Recent Results on Ion-Atorn Collisions at Energies Within the MeV Range

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A selection of recent results on charge-changing collisions involving hydrogen and helium as both projectiles and/or targets, at impact energies within the MeV range, is presented and discussed having as background the development of new ideas and experimental techniques in this very active field of Atomic Physics.

I. A brief survey of the historical background

I-a. The early results

This survey is not intended to be either exhaustive or historically complete. It is here simply to stress the sense of continuity and to give the reader a glimpse of the most relevant literature.

The study, both experimental and theoretical, of basic processes in energetic ion-atom collisions has been the subject of many recent publications, despite the seventy-year-old history of the theme. The growth of our knowledge of charge-changing processes along the past seven decades has not been a particularly smooth function of the time. On the contrary, it has been somewhat spasmodic.

Alpha-particles emitted from natural radioactive sources furnished the first energetic ion beams where charge-(hanging processes were quantitatively observed^[1]. Equilibrium fractions for He⁺ in a beam of MeV-alpha-particles emerging from thin foils were measured by Rutherford^[2] in the early 20's. Theoretical estimates of the cross sections for electron capture date bacli to the calculations of Fowler^[3] and Thomas^[4] in the framework of classical physics. Soon later, Oppenheimer^[5] and Brinkman and Kramers^[6] employed the newly discovered quantum mechanical theory to construct what became known as the OBK approximation Proton beams were first used for experimental woik on charge exchange by Bartels^[7]. In this same year, Bethe's^[8] important contribution to the wave-mechanical theory of the inelastic scattering of

charged particles appeared. These pioneering efforts culminated with the publication, in 1933, of the classical book of Mott and Massey^[9], "The Theory of Atomic Collisions".

This promising enthusiasm was cooled down by the irresistible rise of nuclear physics which made atomic collision physics to remain comparatively dormant for the next twenty years. In this period the experimental work was essentially focused on the problems of hydrogen and helium collisions with gaseous targets (mainly H_2 , O_2 , N_2 and noble gases) within an energy range seldom exceeding a hundred of keV. However, the interest in the theory of electronic stopping in solids continued, now including the energy loss of heavy ions (even fission fragments). The energy-loss mechanisms that dominate the stopping of heavy particles at MeV energies are usually excitation and ionization. They were extensively analyzed by Bohr^[10] in an outstanding paper which appeared in 1948. In his paper Bohr established criteria for the relative importance of the different processes together with approximate scaling rules. He used a classical free collision approximation to calculate cross sections which, by the use of these scaling rules, can be extended to cover a wide range of collision partners and relative velocities.

Just after the World War II, Van de Graaff accelerators became available in many nuclear physics laboratories around the world substituting the old Cockroft-Walton accelerators which were then appropriated by the experimental atomic physicists. Charge-changing and stopping power studies within a new energy range became possible and it was soon noticed that the interpretation of experiments was easier at the moderately higher energies. A summary of the experimental situation until 1958 is given in a review paper by Allison^[11].

The revival of the theoretical activity in atomic collisions was due to the Belfast school (D.R. Bates, G.W. Griffing, A. Dalgarno, B.L. Moisiewitsch among others) which was responsible for a series of illuminating papers published during the fifties. A very important result obtained by Bates and Griffing^[12] at this occasion was to recognize that in the ionization of a target atom by a projectile carrying electrons (dressed projectiles in contrast to bare nuclei) the incident electrons may be more than passive spectators, being true actors playing the role of ionizing agents. This was an early example of the importance of electron correlation in ion-atom collision systems, a matter that is receiving a great deal of attention in the last years and that is one of the points of interest of this paper. Another fundamental result, this time in the electron capture theory, was obtained by Bates and McCarroll^[13]. They recognized that the description of the electron capture process, in the framework of Massey's perturbed stationary states molecular model^[14], requires the inclusion of electron translation factors in the eigenfunction expansions in order to give results that are independent of the choice of the origin. These factors taking account of the momentum associated with the translational motion of the active electron are essential at high projectile velocities (when the collision velocity exceeds the velocity of the electron to be transferred) reducing significantly the capture cross sections. This important improvement was incorporated by all close-coupling calculations ever since. A further improvement in the theoretical description of the electron capture was to consider the influence of the long-range Coulomb potentials of the projectile and target nuclei on the transfer process^[15]. This is at the origin of the continuum distorted-wave approximations.

I-b. The more recent developments

Closing the fifties, two important achievements are related to the description of inner-shell ionization by massive charged particles: the plane-wave Born approximation (PWBA) was applied by Merzbacher and Lewis^[16] and Bang and Hansteen^[17] introduced the semiclassical approximation (SCA). They were the source of inspiration for very many publications appearing till now. As it is well known, it was shown later that the two approaches are equivalent in the calculation of total cross sections^[18]. At this point it is worth mentioning the relevant contributions of Brandt and collaborators^[19] and of Kocbach^[20] to the PWBA and SCA formulations, respectively. The incorporation of a number of corrections in the original formulations permitted the theory to attain a high degree of precision in the description of the experimental data.

A great incentive to sustain the interest in inner shell ionization was the development of analytic techniques for characterization of materials by particleinduced X-ray emission (PIXE)^[21] which demanded precise determination of the relevant cross sections. Other branches of physics where ion-atom or ion-solid interactions play a fundamental role and which began to experience a fast rise in the 60's, stimulating both theoretical and experimental investigations in basic collision processes were: astrophysics, plasma physics, controlled thermonuclear fusion, physics of the atmosphere, ion implantation, radiation damage, and radiation detectors and accelerators design.

It is now time to speak about some experimental developments that very much broadened the field of investigations in atomic collisions. They are related to ion sources and beam preparation, ion accelerators, charged-particle and X-ray detectors, and data acquisition. In the late 50's the Van de Graaff accelerators constructed for nuclear physics research became somewhat outmoded for their original purposes and began to be used for atomic collisions within the MeV-energy region. At these kinetic energies, the projectile velocity is a few tens of the Bohr unit, αc , (v $\simeq \sqrt{40E}$ where v is given in terms of the Bohr velocity, and E in MeV per nucleon; in this paper the velocity will be always given in this unit). In the next sections we will concentrate our attention on processes occurring at a few MeV per nucleon (v \gg 1).

