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Recoil energy distribution of hydrogen isotopes adsorbed on tungsten

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Abstract

The energies of adsorbed H and D recoiled from tungsten surfaces during bombardment with 3 keV Ne^+ at oblique angles of incidence were measured. The energy spectra show structure that extends above the elastic recoil energy. We find that the high-energy structure results from multiple collisions, namely recoil of a H isotope followed by scattering from an adjacent W atom, and vice versa. This scattering assisted recoil process is especially prevalent for H isotopes adsorbed on W, owing to the large mass difference between the scattering partners. Such processes will tend to enhance H isotope recycling from plasma-facing W surfaces and reduce energy transfer to the W substrate.

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1. Introduction

Tungsten shows much promise as a plasma-facing material to use in magnetic fusion energy devices, both on the divertor and first-wall in tokamaks, and on plasma-forming electrodes in various advanced concepts [1–6]. Its refractory nature, moderate neutron activation properties, low retention of hydrogen isotopes and low sputter erosion rate are key features.

Tungsten readily adsorbs molecular hydrogen [7], leading to a saturated monolayer coverage that will exist on all bare W surfaces exposed to the hydrogen isotope fill gas prior to the start of a plasma discharge. Adsorbed hydrogen isotopes rapidly exchange with gas phase molecules and can be efficiently removed by plasma particle bombardment. At low incident energies, e.g., <1 keV, hydrogen isotope atoms and ions have a high probability of reflecting from W surfaces. Taken together, prompt adsorption, desorption and reflection of hydrogenic particles make W a high recycling material.

The term ‘recycling’ as used in this article has a broader meaning than is usually taken. It refers not only to direct reflection of hydrogen isotopes from a plasma-facing surface, but also to the release of adsorbed hydrogen isotopes by the impact of plasma particles on the surface. Thermal desorption is an additional process that releases adsorbed hydrogen and in a general sense contributes to recycling, but will not be explicitly considered further here.

The higher-energy portion (i.e., well above thermal energies) of the recycle flux results from direct reflection of energetic incident hydrogenic particles and from recoil of adsorbed hydrogen isotopes by energetic plasma

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particles. The reflection component of recycling has been well studied with both modeling and experiment, but the recoil component has received less attention and is the subject of this study.

Heavier particles are more efficient in sputtering substrate atoms and in recoiling hydrogen isotopes, so plasma impurities may play a disproportionately large role in plasma materials interactions. An example is Ne, which may be added in minor quantities in a fusion reactor to increase edge radiation. Its effect on erosion and recycling could be significant. We consider the recycling aspect by examining D^+ recoil emission during Ne^+ bombardment of W surfaces being exposed to D_2(g).

2. Experimental

The measurement apparatus consists of a mass analyzed, low-energy ion source, UHV analysis chamber, and an electrostatic ion energy analyzer. It has been previously described [8]. Samples examined were polycrystalline high purity W (99.95%) that had been mechanically polished and single crystal W that had been oriented and polished along the (100) plane. The samples were first cleaned in-situ using a combination of sputter bombardment and annealing. Ion scattering measurements confirmed that the surface of each sample had only minor amounts of contamination (<1% O). The energy spectrum of positive ions emitted from the sample was then measured with a 3 keV Ne^+ beam incident at 18° from the surface in vacuum and during exposure to 1–10 μPa D_2. All measurements were made at room temperature.

