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Spectra of multiply charged hollow ions in the plasma produced by a short-wavelength nanosecond laser

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Abstract. Complex spectral structures located between the resonance lines of H- and He-like MgXII and MgXI ions were recorded in experiments on plasma heating by the radiation of a low-power short-wavelength excimer XeCl laser (12-ns pulses with an energy of 2 J). The above spectral structures were shown to arise from transitions in the so-called hollow multicharged ions, i.e., in ions with an empty 1s-shell, which were previously observed in laser produced plasmas only with ultrahigh-power femto- and picosecond laser facilities having extremely high-contrast laser pulses.

1. Introduction

Complex spectral structures located in the region between the resonance lines of H- and He-like ions were recorded in emission spectra of a plasma heated by laser radiation at the high-power NIKE laser facility (Naval Research Laboratory, USA) [1].

Earlier, only satellites of the resonance line of the H-like ion arising from the radiative decay of doubly excited states of He-like ions (see, e.g., review [2]) were recorded in this spectral range. The satellite transitions, which have been studied in considerable detail both experimentally and theoretically, cannot explain the observed spectra.

Here, we study the transitions in so-called hollow ions, i.e., ions with an empty $1s$ shell, which were observed in laser-produced plasmas only for high ionisation degree employing ultrahigh-power femto- and picosecond laser facilities having extremely high energy contrast ratios (see, e.g., Refs [3 – 6]). At the same time, hollow ions of low ionisation degree were produced previously only in the interaction of high-intensity relativistic ion beams with solids [7].

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The transitions in multiply charged hollow ions can be investigated employing a relatively simple plasma source driven by a laser pulse of nanosecond duration and moderate intensity that should have a sufficiently short wavelength.

2. Experimental

In this work, plasma heating was accomplished using a 0.308-μm excimer XeCl laser [8]. The laser pulse energy was 2 J, and the pulse length was 12 ns. The laser was operated at a repetition rate of 10 Hz. The radiation was focused on the surface of a solid magnesium target to a spot 70 μm in diameter, so that the laser intensity was rather low (4×10^{14} W cm^{-2}).

The x-ray plasma radiation was recorded using a FSSR-2M spectrograph with a spherically bent mica crystal [9, 10]. The spectrograph provided a resolving power $\lambda/\delta\lambda$ of over ~ 5000 simultaneously with a one-dimensional spatial resolution of $\delta x \approx 30$ μm in the direction of plasma expansion (i.e., along the target normal). Fig. 1a gives an example of the emission spectrum of the magnesium plasma in

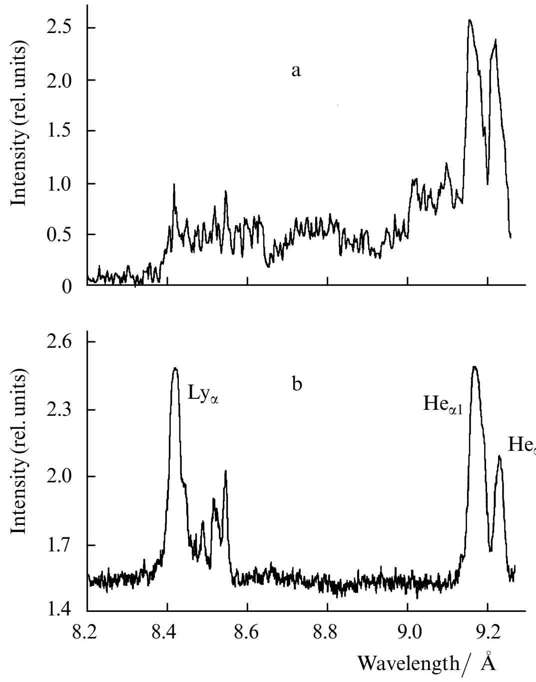

Figure 1. Emission spectra of a magnesium plasma in the 8.4 – 9.3 Å range heated by a XeCl (a) and a Nd (b) laser.

the 8.2 – 9.3 Å range obtained in this work. For comparison, Fig. 1b shows the spectrum of a magnesium plasma heated by a neodymium laser with a pulse length of 1 ns at an intensity of 10^{14} W cm^{-2} [2].

Note that spectra similar to that shown in Fig. 1b were previously observed not only in a laser-produced plasma, but in exploding wire and pinch plasmas and in the solar corona as well (see, e.g., the review [2] and references therein). For the sake of brevity, in the subsequent discussion the spectra of this kind will be referred to as conventional. One can see from Fig. 1 that conventional emission spectra of the magnesium plasma in the 8.2 – 9.3 Å range exhibit the resonance and intercombination lines of He-like MgXI (the $\mathrm{He}_{\alpha1}$ and $\mathrm{He}_{\alpha2}$ lines), the resonance line of H-like MgXII ions (the Ly_α line), and its dielectronic satellites arising from the radiative decay of the autoionising states of He-like MgXI ions (a group of lines in the 8.44 – 8.56 Å range). One can see from Fig. 1 that the spectrum in Fig. 1a differs from the conventional one primarily in that it contains additional quasi-continuous structures residing in the 8.4 – 8.65, 8.67 – 8.92, and 8.94 – 9.15 Å ranges.