Also during the sixties, experimental physicists start to migrate from nuclear to atomic physics, bringing with them their experimental apparatuses and techniques. The influx of so many new ideas, new experimental facilities and new people was, to a great extent, responsible foi the explosive development of the physics of the atomic collisions in the next two decades as witnessed by the increasing number of contributed papers in the biennia International Conference on the Physics of Electronic and Atomic Physics (ICPEAC). The interest deviated rapidly to the new area of inner-shell processes mostly because these neophytes newly converted to Atomic Physics felt themselves much more comfortable in experiments where MeV-beams were sent on a solid target and the resulting X-rays were analyzed with Si-Li detectors. The trajectory of almost all nuclear physicists arriving at Atomic Physics passed through this entrance. The case of our own laboratory^[22] is a typical one.

When a fast beam of projectiles with initial charge q passes through matter and a projectile \mathbf{P} collides with a neutral target \mathbf{T} , different inelastic processes may occur. These include target or projectile excitation, and capture of one or more electrons of the target by the projectile. They can be represented by

$$P^q + T \rightarrow P^p + T^i + (p+i-q)e^-$$

Generally the indices q, p and i can assume values ranging from zero (or -1 when a negative ion can be formed) to Z_P or Z_T , the atomic numbers of the projectile and of the target, respectively. The number of electrons released in the process is (p + i - q). Often several of these processes take place in a single collision as, for example, the loss (ionization) of an electron by the projectile accompanied by the excitation or the ionization of the target. To the process represented above corresponds a total cross section σ_{qp}^{i} . The measurement of this cross section requires the simultaneous observation of both the projectile and the target charge states, i.e., a coincidence experiment. The first experiments of this type^[23] were reported in 1965. When only the projectile final state is identified the measured cross section is the sum over the final target states, $\sum_{(i)} \sigma^i_{qp} = \sigma_{qp}$

However, the charge states are not enough to fully specify the process. For instance, collisions involving neutral target atoms and highly-stripped projectiles have many open channels for capture: any electron from the target can be transferred to the many vacant projectile states. Without a measurement of the projectile or target characteristic X-rays or Auger electrons in coincidence with the emergent-projectile and/or the recoiling-target charge states it is not possible to identify the initial and final atornic orbitals involved in the transition and only a total cross section can be measured which is the sum over all the allowed pairs of electronic states. Thus, for a closer comparison with detailed theoretical calculations on electron capture it is mandatory to select experimentally either the initial or the final state occupied by the electron, if not both. The semiconductor gamma-ray detectors that appeared in the sixties have given a fundamental impulse in the inner-shell studies. In other experiments, Auger electrons are observed instead of X-rays.

New ion sources had permitted studies with relatively low energy multi-charged heavy ions. With the two-stage electrostatic accelerators, secondary beams of heavy ions in practically any charge state throughout the periodic table can be obtained. The very interesting bare, hydrogen-like or helium-like projectiles are nowadays currently produced in many laboratories. Even in larger machines, designed to produce GeV/a.m.u. beams, experiments in atomic physics are usually carried out.

Neutral and negatively-charged hydrogen bearns were, since the beginning, of great interest in atomic collisions. Workable MeV bearns of H^0 and H^- were first produced^[24] in 1958. Charge-changing processes in which a high-velocity hydrogen beam in a given charge state (proton, neutral atom), or the corresponding heavy ion beam, impinges upon an atomic hydrogen target constitute, in principle, the simplest processes. Therefore they are subject of great theoretical interest. However, the difficulties with the preparation of atomic hydrogen targets render these experiments a rather complicated task. The first measurements using atomic hydrogen as a target were made by Fite et al.^[25]. In 1966, an atomic hydrogen chamber was mounted in Belfast^[26] and since then this laboratory has been continually using this facility. Generally speaking, it is a tungsten furnace heated to about 2600K by conduction of an electric current where dissociation of H_2 takes place. However, for the most interesting studies (highvelocity proton and helium bearns on atomic hydrogen) the operation of such an oven is far from being simple

because of the heavier atom impurities released from the heated surfaces. A remarkable result coricerning the fundamental $H^+ + H \rightarrow H + H^+$ reaction, namely, the measurement of its forward angular distribution was published by Vogt et al.^[27] in 1986.

A further step in the experimental investigation of atomic collisions was the possibility of access to the differential cross sections. The dependence of the reaction probability on the impact parameter is an indispensable piece of information to permit a deeper insight into the collision mechanisms. In some cases different processes leading to the same final state present distinct impact-parameter dependence and their separation depends on angular distribution measurements. Position-sensitive solid-state detectors and, more recently, position-sensitive microchannel plates were essential for this decisive experimental advance. A fine example of an early application of this technique is the experimental observation^[28] of the Thornas peak in high-velocity electron capture by protons from He. The differential cross section $d\sigma/d\theta$ was measured as a function of 0 for three different proton energies (2.82, 5.42 and 7.40 MeV) in an interval extending up to 1 mrad. The peak interpreted as due to the double-scattering mechanism of Thomas^[4] appears, as expected, at an angle near $\sqrt{3} m/2M$ where m and M are the electron and the proton masses, respectively. The position of the peak (~ 0.47 mrad) does not depend on the proton energy but the peak becomes increasingly pronounced as the energy is increased. This last feature confirms the theoretical prediction that, in a perturbation expansion, this peak corresponds to a second-order Born processes and that in the limit of high velocities the second-order Born term dominates over the first one^[29]. A few years later, Vogt et al.^[27] obtained much cleaner results using atomic hydrogen as a target, in a true three-body problem.

In the sequel we present a small sample of the rich variety of phenomena encountered in charge-changing collisions to illustrate some recent developments in the understanding of basic processes in this field. In section II problems related to one-electron capture are briefly discussed. Unlike inner-shell ionization theories, electron-capture theories are far from reproducing with the same accuracy the experimental situation in the Alceu G. de Pinho

 $v \gg 1$ regime. Moreover, it is shown that the onset of the radiative capture process renders extremely difficult to decide amongst the many first- and secondorder theories developed to describe the ordinary (nonradiative) capture process. In section III, two-electron processes are presented and scattering-correlation effects are discussed. This is, doubtless, the most active area of atomic-collision physics at present. Special attention is paid to the collisions of structured projectiles.

