Introduction

Optical microscopy has come a long way from Zacharias Jansen's first microscope at the end of the 16*th* century to today's highly developed microscopes. A number of different contrast mechanisms allows broad applicability as a routine tool in biologiy, medicine, materials research, and many more. It is hard to think of any other tool for routine and exploratory tasks that is as widespread as optical microscopy. For an overview of the historical develoment, go see the <u>History of the Light</u> <u>Microscope</u>.

In the late 19th century, the German Ernst Abbe and the Englishman Lord Raleigh introduced a concept that is known as the *diffraction limit of spatial resolution*. This fundamental law states that with light, as with any other wave phenomenon used for microscopy, it is not possible to spatially resolve details that are located closer together than approximately half the probing wavelength. For optical microscopy, typically operating at a wavelength of 500 nm (the visible spectrum ranges from 400 nm to 700 nm), the lateral resolution is thus limited to about 250 nm.

Recently, two different approaches have been demonstrated to break the diffraction limit of resolution. Both approaches differ from conventional optical microscopy in that they don't form images of the object by means of lenses, but rather collect the optical response of the sample point by point.

In *Laser Scanning Confocal Microscopy*, both the sample illumination and the light collection is focussed onto the same spot on the surface of the sample (or even inside the sample). The sample is scanned and at every location the light intensity is recorded by a computer. The increase of resolution is achieved by the overlap of two Gaussian focal profiles, effectively narrowing the profile width and thus improving the resolution.

The second approach, named *Scanning Near-Field Optical Microscopy (SNOM)*, brings a small optical probe very close to the sample surface, in the region called "near-field". Here, at distances smaller than the wavelength away from the surface, also those waves can be probed that do not propagate, but rather decay exponentially perpendicular to the surface. It can be shown fairly easily that it in this evanescent field the k-vectors parallel to the surface can be fairly large, corresponding to small lateral (spatial) dimensions. As opposed to conventional as well as to laser scanning microscopy which are far-field microscopies, SNOM requires the close proximity between probe and sample.

This concept of near-field microscopy had been proposed already in 1928 by Edward Hutchinson Synge, however, the technical difficulties could not be overcome. It is amazing to see how closely the proposed device resembles today's instruments! A number of further proposals followed, probably without knowing of the earlier publications. Finally, after the demonstration of the Scanning Tunneling Microscope (STM), the principle was experimentally demonstrated in 1984. It's fair to say that SNOM is a rather young technique, and by far not yet established in the way conventional optical microscopy is.

With SNOM we have tool in hand that combines the advantages of optical microscopy (a whole variety of different contrast mechanisms, the possibility of spectroscopy for chemical identification, the fact that we are used to *see* (=probe optically) the world around us everyday), with the high resolution capability of scanned probe microscopies such as Scanning Tunneling Microsocopy (STM) and Scanning Force Microscopy (SFM). Today SNOM has a proven spatial resolving power of around 50 nm. That is definitely less than what STM and SFM are capable to, but comes along

with valuable information only accessible with optical contrast. One should look at it as a complementary tool with some room for improvement.



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Instrumentation

Scanning near-field optical microscopy can be performed in many *different ways of operation*. Most common today is the use of aperture probes for transmission microscopy, either in illumination (a) or in collection (b). However, many samples or substrates are opaque, so that working in reflection is necessary (c). The reflected light can be collected by optics close to the tip, or by the fiber probe itself, in which case often uncoated fiber tips are used.

A different approach is taken by the Photon Scanning Tunneling Microscope (PSTM), where evanescent waves are created at the sample surface by oblique far-field illumination (d). The probe tip acts as a scatterer of the evanescent field, leading to homogeneous waves which can be easily detected. Easy to operate, this mode suffers somewhat from difficulties in data interpretation. Of high interest is this arrangement with inverted light path, the i-PSTM or Tunnel Near-Field Optical Microscope (TNOM) or forbidden light near-field optical microscope. Similarly to the PSTM, light can be scattered from the evanescent field by other probe tips, such as a force microscope



tip on a cantilever (e). In the Plasmon Near-Field microscope, surface plasmons are generated at the surface of a sample on a thin film metallic substrate, and scattered by a probe tip (f).

