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Study of *K*-shell Ionization Cross-sections of Atoms by Electron Impact*

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Abstract: In this paper, we give a description of recent progress on the measurements of electron-impact *K*-shell ionization cross-sections of atoms in the keV energy range. We present our experimental method of using thin targets with thick substrates and our measurements taken recently to improve the accuracy of the experimental data by an example of measuring *K*-shell ionization cross-section for Cr element within the incident energy of less than 26 keV. We also compare the *K*-shell experimental data sets available for 8 low-*Z* elements and 16 medium- and higher-*Z* elements in the keV energy range with some theoretical models and empirical formulae. The general comments on the status of measurement and comparison with theories for atomic *K*-shell ionization cross-section by electron impact are given.

Key words: electron impact; *K*-shell ionization cross-section; Rutherford backscattering spectroscopy; Monte Carlo simulation

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1 Introduction

Accurate atomic inner-shell ionization cross-sections by electron impact is of very importance not only in understanding the interaction of electrons and atoms but also in many applied branches such as in plasma physics, astrophysics, radiation physics and electron-matter interaction modelling and in quantitative analysis by electron probe (e. g., electron probe microanalysis (EPMA), Auger electron spectroscopy (AES) and electron energy loss spectroscopy (EELS))[1－3]. Up to now, the study of ionization cross-sections of atomic inner-shells by electron impact is still an interesting subject experimentally[4－11] and theoretically[8, 12, 13]. The accurate measurement of inner-shell ionization cross-sections poses numerous difficulties. In recent years, our group devoted efforts to the measurements of inner-shell ionization cross-sections of atoms by electron impact. We developed a method to measure the electron-impact inner-shell ionization cross-sections of atoms by using thin targets with thick substrates, which has the advantage of avoiding the difficulties of preparing self-supporting thin targets and has been applied to the measurement of inner-shell ionization cross-sections. Recently some experiments have al-

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so been taken to improve the accuracy of our data.

In this paper, we first review the recent progress in this research field. We then present our recently improved experimental method by an example of measuring *K*-shell ionization cross-section for Cr element within the incident energy of less than 26 keV. And we compare our present experimental data of Cr element and some existing experimental data sets with some theoretical models and some empirical or semi-empirical formulae. Finally, the general comments on the status of measurement and comparison with theories are presented.

2 Recent Progress

Powell has reviewed the status of early theoretical and experimental studies of this field[1,2]. Recently, he also reviewed the development of this field before 1996[3]. Therefore, in the present paper, we place our emphasis on the development of recent several years. In addition, we limit our discussion to the incident energy region less than 200 keV. This energy upper limit was chosen because inner-shell ionization cross-sections in the lower energy region are of interest both for many theoretical studies and for many applications (for example, typically about 3—25 keV for AES, 10—50 keV for EPMA, 50—200 keV for EELS)[3].

