Hollow Ion Spectra In Warm Dense Laser-Produced Plasma: Observation And Modeling

Anatoly Ya. FAENOV[1,2], Tatiana A. PIKUZ[1,2], Igor Yu. SKOBELEV[2], Yuji FUKUDA[1], James COLGAN[3], Joe ABDALLAH Jr.[3]

[1]*Kansai Photon Science Institute (KPSI), Japan Atomic Energy Agency (JAEA), Kizugawa-shi, Kyoto, Japan*

[2]*Joint Institute for High Temperatures, Russian Academy of Sciences, Moscow, Russia*

[3]*Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico, USA*

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X-Ray emission plasma spectra are investigated for 2 cases: i) plasma obtained under irradiation of Ar clusters by ultrashort laser pulses and ii) Mg-plasma heated by a short-wavelength long (nanosecond) laser pulse. It is demonstrated that under some experimental conditions very complicated spectra structures appear. Calculations in support of these measurements have been performed using a detailed atomic kinetics model with the ion distributions found from solution of the time-dependent rate equations. The calculations are in good agreement with the measurements and the role of hollow atoms in the resulting complicated spectra is analyzed. The signatures of hollow atom spectra can be identified in the calculations, which are qualitatively supported by the experimental measurements. It was found that the Hollow atoms structures are more clearly pronounced in the case of lower electron temperature and practically solid density plasmas.

Keywords: hollow ions, spectroscopy, cluster target, laser plasma interaction

1. Introduction

It has been shown in [1-5] that the X-Ray emission spectra of warm dense plasma contain some exotic spectral lines caused by radiative transitions in so called “hollow ions”, that are the highly charged ions with an empty inner K-shell (KK-hollow ions) or with vacancies in K- and L-shells (KL-hollow ions).

Such exotic spectra are produced usually by interaction of a high-contrast femtosecond laser pulses with solids or clusters. Another way is to use nanosecond short-wavelength laser pulses to interact with the solid. In the present work both methods were used to investigate the role of the highly charged hollow ions in the X-Ray emission plasma spectra.

2. Experimental set up and results.

The Ar cluster experiments were performed with the JAEA (Kyoto, Japan) Ti: sapphire laser system based on the technique of chirped pulse amplification, which was designed to generate 20 fs pulses and is capable of producing a focused intensity up to 10^{20} W/cm^2 [6]. In this study the amplified pulses were compressed to 30 fs by a vacuum pulse compressor yielding a maximum pulse energy of 360 mJ [7-9]. Two types of experiments were performed. In the first set of experiments, only one double Pockels cell was used after the regenerative amplifier, which allowed us to reduce the laser prepulse by up to $5x10^{-4}$ compared with the main laser pulse. In addition, experiments were performed where after the regenerative amplifier, the laser pulse is passed through two double Pockels cells, which allows the possibility of reducing the laser prepulse even further. The final contrast ratio between the prepulse and main laser pulse, which precedes it by 10 ns, was better than $4.6x10^{-6}$. The experiments were carried out with various laser energies (49 - 115 mJ), and in a wide range of laser pulse durations from 30 up to 1000 fs, which corresponds to laser intensities of about $6x10^{16}$-$2x10^{18}$ W/cm^2.

A cluster-gas target was produced by expanding 60-bar Ar gas into a vacuum using a pulsed valve connected with a special nozzle consisting of three truncated cones with different apex angles. The nozzle has the capability to produce Ar clusters with an average diameter of around 1.5 μm [10]. To minimize the tendency of the laser prepulse to destroy the clusters, we purposely used in our experiments very big clusters, since the rate of cluster decay is primarily determined by the number of atoms in the cluster [11,12]. In such cases, the micron-sized clusters can significantly reduce the cluster's sensitivity to the laser prepulse, preclude low-density preplasma formation, and thus guarantee the direct interaction between the high-density cluster and the main fs pulse. The existence of a dense core region of the clusters was confirmed *via* analysis of the X-ray emissions from the Ar clusters in separate experiments [7-9, 12-14]. It was demonstrated in these studies that, if the laser prepulse was too big, clusters were destroyed, no frozen part of cluster remained, and no X-rays were observed. So, although the prepulse intensity was strong enough to ionize the Ar atoms in the clusters in our

author’s e-mail: faenov.anatoly@jaea.go.jp

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experiments, the ionization and the expansion occur at the cluster peripheral region, and the cluster core can be treated as frozen at all prepulse intensities of about 10^{11} -10^{14} W/cm^2. Indeed, in the case of higher prepulses, the remaining part of the frozen cluster was smaller compared with a higher laser contrast.

The spatially resolved X-ray spectra were measured using focusing spectrometers with spatial resolution [15]. This spectrometer is equipped with a spherically bent mica crystal (R = 150 mm) and a vacuum compatible X-ray charge coupled device camera (DX420-BN, ANDOR). This instrument recorded He-like spectra of Ar including the associated satellite lines.

