Surface-limited deuterium uptake of Ru films under plasma exposure

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ABSTRACT
Blister formation has been an emerging research topic for extreme ultraviolet (EUV) mirrors exposed to hydrogen plasmas. Similar to plasma-facing materials in nuclear fusion reactors, it has been reported that blister formation in EUV mirrors is initiated by hydrogen uptake due to hydrogen ion or atom bombardment. However, the research so far has focused on Mo/Si multilayers exposed to only hydrogen ions or atoms, while the EUV mirror typically has a Ru capping layer facing hydrogen plasmas. We present experimental work to measure plasma-induced hydrogen uptake of Ru films. We bombarded our designed Ru-capped target with a low-temperature deuterium plasma and measured the deuterium retention using elastic recoil detection. Contrary to ion-driven deuterium uptake, the deuterium uptake rate of the Ru film had no dependence on the deuterium ion flux or energy after a period of plasma exposure. A reaction–diffusion model has been built to calculate the time evolution of deuterium retention, which well fits the experimental data. Based on this model, we conclude that the surface composition of the Ru film is the limiting factor for the deuterium uptake, which is seriously weakened when the surface is covered by Ru oxide. After the Ru oxide is reduced by the plasma, the uptake rate is predominantly driven by the deuterium surface coverage on metallic Ru. Our model also indicates that at the deuterium-populated Ru surface, deuterium has a low absorption barrier to penetrate the surface, which is supported by previously reported computational work.

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I. INTRODUCTION
Hydrogen uptake in materials is an important research subject for a variety of applications, and it is essential to understand and control hydrogen uptake to avoid its undesirable effects. Hydrogen can be introduced to steels during electrochemical processes where a high hydrogen concentration can lead to hydrogen embrittlement and, thus, shorten the lifetime of the steels.1 For metals upon exposure to hydrogen gas, dissociation of the molecular hydrogen is typically the rate limiting step of hydrogen uptake. However, metals with catalytic properties can readily dissociate hydrogen molecules into hydrogen atoms, which can penetrate the surface and diffuse into the bulk.2

Hydrogen uptake can also take place when the absorbing material is exposed to a hydrogen plasma.3 Hydrogen can arrive at the material surface in the dissociated form without assistance of a catalytic metal surface. The atomic hydrogen can stick to the surface and enable the steps toward hydrogen uptake.4 Besides hydrogen adsorption on the surface, high-energy hydrogen ions from a plasma can be directly implanted into the bulk.5 This phenomenon has been extensively studied in the context of nuclear fusion reactors since it is one of the primary causes of the hydrogen-isotope retention in the plasma-facing materials.7 The implanted hydrogen atoms can recombine in small voids in the material into gas molecules, accumulate high pressures, and subsequently deform the material surface by forming blisters.3 These blisters near the metallic surface pose potential risks to nuclear fusion reactors and, therefore, have triggered substantial research efforts on materials exposed to fusion-relevant hydrogen plasmas.8 Hydrogen uptake is also an important factor for blister formation in the extreme ultraviolet (EUV) mirrors.9 The EUV optics typically consists of Mo/Si multilayers, and Ru is frequently deployed as the capping material.10,11 In the EUV machine, these Ru-capped mirrors are kept at room temperature12 and exposed to EUV-induced hydrogen plasmas.13 It has been reported that blisters
emerge in Mo/Si multilayers exposed to hydrogen atoms or deuterium plasmas, and hydrogen uptake plays an essential role in this process.\textsuperscript{14} It has also been experimentally demonstrated that the onset of blister formation depends on the energy at which hydrogen ions bombard Mo/Si multilayers.\textsuperscript{15} Density functional theory (DFT) modeling indicates that hydrogen at the Ru surface faces a high-energy barrier to enter the subsurface, and hydrogen uptake of Ru could still occur once the Ru surface is nearly saturated with hydrogen by a hydrogen plasma.\textsuperscript{16} The energy barriers for hydrogen near the surface play a crucial role here because they determine the effective rate of plasma-induced hydrogen uptake.\textsuperscript{17} However, they have not been measured since plasma-induced hydrogen uptake of Ru has attracted low attention, and very few relevant experiments have been carried out. Therefore, it is important to perform model experiments to measure the uptake rate and to study the influence of plasma parameters on the hydrogen uptake.

In this work, we experimentally investigated plasma-induced hydrogen uptake of Ru at room temperature. We designed a Ru-capped film stack as the target to be exposed to a plasma. To systematically study the influence of the plasma parameters, we designed a series of plasma settings and characterized each corresponding plasma using a double Langmuir probe and dual heat flux sensors (DHFSs).\textsuperscript{17} We exposed multiple targets with various fluences in each plasma setting and measured the deuterium content in the target using elastic recoil detection (ERD). To explain the data, we built a reaction–diffusion model to simulate the deuterium retention based on the experimental settings and available literature values. Important parameters for the hydrogen uptake are derived using the model to determine the decisive factors for deuterium uptake of Ru films under a deuterium plasma exposure.

II. EXPERIMENT

Hydrogen uptake can be achieved by ion implantation, radical exposure, and gas exposure based on the energy of the incoming hydrogen. To study the plasma-induced deuterium uptake of Ru, we exposed Ru-terminated targets to only deuterium gas or deuterium plasmas with different ion energies, followed by \textit{ex situ} measurements of the deuterium retention in the targets. The experimental details are described below.