Let's talk about the *illumination mode* SNOM for the moment. A *typical instrument* consists illumination (laser, fiber coupler) and collection optics (high N.A. objectives, filters, photomulitpliers for moderate light levels or photon counters of very low intensities),



fiber tip holder with shear force feedback (oscillator and lock-in amplifier, often based on a tuning fork), an approach scheme (mechanical or motorized), and a scanner (piezo tubes or stacks, it is often advantageous to scan the sample rather than the probe). Digital data acquisition and anti-vibration damping (optical tables, actively or passively dampened) completes the equipment. The microscope shown here sits on a conventional (inverted) light microscope, which allows to localize the sample with low resolution prior to SNOM operation.



The most crucial part of a near-field optical microscope is the *optical probe*. In most cases it is fabricated from an optical fiber which as been tapered (to reduce its size) and coated with metal



from the sides (to make it opaque); leaving only a small hole (the aperture) open at its very end. The tapering is usually achieved by heating and pulling, or by chemical etching, the coating is done from the sides while the fibers are rotated along their axes. When you do everything right, and you are a bit lucky as well, the resulting probe has a small and regular *aperture* (approx. 80 nm in the image to the right), and no holes at the sides where light would leak out and cause an unwanted signal background.

It is important to keep the optical probe at constant distance from the sample, so that changes in optical signal can be attributed to varying sample properties, and not to a mere change in probesample distance. Usually, the damping of the horizontally vibrating SNOM probe (caused by "shear forces") is taken as a measure for this distance. This way the *shear force feedback* keeps the probe tip at constant height. Various ways to measure the vibrating amplitude have been demonstrated, such as scattering an auxiliary light beam of the fiber probe and measure the resulting scattered light by a split detector, or to attach the fiber firmly to a small quartz tuning fork and measure the induced damping signal electrically, without any disturbing auxiliary optics. The amplitude signal is fed back into the control electronics which puts out a signal to the vertical piezo actuator. The image to the right shows the fiber end extending about 2mm over the tuning fork's legs. A tiny light spot from exiting light from a HeNe laser can be seen at the fiber tip





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Applications

The strength of near-field optical microscopy over other scanning probe techniques is that it allows to observe a whole variety of (optical) sample properties. If you are just interested in sample topography, force or tunneling microscopy will do a better job. With SNOM hoewever you have the choice of additional, often complimentary, contrast mechanisms:

- monitoring the <u>light intensity</u> allows to image changes in transmittivity or reflectivity, or in index of refraction
- <u>polarization contrast</u> shows sample birefingence and many aspects of orientation on surfaces
- <u>wavelength contrast</u> (or "fluorecence microscopy") lets one observe luminescence (photo- as well as electro-excited) and (molecular) fluorescence phenomena, as well as to perform spectroscopy for chemical identification

intensity contrast

Monitoring the intensity of light gives you information about the transmittivity and reflectivity of the sample, and of changes of index of refraction in general. Monitoring just the intensity of the signal is particularly succeptible to <u>topography artifacts</u>, and care has to be taken when interpreting the data.



Silver halide tabular grains are very flat samples, which makes topography artifacts unlikely to appear. These crystals are used in photographic emulsions, and their structure and composition is optimized for transposing light into latent images. The AgIBr crystals images here show almost no surface topography (left image). The SNOM signal (in transmission) shows variations which are likely to stem from changes of the index of refraction, caused by a gradient of Iodine concentration inside the crystals. *Image size:* 7.5 μ m x 7.5 μ m.

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polarization contrast

Many objects that to the unaided eye appear optically isotropic show various kinds of contrast when viewed through optical polarizers. illuminated by polarized light. This polarization contrast is induced by dichroism (the material shows selective absorption of one of two orthogonal polarization directions) or birefringence (the material shows differenct index of refraction for the two orthogonal polarization directions). Well-known examples are dichoric polarizers or the birefringent crystal calcite which forms double images of objects viewed through it.

