JOURNAL OF APPLIED PHYSICS VOLUME 91, NUMBER 7 1 APRIL 2002

Optical second harmonic intensity images of a silver grating surface

H. Sano,[a)] M. Kume, and H. Nakagawa
School of Materials Science, Japan Advanced Institute of Science and Technology, Tatsunokuchi, Ishikawa 923-1292, Japan

G. Mizutani
School of Materials Science, Japan Advanced Institute of Science and Technology, Tatsunokuchi, Ishikawa 923-1292, Japan and "Fields and Reactions," PRESTO, Japan Science and Technology Corporation, 4-1-8 Hon-machi, Kawaguchi, Saitama 332-0012, Japan

(Received 2 October 2001; accepted for publication 11 January 2002)

We have obtained optical second harmonic (SH) intensity images of a silver grating surface. A stripe pattern was seen in the SH intensity image when the incident light polarization is perpendicular to the lines of the grooves of the grating. We have found that the strongest SH radiation comes from the bottoms of the grooves. On the other hand, the strong linear reflection comes from the slope of the grooves. We suggest that the enhancement of the electric field at the bottoms of the grooves has led to the observed strong SH radiation.
[DOI: 10.1063/1.1457537]

I. INTRODUCTION

Optical properties of metal microstructures and the electric field enhancement effect in such structures due to surface plasmon polariton (SPP) excitation are of great interest from points of view of both basic and applied research.[1,2] On one hand, the spatial distribution of the electric field will give a clue to understanding the optical response of SPPs in microstructures. On the other hand, the control of the spatial distribution of the electric field on a microstructured metal surface is very important in the development of high power laser and optical parametric generator systems containing metal gratings. This is because the damage of the metal grating can be caused by the light-induced intense electric field on the grating surface. So far, several theoretical studies on the electric field distribution on the illuminated metal grating surface have been reported.[3–7] However, there have been few examples of experimental studies.

In the present work, we have observed optical second harmonic (SH) intensity images of a silver grating surface, in order to investigate the spatial distribution of the electric field on the illuminated metal surface. The SH microscope can probe the surface electric field distribution without disturbing the local field at the surface, because surface SH response is so weak that the intensity of the incident electric field is hardly affected by this nonlinear interaction. Furthermore, the SH microscopy has an advantage of its contrast-enhanced images of field intensity on metal surfaces, since SH intensity is proportional to the square of the fundamental local field intensity.

So far, one trial of observing SH intensity images of a metal grating has been reported.[8] However, no obvious structure corresponding to the grooves of the grating was seen in the observed SH intensity images. In the present study we have obtained SH intensity images of a silver grating with clear stripe patterns.

II. EXPERIMENT

The sample was a blazed holographic grating with a line spacing of 200 lines/mm. A silver film of thickness 100 nm was evaporated on the grating surface by resistive heating in a vacuum jar. The evaporated film was sufficiently thicker than the penetration depth of the incident and SH light fields. In order to obtain the form factors of the grating, the surface profile of the grating was measured with an atomic force microscope (AFM). The observed AFM image and the profile of the grating are shown in Figs. 1(a) and 1(b), respectively. The result indicates that the grating has grooves with the periodicity of 5 μm, the depth of $\sim$1 μm, and the blaze angle of $\sim$14°. It is noted that surface roughness with an amplitude of about 50 nm was seen all over the gentler slopes of the grooves.

The setup of the reflection SH microscope system[9] used in the present study is schematically shown in Fig. 2. We have used a configuration of normal incidence and observation. The incident light for (SH generation) measurements was generated by a mode-locked Nd^{3+}: yttrium–aluminium–garnet laser with the wavelength of 1064 nm, the pulse duration of 30 ps, and repetition rate of 10Hz. It illuminated the area of a diameter of about 75 μm on the sample surface. The reflected SH light from the sample surface was passed through a bandpass filter with center wavelength of 532 nm, and was detected by a charge-coupled-device (CCD) camera with a time gated image intensifier. The accumulation time in the SH intensity image observation was 100 min. In addition to SH intensity images, linear reflection images at the incident wavelength of 532 nm were observed using the same setup. All optical measurements were performed in air at room temperature.

