

# K $\alpha$ radiation from low charge chlorine heated by an ion beam for cold dense plasma diagnostics

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(RECEIVED 18 October 2010; ACCEPTED 4 January 2011)

## Abstract

K $\alpha$  radiation from low charge chlorine is examined for cold dense plasma diagnostics. A relativistic atomic structure calculation shows that the transition energies of K $\alpha$  lines are slightly shifted to a higher-energy side according to the degree of M-shell ionization. Total spectral shift from Cl<sup>+</sup> K $\alpha$  lines up to those with a fully stripped M-shell is about 10 eV. With an assumption that collisional-radiative-equilibrium is valid, spectral calculations were carried out for a C<sub>2</sub>H<sub>3</sub>Cl-plasma heated by an ion beam, and clear deformation among spectral line-shapes is found in the range of the electron temperatures of  $\leq \sim 30$  eV. Contribution to the composite spectra of K $\alpha$  lines with an excited electron in the outer-shell is also briefly discussed. Cl-K $\alpha$  lines with M-shell electrons can be expected to give us distinct understandings for energy deposition by an ion beam in cold dense plasma.

**Keywords:** Diagnostics; Energy deposition; Heavy ion fusion; K $\alpha$  radiation; K-shell ionization; M-shell electron

## INTRODUCTION

In heavy ion inertial fusion research, energy deposition by incident energetic ion beams is one of the critical issues, and many corresponding studies have been intensively done experimentally and/or theoretically (Hoffmann *et al.*, 1990; Peter *et al.*, 1991a, 1991b; Stöckl *et al.*, 1998). In the related experiments, time-of-flight (TOF) is one of the most powerful diagnostics for the purpose in the framework of particle diagnostics (Ogawa *et al.*, 2000; Oguri *et al.*, 2000; Hasegawa *et al.*, 2003). In the category of X-ray diagnostics, which is also one of the promising tools for the above purpose, Goel *et al.* (1998) demonstrated some numerical calculations of K $\alpha$  spectra from low-Z target material heated by a proton beam. In their study, because of cold plasma creation and a small number of M-shell electrons of target material, K $\alpha$  spectra with M-shell electrons do not seem to have enough spectral-shift to diagnose plasma temperature, and K $\alpha$  spectra with a partially ionized L-shell are found to be effective. MacFarlane *et al.* (1993) showed the plasma creation by a proton beam of about 30–45 eV near a target surface with the use of Al<sup>5+</sup> K $\alpha$  radiation. Rosmej

*et al.* (2005a, 2005b) and Rządkiwicz *et al.* (2010) experimentally obtained a charge distribution along Ca projectile trajectory in a SiO<sub>2</sub> aerogel target, and showed direct observation of Si-K $\alpha$  lines with L-shell vacancies and chemical bounding with neighbor oxygen. Their studies may lead us to a fruitful understanding of energy deposition by an ion beam, and have a potential to open a new field of cold and/or warm dense matter physics.

In our previous study (Kawamura *et al.*, 2006), Cl-K $\alpha$  spectra from polyvinyl-chloride (C<sub>2</sub>H<sub>3</sub>Cl), which is often used in laser-produced-plasma (LPP) experiments and chlorine is doped as a tracer, heated by a He<sup>2+</sup> beam were examined for dense plasma diagnostic, and a threshold temperature ( $\sim 85$  eV) is found to trace energy deposition by an incident ion beam traveling in a target. In the K $\alpha$  diagnostics with plasma creation by an ion beam, K-shell ionization and dielectronic capture are competed due to a small density of an ion beam, and the K-shell ionization process governs the kinetics of the K $\alpha$  emission below the threshold. Due to the threshold, there may be some aspects that the variation of the K $\alpha$  radiation with high charge states is not enough for the diagnostics. In such a case, the K $\alpha$  radiation with lower charge states is preferred.

On the other hand, a heating process by laser-produced fast electrons is one of the important issues in an LPP-scheme, and diagnostics using K $\alpha$  radiation with high charge states

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works well to observe energy deposition by fast electrons. There is no competition between atomic processes seen in plasma creation by an ion beam since the density of fast electrons is high enough to be near a critical density for an incident laser wavelength (Kawamura *et al.*, 2002; Nishimura *et al.*, 2003).

