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Laser Writing of a Subwavelength Structure on Silicon (100) Surfaces with Particle-Enhanced Optical Irradiation[1]

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Spherical 0.5-µm silica particles were placed on a silicon (100) substrate. After laser illumination with a 248-nm KrF excimer laser, hillocks with size of about 100 nm were obtained at the original position of the particles. The mechanism of the formation of the subwavelength structure pattern was investigated and found to be the near-field optical resonance effect induced by particles on the surface. Theoretically calculated near-field light intensity distribution was presented, which was in agreement with the experimental result. The method of particle-enhanced laser irradiation has potential applications in nanolithography. © *2000 MAIK "Nauka/Interperiodica".*

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The field of nanoelectronics has evolved into a major area of investigation. Nanolithographic techniques such as atom beams [1, 2], electron beams [3], scanning probe tunneling [4–6], and scanning near-field optical lithography [7–9] are expected to be potential methods in the fabrication of present and future nanodevices. However, due to their incompatibility with the present fabrication processing and their low throughput, the application of these methods is presently confined to the experimental stage. Meanwhile, traditional optical lithography is limited to the diffraction effect and always relates to complex system and high cost. In this letter, we report a novel, low-cost, and simple optical lithography technique using particle-enhanced laser irradiation.

Standard spherical silica particles (Duke [10]) packaged as low-residue aqueous suspensions were used in our experiment. The diameter of the particles is 0.5 µm, with a deviation limited in a range of ±5%. Silicon (100) samples were dipped in 5% hydrofluoric acid for 20 seconds of hydrogen passivation and rinsed with DI water afterwards. Particles were applied on Si (100) surfaces and dried by a hot air jet. The location of the particles on the substrate before laser irradiation was observed with an optical microscope. As shown in Fig. 1a, the particles are spherical and smooth and many of them are free of aggregation.

A 248-nm KrF excimer laser with pulse duration of 23 ns was used. In order to produce an observable laser-induced pattern, beam intensity in the area of the sample was adjusted to about 340 mJ/cm^2. The laser beam was incident normally on the Si (100) surface. The pulse number was 200. The sample, after laser irradiation, was observed again with the optical microscope, as shown in Fig. 1b. Comparison of the two pictures, i.e., before and after irradiation, showed that localized black spots (Fig. 1b) appeared at the original position of the particles (Fig. 1a). In the two pictures, the original particle position (Fig. 1a) and the observed corresponding black spots (Fig. 1b) are marked by curves.

Field-emission SEM was used to track the black spots that corresponded to the original particle position. The SEM image (Fig. 2a) shows the black spot observed in Fig. 1 as a circular hillock, and a curve around the hillock can be clearly seen. This curve around the hillock can also be seen in Fig. 2b, the AFM observation. This indicates that the hillocks were produced due to melting of the "hot point." The size of the hillock is about 100 nm, with height about 10 nm.

The same laser fluence (340 mJ/cm^2) was also applied on clean Si (100) surfaces (without particles), but no damage spots were observed. It is obvious that higher intensity was achieved due to the spherical particles in our experiment, and this resulted in the formation of the hillocks. The mechanism can be explained as the enhancement of light intensity near the contact area [11–13]. Since the characteristic distance between particles and substrate is smaller than the radiation wave-

[1] This article was submitted by the authors in English.

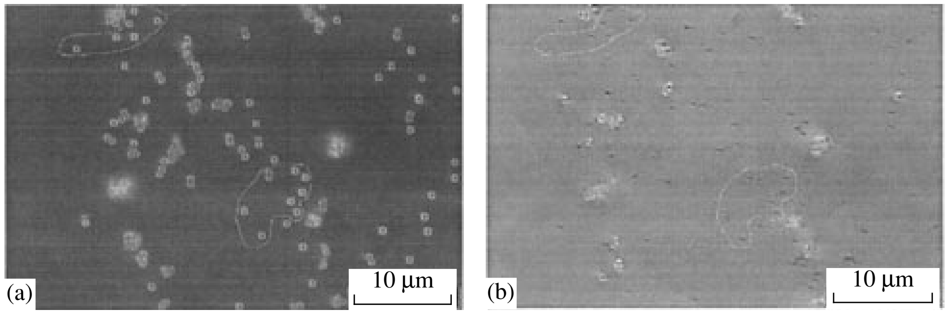

Fig. 1. Optical microscope observation of the localized hillocks induced by particle-enhanced laser irradiation: surface view (a) before and (b) after irradiation.