A. Ru-capped targets

The targets used in this work were Ru-capped Ti films on Si (100) substrates 25.4 mm (1 in.) in diameter. The film stack is shown in Fig. 1(a). This target design deploys the Ti layer as a hydrogen absorption region, which is protected by the Ru capping layer against oxidation. When the target is exposed to a deuterium plasma, deuterium has to permeate the Ru layer before absorption occurs at the Ti layer. In this manner, the deuterium retention in the Ti layer is used to quantify the deuterium uptake rate of the Ru layer. Similar approaches have been successfully applied to other Ru-capped films exposed to hydrogen radicals.\textsuperscript{16,19}

The targets were prepared by magnetron sputtering at Philips Engineering Solutions. The base pressure of the sputtering system was below $5 \times 10^{-7}$ mbar, and argon was used as the working gas. The growth rates were 0.125 and 0.222 nm/s for Ru and Ti, respectively. The Ru and Ti layers both had a nominal thickness of 40 $\pm$ 4 nm. The areal densities of Ru and Ti were (2.9 $\pm$ 0.23) $\times$ 10$^{21}$ atoms/m$^2$ and (1.75 $\pm$ 0.14) $\times$ 10$^{21}$ atoms/m$^2$, respectively, according to our Rutherford backscattering spectroscopy (RBS) measurements at the DIFFER Ion Beam Facility (IBF). With these results, we obtain the Ru lattice density of 7.25 $\times$ 10$^{28}$ m$^{-3}$. Targets with thickness of 24.7 and 34.7 nm were also prepared for experiments to check the influence of deuterium diffusion, which will be discussed later. Grazing incidence x-ray diffraction (GIXRD) measurements were carried out at the incidence angle $1^\circ$ using Bruker D8 ADVANCE ECO to characterize the film structure of the target. The result indicates that the Ru layer was a polycrystalline film with a typical grain size of 21 $\pm$ 2.1 nm, as shown in Fig. 2.

The target surface composition was quantified by x-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha at Nanolab@TU/e) analysis. The Ru target surface has 58 at. % of Ru and 42 at. % of O when we assume a negligible amount of carbon. Based on the Ru 3d$\sub{5/2}$ and 3d$\sub{3/2}$ peaks in the XPS spectrum as

![FIG. 1. (a) The film stack of the target used in this work. The Ru and Ti films were deposited using a magnetron sputtering machine. One monolayer of RuO$_2$ adventitiously formed at the surface according to our x-ray photoelectron spectroscopy (XPS) analysis. (b) A photo of one of the targets clamped on the target holder for ERD measurement. The feature close to the flat on the target surface was the reflected image of a window in the lab by Ru coating.](image1)

![FIG. 2. A typical GIXRD spectrum of our as-deposited Ru-capped target. We use Bruker DIFFRAC.EVA to estimate the grain size based on the Scherrer equation. The major peak Ru (0 0 0 2) gives a grain size of approximately 21 $\pm$ 2.1 nm.](image2)
shown in Fig. 3, we find that the intensity ratio of Ru oxide to metallic Ru is 0.4, and we use this ratio \( R \) to estimate the RuO\(_2\) thickness \( d_{\text{RuO}_2} \) by applying the following formula derived from the Beer–Lambert equation:\(^{21}\)

\[
d_{\text{RuO}_2} = \lambda_A \cos \theta \ln \left( 1 + \frac{R}{R_0} \right),
\]

where \( \lambda_A \) is the attenuation length for the Ru 3d electrons, which are emitted at an angle \( \theta \) to the surface normal. \( R_0 \) denotes the intensity ratio of a thick RuO\(_2\) layer to a thick metallic Ru. Assuming \( \lambda_A \) is the same for the electrons from the RuO\(_2\) and those from the metallic Ru, we take \( R_0 = \frac{N_{\text{RuO}_2}}{N_{\text{Ru}}} \), where \( N_{\text{RuO}_2} \) and \( N_{\text{Ru}} \) are the atom number densities of RuO\(_2\) and Ru, respectively.\(^{21}\)

\[
\lambda_A = \frac{0.65 + 0.007E_A^{0.93}}{Z^{20.38}},
\]

where \( E_A \) is the energy of emitted electrons in eV and \( Z \) is the atomic number of the material. By inserting all the relevant parameters into Eq. (1), we obtain the presence of 0.31 ± 0.03 nm of RuO\(_2\), which corresponds to 0.98 ± 0.09 monolayers. This result is consistent with the previously reported one monolayer of RuO\(_2\) for Ru thin films upon air exposure at room temperature.\(^{22,23}\)

### B. Deuterium plasma exposures

The work on deuterium plasmas was performed on nano-PSI at DIFFER. Nano-PSI is an experimental setup at DIFFER to study plasma surface interactions (PSIs).\(^{17}\) Its basic schematic is shown in Fig. 4. The details of the setup are given in Ref. 17, and a summary is given here. The base pressure of nano-PSI was approximately 10\(^{-7}\) mbar when the system was pumped by a turbomolecular pump. For plasma generation, operation was switched from the turbomolecular to a roots pump, and the typical operation pressure was 45–97 Pa. Plasmas in the system were generated by a cascaded arc source,\(^{24,25}\) and the gas flow into the source was typically 1.2–1.5 slm. During operation of the plasma source, the cathode current was set to 40 A, and the characteristic cathode voltage was 130 V for deuterium plasmas. A target was mounted on a water-cooled target holder, and the target surface was 100 mm away from...
the nozzle of the plasma source. The target had a circular exposure area with a diameter of 18 mm, defined by a mask for clamping the target on the target holder. The diameter of the exposure area was much smaller than the plasma diameter of 67 mm, as measured by a double Langmuir probe, such that the loading of the surface can be considered uniform. A K-type thermocouple was attached to the backside of the target for temperature measurement, and typically, the target remained at room temperature during plasma exposure. With the double Langmuir probe and DHFS, we can measure the electron temperature, the ion flux, and the radical flux for our plasma source settings. In our typical condition, the vessel pressure was 45 Pa, the electron temperature was $0^\circ$C, the ion flux was $(4.2 \pm 0.47) \times 10^{19}$ m$^{-2}$ s$^{-1}$, and the radical flux was $(2.3 \pm 0.32) \times 10^{22}$ m$^{-2}$ s$^{-1}$. We define this plasma source setting as our nominal one.

