

2. Experimental

The STML instrument using a conductive and transparent tip, featuring sub-10 nm spatial resolution in the electroluminescence regime, was reported previously [7]. In this work the STML setup has been complemented with a beamsplitter unit in a configuration typical for the fluorescence microscopy (Figure 1). In order to provide optical excitation the second-harmonic Nd:YAG laser with a 5W maximum output power (Millenia, Spectra Physics), emitting continuous wave (cw) beam at 532 nm, was used. The excitation light is coupled through a single-mode fiber to the beamsplitter input port. From the beamsplitter unit the light is directed to the STM UHV chamber by the multimode, H₂-doped silica fibers (STU-800 outside the UHV chamber and STVH-455M inside the chamber, Mitsubishi Densen) through the silica glass window of a mini-viewport flange.

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The signal is collected through the same transmission line (cf. Figure 1) by the triple-grating spectrometer (SpectraPro, Princeton Instruments) with an attached charge-coupled device (Spec-10 CCD array of 1340x100 pixels, Princeton Instruments) for spectroscopy or by the photomultiplier tube (R3310-02, Hamamatsu) for the photon mapping. The spectroscopy detection range is 700-1000 nm, limited by the CCD sensitivity for the long wavelengths and by the short wavelength cut-off filter from the other side of the spectrum.

The STML tip represents a tapered optical fiber, coated with a conductive and transparent layer of indium tin oxide (ITO) 50-70 nm thick [9]. In order to make it suitable for high spatial resolution optical measurements a metal coating was applied with the formation of a subwavelength aperture at the tip apex (Figure 2). First, Pt-Pd layer 400-500 nm thick was deposited all over the tip using magnetron sputtering. Then the aperture was opened at the tip apex using focused ion beam (FIB) milling (SII, XVision 200TB). The aperture was formed such that the tip apex has taken a pyramid-like shape, which is necessary for STM mode operation with high spatial resolution.

The control sample used here was a p-doped GaAs substrate ($n_p = 10^{19} \text{ cm}^{-3}$) with GaAs/AlAs alternate multilayers of various thicknesses: 50, 20, 10 and 5 nm; six pairs for each thickness. The heterostructures were grown by the metalorganic chemical vapour deposition (MOCVD) technique [8]. These nanostructures can provide topography contrast for STM measurements as well as luminescence contrast for optical measurements due to the direct/indirect bandgap nature of the GaAs/AlAs semiconductor heterostructures.

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The measurements were carried out under UHV condition ($P < 3 \cdot 10^{-8}$ Pa) on the sample surface prepared in the vacuum by cleavage. All the luminescence spectra were corrected by the signal from the optical line for the retracted tip.

3. Results and Discussion

Since in the present configuration the signal is collected through the same transmission line as the excitation is guided in, both the inelastically scattered excitation light and the induced luminescence from the optical line should be minimized. There are some inherent defect states in fused silica, which can cause specific luminescence from the fiber or window components (see e.g. [10]). For example, a ~ 1.9 eV (650 nm) luminescence band in silica can be activated by the 532 nm laser light used here. To avoid the detection of this emission a cut-off filter was used shifting the detection range to the near-infrared wavelengths.

In the optical excitation regime the excitation power from the laser is driven to the tip apex. It was shown that depending on the tip geometry the transmittance of such a tip can vary almost two orders of magnitude due to constructive/destructive interference [11]. Due to the small size of the subwavelength aperture the local power density at the tip apex can reach values sufficient for the sample damage. Thus the appropriate values of the excitation power must be chosen for every tip/sample configuration. The same is true for the electroluminescence regime, where high values of the applied bias can lead to the degradation of sample luminescent properties. Here, a several mW laser power and less than +3 V sample bias voltage were used.

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The optical properties of a substrate affect spatial distribution of the light emanating from a point-like emitter [12]. This is due to the non-uniform distribution of the radiative power for the light emitter located at the interface of two optical media. For higher refractive index substrates less light is emitted in the upper halfspace thus decreasing the measured yield. Although the semiconductor substrate used in this work has a high value of the refractive index ($n_{\text{GaAs}} \approx 3.5$) it was nevertheless possible to detect light emission both in electric and optical excitation regimes.

