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Photoionization of helium above the $n = 2$ photoionization threshold ([†] *)

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Résumé. — L'étude des processus de photoionisation dans l'hélium a été entreprise entre 69 eV et 150 eV par spectrométrie d'électrons à l'aide du rayonnement synchrotron émis par l'anneau de stockage ACO. La probabilité de laisser l'ion résiduel dans l'état excité $n = 2$ a été mesurée en fonction de l'énergie des photons incidents. La section efficace absolue déterminée pour ce processus est en bon accord avec un récent calcul théorique utilisant une approche multiconfigurationnelle dans le cadre de la théorie des perturbations à N corps.

Abstract. — Using the technique of electron spectrometry and the synchrotron radiation of the storage ring ACO, the photoionization of helium has been investigated between 69 and 150 eV. The probability to leave the residual ion in the $n = 2$ excited state has been measured as a function of photon energy. The absolute partial photoionization cross section for this process has been determined and found to be in good agreement with recent Multiconfigurational Many-Body Calculations.

1. Introduction. — Photoionization of atoms may leave the residual positive ion either in the state of lowest energy (so-called ground state when the ionization takes place in the outermost subshell), corresponding to a final ionic state in which a single electron has been ejected from an inner or an outer shell, or in an excited state of higher energy in which at least a second electron has been promoted to some empty orbitals. The production of these excited states of the ion through photoionization has been considered as one of the most illustrative examples of the breakdown of the one-electron model of the atom, since the corresponding processes are strictly forbidden in the independent particle theory; thus they are an explicit manifestation of electron correlations. The various terms used in the past to denominate this kind of processes, like monopole transitions, shake up and

conjugate shake up processes, two-electron transitions, simultaneous excitation and ionization, refer only to various degrees of approximation in the theoretical approach of the problem, but it should be kept in mind that the process to be described is not a two-step process [1]. In fact, any distinction between the various ways the ground state and the excited states of the ion are produced represents only an inadequacy in our way of representing exactly the wave functions of the initial state and of the final state of the system [1, 2, 3]. It is possible to explain the origin of all excited states in a unified approach by considering the N -particle system; in this view, transitions from the initial state to the various excited states of the ion occur exactly the same way as to the ground state of the ion [1-3].

One of the best ways to study experimentally these photoionization processes is to use the technique of photoelectron spectrometry. By measuring the energy and the numbers of photoelectrons ejected from an atomic species by monochromatic radiation, one has a direct access to the various final states of the ion, since each of these final states corresponds to a different kinetic energy of the outgoing electron. Thus, photoelectron spectra produced in this case show generally intense peaks, usually called main peaks, related with

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a final ionic state in which a single hole has been created in the various inner or outer shells, without any additional excitation energy transferred to the ion, and several peaks, generally of lower intensity, on the low kinetic energy side of the main peaks, that are called satellite lines, corresponding to transitions in which another hole has been created in the same or in a different subshell. This is true only when correlations are small enough to have no dramatic effect on the single hole spectrum. An example where this condition is not fulfilled is the photoionization in the 4p shell of xenon, where the photoline corresponding to the creation of a single hole in the $4p_{1/2}$ subshell disappears [4]; this is sometimes referred to as *collective effect* [5], and is a good example of strong correlation effects.

From the theoretical point of view, the configuration interaction method has been mostly used to study these satellites, also called correlation satellites [1, 6, 7]. In this approach, the origin of these satellites has been assigned to Initial State Configuration Interaction (ISCI) and Final State Configuration Interaction (FSCI). Furthermore, FSCI can also be divided in two categories, if one excepts the resonant process of autoionization : Final Ionic State Configuration Interaction (FISCI), which takes into account only the $(N - 1)$ electron system of the residual ion, and Continuum State Configuration Interaction (CSCI) or Interchannel Coupling, which takes into account the total N electron system in the final state, by introducing the interaction between the photoelectron in the continuum and the $(N - 1)$ electrons of the residual ion [1, 3]. In view of the energy dependence of the relative intensity of these satellites, FISCI should give rise to features completely independent of photon energy, ISCI to processes possibly varying with photon energy, while processes originating from CSCI should depend strongly of this parameter, since the coupling between the electron in the continuum and the residual ion is *a priori* important only for low kinetic energies of the photoelectron. Thus, a study of the energy dependence of the correlation satellites is expected to yield valuable information about the origin of the various satellites.