C. Ion beam analysis

In this work, we used Rutherford backscattering spectroscopy (RBS) to determine the Ru and Ti film thicknesses and elastic recoil detection (ERD) to measure the amount of deuterium in the plasma-exposed targets. These ex situ measurements were carried out at the DIFFER IBF. In both RBS and ERD measurements, our incident beam was a $^3$He$^+$ beam at 2.4 MeV. A fraction of the incident beam was backscattered by a so-called chopper for relative ion dose control. After passing by the chopper, the ion beam entered the target at an incident angle $\alpha$, which was controlled by rotating the target about the rotation axis, as shown in Figs. 5 and 6.

In the RBS configuration shown in Fig. 5, we used Cornell geometry and set $\alpha = 4^\circ$. The incident beam, the scattered beam, and the rotation axis were in the same plane. The RBS detector sat at the scattering angle $\theta = 170^\circ$ with respect to the incident ion beam. We used an Si target coated with $6.5 \times 10^{11}$ atoms/m$^2$ of Pt as the RBS reference target to calibrate the ion dose. This way, the Ru and Ti thicknesses were determined by fitting the RBS spectrum of the Ru-capped target with SIMNRA (version 7.03).

In the ERD configuration shown in Fig. 6, we used International Business Machines (IBM) 1 geometry, where the incident and scattered beams were in the same plane perpendicular to the rotation axis. To control the incident angle, the surface normal of the target was rotated toward the direction of the scattered beam. We set $\alpha = 75^\circ$ and the ERD detector sat at the scattering (recoil) angle $\theta = 30^\circ$ with respect to the incident ion beam. A Mylar$^\text{®}$ foil of $1.26 \times 10^{24}$ atoms/m$^2$ was placed in front of the ERD detector to stop the scattered $^4$He at the recoil angle such that we received signals of recoil hydrogen and deuterium particles at the ERD detector.

For quantitative determination of the deuterium retention using ERD, we used a combination of the chopper and an ERD reference target with pre-determined deuterium concentrations. We obtained ERD spectra of the ERD reference and the plasma-exposed targets at the same ion dose controlled by the chopper. The deuterium areal density, $(Nt)_r$, in the plasma-exposed target can be calculated based on the following formula:

\[
(Nt)_r = A_r x \frac{\sigma_i Q_i}{\sigma_f Q_f} (Nt)_i,
\]

where $(Nt)_i$ is the deuterium areal density in the ERD reference target. $A_r, \sigma_i, and Q_i (i = r, x)$, respectively, refer to the spectral area corresponding to deuterium, the reaction cross section, and the ion dose on the target. The subscripts $r$ and $x$ refer to the ERD reference target and the plasma-exposed targets, respectively. When the parameters $(Nt)_i, \sigma_i, and Q_i (i = r, x)$ are pre-determined, this method directly gives the deuterium areal density. In this work, $(Nt)_i$ can be calculated based on the film stack of the ERD reference target as shown in Fig. 7. We used the cross section measured by Kellock and Baglin for $\sigma_i$ and $\sigma_f$ and set $Q_i = Q_f$ using the chopper. This way, the deuterium areal density can be determined based on the corresponding spectral area relative to that of the ERD reference target.
III. EXPERIMENTAL RESULTS

A. Gas exposure and ERD spectra

During the deuterium plasma exposure in nano-PSI, the target faces a much higher molecule flux than the ion or atom flux because the ionization and dissociation degrees are typically less than $10^{-3}$ and $10^{-2}$, respectively. Since hydrogen gas molecules can stick to an Ru surface even above room temperature and could there subsequently dissociate and diffuse inward, we first investigated the influence of deuterium coverage of the Ru surface upon deuterium gas exposure. Therefore, we exposed one target to deuterium gas at 45 Pa and another one to a deuterium plasma in the nominal setting (i.e., also at a vessel pressure of 45 Pa). Both exposures lasted for 4200 s. The two resultant ERD spectra are shown in Fig. 8(b). In the ERD spectra, we observed two ($H_{\text{surface}}$ and $H_{\text{bulk}}$) and three peaks ($H_{\text{surface}}$, $H_{\text{bulk}}$, and $D_{\text{bulk}}$), respectively, for the gas-exposed and plasma-exposed targets.

To understand the spectra, we performed simulations using SIMNRA, and the film stack for simulations is shown in Fig. 8(a). The simulated spectrum well matches the experimental spectrum for the plasma-exposed target. According to the simulation, the three peaks in the ERD spectra result from $3.1 \times 10^{19}$ atoms/m$^2$ of hydrogen at the Ru surface ($H_{\text{surface}}$), $4.1 \times 10^{19}$ atoms/m$^2$ of hydrogen in the Ti layer ($H_{\text{bulk}}$), and $8.8 \times 10^{20}$ atoms/m$^2$ of deuterium in the Ti layer ($D_{\text{bulk}}$), respectively. This indicates that deuterium was retained in the Ti layer after the deuterium plasma exposure. On the other hand, the $D_{\text{bulk}}$ peak was missing in the spectrum of the gas-exposed target. Based on the simulation, we can be confident that the plasma-induced deuterium retention was caused by ions or radicals in the plasma instead of gas molecules.

It is also important to note that we calculate the spectral area $A_i (i=r,x)$ of deuterium [Eq. (3)] by integrating over the $D_{\text{bulk}}$ peak in each spectrum. This method is applied to all the plasma-exposed targets in Sec. III C.