II. Radiative **electron** capture in proton-onhydrogen collisions

It was well known since the early 50's that the first-order theories of electron capture at not too high velocities^[5,6] account for the gross features of the process since the target atom wavefunctions are barely distorted by the fast projectile. The overlap integral of the electronic wavefunctions in momentum space essentially determines the capture probability. The fast decrease of the amplitude of the momentum wavefunctions with increasing electron momentum results in a very fast asymptotic (v^{-12}) decrease in the capture cross section. In the classical description of Thomas^[4], electron capture by a proton proceeds through two successive Coulomb scatterings in the first of which the transferred electron is ejected by the projectile. In the second collision the electron is scattered by the target nucleus and the capture follows if the electron acquires a final vector velocity that matches that of the scattered projectile. In the quantum-mechanical description, this double-scattering process is represented by second order terms in the Born expansion. At very high collision velocities, where the high-momentum components of the bound states are not large enough to facilitate the capture, the double-scattering processes become important mechanisms of electron capture as confirmed by the experimental observation of the Thomas peak^[27,28]. Thus a weaker (v^{-11}) asymptotic velocity dependence is expected as demonstrated in many different secondorder approaches: second Born^[30], continuurn distorted wave^[31,32], strong potential Born^[33,34] and symmetric eikonal^[35] calculations. Meanwhile, it was shown^[36,37] how to improve the first-order calculations by considering the correct asymptotic boundary conditions. The question concerning the high-energy electron capture theories is still controversial, and has given rise to some recent publications^[38-40].

This effervescent theoretical background, however, begged for confrontation with experimental results. It was already mentioned how the Heidelberg group tried to test the theories with a pure three-body collision system, namely $p + H \rightarrow H + p$. They measured the forward angular distribution of the emerging hydrogen atoms^[27] and extended up to 7.5 MeV the measurement of the electron capture cross section^[41]. The structure of the Thomas peak is relatively well reproduced by the second-order theories^[42]. However, in the integral cross section measurement it was observed^[41], at the highest energy, a significant (30%) discrepancy between the measured and the calculated values which was correctly interpreted as due to the contribution of another capture mechanism, the radiative electron capture (REC). Because of the rapid transfer of the electron from the target bound state to the fast-moving projectile bound state, a large change of electron momentum and energy is required which must be transferred to a thirc particle which can be the target nucleus in the case of ronradiative capture (MEC for mechanical electron capture) or a photon in the case of REC. At high impact velocities it can be assumed that the electron is initislly free and the REC is simply the inverse photoelectric effect^[43]. REC was observed for the first time by Schnopper et al.^[44] and its most spectacular exhibition takes place when a fast highly-stripped heavy ion (e.g.^[45], 25 MeV/a.m.u Xe⁵³⁺) traverses a crystal in channeling regime (e.g. $^{[45]}$, 17 μ m thick Si in (110) axial direction). Under channeling conditions the small impact parameter collisions are severely inhibited and the usual nonradiative capture is almost completely suppressed. REC, however, can occur with unbound target electrons and hence it is singled out.

Briggs and Dettman^[46], using the first Born approximation, have shown that the high-velocity limit of the 1s-1s REC cross section is proportional to v^{-5} . The same result was obtained^[47] with the continuum distorted wave (CDW) method. This means that at very high projectile energies it is the REC that gives the leading term. It becomes the dominant mechanism^[46] for $v \gtrsim 19.2 Z_T^{5/7}$. For protons in hydrogen the crossing of MEC and REC cross sections takes place at about 9 MeV. Then, if REC is not experimentally distinguished from MEC it is impossible to investigate the high velocity behavior of the nonradiative capture in this simplest three-body system.

The onset of the REC process in proton on hydrogen molecule collisions was studied recently by this author and co-workers^[48] in the energy range from 8 up to 24 MeV. Momentum analyzed beams of protons (12 to 24 MeV) and deuterons (16 to 23 MeV) were delivered by the electrostatic tandem accelerator of the TANDAR Laboratory at Buenos Aires. The corresponding velocity interval was $18 \lesssim v \lesssim 31$. The collimated beam was directed into a target chamber filled with researchgrade purity hydrogen gas. The equilibrium fraction was measured as a function of the impact velocity. As the equilibrium fraction does not depend on the initial charge state of the beam, the incident beam was permitted to enter the target chamber through a $6.3 \,\mu\text{m}$ thick aluminized mylar window. The emerging beam was charge analyzed and the undeviated beam, after passing a chopper, was detected by a E + AE telescope. For high enough values of v the equilibrium fraction is given by the simple relation $\phi_0 = \sigma_{10}/\sigma_{01}$. The electron loss cross section σ_{01} was obtained from known experimental values^[49,50] and an energy dependente given by $E^{-0.99}$ was obtained.

The resulting values of the total electron capture cross section σ_{10} are presented in Figure 1 together with those of Schwab et al.^[41]. The results obtained with molecular hydrogen were divided by two in order to be compared with experimental and theoretical values obtained with atomic hydrogen. It was believed that at high momentum transfers this procedure can be justified since the hydrogen momentum wavefunctions are not disturbed much by molecular interactions. A dramatic change in the slope of the σ_{10} vs E curve is clearly observed and it seems obvious to attribute this slowing down in the decrease of σ_{10} with E to the onset of the REC process, even in the absence of an experimental observation of the emitted photon. The theoretical curves shown in Fig. 1 correspond to the REC and MEC cross sections as given by the three-body calculations of Gonzalez and

Miraglia^[47] carried out using the CDW inethod for the process $H^+ + H(1s) \rightarrow H(1s) + H^+$. To account for the capture into the outer shells the original calculated values were multiplied by the factor $\sum n^{-3}$ which corresponds to a sum over all final bound states^[43]. The curve labeled "total" was obtained by the addition of the REC and MEC cross sections, disregarding eventual interference effects. The impressive agreement between measured and calculated cross sections seems to strongly support this interpretation.



Figure 1: Total electron capture cross section per atom (in units of a_0^2) as a function of the projectile energy (in MeV/a.m.u.). Dashed curve: REC-CDW calculation. Full curve: Total (MEC+REC) CDW calculation. See Ref. [47] and the text for details.