3. Experimental results and discussion

3.1. Qualitative analysis of the observed spectra

Consider what spectral transitions can fall in the range between the Ly_α line of MgXII ions and the He_α line of MgXI. In the first place, these may be the lines related to the electronic transitions from a state with the principal quantum number $n' = 2$ to the states with the principal quantum number $n = 1$, i.e., transitions of the type

$$(1s)^{n_1}(2l)^{n_2}(3l)^{n_3}(4l)^{n_4}\ldots - (1s)^{n_1+1}(2l)^{n_2-1}(3l)^{n_3}(4l)^{n_4}\ldots, \quad (1)$$

where $n_1 = 0$ or 1 and $n_2 \geqslant 1$. Note that, for $n_3 = n_4 = \ldots 0$ and $n_2 = 1$, we have the resonance line of the H-like ion (for $n_1 = 0$) and the resonance line of the He-like ion (for $n_1 = 1$) (Fig. 2). The transitions with $n_1 = 1$ and $n_2 + n_3 + \ldots > 1$ are the well-studied dielectronic satellites of the resonance line of the He-like ion, and their wavelengths exceed, as a rule, the wavelength of the resonance line itself. These wavelengths may only increase with increasing occupation numbers $(n_2 + n_3 + \ldots)$. Note that some of the dielectronic satellites may be shorter in wavelength than the resonance line owing to the interaction between electron configurations. However, they are exceptions to the general rule, being small in number and residing only in the immediate vicinity of the resonance line.

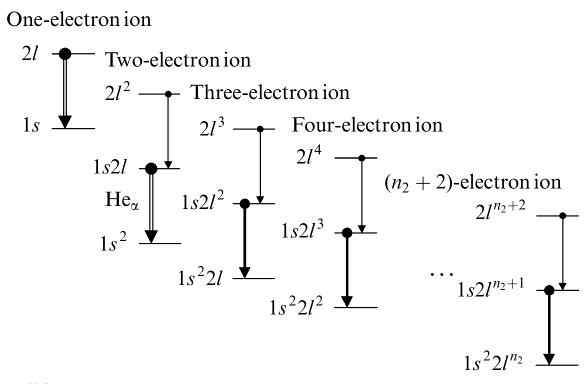

Figure 2. Transitions in multiply charged ions accompanied by changes in the principal quantum number of the optical electron from $n' = 2$ to $n = 1$: resonance lines (double arrows), dielectronic satellites (bold arrows), and transitions in hollow ions (thin arrows).

Therefore, among the transitions of type (1), only transitions with $n_1 = 0$, i.e., the transitions in hollow ions

$$(2l)^{n_2}(3l)^{n_3}(4l)^{n_4}\ldots - (1s)(2l)^{n_2-1}(3l)^{n_3}(4l)^{n_4}\ldots \quad (2)$$

may lie in the range between the Ly_α and He_α lines (and also at a considerable distance from them).

This brings up the question: Which of transitions (2) do reside in the range in question? Strictly speaking, the answer to this question should be derived from detailed atomic structure calculations. However, a simple estimate can be made starting from the presently available data on the satellites of the resonance lines of one- and two-electron ions. We make these estimates for magnesium ions, for it was precisely these ions that were investigated in the experiments conducted.

The resonance Ly_α doublet of the H-like MgXII ion has an average wavelength of 8.421 Å, and the resonance He_α line of MgXI, a wavelength of 9.1697 Å. The calculations for the dielectronic satellites of these lines performed in Refs [11,12] show (and this is in excellent agreement with experimental results) that the addition of one electron to the $2l$-shell causes the wavelength of the $2p - 1s$ transition of the optical electron to shift by 0.13 – 0.25 Å owing to a partial screening of the Coulomb field of the nucleus.

The screening of the nucleus by an electron added to the $3l$-shell is significantly weaker, and the wavelengths shift by 0.02 – 0.04 Å. For a $4l$ electron, the shift is still smaller, being 0.01 – 0.02 Å. This means that a $2l$ electron reduces the effective charge of the atomic core by 0.092, a $3l$ electron by 0.02, and a $4l$ electron by 0.007. (Naturally, the case in point is the effective charge 'sensed' by the optical $2p$ electron; for higher-excited optical electrons, the screening parameter is larger.) Hence, it follows that the $(2l)^{n_2} - (1s)(2l)^{n_2-1}$ transitions for $2 \leqslant n_2 \leqslant 6$, i.e., the transitions in hollow He-, Li-, Be-, B-, and C-like ions, are likely to lie in the 8.42 – 9.17 Å range under consideration. Since the shifts induced by higher orbitals are significantly smaller, the $(2l)^{n_2}(3l)^{n_3}(4l)^{n_4}\ldots - (1s)(2l)^{n_2-1}(3l)^{n_3}(4l)^{n_4}\ldots$ transitions in hollow ions with a larger number of electrons will reside in the same range.

From the above estimates, it follows that one more class of spectral lines may fall in the wavelength range under consideration, namely, the lines arising from electronic transitions from a state with the principal quantum number $n' = 3$ to the states with the principal number $n = 1$. Indeed, the wavelength of the $1s3p - 1s^2$ transition in He-like MgXI is 7.8508 Å. Hence, upon the addition of four or more electrons to the $2l$-shell, the net shift will be large enough for the transitions to fall in the spectral range of interest.

Adding several electrons more to the higher excited ($3l$, $4l, \ldots$) shells does not change the situation, and we conclude that the 8.42 – 9.17 Å range should harbour transitions of the type

$$(1s)(2l)^{n_2}(3l)^{n_3}(4l)^{n_4}\ldots - (1s)^2(2l)^{n_2}(3l)^{n_3-1}(4l)^{n_4}\ldots \quad (3)$$

with $n_2 \geqslant 5$. Note that these transitions are not transitions in hollow ions, and the lines arising from these transitions are dielectronic satellites to the He_β line of the He-like ion, which result from the radiative decay of the autoionising states in relatively low-ionised N-, O-, F-, Ne-, Na-, and Mg-like magnesium ions. These satellite lines have never been observed.

The point is that, for these satellites to be excited efficiently, on the one hand, the plasma temperature should be low enough not to exceed several tens of electron-volts (otherwise, the ions with such a low ionisation degree would be absent in the plasma), and on the other, kiloelectron-volt electrons should be present to excite the satellite transitions. In a laser produced plasma, these requirements are very hard to meet simultaneously. The lines of this type would be observable under heating of solid targets by high-power electron beams. However, as far as we know, these experiments have never been staged.

