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Jingsong Wei

Nonlinear Super-Resolution Nano-Optics and Applications

非线性超分辨纳米光学及应用

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Preface

With the development of electro-optical technology, nano-Sci and Tech., and life science and biology, the light imaging resolution, lithographic linewidth, and optical information mark size are required to reach down to subwavelength or even nanoscale. However, these are restricted by the Abbe limit due to the diffraction effect. Researchers have proposed a number of methods to fight against the Abbe limit. These methods can be classified into two kinds, one is to change the point spread function, such as scanning near-field optical probe microscopy, liquid (solid) immersion lens, and phase-only pupil filter, etc. The other is to improve the resolution through detecting the fluorescence signal, such as photo-activated localization microscopy (PALM) and stochastic-optical reconstruction microscopy (STORM), which are generally used in biomedical and life science. The stimulated emission depletion (STED) is an excellent combination of two kinds of techniques mentioned above. The STED is mainly used in biomedical imaging and life science, and is also explored to apply to nanolithography in recent years.

This book first introduces the principle and technical schematics of common methods for realizing nanoscale spot (Chap. 1), describes the third-order nonlinear effects and characterization methods (Chaps. 2 and 3), and then analyzes the strong nonlinear characteristics (including nonlinear absorption and refraction) of semiconductor and metal-doped semiconductor thin films (Chap. 4). Chapters 5–7 focus on nonlinearity-induced super-resolution effects, including nonlinear saturation absorption-induced aperture-type super-resolution, nonlinear refraction-induced self-focusing and interference-manipulation super-resolution, and the combination of nonlinear thin films and phase-only pupil filters to compress the side-lobe intensity and reduce the main spot size to nanoscale. Applications in high-density optical information storage, nanolithography, and high-resolution light imaging are presented in Chaps. 8 and 9, and some remarks are given at the end of the book.

I hope that this book can drive nano-optics and nanophotonics to continue to advance. The book is helpful for advanced undergraduates, graduate students, and researchers and engineers working in related fields of nonlinear optics, nano-optics and nanophotonics, information storage, laser fabrication, and lithography, and light imaging etc. It is unavoidable that some errors and incorrectness may occur in this book, I hope that the readers can point them out. I also will further correct them and improve my work on future releases.

The work in this book is partially supported by the National Natural Science Foundation of China (Grant Nos. 51172253, 61137002, and 60977004). Here please allow me to express my appreciations to my family, Prof. Fuxi Gan, and Prof. Chenqing Gu due to their support in my work and life. It is a pleasure to thank my colleagues and students for their help. Last but not least, I am delighted to dedicate this book to my son, Yusen. Yusen is a smart boy, and he brings joy and happiness to our family.

Shanghai, China Jingsong Wei

Contents

Chapter 1
General Methods for Obtaining Nanoscale Light Spot

1.1 Introduction

In 1873, the German physicist Ernst Abbe realized that the resolution of optical imaging instruments is fundamentally limited by the diffraction of light. His finding indicated that ultimately the resolution of an imaging instrument is not constrained by the quality of the instrument, but by the wavelength of light used and the aperture of its optics [1]. That is, the resolution of a conventional optical-lens-based imaging system is restricted by the Abbe diffraction limit to $\sim\lambda/2NA$, where λ and NA are the free-space wavelength of the imaging radiation and numerical aperture of the lens, respectively. This diffraction-limited phenomenon hindered the performance of optical microscopy for over a century and was considered a fundamental, unbreakable rule. Beating the diffraction limit and realizing nanoscale light spot have become one of the primary research focuses in modern optics [2]. Realizing nanoscale light spot is also always a hot subject due to its important applications in high-density data storage and nanolithography, etc. According to Abbe diffraction limit, to reduce the light spot size, a short-wavelength laser source and a high NA lens need to be used. However, in current far-field optical system, on one hand, it is difficult to shorten the laser wavelength further because of high product cost and low transmission for optical elements. On the other hand, the NA of a commercially available lens is close to the limit. Under such circumstances, numerous methods and techniques are proposed to overcome the Abbe limit. Let us give a brief introduction to the working schematics and principles of general methods for realizing nanoscale light spot.