It is worth noting that the velocity for which the radiative and nonradiative cross sections become equal do not depend on the projectile atomic number. However, the absolute values of the cross sections scale as Z_p^5 . On the other hand, part of the total energy of the emitted photon increases with Z_p^2 . These circumstances open interesting possibilities of more detailed studies of the velocity dependence of the REC process by using fully-stripped heavy ions impinging upon hydrogen^[51]. A differentially-pumped gas cell and the growth rate method can be used and, moreover, the possibility of observation of the emitted photon and the value of the cross sections render feasible the separation of the REC process in coincidence studies. **III.** Two-electron processes and scattering **cor**-relation

111-a. Transfer and excitation

The basic one-electron processes are excitation, ionization and capture. Usually the projectile is a (charged) ion and the target is a (neutral) atom. The probability of forming a negatively-charged ion being in general very small, the capture proceeds, as a rule, to the projectile. On the other hand, excitation and ionization can occur in both projectile and target. The projectile ionization is called "electron loss".

Two-electron processes result from combinations of the three afore-mentioned basic processes. They are: double excitation (DE), double ionization (DI), double capture (DC), ionization and excitation (IE), transfer and excitation (TE) and transfer and ionization (TI). In all of them the final state differs from the initial one by two spin-orbitals. DF,, DI, DC and IE are selfexplanatory designations and they have been investigated from a long while ago. TE and TI, however, deserve some additional remarks as they were identified much more recently.

The TE process is the electron capture and projectile excitation occurring together in a single encounter^[52]. A dressed projectile captures an electron from the target and simultaneously excites an electron from the ground-state configuration of the ion. The intermediate projectile excited state is de-excited by the emission of a photon or an Auger electron. The TE process has received considerable attention experimentally after the observation by Tanis et al.^[53] of a resonance in the total cross section. It is analogous with dielectronic recombination in which the captured electron is initially free instead of bound. Both proceed via the inverse of an Auger transition and, hence, are resonant when the kinetic energy of the projectile electron matches the transition energy. This resonant condition in the collision velocity, referred to as resonant transfer and excitation (RTE), is a typical example of processes involving electron correlation. A uncorrelated capture and excitation process called nonresonant transfer excitation (NTE) is also observed but it is a two-step process in

which two independent interactions take place in a single encounter. NTE is due to an interaction between the projectile nucleus and a target electron resulting in a capture plus an interaction between the target nucleus and a projectile electron resulting in excitation (see Figure 2). Such combination of independent transfer and excitation events does not depend resonantly on the incident velocity. Nevertheless, NTE does exhibit a maximum in its energy dependence that results from the product of an increasing excitation cross section and a decreasing electron-capture cross section.



Figure 2: Scheniatic representation of the transfer and excitation processes. 2a) The resonant (correlated) process; 2b) The nonresonant (uncorrelated) process. Key to symbols appearing in this figure and in Figures 4 and 5: $P - projectile; \Box - target; C - capture; E - excitation; I - (target) ionization; L - (projectile) loss; solid circumference: spin-orbital occipied in the initial state; dashed circumference: spin-orbital to be occupied in the final state; wavy-line: the scattering correlation interaction; dashed-dotted line: the (screened) nucleus-electron interaction$

Experimentally the RTE has been identified and investigated by measuring either the yield of deexcitation photons in coincidence with projectiles which have captured an electron^[52-54] or the yield of Auger electron emission associated with single capture events^[55,56]. Indeed, it was the use of ion-X-ray coincidence techniques that lec to the discovery of RTE. However, at the present time, the high-resolution zero-degree Auger spectroscopy technique is by far the most powerful instrument of investigation of this process. This technique obviates the need for coincidence measurements and provides rauch more complete information about

the intermediate excited states formed in the collision which decay by autoionization. Projectile electron spectroscopy has been used for many years in the so-called beam foil spectroscopy^[57]. However kinematic line-broadening effects have limited the usefulness of this technique. These broadening effects were substantially reduced by the observation of the emitted electrons at zero degrees with respect to the beam direction^[58]. Furthermore, few-electron systems are expected to provide the best testing grounds for studying the role played by electron-electron interaction. Since the lighter the projectile the smaller the fluorescence yield, the use of Auger spectroscopy became particularly attractive for investigating low-Z projectile excited states.

III-b. Transfer and Ionization

A very comprehensive definition of the transfer and ionization process is a collision wherein the target looses more electrons than are captured onto the projectile. This means that one or more unbound electrons are produced. Ionized electron spectra can be viewed as a continuation of target excitation into the ionization continuum plus a continuation of charge transfer to projectile excited states into the ionization continuum^[59]. In the second case, the wavefunction which describes the motion of the electron after the collision is a projectilecentered continuum (Coulomb) wavefunction. An experimental demonstration of the existence of this second kind of ionized electron spectra in ion-atom collisions can be found in the work of Vane et al.^[60]. They have sent bare C^{6+} and O^{8+} ions in Ar and measured the spectra of the electrons captured in the continuum of the projectile in coincidence with final charge states different from the incident one.

The electron capture to the continuum (ECC) gives rise to a cusp-shaped distribution of electrons in velocity (v,) and angle, emergent from the collision region, and centered very near the vector velocity of the scattered ion. However, the mere observation of a forward peak of unbound electrons is not enough to ascertain that it is an ECC peak^[61]. In fact, ECC gives rise to an asymmetric cusp which is strongly skewed toward lower velocities. The cusp is instead nearly symmetric in the electron loss to the continuum (ELC), a process that may occur when the projectile is not a bare ion. Vane et al.^[60] did not perform a rigorous analysis to distinguish between ECC and ELC. Notwithstanding, they attributed the continuous electron spectra to ECC.

When a projectile with charge q impinges upon a helium atom, the TI process gives rise to a final state consisting of an emergent projectile with charge q-1, a He²⁺ residual ion and an electron in the projectile continuum. Andersen et al.^[62] were the first to use He as a target with the purpose of identifying the TI process. The coincidence of a free electron with the capture of a bound electron necessarily identify a TI event. However, it is the shape of the electron forward peak that permits the distinction between ELC and ECC. Andersen et al.^[62] performed a measurement of the double-differential cross section in the projectile rest frame as a function of the electron energy and concluded that it was characteristic of ELC. Then the TI process involves the formation of a highly-correlated two-electron projectile state by the transfer of an electron pair, with subsequent loss of one electron to the projectile continuum via an electron-electron interaction. More recent experiments^[63] support this interpretation.