B. Plasma source settings

In this work, we study the influence of three plasma parameters, which are the ion energy, the ion flux, and the atom flux. In addition to the nominal setting, we chose three more settings of which the details are shown in Table I. Based on the result shown in Sec. III A, we assume that the difference of the gas pressure does not play a role. For floating targets, the ion energy is estimated to be approximately 1 eV based on measurements in Ref.30 under similar plasma conditions. For biased targets, the ion energy is dominated by the bias voltage and estimated to be 20 eV since the plasma potential is only 0.2 eV.30 The ion flux and the atom flux in various settings were measured, respectively, using the double Langmuir probe and the DHFS. When changing the source setting, we attempt to change only one plasma parameter at a time while
keeping the other two parameters close to those in the nominal setting, as shown in Fig. 9. This approach helps us to systematically study the influence of individual plasma parameters on the deuterium retention.

### C. Deuterium retention

In each source setting, we exposed a series of the Ru-capped targets for various time durations. In addition, we exposed targets with different Ru thicknesses to plasmas in the same setting. After each exposure, the target was taken out of nano-PSI and stored in a desiccator for less than one week before deuterium retention measurements at the DIFFER IBF. The results are shown in Fig. 10. First, we observed no significant influence of the Ru thickness on the deuterium retention in the range of the thickness studied here. This result indicates that the increase of the deuterium retention is not limited by diffusion through the Ru bulk. Second, for all the source settings, the deuterium retention increases with the plasma fluence. For the exposure time of less than 1000 s, the retention increases faster in the high ion energy setting than the other ones. A higher ion energy and a higher ion flux both lead to a higher uptake rate, while the atom flux does not have significant influence. However, after 1000 s, the deuterium retention increases at similar rates for all the source settings. Variation of the plasma parameters listed in Fig. 9 does not lead to a different rate of the retention buildup in this time range.

Despite a low hydrogen solubility of Ru, our results show that upon exposure to a deuterium plasma, deuterium can penetrate the Ru layer and reach the Ti layer. This deuterium transport typically involves overcoming the energy barrier at the Ru surface and subsequently diffusing from the Ru subsurface to the Ti layer. However, since our data show no dependence of deuterium retention on the Ru thickness, as shown in Fig. 10(a), the increase of the deuterium retention is limited by surface-related processes instead of diffusion through the Ru layer. DFT studies by Onwudinanti et al. show that, for a low hydrogen surface coverage, the Ru surface limits hydrogen uptake because hydrogen needs to overcome a high-energy barrier of 1.06 eV to migrate from the surface to the subsurface. However, these DFT studies also show that the surface process can still occur if the Ru surface sites become unavailable due to exposure to a hydrogen plasma, which leads to a high hydrogen surface coverage. In the experiment, we cannot perform operando measurements

<table>
<thead>
<tr>
<th>Source setting</th>
<th>Gas flow (slm)</th>
<th>Pressure (Pa)</th>
<th>Target bias</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nominal</td>
<td>1.5</td>
<td>45</td>
<td>Floating</td>
</tr>
<tr>
<td>High atom flux</td>
<td>1.5</td>
<td>97</td>
<td>Floating</td>
</tr>
<tr>
<td>Low ion flux</td>
<td>1.2</td>
<td>77</td>
<td>Floating</td>
</tr>
<tr>
<td>High ion energy</td>
<td>1.5</td>
<td>45</td>
<td>−20 V</td>
</tr>
</tbody>
</table>

**TABLE I.** An overview of the plasma source settings.

![FIG. 9. The plasma parameters in the various source settings.](image)

![FIG. 10. The comparison between simulated and measured deuterium retention in the Ru-capped targets exposed to deuterium plasmas. (a) The deuterium retention in targets with different thicknesses exposed to plasmas in the high ion energy setting for 720 s. (b) The deuterium retention in targets exposed to plasmas in the different settings for various time durations.](image)
of the deuterium surface coverage to verify the theory by Onwudinanti et al. Instead, we calculate the deuterium surface coverage using a reaction–diffusion model to have a better understanding of the results. This is discussed in Sec. IV.

IV. MODEL

A. Model description

The reaction–diffusion model is built based on the work by Pick and Sonnenberg and Ogorodnikova for ionic and atomic hydrogen–metal interactions. The model focuses on the hydrogen–metal interactions close to a nearly saturated surface, where the hydrogen surface density is comparable with the areal density of the chemisorption sites on the metal surface. In recent years, this approach has become more widely used in research on plasma-induced deuterium uptake, and it has been successfully applied to simulations of self-damaged tungsten exposed to low-energy deuterium atoms. In these cases, deuterium atoms are first adsorbed on the tungsten surface without directly reaching the bulk, and therefore, the surface-related reactions need to be included in the model. Similarly, our model takes into account the reactions related to time evolution of the deuterium concentrations at different depths in the Ru film. The potential diagram of deuterium atoms near the Ru surface is depicted in Fig. 11.

The model is implemented using a python script, which simultaneously solves the system of ordinary differential equations using the Adams/backward differentiation formula (BDF) method. The variables in the model are described below:

1. the concentration of metallic Ru at the surface \( n_{Ru} \) in \( \text{m}^{-2} \),
2. the deuterium concentration at the metallic Ru surface \( n_{surf} \) in \( \text{m}^{-3} \),
3. the deuterium concentration at the metallic Ru subsurface \( n_{sub} \) in \( \text{m}^{-3} \), and
4. the deuterium concentration in the Ti layer \( n_{trap} \) in \( \text{m}^{-3} \).

