

Integration of a Raman spectroscopy system with a low-temperature scanning tunnelling microscope under UHV conditions

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Abstract

This article presents an optical system for integrating Raman spectroscopy with a commercial low-temperature scanning tunnelling microscope (LT-STM) operating under ultrahigh vacuum (UHV) conditions. The external excitation, collection, filtering, alignment and mechanical-support modules were designed and constructed to match the factory-defined 30° optical geometry of the LT-STM platform while avoiding direct mechanical modification of the suspended STM head. The performance of the integrated configuration was evaluated at room temperature using a single-crystal silicon reference sample and compared with a near-normal-incidence Raman configuration operated in a glove-box environment. Analysis of 100 individual acquisitions per configuration showed low short-term acquisition-to-acquisition variability. After normalisation to the incident excitation power measured immediately before the corresponding external window, the mean integrated intensity of the first-order silicon Raman band in the LT-STM configuration was lower than that obtained in the glove-box configuration by a factor of approximately 7.4. The first-order silicon phonon band remained clearly resolved in the LT-STM configuration. The results demonstrate the feasibility of far-field Raman detection under UHV conditions and provide a practical basis for future low-temperature Raman and TERS studies.

1. Introduction

The ability to correlate local atomic structure with vibrational and electronic properties is central to modern surface science and nanotechnology [1]. Scanning tunnelling microscopy (STM) provides real-space imaging with atomic [2] and molecular [3, 4] resolution, while Raman spectroscopy delivers chemically specific vibrational fingerprints in a non-destructive manner [5, 6]. Their integration enables correlated structural and spectroscopic characterisation at the nanoscale. In particular, tip-enhanced Raman spectroscopy (TERS) combines the spatial resolution of STM with plasmonically enhanced Raman scattering localised at the metallic tip apex, enabling chemical sensitivity beyond the optical diffraction limit [7–9].

Raman spectroscopy has become a key analytical technique for investigating the structural and vibrational properties of low-dimensional systems [10, 11]. In our laboratory, it is routinely used for comprehensive characterisation of two-dimensional materials, including the analysis of thickness-dependent, structural, and optoelectronic properties [11–13]. These studies are conducted using a Raman spectrometer developed by our research group and optimised for operation in a controlled argon atmosphere within a glove-box environment, effectively preventing sample degradation and minimising environmental contamination [14–16]. However, to achieve atomic-scale correlation between structural, electronic, and vibrational properties under strictly controlled surface conditions, Raman measurements must be extended to ultrahigh vacuum (UHV) environments.

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UHV conditions are essential for studies of atomically clean surfaces, well-defined interfaces, two-dimensional materials, adsorbed molecular layers, and weakly bound surface species, where even minor contamination may significantly affect the measured properties. Combined with low-temperature (LT) STM, these conditions enable surface-sensitive phenomena, including phase transitions and adsorbate–substrate interactions, to be investigated under precisely controlled cryogenic and UHV environments. Commercial LT-STM systems enable atomic-resolution imaging under cryogenic and UHV conditions, yet their optical accessibility is inherently limited. The literature has extensively reviewed optical architectures for TERS under UHV conditions, ranging from the comprehensive review by Pozzi *et al.* [17] to the overview by Schultz [18]. Several optical architectures have been developed for Raman spectroscopy and TERS under UHV conditions. Fully custom-built instruments commonly employ internal aspheric lenses or high-numerical-aperture parabolic mirrors, providing efficient light collection and enabling nanoscale TERS measurements. Other implementations use fibre-optic probes for far-field Raman monitoring under UHV conditions, whereas more recent approaches are based on optical retrofits of existing commercial scanning probe microscopy platforms. Nevertheless, detailed descriptions of practical optical integration strategies specifically adapted to commercial LT-STM architectures remain limited. Table 1 compares representative systems and their principal characteristics. The comparison in Table 1 shows that large collection solid angles and demonstrated nanoscale TERS performance have generally been achieved using fully custom-built instruments equipped with internal high-NA lenses or parabolic mirrors. In contrast, the present implementation retains the factory-installed internal 30° optical geometry of a commercial LT-STM, while the excitation, filtering, optical-inspection, fibre-coupling, and mechanical-support modules are implemented externally. This approach avoids modifying the STM head and introducing additional custom optical components into the cryogenic UHV region. However, unlike several mature TERS platforms listed in Table 1, the present study validates far-field Raman detection under UHV conditions rather than cryogenic TERS performance.

