FEMTOSECOND MEASUREMENTS COMBINED WITH NEAR-FIELD OPTICAL MICROSCOPY

Artyom A. Astafiev,
Semyonov Institute of Chemical Physics, Moscow, Russian Federation.

Keywords: diffraction limit near-field scanning optical microscopy (NSOM or SNOM), aperture NSOM, pump-probe method, Bethe-Bowkamp equations, GVD precompensation, quantum well, quantum wire, Schottky contact, surface plasmons.

Contents

Summary

This chapter presents an overview of femtosecond near-field optical microscopy (or femtosecond (ultrafast) NSOM) – an experimental method combining pump-probe femtosecond spectroscopy with spatial resolution capabilities of the near-field imaging. Near-field microscopy, which appeared in 1980s, allows achieving spatial resolution lying below the diffraction limit using a miniature tip to probe optical properties of the sample. Its combination with femtosecond technique gives an experimental tool providing simultaneously ultrahigh time and spatial resolution. Such a tool is extremely promising in studies of charge carrier dynamics and relaxation in nanostructured metal or semiconductor systems. However, in application of ultrafast NSOM several issues must be taken into account. The most important are a proper choice of experimental configuration must be made, an improvement of signal/noise ratio to a level allowing measuring realistic signals, and a control on the length of laser pulses to realize a high time resolution. Technical complexity of ultrafast NSOM is a reason of limited applications of this technique. Still beginning from late 1990s a number of research groups managed to use femtosecond NSOM for studying the dynamics of ultrafast processes in mesoscopic systems, such as single nanostructures, single metallic nanoparticles etc. A few examples of such investigations are presented in this article. Thus, ultrafast NSOM proved to be a viable instrument for characterization of nanoscale systems with resolution of the order of 100 fs in time and 100 nm in space. With the improvements of the NSOM resolution and signal to noise ratio, the femtosecond NSOM will widely spread in the field of ultrafast nano-optics.

1. Introduction

Space and time resolved measurements are of crucial importance in experimental physics. The recent interest to mesoscopic systems and their applications in electronics and nanomaterials brought about the need to characterize ultrafast (i.e. picosecond or nanosecond) physical processes in systems of nanometer-scale size. Examples of such processes include charge carriers transport in the bands of semiconductors and electron dynamics in metals. Usually dynamics of these physical processes are studied with ultrafast laser spectroscopy. However, in most experiments involving methods of laser spectroscopy, the information about the dynamic processes is specially averaged over the area of the laser beam. This averaging precludes from resolving inhomogeneities of nanostructured systems. Hence in study of such systems spectroscopic methods with high spatial resolution are essential. Moreover, space and time for small-scale systems are interrelated, since e.g. for small-sized systems carrier charge transport occurs in short times: in one femtosecond an electron moves over a distance on the order of nanometer. Therefore it is desirable to achieve high spatial and temporal resolution simultaneously, the latter being on femtosecond timescale.

A combination of optical microscopy with time-resolved methods looks an extremely promising tool for solving the task described above and offers a possibility of studying dynamics of physical and chemical processes with sub-micrometer spatial resolution. An important obstacle is that conventional optical microscopes have a spatial resolution limited by so-called diffraction limit. The Rayleigh criterion gives the following expression for the smallest distance between two neighboring points which can be resolved: $\Delta r = 0.61\lambda / NA$, where NA is a numeric aperture of the objective and λ is the wavelength used. Even for oil-immersion objectives NA is usually smaller than 1.5, therefore the resolution limit can be taken approximately equal to $\lambda / 2$. In the visible range it corresponds to 200-400 nm, thus the resolution of conventional microscopes does not suffice to spatially resolve inhomogeneities in nanometer-scale structures. However, the resolution limit can be circumvented using so-called near-field scanning optical microscopy (NSOM or SNOM) - a new branch of optical microscopy technique. A characteristic feature of the NSOM is a probe, creating a highly localized light source, which is placed in close proximity to the sample. In the most common aperture NSOM, the probe is a tapered optical fiber covered with a layer of metal (usually aluminum) so that on the very its tip an uncovered aperture remains. Light coupled to the fiber passes through the aperture, illuminating the sample. If the sample is within a small distance from the probe (in the near-field zone), then the divergence of light can be neglected and the illuminated spot on the sample will have the same size as the probe's aperture. The same holds true, if the probe is used to collect light emitted or passing through the sample, only the sample area located right under the probe gives significant contribution to collected signal. Thus, the spatial resolution of the NSOM is determined by the aperture diameter, which can be made of nanometric size, although there are additional factors that limit resolution, such as the penetration of light into the metal screen (skin-effect). In practice the typical resolution of aperture SNOM is from 50 nm and higher. By scanning the probe across the sample surface one can obtain a super-resolution image, an additional advantage being availability of information on sample's topographic structure. Although, NSOM has a number of disadvantages compared with conventional optical microscopes, such as complexity of probe production and its low

