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# NONLINEAR OPTICS

## Laser Field Enhancement at the Scanning Tunneling Microscope Junction Measured by Optical Rectification

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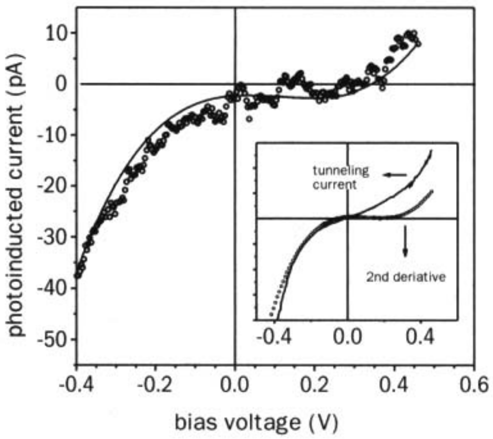
Optical field enhancement due to resonances in nanostructures is a well known phenomenon that has given rise to the development of techniques such as surface enhanced Raman spectroscopy[1] and surface modifications in the nanometric range, among others.[2] Many theoretical works[3] have predicted enhancement factors strongly dependent on the shape of the structure and the optical properties of the nanostructured material, magnitudes that are poorly known.

In a recent letter, we have been able to perform the direct measurement of the magnitude of the field enhancement by means of determining the rectified current induced at the tip of a scanning tunneling microscope (STM) junction.[4] The sharp metallic tip of the microscope acts as the enhancement structure, creating a very small region of a strong electric field. When the sample is brought to the tip, the enhanced field biases the tip-sample junction at the optical frequency giving rise to an electrical current. The nonlinear current-voltage response of the STM junction yields a term proportional to the electric field squared (optical rectification), which has a net DC contribution that can be measured with the instrument amplifier. The simultaneous measurement of both the DC bias current and the optical rectified contribution allows the determination of the voltage drop induced by the optical field across the tip-sample region. In Figure 1, the optically induced current is shown as a function of the biasing voltage, and fitted (solid line) with the second derivative of the DC current. Thermal effects due to the expansion of the sample heated by the laser were also considered.[5] As a guide, the inset in the figure shows the measured DC current and its calculated second derivative. The value of the enhanced field can be extracted from our knowledge of the tip sample distance. The magnitude of the field enhancement achieved for the particular case of the figure is 450, and depends on the particular tip used. Similar values are obtained for highly oriented pyrollitic graphite, indicating that the enhancement is mainly due to the tip.



**Bragas Figure 1.** Photoinduced tunneling current as a function of the STM tip-sample biasing voltage. Dots are measured values and the full line is the fit corresponding to the optical rectification contribution. The inset shows the DC contribution and its second derivative used to fit the data.

This large field enhancement in a very confined region should find applications in diverse fields such as surface nanomodification, particle trapping, field enhanced optical microscopy, nanoscaled nonlinear optics, and testing models for the linear and nonlinear optical response of nanostructures. The possibility of measuring a particular field enhancement for a given structure is a relevant step toward the mentioned applications.

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## Suppression of Multiphoton Fluorescence in Hyper-Rayleigh Scattering

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Hyper-Rayleigh scattering (HRS) has become widely accepted as an experimental technique for the determination of the first hyperpolarizability (second-order nonlinear polarizability), $\beta$, of molecules in solution. Apart from being simpler both theoretically and experimentally than electric-field-induced second-harmonic generation (EFISHG)—applicable to neutral, dipolar molecules only—HRS is the sole technique that gives a $\beta$ value for ionic or octopolar species. The combination of HRS and EFISHG also allows different elements of the hyperpolarizability tensor to be analyzed.

However, because of the incoherent nature of HRS, no discrimination is possible against multiphoton fluorescence at exactly the second-harmonic wavelength. We have now implemented a technique that does just that, enabling the measurement of ionic or octopolar, as well as fluorescent species.