Charge Exchange Signatures in X-Ray Line Emission
Accompanying Plasma-Wall Interaction

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Abstract. Directional flows of well-collimated energetic ions produced at laser-burnt-through foils were used to investigate transient phenomena accompanying the plasma interaction with surfaces of solid targets (generally known as plasma-wall interaction, PWI). The plasma jet launched from the rear surface of the 0.8-μm-thick Al foil irradiated at oblique laser incidence with an intensity of $3 \times 10^{14}$ W/cm² was incident on the quasi-massive C target. The plasma x-ray self-emission was analyzed by focusing survey and high-dispersion spectrometers. The time-integrated, spatially resolved narrow-band spectra recorded close to the C surface exhibit a dip structure in the red-wing profiles of the hydrogenic Al Lyγ line which was attributed to the charge exchange between two stationary Coulomb centers represented by the Al XIII and fully stripped C ions. This identification of the charge exchange signatures in x-ray line emission is supported by hydrodynamic simulations of environmental conditions in the near-wall plasma and by predictions of the dips positions following from complementary theoretical models. The agreement between the experiment and theory validates the first high precision x-ray spectroscopic identification of charge exchange phenomena accompanying the PWI.

1. Introduction

Being subjected to extremely high heat loads and high particle and neutron fluxes, the plasma-facing materials of present-day experimental fusion reactors experience particularly hostile environmental conditions. Many of currently used inner-wall components cannot withstand the plasma scenarios foreseen for the future commercial power plants [1]. Further optimization of plasma-facing materials needed for development of new technological concepts requires a detailed understanding of the PWI processes that occur when solid surfaces are bombarded by intense particle fluxes.

Various mechanisms of the energy transfer, material erosion and migration in the near-wall region include ion deceleration and stopping, shock waves generation, formation of highly excited Rydberg states or hollow atoms with multiple inner vacancies, charge transfer processes, and ion neutralization [for references, see e.g. 2]. The purpose of this paper is to contribute to x-ray spectroscopic diagnostics of charge exchange (hereafter CE) phenomena accompanying the PWI. We describe the experimental configuration used, present high-resolution spectroscopic data, and interpret the observed modulation of Al Lyγ profiles in terms of CE between multicharged atoms. The identification of CE phenomena is supported by hydrodynamic simulations and by predictions of two complementary theoretical models.
2. PALS experiment

The experiment has been performed on the iodine laser system of the PALS Research Centre [3]. The scheme of the experimental setup is shown in figure 1. The laser beam delivering 50-90 J of frequency-tripled radiation (438 nm) in a pulse length of 0.24-0.3 ns (full width at half maximum) was incident onto the primary, jet-producing target (0.8-μm-thick Al foil) at an angle inclined by $\alpha = 30^\circ$ from the target normal.

Figure 1. Schematic drawing of the diagnostic complex used for x-ray characterization of plasma-wall interactions.

The reason for the oblique incidence of the laser beam onto the target is explained in figure 2, where the jets formation in transmission geometry is simulated by the Prague Arbitrary Lagrangian Eulerian hydrocode PALE [4]. The 2D distribution of plasma parameters indicates that the foil burns through well before the laser pulse maximum, i.e., at normal incidence ($\alpha = 0^\circ$) the plasma jet strikes the secondary target pre-ionized by the action of the transmitted radiation. In the oblique incidence case ($\alpha = 30^\circ$), the laser beam does not hit the secondary foil and the expanding plasma jet launched in a direction of normal to the foil surface interacts with the unperturbed target, thus creating a better-characterized environment for PWI studies. The optimum conditions for formation of sufficiently high aspect-ratio jets were found when defocusing the beam to the focal spot radius of 150 μm [5].

The standard diagnostic complex used in the PALS PWI experiments consists of the time-resolved x-ray imaging of the plasma expansion, optical spectroscopy, interferometry and several x-ray spectrometers. The primary diagnostics applied in the present study was a high-resolution vertical dispersion Johann spectrometer (VJS) fitted with a cylindrically bent crystal of quartz (100) [6]. Hereafter we shall concentrate on evaluation of high-resolution spectra obtained by the VJS. The interpretation of experimental data provided by supporting diagnostics can be found in papers [2, 5].