From the theoretical point of view, helium is a good case to study these processes : since the wave functions of the single electron in the final state of the ion are exactly known, only ISCI and CSCI can play a role. It is also a convenient case from the experimental point of view, since the number of satellites is relatively small : there is no possible mixing between photoionization processes originating within different subshells. Thus, a careful study of the variation of the relative and absolute intensities of the satellites at low photoelectron energy, starting as close as possible to threshold, should allow us to delineate the importance of Interchannel Coupling.

When the helium atom is ionized by photons of increasing energy, it may be left in any of the states

of He^+ ($n = j$), $j = 2, 3, 4, \dots$, as soon as the photon energy is greater than the corresponding j threshold : $j = 2$ at 65.4 eV ; $j = 3$ at 72.7 eV ; $j = 4$ at 75.4 eV ; ... The double ionization threshold is at 79.0 eV. The study of correlation satellites in helium requires a high flux of monochromatic radiation, because the cross section to produce He^+ in these various states is rather low (less than 1 megabarn for the $n = 1$ state in the region of the $n = 2$ threshold, less than 0.1 megabarn for the $n = 2$ state ; furthermore these values are decreasing rapidly with increasing photon energy). Few experimental results are available at this time ; the relative probability to leave the helium ion in the $n = 2$ states has been measured at one energy close to threshold (67.4 eV) by Samson [8], using an emission line from multiply ionized ions as the source of radiation, between 110 and 1 500 eV by Krause and Wuilleumier [9, 10] using characteristic soft X-ray lines, and at 1 487 eV by Carlson *et al.* [11] using the $K\alpha$ line of aluminum ; some relative values for producing the $n = 3$ states have also been obtained [10]. The results of two theoretical calculations predicting a different behaviour for the intensity of the $n = 2$ satellite were also available when we started our experiments [12, 13].

These processes were also analysed in (e, 2e) experiments [14], but, in this case, the correlation effects concerned differ from those that are important in photoionization, and a direct comparison between the two methods is not straightforward.

Here, we present the results of an experimental investigation performed between 70 and 150 eV using synchrotron radiation as the excitation radiation.

2. Experiment. — The experiments were carried out using the monochromatized synchrotron radiation of the ACO storage ring in Orsay, with a cylindrical mirror analyser (CMA) to analyse the photoelectrons. The CMA was used with its axis parallel to the incident radiation, which is an experimental set up giving independence from the state of polarization of the incident radiation [15]. The analyser accepted electrons about the magic angle of $54^\circ 44'$, which makes the results independent of any angular distribution effect. The resolution of the CMA was set to about 0.9 %. A detailed description of the apparatus and of the operating procedure can be found elsewhere [16, 17].

Because of the theoretical importance of the problem and of the rather difficult experimental conditions, three sets of completely independent measurements were taken, using different optical systems for the beam line and for the monochromator.

In the first series of experiments, the CMA was mounted behind the exit slit of a grazing incidence Rowland Circle monochromator [18] attached to the beam line A8 of LURE. At that time, a cylindrical mirror was used inside the beam line to deflect the radiation coming from the ring to the monochromator. With this system, the maximum photon flux routinely

Fig. 1. — Typical photoelectron spectra following ionization of helium taken in the first set of experiments, with the Rowland Circle monochromator and a cylindrical mirror in the beam line A8 of LURE. The counting rate on the $n = 1$ photoline is, at most, 150 counts/s, but was, usually, less than 100 counts/s.

available was about 3×10^9 photons/s in a 1% bandpass. An example of spectra obtained in this configuration is shown in figure 1, around 80 eV photon energy. The maximum counting rate was 150 counts/s for the $n = 1$ ionic state photoline and less than 6 counts/s for the $n = 2$ excited state line. The photon flux was too low to observe higher excited states. However, it should be noted that the signal to noise ratio was good enough to allow a quantitative determination of the relative intensity of the $n = 2$ peak, in spite of the low counting rate.