For Mg spectra we used the data obtained in previous experiments [5] carried out using a low intensity XeCl excimer laser facility with an energy of 2 J and a pulse duration of 12 ns (laser intensity on the target is 4 x 10^{12} W/cm^2). Using FSSR spectrometers [15], have allowed us to measure low intensity X-ray spectra simultaneously with high spectral and spatial resolution. These spectra show unusual quasi-continuous line emission produced by the laser irradiation of Mg targets. Such emission occurs at wavelengths just shorter than the He-like resonance line at 9.17 Å. Note that the observed unusual quasi-continuous line emission is lying between the resonance Ly_α line of the H-like ion and resonance lines He_α from the He-like ion, and that they are strongest near the target surface. Typical laser produced plasmas emit satellite lines at wavelengths just longer than the He_α line (see for example, [4,8]). These types of transitions have been studied in detail and relative intensities of spectral lines can often be predicted by collisional-radiative models for a prescribed plasma density and temperature. The satellite lines that are commonly observed in these plasmas are generally due to $1s2pnl$-$1s^2nl$ transitions in the Li-like ion. Lines occurring at wavelengths just between the Ly_α and He_α lines belonging to transitions in He-like ions are rarely observed.

It is clear that the spectra observed are caused by very non-equilibrium conditions because of its unusual characteristics. LTE and typical collisional-radiative modeling would not normally predict such intense spectral structures relative to the resonance line in the wavelength region in question. The first requirement is to identify the emitting species. An initial guess for line emission in this region could be n = 3 to n = 1 transitions in moderately ionized magnesium. Another possibility is n = 2 to n = 1 transitions in KK-hollow ions. Previous theoretical analysis [4,5] confirmed that such complicated structure in the spectral range 8.5-9.1 Å could belong to spectra from such transitions. However, those calculations were incomplete, and questions remain about the mechanisms of hollow atom formation in such a laser-produced plasma and additional modeling is necessary to address such issues.

3. Spectra simulations.

The atomic data used in this model were generated from the Los Alamos suite of computer codes developed over many years to calculate atomic structure and atomic scattering quantities. The CATS code [16], which is an adaptation of Cowan's atomic structure codes [17], was used to calculate wave functions, energy levels, oscillator strengths, and plane-wave Born collision strengths for all ionization stages of Mg and Ar. CATS was used to generate data in the configuration-average approximation. Electron-impact ionization, photoionization, and autoionization cross sections were calculated using the multi-purpose ionization GIPPER code [18]. The latter two processes were calculated explicitly with distorted-wave continuum functions while the electron-impact ionization calculations used scaled hydrogenic cross sections that were designed to reproduce distorted-wave calculations. Collisional de-excitation, three-body, radiative, and dielectronic recombination rates were obtained from detailed balance.

The atomic model used for Mg study included over 1600 configurations among all the ion stages, where more configurations were used for the near-neutral stages, ranging from 15 configurations for the H-like ion stage to over 270 configurations for the neutral atom. Configurations were included with one electron promoted from the K or L shells into the n = 3, 4, or 5 shells. Also included were two-electron promotions, where either both electrons were promoted from the K-shell (KK-hollow ions), or one electron promoted from the K- and one from the L-shell (KL-hollow ions). Finally, we also included exotic configurations where one electron was promoted from the K-shell, two electron were promoted from the L-shell, and all three excited electrons populated the M-shell. Many of these configurations can easily describe a large number of fine-structure levels, which can make a full calculation at the fine-structure level prohibitive. The MUTA (mixed UTA) approach, which retains the strongest lines within a given transition array and the weak lines with a single functional form [19,20], was employed to allow a reasonable calculation while retaining the accurate spectral description required for this study.

The solution of the rate equations for the Mg plasma was carried out using the Los Alamos code ATOMIC, which has been described previously [21]. The MUTA capability allows us to obtain ionic populations at the configuration-average level while retaining the ability to provide a spectral description comparable in accuracy to a detailed (fine-structure) approach, where all lines are included explicitly in the calculation. This development was crucial since it enabled us to include kinetics for configurations where two or more electrons were promoted from the K and L shells within a calculation that could be completed in a timely manner (compared with a full fine-structure kinetics calculation). Since the plasma was formed from a very long laser pulse, the steady-state rate equations were solved for various electron temperatures and densities. A small fraction of the electron density (typically 0.1% or less) was at a temperature of 2 keV to simulate a 'hot electron' tail.

In the case of Ar plasma the effects of hollow atoms on the observed emission spectra was found to be difficult to isolate, due to the close proximity of strong lines from transitions within the F- and O-like ion stages of Ar, as well as the nearby K_α lines (see for more details [22]. Detailed theoretical calculations made using the ATOMIC plasma kinetics code, which include configurations of KL-hollow ions, have allowed exploration of these emission lines.

