ELECTROMAGNETIC FIELD ENHANCEMENT AND LIGHT CONFINEMENT ON THE NANOSCALE

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ABSTRACT

Electromagnetic field enhancement and light localisation associated with small metallic particles or roughness of a metal surface are closely related phenomena that play an important role in linear and especially nonlinear optical processes. Employing electromagnetic field enhancement and nonlinear optical effects such as second-harmonic generation, strongly localised nanoscopic light sources can be achieved which are needed in scanning near-field microscopy for imaging as well as materials modification on the nanoscale.

INTRODUCTION

Nanoscale light sources are widely used nowadays in scanning near-field optical microscopy (SNOM) and have promising applications in nano-photonics, data storage and processing, materials characterisation, etc. A traditional way to achieve such a confined light source is to use a small size aperture at the end of a sharpened optical fibre [1]. More recently, however, another approach is attracting much attention based on apertureless nanoscopic scattering, linear and two-photon fluorescence, or second-harmonic generation (SHG) [2-8]. The nanoscopic volume of fluorescent material has initially been proposed for achieving a localised light source of the wavelength different from the excitation wavelength followed by the implementation of a single molecule as a light source [3,4]. Nonlinear optical processes enable even better light confinement providing a light source the electromagnetic field of which drops faster than $1/r^3$ with a distance from a source [5,7,8].

Light confinement and the electromagnetic field enhancement are closely related to each other. It is the confinement that leads to a strong field enhancement due to small volume of the related electromagnetic modes. Electromagnetic field enhancement can be related to localised surface plasmon (LSP) excitation as well as topological (lightning-rod) effects. With the development of scanning near-field optical microscopy which allows probing the electromagnetic field close to a surface with subwavelength resolution, the studies of these effects have been raised to a qualitatively new level. SNOM measurements can provide direct correlation of a surface topography to the related electromagnetic field distribution as both are imaged simultaneously.

Here we would like to discuss electromagnetic field enhancement effects and related light confinement on an example of near-field second-harmonic generation. First, SNOM studies of second-harmonic generation from rough metal surfaces will be discussed. Next, the applications of the tip-induced second-harmonic generation in novel type of apertureless SNOM will be presented.

LOCALISED SURFACE PLASMONS

Localised surface plasmon is a collective electron plasma excitation on a curved metal surface that can be excited by electromagnetic wave and in turn, decays with light emission [9,10]. Localised surface plasmons are associated with small metallic particles or roughness of a metal surface. Strong field enhancement is also expected in narrow gaps between nanoparticles when the electromagnetic near-field interaction modifies plasma resonances of the individual nanoparticles resulting in the surface plasmon localisation in the gap between the particles [11,12]. The localised surface plasmons have been successfully employed for describing surface enhanced Raman scattering, surface enhanced second-harmonic generation, photon emission from a tunnelling junction of an electron scanning tunnelling microscope, giant enhancement of the electromagnetic field and related optical processes in self-affine structures and fractals, etc. [11-13]. This kind of localised excitation can not be visualised by conventional optical techniques as their electromagnetic field is confined close to the surface, but they can be addressed with SNOM [14-19].

For example, surface plasmons localised in a pit on a thin metal film result in the enhancement and confinement of SHG at the pit (Fig. 1) [16-18]. The SH image looks almost like a ‘negative’ of the topographical image. The SH signal

