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Na 原子及类 Na 离子 Fe^{15+} 价壳层光电离过程的理论研究

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摘 要:采用相对论多组态 Dirac-Fock 理论,计算了 Na 原子及类 Na 离子 Fe^{15+} 基态的光电离截面.对 Na 原子,考虑相对论效应与弛豫效应,本文计算结果与前人计算结果和实验测量结果一致性较好.对 Fe^{15+} 离子的最外价电子,相对论效应对其光电离过程产生主要影响,而轨道弛豫效应则可忽略不计;对中性 Na 原子则情况完全不同:轨道弛豫效应的影响非常显著,但其光电离截面对相对论效应仅有非常微弱的依赖.

关键词:原子光学;光电离;截面;Cooper 极小;相对论效应;弛豫效应
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Theoretical Study on the Valence-shell Photoionization of Na Atoms and Na-like Fe^{15+} Ions

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Abstract: Calculations of photoionization cross sections for Na atoms and Na-like Fe^{15+} ions were carried out in the framework of relativistic multiconfiguration Dirac-Fock method. For Na atoms, comparisons were made with previous theoretical calculations and available experimental measurements and a good or at least reasonable agreement is found if the relativistic and relaxation effects are taken into account. It is shown that, for the outermost valence electrons of medium Fe^{15+} ions, the relativistic effects accounts for a prominent part in the photoionization process and orbital relaxation can be negligible; however it is not the same case for neutral Na atoms where the influence of orbital relaxations is significant but cross sections slightly depend on the relativistic effects.

Key words: Atom optics; Photoionization; Cross section; Cooper-minimum; Relativistic effect; Relaxation effect
OCIS Codes: 020.0020; 020.2930; 020.5580; 300.6210; 260.3230

0 Introduction

The interaction of photons with atoms (ions), i.e., the emission of an electron after the absorption of a photon, is one of the most fundamental atomic processes in nature that occurs in many areas of physics. Reliable knowledge of atomic important parameter values such as photoionization cross sections is crucial for both conventional and modern applications in science and industry. For example, photoelectron

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spectrum is a powerful tool to estimate element abundances, in both nearby and distant universe, with an accuracy limited by the accuracy of the relevant atomic data. The high-precision cross-section data are required for understanding fundamental aspects of the interactions occurring in stellar atmospheres, high-temperature environments such as stars, nebulae, controlled thermonuclear fusion reactors[1]. On the other hand, understanding the photoionization of highly charged ions, such as iron ions, is a premise for the interpretation of astrophysical observations of accretion sources such as active galaxies and the study is largely based on the investigation of photoelectron spectra[2-3].

For ab initio theories, alkali atoms as quasi-one-electron systems have proven to be an important and interesting testing ground for theoretical descriptions of the photoionization process. From the experimental point of view, alkali metals, being easy to evaporate, are a relatively convenient target for studies and have been investigated over the years[4-5]. Furthermore, alkali metals are very reactive; thus, the target densities have to be kept relatively low in order to avoid contamination of the experimental setup. High-resolution studies were started only in the last two decades with the advent of third generation synchrotron radiation sources[5]. Previously, ground state Na was studied employing high-resolution photoabsorption cross section[6] and photoelectron spectra[7], along with a variety of theoretical calculations[6,8-10].

In this paper, sodium atoms and Na-like Fe^{15+} highly charged ions were selected as test case for photoionization because they are simple open-shell systems for theory and removal of an outer valence electron gives only one final states $2p^6$ ($^1S_0$) which make them small enough that one has the possibility of treating the photoionization process completely, even including relativistic effects and relaxation of the bound-state electron density due to the photoemission of photoelectron. The present results are obtained by the fully relativistic Multiconfiguration Dirac-Fock (MCDF) method[11]. The comparison between the present calculations and the previous theoretical calculations[8,12-14] and available experimental measurements[4] show the capability of present method in understanding the near-threshold cross sections where a detailed theoretical description is difficult.

1 Theory

The Multiconfiguration Dirac-Fock (MCDF) method is used in the present work to calculate the photoionization cross sections and thresholds of Na atoms and Na-like Fe^{15+} highly charged ions. Usually, the MCDF method is a very effective many-body methodology since it includes the important relativistic effects and orbital relaxation, as well as the major correlation effects[5, 15]. In the present calculation, only electric dipole transitions were considered using the Babushkin gauge which in non-relativistic limit correspond to the length forms of electron-photon interaction.