Similarly, satellites to the He_{γ}, He_{δ}, etc. lines may also find themselves in the above range for the greatest possible number of screening electrons (neutral and singly or doubly ionised magnesium). However, these lines would be even lower in intensity and still harder to record.

3.2. Numerical simulation of the emission spectra

The above qualitative estimates are confirmed by numerical calculations of the atomic structure of the magnesium ion performed in this work. For a preliminary estimate of the intensities and the positions of different spectral transitions, we used the CATS code, which allows the emission spectra of ions of different ionisation degree to be calculated in the framework of the model of an 'average atomic configuration' [13]. All possible electron configurations constructed of 1*s*, 2*s*, 2*p*, 3*s*, 3*p*, and 3*d* orbitals for ions from MgI to MgXII were taken into account. The emission spectra were calculated in the approximation of local thermodynamic equilibrium (LTE). The results obtained for two plasma temperatures are presented in Figs 3 and 4. Note that the ground state population densities of ions of different ionisation degrees were taken to be equal in these calculations; i.e., the ionisation balance was not calculated.

Fig 3 displays the results of calculations for a relatively low plasma temperature T_e (100 eV). In this case, the emission spectrum, constructed assuming a LTE, is dominated by the satellite structures of type (3) since the population densities of the states of hollow ions are very small. With increasing temperature, the situation changes and transitions of type (2) in hollow ions come to dominate the spectrum (see Fig. 4 constructed for $T_e = 1000$ eV). One can see from Figs 3 and 4 that for each ionisation degree, the emission spectrum exhibits several rather broad bands which differ by combinations of the occupation numbers with retention of their sum.

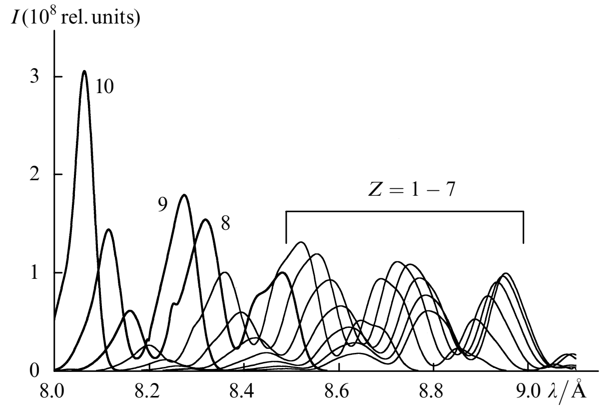

Figure 3. Emission spectrum of the $n' = 3 - n = 1$ transitions in magnesium ions [transitions of type (3)] calculated for $T_e = 100$ eV using the approximation of an average atomic configuration. The numbers at the curves denote the ionic spectroscopic symbols.

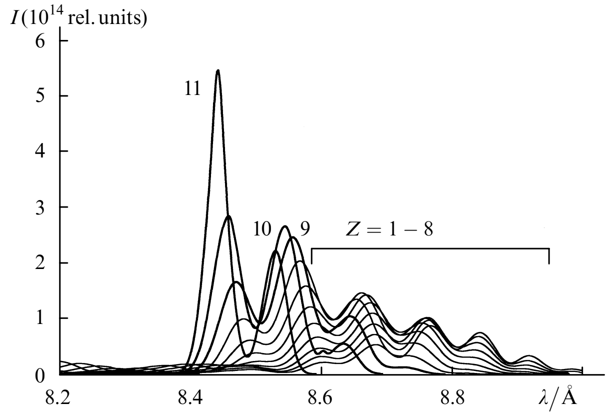

Figure 4. Emission spectrum of the $n' = 3 - n = 1$ transitions in hollow magnesium ions [transitions of type (2)] calculated for $T_e = 1000$ eV using the approximation of an average atomic configuration. The numbers at the curves denote the ionic spectroscopic symbols.

The most significant difference between Fig. 3 and 4 is that the transitions in hollow ions are grouped in the wavelength range above 8.4 Å, and when these transitions dominate, the observed spectrum would have an abrupt edge at a wavelength of 8.4 Å. When the satellites are dominant, no such sharp edge is present and the spectral peaks cover the range from 8 to 9 Å fairly uniformly (see Fig. 3). Since the observed spectra clearly exhibit a sharp edge near 8.4 Å (see Fig. 1), this circumstance alone suggests that the observed structures arise from transitions in multiply charged hollow magnesium ions. The fact that the spectrum observed by us is related to the transitions in hollow ions rather than to the satellite structures of low-ionised ions is also confirmed by the following.

As noted above, a beam of fast electrons is required to excite such structures in a cool plasma. The presence of a fast electron beam should give rise to the production of fast ions, i.e., ions with energies well above the thermal one. Our earlier x-ray spectroscopic investigations showed that the plasma production at the HERCULES facility is not accompanied by the generation of fast ions, and, hence, the occurrence of a fast electron beam is unlikely [14].

The average atomic configuration model used in the construction of Figs 3 and 4 allows us to estimate only the position of the centre of gravity of the envelope of a group of lines emitted upon the radiative decay of one or other configuration but not its shape. In this connection, extensive calculations of atomic characteristics were performed here using the codes that permit determining the wavelength and the probability of each individual spectral transition out of the collection involved. Treated most fully was the 8.4 – 8.7 Å range which harbours, as one can see from Fig. 4, the spectral transitions of hollow ions of the highest ionisation degrees.

Note that the least excited atomic configurations in plasmas are, as a rule, the most populated. The only exceptions to this general rule are, in fact, a dense recombining plasma (in this case, the recombination flux goes through highly excited configurations, thereby effecting their efficient population) and a plasma in which resonance processes occur (charge exchange, photoexcitation or photoionisation by an external narrow-band radiation), which result in the selective population of specific configurations.

