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Scanning near-field optical microscopy utilizing silicon nitride probe photoluminescence

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We describe a simple method for performing high-resolution scanning near-field optical microscopy (SNOM). A commercial Si$_3$N$_4$ tip is illuminated by an intense light source, which causes the tip to emit redshifted (inelastically scattered) light. Part of the redshifted light passes through a sample, allowing transmission light microscopy. By simple modification of a commercial atomic force microscopes (AFM), we are able to image many different samples with high-resolution optical microscopy, achieving 20–30 nm lateral resolution for the best samples. The high resolution of the technique is not only due to the high curvature of the AFM tip, but also to the fact that the intensity of inelastically scattered light transmitted through the sample decays exponentially with the separation between the tip and the sample (decay length ~100 nm). We envisage applications to transmission SNOM, spectroscopic imaging, and imaging of fluorescently labeled bioconjugates. The collection of the optical image does not interfere with the normal operation of the AFM, so deflection, height, or other modes of operation can be captured simultaneously. © 2005 American Institute of Physics. [DOI: 10.1063/1.2136216]
itself is photoluminescent but that gold enhances the luminescence.

Light scattered from the tip was collected by the microscope objective and the incident green light was removed with a dichroic mirror and long-pass filter; the orange light was collected by a photomultiplier tube (PMT) (Hitachi H5784-01). Resolution was improved by the addition of a small aperture placed before the PMT. All images presented here were captured in air, but images can also be captured in water.

The intensity of orange light that passes back through the sample and is captured by a photomultiplier tube decays exponentially with the separation between the tip and the sample, as shown in Fig. 1. The best lateral resolution in optical imaging was recorded when the incident laser light was focused onto the tip and the beam angle was adjusted to give the largest gradient of the inelastically scattered intensity as a function of separation. We note that the high power-density beam causes a large force that pulls the tip toward the sample. The rapid decay of the intensity (decay length \(\sim 100 \text{ nm}\)) keeps the emitted orange light localized to a small region, thereby providing high resolution. We find that silicon nitride tips from several batches and a variety of types produce this inelastic scattering effect. Under the conditions of our experiments, when the AFM is removed, the green light passes through the sample and illuminates the ceiling of our laboratory, clearly indicating that the beam that illuminates the tip is not evanescent. This suggests that the exponential decay of the optical signal is due to a near-field interaction between the tip and the sample. The photoluminescent effect itself is not due to a near-field interaction because there is a component of the photoluminescence that is present at large (millimeter) separations. At this point we do not understand the molecular origin of the photoluminescence in silicon nitride.

We use the inelastic scattering to obtain high-resolution images of samples. While all the other optical components are kept stationary, the sample is scanned under the tip, so that different positions on the sample come into the path of the intense part of the orange light emitted from the tip. Figure 2 shows images of tobacco mosaic virus in which the AFM is used with feedback to the cantilever Z displacement to maintain constant cantilever deflection. At the same time that we collect the high-resolution AFM image, we also record the intensity of light that is captured by a PMT positioned below the sample to obtain the optical image. Note that the virus is revealed as regions of low light intensity. This is in contrast to the commonly observed imaging artifact where sharp, high features produce higher intensity due to failure of the feedback loop to maintain a constant height above the sample. The optical contrast mechanism is made clear by imaging opaque carbon particles in constant height mode, where the tip stays at an approximately constant height above the glass slide (Fig. 3). Particles as small as 20–30 nm in diameter can be resolved. In earlier work by Azoulay et al., apertureless optical images of (opaque) metal islands showed the islands as regions of high optical signal. This was interpreted as arising from interference between the optical signal from the AFM tip and the specular-reflection field. In Fig. 3, the contrast is reversed compared to that observed by Azoulay et al.: a large optical signal arises from the gaps between the opaque particles. This contrast is consistent with contrast arising through absorption, as with transmission light microscopy. The different contrast obtained here is explicable because we are filtering out the
incident 532-nm light and collecting only light that is inelastically scattered from the tip. Therefore our optical scheme is very similar to the regular fiber-probe SNOM, where light is emitted from an aperture. This is further clarified in Fig. 4, an image of KCl crystals, where the large optical signal corresponds to the gaps between the crystals. For optically transparent samples such as the virus and KCl, the diminished intensity of transmitted light cannot arise by absorption, so the contrast must arise from the near-field interaction of the dipolar field of the sample with the tip. Note also that in Fig. 4 there is not a perfect correlation between the optical and deflection images: the optical image is not simply a record of the cantilever deflection.

In conclusion, we present a simple, novel approach to high-resolution optical imaging based on the photoluminescence of commercial silicon nitride AFM tips. After stimulation with intense green light, emitted orange light from the tip can be used to probe the sample. The intensity of this orange light decays exponentially with distance from the sample. We achieve 20–30 nm resolution parallel to a surface and 2–5 nm normal to the surface for different samples. At present our tests of resolution are limited by sample feature size. Our images always produce a large optical signal when the tip is above gaps between adsorbed features, and a lower intensity for adsorbed materials that block light to the tip. We have shown that we can vary the frequency of the scattered light, so spectroscopic imaging is also possible. One can also envisage other modes where the tip is scanned at a constant optical signal, or oscillatory modes.

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