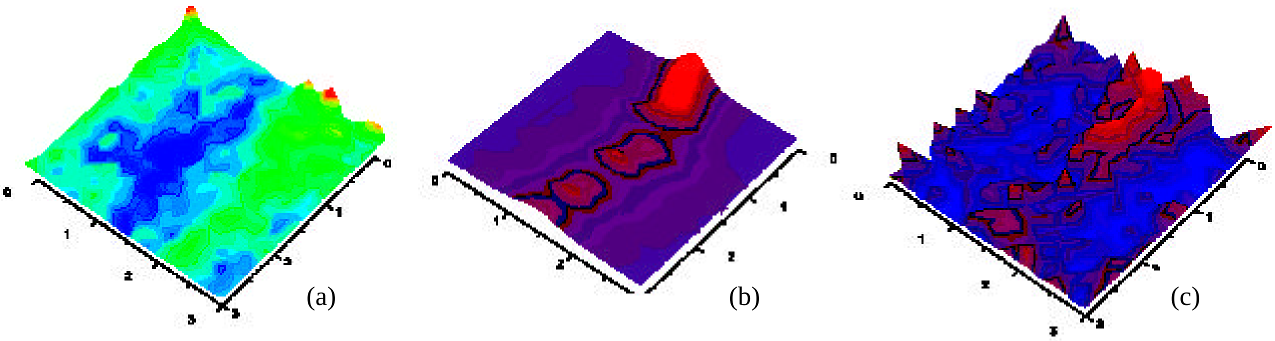

Fig. 1. Topography of the silver film (a), calculated in the model of localised surface plasmons (b) and measured (c) second-harmonic intensity distribution over the surface. Excitation light wavelength is 1064 nm.

measured in the flat regions is smaller than the signal measured near the pit. An increase of the pit width results in a decrease of the observed SH intensity as predicted by the LSP model [18].

TOPOLOGICAL FIELD ENHANCEMENT: LIGHTNING-ROD EFFECT

Even if localised surface plasmons cannot be excited, a lightning-rod effect, a pure electrostatic effect, can result in the significant field enhancement at the surfaces of large curvature [9,10,19]. Under such conditions (no resonant LSP excitation), a local enhancement of the SH intensity up to 10 times at the defects of micron-size lateral dimensions has been observed at the rough gold films while the average SHG enhancement is only of about 1.2 times (Fig. 2) [19]. The enhancement occurs at the conical defects if the excitation light has a component of the electric field parallel to the cone axis.

APERTURELESS SECOND-HARNONIC SNOM

The effect of the SHG enhancement at the surface features discussed above can be turned around and used to perform a novel type of apertureless near-field optical microscopy. A sharp metal tip can be used to achieve a localised second-harmonic light source (cf. Fig. 2). Owing to the field enhancement at the extremity of a probe tip, main source of SH light is situated either at the apex of a tip or at the surface region just under the tip in the case if surface nonlinearity dominates [7,8]. If metallic tip and/or metal surface are used which support surface plasmons, the localised surface plasmons related to the tip-surface junction can lead to additional resonant field enhancement and play a role in the determining of the SNOM resolution which is limited by the LSP localisation range in this case. In addition to a background free detection, this technique provides a much better spatial confinement of the signal field than possible to achieve in conventional scattering or fluorescent apertureless SNOM.

Surface SHG Enhanced by a Metal Tip

In the first experiments, both metal tip and metal film were used [8]. The gold surface exhibits random topographic structure (Fig. 3a). The lateral distribution of second-harmonic intensity induced with a silver tip under the excitation with light having electric field component parallel to the tip axis reveals complex behaviour depending on the surface defect structure but in general reflects the local topography (Fig. 3b). Lateral variation of the SH signal is related to the different degree of the electromagnetic field enhancement due to the interaction between the tip and surface defects. At the places of the film where the defect structure is simple, the SH images show good correlation with the field

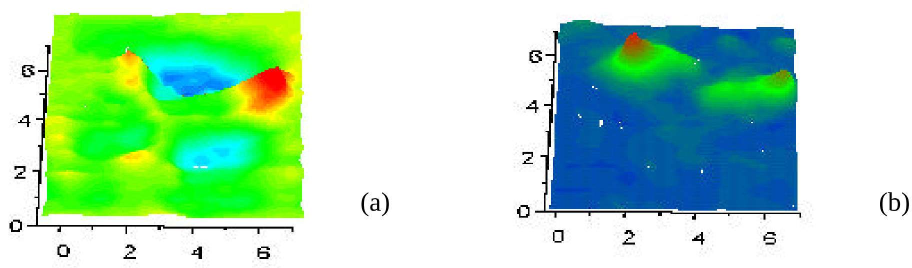

Fig. 2. Topography (a) and corresponding SH image (b) of the rough gold film. Excitation light wavelength is 780 nm.