It is important to note that, besides deuterium concentrations, we also treat the surface concentration of metallic Ru as a variable in the model. We assume that all the variables are zero as the initial condition, which means that the target surface consists of only impurities other than metallic Ru before plasma exposure. This is a valid assumption because our target was exposed to air before plasma exposure in nano-PSI, and our XPS analysis shows that the target surface was covered by a mono-layer of native oxide instead of metallic Ru, as shown in Sec. II A. We assume that deuterium uptake could occur only at the surface of metallic Ru but not of the native oxide since hydrogen desorbs from the Ru oxide surface at a much lower temperature than a metallic Ru surface. Additionally, a recent study shows that a sub-monolayer coverage of oxygen on W prohibits deuterium from reaching the bulk because the W surface sites can be covered by oxygen instead of deuterium atoms. In the model, we, therefore, assume that the deuterium surface coverage is zero on the Ru oxide surface. Previous work by Soroka et al. also suggests that it is necessary to take the native oxide layer into account when studying hydrogen permeation through a Ru film upon exposure to hydrogen radicals. As the target is exposed to a deuterium plasma, the concentration of metallic Ru increases due to plasma-assisted oxide removal. The oxide removal rate, which is also the surface Ru growth rate, is determined by the deuterium ion flux multiplied by the oxide removal yield given the atomic D flux in the \( 10^{-2} \text{ m}^{-2} \text{ s}^{-1} \) range. The concentration of a metallic Ru surface reaches its maximum when the Ru surface coverage becomes unity. We assume that one Ru atom can accommodate one D atom at most; therefore, the maximum of the deuterium surface coverage is also unity.

A brief precis of the main processes in the model is now outlined: deuterium starts to enter the system as metallic Ru emerges as a result of oxide removal. Deuterium at the surface can also leave the system via the desorption and abstraction processes, which follow the Langmuir–Hinshelwood and Eley–Rideal mechanisms, respectively. Given weak deuterium desorption and abstraction from Ru at room temperature, deuterium is expected to stick to the surface of metallic Ru, leading to a high deuterium surface concentration \( n_{surf} \). Next, deuterium can cross the absorption barrier \( E_{absorb} \) of the Ru surface to enter the Ru subsurface, while deuterium could also come from the Ru subsurface back to the Ru surface by overcoming the resurfacing barrier \( E_{resurf} \). Since most of the surface sites are occupied by the incoming deuterium atoms from the plasma, deuterium atoms at the subsurface cannot easily come back to the surface, building up the deuterium concentration at the subsurface \( n_{sub} \). Deuterium at the subsurface can then diffuse to the interface between the Ru and Ti layers, where we assume that all the coming deuterium atoms become trapped in Ti by forming titanium deuteride. These trapped deuterium atoms contribute to the deuterium concentration \( n_{trap} \), which we measured experimentally using ERD. At this point, we also assume that the detrapping rate from the titanium deuteride is zero due to the strong chemical bonds formed.

Below, we describe our model for calculation of four concentrations in the system. Based on oxide reduction and particle
conservation, we have the following differential equations for the concentrations:

$$\frac{dn_{Ru}}{dt} = \Gamma_{\text{removal}},$$

$$\frac{dn_{surf}}{dt} = \Gamma_{\text{adsorp}} - \Gamma_{\text{abstr}} - \Gamma_{\text{desorp}} + \Gamma_{\text{resurf}},$$

$$\lambda_{Ru} \frac{dc_{sub}}{dt} = \Gamma_{\text{adsorp}} - \Gamma_{\text{resurf}} - \Gamma_{\text{diff}},$$

$$d_{Ti} \frac{dc_{trap}}{dt} = \Gamma_{\text{diff}},$$

where the fluxes are defined as

$$\Gamma_{\text{removal}} = \Gamma_{\text{atom}} \xi (1 - \theta_{Ru}),$$

$$\Gamma_{\text{adsorp}} = \Gamma_{\text{atom}} \sigma_{\text{adsorp}} (n_{Ru} - n_{surf}),$$

$$\Gamma_{\text{abstr}} = \Gamma_{\text{atom}} \sigma_{\text{abstr}} n_{surf},$$

$$\Gamma_{\text{desorp}} = 2k_{\text{desorp}} n_{surf}^2,$$

$$\Gamma_{\text{absorp}} = k_{\text{absorp}} n_{surf}^2,$$

$$\Gamma_{\text{resurf}} = k_{\text{resurf}} c_{sub} (\theta_{Ru} - \theta_{Ti}),$$

$$\Gamma_{\text{diff}} = \frac{D_{\text{diff}}}{d_{Ru}} c_{sub},$$

where all the parameters are divided into the fixed ones in Table II and the free ones in Table III. When we fit the deuterium retention in Fig. 10(b) with the model, we vary only the free parameters and keep the fixed ones constant. As shown in Table II, most of the fixed parameters can be measured experimentally or are available in the literature for atomic hydrogen. We use the reported adsorption cross section for atomic hydrogen in Ref. 47, and the abstraction cross section is obtained by extrapolating the temperature-dependent cross sections in Ref. 44. It is also worthwhile to note that since we have a polycrystalline Ru film, as shown in Fig. 2, the diffusion coefficient $D_{\text{diff}}$ is calculated using the following equations:

$$D_{\text{diff}} = \frac{1}{\sqrt{2}} \frac{D_{\text{diff},1} + D_{\text{diff},2} + D_{\text{diff},3}}{3},$$

where $D_{\text{diff}} (i = 1, 2, 3)$ is the extrapolated diffusion coefficient at room temperature based on molecular dynamics simulations in Ref. 48, and the subscripts refer to different Ru lattice structures, where grain boundaries are taken into account. A factor of $\frac{1}{\sqrt{2}}$ is added to Eq. (6) due to the mass difference between hydrogen and deuterium.44 The free parameters to be determined are shown in Table III. We assume that the absorption and resurfacing barriers are constants, while the removal yield is expected to be a function