In MCDF method, the radial charge density of nucleus is calculated based on the Fermi statistical distribution function[11]. The Atomic State Functions (ASFs) for an N-electron system are linear combinations of symmetry adapted functions, so-called Configuration State Functions (CSFs), expressed as

$$\psi(PJM)=\sum_r c_r\varphi(\gamma_r PJM) \tag{1}$$

where P (parity), J (angular momentum) and M are symmetry labels, γ_r contains all the quantum numbers necessary to distinguish states and the mixing coefficients c_r are determined by diagonalization of the Dirac-Hamiltonian. The CSF $\varphi(\gamma_r PJM)$ is linear combination of N-electron Slater determinants made up of one-electron functions.

For the photoionization of medium and heavy elements, it is well-known that the orbital relaxation between initial and final states is an important contribution to matrix elements, such relaxation effects are known to modify the cross sections by up to 30% or even more[5]. In MCDF approximation, the major parts of orbital relaxation can be properly taken into account by the virtue of a separate optimization of the initial atomic and final ionic states. Such a computational procedure leads to two sets of orbital functions for the initial and final states that are no longer orthogonal to each other. Moreover, in order to understand the importance of relativistic effects in photoionization process, we performed two sets of calculations using the relativistic Dirac-Coulomb Hamiltonian Eq. (1) of Ref. [16] and its nonrelativistic counterpart. Atomic units are adopted in this paper unless otherwise stated. In this system of units, the speed of light $c=137.036$. The nonrelativistic results can be obtained by enlarging the speed of light by a factor of 2000[11]. In order to ensure a proper nonrelativistic limit as well as the compare in a unified manner, the calculations are carried out using one-configuration: the initial configuration $1s^22s^22p^63s_{1/2}$ ($^2S_{1/2}$) and final configuration $1s^22s^22p^6$ ($^1S_0$). In this way, the role of relativistic effects will be elucidated in a consistent manner between relativistic and nonrelativistic calculations.

In the present work, the photoionization from the states $^2S_{1/2}$ of ground configurations Na and Fe^{15+} were considered, the process can be written as

$$2p^6 3s(^2S_{1/2}) + h\nu \rightarrow 2p^6(^1S_0) + \varepsilon l \quad (2)$$

where ε and $l = 1$ are the kinetic energy and orbital angular momentum quantum number of the outgoing photoelectron, respectively. The general expression of photonization cross sections from the initial state i to the final state f is given by[17]

$$\sigma_{if}^{PI} = 4\pi^2 a_0^2 \alpha \frac{df_{if}}{d\varepsilon} \quad (3)$$

where α is the fine structure constant and a_0 is the Bohr radius. The oscillator strength density $df_{if}/d\varepsilon$, given by Eq. (6) of Ref. [16], contains the dipole matrix element and describes the dynamics of the photoionization process, where the continuum orbitals have been calculated in an average potential of the core states by keeping the bound orbitals fixed. Numerical calculations of Eq. (3) were performed by using the Relphoto08 component, based on MCDF method and the corresponding package Grasp92[11] and developed by our group[15-16,18], in which the nonorthogonality of radial wavefunctions is included by following Löwdin's expressions[19].

2 Results and discussions

In this section, MCDF results for the 3s photoionization cross sections and thresholds of Na atoms and Na-like Fe^{15+} ions are presented. The photoionization cross sections of Na atoms as a function of outgoing photoelectron energy are shown in Fig. 1 together with the previous theoretical calculations and available experimental measurements. Solid line with open triangles is the results which come from present MCDF calculations in Babushkin gauge. The other theoretical results in length gauge are the Breit-Pauli R-matrix calculations[14] (dash-dotted line), the MCHF calculations[13] (dashed line), the MBPT calculations[8] (solid line), the polarized-orbital approximation (dash-double-dotted line) and exchange-adiabatic approximation calculations[12] (dotted line). The present MCDF cross sections are shown by the solid line with open triangles and the recommended experimental data by Hudson and Carte are displayed by solid line with closed circles[4]. One clear observation from Fig. 1 is that the photoionization cross sections do not change monotonously: for certain photoelectron energies local minima, i. e., Cooper-minima, can be seen in the cross sections. The nonzero minimum is due to the relativistic effects which results in the dipole transition matrix elements of the two final channels $\varepsilon p_{1/2}$ and $\varepsilon p_{3/2}$ to be zero at different energies and has been analysised through the photoionization studies of different atoms theoretically[14, 20]. Our calculation results agree well with the experimental measurements in low energy range[4], especially in the energy range below and at the minimum position. In the energy range above the minimum, the results of the Breit-Pauli R-matrix[14], MCHF[13] and polarized-orbital approximation[12] calculations are in better agreement with the experimental measurements than our calculations, despite the fact that the agreement between the results of the MBPT[8] and exchange-adiabatic approximation calculations[12] and our calculations is excellent.