That is why of all the possible electron configurations we will consider the least excited configurations having the lowest occupation numbers n_i. Specifically, we will restrict ourselves to the following sets of occupation numbers:

(1) for He-like ions ($\Sigma n_i = 2$):
$n_2 = 2$,
$n_2 = 1$, $n_3 = 1$,
$n_2 = 1$, $n_4 = 1$;

(2) for Li-like ions ($\Sigma n_i = 3$):
$n_2 = 3$,
$n_2 = 2$, $n_3 = 1$,
$n_2 = 2$, $n_4 = 1$,
$n_2 = 2$ $n_5 = 1$,
$n_2 = 1$, $n_3 = 2$,
$n_2 = 1$, $n_3 = 1$, $n_4 = 1$;

(3) for Be-like ions ($\Sigma n_i = 4$):
$n_2 = 4$,
$n_2 = 3$, $n_3 = 1$,
$n_2 = 3$, $n_4 = 1$,
$n_2 = 3$, $n_5 = 1$,
$n_2 = 2$, $n_3 = 2$,
$n_2 = 2$, $n_3 = 1$, $n_4 = 1$,
$n_2 = 1$, $n_3 = 3$;

(4) for B-like ions ($\Sigma n_i = 5$):
$n_2 = 5$,
$n_2 = 4$, $n_3 = 1$,
$n_2 = 3$, $n_4 = 2$,
$n_2 = 2$, $n_5 = 3$,
$n_2 = 1$, $n_3 = 4$.

The wavelengths and the oscillator strengths for all transitions from the above configurations were calculated in the approximation of intermediate coupling with the inclusion of the configuration interaction.

For each set of occupation numbers, we have a tremendous (up to several thousand) number of closely spaced lines, which make up the so-called unresolved transition array. (The only exception is the case of two-electron configurations, where the lines are moderate in number and form several well-resolved spectral peaks.) When the plasma density is not too low (in our experiments, the plasma density near the target was 10^{22} cm^{-3}), an LTE is realised within the levels of one configuration and the emission plasma spectrum related to this configuration is described by the sum

$$S(\lambda, n_1, n_2, n_3, \ldots) = \sum_{i,k} g_i A_{ik} f(\lambda - \lambda_{ik}) \exp\left(-\frac{E_i}{kT_e}\right), \quad (4)$$

where g_i and E_i are the statistical weight and the energy of the level; A_{ik} is the radiative transition probability; and $f(\lambda - \lambda_{ik})$ is the profile of an individual transition. Sum (4) is taken over all the levels of a given configuration and represents the spectral function of the configuration. The integral emission $S(\lambda)$ of an optically thin plasma is determined by the following expression:

$$S(\lambda) = \sum_{n_1, n_2, \ldots} S(\lambda, n_1, n_2, \ldots) \frac{N(n_1, n_2, \ldots)}{g(n_1, n_2, \ldots)}, \quad (5)$$

where $N(n_1, n_2, \ldots)$ and $g(n_1, n_2, \ldots)$ are the population density and the statistical weight of the corresponding configuration, respectively.

Sum (5) can be conveniently split into parts corresponding to configurations with a fixed total number of electrons $M = \Sigma n_i$. If, in doing this, we assume that LTE is also realised for all isoelectronic configurations, we can calculate the integral spectral function for each ion. These functions will depend on the plasma temperature (which will determine the relative population densities of different isoelectronic configurations) and the transition profile $f(\lambda - \lambda_{ik})$. The results of calculations of these spectral functions for two-, three-, four-, and five-electron atoms, which were performed for Gaussian profiles of the width 0.004 Å, are given in Figs 5 – 8. They reveal the following general features:

– For each isoelectronic sequence, the transitions in hollow ions are confined to the wavelength range from $\lambda(\mathrm{Ly}_\alpha)$ to some $\lambda_{\max}(M)$, which depends on the number M of electrons in the ion;

– The wavelength $\lambda_{\max}(M)$ is an increasing function of M;

– For a low temperature, the spectral functions are localised near the wavelength $\lambda_{\max}(M)$. As the temperature increases, they spread over the entire range from $\lambda(\mathrm{Ly}_\alpha)$ to $\lambda_{\max}(M)$.

This behaviour of the spectral functions is qualitatively quite natural. Indeed, only the least excited hollow-ion configurations of the $(2l)^M$ type are fairly strongly populated at a low temperature. For these configurations, the wavelength shift is a maximum and, as already noted, increases with M. With an increase in temperature, the $(2l)^{n_2}(3l)^{n_3}\ldots$-type configurations, for which the effect of screening is not so strong, increase in importance and the spectral functions begin to shift to the blue.

It is interesting to compare the relative positions of the spectral functions for ions with a different number of electrons. This comparison is shown in Figs 9 and 10 for two plasma temperatures. One can easily see that the spectral functions of different ions overlap despite a systematic shift to the red with increasing M. Note that narrow peaks of the spectral functions form, as a rule, owing to an accidental coincidence of the wavelengths of a very large number of spectral lines rather than due to one or several strong transitions.

It is hard to attain a very high accuracy of calculations of the atomic characteristics for complex atomic systems with a large number of open shells. That is why the true spectral functions would most likely be devoid of such clearly defined peaks and would more evenly fill the corresponding spectral region. Unfortunately, to date, there is no way to *a priori* determine the accuracy of wavelength calculations for hollow ions with three or more electrons.

One can see from Figs 5 – 10 that the maxima of the spectral functions corresponding to different electron configurations are of the same order of magnitude. This implies that, if the densities of ions of different isoelectronic sequences in the plasma do not differ greatly (i.e., are of the same order of magnitude), all the configurations considered above will make an appreciable contribution to the integral emission plasma spectrum, which will be of a complex quasi-continuous nature.