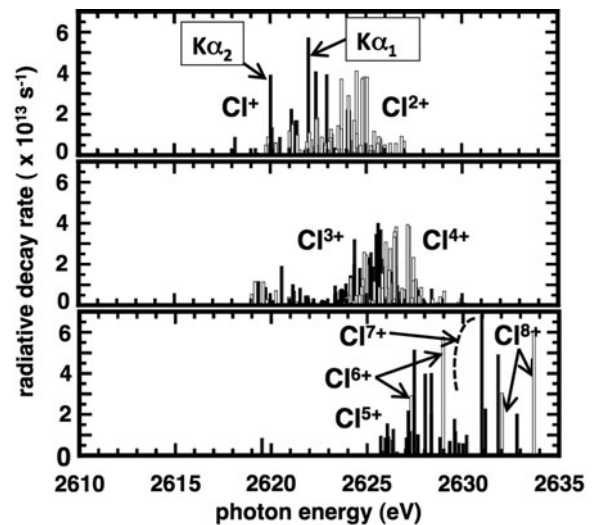
In the above studies, K $\alpha$  radiation with a partially ionized L-shell was intensively focused, resulting in well-separated K $\alpha$  spectra, which are essentially applicable to moderately hot plasmas, for instance, the electron temperature of near 100 eV is suitable for chlorine.

In this paper, the demonstration of K $\alpha$  lines from low charge chlorine with M-shell electrons (including a closed L-shell without M-shell electrons), which are associated with Cl $^+$ –Cl $^{7+}$  (including Cl $^{8+}$ ) and called “cold-K $\alpha$ ” hereafter, is examined for cold dense plasma diagnostics. Ionization between lower charge states results in small spectral shift of line radiation. K $\alpha$  radiation is an atomic transition between L and K-shells. The resultant spectral shift by M-shell ionization of chlorine is much smaller than that by L-shell ionization, and cold-K $\alpha$  is generally emitted at low plasma temperature.

In the calculation, a C $_2$ H $_3$ Cl target is used and the spectral shift from Cl $^+$  K $\alpha$  in accordance with M-shell ionization is clearly shown. Neutral chlorine has the electronic configuration of  $3s^23p^5$  in the M-shell, and total blue-shift due to the ionization from Cl $^+$  up to Cl $^{8+}$  is expected to be enough for distinct spectral diagnostics. Resultant spectral calculation shows the clear deformation of a line-shape at the electron temperatures of  $\leq 30$  eV. Hansen *et al.* (2005) derived such an electron temperature from a laser-irradiated Al-coated Ti foil plasma using low charge Ti-K $\alpha$  radiation. Neutral Ti has the electronic configuration of  $3s^23p^63d^24s^2$  in the outer-shell, and the clear spectral shift may be seen from the charge state of more than  $\sim 5$ , of which outermost  $3p$ -orbital is open. Since the blue-shift of Ti-K $\alpha$  with the ionization state of less than five may be small, the electron temperature must be higher than  $\sim 40$  eV for distinct spectral diagnostics. Chlorine has an open  $3p$ -orbital at neutral, and is expected to be a good candidate for cold dense plasma diagnostics. In addition, recent studies on the topic were done by Sengebusch *et al.* (2007) and Neumayer *et al.* (2009). They observed small spectral shift of Cl-K $\alpha$  radiation. K $\alpha$  radiation with a lower charge state also gives low emission density, which is the same as that with a higher charge state (Kawamura *et al.*, 2006). However, in the study, a resultant volume of plasma creation with an ion-beam-produced-plasma (IPP) scheme is much greater than that by an LPP-scheme, and the above problem may be solved.

## DESCRIPTIONS ON MODELING OF ATOMIC PROCESSES

Emission energies and oscillator strengths of Cl-K $\alpha$  lines are calculated with the use of GRASP92 and RATIP codes,



**Fig. 1.** Radiative decay rates and transition energies of Cl cold-K $\alpha$  lines calculated by GRASP92 and RATIP codes. From the chronological scientific tables of National Astronomical Observatory (2006), K $\alpha_1$ , K $\alpha_2$  are, respectively, 2622.3 eV and 2620.7 eV.

