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## Template stripping and bonding of smooth probes with nanoscale features for tip-enhanced Raman spectroscopy

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### Abstract

The batch fabrication of scanning microscopy probes for tip-enhanced Raman spectroscopy (TERS) is an often discussed issue and remains a challenging task. In our work, we started from a known metal moulding process and developed a dry release process based on metal bonding. After establishing the release process, scanning tunnelling microscopy probes could reliably be fabricated and used for tip-enhanced Raman measurements. The Raman signal enhancement with laser excitation from the side was investigated on substrates covered by malachite green dye molecules, where clear signal from few molecules could be detected. TERS sensitivity was investigated on substrates coated with ultra-thin amorphous carbon layers and compared to standard confocal Raman spectroscopy.

### Highlights

- A bonding-based dry-release process for template stripped microstructures was developed.
- Scanning tunnelling microscopy probes for tip-enhanced Raman spectroscopy were reproducibly fabricated.
- Tip-enhanced Raman signal from few molecules of malachite green dye and ultra-thin amorphous carbon layers was measured.

### Keywords

Template stripping, metal moulding, microstructure dry-release, microstructure bonding, tip-enhanced Raman spectroscopy, amorphous carbon, scanning probe microscopy

### 1. Introduction

Raman spectroscopy utilizes the inelastic scattering of monochromatic light e.g. by molecules to probe for material composition, mechanical properties, crystallinity and a variety of other properties. Due to its small cross section on the order of  $10^{-30}$  cm<sup>2</sup>, Raman scattering is relatively weak compared to Rayleigh scattering, and the lateral resolution is limited by the excitation laser focus, typically around several hundred nanometres. As a possibility to increase the signal intensity and lateral resolution of optical imaging, apertureless scanning near-field optical microscopy (a-SNOM) was developed, the concept of which was first proposed in 1985 [1] and demonstrated for the first time in 2000 [2-4]. Microscopy and spectroscopy on the nanoscale are now playing an increasing role in addressing biological, medical or material questions down to the molecular level [5].

Often, the term tip-enhanced Raman spectroscopy (TERS) is used when apertureless SNOM is combined with Raman spectroscopy. In TERS, a metallic tip is scanned over a sample surface using a scanning probe microscopy (SPM) feedback mechanism while the Raman laser is focused on the tip. At each point a full Raman spectrum is recorded. The tip is expected to work as an antenna for the incoming electrical field as well as for the scattered near-field, which leads to the well-known  $|f(\omega_R)|^2 \cdot |f(\omega_0)|^2$  enhancement using a point dipole approximation, where  $f$  denotes the local field enhancement factor,  $\omega_0$  denotes the laser frequency and  $\omega_R$  the Raman frequency [6]. Resulting from the strong and localized enhancement at the hotspot near the tip apex, the lateral resolution is shown to

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