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# Level-energy-dependent mean velocities of excited tungsten atoms sputtered by krypton-ion bombardment

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Visible emission spectra were acquired from neutral atoms sputtered by 35–60 keV Kr<sup>+</sup> ions from a polycrystalline tungsten surface. Mean velocities of excited tungsten atoms in seven different 6p states were also obtained via the dependence of photon intensities on the distance from the surface. The average velocities parallel to the surface normal varied by factors of 2–4 for atoms in the different 6p energy levels. However, they were almost independent of the incident ion kinetic energy. The 6p-level energy dependence indicated that the velocities of the excited atoms were determined by inelastic processes that involve resonant charge exchange. © 2015 American Vacuum Society. [http://dx.doi.org/10.1116/1.4928250]

#### I. INTRODUCTION

Diverter plates in a fusion device are exposed to high-intensity fluxes of energetic particles. Tungsten (W) is a candidate material for diverter plates in the international thermonuclear experimental reactor because of its high melting temperature, good thermal conductivity, and high threshold energy for erosive sputtering.<sup>1-3</sup> Tungsten retains tritium and deuterium bombarded during hydrogen isotope plasmas. Retention of these isotopes poses safety and economic issues that must be mitigated in fusion reactor operations. Thus, considerable effort is being made to predict hydrogen isotope retention in various forms of tungsten under actual fusion reactor conditions. Furthermore, dust formation in plasmas is attracting extensive attention with regard to the inventory of tritium atoms in a nuclear fusion device.<sup>1,4</sup> Erosion due to sputtering of a diverter plate is one of the critical issues in determining the lifetime of plasma-facing components (PFCs). Sputtered atoms and reflected ions, generated in PFCs by plasma-wall interactions, become impurities that are transported into the core plasma along magnetic field lines. Hence, they determine in large part the condition of the fusion plasma.<sup>5</sup>

One of the disadvantages of tungsten diverters is the large radiative power dissipation ("radiation cooling") by sputtered excited atoms and ions.<sup>6</sup> The sputtered neutrals can penetrate the plasma core across the magnetic field near the divertor.<sup>7</sup> Because the penetration depth depends on the initial velocity, it is essential to know the velocities of the sputtered tungsten atoms. However, these data are usually unavailable because they require technically challenging measurements.

Impurity seeding with Ne, Ar, Kr, Xe, or N<sub>2</sub> has been used to reduce heat loads to PFCs by radiation.<sup>8</sup> These gases actually improve plasma performance; however, they also contribute to surface sputtering. Furthermore, tungsten atoms sputtered by impurity ions cannot be ignored, even at low concentrations (e.g.,  $\sim 0.2\%$  Ar ions<sup>8</sup>), because those sputtering yields are orders-of-magnitude larger than those from impurity D or C ions.<sup>9</sup> Therefore, we have examined the kinetic energies of W atoms sputtered by energetic Kr<sup>+</sup> ions.

Previously,<sup>10</sup> we used an optical method to measure the mean velocity of tungsten atoms excited in the  $5d^56p$  state sputtered by Kr<sup>+</sup> ions at normal incidence on a polycrystalline tungsten surface. No significant variation in velocity was found for the different projectile energies. The main objective of the present study is to verify that the reproduction of the previous results for other seven 6p excited states of W atoms. After a brief explanation of our experiments in Sec. II, the results and discussion are given in Sec. IV.

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#### **II. EXPERIMENT**

Details concerning the experimental procedures are described elsewhere<sup>10</sup> and will be briefly summarized here. As depicted in Fig. 1, the experiments were performed with a medium-current ion implanter (ULVAC IM-200MH-FB) connected to a beam line at the National Institute for Fusion Science (Toki, Japan).