which are based on a multi-configuration Dirac-Fock (MCDF) method (Parpia *et al.*, 1996; Fritzsche, 2001, 2002). Figure 1 shows the radiative decay rates of Cl-K $\alpha$  lines from the charge states of Cl $^+$ –Cl $^{8+}$ , and they come from the ground-states with a vacant K-shell, namely, Cl $^+$ :  $1s2s^22p^63s^23p^5 \rightarrow 1s^22s^22p^53s^23p^5 + h\nu$ , Cl $^{2+}$ :  $1s2s^22p^63s^23p^4 \rightarrow 1s^22s^22p^53s^23p^4 + h\nu$ , ..., Cl $^{8+}$ :  $1s2s^22p^6 \rightarrow 1s^22s^22p^5 + h\nu$ . The resultant K $\alpha_1$  and K $\alpha_2$  are found to be calculated within the accuracy of  $\sim 0.5$  eV compared with one of available data (National Astronomical Observatory, 2006). The K $\alpha$  lines overlap with those of the next charge state, and cold-K $\alpha$  is mainly composed of the lines from Cl $^+$ –Cl $^{6+}$ , which are distributed in 2.62 keV–2.63 keV, and the spectral blue-shift due to M-shell ionization of  $\sim 10$  eV is found. The cold-K $\alpha$  lines can be expected to be good candidates for cold dense plasma diagnostics with the spectral resolution on the order of 1 eV. As is discussed in our previous study, it should be noted again that the non-radiative channels due to Auger transitions must be considered for the estimation of net K $\alpha$  emission as the fluorescence yields of Cl-K $\alpha$  lines with low charge states are  $\sim 0.05$ .

The model of population kinetics considered here is presented in Figure 2. Since the population of 1s-vacant ions is very small, the average charge state of total Cl-ions  $Z_{\text{total}} \approx Z_{\text{bulk}} \approx Z_{1s\text{-vacant}} - 1$ , where  $Z_{\text{bulk}}$  and  $Z_{1s\text{-vacant}}$  stand for the average charge state of bulk-ions and 1s-vacant ions, respectively (Kawamura *et al.*, 2002). According to Figure 6 given in Kawamura *et al.* (2006), an intensity ratio between cold-K $\alpha$  radiation and high charge one is an effective index to deduce an electron temperature  $T_e$  in the range of  $50 \text{ eV} \leq T_e \leq 100 \text{ eV}$ , in which the ratios give almost 0.1–10. For  $T_e \leq 50$  eV, it is suggested that only K $\alpha$  lines with M-shell electrons can be applicable.

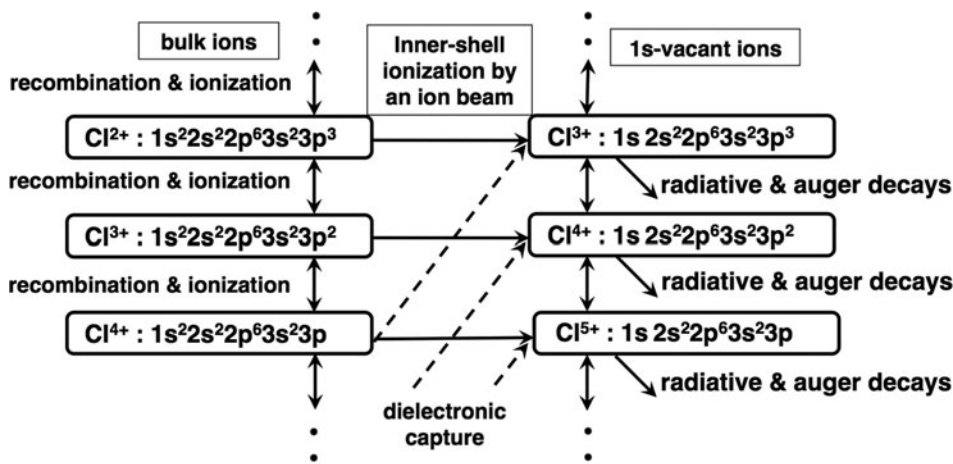


Fig. 2. Population kinetics associated with cold-K $\alpha$  lines.