As an example, Fig. 11b shows a spectrum calculated for $T_e = 100$ eV and the following relative populations of the $(2l)^{n_2}$ configurations: $N[(2l)^2]/g[(2l)^2] : N[(2l)^3]/g[(2l)^3] : N[(2l)^4]/g[(2l)^4] : N[(2l)^5]/g[(2l)^5] = 4:1:1:1$. One can see that the ionic configurations with 2 – 5 electrons considered

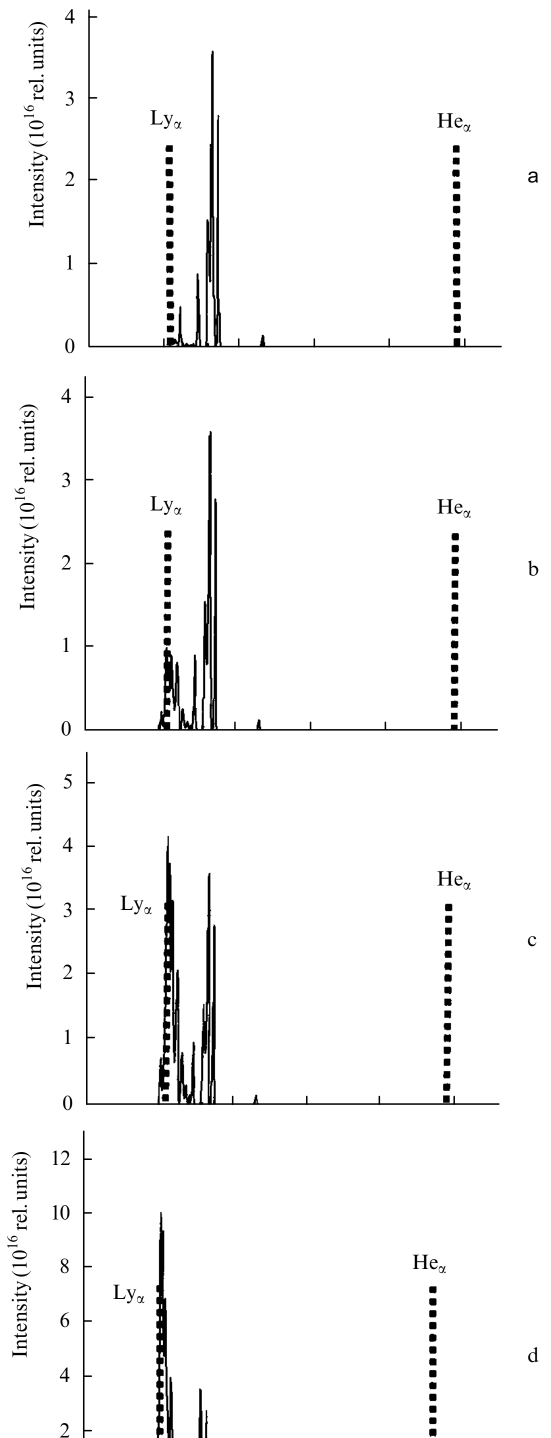

Figure 5. Spectral functions of a He-like MgXI ion calculated in the LTE approximation for a plasma temperature $T_e = 50$ (a), 100 (b), 200 (c), and 500 eV (d).

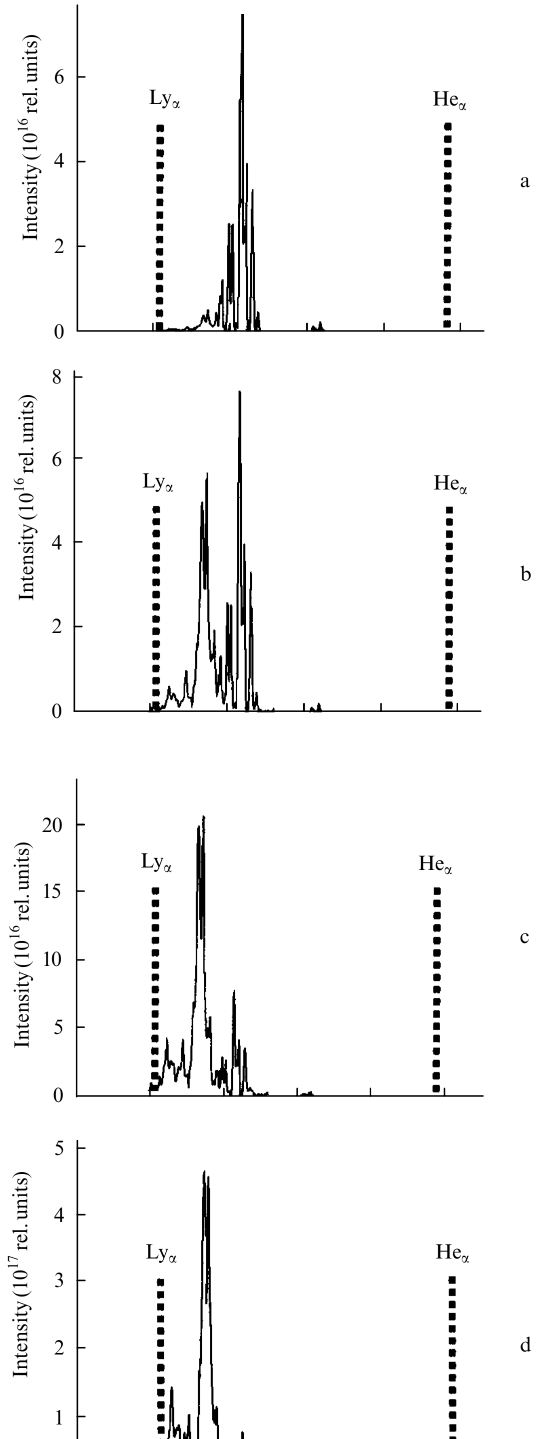

Figure 6. Spectral functions of a Li-like MgX ion calculated in the LTE approximation for a plasma temperature $T_e = 50$ (a), 100 (b), 200 (c), and 500 eV (d).

above form a quasi-continuous transition array in the 8.4 – 8.8 Å wavelength range. Also shown are the experimental spectrum (Fig. 11a) obtained in this work and the emission spectrum arising only from the dielectronic satellites of the Ly_α line of MgXII ions (Fig. 11c). Comparing these three spectra allows the following conclusions:

(1) The emission spectrum of the plasma produced by an excimer laser is by no means attributable, not even qualitatively, to the dielectronic satellites of the Ly_α line, even though at the same time the latter account very nicely for the IR-laser-produced plasma spectra observed previously (see Fig. 1b).