In order to calibrate the absolute positions in the SH and linear reflection images, we have observed a certain remark-

a)Author to whom correspondence should be addressed; electronic mail: h-sano@jaist.ac.jp

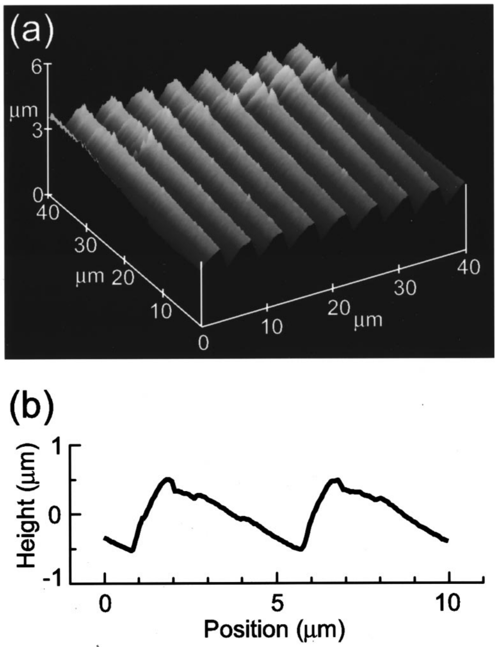

FIG. 1. (a) The AFM image and (b) the profile of a silver grating surface.

able scratch on the grating surface with both AFM and the SH microscope. Figures 3(a) and 3(d) are a photograph and a linear reflection image of the grating surface with the scratch, respectively. The photograph in Fig. 3(a) was taken by a conventional camera placed at the position of the CCD in the setup of Fig. 2. An ordinary light bulb was used to illuminate the sample for Figs. 3(a) and 3(d). Comparing the AFM, the photograph, and the reflection images of the scratch and the grooves, we have determined which parts in the SH and linear reflection images correspond to the tops and the bottoms of the grooves of the grating. The result is shown in Fig. 3(b). Comparing the reflection profile in Fig. 3(c) with the cross section of the grating in Fig. 3(b), we find that bright and dark stripes in Figs. 3(a) and 3(d) originate from the steep and gentle slopes of the grooves of the grating, respectively.

The spatial resolution of the SH microscope equipped with the conventional camera is about 0.5 μm, since fine structures with sizes of about 0.5 μm can be seen in Fig. 3(a). The spatial resolution of the SH microscope equipped with the CCD camera is estimated to be between 0.5 and 1 μm from Fig. 3(d). This spatial resolution of the SH microscope does not differ greatly from the spacing in the grating. However, Fig. 3 demonstrates that we can unambiguously judge from where on the grating the strong reflected light is coming.

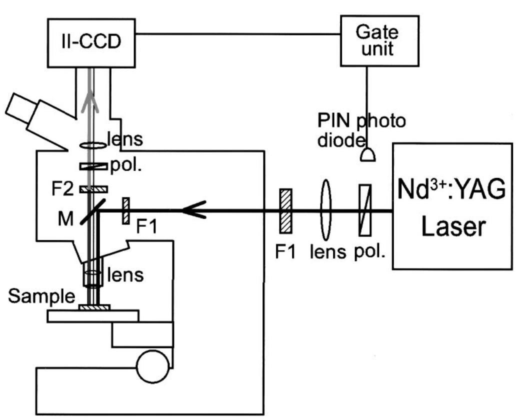

FIG. 2. Reflection SH microscope system: ($F1$) visible cut filter; ($F2$) bandpass filter; and (M) dichroic mirror.