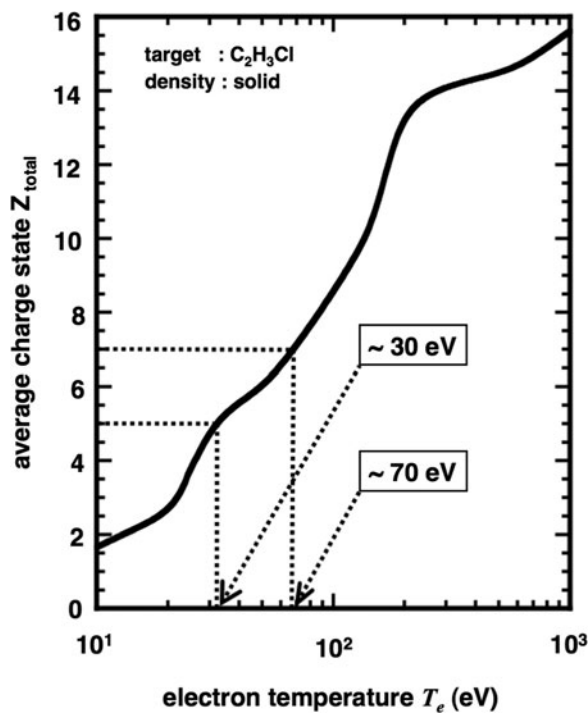


Fig. 3. Dependence of the average charge state of chlorine on electron temperature. Target material is  $\text{C}_2\text{H}_3\text{Cl}$  and the plasma ion density is solid density ( $\approx 8.1 \times 10^{22} \text{cm}^{-3}$ ). The incident beam is a  $\text{C}^{6+}$  beam, of which current density and mean energy, respectively are  $3 \text{ kA/cm}^2$  and  $30 \text{ MeV}$ . The energy spread of the ion beam described by Maxwellian is 10% of the mean energy.

### SPECTRAL DEFORMATION OF K $\alpha$ LINES WITH M-SHELL ELECTRONS

In the viewpoint of an average charge state, we can see a threshold temperature below which K $\alpha$  lines with M-shell electrons are dominant. Figure 3 is the average charge state of chlorine in a polyvinyl-chloride ( $\text{C}_2\text{H}_3\text{Cl}$ ) plasma irradiated by a  $\text{C}^{6+}$  beam, of which beam-current density and mean particle energy are, respectively,  $3 \text{ kA/cm}^2$  and  $30 \text{ MeV}$ , and the energy spread of the ion beam described by Maxwellian is

10% of the mean energy. The mean energy of  $30 \text{ MeV}$  is assumed so as to get enough cross-section of K-shell ionization (Kawamura *et al.*, 2006), and the plasma ion density is assumed to be solid density ( $\approx 8.1 \times 10^{22} \text{cm}^{-3}$ ). The atomic population for  $Z_{\text{total}} \leq 7$ , which corresponds to  $Z_{1s-\text{vacant}} \leq 8$ , is dominant in  $T_e \leq \sim 70 \text{ eV}$ , and that for  $Z_{\text{total}} \leq 5$ , which is  $Z_{1s-\text{vacant}} \leq 6$  and main component of cold-K $\alpha$  lines, is in  $T_e \leq \sim 30 \text{ eV}$ . Therefore, the K $\alpha$  lines from  $\text{Cl}^+ - \text{Cl}^{6+}$  are applicable in  $T_e \leq \sim 30 \text{ eV}$  for plasma diagnostics.

The dependence of a spectral line-shape on the electron temperature is presented in Figure 4. Calculated spectra are convolved by Stark broadening with quasi-static electric-microfield, electron impact broadening using a semiclassical expression, natural and Doppler broadenings (Kawamura *et al.*, 2001, 2003). In the Doppler broadening calculation, the ion temperature is assumed to be equal to the electron

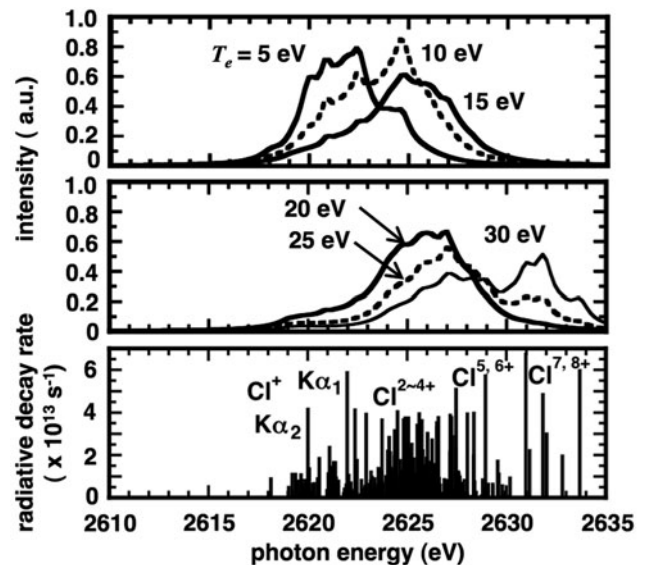


Fig. 4. Deformation of a spectral line-shape and blue-shift of cold Cl-K $\alpha$  radiation from a polyvinyl-chloride ( $\text{C}_2\text{H}_3\text{Cl}$ ) plasma. The plasma ion density is solid density. The incident beam parameters are same as in Figure 3.