### Table II. The fixed parameters in the reaction–diffusion model.

<table>
<thead>
<tr>
<th>Fixed parameter</th>
<th>Symbol</th>
<th>Value</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atom flux</td>
<td>$\Gamma_{\text{atom}}$</td>
<td>$2.3 \times 10^{22}$ m$^{-2}$s$^{-1}$</td>
<td>Experimental data</td>
</tr>
<tr>
<td>Ion flux</td>
<td>$\Gamma_{\text{ion}}$</td>
<td>$4.2 \times 10^{19}$ m$^{-2}$s$^{-1}$</td>
<td>Experimental data</td>
</tr>
<tr>
<td>Target temperature</td>
<td>$T$</td>
<td>23 °C</td>
<td></td>
</tr>
<tr>
<td>Ru layer thickness</td>
<td>$d_{Ru}$</td>
<td>40 nm</td>
<td></td>
</tr>
<tr>
<td>Ti layer thickness</td>
<td>$d_{Ti}$</td>
<td>40 nm</td>
<td></td>
</tr>
<tr>
<td>Ru lattice density</td>
<td>$N_{\text{latt,Ru}}$</td>
<td>$7.25 \times 10^{28}$ m$^{-3}$</td>
<td>Experimental data</td>
</tr>
<tr>
<td>Ru lattice parameter</td>
<td>$\lambda_{Ru}$</td>
<td>$\sqrt{N_{\text{latt,Ru}}}$</td>
<td></td>
</tr>
<tr>
<td>Maximum surface coverage</td>
<td>$n_{\text{max}}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ru surface coverage</td>
<td>$\theta_{Ru}$</td>
<td>$\frac{\lambda_{Ru}}{n_{\text{max}}}$</td>
<td></td>
</tr>
<tr>
<td>D surface coverage</td>
<td>$\theta_{D}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pre-exponential factor</td>
<td>$v_0$</td>
<td>$10^{13}$ Hz</td>
<td>Refs. 4 and 32</td>
</tr>
<tr>
<td>Adsorption cross section</td>
<td>$\sigma_{\text{adsorp}}$</td>
<td>3.2 Å$^2$</td>
<td>Metallic Ru$^{37}$</td>
</tr>
<tr>
<td>Abstraction cross section</td>
<td>$\sigma_{\text{abstr}}$</td>
<td>1.1 Å$^2$</td>
<td>Metallic Ru$^{34}$</td>
</tr>
<tr>
<td>Ru desorption barrier</td>
<td>$2E_{\text{desorp,Ru}}$</td>
<td>1.04 eV</td>
<td>Metallic Ru$^{35}$</td>
</tr>
<tr>
<td>Desorption rate constant</td>
<td>$k_{\text{desorp}}$</td>
<td>$v_0 \lambda_{Ru}^2 \exp\left(-\frac{E_{\text{desorp}}}{k_{B}T}\right)$</td>
<td>Refs. 4, 32, and 33</td>
</tr>
<tr>
<td>Absorption rate constant</td>
<td>$k_{\text{absorp}}$</td>
<td>$v_0 \exp\left(-\frac{E_{\text{absorp}}}{k_{B}T}\right)$</td>
<td>Refs. 4, 32, and 33</td>
</tr>
<tr>
<td>Resurfacing rate constant</td>
<td>$k_{\text{resurf}}$</td>
<td>$v_0 \lambda_{Ru} \exp\left(-\frac{E_{\text{resurf}}}{k_{B}T}\right)$</td>
<td>Refs. 4, 32, and 33</td>
</tr>
<tr>
<td>Diffusion coefficient for Ru</td>
<td>$D_{\text{diff}}$</td>
<td>$5.33 \times 10^{-13}$ m$^2$/s</td>
<td>Ref. 48</td>
</tr>
</tbody>
</table>

### Table III. The free parameters in the reaction–diffusion model.

<table>
<thead>
<tr>
<th>Free parameter</th>
<th>Symbol</th>
<th>Best fit</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oxide removal yield</td>
<td>$\xi$</td>
<td>$2 \times 10^{-3}$ at 1 eV</td>
<td>Ion energy dependent</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Absorption barrier</td>
<td>$E_{\text{absorp}}$</td>
<td>0.87 eV</td>
<td>Refs. 16 and 34</td>
</tr>
<tr>
<td>Resurfacing barrier</td>
<td>$E_{\text{resurf}}$</td>
<td>0.62 eV</td>
<td>Refs. 16 and 34</td>
</tr>
</tbody>
</table>
of the ion energy based on experimental results reported by Dolgov et al.43

Given the parameters discussed above, our model calculates evolution of the concentrations \( n_{Ru}, n_{surf}, c_{sub}, \) and \( c_{trap} \) for all the experimental settings. We compare \( c_{trap} \) with the measured deuterium retention, and, as shown in Fig. 10(b), our model predicts quite well the experimental data with the best \( t \) values in Table III. The model shows that the deuterium surface coverage on the metallic Ru surface stays 0.74 during the plasma exposure, as shown in the inset of Fig. 12. This means once any part of the Ru oxide surface is reduced to metallic Ru, it is immediately covered by deuterium at 0.74 surface coverage. It is also important to note that since the entire surface is reduced to metallic Ru after long exposure, the surface coverage calculated by the model reaches 0.74 for all the plasma settings, as shown in Fig. 12 and depicted in Fig. 13 for clarity. The deuterium uptake rate is determined by this high surface coverage, which is consistent with the previous findings based on DFT.16 Therefore, this model is valid for a quantitative description of the deuterium concentrations in our experiments. In Sec. IV B, we will further discuss the determined free parameters in comparison with previously reported values in the literature.