In 1990, Long et al[14] compiled for the first time the available experimental data for *K*-shell ionization cross-section by electron impact. Later on, Joy[15] compiled an electron-solid interaction experimental database, in which experimental data of *K*-shell ionization cross-sections were included. From above two databases, we can find that at that time the experimental data for *K*-shell ionization for many elements were scarce (even at present the *L*-, *M*-shell ionization cross-section data are still very/extremely scarce) and significant discrepancies between some data sets existed. Therefore, it was difficult to definitely assess the reliability of theoretical models based on the available experimental data. In recent years, some groups involved the measurement of inner-shell ionization cross-sections by electron impact[4—11]. Our group[4,16] made use of thin targets with thick substrates in our experiments. The method we adopted has the advantage of avoiding the difficulties of preparing self-supporting thin targets and has been applied to measure *K*-shell[4], and most recently *L*-shell[6], ionization cross-sections for some atoms in the energy region from threshold to several tens keV. Moreover, Llovet et al reported the measurements for Ni, Cr, Cu[7], Fe and Mn[8] elements in the energy region 6.5—40 keV, the uncertainties of relative cross-sections were of order of ～2%, and the uncertainties of absolute cross-sections were increased to ～10%. Their experimental data were compared with Hippler's[17] and Mayol and Salvat's[18] plane-wave Born approximation (PWBA) theories and Segui et al's distorted-wave Born approximation (DWBA) theory[8] as well as Casnati et al's empirical formula[19]. Most recently, their work was also extended to Lα production cross-section measurements for W, Pt and Au elements by 10—30 keV electrons[9], the targets also consist of very thin films of the studied elements deposited on thick carbon substrates and the experimental data were well corrected by Monte Carlo simulation. Shanker and Hippler[10] measured the *K*-shell ionization cross-sections of S element with uncertainty of 20% within the energy region of 3.5—14.0 keV and compared their data with Hippler's[17] PWBA theory and some empirical formulae. They also studied the effect of molecular environment on the *K*-shell ionization cross-sections of sulphur using SO_2 and SF_6 gas targets. Their experimental data did not show any molecular effect whereas Quarles and Estep[20] observed this effect. This is still an interesting problem that needs to be clarified. Moreover, Schneider et al[11] reported for the first time the absolute cross-sections for *K*- and L_3-shell ionization of Ag and Au targets by positron impact in the energy region of 30—70 keV and observed the difference between positron and electron impacts in this energy region. This difference was successfully explained with the Hippler's[17] PWBA theory in which electron exchange effect and the effect of the nuclear Coulomb field are taken into account. Most recently, the database for experimental K-shell ionization cross-section has been updated by Liu and An et al and is available from Internet[21]. From this new compilation, we can observe that the K-shell ionization cross-section data have been available in a wider range of elements and incident electron energy in comparison with the situation of about 13 years ago. In our recent study[4], we notice that further measurement of cross section data with higher accuracy (i. e., experimental error ~10%) is very desirable for testing theoretical models. Therefore, most recently, we have taken some measurements to try to improve the accuracy of our experimental data[16]. These measurements, which will be described in the following section, include: (1) the measurement of the thin target thickness with Rutherford backscattering spectroscopy (RBS); (2) the electron mean track length correction based on Monte Carlo method; (3) the detection efficiency calibration in the lower energy region using thick carbon target bremsstrahlung by electron impact.

Up to now, a lot of theoretical models[1-3], depending on the various treatment of incident and outgoing wave functions, atomic structure and the treatment of relativistic, Coulomb and electron exchange effects and so on, have been developed since the work of Bethe in 1930[22]. Powell reviewed the early status of this subject in his articles[1,2]. Khare and Wadehra[12], Luo and Joy[13] and Segui et al[8] have carried out the most recent quantum-mechanical calculations for inner-shell ionization cross-sections. In addition, Gryzinski[23] developed a most successful classical model for atomic ionization.

A large number of analytical formulae have also been proposed to represent the calculated and measured inner-shell ionization cross-section data[1-3]. This is because general theoretical calculation needs numerical solution, which is often time-consuming and difficult to give rise to simple analytical formulae whereas analytical formulae can be easily used in algorithms developed for the microanalysis. Many of empirical and semi-empirical formulae are modifications of the Bethe formula[22]. Some early formulae were proposed by many authors[1-3]. Recently, Hombourger[24] proposed an empirical formula by fitting expanded data sets. Deutsch et al[25] also improved the energy dependence in the low-energy regime of their early semi-empirical formula. By fitting the Llovet et al 's[7] most recently measured K-shell ionization cross-section data of Cr, Ni, and Cu elements in the incident energy region of 6.5—40 keV, we also modified the Deutsch et al's early semi-empirical formula[4]. At present, one of the most widely used empirical formulae in the energy region of interest here is Casnati et al's empirical formula[19]. As pointed out by Powell[2], we should notice that none of empirical and semi-empirical formulae might be used without hazard beyond the range of conditions for which they were initially developed.

3 Experimental Method and Data Processing

We briefly present our recently improved experimental method by an example of measuring K-shell ionization cross-section for Cr element within the incident energy of less than 26 keV. The other experimental details can be found elsewhere[4,16].

The electron beams from near threshold to several tens keV were provided by an electron gun and adjusted in accordance with X-ray counting rate. The thin targets used in our experiment were prepared in China Institute of Atomic Energy by evaporating elements of interest on thick aluminum substrates. The thickness of thin targets was thin enough to limit the energy loss of incident electrons less than ~1% of incident electron energy. The thicknesses of thin targets were measured by using RBS in our laboratory. 2 MeV $^4He^+$ ions were provided by an electrostatic accelerator with maximum terminal voltage of 2. 5 MV. The backscattering spectra were analyzed using a computer code GISA3. 3[26]. The thickness value of thin target determined by RBS technique was 5. 5 $\mu g/cm^2$ for Cr element. The uncertainty of thin target thickness determined by RBS was estimated to be about 5%.