In this work, we present our integration of a custom optical coupling system enabling Raman spectroscopy with a commercial LT-STM operating under UHV conditions. The optical layout was specifically adapted to the factory-defined 30° geometry while avoiding direct mechanical modification of the suspended STM head. We provide a quantitative comparison of the integrated Raman intensities of the first-order silicon phonon band obtained in the LT-STM and glove-box configurations. The comparison provides an end-to-end benchmark of the overall Raman response of the two experimental configurations under the stated measurement conditions. The presented system establishes an experimentally validated platform for far-field Raman detection under UHV conditions and a technical basis for subsequent low-temperature Raman and TERS development. Our team fully designed and constructed the external optical coupling and mechanical integration.

2. Methodology

The experimental platform was based on the third-generation LT-STM system manufactured by Scienta Omicron, which was integrated with a Raman spectroscopy unit to enable combined structural and optical characterisation under UHV conditions. While the internal optical elements and 30° geometry are manufacturer-defined, the entire external optical coupling, alignment, filtering, and mechanical stabilisation system was designed and constructed by our team. To minimise mechanical vibrations, the LT-STM system was installed in a vibration-isolated basement laboratory [26]. No systematic before-and-after vibration-spectrum or tunnelling-noise measurements were performed specifically to quantify the influence of the Raman optical retrofit on the STM noise floor. Figure 1 presents a photograph of the LT-STM system employed in this work. The LT-STM setup consists of three independently pumped chambers: load-lock, preparation, and analysis interconnected via UHV-compatible gate valves. This modular architecture enables sequential sample handling, surface preparation, and high-resolution measurements while maintaining strict vacuum integrity throughout the system. The load-lock chamber serves as the entry point for samples and measurement probes. It routinely reaches a base pressure of approximately 10^{-6} mbar. Importantly, it allows direct docking of a vacuum suitcase, facilitating sample transfer from external UHV systems under pressures of approximately 10^{-8} mbar. This capability minimises exposure to ambient contamination and helps preserve well-defined surface conditions during inter-system transfer. The preparation chamber operates at pressures better than 10^{-10} mbar, providing an environment suitable for advanced surface treatment procedures. It is equipped with multiple evaporation sources for thin-film deposition, as well as a sputtering gun for argon ion bombardment used in surface cleaning and modification. The analysis chamber operates at a pressure on the order of 10^{-11} mbar. A multi-axis manipulator enables precise positioning of the sample and incorporates both resistive heating and cooling functionalities, allowing controlled thermal processing prior to measurement. Factory-installed optical elements positioned in close proximity to the STM/AFM head enable *in situ* optical access to the sample under UHV conditions.

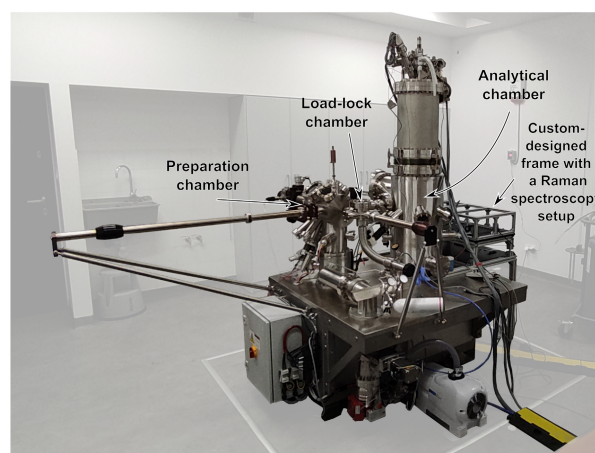


Fig. 1. Photograph of the third generation LT-STM installed in our laboratory.

Table 1.
Comparison of representative optical architectures integrating Raman spectroscopy or related optical techniques with UHV and LT-STM systems.