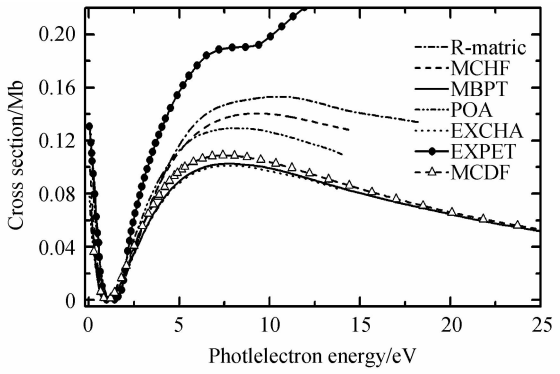

Fig. 1 Photoionization cross sections of the ground state $2p^6 3s(^2S_{1/2})$ of Na atoms

Although our present calculation agrees well with the experiment near the Cooper-minimum, it is possible that the experimental values are too high in the high energy range. As seen clearly, there is an abnormal bump in the experimental results higher than all the theoretical calculations. As already pointed out by Dasgupta and Han[12, 14], this bump could be attributed to the photodissociation processes of Na_2^+, which tends to increase the measured cross sections since the molecular photodissociation cross sections are much larger than the photoionization cross sections of Na atoms, even small amounts of molecular vapor could increase the experimental cross sections to a significant extent, and we believe that such experimental enhancement may also exist in other alkali-metal photoionization processes. On the other hand, the MCDF cross section is too low compared with the experiment in the high energy range. It is expected that more reasonable agreement will be obtained if relevant electron correlations can be included by allowing sufficient large multiconfiguration expansion. However, our main purpose is to investigate the influence of relativistic effects and orbital relaxation in the photoionization processes of Na atoms and Na-like Fe^{15+} ions, such a detailed calculation is beyond the scope of the present work. In particular, as shown in Fig. 1, the agreement manifests that single configuration approximation is enough to explain the observations in experimental conditions and then the discussion below is based mainly on single configuration approximation.

We now discuss the influence of relativistic effects in more detail. For light atomic systems, such as Na atoms, the relativistic corrections are relatively small as generally believed. Fig. 2 presents 3 s large component radial wavefunctions $P(r)$ (linear scale to the left axis) and small component $Q(r)$ (scale to the right axis) in the $2p^6 3s\ (^2S_{1/2})$ configuration of Na atoms. Solid lines are the relativistic results and dashed lines are the nonrelativistic counterparts, respectively. It should be noted that the thick lines are the large component radial wavefunctions. As expected, the relativistic large component has an oscillating structure consistent with that of the nonrelativistic counterpart completely, despite the fact that the difference between the two small components is very obvious as can be seen from Fig. 2. The relativistic (nonrelativistic) photoionization threshold with wavefunctions from single configuration approximation is 4.957 eV (4.951 eV) compared to the NIST value of 5.139 eV[21]. Note that the relativistic result is slightly higher than its norelativistic counterpart, i. e., the influence of relativistic effects is rather weak in this case. In fact, the main features of relativistic effects have been discussed in Refs. [5, 22] and this conclusion is also confirmed there: the relativistic contraction of inner electrons gives rise to a larger screening for the outer electron, then 3s radial density will expand. As expected, more accurate relativistic threshold, 5.145 eV, can be obtained if further multiconfiguration expansion is taken into account.