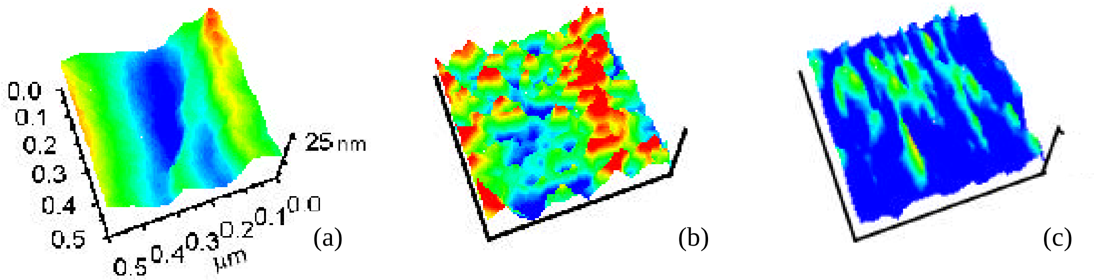

Fig. 3. Topography (a) of the gold film and second-harmonic intensity distributions (b and c) recorded using p- and s-polarised excitation light (λ = 780 nm) with the silver probe tip.

enhancement at the individual defects. At the regions with a complex defect structure, the SH image reflects not individual surface features but the local field distribution in the system of interacting defects. Although localised surface plasmons have not been especially excited in these experiments, at a rough surface where features of all sizes and shapes are present it is always possible to find a defect (or ensemble of defects) and tip position for which localised plasmon can be excited at the tip-surface junction at the excitation or SH frequency. They are the defects that dominate in the recorded SH images. The field enhancement depends dramatically on the geometry, size, and mutual position of the tip and surface defects. Slight variations of the defect shape may result in the significant variations of the enhancement and, as a result, the observed SH signal. By changing polarisation of the excitation light or tip-surface distance, the interaction between the tip and surface and therefore resulting SH intensity can be modified. The contrast in the SH images disappears under the excitation with light having no electric field component parallel to the tip axis (Fig. 3c) or when the tip is retracted in a far-field region.

SHG from a Metal Tip Apex

Polarisation and distance dependencies of second-harmonic generation at a gold tip apex have been studied in the context of applications as a nanoscopic light source in apertureless SNOM [20]. The results shows that, in general, different topological features of a tip are responsible for scattering and second-harmonic generation in the far- and near-field. The near-field interaction between a probe tip and a surface significantly modifies the electromagnetic field enhancement at the tip resulting in the different distance dependencies of near-field scattering and second-harmonic generation. Polarisation contrast obtained with second-harmonic signal significantly exceeds polarisation contrast of linear scattering from the tip. The near-field SHG at a metal tip apex is a relatively easy way (standard STM tip can be used) to achieve a reliable (no bleaching) and strongly confined (due to the field enhancement effects) light source close to the sample surface needed in numerous applications. The second-harmonic light is strongly confined to the tip-surface junction due to the field enhancement effects related to localised surface plasmons and/or lightning-rod effect.

CONCLUSION

Light localisation effects due electromagnetic field enhancement are important from both fundamental point of view and numerous potential applications. Nanoscopic light sources are required for imaging and characterisation of materials and devices on the nanoscale. They can also open up the possibility for local optical and spectroscopic studies of surfaces, surface defects and adsorbates. Field enhancement effects are especially advantageous for investigations of nonlinear optical materials and engineering of nanoscale nonlinear optical devices. Fabrication, characterisation and applications of nanostructured materials and based on them devices also require our ability to manipulate strongly localised electromagnetic fields. These can be used for both nanolithographic fabrication of nanostructures as well as for optical and magneto-optical high-density data storage. Closely related to this is photo-chemistry and photobiology on the nanoscale. Using localised light sources, the photochemical and photobiological trasformation can be induced locally on the level down to individual molecules. Scanning near-field microscopy technique would allow one to image the overall area under investigation, identify the regions of interests, and finally perform optically induced transformations in these specifically chosen regions.

A quest for high-speed all-optical computing and telecommunications has led to the development of a series of optical devices to replace slow electronic transistors, diodes, etc. In most of such devices the electromagnetic field enhancement effects plays determining role and makes the operation of the devices possible via induced (and subsequently enhanced) nonlinear optical processes. The striking example of such phenomenon is recently observed single-photon tunnelling via localised surface plasmon modes [21]. Nanoscale photonic devices and all-optical integrated circuits are now within a reach.

In addition to numerous applications, nano-local light sources provide a tool for the investigations of rich physics in the optical near-field where a number of effects related to multipolar processes and quantum electrodynamics manifestation takes place resulting in the changes in lifetime of excited states, nonlinear susceptibility changes, nonlocal effects, etc.

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