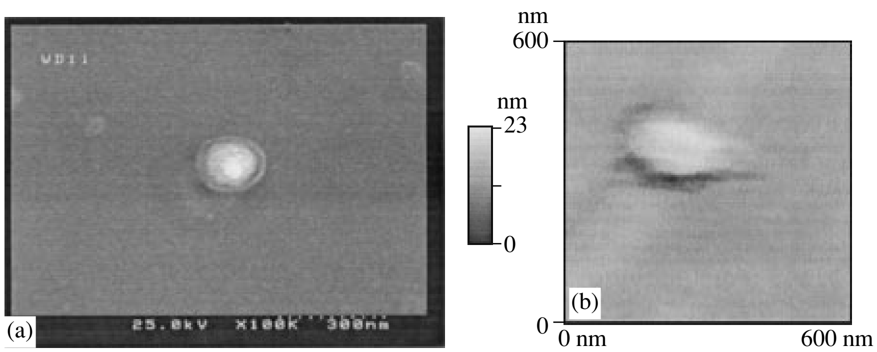

Fig. 2. Image of the hillock structure: (a) SEM and (b) AFM. These pictures present different hillocks.

length, and the particle size is of the order of a wavelength, particles do not simply play the role of microfocusing lens as in far-field, but relate to the optical resonance effect in near-field. The source of the optical resonance is excitation of partial waves (multipole modes of a spherical cavity) [14]. The optical resonance produces a high-intensity zone in the near-field region, and, naturally, when this high-intensity zone is on the substrate surface, it can lead to formation of "hot points."[1] These "hot points" produced the hillocks.

The light intensity on the surface under the spherical particle was calculated by solving the electromagnetic boundary problem "particle on surface" [12, 15, 16]. "Mathematica-4" [17] was used for calculations. Figure 3 shows the intensity distribution within the plane perpendicular to the wave vector of the incident wave. This intensity I is defined as the normalized value of the z-component of the time-averaged Poynting vector. From the picture, a strong optical enhancement can be seen. The intensity of light in the center is 20 times greater than the incident intensity. The achievement of the higher intensity according to calculations was clearly observed in our experiment. This computational result can also explain the work done by Kane and Halfpenny [18], in which the damage threshold of glass substrates was reduced by smaller surface particle coverage. The full width at half maximum (FWHM) in the intensity distribution is shown in Fig. 3. The FWHM is equal to 80 nm, which agrees with the hillock size of 100 nm. This confirmed that a subwavelength structure was achieved in our experiment.

The small nanoparticles have potentially useful optical, optoelectonic, and material properties that might lead to application in nanostructure fabrication [19]. Although the 0.5 μm (submicron) spherical particles discussed in the present paper work by a different mechanism, they too have the same potential in nanofabrication by producing enhanced light intensity in the near field. Further theoretical calculation shows that even higher line resolution below 100 nm can be achieved by selecting a suitable particle size and wavelength. We do not discuss here the technical problem related to particle arrangement and positioning.

In conclusion, we have reported a novel lithographic technique where particles were applied on a silicon surface and a subwavelength structure was achieved with particle-enhanced laser irradiation. The mechanism was found to be the near-field optical resonance effect induced by particles on the surface. Calculation result was presented by solving the electromagnetic boundary problem. Compared to other nanolithographic techniques, the method of particle-enhanced laser irradiation does not need a complex system and uses neither a

[1] It follows from the Mie theory calculations [11–13], as well as from more precise calculations [12], which take into account the secondary scattering of radiation reflected by the substrate.

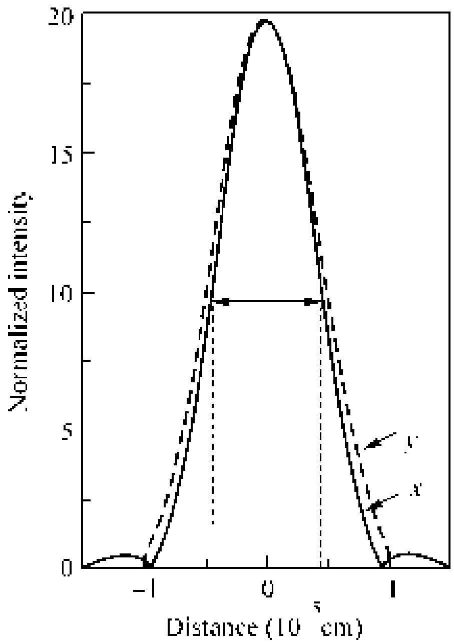

Fig. 3. The light intensity distributions (z-component of the averaged Poynting vector) on the substrate surface under the spherical particle (calculations with the Mie theory). Vector **E** of the incident plane wave is directed along the x axis, vector **H** along the y axis. Distribution of the surface intensity along the x axis is shown by a solid line and along the y axis by a dashed line.

mask nor a resist, but simply relies on the near-field optical resonance effect to achieve higher resolution and intensity. Hence, it, may have greater potential in nanofabrication.

Addition: When this paper was ready for publication, we received information from the Konstanz and Madrid groups that they had also found a similar optical field-enhancement effect using shorter laser pulses (from 150 fs to 6.5 ns) [20].

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