The characteristic X-rays emitted from the target atoms were detected by an Ortec Si(Li) detector. The X-ray spectra were recorded by a PC computer-based Ortec multichannel analyzer MCA916. The efficiency calibration of this detection system in the energy region down to 0. 58 keV was performed using the thic*K*-carbon-target bremsstrahlung by 19 keV electron impact[27]. This method was proposed by Tschischgale et al[28] and Wolters et al[29] and was successfully used in the efficiency calibration of their Si(Li) detectors in the energy region down to ~0. 5 keV. In this method, the Wentzel's formula was employed for the theoretical thick target bremsstrahlung calculation, and the self-absorption correction and the convolution of detector's response function with the bremsstrahlung spectrum had also simultaneously been taken into account. The shape of the efficiency calibration curve was determined from the ratio of experimental and theoretical thick carbon target bremsstrahlung spectra, and the absolute value for the efficiency calibration was obtained by using ^{241}Am radioactive standard source. The accuracy for the efficiency calibration with this method was estimated to be ~6%. In fact, best model for the theoretical thick target bremsstrahlung calculation would be a full Monte Carlo calculation. PENELOPE is an excellent computer code system for Monte Carlo simulation of coupled electron-photon transport in arbitrary materials for a wide energy range, from a few hundreds eV to about 1 GeV[30]. Most recently, a new algorithm for the simulation of bremsstrahlung emission by fast electron[31], based on the most reliable numerical values of the shape function calculated by Kissel et al[32], has been implemented into the PENELOPE code. Our preliminary calculation shows that the theoretical bremsstrahlung spectra based on the Wentzel formula and PENELOPE code are apparently different in the energy region below ~4 keV. The efficiency calibration of our Si(Li) detector by using PENELOPE code is in progress, and we believe that it will improve the accuracy of the efficiency calibration in the low energy region and will benefit the ionization cross-section measurement of *K*-shell for lower-*Z* elements and of *L*-, *M*-shells for medium- and higher-*Z* elements, which is also in progress in our laboratory[6].

Moreover, the incident electrons will experience scattering and not penetrate straight through the thin target[7]. The mean track length correction of incident electrons was calculated using Monte Carlo EGS4 code[33], i. e. , the actual thin target thickness values were replaced by the energy-dependent electron mean track lengths in data processing. In addition, the measured cross-section data should be corrected due to the electron reflection from the thick substrate. The correction method has been described in Ref. [34]. But, to make a crosscheck, the electron reflection spectra needed in the corrections were calculated using EGS4 code instead of the previously used bipartition transport theory[34]. We found that the results from the electron reflection spectra calculated using EGS4 code and the bipartition transport theory were consistent within the accuracy of less than 4%. The estimated error from the correction method was also about 4%. The characteristic X-ray self-absorption correction can be negligible.

With the experimental procedure described above, we had performed the measurements for Cr element from the threshold energy up to ~ 26 keV[16]. The fluorescence yield was taken from the tabulation of Hubbell et al[35]. The adopted ratio of K_β to K_α intensity was the most probable value reported by Khan and Karimi[36]. Experimental uncertainty mainly comes from net peak counts (~3%), detection efficiency (~6%), fluorescence yield (~2%), target thickness (~5%), inhomogeneity of the target (~4%) and correction method (~4%). Therefore, the total uncertainty is estimated to be ~10%.

4 Comparison between Theory and Experiment

Firstly, we compare our present experimental data for Cr element with Hippler's[17] and Khare and Wadehra's theory (denoted as PWBA-C-Ex)[12], Luo and Joy's theory[13] and the results of Casnati et al's empirical formula[19] and Hombourger's empirical formula[24]. The comparison is shown in Fig. 1. Q_K, E_K and U_K represent the ionization cross section, ionization threshold and overvoltage for K-shell, respectively. The previous experimental data for Cr element[7, 34, 37] are also plotted. From Fig. 1, we can see that within the uncertainties our present experimental data are in good agreement with the results of PWBA-C-Ex theory and Luo and Joy's theory and also in reasonable agreement with the results of Casnati et al's and Hombourger's empirical formulae. We also observe that our present experimental data are in reasonable agreement with Luo et al's[34] and Llovet et al's[7] data for Cr element except He et al's[37] data. The same conclusion has also been obtained from the analysis of experimental data for Ti element, which were measured with the same method described above[16]. It indicates that our efforts to improve the experimental accuracy are very effective. We also observe that the data of He et al[37] are in general smaller than the other experimental data or theoretical predictions by approximately a factor of ~1.4, this might be attributed to the determination of target thickness by weighing.