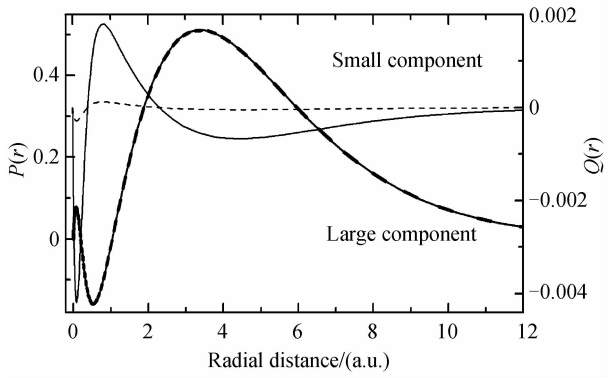

Fig. 2 Large and small component radial wavefunctions of 3s electron of Na atoms

Fig. 3 depicts the calculated photoionization cross sections of the ground states $2p^6 3s(^2S_{1/2})$ of Na atoms and Fe^{15+} ions. Solid lines are the results which come from present MCDF calculations, dashed lines are the corresponding nonrelativistic results and dotted lines are those from MCDF calculations without relaxation. It is evident that, for neutral Na atoms, all cross section curves agree with each other qualitatively and show a Cooper-minimum in low energy range. Although the difference between the relativistic and nonrelativistic small component radial wavefunctions is evident, relativistic effects do not produce substantial change in photoionization cross section. On the other hand, in order to investigate the influence of orbital relaxations, i. e., rearrangement of the bound-state electron density due to the photoemission of outer 3s electrons, it is useful to make a comparison of cross sections with relaxation and without relaxation. For this purpose, Fig. 3 also shows the cross sections when the orbital relaxations are not included in MCDF calculation, i. e., the bound one-electron orbitals of initial state are also used as the unperturbed one-electron orbitals of final state. Obviously, the difference between the photoionization cross sections with relaxation and without relaxation demonstrates that the orbital relaxation has notable influence on the cross section especially in low energy range, as well as on the photoionization threshold, i. e., such relaxation effects increase the threshold from 4.957 eV to 4.962 eV. This indicates that the one-electron orbitals of final ion do not collapse enough in unrelaxed calculations as generally accepted.

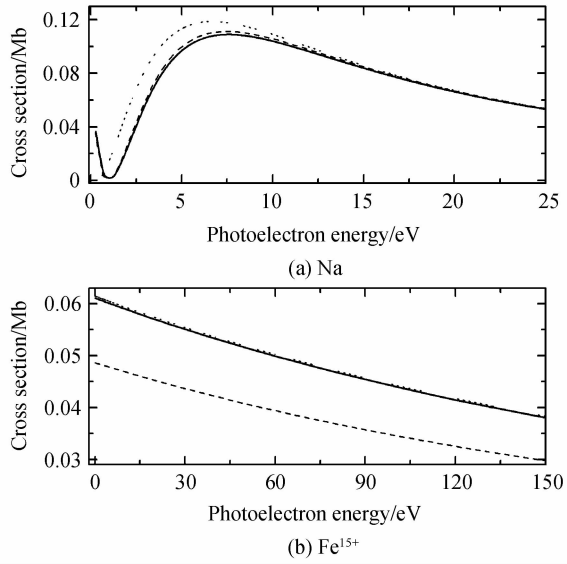

Fig. 3 Photoionization cross sections of the ground states $2p^6 3s(^2S_{1/2})$ of Na atoms and Fe^{15+} ions

For Na-like Fe^{15+} ions, as seen clearly, all cross section curves show a smooth, monotonically decreasing behavior. The nonrelativistic cross section, however, differs visibly from the MCDF calculations, whereas the cross section without relaxation matches the MCDF calculations almost perfectly. Then, one can conclude that, as generally expected, single configuration approximation suffices to describe the photoionization process of highly charged ions, or, vice versa, electron correlations has a negligible effect on the cross section. In addition, nonrelativistic threshold (485.033 eV) is found to be 0.78% below the

relativistic value (488.866 eV), while the calculated threshold without relaxation (488.924 eV) remains unchanged essentially. Thus, as mentioned above, the orbital relaxation is small and often leads to an increase of the photoionization threshold, the significant difference between the relativistic and nonrelativistic results demonstrates the importance of relativistic effects in the photoionization process of highly charged ions.

3 Conclusion

In the present work, the photoionization processes of neutral Na atoms and Na-like Fe^{15+} highly charged ions are investigated by relativistic multiconfiguration Dirac-Fock method. A detailed discussion is performed by analyzing the influence of relativistic and relaxation effects on the cross sections and thresholds. For Na atoms, a good or at least reasonable agreement between the present MCDF calculations and previous theoretical calculations as well as available experimental measurements is found if relativistic effects and orbital relaxations are taken into account. Our study provides clear evidence that, for the photoionization process of highly charged ions such as Na-like Fe^{15+}, the relativistic effects are very prominent but orbital relaxation can be neglected. This is in contrast to neutral Na atoms where significant influence of orbital relaxations can be found in the photoionization process but there is no essential discrepancy for the relativistic and nonrelativistic results.

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