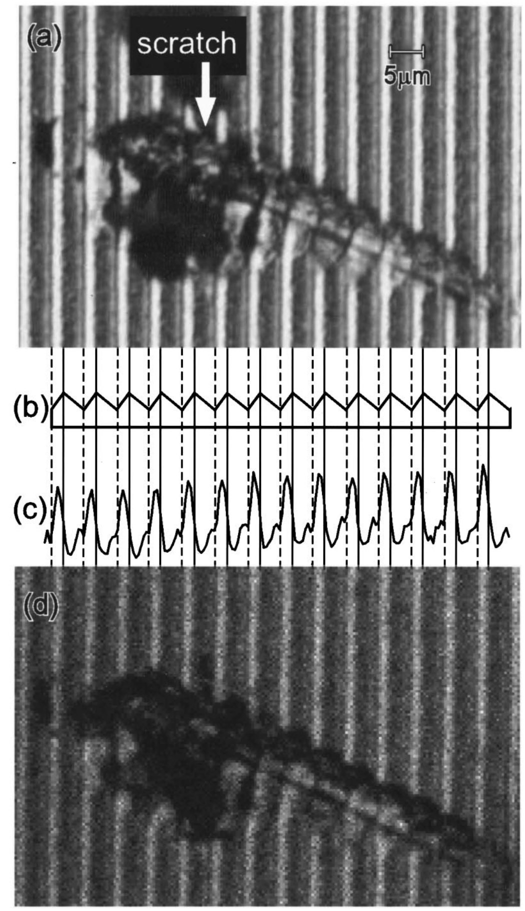

FIG. 3. (a) Photograph and (d) linear reflection image of a scratch on a grating surface, (b) cross section of the grating, and (c) profile of the linear reflection. Vertical solid and dashed lines on the cross section indicate the position of the top and the bottom of the grooves, respectively.

III. RESULTS AND DISCUSSION

Figures 4(a) and 4(b) are linear reflection images observed with the incident light polarizations perpendicular and parallel to the lines of the grooves, respectively. White dots in the images indicate observed photons. Clear stripe patterns are seen in both polarization configurations. These linear reflection images in two polarization configurations show no difference from each other in their patterns or their intensities. Here we note that weak replica images shifted upward are overlapped with the main images. These replica images result from multireflection of an optical filter in the microscope system. This effect leads to no serious problem in the later discussion, because the overlapped images are shifted in the direction exactly parallel to the grooves.

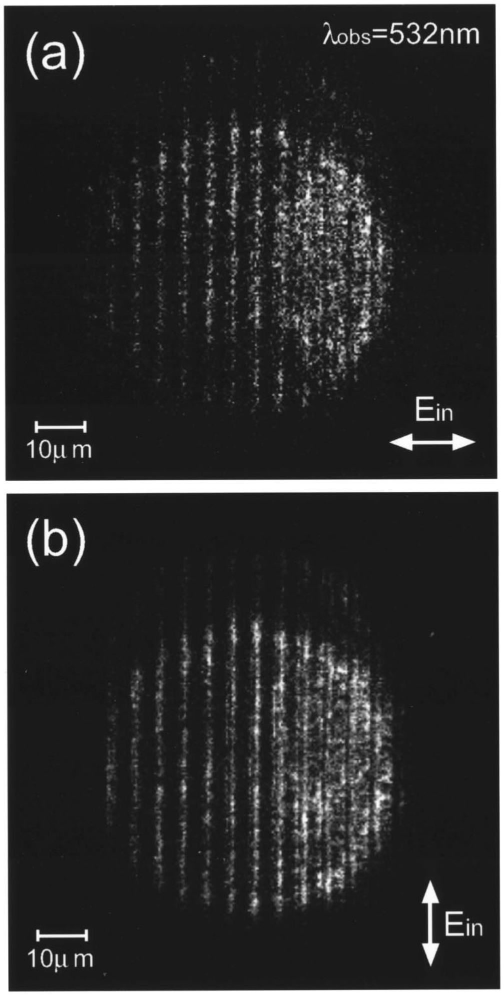

FIG. 4. Linear reflection images of a silver grating surface for the incident light polarizations: (a) perpendicular and (b) parallel to the lines of the grooves. The wavelength of the incident light was 532 nm.

