ELSEVIER

Ultramicroscopy 57 (1995) 309–312

ultramicroscopy

Optically trapped non-linear particles as probes for scanning near-field optical microscopy

H.M. Hertz [a], L. Malmqvist [a], L. Rosengren [b], K. Ljungberg [b]

[a] *Department of Physics, Lund Institute of Technology, P.O. Box 118, S-221 00 Lund, Sweden*
[b] *Department of Technology, Uppsala University, P.O. Box 534, S-751 21 Uppsala, Sweden*

Received 10 November 1993; accepted 28 December 1993

Abstract

We use the frequency doubled light from an optically trapped lithium niobate particle for non-intrusive scanning near-field optical microscopy. The detected power from this 50–100 nm diameter probe is currently tens of pW and is expected to approach nW with an improved detection system. The current experimental resolution is approximately 0.5 μm, while the ultimate theoretical resolution is 70–90 nm. An acoustic trap which potentially allows higher resolution imaging is briefly described.

1. Introduction

In scanning near-field optical microscopy (SNOM) [1] subwavelength resolution is obtained by scanning a microscopic optical probe in close proximity to the object. Current probes include etched apertures, protrusions [2], fluorescence tips [3] and pulled fiber tips [4]. However, biological objects with intervening membranes or rough surfaces are not always accessible for such studies since the positioning of the probe requires mechanical access to the object. Trapped particle optical microscopy (TPOM) [5] eliminates this restriction by using a non-intrusive optical trap [6] to position and scan the microscopic optical probe. In the first TPOM demonstration experiments, scattered light (λ = 514 nm) from optically trapped 290 nm SiO_2 particles was used for the imaging [5]. However, the resolution was limited to approximately 2 μm due to the scattering of the trapping laser beam by the test object, resulting in a low signal-to-noise ratio at small particle–object distances. In our current arrangement we eliminate this limitation by trapping a microscopic lithium niobate ($LiNbO_3$) particle and using the frequency doubled light from the non-linear crystal for the microscopy [7–9]. In addition to its non-intrusive character, the use of an optically trapped particle as probe has several other advantages compared to conventional SNOM probes. Accidental collisions with an object will not result in probe or object destruction due to the elastic nature of the optical trap. Loss of particle or probe deterioration does not present a problem since plenty of particles are available for injection in the trap. Finally, the non-linear wave-

0304-3991/95/$09.50
SSDI 0304-3991(94)00158-8

length conversion in the lithium niobate particles show no sign of the bleaching known to occur in many fluorescent probes.

2. Experiments and discussion

The optical trap is shown in Fig. 1. A Nd : YAG laser beam ($\lambda = 1.06$ μm) is focused by a NA = 1.25 water immersion microscope objective into a water cell. Due to radiation pressure [5,6] $LiNbO_3$ particles with a diameter of approximately 50–100 nm are non-intrusively trapped just below the focus. The chopped CW trapping beam is frequency doubled in the trapped particle, resulting in approximately a pW of visible emission ($\lambda =$ 532 nm) for an average input power of ≈ 50 mW. The studied test object is positioned below the particle and the trapped particle may then be accurately scanned in immediate proximity to the object. By detecting only the visible light, the scattering problem is circumvented and smaller particle–object distances than those in Ref. [5] may be used. The infrared trapping wavelength was chosen due to its low absorption in most biological materials, thereby avoiding thermally induced turbulence and damage to the studied object. The method is experimentally demonstrated on freestanding etched silicon bridges, which exhibit high absorption at visible wavelengths and low in the IR. The 1 mm long and ≈ 10 μm thick bridges were produced by anisotropic etching in KOH. The test edge was

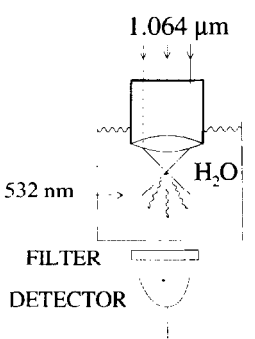

Fig. 1. Experimental arrangement for optical trapping and frequency doubling in lithium niobate microparticles.