Polarization contrast SNOM is not only interesting since its resolution is potentially better than that of conventional optical microscopy. The combination of optical and topographic information obtained by shear-force imaging is equally

attracitive.

To experimental schemes of polarization SNOM can be distinguished, depending on whether the direction of input polarization is kept fixed, or whether it is being modulated.

SNOM with fixed input polarization

Working with one fixed input polarization direction is easier to achieve for technical reasons. Ususally, fiber probes show asymmetries in taper shape and aperture, which leads to a partial depolarization of the injected light. Often, one direction of polarization can be found where the fiber depolarization is small, allowing to perform polarization contrast imaging with reasonably small instrumental signal background.

Micropatterned Liquid Crystal Films

Liquid crystals deposited on polyimide substrates orient according to the polymer orientation. This effect is exploited e.g. for the fabrication of liquid crystal display cells where the substrates are oriented by rubbing with a velvet roll. For this sample, additionally to the velvet roll procedure, small areas of substrate material have been oriented in a direction different to that of the surrounding matrix, with the help of a force microscope tip. The deposited liquid crystal film thus shows small domains of different orientation of the main axis of the linear birefringence, as can be

verified by conventional optical microscopy under crossed polarizers. Devices based on this technique are potentially interesting for producing small light guiding structures.

M. Rüetschi, J. Fünfschilling, and H.-J. Güntherodt. "Creation of submicron orientational structures in thin liquid crystal polymer layers", J. Appl. Phys. 80, 3155 (1996).



was used that could be photopolymerized in order to make its free surface accessible for the SNOM probe. The topography image (left) shows a small depression where the force microscope patterning took place. The contrast in the optical image can be adjusted by choosing the analyzer position. Maximum contrast can be achieved between the liquid crystals in the domain and in the matrix (top right), but also inside the domain walls (bottom right). Some of the features seen in the optical image correspond to topographic features which are known to scale with different molecular orientation angles inside and outside the domains. These optical features might be caused by the topography artifact. There are, however, optical features where the topography does not show any correspondence, strongly suggesting optical contrast. Most likely, this contrast stems from singularities in

For SNOM imaging a liquid crystal

index of refraction, resulting from a change of clockwise and counter-clockwise re-orientation of the liquid crystals along the domain wall.

Th. Lacoste. "Scanning Near-field Optical Microscopy and Polarization Contrast", Ph.D. thesis (german language), University of Basel 1997.

SNOM with Polarization Modulation

Varying the linear input polarization over all possible directions offers additional information over working with one fixed polarization. In a modulation scheme, the input polarization is varied continuously over 180 degrees, and the sample response to all these different linear polarization states is recorded. Experimentally, the modulation is realized by an electro-optical modulator and a quarter-wave plate. Fiber birefringence compensation is even more important than for fixed polarization experiments, and depolarization by the SNOM probes is a severe problem. This technique has been pioneered by <u>Paul Barbara's group at U Minnesota (online newletter)</u>, see also "Polarization-Modulation Near-Field Scanning Optical Microscopy of Mesostrucured Materials" by D. A. Higgins, D. A. Vanden Bout, J. Kerimo, and P. F. Barbara, J. Phys. Chem. **100**, 13794 (1996).

Dye Crystals



Crystals of the dye naphtol blue black, a dye that is absorbing in the red, have been imaged using polarization modulation. The topography image (top left) shows longish crystallites, surrounded by an apparent roughness which is probably due to contamination. The averaged transmission image (top right) shows increased absorption at the location of the crystallites. Each crystallite shows maximum absorption when the polarization direction and the orientation of its transition dipole moment coincide. Lock-in detection of the transmitted light allows to simultaneously generate an image of the optical anisotropy (bottom left, lock-in amplitude signal), which is the maximum absorption of the sample of any of the illuminating input polarization states. Similarly, an image of the direction of the transition dipole moment (bottom right, lock-in phase signal) shows the direction of the polarization where

maximum absorption occurs.