Reference/ system	STM platform	Optical architecture and geometry	Operating conditions	Experimentally demonstrated capability	Main distinction from the present work
Steidtner and Pettinger (2007) [19]	Custom UHV STM-TERS	High-NA parabolic mirror integrated between the STM scanner and sample, and used for both focusing and collection; NA = 1	UHV; operation in a controlled gas atmosphere	Raman and TERS of Si and dye layers; enhancement of 10^6 for dye adsorbates	High-NA excitation and collection, but requiring extensive integration of the optical system inside the UHV chamber
Kuhnke <i>et al.</i> (2010) [20]	Custom LT-UHV STM	Three independently selectable optical paths equipped with <i>in situ</i> adjustable aspheric lenses; NA = 0.42 per lens; optical axes inclined by 30° relative to the sample plane	4.2 K, UHV	Optical excitation, collection and imaging of the tunnel junction; characterisation of tip-induced plasmon emission, including angular- distribution and polarisation measurements	Three independently adjustable cryogenic optical paths inside UHV, compared with the single excitation- and-collection path used for Raman measurements in the present work
Jiang <i>et al.</i> (2012) [21]	Custom UHV STM-TERS	Laser-focusing and Raman- collection optics positioned externally to the UHV-STM	UHV	UHV-TERS of CuPc/Ag(111) enhancement factor of 7.1×10^5	System based on a custom UHV-STM platform
Sheng <i>et al.</i> (2018) [22]	Custom LT-UHV STM with molecular beam epitaxy (MBE)	Adjustable aspheric lens mounted inside the UHV chamber beside the STM head; side-illumination and backscattering collection geometry; NA not reported	Low temperature, UHV	<i>In situ</i> TERS of silicene/Ag(111); enhancement approx. 10^9 ; spatial resolution down to 0.5 nm	Fully custom LT-UHV STM-TERS-MBE platform with internal adjustable optics and experimentally demonstrated nanoscale TERS
Peis <i>et al.</i> (2023) [23]	Home-built LT-UHV STM-TERS	Off-axis parabolic mirror with a large working distance; NA $\approx$ 0.85; external control of the incident and detected light polarisation	15 K; UHV	Polarisation-dependent TERS measurements of vibrational modes in carbon nanotubes; volume-normalised gain factors of approximately 10^5 – 10^7	High-NA parabolic-mirror collection and polarisation control in a fully custom- built cryogenic UHV instrument; the present work uses the factory- installed lens geometry of a commercial LT-STM
Khelifi <i>et al.</i> (2023) [24]	Dedicated UHV Raman apparatus used in correlation with LT-STM	Fibre-optic Raman probe with a collection lens inserted directly into the UHV chamber; backscattering geometry; NA not reported	UHV; Raman measurements performed during surface preparation	<i>In situ</i> far-field Raman monitoring of the cleaning of III–V semiconductor surfaces, with the final surface reconstruction assessed by LT-STM	Similar far-field Raman scope, but the optical probe is inserted directly into UHV and Raman measurements are correlated with subsequent LT-STM characterisation rather than integrated with the LT-STM optical axis
Cirera <i>et al.</i> (2025) [25]	Commercial Omicron variable- temperature (VT) SPM upgraded for optical spectroscopy	Retractable focusing lens mounted inside the UHV chamber on a custom support and actuated using a wobble stick; NA = 0.45; external optical cage with camera and fibre coupling	UHV, variable- temperature and cryogenic operation	Electroluminescence, micro- Raman spectroscopy and TERS using Ag and Au tips; vibrational spectroscopy of C ₆₀ /Ag(111) with lateral resolution of approx. 2 nm	Closest retrofit analogue: an internal retractable lens was added to an existing commercial VT-SPM
Present work	Commercial Third Generation LT-STM (Scienta Omicron)	Factory-installed movable high-NA lens with cryogenic three-dimensional positioning, a manufacturer-specified NA greater than 0.45, a nominal axial travel of 2 mm, a nominal lateral travel of 2 mm $\times$ 2 mm and a 30° optical axis, combined with custom external excitation, filtering, inspection, collection and fibre-coupling modules	UHV; Si Raman at room temperature	Far-field Raman detection of the Si phonon band and an end-to-end comparison with a glove-box Raman configuration	The factory LT-STM optical access is retained, while all external Raman coupling and inspection modules were developed by the authors; no modification of the STM head or additional custom optics inside UHV was required