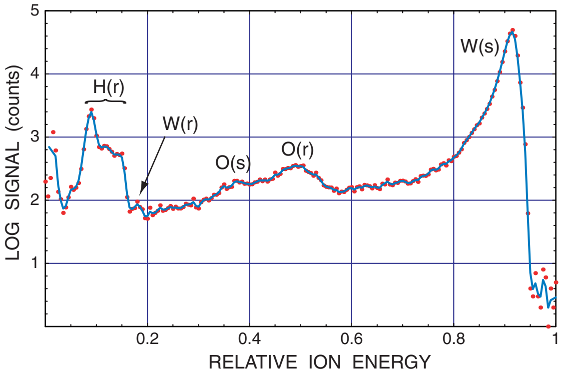

Fig. 1. Ion energy spectrum recorded from W(100) at a 45° laboratory observation angle during 3 keV Ne^+ bombardment along the [100] azimuth. The energy scale is relative to the incident ion energy. Peaks in the distribution (occurring at relative energies shown in parentheses) are assigned to recoil (labeled r) emission of H (0.09), O (0.49), and W (0.18) and scattering (labeled s) of the incident Ne^+ by O (0.36) and W (0.92) atoms. The signal intensity at the lowest energies results from secondary ion emission.

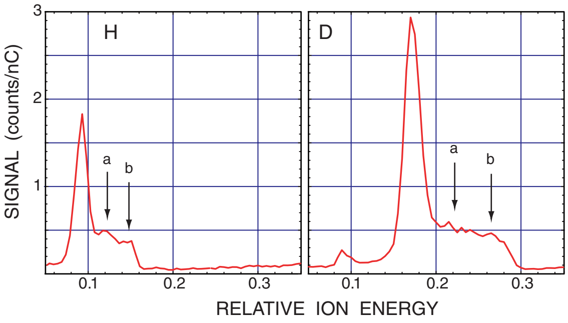

Fig. 2. Detail of ion energy spectrum of the recoil emission of H^+ from a sputter clean W sample in vacuum (left panel) and of D^+ from a sputter cleaned W sample during exposure to 10 μPa D_2(*g*) (right panel). The main peak in each case is from direct recoil, while the high-energy structure and weak peaks labeled *a* and *b* are attributed to multiple collisions.

3. Results and discussion

The energy distribution of positive ions emitted from a W(100) sample during 3 keV Ne^+ bombardment is shown in Fig. 1. A classical kinematic analysis shows the structure observed in the spectrum to result from quasi-elastic binary collisions involving H, O, and W surface atoms. The main peak is Ne^+ which has been scattered from surface W atoms. Other peaks are identified as Ne^+ scattered from surface O atoms, O^+ recoils, H^+ recoils, and possibly W^+ recoils. The sample had been sputter cleaned in UHV prior to recording the spectrum, so the presence of H and O on the surface is attributed to adsorption of residual gases in the chamber or intrinsic impurities in the sample rather than a native contamination layer. The H^+ recoil peak has an unusual structure compared to the other peaks in the spectrum. As shown in Fig. 2, there is a main peak that has an energy close to the elastic recoil energy for H recoiled by Ne at a 45° angle and a higher-energy component that appears to have two subsidiary peaks. We endeavored to understand the origin of this structure.

An isotope exchange experiment using D_2 demonstrated that the high-energy structure above the H^+ recoil peak is associated with the presence of H and is not due to other species on the surface. When the sample was exposed to D_2(*g*) at 10 μPa, the H^+ recoil signal nearly disappeared and a new peak appeared at a higher energy. The new peak was identified as resulting from D^+ recoils. Interestingly, the D^+ recoil signal had a structure similar to that observed for H^+ recoils, as shown in Fig. 2. When the D_2(*g*) supply was shut off, the D^+ recoil signal eventually disappeared and was replaced by a H^+ recoil peak. The origin of the adsorbed H is uncertain; it arose either from adsorption of H containing residual gases in the analysis chamber (e.g., H_2, H_2O, CH_4) or diffusion to the surface of bulk H contained in the sample.

Examination of the H^+ and D^+ recoil signals suggests that three peaks may be present in the recoil energy distribution for each isotope, as indicated in Fig. 2. The main peak is located at the elastic recoil energy and is simply explained as resulting from direct recoil of a H or D atom in a binary collision with an incident Ne. The upper two peaks (labeled *a* and *b*) cannot be the result of an inelastic energy loss process, since they occur at an higher energy than the elastic recoil energy.