Figure 2. Alternate schemes of the plasma jets production and their interaction with secondary targets. The simulated distribution of the plasma electron density $n_e$ corresponds to the laser pulse maximum.
3. Correlation of observed spectra and simulated environmental conditions

One of characteristic properties of the VJS is a simultaneous production of two sets of mirror symmetric spectra with one-dimensional (1D) spatial resolution perpendicular to a direction of plasma observation. The existence of the axis of symmetry facilitates the computational reconstruction of the spectra and considerably increases the reliability of identification of tiny spectral features assuming that they are observed in both parts of corresponding spectral records [6].

Figure 3. Spatially resolved spectra of Al Lyγ, δ self-emission from double-foil Al/C target recorded by the VJS. The laser strikes the primary Al target at oblique incidence from above.

The spatially resolved x-ray spectrum presented in figure 3 was recorded at the oblique incidence of the laser beam (57 J, 0.27 ns, 2.5×10¹⁴ W/cm²) striking the primary Al target from above. The Al foil and the secondary quasi-massive C target (250-μm-thick pyrolytic graphite foil) were separated by a distance of 590 μm. An expansion of the plasma jets produced at both front and rear surfaces of the Al foil (spatial coordinate 0) is visualized via the emission of the Lyman series members γ and δ of the H-like Al. The recorded line profiles are characteristic for three zones marked in figure 3. In zone I extending within approximately ±100 μm from the Al target, the emitted lines display distinct Doppler shifts due to directional reversal of the plasma jets outflow at opposite sides of the burnt-through foil. Close to the Al target, the optical thickness of the Al Lyγ emission increases, thus the observed shifts result from a combination of the lateral and normal plasma expansion with respect to the target surface. The detailed evaluation of the line shifts at the distances of ±25 μm from the Al foil provided the effective lateral velocity component ~1.5×10⁷ cm/s which agrees with the near-surface semispherical plasma expansion observed in interferometric measurements [5].

In zone II extending up to the distance of about 450 μm from the Al foil, the plasma propagates in a laminar flow and no large-scale plasma turbulences are visible [5]. Consequently, the distribution of the measured effective plasma parameters follows predictions of a simple 1D hydrodynamic modeling. With the increasing distance from the primary target, the intensity of the observed Al Lyγ emission gradually decreases but then it again intensifies in the zone III. The enhanced emission close to the C surface indicates deceleration, trapping and subsequent dissipation of Al ions at the secondary target.

The interpretation of the spectra emitted from the near-wall zone requires the knowledge of the environmental plasma conditions. The analysis of the spectra recorded by the spherically bent mica crystal spectrometer [7] in the zone III provided the electron temperature $T_e \approx 450–550$ eV and electron density $n_e$ within 0.5–5×10²¹ cm⁻³. The limited spatial resolution of the spectrometer (~20 μm) does not permit the more detailed determination of the plasma parameters distribution in the near-wall zone; to remedy this, a series of simulations has been performed to evaluate the time-dependent distribution of the macroscopic plasma parameters.
Interpenetrating plasmas were modeled by a combination of the 2D hydrodynamic code PALE with the 1.5 D multi-fluid code MULTIF [8]. The PALE code provided the temporally resolved 2D maps of hydrodynamic quantities. For time $t = 340$ ps after the laser pulse maximum, the simulated plasma outflow approaches closely the secondary target. From relevant 2D distribution of plasma parameters, the 1D profile corresponding to the central cylindrical axis of symmetry was extracted and used as input for the MULTIF description of the Al plasma interaction with the cold C surface, production of the counter-streaming C plasma and subsequent collision and interpenetration of both plasmas [9].

Immediately after the impact of Al jet onto C surface, the simulations indicate a fast creation of a relatively thin (~20 $\mu m$) near-wall layer of highly ionized, strongly interpenetrating Al and C ions. The resulting temporal-spatial distribution of the electron density and temperature is shown in figure 4; as discussed in more detail in section 4, the found environmental conditions are favorable for observation or charge exchange phenomena in the near-wall layer. The validity of these simulations is limited by the 1D simplification used when running the MULTIF and by neglecting the C-surface preheat due to combined effects of the scattered laser light, fast particles and shock waves. On the other hand, the simulations provide a good qualitative insight into processes occurring within the inter-target space.