The 35–60-keV Kr<sup>+</sup> ion beam was introduced into the vacuum chamber after mass/charge separation and was transmitted through a 5-mm-diameter aperture before impinging normally on a polycrystalline tungsten surface mounted on a linear-motion manipulator. A bias voltage of ~100 V was applied to the disk, with an aperture hole used to retard secondary electrons emitted from the tungsten surface. The base pressure of the vacuum chamber was maintained at  $\sim 1 \times 10^{-6}$ Pa without ion beam. The pressure reached up to  $\sim 3 \times 10^{-5}$ Pa during ion-beam irradiation of the tungsten surface. In addition, the surface coverage of oxygen on the sample surface was estimated to be less than 0.04 during irradiation as a result of the high current density ( $\sim 150 \ \mu A/cm^2$ ).<sup>10</sup>

Visible spectra of the sputtered plume were acquired with a monochromator equipped with an electronically cooled charge coupled device (CCD). The normalized photon



FIG. 1. Schematic of the experimental apparatus. IS, ion source; MS, mass separator; QL, quadruple lens; EL, einzel lenses; BM, beam-profile monitor; DP, deflection plates; AH, aperture hole; TS, target sample; LM, linear-motion manipulator; QW, quartz window; OL, optical UV lens; and MM, monochromator equipped with a CCD.

intensity I(z) of a specific line spectrum was recorded as a function of distance z from the tungsten surface. The mean velocity  $\langle v_z \rangle$  parallel to the surface normal was determined by analyzing multiexponential functions approximated by the following equation:

$$I(z) \cong \sum_{k=1}^{n} I_k \exp\left(-\frac{z}{z_k^*}\right),\tag{1}$$

where  $I_k$  is the photon intensity (normalized by the ion-beam current) of the *k*th component at z = 0 (surface), when the survival probability of the excited state is unchanged.<sup>11,12</sup> The origin of z was defined as the position where I(z) was maximum. The quantity  $z_k^*$  is the characteristic distance equal to the product of  $\langle v_z \rangle$  and the lifetime  $\tau_k$  of the excited state; i.e.,  $z_k^* = \langle v_z \rangle \tau_k$ . The value *n* was determined by the number of straight lines drawn in a semilogarithmic plot of I(z). Data near the surface may be affected by short-lived cascade transitions,<sup>10</sup> nonradiative transitions,<sup>10,12</sup> selfabsorption of resonance lines by sputtered atoms, and light scattering. Therefore, we did not include data for z < 0.6 mm. The linear dependence of the observed photon intensity with the ion beam current indicated that no secondary processes occurred in the collisions.<sup>10</sup> Spectra were calibrated with H<sub>2</sub>, He, and Hg discharge lamps.

#### **III. RESULTS AND DISCUSSION**

#### A. Optical emission spectra of tungsten atoms

Figures 2(c) and 2(d) show optical emission spectra acquired from the tungsten surface (z = 0) bombarded by a 35-keV Kr<sup>+</sup> ion beam. The spectra were obtained by integrating the monochromator/CCD data in Figs. 2(a) and 2(b), respectively, along the *y*-direction. The background was not subtracted, and the W and Kr atomic lines are labeled. The wavelengths, transitions, and excited-state lifetimes are summarized in Table I.

Sixteen sharp W(I) lines and four slightly broadened Kr(II) lines were observed. Doppler broadening of the Kr(II) lines indicated that the backscattered Kr<sup>+</sup> ions had higher velocities in the direction parallel to surface than the sputtered tungsten atoms,<sup>15</sup> as shown in the insets of Figs. 2(c) and 2(d). However, the elongated lines in the tungsten spectra along the *y*-direction in Figs. 2(a) and 2(b) indicate large-angle atom sputtering with respect to the normal direction (*z* axis). However, the absence of W(I) spectra extending beyond the CCD-image area verifies that most of the sputtered atoms were observed.

The energy-level diagram of atomic tungsten is shown in Fig. 3, where  $\phi = 4.55 \text{ eV}$  is the work function of bulk polycrystalline tungsten,<sup>16</sup>  $I_{ex}$  is the atomic ionization potential of the excited state, and  $\Delta U = \phi - I_{ex}$ . There are four transitions with a common excited state: group 1 (W2 and W3) with the excited state of 5d<sup>4</sup>6s(<sup>6</sup>D)6p <sup>7</sup>D<sub>4</sub>; group 2 (W5 and W14) with 5d<sup>4</sup>6s(<sup>6</sup>D)6p <sup>7</sup>D<sub>3</sub>; group 3 (W12, W15, and W16) with 5d<sup>4</sup>6s(<sup>6</sup>D)6p <sup>7</sup>D<sub>2</sub>; and group 4 (W9 and W11) with 5d<sup>5</sup>(<sup>6</sup>S)6p <sup>7</sup>P<sub>3</sub>. Atoms with a common excited state should have equal  $\langle v_z \rangle$ .