The results are summarized in Figure 3. A tip for all-optical regime was prepared using metal deposition and FIB milling as described above. Here, however, the tip aperture size was made smaller, around 150 nm in diameter as seen in Figure 3 (top, left). The pyramid-like shape of the tip is not clearly visible since the pyramid slope was intentionally made gentle to reduce the opening size. Nevertheless, the successful operation in the STM mode with this tip was possible as demonstrated in Figure 3 (top, middle). Although the tip apex is not as sharp as for an ordinary STM metal tip the processing of the tip apex with a FIB and a standard STM built-in piezo positioning and aligning system allow STM operation with high spatial resolution. Indeed, 5 nm thick GaAs/AlAs multilayers can be resolved in this mode as shown on the inset. Thus the area of interest can be located by operating the microscope in STM mode before switching to the optical characterization. The last figure in the top row is the photoluminescence spectra recorded with this tip at room and liquid nitrogen temperatures from GaAs. The smaller bandgap (longer emission wavelength) at higher temperature and the sharper luminescence line at low temperature confirm the origin of luminescence being that of bandgap recombination in the bulk semiconductor. It is also important to note that luminescence at low temperature is more efficient due to the suppressed diffusion of carriers to non-radiative recombination centres.

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The lower row in Figure 3 represents results of the electrical excitation experiment. The tip was a tapered silica fiber with conductive and transparent ITO layer on top. Again, GaAs/AlAs multilayers from 50 to 5 nm thickness can be resolved in the STM operation mode (middle, bottom). However, the signal-to-noise ratio for the low temperature electroluminescence spectrum of GaAs is worse than that of measured by optical excitation (right, bottom). This is due to the smaller area probed in this regime. The spatial resolution for electroluminescence measurements is limited by the excitation area on the sample. Indeed, the 50 nm thick multilayers can be clearly resolved by the photon mapping as shown on the inset. From this image the spatial resolution can be estimated to be less than 10 nm [8]. However for the optical excitation the spatial resolution is typically limited by the aperture size [1]. Thus the excitation area is ~ 200 times larger for these tips, resulting in visibly better signal-to-noise ratio for the photoluminescence spectrum. The concrete values for the typical trade-off between the spatial resolution and the collected yield for this system need to be investigated further.

Finally, tips can be routinely exchanged inside UHV chamber using magnetic coupler transfer rods. Thus the switching between different microscopy regimes (STM, STML, and SNOM) can be realized by mounting the appropriate tip without breaking the vacuum. In addition, not only the same sample can be probed in different regimes in this system, but also the same area on the sample. It can be realized, first, by localizing the area of interest in the STM mode and then targeting it optically or electrically. The most straightforward way, however, is to use the same tip for all the regimes mentioned. In Fig. 4 we show electrically and optically excited luminescence spectra measured with the tip shown in Fig. 3, top, left. Thus metal-covered tips with a FIB prepared subwavelength aperture can operate in all three regimes targeting the same area on the sample. Also

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note that in this case the contribution of the near-field coupling in electroluminescence regime is stronger than in Fig. 3, bottom due to the subwavelength size of the light-collecting aperture.

In brief, a new microscope combining optical and electric excitation into a single STM-based unit was described with the proof-of-concept measurements performed on GaAs and GaAs/AlAs heterostructures at varying temperature. This approach may find its niche not only where all-optical measurements are required in addition to STM and STML characterization, but also where electrical modification with subsequent *in situ* optical probing is desirable.

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FIGURE CAPTIONS

FIG. 1 Schematic representation of the STML setup with an addition of the optical excitation. Excitation beam is guided through optical fibers via a beamsplitter unit to the tip inside an UHV chamber. The signal, collected by the subwavelength aperture tip, propagates through the same transmission line back to the spectrometer with CCD for spectroscopy or to the photon counter for luminescence microscopy measurements.

FIG. 2 Tip preparation for all-optical measurements. (a) A metal film is deposited all over an STML tip. SEM images before and after metal deposition are superimposed for the aperture design. (b) Focused ion beam milling is used to create a pyramid-like shape at the tip apex. A sharp apex is necessary for successful STM operation while a subwavelength aperture is needed for high spatial resolution optical measurements.

FIG. 3 Results of the microscope operation in optical (top) and electric (bottom) excitation regimes. STM (middle) and luminescence (right) modes are shown. SEM images of the tip before and after FIB processing are superimposed to show the subwavelength aperture for all-optical operation (top, left). Alternate GaAs/AlAs multilayers with thicknesses of 50, 20 and 10 nm can be discerned in STM images acquired for both regimes (middle); 5 nm multilayers can also be seen in the optical tip STM image (top, middle, inset). Luminescence spectra of GaAs at 80 K and room temperature were obtained using 2 mW 532 nm optical excitation (top, right) and at 80 K for the electrical excitation (bottom, right). Electroluminescence photon mapping of the 50 nm thick GaAs/AlAs multilayers (bottom, right, inset) indicates sub-10 nm spatial resolution [7]. Signal-to-noise ratio for the electric excitation spectrum is worse due to the smaller excitation area.

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FIG. 4 Optical and electrical excitation luminescence spectra from GaAs measured at low temperature (82 K) with the SNOM/STM tip shown in Fig.3, left, top. Laser excitation power was 5.3 mW for optical measurements and sample bias was +1.5V with tunneling current 0.2 nA for the electrical excitation regime.

FIG. 1

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