In the second set of experiments, the same monochromator was used, but the first mirror in the beam line was replaced by a toroidal mirror, increasing the photon flux available in the source volume of the CMA by a factor 3 to 4. In addition, at that time, all optical surfaces were coated with platinum as frequently as possible. Under these conditions, we obtain photon fluxes up to 2×10^{11} photons/s, as the maximum value in 1% bandpass [19]. However, since this Rowland Circle monochromator is operated in the 10^{-6} torr range, the three optical surfaces (pre-focussing mirror, grating, plane mirror at the exit slit) are quickly contaminated by deposition of hydrocarbon layers, which reduces this flux by a factor about 10 in routine operating conditions. Since the grazing angle is continuously variable in this Rowland Circle monochromator [18], we were able to cover the whole energy range of interest, up to 150 eV, in the best experimental conditions. At the highest photon energies of the domain, the excited states $n = 3$ and $n \geq 4$ were observed, as can be seen in figure 2 which shows an example of spectra obtained in this configuration. The signal to noise ratio was not as good as in the preceding case, because the channeltron was operated with a positive voltage on the cone, in order to correct directly for a possible discrimination of the

Fig. 2. — Examples of photoelectron spectra obtained with the Rowland Circle monochromator and a toroidal mirror in the beam line A8 of LURE. The two extreme photon energies investigated in this second set of experiments are presented here : 74.0 eV and 150.3 eV. At 74 eV, the insert shows the $n = 2$ photoline after background subtraction. Between 100 and 150 eV photon energy, higher excited states ($n \geq 3$) have been observed and their relative intensity has been determined. Typical counting rate of 250 counts/s at low photon energies have been obtained routinely in this series of measurements.

channeltron efficiency against electrons of different kinetic energies.

In the third set of measurements, the electron spectrometer was mounted behind a 3 meter toroidal grating monochromator [20]. The monochromatized light was refocussed in the source volume of the CMA with a toroidal mirror placed after the exit slit. This monochromator is operated in the 10^{-9} torr range and the high photon flux initially observed (about 7×10^{11} photons/s, within a factor 2, in a bandpass of 0.6 eV at 75 eV with a toroidal grating mounted for about eighteen months), did not decay because of hydrocarbon contamination. The increased efficiency enabled us to use lower bandpass (in the 0.3 eV range), lower pressures in the source volume (about 10^{-4} torr) and shorter counting times. An example of the spectra obtained in this configuration is shown in figure 3. Effective counting rates are 5 000 counts/s for the $n = 1$ peak and 150 to 200 counts/s for the $n = 2$ photoppeak. Because of the high photon flux, we were able

Fig. 3. — Typical photoelectron spectra taken with the toroidal grating monochromator in the low energy part of the domain under investigation. A smaller band pass of the monochromator could be used (in the 0.3 eV range) while still providing an electron counting rate of about 5 000 counts/s on the $n = 1$ photoline and of 150 to 200 counts/s on the $n = 2$ photoline. The upper part shows the spectrum at the lowest photon energy (69 eV) we have worked in these experiments.

also to obtain data at lower kinetic energies for the satellite line. In fact, we have measured the relative intensity of the $n = 2$ satellite line down to a kinetic energy of 3 to 4 eV. Even smaller bandpasses could be used, with this monochromator, but since we had access to it for a short period of time, we only had time to check the consistency of these data with our earlier measurements.

3. Results and discussion. — The relative intensity of the satellites was determined as the ratio of the respective area under the corresponding photolines, after background subtraction and dispersion correction. Since the photoelectron lines corresponding respectively to the $n = 1$ and to the $n = 2$ states of He^+ are 40.8 eV apart, a careful study was carried out of all effects leading possibly to discrimination by the apparatus against electrons of different kinetic energies (as different as 4 and 44 eV in the worst case). We carefully checked the transmission of the electron spectrometer, the effect of scattering of the photoelectrons on their way from the source volume to the

channeltron (pressure effect), and the detection efficiency of the channeltron. The calibration curve of the channeltron was determined, in particular, for kinetic energies of the photoelectron corresponding respectively to the $n = 2$ and to the $n = 1$ states of He^+ . These curves allowed us to correct the data of the first set of measurements (taken with zero voltage on the channeltron cone, because of the low photon flux and the higher background when it was operated with a positive voltage) as well as to check the consistency of the two other sets of data taken with various positive voltages on the cone. At high flux levels, the main source of error comes from this correction and not from the statistical errors.