lifetime, a need in complex setup in maintain sample-to-probe distance and low throughput of the probe's aperture, it is still an efficient instrument for obtaining a subwavelength spatial resolution.

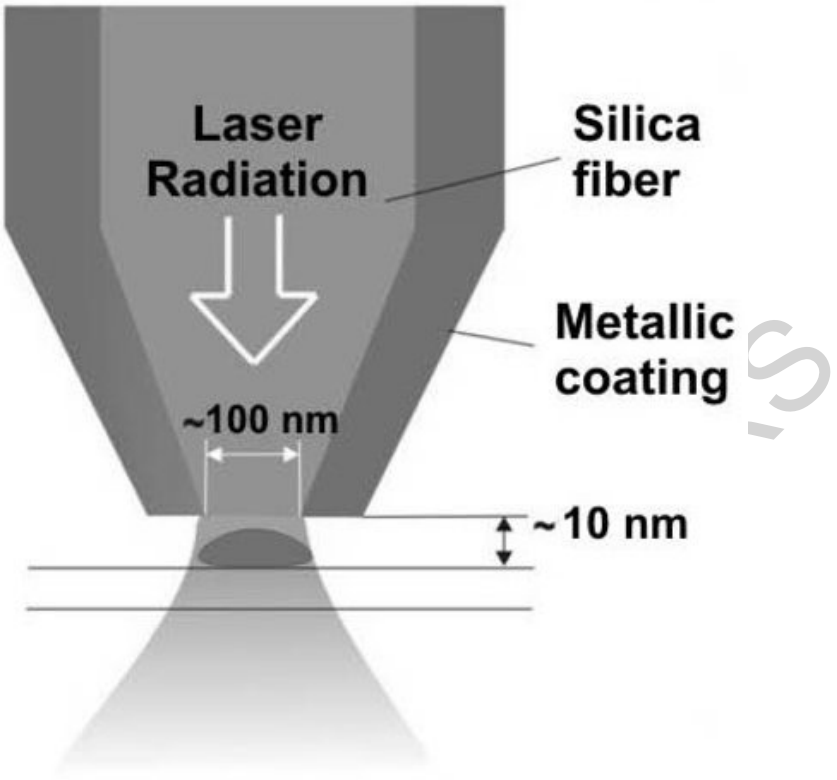

Figure 1. Schematic representation of the NSOM aperture probe

Thus, the near-field scanning optical microscopy is the technique, providing the highest achievable optical resolution, and its combination with femtosecond laser technique promises the best tool for studies of ultrafast processes on nanoscale. Several ways of such a combination can be conceived.

One possible method involves time-counting electronic devices for time-correlating photon measurement and was successfully applied for measuring lifetime of excited states of fluorescent molecules. The technical realization includes a time-to-amplitude converter (TAC) which is started by a laser impulse and stopped by a signal from a fluorescence detector registering photons emitting by a studied systems, so that the time span between the events of laser excitation and photon emission is measured. Accumulating necessary statistics one can obtain a number of emitted photons depending on time passed after the excitation and build a decay curve. However, the responding speed of electronics limits the temporal resolution of this method to a picosecond range, which is enough for measuring fluorescence decay, occurring in molecular systems within nanoseconds, but not sufficient for studying faster processes, e.g. carrier dynamics in semiconductors or electronic states dynamics in photoexcited molecular systems.