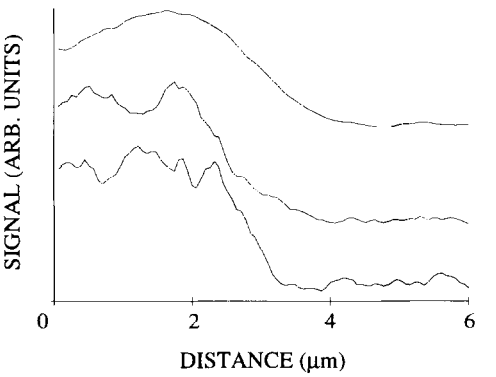

Fig. 2. One-dimensional scans over an absorbing silicon edge as the particle approaches the object. The particle–object distance is decreased going from the top curve to the bottom curve.

determined by the 90° angle between the (100) and the (010) crystallographic planes. The edge sharpness was measured to $< \pm 50$ nm by scanning electron microscopy. Fig. 2 shows three one-dimensional scans over the silicon edge. Going from the top to the bottom curve, the sharpening of edge scans as the particle approaches the object is illustrated. The lower edge scan indicates an experimental resolution of approximately 0.5 μm [9]. The resolution in the present experiment is limited by several factors of which surface forces dominate. The high refractive index of silicon results in strong van der Waals forces [10] between the surface and the particle, making it difficult to position the probe closer than a few hundred nm from the silicon bridge [9]. At closer distances, the forces of the optical trap ($\approx$ pN) are exceeded by the surface forces. Using test objects more similar to biological objects, with low refractive index, the surface forces should be reduced by one to two orders of magnitude, making high-resolution experiments possible.

In addition to resolution, the emitted power of the probe is of great importance for a functional SNOM microscopy system. The ≈ 1 pW used above is sufficient to image highly absorbing objects with reasonable signal-to-noise ratio. However, fluorescent imaging requires a few nW [11]. In order to increase the emitted green intensity while still keeping the average IR trapping beam below the approximately 100 mW biological dam-

age threshold [12], the IR beam was pulsed, thereby taking advantage of the quadratic intensity dependence of the frequency doubled light. The Nd:YAG laser was acousto-optically Q-switched at 25 kHz resulting in 400–800 ns long pulses depending on pump power. The size of the trapped particle was determined by measuring the scattered IR light and comparing it to the scattered light from monodisperse 60 nm diameter silicon dioxide particles. In this size range Rayleigh scattering can be assumed, making the scattered intensity a simple function of particle diameter and refractive index [5,13]. Fig. 3 shows the average detected frequency doubled power as a function of average input IR power for a 75 nm diameter $LiNbO_3$ particle. The emitted light was collected with a NA = 0.12 detection system. The power increases slightly more than the expected quadratic intensity dependence due to shortening of the Q-switched IR pulses as the input power is increased. In recent experiments, using KTP ($KTiPO_4$) particles, an order of magnitude higher visible emission than for $LiNbO_3$ was observed. Assuming that the detection efficiency is increased by using a NA = 1 objective, the detected visible power at 100 mW average input power would then approach one and ten nW for $LiNbO_3$ and KTP, respectively. It seems reasonable to expect that even higher power may be reached with short-pulse high-repetition-rate modelocked lasers, making fluorescence studies feasible.

The approximately 0.5 μm resolution in the test experiment is still less than the conventional far field diffraction limit. Theoretically, the resolution is ultimately determined by the physical size of the particle convolved with its displacement due to Brownian motion in the optical trap. Calculations following Refs. [5,8] indicate that a resolution of approximately 90 nm is possible using a 100 mW, $\lambda = 1.06$ μm trapping beam and 75 nm $LiNbO_3$ particles. With $\lambda = 800$ nm radiation, which also exhibits low absorption in biological material, the theoretical resolution may be increased to approximately 70 nm with 60 nm particles. In order to further increase the resolution we are developing a new type of non-intrusive trap based on a confocal ultrasonic cavity [14] as depicted in Fig. 4. Particles with a different acoustic impedance than the surrounding liquid are trapped in the velocity antinodes of the standing acoustic wave in the cavity. The trapping force is due to the gradient in the time-averaged Bernoulli pressure. In a demonstration experiment with an 11 MHz ultrasonic frequency cavity, 2 μm diameter glass spheres are stably trapped. The measured trapping force was found to agree with theoretical calculation based on Ref. [15]. Extending these calculations to 1 GHz ultrasonic frequency indicates that approximately 50 nm resolution imaging should be possible using the ultrasonic trap [14]. Furthermore, by using acoustic waves for the trapping and optical waves for the detection, the trapping and detection systems are decoupled allowing more flexibility in the design of a trapped particle optical microscope. In order to avoid the scattered light problem, the trap may be used either with nonlinear particles, as discussed above, or with fluorescent particles.

