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Plasma-assisted laser ablation of tungsten: Reduction in ablation power threshold due to bursting of holes/bubbles

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Nanosecond laser ablation of tungsten (W) exposed to helium plasma is investigated using optical emission spectroscopy. Submicrometer-sized holes/bubbles are formed on the surface of W when it was exposed to the helium plasma at a sufficiently high temperature ($\gtrsim$1500–1600 K). The emissions from a virgin W (before the helium plasma irradiation) cannot be detected when the fluence is $<1\ \mathrm{J/cm^2}$; however, the threshold fluence for the detection of neutral W emission after it was exposed to the helium plasma is $\sim 0.2\ \mathrm{J/cm^2}$. The physical mechanism of laser-induced bursting of holes/bubbles is proposed for achieving a significant reduction in ablation power threshold. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2824873]

Tungsten (W) metal has many useful properties such as a high melting point and low sputtering yield. It has been used in areas where high heat and particle loads are concentrated. However, it has been revealed that the irradiation of helium (He) plasma/ions damages the tungsten surface by forming submicrometer-sized structures even if the incident ion energy is significantly lower than the physical sputtering energy.[1] Formation of holes/bubbles on tungsten surface is observed for surface temperatures higher than ~1600 K and a helium ion fluence of more than 10^{25}–$10^{26}\ \mathrm{m^{-2}}$.[2,3] Under some other conditions, it has been demonstrated that nanostructured tungsten with an arborescent shape is formed on a material surface.[4,5] Such damages degrade the optical reflectivity[6] and thermophysical properties of the surface.[4] In fusion experiments, such damages can pose problems with regard to in-vessel metal mirrors[7] and plasma-facing components. On the other hand, the synergetic effects of plasma and laser irradiation may aid in extending the practical application of the laser ablation technique, which is a powerful technique for the deposition of thin films,[8] formation of nanoparticles, welding and bonding of metal parts, and so on. It has thus far been shown that synergetic plasma irradiation can assist the crystal growth in the vapor phase for the laser ablation of boron nitride.[9] In the present study, the synergetic helium-plasma irradiation effects on the laser ablation of W are studied by optical emission spectroscopy, particularly from the viewpoint of surface roughening by the plasma irradiation. It is noteworthy that the synergetic irradiation of the helium plasma significantly reduces the threshold power for the laser ablation of W.

Figure 1(a) shows the schematic of the experimental setup in the linear plasma device Nagoya divertor simulator (NAGDIS)-II.[10] A high-density plasma is generated by a dc arc discharge between a LaB_6 cathode and anode with helium, hydrogen, or deuterium gas as the plasma source. Powder metallurgy W (PM-W) samples with a thickness of 0.2 mm were used as specimens. The specimens were inclined at an angle of approximately 45° to the magnetic field line. The second harmonic of a neodymium-doped yttrium aluminum garnet laser ($\lambda = 532$ nm) with a Q switch (Continuum: SLII-10) was used for the experiments. The pulse width and repetition frequency of the laser are 5–7 ns and 10 Hz, respectively. In the experiments, the laser fluence was varied in the range of $<2\ \mathrm{J/cm^2}$. The actual pulse energy was <0.5 J, and the laser diameter was approximately 5 mm. The transient emission around the specimen in response to the laser pulses was detected using an image-intensified charge coupled device (Hamamatsu: C-3554) with a narrow-band optical filter. Two types of optical filters were used, one for the measurement of W I ($\lambda = 400.9$ nm) and the other for the measurement of He I ($\lambda = 706.5$ nm). Figures 1(b) and 1(c) show the typical W I and He I spectra observed with a Czerny-Turner-type spectrometer in the helium plasma. The values of the full width at half maximum of the transmittance are ~1, and ~4 nm for W I and He I, respectively.