The paradigms of TI are the simple four-body reactions $H^+ + He \rightarrow H + He^{2+} + e^-$ and $He^{2+} + He \rightarrow$ $He^+ + He^{2+} + e^-$. Not very much is known about them for energies higher than a few hundred keV. Recently, Pálinkás et al.^[64] published a very interesting result concerning the first reaction at 1 MeV proton impact. They were able to distinguish between a correlated TI process involving explicitly an electron-electron interaction and a uncorrelated process in which the electron capture to a projectile bound state is described by the first term of the Born expansion (the target nucleus gets the momentum released by the capture) and the second electron is transferred to the continuum by a binary-encounter with the projectile. For the correlated TI process the following two-step process was proposed: the electron to be captured first collides with the projectile and then scatters into a projectile bound state through a collision with the other electron which is sent to the continuum. The quantum-mechanical description of this two-step process (in some sort similar to the Thomas process) was given by Briggs and Taulbjerg^[65] through a second Born calculation and the equivalent

of the classical Thomas peak is expected to appear in the angular differential scattering cross section of the emergent neutral beam. However, a more interesting feature of the correlated TI process was predicted by Briggs and Taulbjerg and later confirmed by calculations of Ishihara and McGuire^[66]. They predicted that a peak should appear in the energy spectrum of electrons ejected at 90° with respect to the beam when the electron velocity matches the projectile velocity. Pálinkás et al.^[64] looked for the corresponding peak at 90° in the angular distribution of $v_e = v$ electrons ejected in coincidence with the emergent neutral beam. A peak was effectively found at 90°, sitting on a continuous background attributable to the uncorrelated events.

Then, in TE as well in TI the two-electron transitions may result from either correlated or uncorrelated processes. This is generally true for all twoelectron processes. The two-electron transition is correlated when an electron-electron (e-e) interaction is present. The uncorrelated process is due to two independent one-electron transitions produced by the electron-nucleus (e-n) interaction. Both processes may occur in a given reaction leading to the same final state and then interference is possible. Hence, it is often very difficult to single out experimentally the correlation effects. Since the early eighties considerable effort has been devoted to the search of correlation effects in twoelectron processes which can be experimentally disentangled from the competing uncorrelated (one-electron) processes.

III-c. Static and scattering correlations

One must distinguish between static and scattering correlations^[67]. Static correlation occurs in separated atoms. An example of static correlation is that associated with the difference between the "exact" and the corresponding Hartree-Fock total energy. In other words, that part of the total energy not incorporated in the "best" independent-particle-model calculation. In the collision problem, the static correlation corresponds to deviations of the asymptotic bound-state wavefunctions from a product of single electron wavefunctions. In any way, static correlation is the residual effect that cannot be described by the independent-particle approximation.

In some cases the asymptotic states are not stationary with respect to electron correlation. For instance, one of the collision partners may be left, after collisional excitation, in a metastable state that decays as, e.g., by Auger effect in the TE process. Another example is the shakeoff meef anism in which the correlation is modified by the removal of an electron by the projectile, leading to rearrangement in the target final state with the ionization of an electron, as observed in the multiple ionization of outer shells. This kind of correlation, occurring in the asymptotic final-state wavefunction, is referred to as dynamic correlation^[67].

Scattering correlation is an e-e interaction in the time-dependeiit two-center Coulomb field of the collision partners. In this way it is correlation intrinsic to the scattering process itself, i.e., correlation occurring during the collision. McGuire^[67] characterized the scattering correlation as that corresponding to deviations of the time-dependent evolution operator from a product of single particle-operators. Single-particle operators lead to the independent-event approximation.

Stolterfoht^[68] stressed the distinction between electron correlation and screening in the framework of the Hartree-Fock method. He showed that the e-e interaction may be partitioned into two terms: one is an one-electron operator 'that can be absorbed into an independerit-particle Hamiltonian and the other is a two-electron operator responsible for the correlation interaction. Then the e-e interaction contains contributions chacacteristic of one-electron operators (the screening effect is an example) beside those characteristic of two-electron operators (that couple states differing by two spin orbitals). Electron-correlation effects are then closely related to two-electron processes.

In the nest sub-sections, results on some twoelectron processes will be comminted. In some of them correlation effects are made experimentally visible but not in others, specially when only integral and total (summed over all final states) cross sections are measured.

III-d. Double Capture

In Stolterfoht's classification^[68] of the scattering correlations the process of double capture is described

as a bicentric scattering because the correlation process occurs during the exchange of the electron pair between the two centers. The four-body problems corresponding to the collisions $H^+ + He \rightarrow H^- + He^{2+}$ and $He^{2+} + He \rightarrow He + He^{2+}$ are of particular interest. Total cross sections were measured for both of them, a few years ago, by this author and co-workers. The maximum impact energy was 1 MeV in the first case^[69] and 3 MeV in the second^[70].

Motivated by the new He^{2+} + He data, Gayet et al.^[71] have made calculations in the framework of the independent event approximation (where the electrons are assumed to evolve independently in time) using an independent electron description for the bound states. The transition amplitude was evaluated through either the CDW approximation or the CDW-eikonal initial state approximation (in which the initial bound wavefunction is distorted by the eikonal Coulomb phase factor due to the projectile). An excellent agreement between the results of the CDW-EIS calculation and the experimental data was found in the absence of any correlation (see Figure 3).



Figure 3: Total double capture cross section for He^{2+} + He collision (in cm²). The curve is the CDW-EIS calculation of Ref. [71] for the resonant collision $(1\varsigma^2 \rightarrow 1s^2)$.

More recently Schuch et al.^[74] have extended the impact energy up to 6 MeV. Their results were compared with a new CDW calculation^[75] using now electron configuration interaction wavefunctions in both

the initial and the final bound states. This static correlation has nothing to do with the scattering itself since a quantum-mechanical bound state of the target is by definition prepared in the absence of the incident beam. There is only a very small difference between the calculation including configuration mixing and that in an independent particle model with the $(1s)^2$ configuration only. Moreover, no significant quantitative difference with the results of Gayet et al.^[71] was obtained, confirming that there is no evidence for scattering correlation in the total cross section.

The differential cross section for 1.5 MeV He²⁺ on He was also studied by Schuch et al.^[74] with the hope of finding some structure in it as, for instance, evidences of a Thomas peak. No clear-cut indication of scattering correlation effects was observed too.