Fig. 2. (Color online) (a) and (b) Monochromator CCD images of the tungsten surface (z = 0) bombarded by 35-keV Kr<sup>+</sup> ions. (c) and (d) Optical emission spectra corresponding to integration of the CCD images (a) and (b) along the *y* direction, respectively.

#### B. Decay curves of photon-emission intensities

Figure 4 is a semilogarithmic plot of the normalized intensities of group 1 as a function of distance from the

surface. W2 and W3 are nearly identical, and the data can be fit with a double exponential with two characteristic distances  $z_k^*$  in Eq. (1); i.e., n = 2. This suggests that W atoms in the 5d<sup>4</sup>6s(<sup>6</sup>D)6p <sup>7</sup>D<sub>4</sub> state are produced *via* two paths that

TABLE I. Wavelengths, transitions (Ref. 13), and excitation lifetimes (Ref. 14) of assigned spectra from a tungsten surface bombarded with 35-keV Kr<sup>+</sup> ions.

Specie	Wavelength (nm)	Transition	Lifetime $(\tau_1)$ of the upper level (ns)	Label
W I	386.80	$5d^46s(^6D)6p\ ^7D_4 \rightarrow 5d^5(^6S)6s\ ^7S_3$	185	W2
	400.88	$5d^{5}(^{6}S)6p^{7}P_{4} \rightarrow 5d^{5}(^{6}S)6s^{7}S_{3}$	59.4	W1
	404.56	$5d^46s(^6D)6p \ ^5F_2 \rightarrow 5d^5(^6S)6s \ ^7S_3$	182	W6
	407.44	$5d^{5}(^{6}S)6p^{7}P_{3} \rightarrow 5d^{5}(^{6}S)6s^{7}S_{3}$	86	W9
	410.27	$5d^46s(^6D)6p\ ^5P_3 \rightarrow 5d^46s^2\ ^5D_4$	50	W10
	413.75	$5d^{5}(^{6}S)6p^{7}P_{3} \rightarrow 5d^{4}6s^{2} {}^{5}D_{2}$	86	W11
	417.12	$5d^46s(^6D)6p\ ^7D_4 \rightarrow 5d^46s^2\ ^5D_3$	185	W3
	424.44	$5d^46s(^6D)6p\ ^7D_5 \rightarrow 5d^46s^2\ ^5D_4$	695	W7
	426.94	$5d^{5}(^{6}S)6p \ ^{7}P_{2}{}^{a} \rightarrow 5d^{5}(^{6}S)6s \ ^{7}S_{3}$	315	W8
	429.46	$5d^{5}(^{6}S)6p \ ^{7}P_{2}^{\ b} \rightarrow 5d^{5}(^{6}S)6s \ ^{7}S_{3}$	76.1	W4
	430.21	$5d^46s(^6D)6p\ ^7D_3\to 5d^5(^6S)6s\ ^7S_3$	161	W5
	448.42	$5d^46s(^6D)6p \ ^7D_2 \rightarrow 5d^46s^2 \ ^5D_1$	250	W12
	465.99	$5d^46s(^6D)6p \ ^7D_1 \rightarrow 5d^46s^2 \ ^5D_0$	275	W13
	468.05	$5d^46s(^6D)6p\ ^7D_3 \rightarrow 5d^46s^2\ ^5D_3$	161	W14
	475.75	$5d^46s(^6D)6p^7D_2 \rightarrow 5d^5(^6S)6s^7S_3$	250	W15
	484.38	$5d^46s(^6D)6p\ ^7D_2\to 5d^46s^2\ ^5D_2$	250	W16
Kr II	435.55	$4s^24p^4(^3P)5p \ ^4D_{7/2} \rightarrow 4s^24p^4(^3P)5s \ ^4P_{5/2}$		Kr1
	457.72	$4s^24p^4(^1D)5p\ ^2F_{7/2} \rightarrow 4s^24p^4(^1D)5s\ ^2D_{5/2}$		Kr2
	473.90	$4s^24p^4(^3P)5p \ ^4P_{5/2} \rightarrow 4s^24p^4(^3P)5s \ ^4P_{5/2}$		Kr3
	476.57	$4s^24p^4({}^3P)5p\ {}^4D_{5/2} \to 4s^24p^4({}^3P)5s\ {}^4P_{3/2}$		Kr4

<sup>a</sup>Lande g = 0.87.

