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Influence of target chemical activity on Balmer lines emission from backscattered hydrogen

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Abstract

We have investigated the influence of implanted hydrogen on Balmer lines emission from backscattered particles under D^+ irradiation to Si target for an incident energy ranging from 5 to 25 keV at a target temperature from room temperature (RT) to 600 K and the experimental results are compared with Monte Carlo simulation by the TRIM92 code. For clean Si surface, D_α photon intensity was proportional to the backscattering coefficients calculated by the TRIM92 code for all incident energies. The intensity stayed constant with increasing the fluence for incident energies of 15 and 25 keV in accordance with the simulation that the backscattering coefficient is not so much influenced by implanted deuterium because the mass of D is much less than that of Si. At 5 keV incidence, the D_α photon intensity gradually increased with the fluence until saturated after prolonged irradiation. The difference between the initial intensity and that after saturated was reduced with increasing the target temperature and disappeared above 600 K. The decay of the D_α photon intensity accompanied by the thermal release of implanted deuterium was also observed above 370 K. According to the TRIM92 calculation, the D/Si atomic ratio at the top surface for 5 keV incidence with the fluence of 4×10^{17} D^+/cm^2 exceeds 0.4 which is the maximum hydrogen concentration of amorphous hydrogenated Si (a:Si–H), whereas that for 15 keV incidence remains below 0.1 even over the 7×10^{17} D^+/cm^2 irradiation. All these results made us to conclude that the D_α intensity at 5 keV incidence is enhanced by retained deuterium near the top surface through modifying the electron capture process of backscattered deuteron.

1. Introduction

Hydrogen recycling process at a limiter or divertor target in fusion system plays an important role not only in boundary plasma physics but also in engineering aspects. To determine a hydrogen recycling coefficient, the Balmer lines emission of hydrogen (H_α, H_β, H_γ...) in plasma has been utilized [1–3]. Generally these lines emission is assumed to be caused by the electron excitation of neutral atoms and molecules in the boundary plasma. However there is a certain contribution of hydrogen directly backscattered in excited state from the target surface. Although backscattering of energetic hydrogen at the solid surfaces has been extensively studied [4,5], such lines emission from the backscattered hydrogen atoms has not been systematically investigated. Recently we have shown that atoms are backscattered in excited states and directly emit Balmer lines [6,7]. Since the excited atoms are produced through an electron capture at the top surface, the emission must be influenced by the hydrogen implanted at the top surface. Therefore it is not possible to tell how large the contribution of direct emission in the total Balmer lines in the boundary plasma is and how the implanted hydrogen in the first-wall influences the intensity of the emission.

In this work we have investigated the influence of deuterium accumulated near the target surface on the Balmer lines emission from the backscattered atoms under irradiation of D^+ to Si target, of which electronic state is known to be modified by implanted hydrogen.

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Although pure silicon may not be the first wall mounted in TEXTOR, siliconization of graphite wall has been occasionally done and gives good results [8].

2. Experimental

Experiments were carried out in a standard vacuum chamber equipped with an ion source and an optical spectrometer [6]. A differential pumping system allows the pressure in the target chamber to be kept below 10^{-6} Pa during the irradiation. Mass analyzed deuterium ions (D^+) accelerated up to 25 keV were injected to a Si target (1 0 0) through a slit of 5 mm diameter with an incident angle of 45° with respect to the target normal. The injected ion flux, monitored by a Faraday cup inserted to the beam line in front of the target, was ranging from 1×10^{13} to 1×10^{14} D^+/cm^2s. The specimen temperature was controlled by a heater behind the target in temperature range from room temperature (RT) to 600 K. The emitted photons from backscattered particles were focused into an optical spectrometer (Spectra Pro 275) whose optical axis was normal to the ion beam and detected with a multi-channel analyzer (OMAIII). The Balmer lines of D_α, D_β, D_γ were simultaneously measured in a wavelength region from 350 to 750 nm while changing the incident ion energy, fluence, flux and target temperature. The effect of surface oxygen was also examined.

With using the TRIM92 code, backscattering of implanted deuterium and depth profiles of both displacement damage and implanted deuterons in Si target were calculated. To see the effect of the implanted deuteron on the backscattering, calculations for amorphous deuterated Si (a:Si–D_x) with x ranging from 0 to 4 were also carried out.

