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To cite this article: Yasushi Oshikane et al 2007 Sci. Technol. Adv. Mater. 8 181

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Observation of nanostructure by scanning near-field optical microscope with small sphere probe

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Received 28 October 2006; received in revised form 27 February 2007; accepted 28 February 2007

Available online 25 April 2007

Abstract

Step and terrace structure has been observed in an area of $1\,\mu m \times 1\,\mu m$ on the cleaved surface of KCl–KBr solid-solution single crystal by scanning near-field optical microscope (SNOM) with a small sphere probe of 500 nm diameter. Lateral spatial resolution of the SNOM system was estimated to be 20 nm from the observation of step width and the scanning-step interval. Vertical spatial resolution was estimated to be 5–2 nm from the observation of step height and noise level of photomultiplier tube (PMT). With applying a dielectric dipole radiation model to the probe surface, the reason why such a high spatial resolution was obtained in spite of the 500 nm sphere probe, was understood as the effect of the near-field term appeared in the radiation field equations.

Keywords: Scanning near-field optical microscope (SNOM); Polystyrene small sphere; Evanescent light; S-polarized light; He–Ne laser; Optical feedback; Solid-solution single crystal; Step-terrace structure; Electric dipole radiation

1. Introduction

An optical microscopy is limited by the so-called Rayleigh diffraction limit, which means that the dimensions of a focusing spot are always about half size of the wavelength used. Therefore, the resolving power with visible light illumination has an upper limit of several hundred nanometers. It is possible to use other microscopic techniques such as scanning probe microscopy or electron microscopy, but the information obtained by visible light can be a valuable tool in scientific studies. In 1984, Lewis’s group and Pohl’s group have developed near-field microscopy independently [1,2]. Since then, there have been many reports of different types of scanning near-field optical microscopes (SNOM). However, the resolving powers of their probes are not over those of atomic force microscopes (AFM) or scanning tunneling microscopes (STM). The most advanced SNOMs, which have lateral resolutions less than 50 nm, have accepted the following probes: (1) optical fiber tip with a small aperture on the coated Al film as shown in Fig. 1(a) [3], (2) apertureless silicon tip with 2 nm radius in Fig. 1(b) [4], (3) polystyrene small sphere illuminated by evanescent light as shown in Fig. 1(c) [5], and (4) tetrahedral glass tip covered with gold thin film illuminated by surface plasmon in Fig. 1(d) [6]. Even if we use such smart probes, probe–sample distance control is an important issue in SNOM, and several kinds of the regulation methods have been developed. The most frequently used method is shear-force feedback used in AFMs. Such auxiliary control could lead to get artifacts in SNOM image [7], and without such control, high-resolution SNOMs have been developed by several groups [3,5].

In this article, we have observed the nanosteps on the flat cleaved facet of single crystal by using the SNOM system with a protrusion probe drawn in Fig. 1(c), and the lateral and vertical spatial resolutions are discussed through the obtained SNOM images.
2. Instrumentation

Fig. 2 shows the details of protrusion probe with a small polystyrene sphere used in the SNOM system. A polystyrene sphere with the diameter of 500 nm is located on top of a quartz substrate with quadrangular pyramidal shape. Since the incident laser beam totally reflected at the internal surface of apex of the pyramid, the generated evanescent wave illuminates the polystyrene sphere. When the sphere approaches very closely to the sample, the near field around the sphere excites dipoles on the sample surface and the far-field light wave is emitted into an objective lens. The intensity of this emission drastically changes depending on the distance between the sphere and the sample surface. It is particularly worth noticing that this optical system takes control of the probe–sample distance by optical feedback only. In other words, vertical position of the probe is controlled to keep the intensity of scattered light from the probe constant.

The schematic view around the probe for our SNOM system is shown in Fig. 3. The laser light source is a 5 mW He–Ne laser with the wavelength of 632.8 nm. The s-polarized laser beam is gradually focused on the apex of quartz substrate. A weak light scattered from the probe is detected by a photomultiplier tube (PMT) through microscope optics based on a conventional industrial microscope system. The sample surface is elevated by micro-mechanical stage. When the probe–sample distance becomes less than a few micrometers, interference fringes appears in the microscopic field with incident white light source. The intensity of scattered laser light from the probe increases drastically in the near-field region where the probe–sample distance is less than a few hundred nanometers. Then, we could start to measure a topographic image of the sample by keeping the intensity of scattered light constant. In this feedback system, (1) the PMT signal is smoothed by lowpass filter, (2) personal computer (PC) reads the PMT signal and compares it to a reference value, and (3) the PC adjusts the height of piezo stage to equalize the PMT signal with the reference value. This scan mode is nearly equal to a constant height mode, and we could obtain the topographic image of the sample surface [5].