These components include: (a) a factory-installed high-numerical-aperture movable lens assembly designed for optical experiments, including low-temperature tip-enhanced Raman and luminescence spectroscopy. The lens assembly was mounted on the left-hand side of the LT-STM head at an angular position of 90° relative to the tip/sample transfer axis, with its optical axis inclined by 30° relative to the sample plane. According to the manufacturer's specifications for the delivered configuration, the lens had a numerical aperture greater than 0.45, enabling light collection over a relatively large solid angle. The lens was mounted on a cryogenic three-dimensional piezoelectric positioning system manufactured by SmarAct. The positioning system provided a nominal travel range of 2 mm along the optical axis and 2×2 mm in the sample plane, enabling precise alignment of the optical focus with the STM tip position under low-temperature and UHV conditions. The assembly also included the required control electronics. The lens-positioning mechanism can be operated under UHV conditions over the temperature range from 4.5 to 300 K; (b) a factory-installed static confocal lens assembly mounted on the right-hand side of the LT-STM head, opposite the movable high-NA lens, at an angular position of 270° relative to the tip/sample transfer axis. Its optical axis was inclined by 30° relative to the sample plane. The lens holder was rigidly attached to the STM head to minimise relative displacement between the lens and the tunnel junction, while the focal position could be adjusted *ex situ* by axial translation of the lens holder; (c) four optical flanges equipped with vacuum-compatible viewports, two of which are aligned with the 30° optical axes defined by the internal lens geometry, enabling efficient excitation and collection of Raman-scattered light; (d) the cryostat incorporated four optical ports equipped with exchangeable infrared-blocking quartz windows. These ports provided optical access to the microscope head while maintaining a manufacturer-specified base temperature below 5 K, provided that the infrared filters were inserted, the evaporation and tip/sample-transfer ports were closed, and the illumination source was switched off.

This optical configuration enables Raman scattering experiments directly within the UHV analysis chamber. The LT-STM platform allows cooling of the sample to liquid-nitrogen or liquid-helium temperatures, providing the hardware capability required for future temperature-dependent Raman measurements. The proximity of the STM tip to the optically accessible sample region provides the geometrical basis for future TERS experiments. Figure 2 schematically shows the optical configuration of the Raman scattering measurement setup integrated with the LT-STM system.

Next, we present the Raman spectroscopy unit integrated with the LT-STM system. The Raman spectroscopy unit is constructed around a 520 mm focal length imaging Czerny–Turner monochromator-spectrograph (MS5204i, SOL Instruments). The spectrograph is coupled to a thermoelectrically cooled HS 101H-2048/122-HR2 CCD camera (SOL Instruments), equipped with a Hamamatsu back-thinned FFT-CCD sensor and operated at -40°C to minimise dark current and improve signal-to-noise performance during long acquisition times. The entrance slit width of the monochromator-spectrograph was set to 100 μm for the Raman measurements presented in this work. The

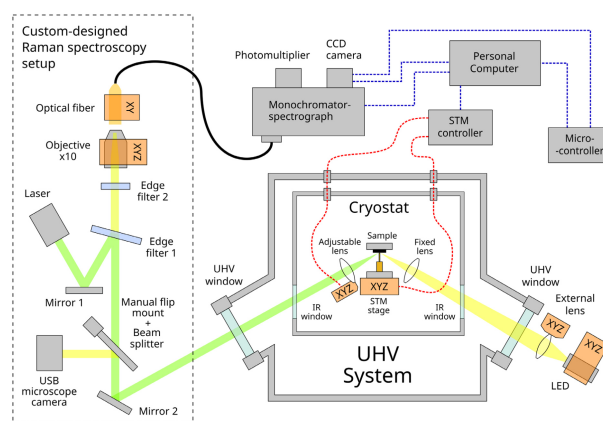


Fig. 2. Schematic diagram of the Raman scattering measurement setup integrated with the LT-STM system. Components designed and constructed by our team are enclosed by the dashed line.

monochromator-spectrograph incorporates a motorised four-position grating turret, allowing flexible spectral resolution and wavelength coverage. The installed diffraction gratings include 1800 lines·mm⁻¹, 600 lines·mm⁻¹, and 150 lines·mm⁻¹ gratings for variable spectral dispersion, as well as a 75 lines·mm⁻¹ Echelle grating optimised for excitation at 532 nm. This configuration enables both high-resolution and broadband spectral measurements, depending on experimental requirements. Raman excitation is provided by a diode-pumped solid-state laser operating at a wavelength of 532 nm. The excitation beam is conditioned within the Raman spectroscopy unit prior to delivery to the sample. The excitation power was adjusted using a variable neutral-density filter with an optical-density range of 0–3. A manually adjustable $2\times 10\times$ beam expander was used to control the beam diameter; detailed manufacturer specifications were not available. A Semrock LP03 532RE-25 RazorEdge® long-pass edge filter was used to suppress the 532 nm excitation line while transmitting the Stokes-shifted Raman signal. According to the manufacturer specifications at an angle of incidence of $0 \pm 2^\circ$, the filter provides an optical density greater than 6 at 532 nm and an average transmission greater than 93% over the wavelength range from 535.4 to 1200 nm. Its specified transition width is 2.7 nm, corresponding to 93.5 cm⁻¹. These manufacturer-specified spectral parameters refer to near-normal incidence; the spectral edge may shift when the filter is operated at an oblique angle. The filter has an outer diameter of 25 mm, a clear aperture of at least 22 mm, and a fused-silica substrate with a hard ion-beam-sputtered coating. It is mounted in a 3.5-mm-thick, black-anodised aluminium ring. The optical path also included two silver-coated steering mirrors (10D620ER.2, Newport) mounted in M1 mirror mounts (Newport), enabling precise adjustment of the excitation and collection paths within the constrained geometry of the UHV chamber.