Figure 2 shows the scanning electron microscopy (SEM) micrographs of the tungsten surface exposed to the helium plasma at a surface temperature of 2200 K. The temperature was measured with a radiation pyrometer. The fluence and incident energy of the helium ions were $\sim 8 \times 10^{25}\ \mathrm{m^{-2}}$ and 25 eV, respectively. We can observe many holes on the surface. The mechanism of the hole formation is thought to be the swelling of the nanometer-sized bubbles formed by the interaction of the helium atoms with the thermal vacancies.[1] This study focuses on the interaction between the laser

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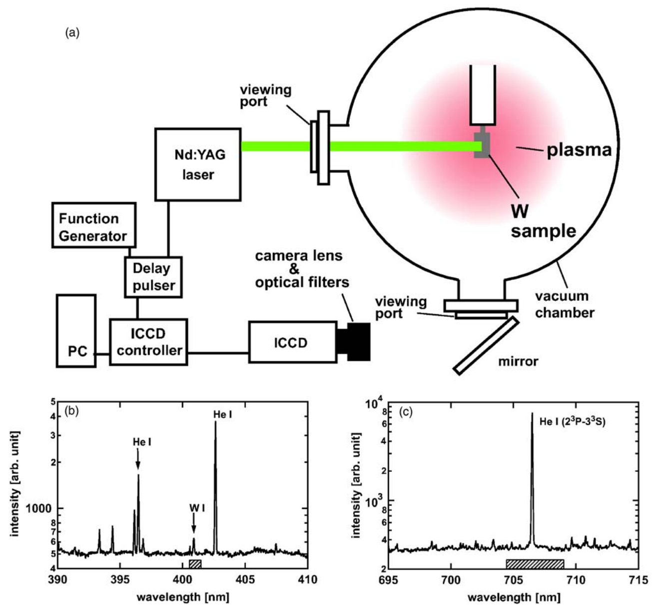

FIG. 1. (Color online) (a) Schematic diagram of experimental setup in the linear plasma device NAGDIS-II. Typical spectra of (b) W I and (c) He I used for the measurement.

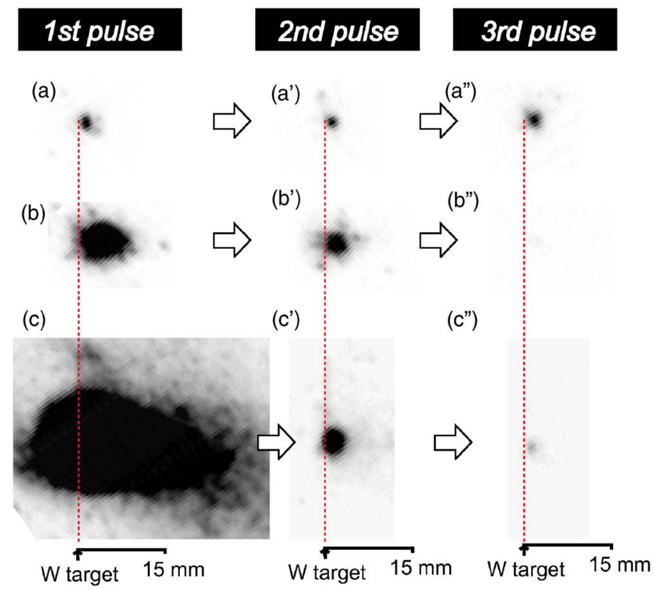

FIG. 3. (Color online) Emission profiles of the ejected particles from the target in response to three sequential laser pulses. [(a)–(a″)] Emission of W I in vacuum from the specimen preexposed to D_2 plasma, [(b)–(b″)] emission of W I in vacuum from the specimen preexposed to He plasma, and [(c)–(c″)] emission of He I in D_2 plasma from the specimen preexposed to helium plasma. The laser pulse energies in (a)–(a″), (b)–(b″), and (c)–(c″) were 1.2, 0.8, and 1.4 J/cm^2, respectively.

pulses and the damaged W material with submicrmeter-sized holes shown in Fig. 2.

