Charge state distributions after sub-shells ionization in Lead atom

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Abstract

Monte Carlo technique has been used to simulate the atomic reorganization vacancy cascades in single ionized and state ions of lead (Pb^{q+} where q=l, 2...70 ionization fold). The highly charged ions and average charge state distributions are carried out after inner-shell ionization. The computer program is made on base of tracing successive x-ray; Auger and Coster-Kronig transitions to filling the holes that exist in atomic subshells. For systematic investigations the problem occurs, that for the most elements consistent sets of atomic data (radiative and non-radiative transitions) are not available. Therefore, we calculate radiative transitions probabilities (x- ray transitions) with program using Multiconfiguration Dirac Fock (MCDF) wave functions and non-radiative transitions probabilities with code using Dirac Fock Slater (DFS) wave functions. The shake-off processes and the closing of energetically Forbidden Coater-Kronig channels during the cascade development are considered. It's found that the heavier atom such that lead atom (Pb) and the lower shell or subshell in which an initial vacancy is produced, the higher will be the average ion charge and the more complex will be the charge spectrum.

Introduction

Inner-shell ionization processes were analyzed under different points of view. Thereby, basic investigation of atomic properties as well as investigation in connection with other disciplines as solid state physics, plasma physics and astrophysics was of interest. Astrophysical plasma, high temperature laboratory and ion-atom collisions are important sources of x-ray radiation. The density and temperature of high temperature plasmas can be determine from an analysis of the emission spectra. The atomic ionization of some astrophysical environmental can be significantly modified by radiative and non- radiative following inner-shell photo ionization.

The ionization of an inner-shell electron leaves the atomic system in a very unstable electronic configuration that may decay by either radiative or non-radiative transitions. The filling of an inner-shell vacancy in an atom or ion may produce a new vacancies in higher shells or subshells, which can then be filled by farther radaitive or / and non-radaitive transitions. This successive transitions during the atomic reorganization called vacancy cascade process, which continues until all vacancies reach the outermost occupied shell. Thus highly charged ion states are ultimately produced. During the vacancy cascade development many photons and Auger electrons may be emitted. In addition to the vacancy filling processes (x-ray and Auger transitions), there is an electron shake off process. It can be interpreted as, when a vacancy is created, there is an a small probability that electron in another shell is excited to an unoccupied bound state or ejected into the continuum as the result of the sudden change of atomic potential. This secondary process causes additional vacancies during the vacancy cascade development and plays an important role in the final charge state distribution (CSD) of ions. During the cascade development some non-radiative channels (Coster-Kronig channels) become energetically forbidden, this process results from the transition energy shifts after multi-electron emission in the atomic structure.

The investigation of vacancy cascade has been of considerable interest in different field of research. Thus thermal multi-charged ions can be produced on intensive sources of synchrotron radiation by sequential K-, L-, and M subshells photoionization [1,2,3,4]. Complete Auger decay pathways of Kr $3d^{-1}$ hole levels including direct double processes are investigated by Zeng el at.[5]. Absolute cross sections for electron impact single and multiple ionization of C^+, N^+ and O^+ ions leading to the formation of C^{q+} (q = 2–4), N^{q+} (q = 2–5) and O^{q+} (q = 2–5) are reported [6]. The so produced thermal ions allow fundamental investigations on the collision physics in different plasma as well as precision spectroscopy for testing QED effects and quantum mechanical electron transfer theories. Vacancy cascades in large molecules leading to molecule decomposition (molecule fragmentation) [7,8,9,10]. Corresponding investigations are also of importance for understanding material damages after irradiation, for understanding detection mechanisms of ionizing radiation and for investigations of solid state effects at photoelectron emission, electron loss spectroscopy. The energy dependence of the resulting charge spectrum from the excitation energy near the absorption edges is of importance [11,12,13]. The studies of inner-shell ionization processes are important in X-ray Fluorescence Spectroscopy (XRFS). Because each atom has a unique set of energy levels, each element produces characteristic x-rays at a unique set of energies, allowing one to non-destructively measure the elemental composition of a sample. Since this method is fast and non-destructive to the sample, it is the method of choice for field applications and industrial production for control of materials. Auger Electron Spectroscopy (AES) a surface specific technique utilizing the emission of low energy electron in the Auger process. It is one of the most commonly employed surface analytical techniques for determining the composition of the surface layers of a samples, and simply a technical problem of detecting charged particles with high sensitivity, with the additional requirement that the kinetic energies of the emitted electrons must be determined. It is useful to briefly review the electronic structure of atoms and solids.