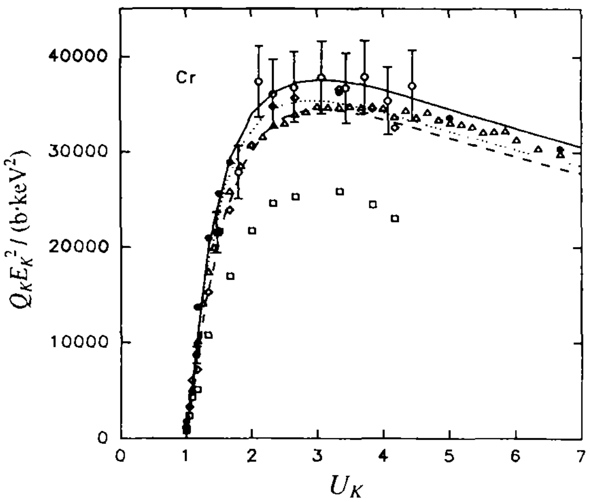

Fig. 1 Plots of $Q_KE_K^2$ versus U_K for Cr element.
○ denotes the present experimental data. ● Luo and Joy's theoretical result. △, □ and ◇ the experimental data for Cr element of Llovet et al[7], He et al[37] and Luo et al[34], respectively. —, --- and ··· the results of PWBA-C-Ex theory, Hombourger's and Casnati et al's empirical formulae, respectively.

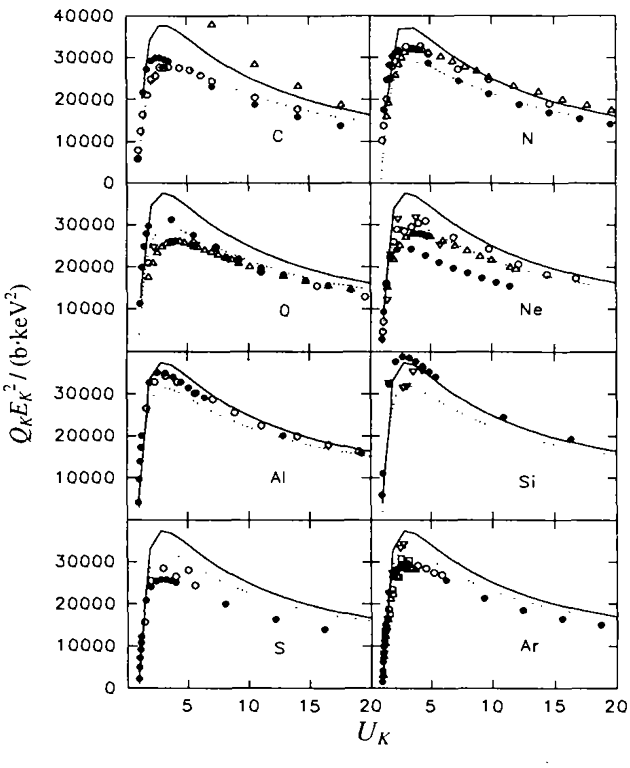

Fig. 2 Plots of $Q_KE_K^2$ versus U_K for C, N, O, Ne, Al, Si, S and Ar elements.
The experimental data for C, N, O, Ne, Al, Si and Ar are taken from the compilation of Liu and An et al[21] and the data for S are read from the paper of Shanker and Hippler[10]. All experimental data have been reevaluated based upon the fluorescence yield compilation of Hubbell et al[35]. ○ the experimental data. ● Luo and Joy's theoretical result. — and ··· the results of PWBA-C-Ex theory and Casnati et al's empirical formula, respectively.