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1.2 Near-Field Scanning Probe Method

In far-field optical systems, the optical resolving power is generally constrained to half-wavelength, namely half-wavelength limit. This is because the high spatial frequencies of objects are such that the waves become evanescent in the certain direction. If the evanescent waves containing fine structure information can be used to illuminate the object, the resolving power of half-wavelength limit can be overcome. The generation of evanescent wave is key to break through the Abbe limit. In 1972, Ash and Nicholls [3] proposed a concept of the super-resolution aperture scanning microscope, where one can fabricate a small aperture of radius $r, (r/\lambda \ll 1)$ in a thin diaphragm, and the object information of a comparably small area can be obtained only if the object is located very close to the aperture. The super-resolution aperture scanning microscope is actually a primary configuration for generating and obtaining the evanescent wave because the distance between the aperture and object surface is smaller than λ.

In order to observe smaller fine structure of objects, Betzig and Trautman [4] proposed to establish the near-field optical microscopy where the irradiation wavelength is in the visible light. The basic configuration is a near-field pinhole probe, as shown in Fig. 1.1. Figure 1.1a is a schematic of the pinhole probe, where the fiber probe is coated with an Al thin film, and an aperture with a diameter of far smaller than incident light wavelength is at the apex of probe. The incident light passes through the fiber probe, and a very small light dot is formed at the aperture of the apex of the probe. The light dot size is determined by the aperture diameter (also see Fig. 1.1b). The light dot is evanescent wave and decays exponentially along the optical axis. In real applications, the distance between the light dot and object needs to be remained in the near-field range of less than the light wavelength, or else it is difficult to obtain a resolution below the diffraction limit.

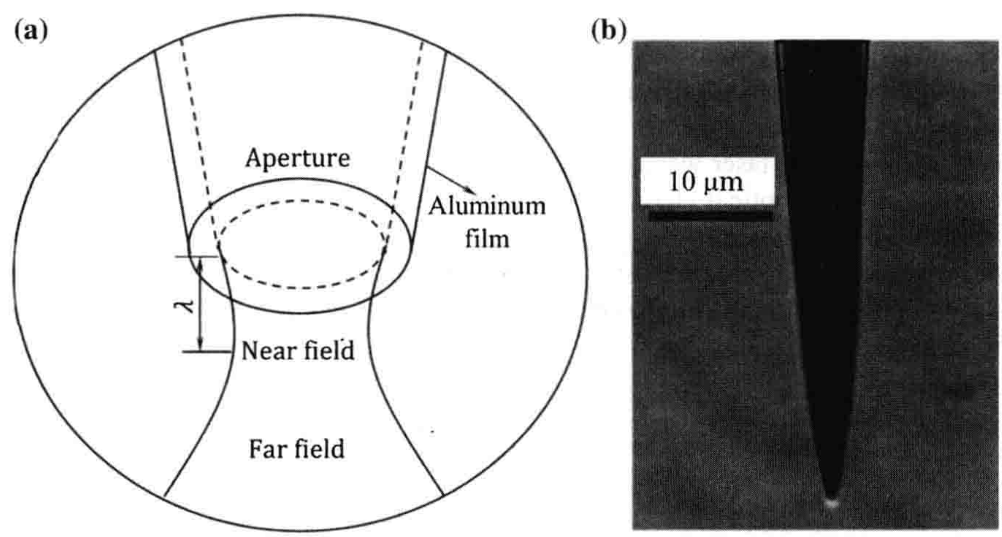

Fig. 1.1 Near-field fiber probe, **a** schematic configuration, and **b** real fiber probe

1.2.1 Aperture-Type Probe

In the configuration of near-field optical microscopy, the design and fabrication of the near-field probe are critical. The aperture-type probe is one of the probes that are most widely used. The aperture-type probe can be fabricated by heating and subsequent pulling method. An alternative method is by wet chemical etching, where bare optical fibers are dipped in an etchant solution to produce a taper with a sharp endpoint. Generally speaking, the probe surface needs to be coated with Al to create an aperture smaller than 100 nm. The advantages are follows: (1) the probe sharpness can be reduced down to 20 nm; (2) the light is easily delivered to the end of the probe; (3) the flat endpoint is beneficial in imaging; (4) the small lateral dimensions of the probe results in a good access to corrugated samples. However, some drawbacks are difficult to overcome, such as the grainy structures on the aluminum coatings resulting in poor optical imaging, highly asymmetric polarization behavior, pinholes at the taper region disturbing the polarization behavior of the tip, and low optical throughput and brightness etc.