<sup>b</sup>Lande g = 1.84.



FIG. 3. (Color online) Energy level diagram of atomic tungsten. W1–16: transitions labeled in Table I, where S, P, D, and F refer to the total angular momentum. The dashed line refers to the Fermi level of bulk tungsten.

were previously verified.<sup>17–19</sup> One is direct excitation with a short lifetime  $\tau_1$ , while the other is a cascading excitation with a long lifetime  $\tau_2$ .

Dzioba et al.<sup>17</sup> observed that cascades from upper states having longer lifetimes than those of the excited state of interest cause tails in the z dependence of the photon intensity. Later, Szymonski et al.<sup>18</sup> proposed a modified model that used a double exponential with two different lifetimes and a common velocity. Doger and Qayyum<sup>19</sup> demonstrated that the lifetimes for cascading transitions in C I and C II line spectra from 10-keV Ar<sup>+</sup> bombardment of graphite can be estimated by the "cascade-correction model,"<sup>20,21</sup> and were in good agreement with experimental data.<sup>22</sup> The function used in that model was equivalent to the double exponential (n=2) mentioned above, implying that  $\tau_1$  and  $\tau_2$  in Eq. (1) correlate with the direct and cascading lifetimes, respectively.<sup>19–21</sup> Furthermore,  $I_1$  and  $I_2$  in Eq. (1) can be expressed in terms of the direct  $N_1$  and cascade  $N_2$  level populations,19-21



FIG. 4. (Color online) W2 and W3 intensities from 35-keV Kr<sup>+</sup> ion bombardment as a function of distance z from the surface. • W2 (386.80 nm), W3 (417.12 nm), ---- curve fit with  $z_1^* = 0.6$  mm, --- curve fit with  $z_2^* = 1.9$  mm.

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$$I_1 = I(0) \frac{N_1(\tau_2 - \tau_1)}{N_1(\tau_2 - \tau_1) + N_2 \tau_1},$$
(2)

$$I_2 = I(0) \frac{N_2 \tau_1}{N_1 (\tau_2 - \tau_1) + N_2 \tau_1}.$$
(3)

Kinetic energies of sputtered excited atoms that are estimated by the model without cascade correction are large relative to those obtained by sputtering theory.<sup>23</sup> However, the velocities of sputtered excited atoms estimated with consideration of the cascade effect were 30%-50% lower than those estimated without considering this effect.<sup>20,21</sup> We obtained a good fit to the decay curve by substituting  $\tau_1$ (from Table I),  $\langle v_z \rangle$ , and  $\tau_2$  into Eq. (1). The results for the W1-W7 and W9 spectra are summarized in Table II. The error bars for  $\langle v_z \rangle$  include a ~10% fluctuation<sup>10,11</sup> because of the low oxygen coverage noted in Sec. II, as well as from fitting errors of the double exponential functions. The  $\tau_2$ error bars are due only to fitting. The mean normal velocities averaged over the 35-60-keV ion bombardment range varied significantly, even among transitions from excited states with the same 6p active electron.

Figures 5 and 6 plot the dependences on Kr<sup>+</sup> bombardment energy of the mean normal velocities  $\langle v_z \rangle$  of sputtered W atoms in the W2, W3, W4, W5, W6, W7, and W9 excited states. In addition, the *z*-component of the most probable speed  $v_z^{\text{mps}}$  calculated by the sRIM–2013 code<sup>24</sup> is plotted in Fig. 6. All the values of  $\langle v_z \rangle$  in Figs. 5(a)–5(d) and 6(a)–6(c) were independent of the Kr<sup>+</sup> energy, within experimental error.