In the present work, we calculate the charge state distribution (CSD) probabilities P(q) after K-, L-, and M- inner-shell ionization of neutral lead Pb and it's ground state ions Pb^{1+}, and Pb^{2+}. The Pb atom is selected heir, since the core electrons remain practically undisturbed in most of inner-shell transition processes. The mean ion charge state distributions P(q) following K, L-, and M- inner-shells ionization of Pb^{q+} ion ground states up to q= 70 ionization degree are pointed out. The modeling of vacancy cascades are carried out using developed Monte Carlo technique [14,15,16]. We calculate radiative transitions probabilities (x-ray transitions) for neutral and ionized lead atom up to 70 ionization fold with an computer code developed by Reiche [17] using MCDF wave functions [18,19] and non-radiative transitions (Auger and Coster-Kronig transitions) with an code written by Lorenz and Hartmann [20]. Shake-off process was calculated with computer code by El-Shemi [21]. The electron shake-off probabilities and closing of energetically forbidden Coster Kronig channels during the cascade development are considered in the present work.

Method of calculation

We have used the Monte Carlo method for simulated vacancy cascades following inner- shell ionization considering x-ray emission, Auger and Coster-Kronig transitions and electron shake-off process. An analysis of each cascade begins with the consideration of all possible electron transitions that may fill an initial vacancy. In the following development of the cascade in each step the computer program selects one of the possible processes according to their relative probabilities by use of random number. After realization of the selected transition, a new configuration of vacancy appears. For each vacancy, first we check whether the electron shake-off will take place or not. If an electron shake off process takes place an electron will be eject from the atomic shells. In a next step we decide from the fluorescence yield whether the transition is radiative or non-radiative transitions. When it is radiative, the new position of the is selected from the partial radiative transition probabilities. In the case of non-radiative transitions two new vacancies are generated according to the relative transition probabilities of the energetically allowed Auger channels. For each new vacancy the computer program goes back the first step described above. The appearance of new vacancy configuration continues until all these vacancies have reached the outer shell. Then the number of vacancies are recorded and got stable results for the final charge state distribution of ions. The calculation are made under assumption that the transition rates for an atom with several vacancies during the cascade development are given a statistical weight deduced from the scaling procedure proposed by Larkins [22]. A more detailed description of the calculation for vacancy cascades after inner-shell ionization of atoms is given in [14,15,16]

