



1,600 1,800 2,000 2,200 2,400 Distance, d (pm)

Incident laser power, I, (mW)

 $I_{TERS} \propto I_0^n$

n=1.2(1)

G-band

TERS intensity (a. u.)

2.2 nm

STM-controlled TERS spectra of isolated single CNTs on Ag(111) using Ag tips were investigated. The selection of Ag as both tip and substrate materials is found to produce a large TERS enhancement factor up to about 108, originating from the strong enhancement of gap-mode plasmons.

The highly confined plasmonic field in the nanogap also drives the spatial resolution of TERS imaging on a CNT down to about 0.7 nm. Such subnanometer resolution allows us not only to visualize directly the spatial extent of defect-induced D-band scattering in real space, but also to fine track the spectral evolution of the G-band with the strain variations due to the bending and the local environment.

Furthermore, the ability to resolve the TERS spectral difference between the inner and the outer sides of a bent CNT helps to provide insights into the origin of G-band. To put it in perspective, our results demonstrate that TERS can go beyond chemical identification and serve as a powerful tool to investigate the defect and strain in low dimensional nanostructures and materials at the subnanometer scale.

