Observation and modelling of hollow multicharged ion x-ray spectra radiated by laser produced plasma

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Abstract. The role of the highly charged hollow ions in X-Ray emission plasma spectra is investigated for 2 cases: 1) plasma obtained under irradiation of Ar clusters by ultrashort laser pulses and 2) Mg-plasma heated by a short-wavelength long (nanosecond) laser pulse. Experimental measurements are presented. 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.

1. Introduction

Plasmas of multicharged ions (Z =10 – 20) with electron temperatures 10 – 100 eV and electron densities $10^{22} – 10^{23}$ cm$^{-3}$ are produced usually by interaction of high-contrast femtosecond laser pulses with solids or clusters. The same plasma parameters are also realized when nanosecond short-wavelength laser pulses (KrF or XeCl or 3$^{rd}$, 4$^{th}$ harmonics of Nd:glass lasers) interact with solids. Highly charged ions in a plasma with such parameters are already weakly- coupled and it is very interesting to investigate their radiation properties. It has been shown in [1-7] that the X-Ray emission spectra of such plasmas 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).

In the present work the role of highly charged hollow ions in X-Ray emission plasma spectra is investigated for 2 cases: 1) plasma obtained under irradiation of Ar clusters by ultrashort laser pulses and 2) Mg-plasma heated by a short-wavelength long (nanosecond) laser pulse.

2. The role of hollow ions in the spectrum of an ultrashort-pulse-laser-driven Ar cluster target

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$^2$ [8]. In this study the amplified pulses were compressed to 30 fs by a vacuum pulse compressor yielding maximum pulse energy of 360 mJ. 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 $5 \times 10^{-4}$ 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 $4.6 \times 10^{-6}$. 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 $6 \times 10^{16}$ up to $2 \times 10^{18}$ 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. 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 [9,10]. In such cases, the use of micron-sized clusters can significantly reduce the cluster's sensitivity to the laser prepulse, preclude low-density preplasma formation, and thus guarantee direct interaction between the high-density cluster and the main fs pulse.

The spatially resolved X-ray spectra were measured using focusing spectrometers with spatial resolution [11]. This spectrometer recorded He-like spectra of Ar including the associated satellite lines and neutral Kα.

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 Kα lines. Detailed theoretical calculations made using the ATOMIC plasma kinetics code [12], which include configurations of KL-hollow ions, have allowed exploration of these emission lines.

It is found (see for more details [13]) that the closest agreement with the experimental measurements comes from a calculation where the electron temperature is 50 eV, with 3% of these electrons at 5 keV, and an atom density of $10^{22}$/cm$^3$ (see Fig. 1). Transitions in the hollow ions give significant (about 50%) contribution only to the strong peak at 4.17 Å. The smaller peak is formed by transitions in F- and O-like ions.

![Fig. 1. Comparison of theory (1) and experiment (2) for Ar spectra.](image1)

**3. Mixed Unresolved Transition Array (MUTA) calculations of a Mg hollow ion spectrum**

Previous experiments [6] 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 $4 \times 10^{12}$ W/cm$^2$) using FSSR spectrometers [11], have allowed us to measure low intensity X-ray spectra simultaneously with high spectral and spatial resolution (see Fig. 2). These spectra (especially near the target) show unusual quasi-continuous line emission produced by the laser irradiation of Mg targets. This 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\(\alpha\) line of the H-like ion and resonance lines He\(\alpha\) 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\(\alpha\) line. 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\(^2\)nl transitions in the Li-like ion. Lines occurring at wavelengths just between the Ly\(\alpha\) and He\(\alpha\) lines belonging to transitions from autoionizing states in He-like ions are rarely observed.

It is clear that the spectra observed are caused by very non-equilibrium conditions because of their unusual characteristics. Local thermodynamic equilibrium (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 [6] confirmed that such complicated structure in the spectral range 8.5-9.1 \(\text{Å}\) 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.

![Fig. 2. Traces of spatially resolved X-ray emission spectra of Mg plasma in the spectral range of 8.4 - 9.3 Å heated by a low intensity XeCl nanosecond laser. The strongest spectral lines belong to resonance Ly\(\alpha\) line of Mg XII and the resonance (R) and intercombination (I) lines of Mg XI as marked.](image)

The solution of the rate equations for the Mg plasma was carried out using the Los Alamos code ATOMIC, which has been described previously [12]. 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 this paper we have used the latest developments in atomic kinetics modeling [14], 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. 3). It should be emphasized that the traditional kinetic model without KK- and KL-hollow ion transitions gives no spectral lines in the region 8.55 – 9.15 Å. Inclusion of a small fraction of hot electrons is necessary to reproduce the experimental spectra, although we must also make the assumption of quite large temperature and density gradients within the bulk electron distribution. 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 [15]). Even at the high densities that we have assumed in our plasma, the line broadening of the satellite lines on which this study is focused is fairly small. We look forward to applying similar calculations to other experimental studies of hollow ion spectral lines, since the investigation of these exotic configurations is increasingly becoming accessible using modern laser technology and X-ray spectrometers with high spectral and spatial resolution.

Fig. 3. (1) - Mg spectra from mixed ATOMIC calculation. (2) - Mg spectra from the experimental measurements of [6]

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