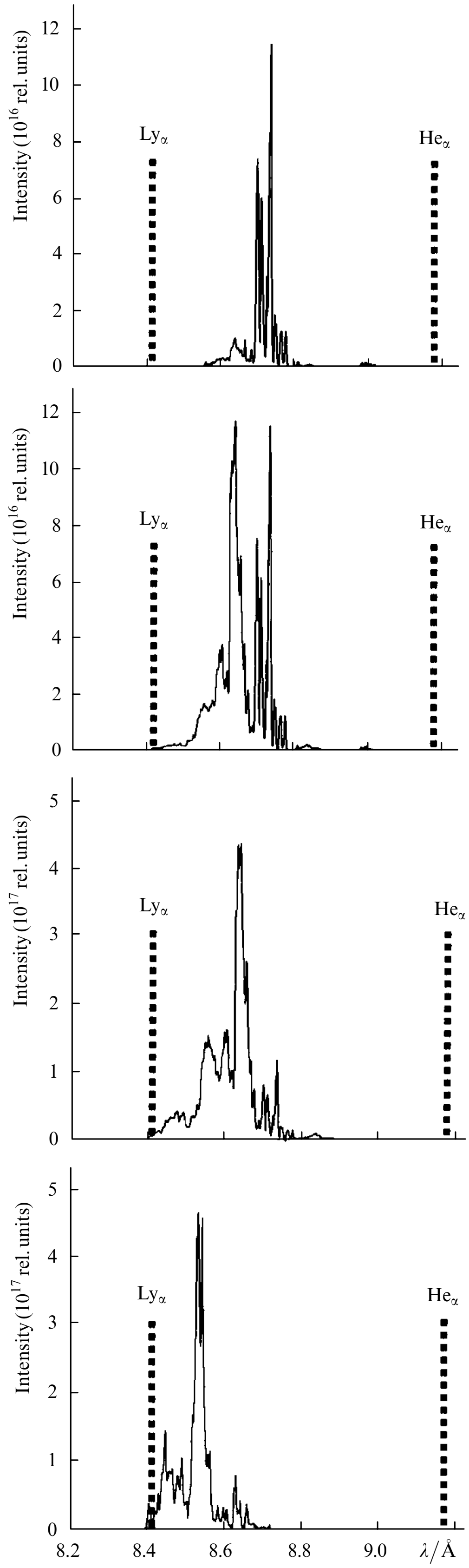

Figure 7. Spectral functions of a Be-like MgIX ion calculated in the LTE approximation for a plasma temperature $T_e = 50$ (a), 100 (b), 200 (c), and 500 eV (d).

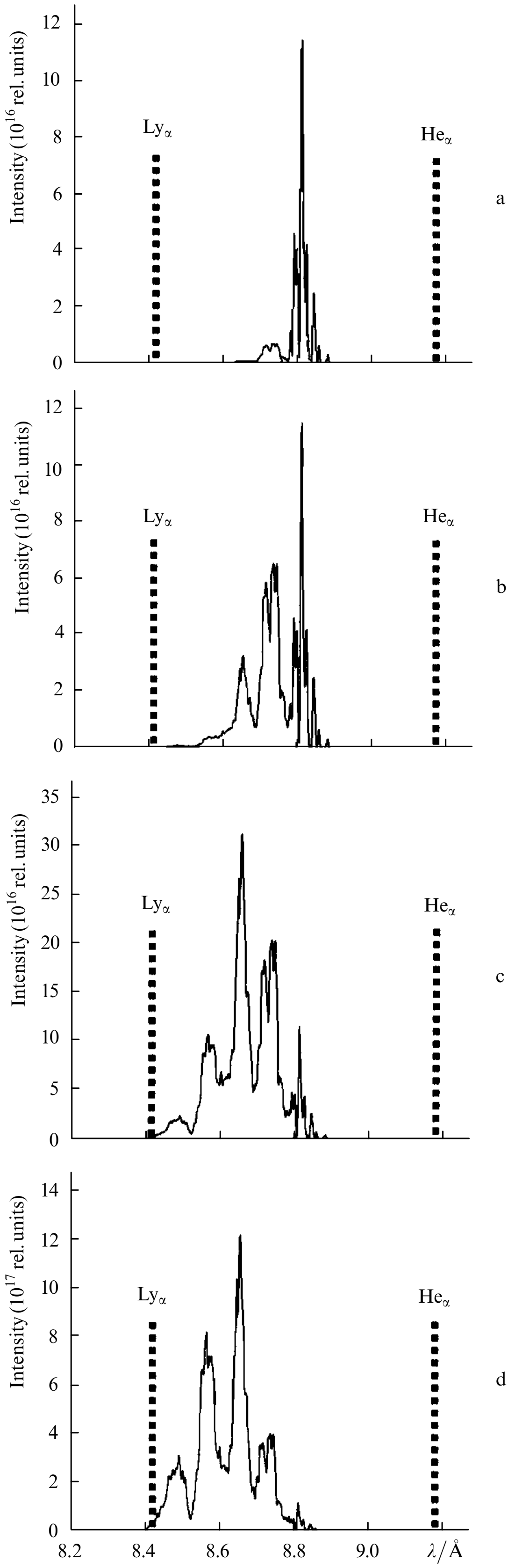

Figure 8. Spectral functions of a B-like MgVIII ion calculated in the LTE approximation for a plasma temperature $T_e = 50$ (a), 100 (b), 200 (c), and 500 eV (d).