B. Discussion

By fitting the experimental data with our reaction–diffusion model, we obtained the oxide removal yield of \( 2 \times 10^{-3} \) and \( 10^{-2} \) at ion energies of 1 and 20 eV, respectively. Similar coefficients have been experimentally measured by Dolgov et al. using hydrogen plasmas at ion energy larger than 15 eV.43 Dolgov et al. found that for ion energy between 15 and 40 eV, the oxygen removal rate increases with the hydrogen ion flux and seems to level off at \( 5 \times 10^{17} \) atoms/(s m\(^2\)) when the hydrogen ion flux reaches \( 10^{19} \) ions/(s m\(^2\)).43 To compare our measured yields with their values, we need to convert our oxide removal yields to oxygen removal rates. We do this for our plasma setting of "High ion energy" because the ion energy was within the energy range of the experiments by Dolgov et al. In this setting, our deuterium ion flux was \( 4.2 \times 10^{19} \) ions/(s m\(^2\)), and our oxide removal yield was \( 10^{-2} \). Since the Ru surface was initially covered by RuO\(_2\) before plasma exposure as shown in Sec. II A, two oxygen atoms need to be removed for each Ru atom to form a metallic Ru surface. As a result, our oxygen removal rate was \( 8.4 \times 10^{17} \) atoms/(s m\(^2\)) at an ion energy of 20 eV, which is in reasonable agreement with the value measured by Dolgov et al. This could indicate that the mechanism of oxide removal in our experiments is similar to the process

FIG. 12. The simulated deuterium surface coverage over time for the various plasma settings. The inset shows the deuterium surface coverage on the metallic Ru surface \( \theta_{D} \), which remains 0.74 throughout the plasma exposure for all the plasma settings.

FIG. 13. Time evolution of the Ru-capped target upon plasma exposure. The circles on the surface refer to the deuterium atoms, and the width of the arrow from the surface to the Ti layer represents the magnitude of the deuterium uptake rate. (a) The target surface is initially covered by a monolayer of Ru oxide. (b) Upon plasma exposure, the deuterium uptake of Ru takes place at the location where the Ru oxide is reduced and the surface is populated by D atoms. (c) After the Ru oxide is fully reduced, the deuterium uptake rate reaches its maximum, limited by the surface coverage.
proposed by Dolgov et al. for ion energy between 15 and 40 eV. They suggested that in this process, the excess energy from the ion dissociates the Ru–O bond, making OH formation highly probable in the presence of hydrogen atoms from the plasma. Then oxygen atoms are removed by forming water vapor and its desorption from the surface.13,19 Similar synergistic effects have been found in plasma-assisted surface processing.20

It is also worthwhile to check the possibility of extrapolating the model proposed by Dolgov et al. for Ru oxide removal. They suggested that the oxide removal yield is a function of the hydrogen ion energy, following an Arrhenius equation as shown below:43

\[
\ln \xi = \ln A_1 - \frac{E_a}{k_B T_s} = \ln A_1 - \frac{E_a}{\gamma_{eff} E_{ion}}, \tag{7}
\]

where \(\xi\) denotes the oxide removal yield and \(A_1\) the prefactor of the Arrhenius equation. \(E_a\), the activation energy of the Ru oxide removal, is equal to 0.48 eV.43,49 \(T_s\) is the surface temperature and \(E_{ion}\) the hydrogen ion energy. \(\gamma_{eff}\) is a coefficient to calculate the effective energy transferred from the incoming ions to the oxide surface after energy loss due to collisions. Dolgov et al. found \(\gamma_{eff} = 0.32\) in their experiment for ion energy between 15 and 40 eV, and Eq. (7) can be simplified as below:

\[
\ln \xi = \ln A_1 - \frac{1.5}{E_{ion}}, \tag{8}
\]

where 1.5 is the slope of the equation in an Arrhenius plot and \(E_{ion}\) is expressed in eV. Note that a certain part of our experiments was performed at an ion energy of 1 eV, which was outside the energy range of the experiments by Dolgov et al. However, we can calculate the slope of Eq. (8) in an Arrhenius plot using the oxide removal yields we obtained. By defining \(E_1 = 1\) eV, \(E_2 = 20\) eV, \(\xi_1 = 2 \times 10^{-3}\), and \(\xi_2 = 10^{-2}\), we then obtain the slope \(\frac{\ln \xi_1 - \ln \xi_2}{E_2 - E_1}\) = 1.7 for the ion energy between 1 and 20 eV. Interestingly, this number is reasonably close to the slope 1.5 for ion energy between 15 and 40 eV, which could suggest that the mechanism proposed by Dolgov et al. is valid for oxide removal at ion energy down to 1 eV. In addition, since the oxide removal rate in our model is only dependent on the ion flux and not the atom flux, we could conclude that the oxide removal rate predominantly depends on the hydrogen ion flux at ion energy larger than 1 eV.

We also obtained the absorption and resurfacing barriers (\(E_{absorp}\) and \(E_{resurf}\)) of 0.87 and 0.62 eV, respectively, by fitting the model to our experimental data. These values have been theoretically computed by Onwudinanti et al. using DFT14 for hydrogen atoms and Ru. Their work shows that octahedral sites are energetically favorable for hydrogen in the Ru subsurface, and \(E_{absorp} = 1.06\) eV and \(E_{resurf} = 0.27\) eV for a \(\frac{1}{2}\) monolayer of hydrogen surface coverage. These computed values are different from the parameters we obtained. To check the influence of these differences, we vary \(E_{absorp}\) and \(E_{resurf}\) in our model and calculate the deuterium retention for the nominal plasma source setting, as shown in Fig. 14. We discover that the simulated retention significantly deviates from the measured data as we increase \(E_{absorp}\) toward the computed value 1.06 eV or decrease \(E_{resurf}\) toward the computed value of 0.27 eV. This shows that both \(E_{absorp}\) and \(E_{resurf}\) have notable impacts on the simulated retention.