A possible explanation for the recoil structure is that the high-energy peaks result from multiple collisions with surface atoms. This speculation is motivated by the fact that two or more weak collisions generally result in a smaller energy loss in comparison to a single collision giving the same deflection angle. Two likely collision sequences are (1) scattering of incident Ne by W followed by recoil of a H or D atom by Ne and (2) recoil of H or D by incident Ne followed by scattering of H or D by a W substrate atom. The calculated final H and D energies resulting from these sequences for the experimental observation angle (45°) is plotted in Fig. 3. Also shown are the observed *a* and *b* peak energies for H and D recoil, plotted as horizontal lines. The intersections of these lines with the calculated energy curves indicate the recoil angles in each collision sequence. It can be seen that for both H and D, the recoil energies for peaks *a* and *b* correspond to recoil angles of 35° and 27°, respectively. This coincidence indicates that the same multiple collision sequence occurs for both hydrogen isotopes. Further, since the collision order has little effect on the final particle energy at recoil angles >25°, either collision sequence (1) and (2) produces the same result. Consequently, it is not possible to determine whether or not one particular collision sequence dominates in producing the observed recoil ion energy spectrum. However, it seems reasonable, based on the equivalency principle, that both sequences are likely to occur. Perhaps the similar final recoil ion energies of the two sequences helps to accentuate the observed peak structure.

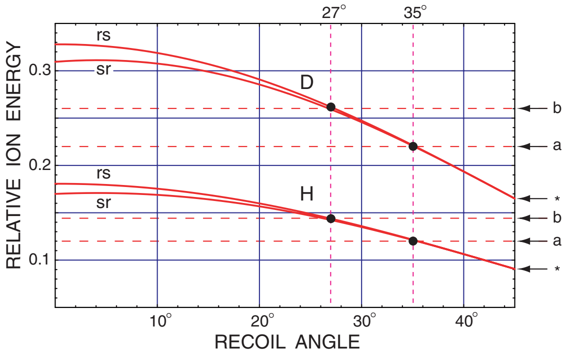

Fig. 3. The calculated final particle energy of H and D emitted from a W surface following scattering and recoil (sr) or recoil and scattering (rs) from the substrate, plotted as a function of the recoil angle. The total deflection angle for the two sequential collisions is 45°. The observed peak energies (∗, *a*, *b*) are shown on the ordinate and recoil angles of 27° and 35° are inferred from the kinematic curves.

Information about the location of adsorbed H and D on the W surface can be obtained from the inferred recoil angles. This was done by constructing recoil ion trajectories for H and D atoms placed in various adsorption sites and comparing the calculated emission angles with the observations. It was found that the high energy peaks in the distribution appear to correlate with recoil of H and D from high-symmetry binding sites on the W(100) surface. This is illustrated in Fig. 4, which shows two trajectory paths consistent with the experimental data. In one case, the H or D atom is located in the bridge site and in the other it is located in the hollow site on the W(100) surface. This suggests that peak *a* in the recoil energy distribution corresponds to recoil from hydrogen isotopes adsorbed in the bridge site and peak *b* to hydrogen isotopes adsorbed in the hollow site. It is interesting to observe that both sites appear to be occupied on the W(100) surface, in agreement with calculations that predict the two sites to have similar chemisorption bond strengths.

The occurrence of scattering-assisted H recoil is favored on W surfaces due to the large mass difference between the collision partners, which results in a small transfer of momentum from H to W and a large scattering cross section of H from W. This enhances both the energy and particle reflection coefficients for H isotope recoils from W. As a result, one expects that in a fusion reactor less incident particle energy would be transferred to a W wall and more of the energy reflected back into the plasma than would be the case for lighter wall materials. So, it may be said that the scattering assisted recoil process contributes to the H isotope recycling properties of tungsten.