Results and Discussion

The x-ray transition rates and electron shake-off probabilities for single ionized Pb, and ion ground states Pb^{q+} up to 70 ionization fold (fully ionized 3s orbital) were calculated using Multiconfiguration Dirac Fock (MCDF) wave functions. The values of non-radiative transitions (Auger and Coster Kronig channels) have been obtained with Dirac Fock Slater (DPS) wave functions. Auger transition energies are determined by the (Z+l) rule using binding energy. These Auger transition energies are important to determine the allowed and forbidden non-radiative channels during the program development. With increasing number of vacancies during cascade process some Auger channels turn out to be energetically forbidden due to the change in electron binding energies. The electron shake-off causes additional vacancies during cascade reorganization of atomic shell. The including of shake-off process and considering of the energy prohibition of Auger transitions leads to more and highly accuracy of results for charge state distributions p(q) and average charged ion $\langle q \rangle$. We now investigated vacancy cascades of a wide verity of ground state ions of Pb^{q+} (q= 1,2,.... .70) ionization fold. Fig. 1 shows the average charge state distributions for lead after initial vacancy in K and L_1 sub-shells as function of ionization degree up to 70 ionization fold (fully ionized 3s orbital). It's found, that the localization of a primary vacancy decisively determines the development of a vacancy cascade. Deeper vacancies lead to higher mean ion charge states and vacancies in outer shells produce only ions with some few additional vacancies because of the restricted number of possible de-excitation channels. As anticipated, the mean charge is found to increase with increasing binding energy of the sub-shell in which the initial vacancy is created. You can see from the fig., an exception to this trend occurs in the case of K- shell vacancy, which give rise to a lower mean charge than an L_I sub-shell vacancy. This exception is attributable to radiative transition (x-ray transitions) which transfer the initial K shell vacancy to $L_{II}(K_{\alpha2})$ or $L_{III}(K_{\alpha1})$, or to the higher sub-shells M and N without ionization (no additional new vacancies are corresponding to this previous transitions), only the primary vacancy in K shell will goes up to the final state of transition. The fact, that non-radiative transitions (Auger transitions) are more probable for L_I sub-shell than for K shell. Thereby the initial L_I sub-shell vacancy may be filled by Coster Kronig transition (L_I- LM) or (L_I - LN). This process can create new vacancy in final state of transition, this new vacancy well be fill again by further radiative and non-radiative transitions. At first clear bend is seen in the curve at $\langle q \rangle$ = 10.9 after L_I and 10.02 after K vacancies, where the 6s is completely ionized. In the range ionization degree of q= 2... 30 the 5d, 4f, 5p, and 5s orbital are ionized step by step. We find, that only small difference in the mean number of ejected electrons $\langle q \rangle$ from ions of corresponding charge states. Beginning with 4d ionization the number of ejected electrons decrease until the fully ionized N- sub-shell is reached at q= 56. The next significant decrease in the average ion charge states reached occurs at about q = 62 (beginning of L sub-shell ionization). The increasing of mean charged ions state at Pb^{62}+ occurs because the transition rate for K- LL Auger processes are three orders of magnitude higher that for the x ray K- L transitions.

The multiple ionization probabilities resulting from the cascade decay of single K-, L_I- and M_1 inner-shell vacancies in neutral Pb^+ are displayed in Fig. 2 as function of ejected number of electrons q by vertical bars located above each value of the final charge. The ionization probabilities are normalized to unity. The mean charge state ions $\langle q \rangle$ are given on the figures. For each primary vacancy the average charge state ions are calculated with the relation $\langle q \rangle = \sum qP(q)$, where q is the number of electron and P(q) is charge state distributions (CSD). The heavier atoms such as Pb and the lower shell in which an initial vacancy is produced, the higher will be the average ion charge and the more complex will be the charge spectrum. The intensities of the remaining ions are more or less symmetrically distributed for these complex spectra. The probabilities of law ion charge e.g. one and two ion charge states are less in heavy atoms after inner shell ionization, but the higher ion charge states such as q=15 the probability is high. You can see in the figures the higher probability of remaining ions for Pb^{0+} after K vacancy is q = 14. The higher charge spectrum is found in the charge state distributions of Pb^{0+} result following L_1- vacancy (q =15). When the location of primary vacancy in outer shell, e.g. M shell, the charge state distributions are no more complex. The highest charge ion spectrum after M shell vacancy is q=12.