In order to improve the probe performance, Veerman et al. [5] developed focused ion beam (FIB) fabrication method. The steps are as follows. (1) The fiber tips are pulled with a fiber puller, and the tip sharpness determines the minimum obtainable aperture size of the finished probe. (2) The Al is deposited on the probes using e-beam evaporation. It is noted that evaporation at an angle of approximately 75° with respect to the probe axis creates an aperture, while evaporation at an angle of 90° covers the entire fiber end. (3) The probes are then put into the FIB machine. In the FIB machine, the Gallium ions are used to remove a very thin slice of material from the tip end, which results in the formation of flattened aperture. The aperture size can be tuned by simply changing the thickness of the slice that is milled. Figure 1.2 shows images of two FIB-etched tips. Figure 1.2a is a flat-end face and well-defined circular aperture with a diameter of 120(±5) nm, a much smaller aperture of about 35(±5) nm is fabricated by tuning the slice thickness in Fig. 1.2b.

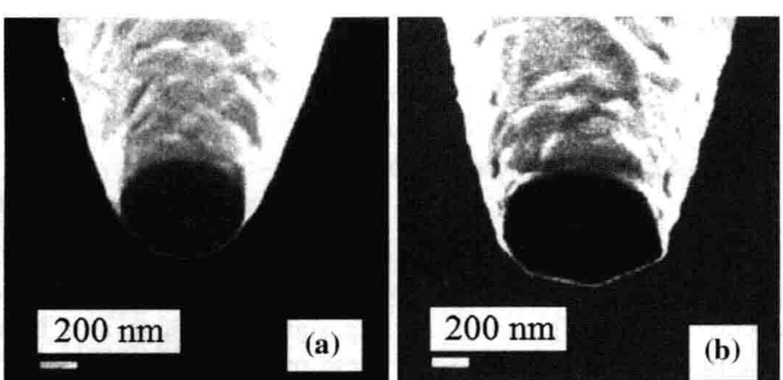

Fig. 1.2 The images of probes fabricated by FIB method, the aperture diameter of **a** 120 nm and **b** 35 nm. Reprinted with permission from [5]. Copyright 1998, American Institute of Physics

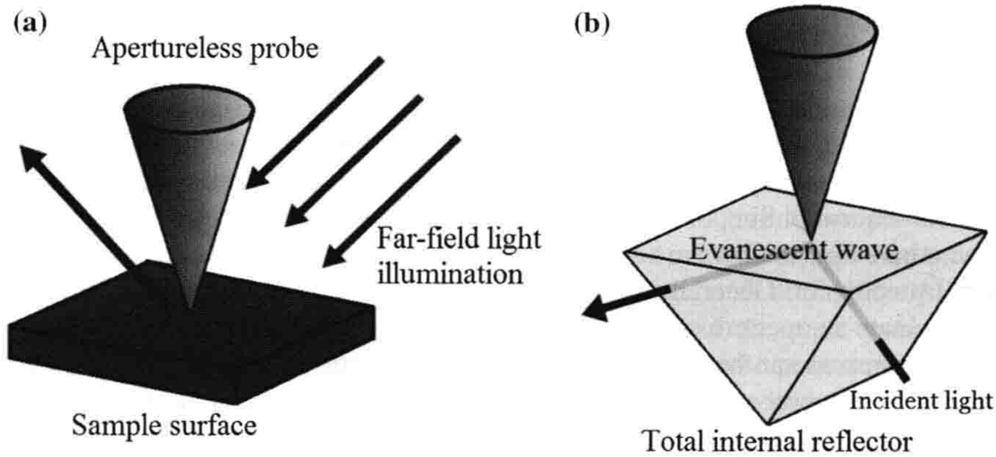

Fig. 1.3 Configurations of apertureless-type metal probe, the sample is illuminated **a** by far-field light, and **b** by an evanescent wave

1.2.2 Apertureless-Type Metal Probe [6]

An alternative to design the probe is a scattering-type tip operated in the apertureless configuration, where a sharp tip is illuminated by far-field light and scatters the light field locally at tip apex. The tip apex actually acts as a local source, as shown in Fig. 1.3a, where the relevant near-field light signal needs to be extracted from a large background of unwanted light from the sample surface.