3. Results and discussion

3.1. Emission intensity

Fig. 1(a–c) show the observed spectra from a clean Si target under D^+ irradiation with an incident energy of 25, 10 and 5 keV, respectively. One can clearly see three Balmer lines of D_α, D_β and D_γ emitted from the backscattered atoms. In addition to these lines, broad band emission from the target was observed but only when the target surface was contaminated. As already demonstrated [6,7] the Balmer emissions were from the backscattered particles in excited states. The line intensity decreased with the incident energy corresponding to the decrease of the backscattering coefficient as seen in the figure. The line intensities were found to be proportional

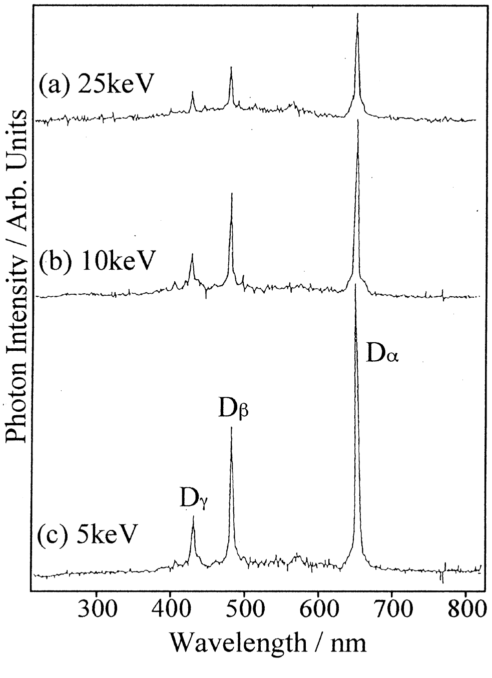

Fig. 1. Observed photon emissions from backscattered D atoms, for incident energy of (a) 25, (b) 10 and (c) 5 keV. Only Balmer series emissions of D_α, D_β, D_γ are appreciable for clean Si surface.

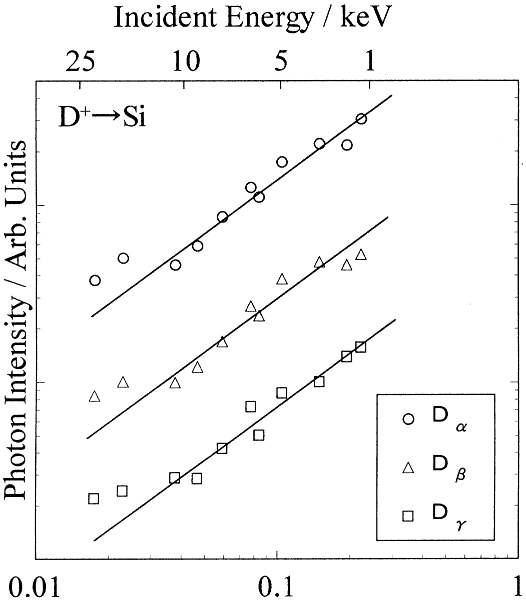

Fig. 2. Intensity of Balmer lines vs. backscattering coefficients that were calculated by TRIM92 code for each incident energy. The lines are only for guide to the eye.

to the flux density (not shown here). The relation between the line intensity and backscattering coefficient is given in Fig. 2 where the intensities of the Balmer lines are plotted as the function of the backscattering coefficients calculated by the TRIM92 code.

According to the TRIM92 calculation, most of the backscattered deuterium leaves the surface after several collisions with Si atoms in the target, and the energy distribution of the backscattered deuterium (see Fig. 3(a)) peaked at lower energies compared to the deuterium backscattered at higher mass targets, such as Mo and W [6]. The TRIM92 calculation also showed that the backscattering process is hardly influenced by implanted deuterium, remaining the similar energy distribution of the backscattering deuterium as pure Si, because the deuterium mass is significantly smaller than Si.

Fig. 4 shows the D_α photon intensity as a function of irradiation time for different incident energies. As expected, the D_α photon intensity stayed constant except for the incident energy of 5 keV. In the previous experiments [6,7], we have observed that HeI (at 578.8 nm (3^3D–2^3P)) line emission under He^+ incidence with any incident energy remains constant with irradiation time as well.