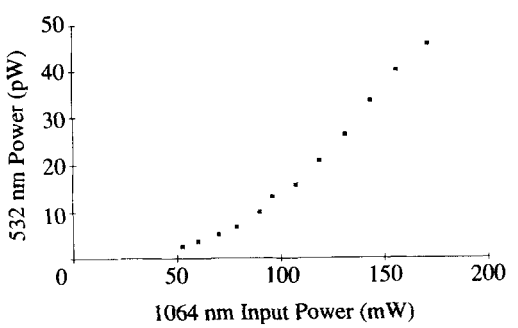

Fig. 3. Detected average power at $\lambda = 532$ nm as a function of incident power at $\lambda = 1.06$ μm. The incident beam was Q-switched at 25 kHz.

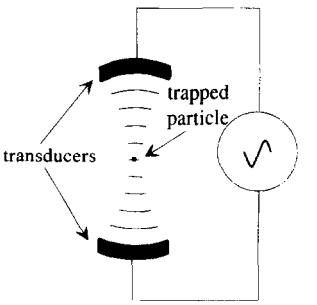

Fig. 4. Experimental arrangement of confocal acoustic cavity for non-intrusive positioning of microparticles.

3. Conclusions

In conclusion, we have shown that optically trapped lithium niobate particles may be used as high-brightness probes in scanned probe optical microscopy. Such non-intrusive probes should be interesting for studies of, e.g., biological material where intervening membranes or rough surfaces often prohibit the use of conventional probes. The resolution in the demonstration experiments on silicon bridges is still limited by the surfaces forces and other factors, which prevent the positioning of the trapped particle probe sufficiently close to the object. Work is in progress to minimize this problem by developing test objects more similar to biological objects. The particle–surface forces are much lower for such low refractive index objects, making it possible to reach the < 100 nm theoretical resolution limit.

The measurements of emitted power from the non-linear crystal fragments indicates that high-resolution fluorescence studies of such objects should be possible using a Q-switched trapping beam. An obvious alternative use of the high-brightness emission from the non-linear particles is to position such a particle on a fiber tip, and perform conventional SNOM in a similar way as fluorescent tips are used in Ref. [3]. This approach would avoid the bleaching problem of many fluorescent substances.

Acknowledgements

The authors gratefully acknowledge the help from Kristina Georgsson, Fredrik Laurell and Lars Rymell. This work was financed by the Swedish Natural Science Research Council, the Swedish Research Council for Engineering Sciences, the Swedish Board for Industrial and Technical Development, the Carl Trygger Foundation and the Crafoord Foundation.

References

[1] D.W. Pohl, in: Advances in Optical and Electron Microscopy, Vol. 12, Scanning Near-field Optical Microscopy (SNOM) (Academic Press, New York, 1991) p. 243.
[2] U.Ch. Fischer and D.W. Pohl, Phys. Rev. Lett. 62 (1989) 458.
[3] A. Lewis and K. Lieberman, Nature 354 (1991) 214.
[4] E. Betzig and J.K. Trautman, Science 257 (1992) 189.
[5] L. Malmqvist and H.M. Hertz, Opt. Commun. 94 (1992) 19.
[6] A. Ashkin, J.M. Dziedzic, J.E. Bjorkholm and S. Chu, Opt. Lett. 11 (1986) 288.
[7] S. Sato and H. Inaba, Electron. Lett. 28 (1992) 286.
[8] L. Malmqvist and H.M. Hertz, in: Near Field Optics, Eds. D.W. Pohl and D. Courjon (Kluwer, Dordrecht, 1993) p. 141.
[9] L. Malmqvist and H.M. Hertz, Two-color trapped particle optical microscopy, Opt. Lett. 19 (1994) 853.
[10] J. Israelachvili, Intermolecular and Surface Forces (Academic Press, New York, 1992).
[11] J.K. Trautman, E. Betzig, J.S. Wiener, D.J. DiGiovanni, T.D. Harris, F. Hellman and E.M. Gyorgy, J. Appl. Phys. 71 (1992) 4659.
[12] S.M. Block, in: Noninvasive Techniques in Cell Biology, Optical Tweezers: A New Tool for Biophysics (Wiley, New York, 1990) p. 375.
[13] M. Kerker, The Scattering of Light (Academic Press, New York, 1969).
[14] H.M. Hertz, Acoustic Trap for Non-intrusive Positioning of Microparticles, manuscript in preparation.
[15] L.A. Crum, J. Acoust. Soc. Am. 50 (1971) 157.