4. Discussions and conclusion.

It is found that in the case of Ar spectra the closest agreement with the experimental measurements come from a calculation where the electron temperature is 50 eV, with 3% of hot electrons at 5 keV, and an atom density of 10^{22} cm^{-3}. The dominant features in the region 4.17-4.18 Å still arise from transitions in the F-like ions, and in the region 4.14-4.16 Å arise from transitions in the O-like ions, but some spectra solely due to hollow atom transitions are also identified. It can be seen from Fig. 1 that the role of the hollow ion spectral lines is very important for time moments when plasma is non-stationary. From this experimental study and theoretical modeling of the measured X-ray spectra of Ar, we could see that the populations of hollow ions are increased in the case of laser interaction with dense cold targets, compared with the cases when the same laser interacts with less dense preplasma. Analogous behavior of hollow atom production was previously observed for fs laser interaction with solid matter (see for example, [2,3,23]).

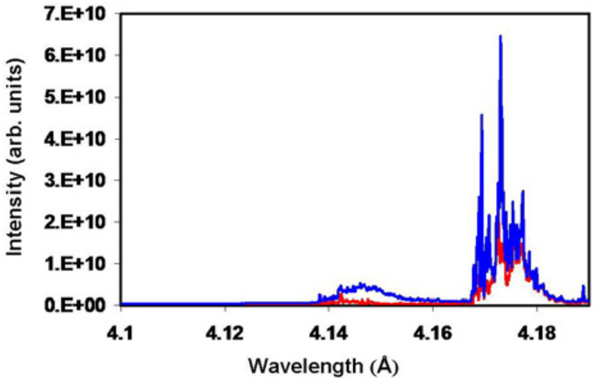

Fig. 1. ATOMIC calculations of the Ar spectra for time 600 fs at an electron temperature of 100 eV and at an atom density of 10^{22} cm^{-3}. Red line – calculation without hollow ions, blue line – all configurations are included.

It should be noted that the Ar plasma with $T_e \sim 50$ eV, with an average ionization of 6, and and ion density $N_i \sim 10^{22}$ cm^{-3} is weakly coupled. For these plasma parameters the electron-electron coupling factor $\Gamma_{ee} \sim 0.11$ and the electron-ion coupling factor $\Gamma_{ie} \sim 0.37$. Generally speaking, strongly-coupled plasmas can modify the probabilities for various collisional processes, for example, the three-body recombination rates. As has been shown in [24], for a strongly-coupled plasma, the three-body recombination rate can be decreased by a factor of $\Gamma_{ie}{}^{5/2}$ compared with the rate for an isolated atom. In our experiments $\Gamma_{ie} < 1$ and this effect will not be large. So, our use of the isolated-atom approximation for the description of the collisional processes within the plasma is justified. However, the interaction of super contrast femtosecond laser pulses with condensed media may lead to the creation of an even more dense and, consequently, more strongly-coupled plasma. In these cases the influence of plasma effects on the elementary atomic processes should be taken into account.

In this paper we have used the latest developments in atomic kinetics modeling [19], coupled with the use of a comprehensive configuration dataset, to model complex spectra created from the interaction of a nanosecond XeCl laser on a Mg target. The experimental spectral lines which arise from 1s-3l transitions, 1s-4l transitions, and transitions involving hollow ions are, on the whole, quite well reproduced by the calculations (see Fig. 2). It should be emphasized that the traditional kinetic model without 1-3, 1-4 and the hollow ion transitions gives no spectral lines in the region 8.55 – 9.15 Å. We find a reasonable reproduction of the experimental spectra by assuming small parts of the plasma are relatively cool and dense, and larger parts of the plasma are hot and less dense (see for details [25]). The atomic model included over 1600 configurations and tens thousands radiative transitions. Therefore it is not possible to identify each spectral components in the spectra observed. Note only, that the hollow ion transitions lie mainly near 8.55 and 9.05 Å, and the emission in the region 8.6 – 9 Å are caused mainly by C-, N-, O- and F-like Mg ions.

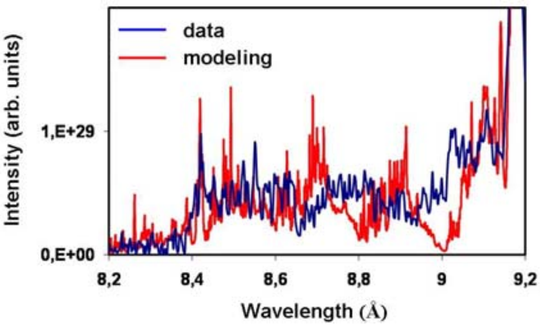

Fig. 2. Blue line: Mg spectra from the experimental measurements of [5]; Red line: Mg spectra from mixed ATOMIC calculation (5% of the spectra from the 10 eV and 20 eV calculations for $N_e = 10^{23}$ cm^{-3}, 10% of the spectra from the 30 eV, 6x10^{22} cm^{-3} calculation, 40% of the spectra from the 50 eV, 6x10^{22} cm^{-3} calculation and 40% of the spectra from 200 eV, 10^{21} cm^{-3} calculation). All calculations include 0.05% of hot electrons at a temperature of 2 keV..

5. Acknowledgments

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