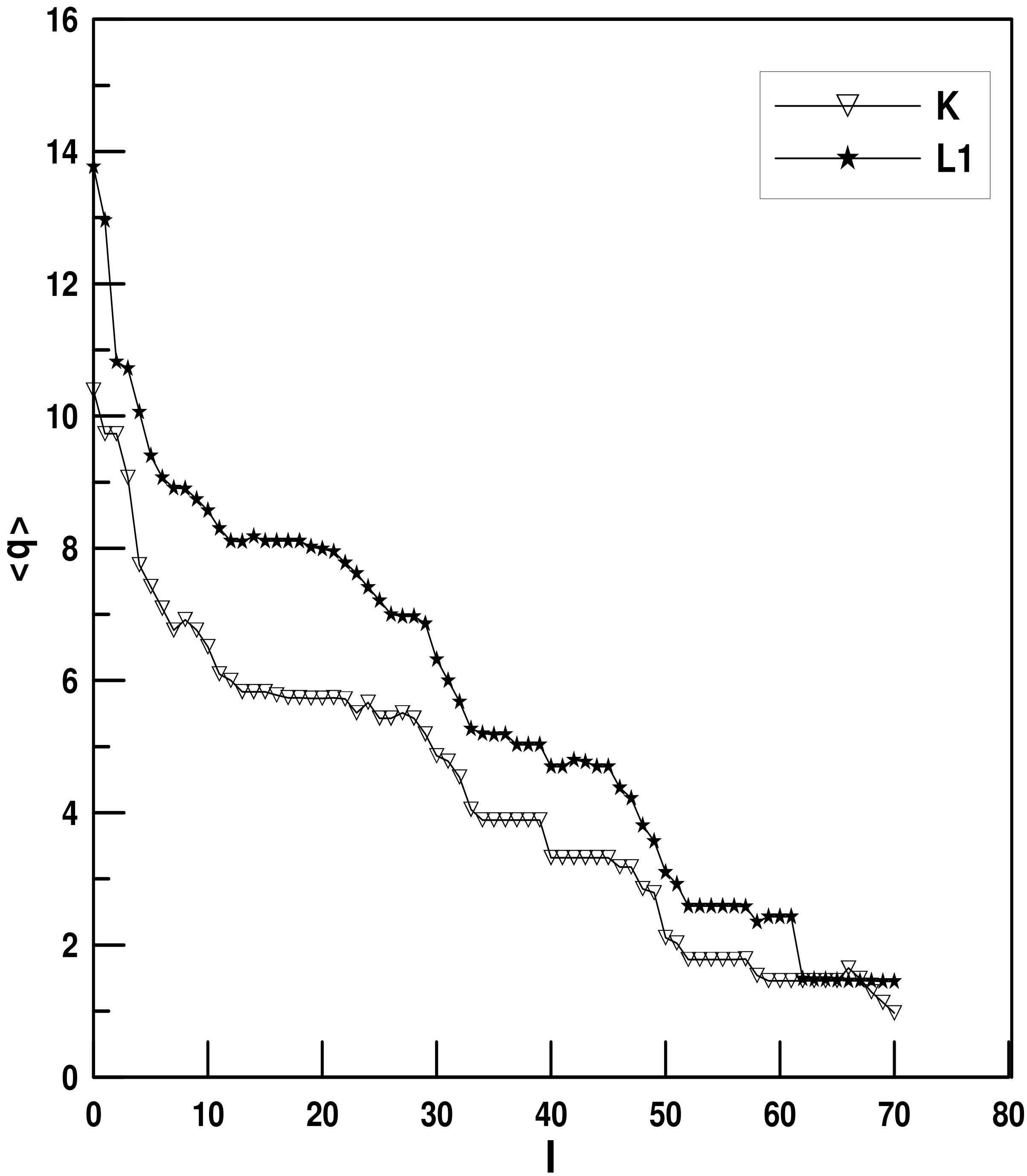

Figure 1: Average number of ejected electrons after K- and L_I shells ionization as function of ionization degree I for lead atom