Figures 3(a) and 3(a″) show the emission profiles of W I in vacuum in response to three successive laser pulses from the W specimen not exposed to the helium plasma. The laser fluence is 1.2 J/cm^2, and the gate width of the image intensifier is 10 μs, which is sufficiently large to detect the total emission due to a laser pulse. The W I emission is observed in vacuum, indicating that not only the vaporization but also the laser ablation accompanying electronic excitation occur in response to the laser pulses. Figures 3(b) and 3(b″) show the W I emission profiles in response to three successive pulses incident on a tungsten specimen that had been preexposed to the helium plasma. The W I emission is clearly observed in response to the first pulse in Fig. 3(b), even though the fluence is 0.8 J/cm^2, which was lower than the onset laser fluence to observe the emission from the W specimen without the helium-plasma irradiation. Figures 3(c)-3(c″), show the emission profiles of He I in D_2 plasma from the specimen preexposed to the helium plasma. The D_2 plasma is used to aid the visualization of the helium atoms emitted from the specimen. It can be observed that a large amount of helium gas is released with a bright emission in a relatively large area in response to the first laser pulse. Such an emission that expands to a large area cannot be observed with a W I filter, even though the irradiation is carried in the D_2 plasma at the same laser fluence as the case shown in Fig. 3(c). Thus, the emission in a large area shown in Fig. 3(c) can probably be attributed to the difference in the atomic masses between helium and tungsten. In both Figs. 3(b)-3(b″) and 3(c)-3(c″), the emission intensity decreased at the second pulse; the emission could not be observed thereafter. It is speculated that the surface holes/bubbles disappeared or significantly reduced after several laser pulses; consequently, the deposited helium gas was released, and the ablation threshold may recover to the original W level.

When the specimen was simultaneously irradiated with the laser pulses and helium plasma, emission was continuously observed in response to the laser pulses. This can be attributed to the repetitive formation and bursting of the holes/bubbles on the surface.[11] Figure 4(d) shows the laser fluence dependence of the emission intensity of W I. In cases (i)-(iii), the specimens were preexposed to the hydrogen plasma for 1800 s at a surface temperature of 1500 K. Additionally, in cases (ii) and (iii), the specimen was preexposed to helium-hydrogen plasmas with ion fluxes of $4.3\times10^{22}\ m^{-2}\,s^{-1}$ for 1800 s and $7.0\times10^{22}\ m^{-2}\,s^{-1}$ for 5400 s, respectively. Figures 4(a)–4(c) show the SEM images of the nonlaser-irradiated part of the surface after experiments (i), (ii), and (iii), respectively. The surface is almost flat in Fig. 4(a), whereas it becomes rough with many pinholes when the specimen is exposed to the helium plasma. The position of the laser irradiation spot did not change during the experiments, and the fluence gradually increased from ~0.1 to ~1.5 J/cm^2. In case (i), the emission is clearly observed from >1 J/cm^2, while the onset laser fluence decreases to ~0.5 J/cm^2 in case (ii) and to ~0.2–0.3 J/cm^2 in case (iii). The irradiation of the helium plasma significantly reduces the threshold laser fluence for the ablation.

The ideal specular reflectance of the W surface at 532 nm at room temperature is approximately 50%;[12] therefore, the deposited energy should be comparable to or greater

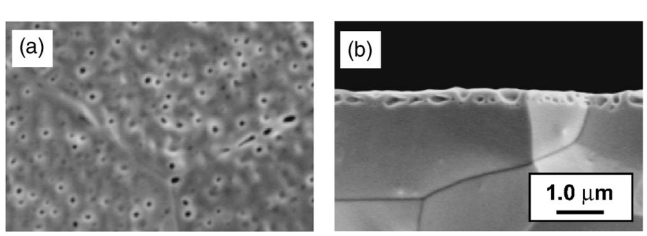

FIG. 2. SEM micrographs of tungsten surface exposed to helium plasma. (a) Top view and (b) cross-sectional view.