In the previous review article of Powell[1,2], comparison has been made of some *K*-shell experimental data with many early theoretical models. Recently, Powell[3] and An et al[4] compared the theories of Luo and Joy[13] and of PWBA-C-Ex[12,17] with experimental data for some elements, and observed that for medium-*Z* elements the two theories can give almost the same results which are in good agreement with the experimental data. But, for low-*Z* elements PWBA-C-Ex theory overestimated the experimental data and Luo and Joy's theory showed better agreement. For higher-*Z* elements (for example, Ag element), the PWBA-C-Ex theory seemed to describe the experimental data better than Luo and Joy's theory. Khare and Wadehra[12] also compared their theory (PWBA-C-Ex) with experimental data and Scofiled's theory for some elements from Al to U in the energy region from ionization threshold to 1 GeV. Good agreement was obtained for *K*- and *L*-shell ionization cross-sections. Llovet et al made a comparison of their *K*-shell ionization cross-section data for Cr, Ni, Cu[7] and Fe, Mn[8] elements in the energy region of 6.5—40 keV with the Mayol and Salvat's theory[18], Segui et al's DWBA theory[8] and PWBA-C-Ex theory[12,17]. PWBA-C-Ex and Segui et al's DWBA theory can provide a good description both in shape and in magnitude for their experimental data. Based on the previous theoretical work, the theoretical models developed recently have made progress in the description of experimental data in wider valid ranges of incident energy and atomic number although some work still needs to be done to improve these models in some aspects.

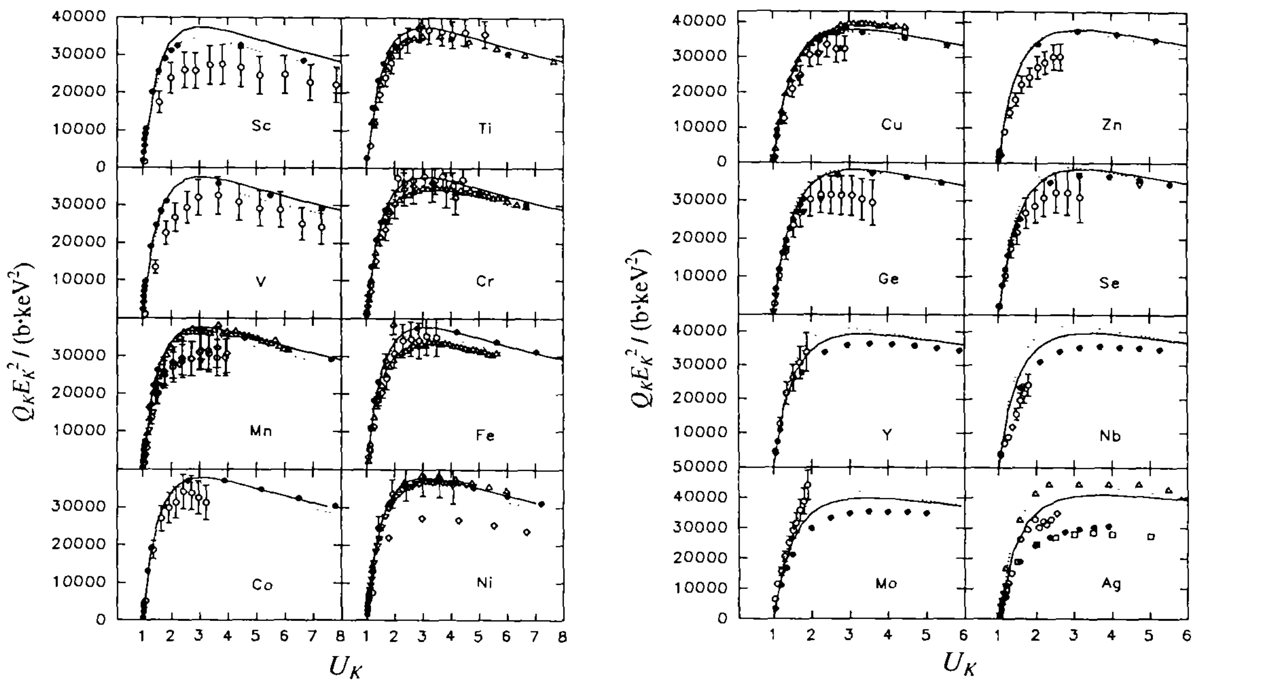

Fig. 3 Plots of $Q_K E_K^2$ versus U_K for 16 medium- and higher-*Z* elements from Sc to Ag (Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ge, Se, Y, Nb, Mo and Ag).