All external optical components were mounted on an MB4575/M solid aluminium optical breadboard (Thorlabs) measuring $450 \times 750 \times 12.7$ mm and equipped with M6 tapped mounting holes. The breadboard was attached to a custom support frame constructed from modular structural profiles with a 40×40 mm cross-section. The frame is rigidly integrated with the table

supporting the LT-STM microscope. The present study did not systematically quantify the long-term stability of the optical alignment or the repeatability of the Raman signal intensity over multiple experimental sessions. Nevertheless, because optical alignment and fibre coupling are performed manually, readjustment may be required after sample exchange, mechanical intervention, prolonged interruption of operation, or thermal cycling. A quantitative evaluation of long-term stability will require monitoring the power-normalised Raman response of a reference sample over extended periods and following multiple independent realignment and thermal cycles.

Figure 3 shows the completed experimental setup. Raman excitation was provided by a continuous-wave 532 nm diode-pumped solid-state laser (Civillaser), operated using an ADR-1805 laser driver and temperature controller. The laser system had a maximum rated output power of 500 mW. The excitation beam was first reflected by the first silver-coated steering mirror (10D620ER.2, Newport), mounted in an M1 mirror mount (Newport), and then directed onto the primary Raman edge filter RazorEdge LP03-532RE-25.

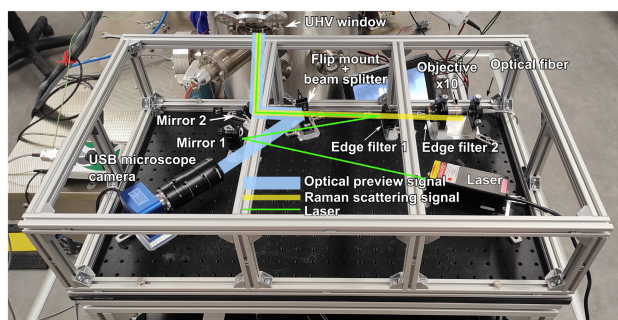


Fig. 3. Photograph of the Raman spectroscopy setup designed and constructed by our team and integrated with the UHV system.

During optical alignment and visual inspection, the 532 nm excitation beam reflected by the primary edge filter was subsequently directed towards a removable beam splitter positioned in the common excitation and imaging path. A UV-grade fused-silica plate beam splitter BSX10R (Thorlabs), measuring 25×36 mm and 1 mm in thickness, with a broadband coating specified for 400–700 nm and a nominal reflection-to-transmission ratio of R:T = 90:10, was installed in an RFM90 manual 90° flip mount (Thorlabs). Lowering the flip mount introduced the beam splitter into the optical path. In this configuration, approximately 10% of the incident 532 nm excitation beam was transmitted towards the sample, whereas approximately 90% of the returning optical light carrying the image of the STM tip-sample region was reflected towards the inspection camera. During alignment and visual inspection, the beam splitter therefore attenuated the excitation beam while directing most of the returning optical light towards the inspection camera.

With the beam splitter inserted, the sample surface and the STM tip could be visualized through the same factory-installed movable lens. The optical image was recorded using a DLT-Cam PRO 20 MP USB 3.0 microscope camera (Delta Optical) equipped with a Sony Exmor IMX183 CMOS sensor.

After optical alignment and visual inspection, the beam splitter was removed from the optical path before Raman acquisition. The Raman-scattered light collected by the same factory-installed movable lens was directed back through the UHV viewport towards the external collection path. After reflection from the second silver-coated steering mirror of the same type, the collected light was directed towards the primary edge filter. The residual 532 nm excitation radiation was reflected by this filter, whereas the Stokes-shifted Raman signal was transmitted towards the secondary clean-up edge filter (Iridian 532 LPF Nano Cutoff, 0.5" diameter, cutoff at 38 cm^{-1}), which provides additional suppression of the residual 532 nm excitation line. The Raman-scattered light collected from the sample was focused onto the optical fibre using a PLCN10 $\times$ /0.25 Plan Achromat objective (Olympus), with a numerical aperture of 0.25 and a working distance of 10.6 mm. The objective was mounted in an LP-05A five-axis lens positioner (Newport), which provided independent translation along the X, Y, and Z axes and angular adjustment about two orthogonal axes, enabling precise alignment of the objective relative to the fibre input. The 10-m-long fibre had a low-OH silica core with a diameter of 100 μm , a numerical aperture of 0.12, and a specified operating spectral range of 300–2200 nm. Its collection end was mounted in a stainless-steel cylindrical holder with an outer diameter of 10 mm and a length of 50 mm. The holder was installed in an LA1V-XY compact two-axis positioner (Newport), enabling independent adjustment of the fibre position along the X and Y axes relative to the coupling objective. The fibre transmitted the collected Raman signal to the entrance of the monochromator-spectrograph.