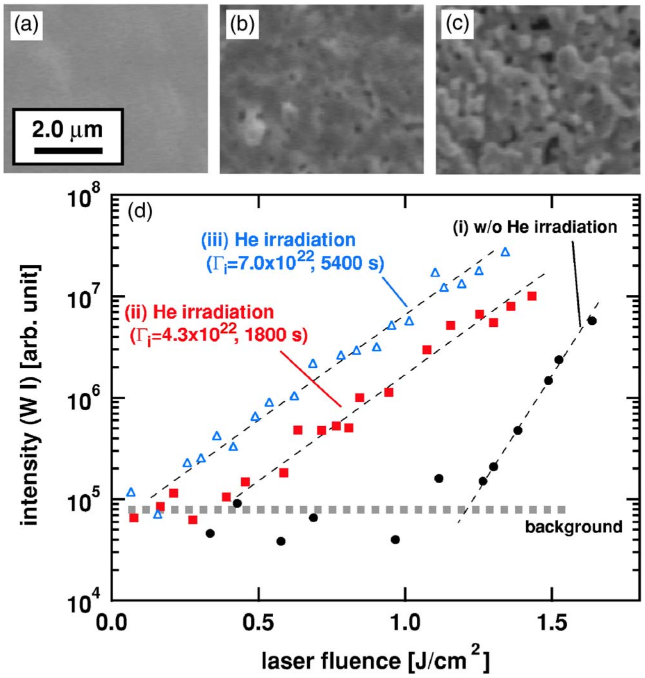

FIG. 4. (Color online) SEM micrographs of PM-W after the irradiation in (a) hydrogen plasma and [(b) and (c)] helium-hydrogen mixture plasma. (d) Laser power dependence of the emission intensity.

than 50% of the incident energy when a nonspecular surface is considered. It has been pointed out that the plasma irradiation reduces the surface reflectivity.[6] However, the threshold reduces to considerably less than half of that in Fig. 4(d), indicating that the reduction in the threshold laser fluence cannot be explained by only the increase in the energy deposited on the surface. It can be said that the reduction in the power threshold is deeply related to the existence of the holes/bubbles. Figures 5(a)–5(c) show the schematic of the bursting of a hole containing high-pressure helium gas. If the temperature of the lid of hole is sufficiently high, the bursting occurs because the stress exerted on the lid of the hole due to the high pressure in the holes may exceed the tensile stress of tungsten.[11] For example, for a hole with a radius of 100 nm, a lid thinner than 100 nm can easily burst when the lid temperature is ~2400 K.[11] Figure 3(c) shows the direct evidence of the bursting of the helium holes in response to the laser pulse irradiation. It is suggested that the tungsten droplets possibly splash on the plasmas, accompanied by the bursting of the holes/bubbles. It is thought that superheating of the droplets and the extremities of the surface structures formed by bubble bursting could lead to an increased ablation efficiency.

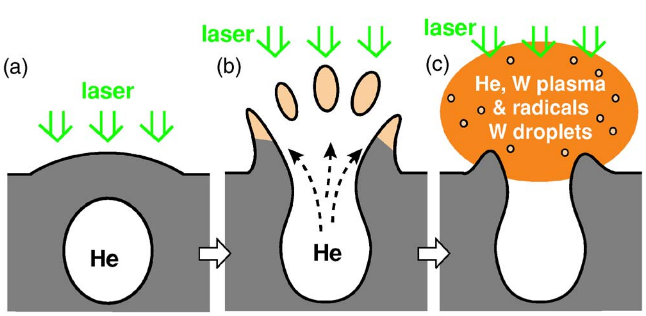

FIG. 5. (Color online) Schematic of the bursting of a hole containing high-pressure helium gas.

Regarding the difference between cases (ii) and (iii), two factors should be taken into account. One is the difference in the helium fluences before the laser irradiation. The fluences in cases (ii) and (iii) are 7.7×10^{25} and 3.8×10^{26} m^{-2}, respectively. A fraction of the holes/bubbles might remain after the laser irradiation when the laser fluence was not high, and the remaining preexposure record might cause the difference. The other factor is the difference in the fluxes. In case (ii), since the flux is approximately two times more, more helium holes/bubbles must be formed in between the laser pulses. It is noteworthy that the emission intensities are different for every pulse in cases (ii) and (iii), probably because the extent of bursting of the holes/bubbles changes on pulse-by-pulse basis. Thus, in Fig. 4(d), the intensity was obtained by averaging 50 images.

The reduction in the ablation threshold may pose a problem for the in-vessel laser transmission mirror in fusion experiments such as ITER and in the inertial fusion reactors. On the other hand, it is expected that the synergetic irradiation of plasmas and lasers can be used for enhancing the efficiency of the pulsed-laser deposition. Thus, for future studies, the investigations using different materials such as molybdenum, gold, and silver for the in-vessel mirror material are important. Moreover, the synergetic effect using materials such as $YBa_2Cu_3O_{7-x}$ superconductors and $Ba_xSr_{1-x}TiO_3$ ferroelectric thin films is interesting for industrial applications.

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