For the incident energy of 5 keV, however, the D_α photon intensity increased with irradiation time and saturated after prolonged irradiation. Such increase in

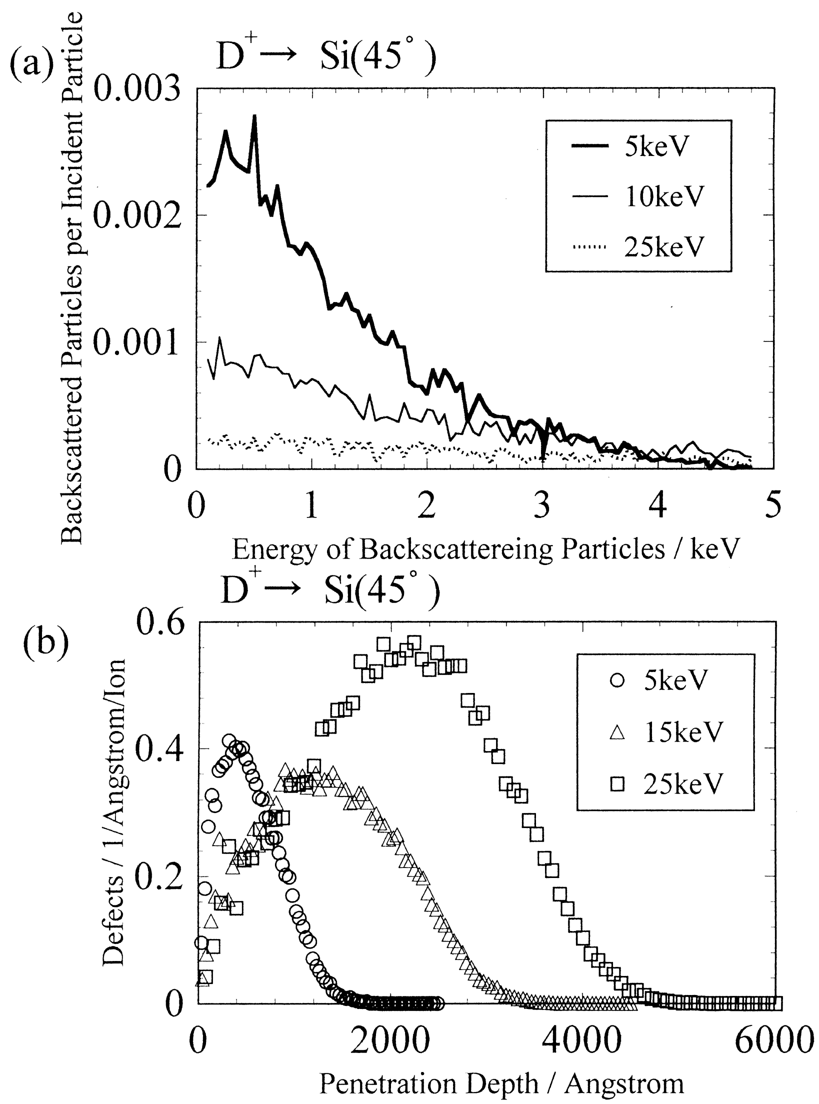

Fig. 3. (a) Energy distribution of backscattered particles for the geometry of the present experiment and (b) depth distribution of defects calculated by the TRIM92 code.

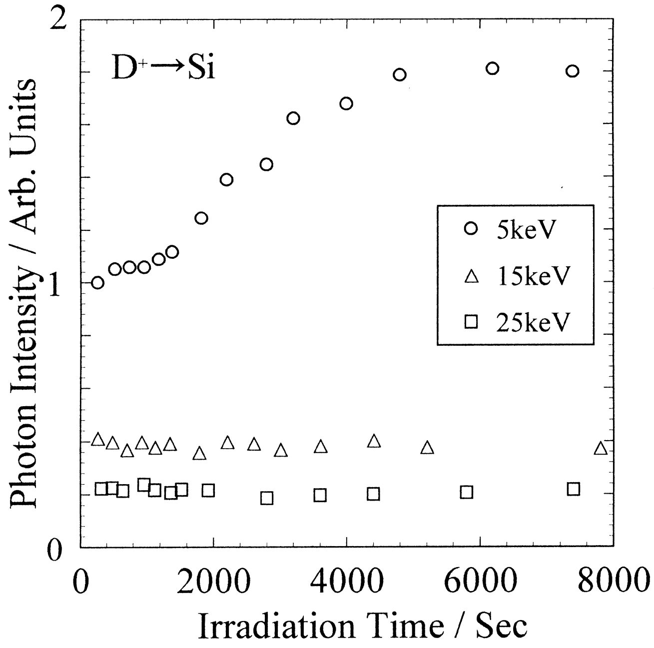

Fig. 4. Sequential change of the emission intensity with irradiation time for different incident energies of 5, 15 and 25 keV (the flux is 8×10^{13} D^+/cm^2s).

the intensity must be attributed either to an increase of the backscattering coefficient or/and a change in the electron capture probability of the backscattered atom at the top surface.