4. Charge exchange signatures in spatially resolved spectra of Al Ly$\gamma$

The Al Ly$\gamma$ profiles of the near-wall-emitted spectra from figure 3 are presented in figure 5a. The mirror-symmetric line profiles display a reproducible fine structure, the development of which is more distinct in the 10-points FFT-smoothed right-hand side spectra shown in figure 5b. As the distance from the C target surface decreases to less than 10 $\mu m$, a depression surrounded by two peaks appears in the very center of the line. This is a manifestation of a significant self-absorption in agreement with the strong density gradient predicted by the plasma simulations; this depression helps to identify the center of the line. Second, with the decreasing distance the line centers shift distinctly to the red. Assuming that these shifts can entirely be ascribed to the so called plasma polarization shift [10], its evaluation in terms of quantum-mechanical impact theory would indicate steep density gradients (starting from $n_e \approx 4 \times 10^{22}$ cm$^{-3}$ observed for the distance of 5 $\mu m$ below the target up to $n_e \approx 4 \times 10^{21}$ cm$^{-3}$ in the distance of 37 $\mu m$ above the C surface). On the other hand, the mechanisms responsible for the observed line shifts may also include the effects of the ions deceleration, radiative transfer in the turbulent plasma close to the target surface, and an enhanced satellites emission, thus the above given estimates should be taken like the upper limit for the density estimates. Both these features however indicate that the plasma-wall experiments exhibit high-density effects.

The third most exotic feature consists in the appearance of prominent dips in the near red wing, whose separations from the line center show a weak dependence on the distance from the target surface, and thus on $n_e$. This feature is a quasi-molecular effect. A weak density dependence of their positions is a characteristic of the charge-exchange-caused dips (X-dips, for brevity), in distinction to the Langmuir-waves-caused dips (observed, e.g., in [11]) whose positions scale as $n_e^{-1/2}$. 

Figure 4. Temporal-spatial distribution of plasma parameters simulated by codes PALE and MULTIF.
Physically, X-dips correspond to anti-crossings of energy terms of the one-electron quasi-molecule of nuclear charges $Z_1$ and $Z_2$: anti-crossings enhance charge-exchange. For the quasi-molecule Al$^{13+}$eC$^6+$ ($Z_1 = 13$ and $Z_2 = 6$) and the energy terms of Al XIII perturbed by C$^6+$ and the terms of C VI perturbed by Al$^{13+}$, the theory predicts three anti-crossings at different inter-nuclear distances (depending only weakly on $n_e$) and thus up to three X-dips to observe. For a clear observation of a particular X-dip, the inter-ionic distance, where the corresponding anti-crossing of terms occurs, should be close to the most probable inter-ionic distance (controlled by $n_e$) [12]. Therefore it is unlikely to observe all three X-dips in the same line profile. The present experiment is favorable for observing dips 1 and 2 (dip 3, the farthest from the line center is unlikely to be observed) while in previous experiments at LULI facility [13,14], only dips 2 and 3 could be observed.

The positions of the X-dips 1 and 2 were carefully determined with respect to the shifted centers of the spectral profiles. At distances $\pm 5 \, \mu m$ from the target surface, the found positions are $3.1 \pm 0.2 \, mA$ for dip 1 and $6.6 \pm 0.2 \, mA$ for dip 2; at distances $\geq 10 \, \mu m$, dip 2 slightly shifted to $6.2 \pm 0.2 \, mA$. With respect to simulated density profiles close to the C surface, this means that only a weak dependence of the dips position on the electron density has been detected. This dependence could be supported by the theoretical analysis of charge-exchange phenomena and two complementary theoretical models.

In the first model [12,15], the dip positions were calculated analytically in the parabolic coordinates using the multipole expansion for the interaction of the hydrogenic ion Al XIII with the fully-stripped ion C VI, and without taking into account the screening by plasma electrons ($n_e=0$). We retained a large number of terms in the multipole expansion to achieve the accuracy adequate for the comparison with experiments. Using this model the predicted positions of the dips 1 and 2 are $3.1 \, mA$ and $6.7 \, mA$.

In the second model [14,16], we calculated the dip positions for the quasi-molecule numerically by diagonalizing the Hamiltonian in the elliptical coordinates, in which the problem of two Coulomb centers allows the separation of variables (simulations by the quasi-molecular code IDEFIX ). These simulations yield exact terms of the quasi-molecule Al$^{13+}$eC$^6+$ with the subsequent allowance for the plasma electron screening (depending on $n_e$). As explained below, the usage of the more realistic
quasi-molecular model IDEFIX shifted the x-dips positions slightly further to the red, while the allowance for the electron screening shifted them to the blue.