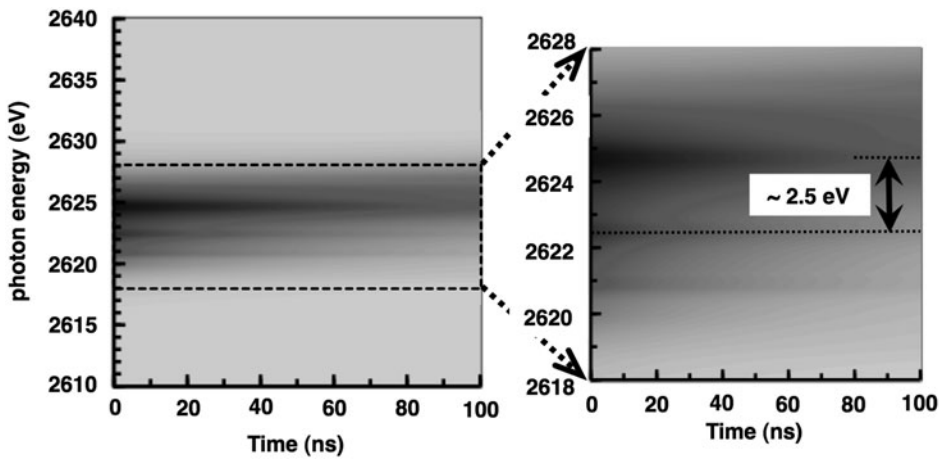


Fig. 5. Demonstration of a time-evolution of Cl-K $\alpha$  spectral deformation from a polyvinyl-chloride ( $C_2H_3Cl$ ) plasma. The plasma ion density is solid density, and the electron temperature is raised from 10 up to  $\sim 15$  eV heated by a  $C^{6+}$  beam, which is assumed to be flat-top with a width of 100 ns. The other beam parameters are same as in Figure 3.

temperature. At  $T_e = 5$  eV, the spectrum consists of mainly two K $\alpha$  components of  $Cl^+$  and  $Cl^{2+}$ . The 1s-vacant states of  $Cl^+ - Cl^{3+}$  contribute to the composite spectrum at  $T_e = 10$  eV. With increase in  $T_e$ , the charge states of main cold-K $\alpha$  components are  $Cl^{2+} - Cl^{4+}$  in  $T_e = 15 - 20$  eV, and the K $\alpha$  lines from  $Cl^{5+} - Cl^{7+}$  have a large contribution to the spectra in  $T_e \geq 25$  eV. Finally, the total blue-shift of the K $\alpha$  lines from  $T_e = 5$  eV up to 30 eV is  $\sim 10$  eV, and the clear spectral deformation can be observed.

Figure 5 also gives a time-evolution of K $\alpha$  spectra for the demonstration of blue-shift with an assumption that population kinetics can be described by a collisional-radiative equilibrium (CRE) condition. The conditions of an incident ion beam are the same as the above calculation, and a flat-top pulse beam with a width of 100 ns is assumed. The energy deposition is approximated by a conventional Bethe's formula with some corrections for bound electrons (Mehlhorn, 1981; Ziegler, 1999), and a formula studied by Peter *et al.* (1991a, 1991b) for plasma free-electrons. An initial plasma ion density and electron temperature are, respectively, solid density and 10 eV. With the incident  $C^{6+}$  pulse beam, the electron temperature is raised from 10 eV up to about 15 eV assuming that created plasma is ideal, resulting in the spectral blue-shift of  $\sim 2.5$  eV and sharpening. Such a time-evolution can be diagnosed using cold-K $\alpha$  spectra with the spectral accuracy on the order of a 1 eV.