3. Experiment

To estimate the spatial resolution of the SNOM system, authors observed the steps in river pattern on cleaved surface of KCl–KBr solid solution. This single crystal contained 40 mol% of KCl and 60 mol% of KBr, respectively. The KCl–KBr was grown from the melt of reagent-grade powders by the usual Kyropoulos method in air [8]. Cleavage of a single crystal along the symmetrical crystallographic planes \{1 0 0\} by impact forms “flat cleavage facets” on the resulting pieces. The cleaved surface of KCl–KBr was covered with a vacuum-deposited thin film of gold with the thickness of 50 nm, to enhance the light scattering caused by the interaction between probe and sample surface.

Fig. 4 shows the microscopic appearance of the cleaved surface of KCl–KBr. The cleaved surface had river pattern like dried-up creeks in the desert. In this microscopic field, we could recognize the small sphere probe illuminated by He–Ne laser because the s-polarized illumination produced
maximum scattered light from the sphere to eyepiece or PMT detector. Hence, the scan area was easily fixed, and this feature is one of the advantages, which arises from the coupling between conventional microscope and small sphere probe. On one of the lines in the river pattern, the observation area of $1 \times 1 \text{ mm}^2$ was scanned. The number of sampling was 50. The scan time was 100 min.

4. Results and discussion

Fig. 5 shows the magnified SNOM image of the small area in one of the river patterns shown in Fig. 4. Many white lines are observed from upper left to lower right in the scan area of $1 \mu \text{m} \times 1 \mu \text{m}$. These lines are believed to be steps at nanometer scales, which was produced by cleavage. The direction of the steps observed by SNOM was same as the direction of the river pattern observed in the microscopic field. Without the gold film, the sharper steps could be observed. In other words, when the film thickness is taken into account, the spatial resolution of the SNOM might be better value. Fig. 5 is a real topographic image because this image rotates by rotating the sample surface. Moreover, in Fig. 5, three parallel vague bands appeared in the vertical direction. Authors infer from stray lights coming from glass substrate as shown in Fig. 2 that the vague bands are caused by interference between them.
Because (1) they are vertical to the direction of the incident light, (2) the interval of the bands is almost 0.5λ, and (3) they get smaller when stray light is restrained.

In order to clarify the observed structure, the inclination of image in Fig. 5 was adjusted so that the terrace width might become the widest. As a result, the image plane was tilted at about 6°, and the resultant image is shown in Fig. 6. This tilt angle is acceptable because the cross section of river pattern might have an ensemble of many steps at nanometer scale and the sample surface might not be quite horizontal. Hence, the height difference between upper right corner and lower left corner becomes about 150 nm in Fig. 6. The contour lines are drawn at every 1 nm interval. The dense and sparse regions of contour lines make clear distinction between step and terrace. This image confirms that steps and terraces construct the river pattern on the cleaved surface, and the part of the terrace shows a (1 0 0) line-shaped structure to the horizontal direction. Hence, the steps were zigzag with an influence of the line structure. The difference of elevation of the step shows step height, and becomes the evaluation value of the vertical spatial resolution. The width or blur of the step part corresponds to the spatial resolution of the horizontal direction. Since the contour lines on the steps have a width of 20 nm, which is equal to the scanning step interval, the lateral spatial resolution of the present SNOM is estimated to be this value. If the scanning step interval is reduced, the limit of lateral resolution will be smaller than 20 nm. On the other hand, since the steps seem to have a height of about 5 nm, the vertical resolution must be 5 nm or less, but it seems to be larger than 2 nm by considering a noise level of PMT.