Figure 4(a) shows the standard view of the STM tip and sample surface obtained through the UHV window as provided by the STM system, while Figure 4(b) presents the image of the tip apex reflected from the silicon surface, captured using the optical preview through the movable lens. However, initial attempts to use conventional illumination through the same lens employed for Raman signal collection (as in the test configuration shown in Fig. 4) proved unsuccessful. The camera was saturated by intense light reflected from the first UHV window, which prevented proper visualisation. Consequently, unlike the preliminary measurement setup and the configuration developed for the glove-box system, the final design provided optical-preview illumination with a white LED source combined with a dedicated lens mounted in an SM1M10 lens tube (Thorlabs) on the opposite UHV flange. A single-crystal Si(100) wafer (Si-Mat) was used as

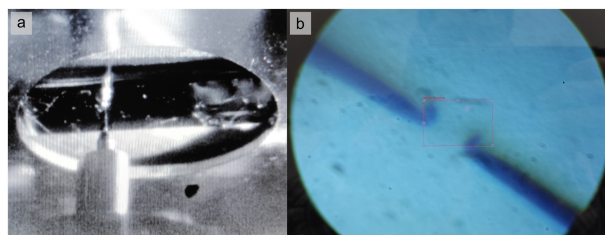


Fig. 4. (a) Optical view of the STM tip and sample surface through the UHV window. (b) Optical image of the STM tip apex reflected from the silicon surface using the implemented inspection setup.

the reference sample for the Raman measurements. Before quantitative analysis, the Raman-shift scale was calibrated against the first-order Si phonon band using the same procedure for both measurement configurations. Quantitative analysis was performed on the 100 individual acquisitions recorded for each configuration rather than on the averaged spectra shown in Fig. 5. For each acquisition, the position of the first-order Si Raman-band maximum was identified. A local linear baseline was fitted using two 20 cm^{-1} -wide spectral intervals located 40–60 cm^{-1} below and above the band maximum. After baseline subtraction, the Raman band was numerically integrated over a 60 cm^{-1} -wide interval centred on the band maximum using the trapezoidal rule. The same procedure was applied to all 100 LT-STM and 100 glove-box acquisitions. The mean integrated intensities were normalised to the excitation powers of 5.0 mW and 3.0 mW for the LT-STM and glove-box configurations, respectively. Short-term repeatability was evaluated using the mean integrated intensity, standard deviation (SD), and coefficient of variation (CV). The statistical uncertainty of the ratio of the power-normalised mean integrated intensities was evaluated by non-parametric bootstrap resampling. For each configuration, 100 individual acquisitions were resampled with replacement, and the power-normalised ratio was recalculated. This procedure was repeated 200 000 times, and the 95% confidence interval was defined by the 2.5th and 97.5th percentiles of the bootstrap distribution.

3. Results

Raman measurements were performed at room temperature using a single-crystal silicon reference sample. Figure 5 compares the spectra acquired in the LT-STM configuration