Double capture in more complicated systems leading to negative final charge states have been reported in the last few years^[69,76], namely, $H^+ + He \rightarrow H^- + He^{2+}$ and $He^+ + He \rightarrow He^- + He^{2+}$. So far as we know, there is not at the present time any available theory, starting from first principles, with the purpose of calculating the cross section for formation of negatively charged ions through double capture. The production of $H^$ ions in collisions of H^+ with Ne and Ar atoms was also examined by Almeida et al.^[65]. A reasonable description of the cross sections in the interval 0.1–4 MeV was obtained assuming the independent event approximation and using the strong potential Born results^[33] for K-shell one-electron capture extended by OBK-type scaling rules to the outer shells^[69].

111-e. Structured projectiles and the e-e interaction

When the projectile also carries electrons, in addition to the projectile nucleus, the accompanying electrons may influence the scattering process. Some aspects of the e-e interaction in collisions with dressed projectiles were already mentioned, e.g., in the RTE process. In this subsection the screening and antiscreening effects will be considered. It is generally possible to consider a projectile with atomic number Z_P carrying N electrons as an equivalent point particle with an effective charge Z'_P . Two extseme situations are easy to visualize. Firstly, when all the electrons are tightly bound the square of the effec-

tive charge is simply given by the screened nucleus, $(Z'_P)^2 = (Z_P - N)^2$. Secondly, when they are all loosely bound then $(Z'_P)^2 = Z_p^2 + N$, i.e. the nucleus and the electrons act incoherently as assumed by the free collision model (FCM) of Bohr^[10]. In the intermediate situations the effective charge is given in the first Born approximation as a function of the momentum transfer^[77]. The fully-screened situation corresponds to small values of the momentum transfer or to large values of the impact parameter, b, and, conversely, the totally incoherent situation corresponds to small values of b. A beautiful illustration of the FCM was given by Wang et al.^[78] in the collision of fast Rydberg hydrogen atoms with different targets. They have shown that the cross section for collision of a Rydberg hydrogen atom is equal to the sum of the cross sections by impact with a free proton and with an equal-velocity free electron.

A similar situation refers to excitation or ionization of a target atom by a projectile carrying electrons. As mentioned in Section I, since the work of Bates and Griffing^[12] it is known that the incident electrons, besides playing a passive role in screening the projectile may also participate actively in the process being the active agent in the excitation or ionization of a target electron. The screening (S) effect decreases the ionization or excitation probability through the presence of an effective nuclear charge of the projectile. On the other hand, when the electrons play an active role they contribute to increase the target ionization or excitation probability. This is known as the antiscreening (AS) effect. An example of the contribution of the active electrons is the study of the collisional ionization of He and Ar by fast H and H⁻ projectiles^[79]. The FCM generally describes single ionization quite well, the collision with the composite projectile being regarded as a collision with the constituents of the projectile which scatter independently on the target. Even in the case of double ionization the agreement with the FCM is surprisingly good despite the important role played by the target-electron correlation^[79].

Finally, another aspect of the same process is the projectile excitation or ionization (electron loss) due to the target electrons. As seen from the projectile frame the target electrons are impinging on the projectile with nearing the same velocity as the target nucleus. The transition of the projectile electron from the ground to a higher state (bound or free) can be induced by the projectile electron-target nucleus interaction or by a projectile electron-target electron interaction. In the second case, when both electrons are removed from their original states, the two-particle aspect of the *e*e interaction becomes evident. In the classification of Stolterfoht^[68] this AS effect is a two-center scattering correlation since it corresponds to the scattering of electrons located initially at different centers. It is then a genuine scattering correlation since it does not exist before the collision. It must be emphasized that in the two-center scattering correlation the *e*-e and the *e*-n interactions may be treated separately in first order^[12,68].

The calculation of the S-AS effects has been carried out usually in the PWBA^[12,77] where the cross section can be expressed as a product of two form factors, one connecting the initial and the final electron states of the projectile and the other doing the same for the target electrons. In the Bates-Griffing approach (the H+H case) all the possible excited states are known and an exact result can be obtained. When the final state of the target is not observed, it is necessary to consider all the possible excited target states. Going from the hydrogen atom to many-electron atoms, McGuire et al.^[77] simplified the problem by using the closure approximation to avoid explicit summations. Anholt^[80] was the first to propost: a simple way to circumvent a flaw in the use of the closure approximation. He introduced explicitly the idea that in the projectile frame of reference the ionization of tlie projectile is due to an impinging beam of electrons in addition to the target nucleus. The small value of the ratio m/M implies that at low incident energies the target electrons do not dispose of enough kinetic energy in the projectile frame to ionize or even excite the projectile electron. Hence, the contribution of the e-e interaction should be severely reduced at low impact energies to take into account the threshold effect in the AS part of the cross section. If the target electrons were completely free this threshold would be sharply defined at the impact energy E = (M/m) A Ewhen AE is the excitation or ionization energy of the projectile. Because of the momentum distribution of the target electrons the threshold is smeared out.

The first experiment to distinguish the e-e from the e-n interaction has been worked out by Zouros et al.^[81].

They observed by Auger spectroscopy the production of $1s 2s 2p^4 P$ projectile states excited in collisions of Lithium-like $(1s^2 2s) O^{5+}$ and F^{6+} ions with He and H₂ targets. The $1s 2s 2p^4 P$ excited state could not be produced by direct e-n interaction since this would require a forbidden spin-flipping transition. To the transition energies AE ~ 560 and 721 eV for O^{5+} and F^{6+} , respectively, correspond threshold impact energies of 16.3 MeV for O^{5+} and 25.0 MeV for F^{6+} . The production of the ⁴P autoionizing state was monitored by high-resolution zero-degree Auger spectroscopy. The production cross section was then determined as a function of the impact energy and was found to increase sharply for $(E/A) \gtrsim 0.75$ MeV/a.m.u.. The threshold effect is especially visible with the H₂ target.

A few months later. Hülskötter et al.^[82] observed the same threshold effect in the cross sections for projectile K-shell ionization measured for 0.75-3.5 MeV/a.m.u. C⁵⁺ and O⁷⁺ projectiles in collisions with H₂ and He targets. A PWBA calculation based on Ref. [80] and taking into account the e-n and e-e interactions reproduced well the data. Similar effects were observed by Shah and Gildoby^[83] in the cross sections for one-electron loss by Li⁺ and Li²⁺ in H, H₂ and He within the range 0.3-2.7 MeV. Other recent experimental studies of the onset of the AS effect have been also directed to the measurement of total cross sections for electron $loss^{[84-86]}$. It was systematically verified that, once the threshold is reached, the e-e and the e-n interactions begin to compete on an equal footing, for light-target collisions.