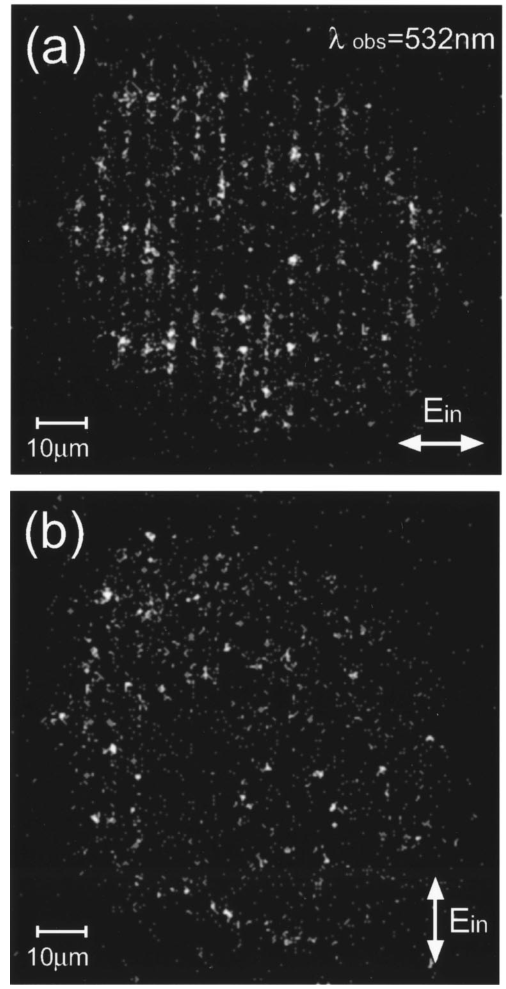

FIG. 5. SH intensity images of a silver grating surface for the incident light polarizations (a) perpendicular and (b) parallel to the lines of the grooves.

SH intensity images of the silver grating observed with the incident light polarizations perpendicular and parallel to the lines of the grooves are shown in Figs. 5(a) and 5(b), respectively. A stripe pattern is seen in Fig. 5(a), while no obvious structure is seen in Fig. 5(b). Total SH intensity in Fig. 5(a) is larger than that in Fig. 5(b).

Let us discuss the SPP excitation as the candidate origin of this polarization dependence of the SH intensity images. SPP has a large momentum component parallel to the metal surface and its electric field is polarized in the plane including the momentum vector and the surface normal. Since the corrugation of the grating gives a large momentum component in the direction perpendicular to the lines of the grooves, SPP can be excited by the incident light with the polarization perpendicular to the lines of the grooves. Thus the enhancement of SH intensity due to SPP is expected in the result shown in Fig. 5(a).

Light intensity profiles obtained by integrating the signal intensity in the direction of the lines of the grooves are shown in Fig. 6. Figures 6(a), 6(b), 6(c), and 6(d) are SH intensity and linear reflection profiles obtained from the images in Figs. 5(a), 5(b), 4(a), and 4(b), respectively. The absolute peak positions in Fig. 6 were determined in the same way as in Fig. 3. Since peak positions in Fig. 6(a) do not agree with those in Figs. 6(c) and 6(d), the SH intensity pattern cannot be explained by the spatial variation of the linear reflectivity from the grating surface alone. There must be a nonuniform field enhancement effect. Because the peaks of the SH intensity are located at the bottoms of the grooves in Fig. 6(a), the electric field is probably enhanced at the bottoms. This result is surprising because it is said that the local electric field is generally the largest near the top of a metallic protrusion.[10]

Now, let us discuss the mechanism of the field enhancement at the bottoms of the grooves. Since it is well known that roughness on a metal surface induces the SPP excitation and enhances the electric field,[11] the origin of the field enhancement may be surface roughness distributed near the bottoms of the grooves. However, as mentioned in Sec. II, the measured AFM image indicates that roughness is distributed over the whole surface of the gentler slopes of the grooves. Thus the surface roughness is not the origin of the field enhancement at the bottoms in the present case.

Next we consider several theoretical studies on the spatial distribution of the electric field on the metal grating. Reference 7 shows that the electric field on the metal sinusoidal grating with the periodicity of 800 nm and the ampli-

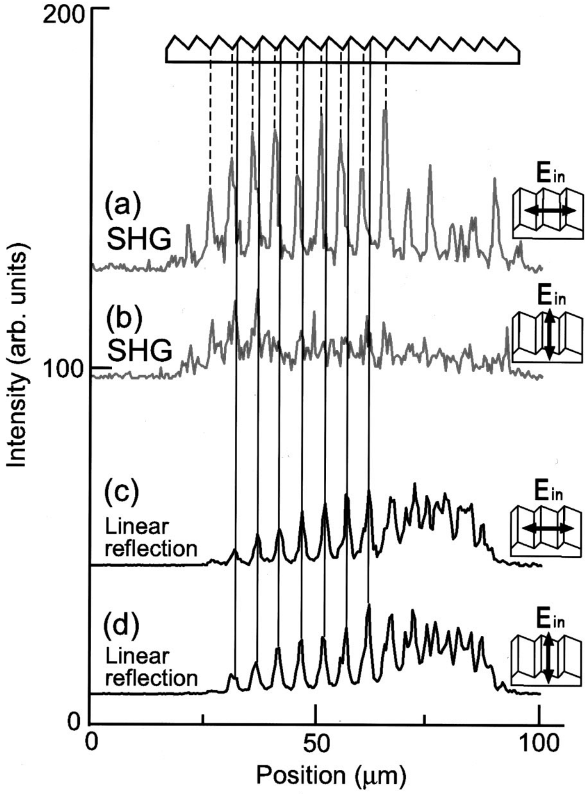

FIG. 6. Profiles of the SH intensity measured with the incident light polarizations: (a) perpendicular and (b) parallel to the lines of the grooves, and profiles of linear reflection measured with the incident light polarizations (c) perpendicular and (d) parallel to the lines of the grooves.