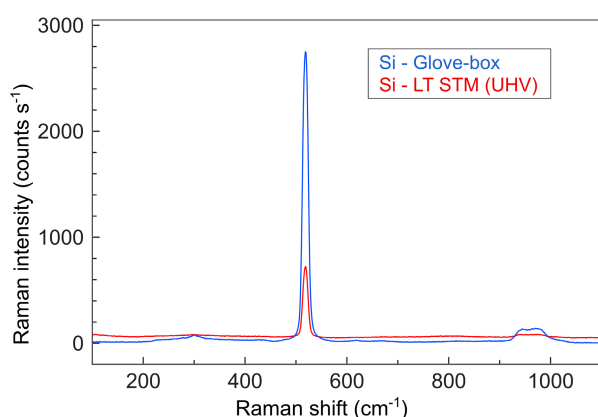


Fig. 5. Raman spectra of single-crystal silicon measured at room temperature in the glove-box and LT-STM configurations. Each spectrum represents the average of 100 individual acquisitions recorded with an acquisition time of 1 s using a 600 $\text{lines}\cdot\text{mm}^{-1}$ grating and an entrance slit width of 100 μm . The excitation powers were 5.0 mW and 3.0 mW for the LT-STM and glove-box configurations, respectively. The same detector dark-signal correction was applied to both datasets. The spectra shown were not normalised to the excitation power, and no additional baseline subtraction, vertical offset, or intensity scaling was applied. Quantitative integrated-intensity analysis was performed separately on the 100 individual acquisitions from each configuration using the baseline-subtraction and integration procedure described in the Methodology.

(red curve) and in the glove-box configuration (blue curve). Each spectrum represents the average of 100 individual acquisitions recorded with an acquisition time of 1 s using 600 $\text{lines}\cdot\text{mm}^{-1}$ grating and an entrance slit width of 100 μm . For quantitative comparison, the 100 individual acquisitions recorded for each configuration were analysed separately using the procedure described in the Methodology. The coefficient of variation of the integrated first-order Si Raman-band intensity was 2.13% for the LT-STM configuration and 0.86% for the glove-box configuration, indicating low short-term acquisition-to-acquisition variability of both measurement series. All 100 acquisitions from each configuration were retained in the statistical analysis. After normalisation of the mean integrated intensities to the corresponding excitation powers of 5.0 mW for the LT-STM configuration and 3.0 mW for the glove-box configuration, the Raman response obtained in the LT-STM configuration was lower than that obtained in the glove-box configuration by a factor of 7.40. Non-parametric bootstrap resampling yielded a 95% statistical confidence interval of 7.36–7.43 for this ratio. This interval reflects acquisition-to-acquisition variability only and does not include systematic uncertainty associated with the excitation-power measurements or other systematic differences between the two optical configurations. In both configurations, the excitation power was measured using a Sanwa LP1 laser power meter with a silicon-photodiode sensor, a specified wavelength range of 400–1100 nm, and a measurement range of 0.01 μW –39.99 mW. The power was measured immediately before the external UHV viewport in the LT-STM system and immediately before the quartz window in the glove-box system. Although the same detector dark-signal correction was applied to both datasets, the LT-STM spectrum exhibits a higher residual baseline than the glove-box spectrum. This difference may originate from configuration-dependent optical background, including residual scattering and reflections from the additional optical interfaces in the UHV optical path and differences in fibre-coupling conditions. Because these contributions were not measured separately, the origin of the residual background cannot be assigned unambiguously to a single mechanism. The lower Raman response in the LT-STM configuration likely reflects the combined influence of the optical geometry and the additional optical elements required for UHV operation. A likely contributing factor is the excitation and collection geometry. In the glove-box Raman system, the optical axis is oriented close to the surface normal, which provided efficient overlap between the excitation volume and the collection solid angle. In contrast, the LT-STM system operates with a fixed 30° optical axis imposed by the microscope internal optical design. This oblique geometry is expected to increase the projected excitation area and may therefore reduce the local excitation density. Additional losses may arise from the increased number of optical interfaces present in the UHV configuration. In the LT-STM setup, both the excitation beam and the Raman-scattered light must pass through a vacuum viewport mounted on the chamber, as well as an additional optical window located inside the cryostat. These elements are necessary to maintain vacuum integrity and suppress thermal radiation but may introduce additional transmission losses through reflection and absorption.

Further signal reduction can occur during coupling of the collected Raman radiation into the optical fibre connected to the spectrograph. Efficient fibre coupling requires precise alignment and mode matching, which becomes more challenging in the off-axis 30° geometry. The experimentally observed decrease in Raman intensity is therefore consistent with the combined influence of geometrical constraints, additional optical interfaces, and fibre-coupling conditions. Despite the lower Raman response, the first-order silicon phonon band is clearly resolved in the LT-STM configuration, demonstrating the feasibility of far-field Raman measurements through the optical path of the UHV LT-STM platform. Further optimisation of fibre coupling, including improved mode matching and focusing, could partially mitigate throughput losses. Anti-reflection-coated UHV viewports could additionally reduce Fresnel losses.