Th. Lacoste, Th. Huser, R. Prioli and H. Heinzelmann. "Contrast Enhancement using Polarization-Modulation Scanning Near-field Optical Microscopy (PM-SNOM)", Proc. NFO-4, 1997. To be published in Ultramicroscopy.

Magneto-Optical Imaging

Magneto-optics describes the interaction of optics with magnetism. The Faraday effect describes the change of polarization of light transmitted through a magnetized sample; the analogous effect in reflection is the magneto-optic Kerr effect which has huge applicability in technology, such as for magneto-optical storage (MO disks). The Faraday effect is a circular birefringence effect:

depending on the magnetization and the material's properties and thickness, a rotation of the



orientation of the transmitted linearly polarized light can be observed.

Bismuth-doped yttrium-irongarnet film (YIG) films exhibit perpendicular magetization, their domains can be imaged by applying the polarization modulation scheme. In contrast to the experiments on the dye crystals described above, this sample displays no polarizationdependent absorption, and an

optical analyzer has to be placed in front of the detector. During the experiment, the light signal will be sinusoidally modulated. The magnitude of the Faraday rotation angle is given by the change of the phase angle between incident and transmitted light, and can be monitored by recording the phase output from a two-channel lock-in amplifier.



Except for some dust particles the sample topography is flat (left image). In the simultaneously recorded optical image (lock-in phase), the domain structure of the YIG film is clearly visible (middle image). The Faraday rotation angle ranges over 2.3° across the whole image, i.e. between up and down domains. For comparison, measurements with fixed polarization were taken at the same sample location (right image). Determining the magnitude of the Faraday rotation angle with a fixed polarization method is a tedious task since it requires to change the analyzer setting until contrast reversal is observed.

Th. Lacoste, Th. Huser and H. Heinzelmann. "Faraday Rotation Imaging by Near-field Optical Microscopy", Z. Phys. B 104, 183 (1997).

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wavelength contrast

The phenomenon of luminescence describes the fact that certain samples absorb light of a specific wavelength, and re-emit light of a longer wavelength (i.e. of less energy). Depending on the time scales involved, one distinguishes between fluorescence (fast decay of the excited state: 10^{-8} sec and less) and phosphorescence (slow decay of the excited state: μ sec to hours).

LiF, such as many other alkali halides, is known to form color centers when irradiated with high energy electrons. The images show topography (left), excitation light at 458 nm in transmission (middle), and photoluminescence signal (right). All signals are taken simultaneously. The luminescence seems to stem from the rims of the grains visible in the topography image. The fact that the excitation light doesn't show this feature indicates that this appearance is not due to a topography artifact. *Image size:* $5 \ \mu m \ x \ 5 \ \mu m$.



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Topography Artifacts



Care has to be taken when SNOMimages are acquired while the fiber probe follows the surface topography. The near-field optical signal is strongly dependent on the distance of the optical probe to the sample surface. Therefore, only slight changes of this distance are already enough to considerably change the recorded optical signal. So far, so good. However, the optical fiber probe is

However, the optical fiber probe is a structure usually some 300 nm in size, and often a grain of the rough Al coating is its most protruding point. This part of the tip will be responsible for shear-force imaging, and thus for the vertical motion of the tip. It is obvious that this motion which keeps the Al grain at a constant distance to the sample, will at the same time induce a change of distance of the optical aperture to the surface underneath, and the optical signal will change. This effect, where the high resolution of force microscopy induces a seemingly high resolution change in the near-field optical signal, is usually referred to as topography artifact.

Since the theoretical treatment of the tip-sample arrangement in near-field optics is rather complicated (it comes down to running numerical calculations in a volume several wavelengths in size and with very low symmetry), there is no easy way to estimate the size of topography artifacts in order to subtract it from the experimental data.

The best way around the perils of topographic artifacts in SNOM imaging is to stay away from moving the tip vertically. This can either be done when the sample is extremely flat and there is no need for feedback action, or by scanning at constant height. The latter unfortunately requires that the tip is at larger distances to the sample in order to be safe. For an experiment where constant height scanning was essential, have look at the <u>forbidden light experiments</u>.

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