#### C. Discussion

The kinetic energy distribution (KED) of sputtered tungsten atoms calculated by the ACAT code<sup>25</sup> has a peak at ~4.45 eV for Kr<sup>+</sup> ions with incident energies >1 keV. This peak energy is equal to half of the 8.9-eV surface-binding energy  $E_b$  of tungsten, implying that the Thompson formula<sup>26</sup> is applicable to the KED data, except for overestimations in the high energy tail.<sup>10</sup> It also corresponds to a most probable speed  $v_z^{\text{mps}} = 2.2 \text{ km/s}$ , which is close to the mean normal velocity  $\langle v_z \rangle = 2.5 \text{ km/s}$  of the W3 line and the averaged value  $\overline{v_z^{\text{mps}}} = 2.6 \text{ km/s}$  of the SRIM simulations. Although

TABLE II. Mean normal velocities  $\langle v_z \rangle$  of sputtered excited tungsten atoms and cascade lifetimes  $\tau_2$  of the upper levels for the W1–W7 and W9 spectra observed on a tungsten surface under Kr<sup>+</sup> ion bombardment.

$\tau_2 (\mu s)$
$0.30 \pm 0.11^{b}$
$0.91 \pm 0.49$
$0.91\pm0.28$
$0.36 \pm 0.09$
$0.81 \pm 0.33$
$1.1 \pm 2.2$
$3.3 \pm 2.2$
$0.43\pm0.11$

<sup>a</sup>Averaged value over the 35–60 keV bombardment range. <sup>b</sup>Reference 10.



FIG. 5. (Color online) (a)–(d): Mean velocities ( $\bullet$  experiment and — average) of W2, W3, W4, and W5 transitions as a function of the incident Kr<sup>+</sup> bombardment energy  $E_i$ .

there was no significant variation of  $v_z^{\text{mps}}$  over the 35–60 keV incident energy range, as shown in Fig. 6(d), the mean normal velocity of the sputtered atoms is expected to increase with  $E_i$ . The component of the high-energy tail due to binary collisions between Kr<sup>+</sup> ions and surface atoms contributes to this increase. The maximum velocity of sputtered particles ( $v_s^{\text{max}}$ ) acquired in an elastic binary collision is given by<sup>27</sup>

$$v_{\rm S}^{\rm max} = \frac{\sqrt{8m_{\rm P}E_{\rm i}}}{m_{\rm T} + m_{\rm P}},\tag{4}$$

where  $m_{\rm T}$  and  $m_{\rm P}$  are the masses of the W atoms and Kr<sup>+</sup> ions, respectively. Thus,  $v_{\rm S}^{\rm max}$  is proportional to  $\sqrt{E_{\rm i}}$  for elastic collisions, as plotted in Fig. 6(d). However, this is not found in the experimental  $\langle v_z \rangle$  in Figs. 5(a)–5(d) and 6(a)–6(c), suggesting that the velocity of the excited W atoms is instead determined by inelastic processes.

Figure 7 plots  $\langle v_z \rangle$  for W1,<sup>10</sup> W2–7, and W9 as a function of the potential energy difference  $\Delta U = \phi - I_{ex}$  defined in Fig. 3. The energy level dependence of  $\langle v_z \rangle$  has a maximum at  $\Delta U \approx 0$  eV, where resonant charge exchange can easily occur, regardless of the impact energies. This trend is consistent with electron tunneling processes such as those in the resonant electron transfer model (RET).<sup>28–32</sup>



FIG. 6. (Color online) (a)–(c) Mean velocities ( $\bullet$  experiment and — average) of W6, W7, and W9 transitions. (d) *z*-component of the most probable speed  $v_z^{\text{mps}}$  ( $\Box$  SRIM and — average) simulated by the sRIM-2013 code (Ref. 24) and the maximum velocity  $v_{\text{S}}^{\text{max}}$  (dashed line) calculated with Eq. (4) as a function of the incident Kr<sup>+</sup> bombardment energy  $E_i$ .

The ejection of metal ions immersed in a sea of valence electrons is the starting point of the RET model. A sputtered particle from a collision cascade is assumed to be a positive ion that can be subsequently neutralized by picking up one



FIG. 7. (Color online) Mean normal velocity  $\langle v_z \rangle$  for various excited states [W1 (Ref. 10), W2–7, and W9] created by 35-keV ( $\bullet$ ), 45-keV ( $\blacktriangle$ ), 55-keV ( $\blacksquare$ ), and 60-keV ( $\blacklozenge$ ) Kr<sup>+</sup> ions, as a function of  $\Delta U = \phi - I_{\text{ex}}$ .