In the meantime, considerable progress was achieved in the theoretical description of the AS mode^[87-88]. Anholt's results^[80] are in good agreement with the exact ones whenever they can be obtained^[12] except very close to threshold where they exhibit the characteristic FCM discontinuity in the slope of the curve $\sigma_{loss}(E)$. Firstly, it was shown by Montenegro and Meyerhof^[87] how to eliminate this unphysical discontinuity. A sum rule for the stopping power due to Bethe was used to perform a more exact summation over the targetelectron states resulting in an electron loss cross section that is the sum of the S and AS contributions and exhibits a smooth transition from the S- to the ASdominant region. In their next paper, Montenegro and Meyerhof^[88] have studied the impact-parameter dependence of the screening effect using the time-dependent SCA. After integration over the impact parameter they have shown that the SCA-ionization cross section with screening is equivalent to the screening contribution to the total-ionization cross section in PWBA. Finally, in a third paper, Montenegro and Meyerhof^[89] have applied the time-dependent SCA to obtain the impact-parameter dependence of the AS effect. It was shown that the AS-mechanism has a much broader probability distribution P(b) than that of the S-mechanism, allowing significant contributions to projectile electron loss for large internuclear distances.

III-f. The separation of the S- and AS-mechanisms

This theoretical effort stimulated an experiment^[90,91] with the purpose of separating the S-from the AS-mechanism. The experiment consisted in measuring the emergent charge states singly and in co-incidente with the recoil ions, in collisions of He⁺ (1.5–4.0 MeV) with H₂ and He targets. The following charge-changing reactions were studied

- (A) $\operatorname{He}^+ + T \rightarrow \operatorname{He}^+ + T^+ + e^-$
- (B) He⁺ + T He²⁺ + ~ + e -
- (C) $\text{He}^+ + T + \text{He}^{2+} + T^+ + 2e_-$

where T stands for H_2 or He. The first reaction is simply target ionization, the second is projectile ionization (electron loss) without target ionization and the last corresponds to the ionization of both the projectile and the target. One can reach the final charge state of B by three different processes pictured in Figures 4 and 5 as B_1 , B_2 and B_3 . Similarly, the final charge state of C can be attained by the processes C_1 or C_2 . The AS-mode occurs whenever the *e-e* interaction (represented in Figure 5 by a wavy line) is present. Otherwise the process is of the S-type (Figure 4).

Processes A and C are given independently by two coincidence measurements (emergent He⁺ or He²⁺ beam with the T^S recoil-ion). A measurement of He²⁺ singles gives the sum of all the processes represented in Figures 4 and 5 and permits to obtain the total cross section for electron loss, σ_{12} . Figure 6 gives the σ_{12} cross sections^[91] together with those of other authors^[70,92-94] for He target. The values given by de Castro Faria et al.^[70] are systematically below those published by other groups. Since they were re-measured recently at the same laboratory^[91] they were normalized by means of a factor of 1.5 obtained by using the four highest impact energies points of Ref. [91], those with the smallest error bars. Once σ_{12} and the cross section for reaction C are known, the cross section for electron loss not accompanied by target ionization (process B) is obtained by subtraction. Figure 7 shows the experimental results of Montenegro et al.^[90] for reaction C in He and Figure 8 the same for reaction B. Whenever results from other authors are known, they are also shown.







Fig. 2.



Figure 5: Schematic representation of the correlated (AS) processes leading to the projectile electron loss in He⁺ + He (g.s.) collision. 5a) C₁-final state: He⁺⁺ + He⁺ + 2e⁻; 5b) B₃-final state: He⁺⁺ + He^{*} + e^- . The key to symbols is as in Fig. 2.





Figure 6: Total electron-loss cross section σ_{12} (in Mb) of He⁺ in He. The theoretical curves are from Ref. [90]: AS, antiscreening; S, screening; σ_{12} , total cross section for processes B+C. The data from Ref. [70] were multiplied by 1.5, as explained in the text.



Figure 7: Cross sections for electron loss (in Mb) of He⁺ followed by target onization in He. The theoretical curve^[90,91] corresponds to the sum of processes C_1 and C_2 or, equivalently, to $AS-B_3 + C_2$ (Reaction C).

Figure 8: Cross sections for electron loss (in Mb) of He⁺ not followed by target ionization in He. The theoretical curve^[90,91] corresponds to the sum of processes B₁, B₂ and B₃ or, equivalently, to $S+B_3 - C_2$ (Reaction B).

theoretical curves were obtained The as follows^[90,91]. The S and AS cross sections shown in Figure 6 were calculated from the results given in Ref. [87] which are based on the extended sum rule method including bound and unbound states. To obtain the cross section for reaction C it is necessary to consider the diagrams C_1 and C_2 . The final states of these processes are indistinguishable from each other. Hence interferente may occur and the two amplitudes should be combined coherently. However, as shown in Refs. [88] and [89], the impact-parameter distributions of the AS and the S processes are very different from one another, the peaks of these distributions lying at quite distinct impact parameters. Then, it is justifiable to neglect interference effects and to add the two individual cross sections. Since the AS-cross section is given by the sum of the cross sections for the C_1 and B_3 processes, it can be written symbolically that $C_1 + C_2 = AS - B_3 + C_2$. The B₃ and C₂ cross sections were calculated in Ref. [90]. Curve C in Figure 7 is the result of this calculation. Curve B, in Figure 8, was obtained from the surn $B_1 + B_2 + B_3$ which is equivalent to $S + B_3 - C_2$. The agreement between theory and experiment is quite impressive but there are still some theoretical problems to be solved specially at the low energy side of the maximum appearing in the cross section curves.

The four-body problems represented by the reac-

tions

$$P^{q} + H \rightarrow P^{q} + H^{+} + e^{-}$$

$$P^{q} + H \rightarrow P^{q+1} + H + e^{-}$$

$$P^{q} + H \rightarrow P^{q+1} + H^{+} + 2e^{-}$$

where P^q stands for He^+ (q = +1) or H (q = 0) are particularly attractive from a theoretical viewpoint because calculations are minimized, as the exact electronic wavefunctions in the initial and final states are known. Coincidence measurements in these systems are welcome. The H+H system was already studied but without the identification of the charge state of the recoils^[95]. For this reaction, the energies of interest lie in the interval 1–300 keV. The He⁺ +H system is still an open problem in spite of the maximum in the electron loss cross section to occur in a more comfortable energy range.