According to the study by Kauffman *et al.* (1975), the contribution of the multiple ionization  $KL^n$  ( $n$  stands for a number of L-shell vacancies.) of neon was discussed. In their study, colliding particles come from an incident ion beam. On the other hand, in our study, even if the multiple ionization associated with M-shell  $KM^n$  is occurred to chlorine by an incident ion beam, because of high density plasma free-electrons up to  $\sim 10^{23} \text{ cm}^{-3}$ , the M-shell ionization is governed by the surrounding plasma free-electrons. The particle density of an incident ion beam is at most  $10^{13-14} \text{ cm}^{-3}$  and the difference of the colliding particle densities between free-electrons and incident ions is up to 9–10 digits. The contribution of the multiple ionization  $KM^n$  can be neglected. Concerning the multiple ionization associated with the L-shell of chlorine, the binding

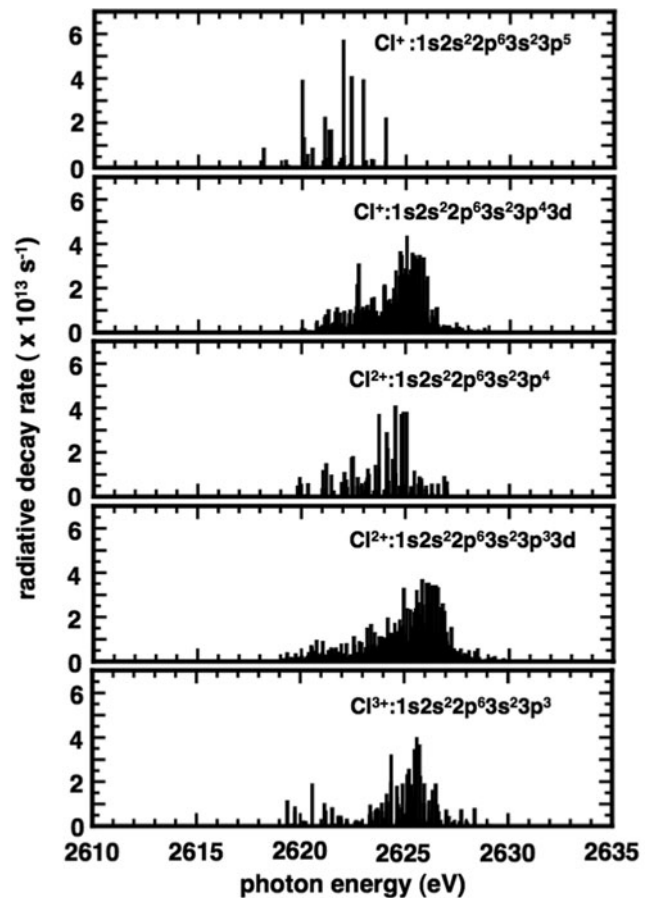


Fig. 6. Radiative decay rates and transition energies of the satellite lines of Cl cold-K $\alpha$  lines, which are also calculated by GRSP92 and RATIP codes.

energies of the L-shell electrons of each low charge state are about 10 times those of the M-shell electrons. Those are about 200–300 eV. Therefore, the contribution of the multiple ionization  $KL^n$  by an incident ion beam is much less than that of  $KM^n$ , and the well-defined spectra for temperature diagnostics without any consideration of the multiple ionization may work well.



### CONTRIBUTION OF SATELLITE LINES AND OPACITY EFFECT TO K $\alpha$ LINES WITH M-SHELL ELECTRONS

Concerning satellite lines, atomic states with an excited electron in the outer-shell ( $\text{Cl}^{n+}; 1s2s^22p^63s^23p^{(5-n)}nl$ ) may have a contribution to the composite spectra. In Figure 6, the K $\alpha$  lines from the excited states  $1s2s^22p^63s^23p^{(5-n)}3d$  strongly overlap with those from  $\text{Cl}^{n+}; 1s2s^22p^63s^23p^{(6-n)}$  and  $\text{Cl}^{n+1}; 1s2s^22p^63s^23p^{(5-n)}$ . However, due to a large contribution of continuum lowering at solid density, an ionization energy of bound electrons is reduced and population of atomic states with a shallow ionization potential is estimated to be small. In Figure 7, the dependence of average continuum lowering  $\Delta E$  (Stewart & Pyatt, 1966) on the electron temperature  $T_e$  is given, and a comparison with the average ionization energies  $I_p$  of  $\text{Cl}^{n+}; 1s2s^22p^63s^23p^{(6-n)}$ , which are the ground-states of 1s-vacant states, is made.  $I_p$  is shown on the average with respect to a total quantum number  $J$  associated with a  $nl$ -electronic-configuration. With increase in  $T_e$  up to 30 eV,  $\Delta E$  can be raised to  $\sim 60$  eV, which is near the average ionization energy of  $\text{Cl}^{3+}$ . Since an ionization degree is affected not only by local electron-temperature but also by continuum lowering,  $(T_e + \Delta E)$  is a good index to predict the degree. From Figures 4 and 7, the spectral deformation can be seen with increase in the index, and such atomic excited states as  $\text{Cl}^{n+}; 1s2s^22p^63s^23p^{(5-n)}nl$  may be neglected at solid density.