3.2. Influence of implanted deuterium

In order to find the cause of the intensity increase at 5 keV incidence, we have performed additional

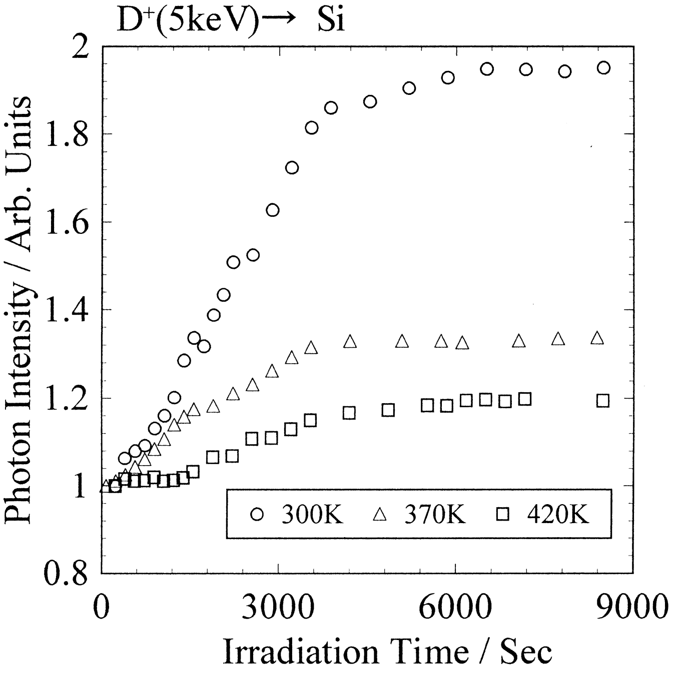

Fig. 5. The D_α intensity against irradiation time for different temperatures of RT, 370, 420 K (the flux is 1×10^{14} D^+/cm^2s).

experiments changing the target temperature and the results are given in Fig. 5. With increasing the target temperature the intensity increment between the initial intensity and the saturated one decreased. Above 600 K the intensity increase disappeared. According to thermal desorption studies, most of the hydrogen implanted in Si is released above 600 K [9–11]. Therefore the implanted deuterium is very likely the cause of the intensity increase. Actually after 600 K heating all results presented here were reproduced, which means the target Si returned to the initial state.

An additional experiment was carried out to see the deuterium release after the implantation as following. After the D_α photon intensity was saturated at a fixed temperature, we stopped the beam, but monitored the D_α intensity by injecting the beam in only during very short time period. The results are given in Fig. 6. One can clearly see that the D_α intensity decays with time. After the intensity returning to the initial value we could reproduce the intensity increase by deuterium injection. The time period needed for returning to the initial intensity became shorter with increasing the target temperature.

We have also looked for a probable surface impurity effect by introducing oxygen in the target chamber, but oxygen did not influence the result except very slight increase of the broad band emission.

Thus the increase of the intensity may be caused by the implanted deuterium. However, a question remains why this happens only for 5 keV incidence and not for higher energies?

Taking into account that electron capture of the backscattering ions occurs at the topmost surface, the electronic structure at the top surface layer dominates the electron population in backscattered deuterium atoms. Generally medium energy (1∼100 keV) hydrogen implantation in Si is known to result in amorphization or a:Si–H layer formation with the saturation concentration of around 0.4 in H/Si atom ratio. As shown in Fig. 3(b), however, the implantation is not necessary to produce the homogeneous damage profiles and implantation in depth. At RT diffusion of hydrogen (deuterium) in Si is very slow. Nevertheless the observed profile of implanted hydrogen was not the same to the implanted profile but was very similar to that of the damage profile given in Fig. 3(b), because the hydrogen solubility in Si is very small and most of the implanted hydrogen moves to the damaged region assisted by defects [12–14]. As a result a:Si–H layer is produced in the vicinity of the damaged region. However, higher fluence of the implantation often results in the blistering with the thickness of the blister cover similar to the peak damage depth [12,15], instead of increasing the thickness of the a:Si–H layer to the surface direction. Therefore hydrogen concentration at the top surface is not likely to be 0.4 in the H/Si ratio at higher energy incidence.