The relative probability of leaving the He^+ ion in the $n = 2$ state to the one of leaving the ion in the $n = 1$ state is shown in figure 4, as extracted from our

Fig. 4. — Variation of the ratio $\text{He}^+(n=2)/\text{He}^+(n=1)$ as a function of the photon energy. All synchrotron radiation data are from this work : $\circ$ values determined with the RCM and the cylindrical mirror; $\bullet$ values determined with the RCM and the toroidal mirror; $\blacksquare$ values obtained with the TGM. $\square$ Result obtained by Samson close to threshold, using a line from multiply ionized ions (ref. [8]); $\triangle$ previous values measured by Krause and Wuilleumier, using characteristic X-ray lines (ref. [9]); ∇ theoretical value extrapolated at threshold by Fano *et al.* (ref. [33]). The solid curve is the result of ISCI calculations (Salpeter and Zaidy, ref. [12]), the dashed curve was obtained in close coupling calculations by Jacobs and Burke, reference [13].

analysis of the data. In general, our three sets of results are in good agreement, taking into account the different limits of error. Except for the point at 110 eV photon energy, our new results are compatible with the previous set of data taken with soft X-ray lines [9]. The discrepancy at this photon energy might be due to an overcorrection of the channeltron efficiency

effect for the data taken with the Be $K\alpha$ line [9]; it should also be kept in mind that this X-ray line is, in fact, a broad band (about 5 eV FWHM) and, in this case, it is difficult to accurately determine the underlying background for the $n = 2$ photoelectron line.

The two published theoretical results are also shown on the figure. The curve calculated by Salpeter and Zaidy [12] only takes into account initial state correlations of the atom; they used a highly correlated Hylleraas type wave function for the initial state and hydrogenic wave functions for the final state, neglecting any possible coupling between the photoelectron and the residual positive ion. The second curve, calculated by Jacobs and Burke [13], exhibits a broad maximum in this energy range; they explicitly took into account this coupling between the photoelectron and the ion, while using also a highly parametrized wave function for the initial state and a close coupling expansion for the final state. Our results do not confirm the existence of a broad maximum. They are rather suggesting a kind of plateau, starting from threshold, followed by a smoothly decreasing relative probability at higher photon energies (the slope of the curve depends, of course, drastically on the energy scale chosen for the graphical representation). This behaviour is similar to the one observed recently by Woodruff and Samson, with a quite different technique [21]; using the synchrotron radiation of the Stoughton storage ring, they observed the fluorescence light emitted in the decay of the $n = 2$ excited state of He^+ to the ground state. Their determination of the relative probability of creating the state $n = 2$ is only relative, but, by normalization to our data, a reasonable agreement seems to exist between the two new sets of data [21].

Even more interesting than the cross section ratio is the variation of the absolute cross section for leaving He^+ in the $n = 2$ state. To determine such a partial cross section, especially when it is weak compared to other cross sections in the same atom, all contributions to the total photoabsorption cross section have to be known on a relative scale. Our results give the relative contribution for ionization processes leaving He^+ in various excited states. For the $n = 3$ state, we adopted a relative value $\text{He}^+ (n = 3)/(n = 1)$ of 2(1) % up to about 100 eV and 1.5 (5) % between 100 and 150 eV, as determined in some of our experiments, in agreement with data previously obtained in Oak Ridge at 132 eV and 151 eV. For the higher excited states ($n \geq 4$), we adopted the value measured here at 150 eV, with a large error limit : 1(1) %, to take into account the lack of data at low photon energy. Finally, for the double ionization cross section, we extracted its relative contribution from a critical analysis of the data obtained with synchrotron radiation [22], with the pseudo-photon method [23], and with some characteristic X-ray lines [24]; the actual values of the relative probability of double ionization range from 0 % at threshold up to 4 % at high photon energy.