It is useful to consider the magnitudes of the various recycling processes. With respect to the present results, a basic question is: does scattering assisted recoil make a significant contribution to overall H isotope recycling? In many cases, e.g., below a few hundred electron volts, the major fraction of the recycle flux is due to reflection of incident particles and the balance is accounted by recoil emission. The specific apportionment between reflection and recoil depends upon many parameters, including the composition of the incident flux, its energy spectrum, impact angle, and the surface concentration of adsorbed H isotopes. In considering cases in which scattering assisted recoil could be a significant process, the

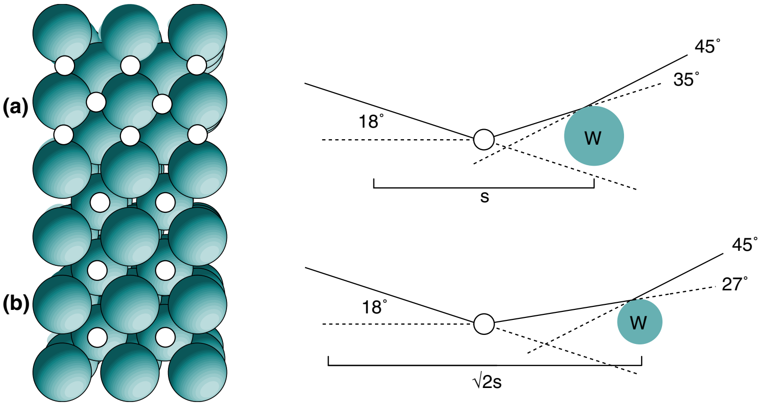

Fig. 4. Constructed ion trajectories and interpretation of the observed high energy recoil peak structure and inferred recoil angles. Peak *a* corresponds to recoil of H and D adsorbed in bridge positions on the W(100) surface and peak *b* corresponds to recoil from hollow positions. In the diagrams, *s* is the W lattice parameter and the size of the W atom is set equal to the impact parameter.

following conditions are expected to accentuate the proportion of recycled hydrogen that arises from scattering assisted recoil: a high fraction of impurities in the incident flux, a high partial pressure of thermalized gaseous hydrogen isotopes in the vicinity of the W surface, and a W surface temperature below that needed for thermal desorption of chemisorbed hydrogen isotopes. Such conditions may be encountered at the first wall during startup and soon after gas-puffing, pellet injection, or the start of neutral beam injection. A preliminary estimate for a incident hydrogen flux containing 3% Ne, based on the experimental data along with calculated recoil and scattering cross sections, indicates that 1–10% of the recycle flux could originate from scattering assisted recoil if more than a half monolayer coverage of adsorbed hydrogen isotopes is present on the W surface. While it is not the main source of recycled hydrogen isotopes, scattering assisted recoil is prevalent enough to merit consideration in detailed models of recycling, as it will affect both the total number and energy of recycled particles.

Acknowledgment

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References

[1] K.S. Zhen, Y.X. Zeng, W.D. Xi, L. Xiang, M.Z. Nian, Surf. Coatings Technol. 131 (2000) 109.
[2] M.A. Ulrickson, J.N. Brooks, D.E. Driemeyer, et al., Fus. Eng. Design 58&59 (2001) 907.
[3] A. Hassanein (Ed.), NATO Science Series II: Mathematics, Physics, and Chemistry, vol. 54, 2002, p. 235.
[4] R.A. Causey, J. Nucl. Mater. 300 (2002) 91.
[5] T. Hirai, M. Rubel, V. Philipps, et al., Phys. Scr. T 103 (2003) 59.
[6] Y. Ueda, K. Tobita, Y. Katoh, J. Nucl. Mater. 313–316 (2003) 32.
[7] K. Heinz, L. Hammer, Phys. Status. Solidi. 159 (1997) 225.
[8] R. Bastasz, J.A. Whaley, Fus. Eng. Des. 72 (2004) 111.