The present implementation has several operational and methodological limitations that should be considered when interpreting its performance. The optical geometry is defined by the factory-designed side access of the LT-STM, with the optical axis inclined by 30° relative to the sample plane. Compared with near-normal incidence, this oblique geometry is expected to produce an asymmetric excitation footprint and may distribute the optical power over a larger effective surface area, thereby reducing the local excitation density. It also restricts the available excitation and collection angles and limits independent optimisation of the illumination direction, collection efficiency, and polarisation relative to the STM tip and the crystallographic axes of the sample. The measured difference in the power-normalised integrated Raman intensity between the LT-STM and glove-box configurations should therefore be interpreted as an overall system-response ratio rather than as a measure of any individual loss mechanism. The comparison includes the combined effects of the different optical geometries, effective excitation areas, collection solid angles, transmission through the external UHV viewport and internal cryostat window, reflection and absorption losses in the optical components, spectral filtering, and coupling of the collected signal into the optical fibre. These contributions could not be separated quantitatively in the present study. Furthermore, the excitation power was measured immediately before the corresponding external window rather than at the sample position. The power-normalised Raman intensities therefore do not explicitly account for losses introduced by the viewports, cryostat windows, and internal optics. Accordingly, the reported comparison represents an end-to-end benchmark of the two experimental configurations under the stated measurement conditions rather than an absolute determination of the Raman collection efficiency at the sample. A further operational limitation is that optical alignment and fibre coupling are performed manually. Alignment requires adjustment of the internal movable lens, external steering mirrors, collection objective, and fibre position. Small changes in these components may affect the detected Raman intensity, and realignment may be required after sample exchange, mechanical intervention, prolonged interruption of operation or thermal cycling. Although the optical inspection system facilitates localisation of the STM tip and laser spot, the current configuration does not include

active beam-position stabilisation, automatic focus control or real-time feedback for maintaining the fibre-coupling efficiency. Long-term alignment stability and signal repeatability over multiple independent measurement sessions have not yet been systematically quantified. The present measurements demonstrate far-field Raman detection through the optical path of the UHV LT-STM using a single-crystal silicon reference sample at room temperature. Although the LT-STM platform is capable of cryogenic operation, this study did not investigate Raman measurements at cryogenic sample temperatures. Consequently, the performance and repeatability of the Raman configuration during cooling and warming cycles remain to be established. Finally, although the optical geometry allows the excitation beam to be focused near the STM tip apex and therefore provides the geometrical basis for future TERS experiments, the present study has not demonstrated tip-induced Raman enhancement. Further work will be required to demonstrate reproducible enhancement, determine the enhancement factor, evaluate the spatial resolution, and verify stable tunnelling operation under laser illumination. Despite these limitations, the present configuration establishes a functional optical pathway for far-field Raman measurements under UHV conditions. The side-access geometry, three-dimensional internal focusing capability, external filtering, and fibre-coupling modules provide a technical basis for subsequent low-temperature Raman and TERS development.

4. Conclusions

We demonstrate the integration of a Raman spectroscopy system with a commercial low-temperature scanning tunnelling microscope operating under UHV conditions. The system is based on a custom external optical coupling scheme adapted to the microscope fixed 30° optical geometry while avoiding direct mechanical modification of the suspended STM head. The optical layout enables excitation and collection of Raman-scattered light through UHV viewports and cryostat windows without modification of the internal STM head. A fibre-coupled spectrometer enables signal transfer despite the restricted optical access typical of commercial LT-STM platforms. The systems performance was verified using a single-crystal silicon sample and compared with near-normal-incidence Raman configuration operating in a glove-box environment at room temperature. Quantitative analysis of 100 individual acquisitions per configuration yielded coefficients of variation of 2.13% and 0.86% for the LT-STM and glove-box configurations, respectively, indicating low short-term measurement variability. After local baseline subtraction and normalisation to the excitation power, the mean integrated intensity of the first-order Si phonon band in the LT-STM configuration was lower than that obtained in the glove-box configuration by a factor of approximately 7.4. This difference represents the combined response of the two complete experimental configurations under the stated measurement conditions and should not be attributed to any individual optical loss mechanism. Nevertheless, the first-order silicon phonon band was clearly resolved in the LT-STM configuration. The developed platform establishes optical access for far-field Raman measurements under UHV conditions and

provides the technical basis for future correlated STM–Raman and TERS experiments at low temperature.

Authors' statement

Research concept and design, hardware development, M.P.; tests and validation, M.P., M.R., P.D., I.L., W.K., P.K.; data analysis, M.P., P.K.; writing the article, M.P., P.K.; critical revision of the article, P.K., P.J.K. All authors reviewed and approved the final version of the manuscript.

Competing interests

The authors declare no competing interests.

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