Investigating the probe-tip influence on imaging using scanning near-field optical microscopy: supplement

Panji Achmari,1,2 Arif M. Siddiquee,1,2,3 Guangyuan Si,4 Jiao Lin,5 Brian Abbey,1,2 and Shanshan Kou1,2,*

1Department of Chemistry and Physics, La Trobe Institute for Molecular Science (LIMS), La Trobe University, Victoria 3086, Australia
2The Centre of Excellence for Advanced Molecular Imaging, La Trobe University, Victoria 3086, Australia
3Department of Electronic Science, Xiamen University, Xiamen 361005, China
4Melbourne Centre for Nanofabrication, Victorian Node of the Australian National Fabrication Facility, Victoria 3168, Australia
5School of Engineering, RMIT University, Victoria 3000, Australia
*skou@latrobe.edu.au

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In this supplemental document, the results of the wavelength variation using the simulation are presented. In addition, the fitting results of the intensity profiles using pseudo-Voigt function are shown. These include the calculation of the resolution estimate of the system from the intensity gradient of the experimental intensity profiles.

1. THE INTENSITY PROFILES WITH DIFFERENT WAVELENGTHS

In our study, a monochromatic source with $\lambda = 640$ nm was used. The probe-tip influence in the SNOM imaging system was investigated using this wavelength in both the experiments and in simulations. The FDTD simulations were used to investigate how changing the wavelength modifies the expected intensity profile. The range of the wavelengths tested was from 500 nm to 640 nm. The probe parameters: $D = 300$ nm and $h = 10$ nm were used, identical to the parameters used for the simulation with $\lambda = 640$ nm presented in the main manuscript. The simulations were carried out for both single-slit and double-slit apertures. The resulting simulated intensity profiles are shown in Fig. S1.

The simulated profiles in Fig. S1 show only slight variations in the profile shape as the wavelength is varied from 640 nm to 500 nm. The oscillations in the intensity profile at the edges of the slits, that appear in the simulations without the probe-tip, become smoother at shorter wavelengths. The rapid decay in the intensity profile at the edges of the slits, however, remains unchanged. The simulated profiles with the probe-tip included also show minor variations as a function of wavelength. The quantitative values for FWHM etc. extracted from the simulated profiles of the single and double-slit apertures are summarized in Tables S1 and S2.

Table S1. Summary of simulated intensity profiles for the single-slit aperture with $w = 180$ nm for 5 different incident wavelengths. The relative intensity amplitudes were normalized such that the integral over the distance ($x$) (i.e. area under the profile) are the same for all profiles. FWHM is calculated from the pseudo-Voigt fitting.

| $\lambda$ (nm) | relative intensity amplitude without probe | relative intensity amplitude with probe | FWHM (nm) with probe |
|---------------|------------------------------------------|---------------------------------------|----------------------|
| 500           | 0.85                                     | 1.63                                  | 276                  |
| 530           | 0.87                                     | 1.57                                  | 271                  |
| 550           | 0.88                                     | 1.61                                  | 270                  |
| 600           | 0.82                                     | 1.51                                  | 277                  |
| 640           | 1.00                                     | 1.45                                  | 252                  |

In the range of the wavelengths tested (500 nm to 640 nm), the FWHM varies within 25 nm of the value obtained at $\lambda = 640$ nm for both single-slit and double-slit apertures. The value for the FWHM does not appear to decrease significantly at shorter wavelengths. In addition, the relative maximum intensity of the profiles does not show significant differences as a function of wavelength. For the double-slit apertures, the peak-to-peak distances are consistent across the wavelength range.

Although our current experiment study does not include a tuneable source, our simulations indicate that the probe-tip influence on SNOM image formation is relatively insensitive to changes in the visible incident wavelength over the visible range from 500 nm to 640 nm. Based on these
Fig. S1. Simulated intensity profiles across a single-slit aperture with \( w = 180 \text{ nm} \) and double-slit apertures with \( w = 470 \text{ nm} \) and \( s = 140 \text{ nm} \). The results from five incident wavelengths between 500 nm and 640 nm are shown with the probe-tip included (solid lines) and without the probe-tip included (dashed lines). The yellow shaded areas indicate the physical extent of the slit(s). The profiles were normalized such that the integral over the distance \( (x) \) (i.e. area under the profile) was the same in each case.

Table S2. Summary of simulated intensity profiles for the double-slit apertures with \( w = 470 \text{ nm} \) and \( s = 140 \text{ nm} \), for 5 different incident wavelengths. The relative intensity amplitudes were normalized such that the integral over the distance \( (x) \) (i.e. area under the profile) are the same for all profiles. FWHM and peak-to-peak distance are calculated from pseudo-Voigt fitting.

| \( \lambda \) (nm) | relative intensity amplitude | FWHM (nm) | peak-to-peak (nm) |
|-------------------|-----------------------------|-----------|-------------------|
|                   | without probe | with probe | with probe | with probe |
| 500               | 0.95            | 1.09       | 406          | 627        |
| 530               | 0.96            | 1.05       | 407          | 627        |
| 550               | 0.99            | 1.05       | 395          | 628        |
| 600               | 0.98            | 1.17       | 418          | 631        |
| 640               | 1.00            | 1.08       | 422          | 628        |

simulations we conclude that the result presented in the manuscript for \( \lambda = 640 \text{ nm} \) provides a good representation of the key characteristics of the influence of the probe-tip on SNOM image formation.
2. FITTING THE INTENSITY PROFILES USING THE PSEUDO-VOIGT FUNCTION

A. Fitting the experimental and simulated data

A comparison between the experimental and simulated intensity profiles is given in the manuscript, with the quantitative analysis of the profiles summarized in Tables 2 and 3. The results of fitting these profiles using the pseudo-Voigt function to extract the quantitative parameters (e.g. peak FWHM) are shown in Fig. S2. The results include the double-slit apertures with $s = 80$ nm which could not be resolved, and hence are omitted from Table 3. We find that the fitted curves are able to accurately reproduce the shape of the intensity profiles for both the experimental and the simulated datasets.

![Fig. S2.](image)

B. Estimating the resolution of the SNOM system

Figure S3 shows the data used for estimating the resolution of SNOM. To estimate the resolution, we use the experimental intensity profile from single-slit apertures with $w = 150$ nm and 180 nm (solid-blue line). We calculated the intensity gradient ($dI/dx$) and found two peaks corresponding to the edges of the slit and took the absolute values of the curves. Because the peaks are asymmetric, the left and right parts of each peak (with respect to its maximum) are fitted with different FWHM values, and the combined asymmetric FWHM values are calculated and shown in the figure. These two peaks are fitted separately, which effectively represent the estimate resolution from the left and right edges of the slits. The average FWHM from the calculation is 185 nm, which we mentioned in the manuscript as the resolution estimate of the SNOM system used in this study.

Figure S3 shows example data used to estimate the spatial resolution of the SNOM images. To determine the resolution, we used the experimental intensity profiles from the single-slit apertures with $w = 150$ nm and 180 nm (solid-blue line). We then calculated the intensity gradient ($dI/dx$) corresponding to these two single-slit apertures. Because the single-slit transmission peaks are asymmetric, the two peaks in the derivative of the intensity profile have slightly different FWHM values as shown in Fig. S3. The final image resolution, estimated by taking the average of these FWHM values obtained from the derivative of the experimental data, was 178 nm ± 14 nm.
Fig. S3. Estimating the spatial resolution of the SNOM using the derivative of the intensity profiles to determine the FWHM associated with the sharp edges of the slits. The intensity profiles are taken from single-slit apertures of width of $w = 150$ nm and $w = 180$ nm.