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or more electrons from the valence band. Because resonant electron transfer is the most likely neutralization process, the excited-state population decreases exponentially with the energy difference,  $\Delta U$ . Furthermore, the RET model successfully predicts state-specific KED (SS-KED) for ground and metastable states of Co, Ni, Sr, and Ag atoms detected with resonance laser ionization mass spectrometry (RIMS)<sup>28-32</sup> during 15-keV Ar<sup>+</sup> sputtering of polycrystalline targets. Different SS-KEDs were observed with RIMS for different electronic states. This was a consequence of the neutralization probability determined by the coupling between the initial metal state and the final atomic state, which is strongly dependent on the interaction time or the velocity of atoms leaving the surface. Thus, key quantities are the coupling strength and the spatial overlap of the atomic and metallic wavefunctions, and the surface-normal velocity of the sputtered atoms.<sup>31</sup> One of the most important parameters that affect the coupling strength and the spatial overlap is  $\Delta U$ . The energies of all the W excited states are within -0.07 to +0.38 eV, which are within -2.4 to +0.74 eV for Co, Ni, Cu, Sr, and Ag atoms in the RIMS experiment.<sup>31</sup> The RET model assumes collision cascades, but does not consider interactions between sputtered atoms and incoming ions. In the SRIM calculation,  $v_z^{\text{mps}}$  for Co, Ni, Cu, Sr, and Ag atoms sputtered by 15-keV  $Ar^+$  are 3.1, 3.1, 2.8, 2.2, and 2.6 km/s, respectively. These velocities are similar to those of W sputtered by 35-60-keV Kr<sup>+</sup> [Fig. 6(d)]. Therefore, the RET model is consistent with our experimental results.

The assumption in the RET model that the excited states of sputtered ions lie below, yet close to, the Fermi level is in conflict with our experimental data. As shown in Figs. 3 and 7, several of the excited states with  $\Delta U > 0$  lie slightly above the Fermi level. However, because the energy differences are small (<0.4 eV) relative to  $\phi$  and  $I_{ex}$ , and given the wide density of states of the 5d valence band for bulk tungsten,<sup>33</sup> the broadened atomic level near the surface is essentially in resonance with the occupied valence band states. The survival probability *P* of an excited atom escaping from the solid surface exponentially decreases with the ratio between survival coefficient (*A/a*) and  $v_z$ , where *A* and *a* are quantities related to the frequency and inverse distance of the nonradiative transitions, respectively,<sup>34</sup>

$$P = \exp\{-(A/a)/v_z\}.$$
 (5)

In Eq. (5), a large  $v_z$  is required for light emission from the excited atoms when the survival coefficient or the probability of nonradiative transition is small. The W1, W4, and W9 lines with 5d<sup>5</sup>(<sup>6</sup>S)6p excited-state configurations have larger  $\langle v_z \rangle$  than the W2, W3, W5, W6, and W7 lines having 5d<sup>4</sup>6s(<sup>6</sup>D)6p configurations. Thus, the probability of nonradiative transitions involving 5d<sup>5</sup>(<sup>6</sup>S)6p excited states is larger than that involving 5d<sup>4</sup>6s(<sup>6</sup>D)6p excited states. The electronic configuration in the excited state may therefore affect the survival coefficient. Both the survival coefficient of the excited states and the electron-pickup process assumed in the RET model are important in the sputtering of excited neutrals.

#### **IV. SUMMARY AND CONCLUSIONS**

The mean normal velocities of various excited W atoms during 35–60 keV Kr<sup>+</sup> ion bombardment were measured by optical emission spectroscopy. The data provide fundamental knowledge concerning plasma-wall interactions. The velocities of atoms with 5d<sup>5</sup>6p or 5d<sup>4</sup>6s6p configurations were found to be dependent on the energy difference between the work function and the ionization potential of the excited state. However, they were independent of the incident beam energy. The velocities varied over 1.2-5.2 km/s for the different energy levels energies of the excited atoms. This suggests that inelastic processes, such as resonant electron transfer, are important in the excitation of sputtered atoms. Furthermore, inelastic processes are even more important for lower collision energies than those discussed here. These results will impact not only nuclear fusion but also general plasma science.

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