To explain the difference in \(E_{absorp}\) and \(E_{resurf}\), we could consider the influence of the hydrogen surface coverage. In Ref. 34, the absorption and resurfacing barriers were computed for a \(\frac{1}{2}\) monolayer of hydrogen surface coverage. However, it has been reported in Ref. 16 that for diffusion into the Ru bulk, hydrogen at the Ru subsurface has a lower energy barrier to overcome for a hydrogen-adsorbed surface than a hydrogen-free one. Besides, it is unfavorable for hydrogen at the subsurface to escape from Ru...
when the surface site is occupied by hydrogen. These indicate that the hydrogen uptake rate could increase with the hydrogen surface coverage, which means that the absorption barrier effectively decreases and the resurfacing barrier increases as the hydrogen surface coverage increases. This effect of the hydrogen surface coverage on the absorption barrier has also been reported for Co and W surfaces using DFT.\textsuperscript{3,25} Furthermore, this finding for W has been verified using a reaction–diffusion model based on experimental data.\textsuperscript{54} Therefore, for the Ru surface, dependence of the absorption and resurfacing barriers on the hydrogen surface coverage could explain the different $E_{\text{ah}}$ and $E_{\text{resurf}}$ determined by our reaction diffusion model and the DFT computation in Ref. 34.

Besides the free parameters determined by fitting, it is also worthwhile to note the negligible effect of direct ion implantation into Ru in our model. Influence of ion penetration on the deuterium retention in plasma-exposed W has not been fully understood.\textsuperscript{53,54} It has been reported that the deuterium retention increases with the ion energy for W exposed to a deuterium plasma.\textsuperscript{2,25} These experiments were carried out at ion energy of several tens of eV. When the ion energy is high enough, the effect on deuterium retention can be attributed to penetration of the W surface by energetic deuterium ions.\textsuperscript{55} In this regime, Baldwin \textit{et al.} showed a possible relationship between the penetration flux into W and the incoming ion flux from the plasma.\textsuperscript{57} If we apply this relationship to our experimental condition, the penetration flux is approximately ten times lower than the ion flux, leading to a flux in the range of $10^{18}$ m$^{-2}$s$^{-1}$ into the subsurface. However, we do not observe this effect in our experiments. After removal of the RuO$_2$, the steady-state deuterium uptake rate is only $2.1 \times 10^{15}$ m$^{-2}$s$^{-1}$ regardless of the ion flux, as shown in Fig. 10(b).

The ion energy also plays an unimportant role based on the similar deuterium uptake rates for the plasma source settings of “Nominal” and “High ion energy.” Our results show that the Ru subsurface is not directly filled by the incoming ion flux with 20 eV. We cannot explain this based on the penetration depth of D$_3^+$ ions at 20 eV into Ru, calculated by SRIM to be approximately twice the Ru lattice parameter $\lambda_{Ru}$. It has been reported by ‘t Hoen \textit{et al.}\textsuperscript{17} that direct penetration of ions with energy less than 5 eV could be largely suppressed by the deuterium surface coverage. Although the deuterium surface coverage reaches 0.74 upon plasma exposure in our model, we still cannot rationalize the insignificant ion penetration using the argument by ‘t Hoen \textit{et al.} because the ion energy in our experiment was 20 eV. However, van den Bos \textit{et al.} reported that hydrogen ion energy has a significant influence on blister formation in Si irradiated by hydrogen ions and observed no blisters for an ion energy of 20 eV.\textsuperscript{15} This was explained by the low hydrogen uptake rate because of the shallow penetration depth of 20-eV hydrogen ions, which lead to a very low hydrogen concentration very close to the surface and possibly a large diffusion coefficient toward the vacuum than into the bulk.\textsuperscript{58} More experiments and even computational studies are needed to understand the influence of the ion energy on the deuterium uptake of Ru. This also shows the importance of near-surface processes for investigation of hydrogen uptake of plasma-facing materials.

V. CONCLUSIONS

Blistering of EUV mirrors originates from hydrogen uptake of the Ru surface exposed to a hydrogen plasma. In this work, we performed both experiments and modeling to study the influence of plasma parameters on deuterium uptake on a Ru surface. The designed targets were exposed to deuterium plasmas with pre-measured plasma parameters, and the deuterium retention in the targets was measured using ERD. In contrast to ion-driven hydrogen uptake, the deuterium uptake rate of the Ru film is nearly independent of the plasma parameters after a plasma exposure duration, whereas the onset of deuterium uptake depends on the ion energy and flux from the plasma. To understand the mechanism, we built a reaction–diffusion model, which includes a time-evolving surface composition and can quantitatively describe the experimental data. Based on the model, we conclude that the deuterium uptake is limited by the surface composition of the Ru film, covered by one monolayer of Ru oxide as deposited. Since deuterium sticks readily to metallic Ru but not to Ru oxide at room temperature, the onset of the deuterium uptake is governed by removal of Ru oxide at the surface. Once the metallic Ru surface emerges, it quickly becomes populated by deuterium due to neutral atom bombardment, and the high deuterium surface coverage lowers the activation energy of deuterium absorption into the bulk. As a result, in the steady state, the deuterium uptake rate is predominantly determined by the deuterium surface coverage and the absorption energy barrier. This study shows the importance of investigating influence of the surface properties on the plasma-induced deuterium uptake, to which we successfully apply a reaction–diffusion model, including a time-evolving surface. The results suggest that for EUV mirrors, the chemical state of Ru at the surface plays a vital role in the hydrogen uptake rate and the subsequent blister formation. Our approach is in line with the emerging trend in the research on deuterium uptake, and our study could be helpful to further understand effects of plasma surface interactions on deuterium uptake.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.
Author Contributions

S. C. Wang: Conceptualization (lead); Data curation (lead); Formal analysis (lead); Investigation (lead); Methodology (lead); Resources (lead); Software (lead); Validation (lead); Visualization (lead); Writing – original draft (lead). E. Zoethout: Data curation (supporting); Formal analysis (supporting); Investigation (supporting); Writing – review & editing (supporting). M. van Kampen: Conceptualization (supporting); Investigation (supporting); Project administration (supporting); Resources (supporting); Supervision (equal); Writing – review & editing (equal). T. W. Morgan: Funding acquisition (lead); Supervision (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are openly available in 4TU.ResearchData at https://doi.org/10.4121/20774977, Ref. 60.

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