As clearly seen in Fig. 6, only the dynamic retention at the top surface, which is the retention just under the

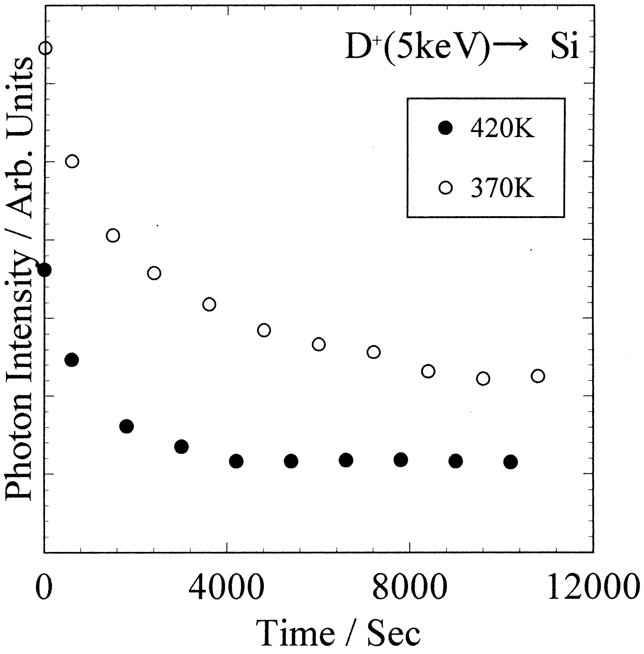

Fig. 6. Time evolution of D_α intensity decay after the termination of beam (only very short period for the emission measurement, the flux is 1×10^{14} D^+/cm^2s).

implantation and is often missed in a conventional thermal desorption study because of the spontaneous release, is important. Suppose hydrogen moves to the damaging profile, the TRIM92 calculation shows that D/Si ratio in the depth range top surface to 30 Å is less than 0.06 after the injection of 4×10^{17} D^+/cm^2 for 15 keV deuterons, whereas it exceeds 0.4 for 5 keV.

All above discussion leads us to conclude that only for the 5 keV implantation, the dynamic deuterium retention at the top surface layer is enough to modify the electronic structure and consequently influences the electron capture process of the backscattering deuteron. The reason for the observation that chemisorption of oxygen or oxidation at the top surface did not influence the Balmer emission is probably that 2s and 2p levels of chemisorbed oxygen or in the oxide (around 20 eV below the Fermi level) are too deep to occur the resonance capture of deuteron. In previous work [6,7], we have observed that the surface oxygen on various metals enhanced HeI emission from backscattered He (The ionization energy of He is around 24 eV, whereas that of H 13.6 eV).

The above discussion is still speculative, the modification of surface electronic structure by the dynamic retention must be monitored by various spectroscopic measurements like photoluminescence and XPS in future.

4. Conclusion

We have performed detailed investigation of Balmer line emission from backscattered atoms under the irradiation of a Si target with D^+ ions. For the clean surface, the emission intensity was proportional to the incident ion flux and the backscattering coefficient. For incidence of 15 and 25 keV, the intensity was not changed with increasing the ion fluence. This is in accordance with the calculated results of the TRIM92 code that the backscattering is not influenced very much by implanted deuterium in Si because the mass of D is much less than Si. At 5 keV incidence, however, the intensity gradually increased with the ion fluence and saturated after prolonged irradiation. The increment of the intensity between the initial and after saturated ones decreased with increasing the target temperature. Above 600 K the initial intensity was kept even after prolonged irradiation. Taking into account the hydrogen behavior in Si previously reported, the increase of the intensity is attributed to the dynamically retained deuterium near the top surface. The surface electronic structure of pure Si is changed to that of amorphized deuterated silicon (a:Si–D) by the retained deuterium. Consequently resonant electron capture process of backscattered deuteron is modified. For higher incident energies dynamic deuterium concentration near the top surface is not likely raised enough to modify the surface electronic structure.

The present conclusion is still speculative. Nevertheless it is clearly demonstrated that the Balmer emission from backscattered deuteron is modified by the surface chemical change. This should be taken into account for the estimation of hydrogen recycling properties by the Balmer emission measurement in fusion devices. The effect could be also injection for carbon because of the similarity of the electronic structure with Si.

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