One can decrease the stray light by using lock-in detection method, where the tip is vibrated at certain frequency, and the detected near-field light signal is demodulated with a lock-in scheme. Another method to reduce the unwanted scattered light is using evanescent wave illumination by total internal reflection technique, as shown in Fig. 1.3b, where the evanescent wave is generated over the sample surface and scattered by the probe, and the scattered light is collected by external optics. Apertureless-type metal probe can obtain much better resolution of below 10 nm by concentrating light field close to the tip apex, and the resolution is determined by the apex radius.

1.2.3 Tip-on-Aperture-Type Probe

For apertureless-type metal probes, the far-field illumination by a focused laser beam exposes a large area around the tip apex, which causes some problems, such as the generation of background scattering signal and large area bleaching for fluorescence imaging. In order to solve these problems, Frey et al. [7] designed and fabricated tip-on-aperture-type probe, where the light fields can be optimally concentrated at tip apex by fully exciting localized surface plasmons or antenna resonance effect. The tip-on-aperture-type probe combines the advantages of

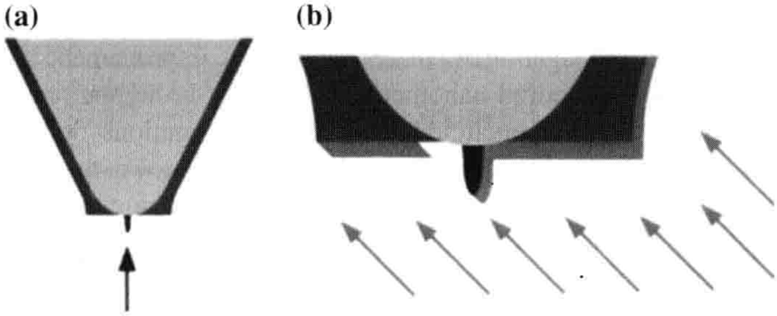

Fig. 1.4 Schematic of fabricating process of tip-on-aperture-type probe. **a** An electron-beam-deposited tip is grown on the aperture of a metallized glass fiber probe in a scanning electronic microscopy, **b** one-sided deposition of Al metalizes the tip and forms an elongated and reduced aperture in tip shadow. Reprinted with permission from [7]. Copyright 2002, American Institute of Physics

aperture and apertureless probes and achieves low background noise and high light throughput as well as topographical resolution. The fabrication process of tip-on-aperture-type probe is as follows.

(1) An optical single mode fiber is thinned in an etching solution.
(2) The fiber is etched in a fourfold diluted solution to reduce the diameter to 15 μm, which makes a 2 μm length sharp tip be obtained because the fiber core is etched more slowly than the cladding.
(3) The tip is covered with Cr adhesion layer with several nanometers and Au layer of about 200 nm due to good contrast in the scanning electron microscope.
(4) An aperture can be generated by pressing the tip on a glass surface and monitoring the far-field light throughput.
(5) The tip is obtained by focusing the electron beam to the center of the aperture for several seconds at 8 kV acceleration voltage, also see Fig. 1.4a.
(6) Cr thin film of 3.5 nm and A1 thin film of 33 nm are then sequencially deposited on the probe surface by evaporation technique at 45° incidence angle (also see Fig. 1.4b).

In this way, the electron-beam-deposited tip is metalized on one side, and the original aperture is reduced to a small and elongated aperture left by the tip's shadow, resulting in an asymmetric tip–aperture arrangement. The position of the tip coincides with one edge of the new aperture.

1.2.4 C-Aperture Encircled by Surface Corrugations on a Metal Film [8]

As reported, the light throughput of the 60-nm circular aperture follows an exponential decay with the distance from aperture due to evanescent wave effect.