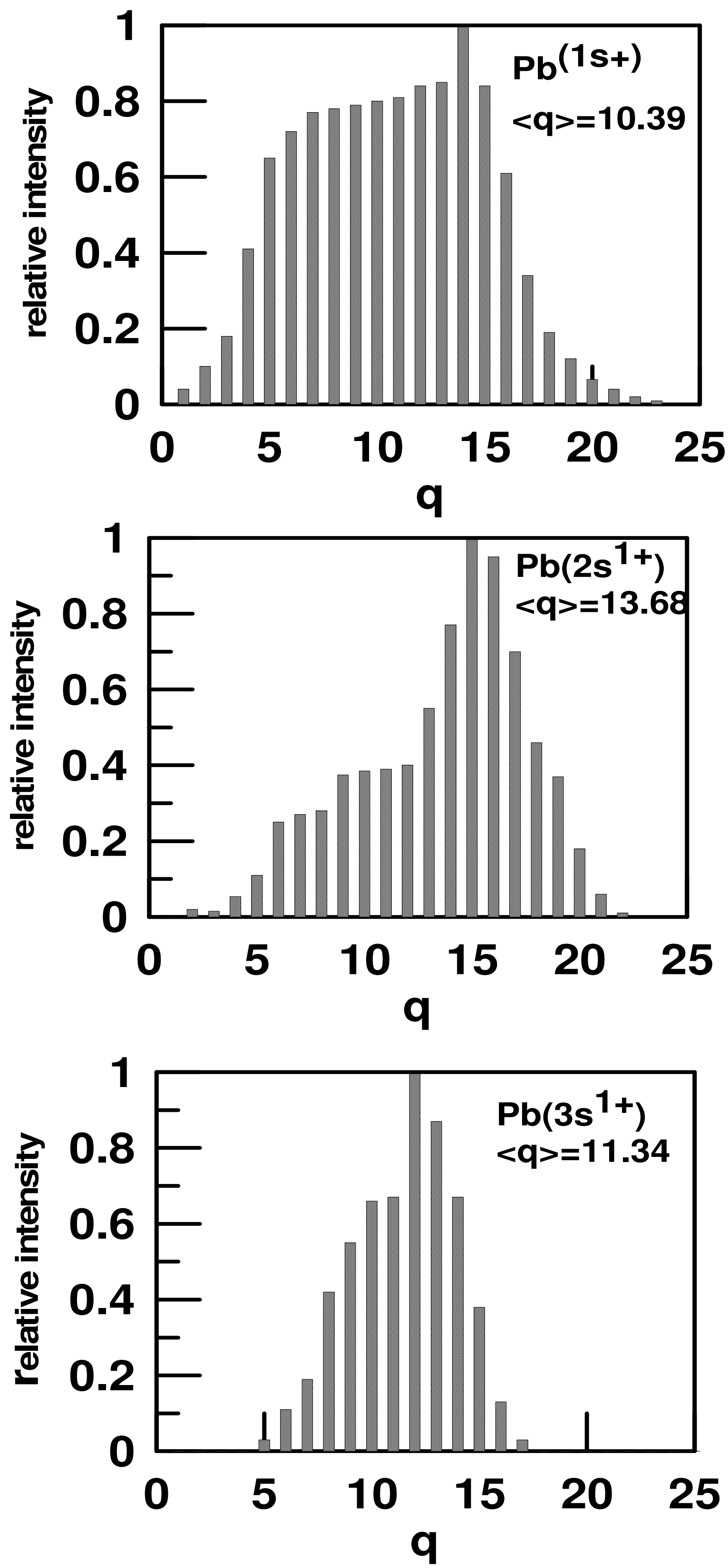

Figure 2: Ion charge state distributions (CSD) following de-excitation decay after K, L_I and M_1 subshell vacancy production in Pb atom.

Conclusion

An overview on ion charge state distributions (CSD) and mean charge state ions in the course of cascading de-excitation of inner-shell vacancies in Pb^{q+} are calculated. The computer program is made on base of tracing successive radiative, non-radiative transitions and electron shake-off process to filling the holes that exist in atomic level. Atomic data (radiative, non-radiative and electron shake-off) for single ionized Pb^{q+} are calculated with Multiconfiguration Dirac Fock (MCDF) wave functions and Dirac Fock Slater (DFS) wave functions. The electron shake-off process and closing of Coster-Kroing channels as result of energetically forbidden channels during cascade development are investigated and considered. Calculation results show a permanent increase of the number of ejected electrons with increasing complexity of atomic structure. The heavier atoms such as lead atom and the lower shell in which an initial vacancy is produced, the higher will be the average ion charge and the more complex will be the charge spectrum Thus, For atoms, a single ionization in inner shell is a possible method to create alternative to high energetic collision processes highly charged ions without high momentum transfer.

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