To utilize the cold-K $\alpha$  lines for cold dense plasma diagnostics, opacity effect is also one of the critical properties.

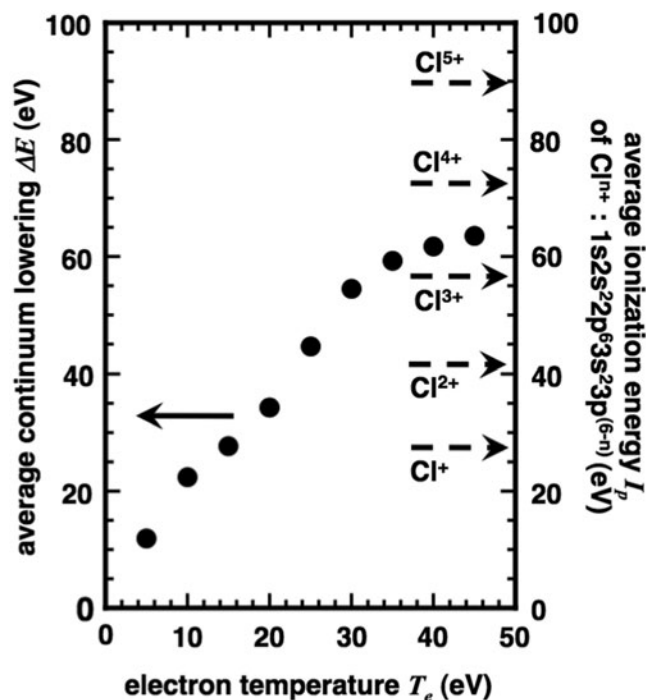


Fig. 7. Comparison between the average continuum lowering and the ionization energies of 1s-vacant  $\text{Cl}^{n+}; 1s2s^22p^63s^23p^{(6-n)}$ .

In one of our past studies (Kawamura *et al.*, 2003), highly charged major K $\alpha$  lines at a certain electron temperature are saturated at the plasma thickness of a few microns, and those minor ones at a few tens microns. This is because the final atomic states associated with the K $\alpha$  lines belong to bulk ions. However, the final atomic states of the K $\alpha$  lines with M-shell electrons discussed here are  $\text{Cl}^{n+}; 1s^22s^22p^53l^{(8-n)}$  ( $1 \leq n \leq 6$ ). As discussed above, the binding energies of the L-shell electrons are about 200–300 eV in accordance with M-shell ionization, and the temperature of bulk-electrons is too low to ionize the electrons. Therefore, those states have a small contribution to determine overall population kinetics, resulting in small opacity for the cold-K $\alpha$  lines. This is a qualitative suggestion at the moment, and quantitative consideration will be done in near future.

### CONCLUSIONS

The plasma diagnostics with cold-K $\alpha$  radiation is proposed. To utilize the radiation, clear blue-shift of cold-K $\alpha$  lines has to be examined, and the lines show the shifts of about 10 eV in accordance with M-shell ionization. From the spectral calculation, the clear deformation of the spectral shape is found in the electron temperatures of  $\leq 30$  eV. Concerning the K $\alpha$  lines from the 1s-vacant states with an excited electron in the outer-shell, it may be expected that they have a small contribution to the composite spectra due to a large continuum lowering at solid density. Opacity effect on the cold-K $\alpha$  radiation is briefly discussed, and give a suggestion qualitatively. Finally, cold-K $\alpha$  lines are one of promising tools for cold dense plasma diagnostics. The opacity effect will be quantitatively studied, and reported somewhere.

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