Within the quasi-molecular model and the quasi-static approximation (valid in the line wings), the Al Ly\(\gamma\) transition emitted from a dicenter plasma Al-C is controlled by the energy difference \(E_u(R) - E_l(R)\) between the upper \(u\) and lower \(l\) levels concerned (\(R\) being the inter-ionic distance). For spectral line shapes the average over \(R\) using the nearest neighbor probability distribution is required.

Let us define the center of the line when the electron screening is neglected,
\[\Delta E_0 = E_u(R \to \infty) - E_l(R \to \infty).\]
This reference is the same for the analytical model "analytic" and the quasi-molecular model without taking into consideration the electron screening "mol" (\(mol\) stands for molecular). For the quasi-molecular model with the allowance of the electron screening "mol + PPS" (\(PPS\) stands for plasma polarization shift), the line center of the line is
\[\Delta E_{0}^{PPS} = \Delta E_{u}^{PPS}(R \to \infty) - E_{l}^{PPS}(R \to \infty).\]

It is well known that the PPS is red, thus \(\Delta E_{0}^{PPS} - \Delta E_0 < 0\). Now we focus on charge exchange corresponding to the anti-crossing at the position \(R_{CE}\). This anti-crossing is responsible of an X-dip.

Let us introduce the position \(\Delta E_{CE}^{analytic}\) of the anti-crossing level with respect to the level \(l\) at the anti-crossing distance \(R_{CE}^{analytic}\) for the analytical model. In the same way \(\Delta E_{CE}^{mol}\) and \(\Delta E_{CE}^{mol+PPS}\) stand for the positions of the anti-crossing levels with respect to the level \(l\) at the distances \(R_{CE}^{mol}\) and \(R_{CE}^{mol+PPS}\), respectively, for the two versions "mol" and "mol+PPS" of the quasi-molecular code IDEFIX [14,16].

The positions of the X-dips with respect to the line center are \(\Delta E_{CE}^{analytic} - \Delta E_0\), \(\Delta E_{CE}^{mol} - \Delta E_0\), and \(\Delta E_{CE}^{mol+PPS} - \Delta E_0\) within the three models "analytic", "mol", and "mol+PPS", respectively.

When compared to the "analytic" model, the model "mol", valid for any electron density, yields a red correction \(C_1\) for the distance of the X-dip with respect to the line center
\[C_1 = \Delta E_{CE}^{mol} - \Delta E_{CE}^{analytic} < 0.\]

The model "mol+PPS" depending on \(n_e\) gives an added correction \(C_2\) with respect to the "mol" model
\[C_2 = (\Delta E_{CE}^{mol+PPS} - \Delta E_{CE}^{mol}) + (\Delta E_0 - \Delta E_0^{PPS}).\]

The second term in this sum is positive and increases with \(n_e\). The first term is negative and its absolute value is negligible with respect to the second term because it relates to small inter-ionic distance not very sensitive to the electron screening. Thus the correction \(C_2\) is always blue (\(C_2 > 0\)).

For the dip 2, the positions 7 mÅ and 6 mÅ, calculated for \(n_e = 1 \times 10^{22} \text{ cm}^{-3}\) using the models "mol", and "mol+PPS", respectively [14,16], agree with the previous discussion (\(C_1 < 0\) and \(C_2 > 0\)). The fact that the experimental X-dip 2 position 6.2±0.2 mÅ measured at distances ≥10 μm is closer to the prediction of the "mol+PPS" model (6 mÅ) than to the prediction of the "analytic" model (6.7 mÅ), is the first experimental evidence of some (weak) dependence of the X-dip positions on \(n_e\).

In the close near-wall area, i.e., at distances ±5 μm from the C-foil surface, the measured positions of both dips, 3.1±0.2 mÅ for dip 1 and 6.6±0.2 mÅ for dip 2, respectively, are close to the "analytic" values 3.1 mÅ and 6.7 mÅ, confirming that \(C_1\) increasing with \(n_e\) can cancel \(C_1\).

To conclude, the experiment performed at PALS is the first to yield with a high precision the X-dips positions in Al XIII Ly\(\gamma\) red wing. Their appearance in both mirror-symmetric parts of the spectra and their small density evolution, confirmed by the theoretical models, support the spectroscopic manifestations of charge-exchange phenomena in plasma-wall interaction.

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