The experimental data are taken from the compilation of Liu and An et al[21] except that the data for Ag are read from Schneider et al's paper[11]. All experimental data have been reevaluated based upon the fluorescence yield compilation of Hubbell et al[35]. ○ the experimental data. The data measured by our group are plotted as symbols with error bars. ● Luo and Joy's theoretical result. — and ··· the results of PWBA-C-Ex theory and Casnati et al's empirical formula, respectively.

Shown in Figs. 2 and 3, we compare the *K*-shell experimental data sets available for 8 low-*Z* elements (C, N, O, Ne, Al, Si, S and Ar) and 16 medium- and higher-Z elements from Sc to Ag (Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ge, Se, Y, Nb, Mo and Ag) in the energy region of interest here with the theoretical models of Luo and Joy[13] and PWBA-C-Ex[12, 17] as well as Casnati et al's formula[19]. Our measured data are plotted as symbols with error bars. Except Cr element, our experimental data for other elements are taken from the compilation[21], which were measured before. All experimental data sets have been reevaluated based on the fluorescence yield compilation of Hubbell et al[35]. From the comparison between the theoretical models and experimental data sets, the conclusions stated before are confirmed. In addition, we also notice that in some cases the accuracy of our experimental data measured before is not so satisfactory, this is the reason why we are constantly trying to take measures, for example, described above, to improve the experimental accuracy. In general, we can find overall good consistency in these data sets, especially in the data sets measured recently by some authors, although discrepancies among some experimental data sets are still existing. We agree with the conclusion obtained by Powell[3] that this implies that the K-shell ionization cross-section data can be measured with combined standard uncertainties of about 10%. We also notice that the experimental data for many elements were measured by only one group in limited energy ranges and few data exist for the higher-Z elements. Therefore, additional measurements are still needed in a wider energy range, especially with an accuracy less than ~10% and for the higher-Z elements.

5 Conclusions

From the description above, some conclusions can be drawn as bollawing.

(1) We introduce our experimental method by an example of measuring the K-shell ionization cross-sections for Cr element in the energy region less than 26 keV. The measures taken here prove to be effective in the improvement of accuracy of experimental data.

(2) From some recent measurements, we conclude that the K-shell ionization cross-section data can be measured with combined standard uncertainties of about 10% although discrepancies among some experimental data sets are still existing.

(3) Additional measurements of K-shell ionization cross-sections are still needed, especially with higher accuracy less than ~10% and for the higher-Z elements. In addition, the measurements for L- and M-shells, which are more complicated and more difficult, should be paid more attention due to the very scarce available data and also as a stringent test for theoretical models.

(4) Within the uncertainty of about 10%, the experimental data of K-shell ionization cross-section in the energy region of interest here for low- and higher-Z elements are able to distinguish which theories developed in recent years are better in the corresponding element regions. Some theoretical work still needs to be done for improving the agreement between theories and experiments. In general, each theoretical model has its own valid range either in terms of incident energy, atomic number, or electron shell considered. It is the hope that we could have a theoretical model that has a wide valid range. The theoretical models developed recently have made progress in this aspect.

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电子原子碰撞 *K* 壳层电离截面研究*

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摘　要：近年来，研究组发展了一套利用厚衬底薄靶测量电子原子碰撞内壳层电离截面的方法. 该方法具有避免制作自支撑薄靶困难的优点. 最近，又采取了一些措施来提高该方法测量数据的精度. 首先评述了近年来该领域的一些研究进展，并通过测量 Cr 元素在小于 26 keV 能量范围 *K* 壳层电离截面的例子介绍了研究组的实验方法. 同时，也对 8 种低 *Z* 元素和 16 种中、高 *Z* 元素在 keV 能区的实验数据和理论模型、经验公式进行了比较，并对目前电子原子碰撞 *K* 壳层电离截面的实验测量和理论研究的现状及需进一步开展的工作进行了讨论.

关 键 词：电子原子碰撞；*K* 壳层电离截面；卢瑟福背散射；蒙特卡罗模拟

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