Far more promising for achieving ultrahigh temporal resolution is a combination of optical microscopy with femtosecond pump-probe method, in which optical response is measured as a function of time delay between the pump pulse exciting the researched system and the probe pulse. In this case the available temporal resolution is limited by the laser pulse duration, nowadays researchers routinely working in a range of tens of femtoseconds. The idea of combining near-field optical microscope with femtosecond spectroscopy methods was first proposed by A. Lewis *et al.* in 1995, the same authors discussed possible ways of experimental realization of this idea and demonstrated the feasibility of coupling femtosecond laser pulses to the near-field optical probe. However, thus far a limited number of groups managed to implement such a combination. This fact should be attributed to complexity of experimental arrangement and the need to resolve technical issues, aspects of which are considered below.

-

-
-

Bibliography

Review articles:

Dunn R.C. (1999) Near-field scanning optical microscopy. *Chem.Rev*., 99, 2891-2927

Higgins D.A., Mei E. (2001) *Near-Field Scanning Optical Microscopy*, in *Scanning Tunneling Microscopy and Spectroscopy*, D.Bonnel ed., Wiley, 371-420 [These two works are general reviews of near-field optical microscopy, its principal features, experimental methods and areas of application]

Li Zhi., Zhang JiaSen, Yang Jing and Gogn QiHuang (2007) Ultrahigh spatiotemporal resolved spectroscopy. *Science in China Series G: Physics, Mechanics& Astronomy*, Vol. 50, 6, 681-690 [A review of progress in ultrafast NSOM by the year of 2007, the principal experimental application are described and issues in femtosecond NSOM are analyzed. The article also contains a useful collection of references to works involving ultrafast NSOM published by 2007]

Articles:

Achermann M., Morier-Genoud F., Seifert W., Wernersson L.-E., Siegner U., Keller U. (2002) Carrier and field dynamics around nanoscale Schottky contacts investigated by ultrafast near-field optics *Phys. Rev.* B, Vol. 65, 045322, 1-9. [The article presents detailed results of studies of Schottky contact embedded in GaAs with femtosecond NSOM in comparison with theoretic model.]

Imura K., Nagahara T., Okamoto H. (2004) Imaging of surface plasmons and ultrafast dynamics in gold nanorods by near-field microscopy. *J. Phys Chem* B, 108, 16344-1647 [The article presents the result of near-filed imaging of local density of states in a single gold nanorod and their temporal dynamics following photo-excitation]

Lewis A., Ben-Ami U., Kuck N., Fish G, Diamant D., Lubovsky L., Lieberman K., Katz S., Saar A., Roth M.(1995) NSOM the fourth dimension: Integration nanometric spatial and femtosecond time resolution.

Scanning, Vol. 17, 3-13 [The pioneering work which first proposed a combination of femtosecond spectroscopy and NCOM, possible experimental configurations are discussed]

Nechay B.A., Siegner U., Achermann M., Bielefeldt H., Keller U. (1999) Femtosecond pump-probe near-field optical microscopy. *Review of Scientific Instruments*, Vol. 70, No. 6, 2758-2764 [This is one the first work in which successful implementation of femtosecond NSOM was reported, a detailed description of experimental setup and issues involved is given]

Siegner U., Achermann B., Keller U. (2001) Spatially resolved femtosecond spectroscopy beyond the diffractions limit. *Measurement Science and Technology*, 12, 1847-1857 [This article describes applications of femtosecond NSOM in study of charge carriers dynamics in semiconductor nanostructures (locally patterned quantum well, single quantum wire, nanoscale Schottky contacts) and the principal results obtained are discussed]

Biographical Sketch

Astafiev Artyom Aleksandrovich (born 1982), graduated from the Moscow Institute of Physics and Technology (MIPT) in 2005, received candidate (PhD) degree in physics in 2008. From 2002 to the present time he worked in the Semyonov Institute of Chemical Physics, Russian Academy of Sciences. Principal interests include: near-field and atomic force scanning microscopy, two-photon fluorescent microscopy, fluorescent and Raman spectroscopy, plasmonic effects in metal-semiconductor nanostructures. A.A.A. published up to 10 articles in scientific journals, participated in scientific conferences.