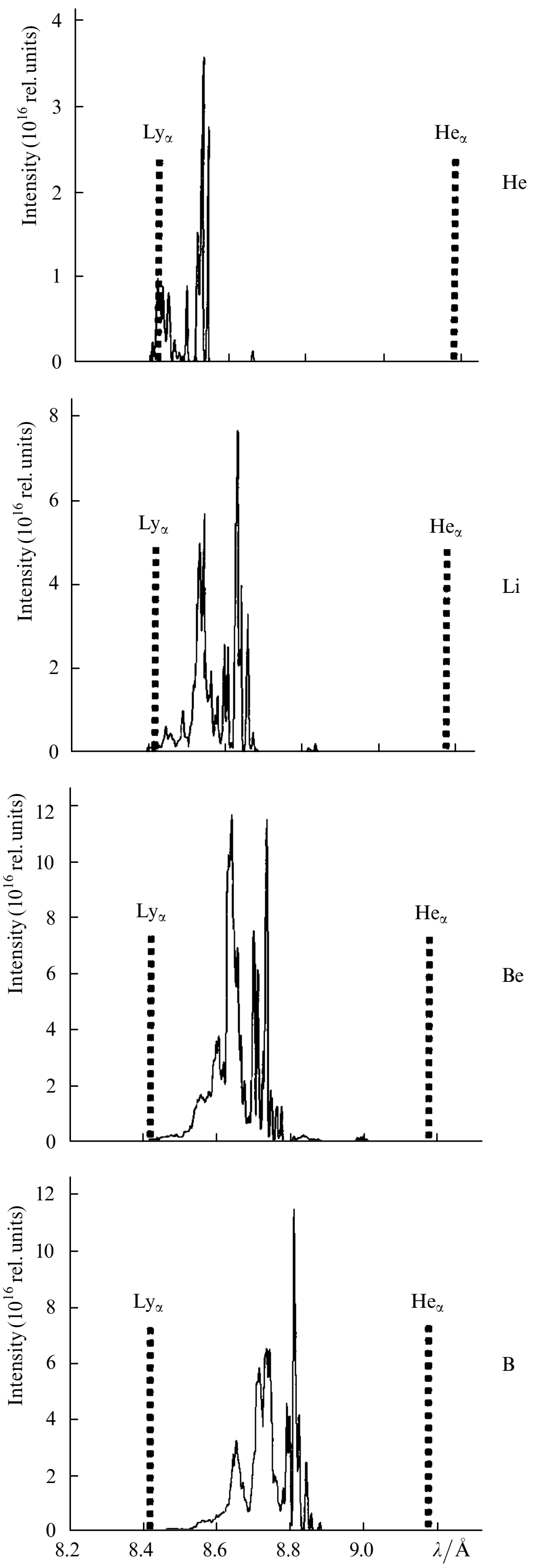

Figure 9. Spectral functions of magnesium ions of different isoelectronic sequences for $T_e = 100$ eV.

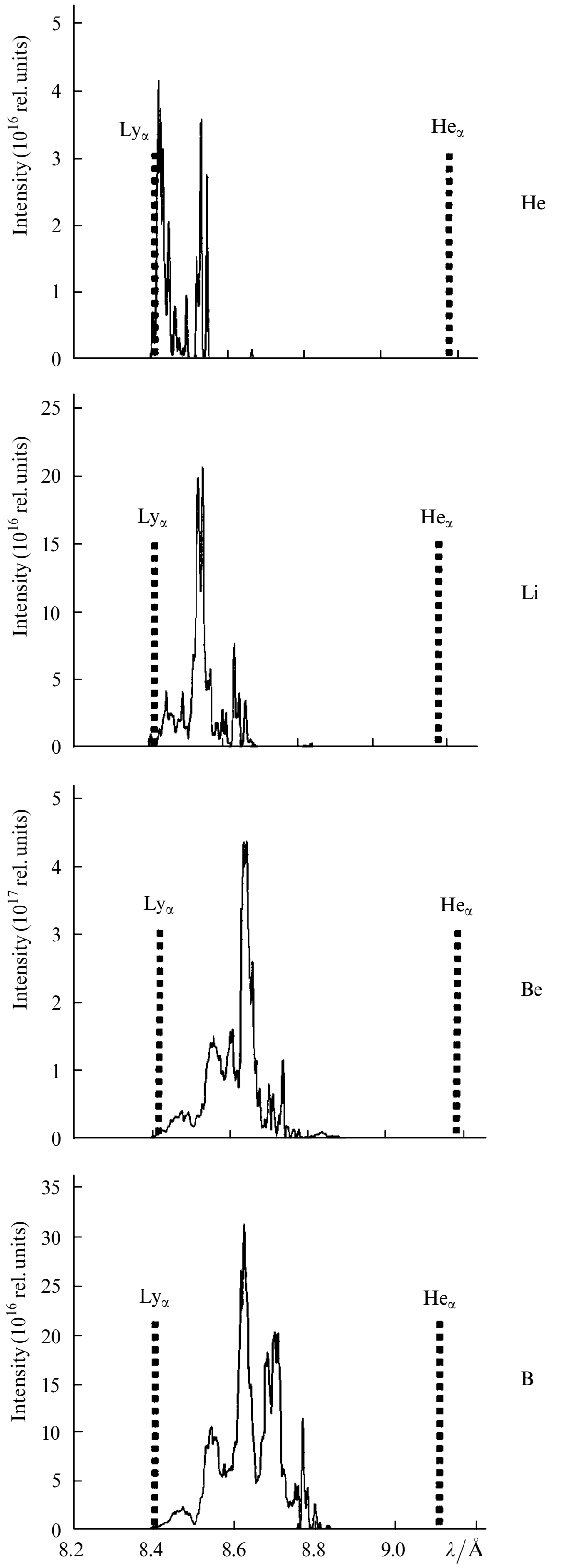

Figure 10. Spectral functions of magnesium ions of different isoelectronic sequences for $T_e = 200$ eV.