III-g. Other manifestations of the antiscreening effect

Another manifestation of the AS effect is related to the transfer and excitation process. In the collision of a hydrogen-like projectile with an atom, a target electron excites the projectile and a second independent target electron is captured into the projectile which is transformed into a helium-like ion in an excited state^[96]. No free electron is present before an eventual decay by Auger effect occurs. Obviously the process is nonresonant and presents a threshold which corresponds to the projectile electron excitation energy. The prototype of this reaction is $He^+ + He \rightarrow [He]^* + He^+ \rightarrow$ $He^+ + e^- + He^+$. The e-e interaction involving the active projectile electron can result in loss instead of excitation. If a second target electron is transferred to the projectile in the same encounter the initial and final charge states of the projectile are the same^[86]. The reaction final state is then indistinguishable from the double ionization of the target produced by the dressed projectile.

A much more stringent test of the SCA theory for screening^[84] and antiscreening^[85] is the investigation of the impact parameter dependence of the e-e interaction. Montenegro et al.^[97] have studied collisions of the hydrogen-like projectiles Li²+ and C⁵⁺ on H₂ and He targets (0.17 $\leq Z_T/Z_P \leq 0.67$) within the velocity interval $Z_P \leq v \leq 2.2Z_P$. The first pair of

inequalities concerns the applicability of the SCA for the calculation of the S amplitude and the second one do tlie same for tlie AS amplitude. The general trend of tlie experimental results confirmed the expectations and demonstrated the increasing importance of the AS process at larger impact parameters.

Additional information concerning the impactparameter dependence of the S and AS probabilities comes from the study of electron emission occurring as a result of structured particle impact. Experiments with intermediate-energy He, He⁺, H⁻ and H projectileâ colliding with He have been reported by DuBois and co-workers^[98]. Electrons created by ionization of the target or the projectile, after exiting the target gas cell, were energy analyzed at different laboratory ernission angles and detected in coincidence with the emerging projectile in a given charge state. Cross sections, differential in emission angle and energy, obtained for a fixed emission angle and in coincidence with projectiles having lost an electron, exhibit a large peak for electron energies such that the ejected-electron and projectileion velocities are nearly equal. This peak, which corresponds to the ELC process, sits on a background due to the simultaneous ionization of both collision partners. Theoretical analyses^[98-100] of these results show that the electron loss by the projectile accompanied by an electron transition in the target is indeed a very important process, irrespective of the emission angle and energy of the ejected electron. As usual, there are two contributions to this process: one correlated due to the e-e interaction, the other uncorrelated due to the projectile nucleus-target electron interaction. The correlated process is important for small momentum transfers to the projectile electron, its contribution increasing with decreasing electron energy and emission angle^[100]. Hence it becomes the dominant process in the low-energy side of the electron spectra, which corresponds to large impact parameters. To associate this part of the spectra with the AS contribution to the simultaneous ionization results in a consistent picture of all known facts about the AS part of the interaction of structured projectiles with atoms and the predictions of Ref. [89].

An alternative approach to describe the correlated projectile electron loss has been recently developed by Lee et al.^[85], based on the impulse approximation (IA).

The IA provides a simple formula for calculating the contributions of the e-e interaction to projectile ionization. Combined with a PWBA calculation of the e-n interaction contributions, the final result agreed well with the experimental data obtained^[85] in the collisions of fast O^{4+} and C^{2+} ions with H_2 and He targets. However, no attempt was made to isolate experimentally the AS from the S mechanism.

Finally, it is worth noticing that the AS enhancement of the ionization cross section must be considered in the calculation of the stopping power as has been recently recognized by Schiwietz and Grande^[101]. Near the stopping power maximum the stopping cross section has to be calculated for each projectile charge state separately. For instance, in the calculation of the electronic stopping of protons in H_2 , the AS process is responsible for a non-negligible contribution to the stopping cross section, when the H+H collision is considered. This is still an open problem for heavier projectiles, when the presence of structured projectiles inside the target is a much more probable situation.

IV. Final remarks

We have seen in this brief review that, despite a 70-year-old history, charge-changing collisions are still an extraordinarily vivid area of atomic physics. New phenomena have been discovered in this last decade that are challenging both experimentalists and theorists. Experirrent is far ahead of theory in most cases but major systematic experimental efforts are also required to provide some highly accurate benchmark-type measurements in a few selected basic processes, especially those related to few-electron problems. In other cases the measured integral cross sections are far from forming a fully satisfactory data base necessary to stimulate more complete and complex theoretical calculations and much experimental effort is required to fill in the existing gaps. This is specially true when multichannel effects are present, a situation that requires coupled-channel calculations. Since total cross sections provide only 1 mited information on the details of almost all processes, double or triple differential measurements are required. Novel experimental techniques are constantly being developed enabling the determination

of increasingly subtle aspects of the charge-changing processes. A major theoretical challenge lies in the area of low incident energies where a molecular description of the entire colliding system (i.e., the incident projectile, the recoiling target and the eventually ejected electrons) is necessary since all partners interact strongly. However, even at relatively high energies many problems persist as is clearly evidenced by the systematic study of Berg et al.^[102] on the helium double ionization by fast (12 < v < 200) and highly-charged heavy-ion impact. Double ionization is a typical example of a single-center scattering correlation in Stolterfoht's classification^[68]. The electrons are located initially at one center and the e-e interaction cannot be treated independently from the n-e interaction. Ford and Reading^[103] developed a "forced impulse method" which was applied to describe the interplay between the e-e and n-e interactions. It was very successful to explain the differences observed with proton and antiproton beams but the situation with energetic highly charged ions is far from being well explained by any available theory.

In the last decade, the interest moved to the study of the electron-correlation effects. Electron correlation is very important in ion-atom collisions at low impact velocities, relatively unimportant at intermediate velocities but, quite surprisingly, become important again in some processes at high velocities. Many new phenomena whose interpretation seems to involve the concept of scattering correlation were identified. However the unambiguous identification of scattering correlation effects is by no means straightforward. It is required to rule out processes concerning electron correlation in the separated atoms or produced by independent singleelectron transitions. The fact that two electrons undergo a transition in an atomic collision is not enough to guarantee the presence of scattering correlation. However, it was shown how the combined efforts of experiment and theory have permitted to understand the role played by electron correlation in some two-electron transitions.

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