The highly resolved SNOM image gives us a question, “Why does a sphere probe of about 500 nm increase the microscopic resolution below a few tens of nanometers?” To find the answer, we treat the problem as electric dipole radiation on the probe surface. The dipole radiation travels as waves of oscillating electric and magnetic fields arising from an electric dipole. The electric fields \( E_\theta \) and \( E_z \) and the magnetic field \( H_\phi \) at the time of \( t \) are expressed by the following equations [9]:

\[
E_\theta = \frac{p}{4\pi}\frac{k^3}{\epsilon_0}\frac{\sin \theta}{r^3} + \frac{i}{k^2} - \frac{1}{k^3} \text{e}^{-i(\omega t - kr)} \tag{1}
\]

\[
E_z = \frac{p}{4\pi}\frac{k^3}{\mu_0}\frac{2\cos \theta}{r^3} + \frac{i}{k^2} - \frac{1}{k^3} \text{e}^{-i(\omega t - kr)} \tag{2}
\]

\[
H_\phi = \frac{p}{4\pi}\frac{k^3}{\epsilon_0}\frac{\sin \phi}{r^3} + \frac{i}{k^2} - \frac{1}{k^3} \text{e}^{-i(\omega t - kr)} \tag{3}
\]

where \( r, \theta, \) and \( \phi \) are spherical polar coordinates, and the electric dipole is placed at \( r = 0 \). The terms \( p \) and \( k \) are the electric dipole moment and the propagation constant of the wave, respectively. The terms \( \omega, \epsilon_0, \) and \( \mu_0 \) are the wave frequency, dielectric constant of vacuum, and permeability of vacuum, respectively. When \( kr \ll 1 \), the terms that vary inversely with the highest power of \( kr \) are dominant in Eqs. (1)–(3). The terms, which are proportional to \((kr)^{-3}\), are included in only Eqs. (1) and (2). As a result, only the electric fields \( E_\theta \) and \( E_z \) are dominant in the near-field region or the extreme narrow gap between sphere probe and sample surface. Therefore, we have checked the behavior of the term “\((kr)^{-3}\)” as shown in Fig. 7.

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**Fig. 6.** Topographical contour image obtained by tilting the image plane of Fig. 5 at an angle of few degrees. The dense and sparse contour-line regions correspond to step and terrace, respectively.

**Fig. 7.** Estimation of electric field based on dipole radiation theory between probe and sample surface: (a) actual configuration of probe surface near the sample surface and (b) curves of \((kr)^{-3}\) instead of near-field component \((kr)^{-3}\) in electromagnetic equations.
Fig. 7(a) is an actual dimension figure of the configuration of probe surface and sample surface, and Fig. 7(b) shows a normalized curve of the term “$$ (kr)^{-3} $$” versus horizontal position x. Instead of r, the curve is calculated with the vertical gap distance of z in each of x positions for simplicity. The vertical gap distance at x = 0 nm is indicated as $$ z_0 $$ in legend of Fig. 7(b). As shown in Fig. 7(b), authors regard an effective interaction area between the probe and sample surface in horizontal direction as FWHM of the curves. When a probe gets across a step edge, the probe could detect differences in the height of sample surface at the points 50 nm from the step edge in horizontal direction because the probe–sample configuration could be seemed to be same as shown in Fig. 7(a). In the horizontal interaction area, whose length is about 50 nm, the probe could detect a step edge in horizontal direction. That is to say, the horizontal spatial resolution is estimated to be 50 nm or less. This value is less than one-tenth of the probe diameter and agrees roughly with the spatial resolution achieved in the experiment. The rapid changes between three curves in Fig. 7(b) are caused by only 2 nm steps in vertical direction. Therefore, the vertical spatial resolution, which may be smaller than a few nanometers, can be also understood. There is a close relationship between the vertical and lateral resolution because the electric dipoles excited on the surface of sphere probe might actually interact with the sample surface tightly in such the small interaction area below diffraction limit.

5. Conclusions

Step and terrace structure have been observed in an area of 1 μm \( \times \) 1 μm on the cleaved surface of KCl–KBr solid solution. Vertical spatial resolution of the SNOM system is estimated to be 5 nm or less, but it seems to be more than 2 nm because of a noise level of PMT. Lateral spatial resolution of the system is estimated to be 20 nm or less. The exact value is not clear because the sample scanning step interval was 20 nm. The limit of lateral resolution will be studied by reducing the scanning step in the near future. In addition, the shot noise of light detection system will be suppressed by using the high-power laser light.

Acknowledgments

This work was partly supported by Grand-in-Aid for Scientific Research and 21st COE program of Ministry of Education, Culture, Sports, Science, and Technology of Japan.

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