tude of 440 nm is enhanced at the top and the middle parts of the slopes of the grooves. Although the ratio of the height to the periodicity of the grooves in this calculation is not far from that in the present study, the calculation does not agree with our experimental data. According to theoretical studies,[3–6] the field enhancement at the bottoms of the grooves on a metal grating occurs in two cases: (1) when the height of the grooves is larger than the periodicity,[3,5,6] and (2) when a wave vector of the grating is twice as large as that of the SPP mode.[4] In case (2), the photonic band gap is created in the dispersion by Bragg scattering of the SPP, and the field maxima of the SPP standing wave of the high energy branch are located at the bottoms of the grooves. However, experimental parameters in the present study are far from the ones of the above two cases. Namely, the grating used in this study has grooves with a height of 1 μm and periodicity of 5 μm so that these form factors do not match the condition in case (1). The wave vector of the grating, 2000 cm^{-1}, is much less than that of the SPP mode estimated to be $\sim$9530 and $\sim$19880 cm^{-1} for the incident and SH light excitations, respectively. These parameters do not match the condition in case (2). Nevertheless, it is possible that these two effects partly contribute to the observed SH enhancement.

At present it seems to us that all the electromagnetic calculations we have referred to[3–7] contradict our experimental result. A probable reason for this contradiction is that the near-field effect or higher-order electromagnetic multipolar effect might not have been correctly estimated in the calculations. The electromagnetic multipolar interactions in the nonlinear optical process[12] usually give very small contribution to the SH response, but this multipolar effect might be enhanced at the bottoms of the grooves in the present case. In order to understand the result in the present study, further theoretical studies taking these effects into account are necessary.

IV. CONCLUSION

We have obtained optical SH intensity images of a silver grating surface. It has been shown that intense SH light is generated at the bottoms of the grooves of the grating. This result cannot be explained by geometrical optics of the SH light generated on the grating surface alone. The field enhancement at the bottoms of the grooves is suggested. At present, the mechanism of this field localization at the bottoms of the grooves is not completely understood.

ACKNOWLEDGMENT

The author would like to acknowledge valuable discussions with Associate Professor T. Inaoka of Iwate University.

[1] *Surface Polaritons*, edited by V. M. Agranovich and D. L. Mills (North-Holland, Amsterdam, 1982).
[2] H. Kano and S. Kawata, Appl. Opt. **33**, 5166 (1994), and references therein.
[3] D. Agassi and T. F. George, Surf. Sci. **172**, 230 (1986).
[4] W. L. Barnes, T. W. Preist, S. C. Kitson, J. R. Sambles, N. P. K. Cotter, and D. J. Nash, Phys. Rev. B **51**, 11164 (1995).
[5] F. J. García-Vidal and J. B. Pendy, Phys. Rev. Lett. **77**, 1163 (1996).
[6] M. B. Sobnack, W. C. Tan, N. P. Wanstall, T. W. Preist, and J. R. Sambles, Phys. Rev. Lett. **80**, 5667 (1998).
[7] R. A. Watts, T. W. Preist, and J. R. Sambles, Phys. Rev. Lett. **79**, 3978 (1997).
[8] K. L. Haller, L. A. Bumm, R. I. Altkorn, E. J. Zeman, G. C. Schatz, and R. P. Van Duyne, J. Chem. Phys. **90**, 1237 (1989).
[9] H. Sano, T. Shimizu, G. Mizutani, and S. Ushioda, J. Appl. Phys. **87**, 1614 (2000).
[10] D. P. Woodruff and T. A. Delchar, *Modern Techniques of Surface Science*, 2nd ed. (Cambridge University Press, New York, 1994), pp. 414–417.
[11] A. Otto, *Light Scattering in Solids IV* (Springer, Berlin, 1984), pp. 289–418.
[12] P. Guyot-Sionnest and Y. R. Shen, Phys. Rev. B **38**, 7985 (1988).