Using the total photoabsorption cross section as tabulated by West and Marr [25], we obtained the results shown in figure 5. On this absolute scale, the partial photoionization cross section for leaving He^+

Fig. 5. — Variation of the absolute value of the partial cross section for photoionization of helium leaving the residual ion in the $n = 2$ state. All data points are from this work. — ISCI calculations, Salpeter and Zaidy, reference [12]; - - - - close coupling calculations, Jacobs and Burke, reference [13]. The dash-dotted curve — · — · — is the result of a quite recent calculation by Chang (ref. [26]).

in the state $n = 2$ decreases continuously from threshold, as do the two theoretical calculations already mentioned. Our data remain between both curves at low energy, reaching the values of Salpeter and Zaidy at higher photon energies. However, our data agree best with a recent calculation by Chang [26], who used a multiconfigurational Many-Body approach.

In comparing experimental and theoretical results, it should be kept in mind that there are, in fact, two possible final states, almost degenerate, for the $n = 2$ state of He^+ : the $\text{He}^+ 2s$ and $\text{He}^+ 2p$ configurations are separated only by 5×10^{-4} eV and are undistinguishable in this type of experiments. While Salpeter and Zaidy found that only the $2s$ final state is created with some probability, Jacobs and Burke obtained a threshold ratio of 3 : 1 for the probability of leaving the ion in the $2p$ final state compared to the probability of leaving the ion in the $2s$ final state; this ratio decreases continuously with increasing photon energy [13]. The answer to this question could come from angular distribution experiments, since the ϵp electrons, corresponding to the $2s$ final state, are expected to present an angular behaviour different than that of either ϵs or ϵd electrons. Such measurements begun between 150 and 110 eV with characteristic X-ray lines and the results did not follow completely the close coupling predictions at 110 eV, suggesting already a possible deficiency in the theory [9]. However, the main region still to be investigated is the 65-110 eV region. There is now some hope, with the availability of high throughput grazing incidence monochromators [20], to achieve such an experiment in the near future. It is also possible to separately measure the ratio $\text{He}^+ (2s)/(2p)$ in a beam experiment, by observing the radiative decay of $\text{He}^+ (2p)$ in the ionization

region and of He^+ (2s) in an adjacent quenching region.

From a theoretical point of view, it should be noted that electron impact excitation [27] of the $n = 2$ state of He^+ ion yielded experimental cross section values less than those provided by the close coupling calculations [28]. Further refinements in the theory, which included more pseudo-states in the close coupling expansion, lowered the theoretical values and brought them in better agreement with experiment [29]. However the new approaches of Chang, using a multiconfigurational description for the initial and the final state [26], and of Starace, using the hyperspherical coordinates [30], seem to be more promising.

Finally, we would like to point out that, in the energy region between 70 and 75 eV, there are autoionizing doubly excited states of the $(sp, 3n+)$ and of the $(sp, 4n+)$ resonance series. These states were observed a long time ago by Madden and Codling [31] in photoabsorption, and more recently in fluorescence experiments by Woodruff and Samson [21]. In this energy region, the total photoabsorption cross section is strongly affected by the competition between the direct ionization process and the simultaneous excitation of the two electrons into some levels of these series, followed by autoionization of the excited atom into one of the several channels that are available in the continuum. This competition gives rise to interference effects seen in the Fano profile of the absorption lines. This effect might occur, although to a lesser extent, up to the double ionization threshold (79.0 eV). At some photon energies, where the cross section is

varying rapidly within the small bandpass of the monochromator, we have observed a double peak for the $n = 2$ photoline [32]. Even when the $n = 1$ photoline appears as a single peak in this energy region, the ratio $n = 2/n = 1$ (He^+) has been found to deviate sometimes from the *continuum* branching ratio, as determined from the values obtained outside, but very close to this energy region [32]. Thus, we have included in figure 4 only those values that we believe are not much influenced by the resonances, taking into account the positions of the resonance lines relative to the photon energy, and the shape and the width of the autoionizing lines [21] compared to the bandpass of the monochromator. A detailed study of this energy region is in progress, with a smaller bandpass of the monochromator.

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