(2) The transitions in hollow magnesium ions with 3 – 5 electrons are responsible for a quasi-continuous spectrum, which lies not only in the spectral range between the Ly$_\alpha$ line and its dielectronic satellites, but also extends beyond its long-wavelength boundary.

(3) The emission spectrum in the 8.4 – 8.8 Å range is formed both by the dielectronic satellites and the transitions in Li-, Be-, and B-like hollow magnesium ions.

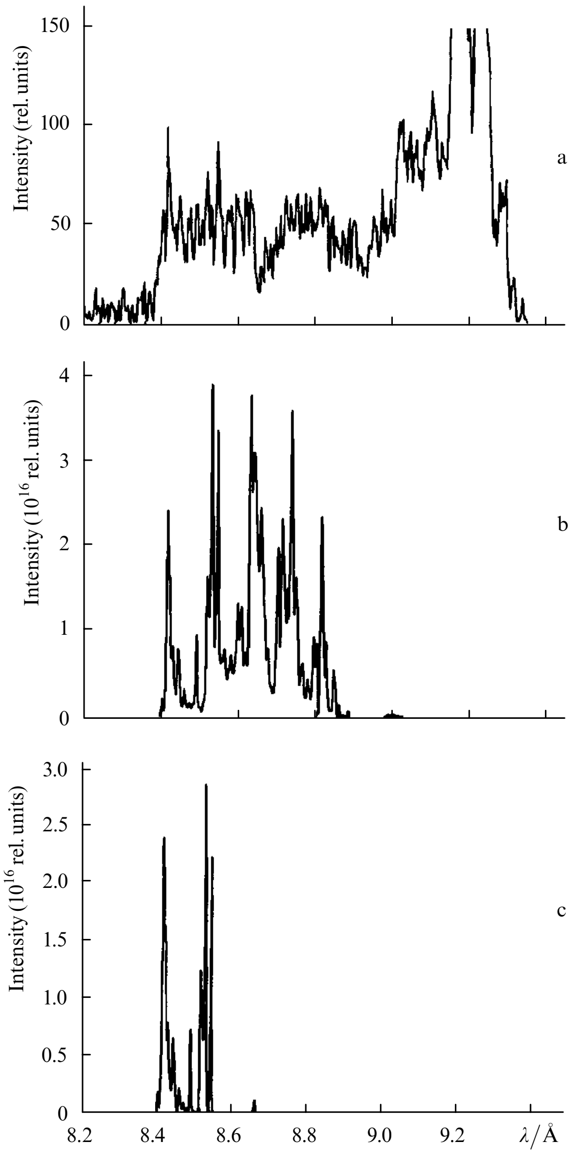

Figure 11. Emission spectra of a magnesium plasma in the 8.4 – 9.3 Å range: spectra observed in a plasma heated by an excimer laser (a) and also those obtained according to the theoretical model that includes the transitions in hollow ions with a number of electrons $M < 6$ (b) and according to the traditional model that includes only the dielectronic satellites of doubly excited states of He-like ions (c).

(4) The spectrum in the $\lambda > 8.7$ Å range may be ascribed only to the transitions in hollow ions with a number of electrons $M \geqslant 4$.

Naturally, the model spectrum shown in Fig. 11b cannot pretend to provide a quantitative description of the observed spectrum in the entire 8.4 – 9.2 Å range for a variety of reasons, the main ones being as follows:

First, we consider only the ions with a number of electrons $M < 6$. It is not the transitions in hollow C-, N-, and O-like ions under our examination that should, on the one hand, form the emission spectrum in the 8.7 – 9.2 Å range and, on the other, make some (moderate) contribution to the shorter-wavelength 8.4 – 8.7 Å range.

Second, the calculated spectral functions of multielectron configurations are, as noted in the foregoing text, in error (the errors are introduced primarily into the positions of individual spectral transitions), which may tangibly alter their spectral shapes.

Third, the experimental spectrum is the wavelength dependence of the plasma luminosity averaged over its entire lifetime. In the course of time evolution of the plasma, its parameters (density, temperature, ion composition) vary strongly. Referring to Figs 5 – 8, a variation of only the temperature has a dramatic effect on the spectral functions related to a given ion. Averaging the spectral functions over the temperature would yield a structurally more 'loose' spectrum with less clearly defined peaks, which is what we see in the observed spectrum.

4. Conclusions

The main result of our work is a demonstration that the unusual emission spectra of the excimer laser-produced plasma, located between the resonance lines of H- and He-like ions MgXII and MgXI, arise from transitions in hollow multicharged ions with a number of electrons $M > 2$. Elucidation of the mechanisms of hollow-ion formation in a laser-produced plasma calls for systematic experimental x-ray spectroscopic investigations as well as extended calculations of collisional-radiative ion kinetics in a transient nonuniform plasma with the inclusion of possible photoionisation by intrinsic emission and ion charge exchange.

It should be stressed that the spectral range between the satellites of the resonance line of an H-like ion and the resonance line of a He-like ion is, as shown above, best suited for experimental studies, because the spectra of hollow configurations in this range are not blended by other transitions. Until very recently, this range was believed to harbour no spectral transitions, and therefore no systematic investigations of the emission of a laser-produced plasma were pursued in this range.

We emphasise once again that pursuing these investigations does not, as shown in our work, necessarily require costly high-power laser or accelerator facilities. Advantage can be taken of relatively simple plasma sources on the basis of a moderately high-power short-wavelength laser with a pulse of nanosecond duration.

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