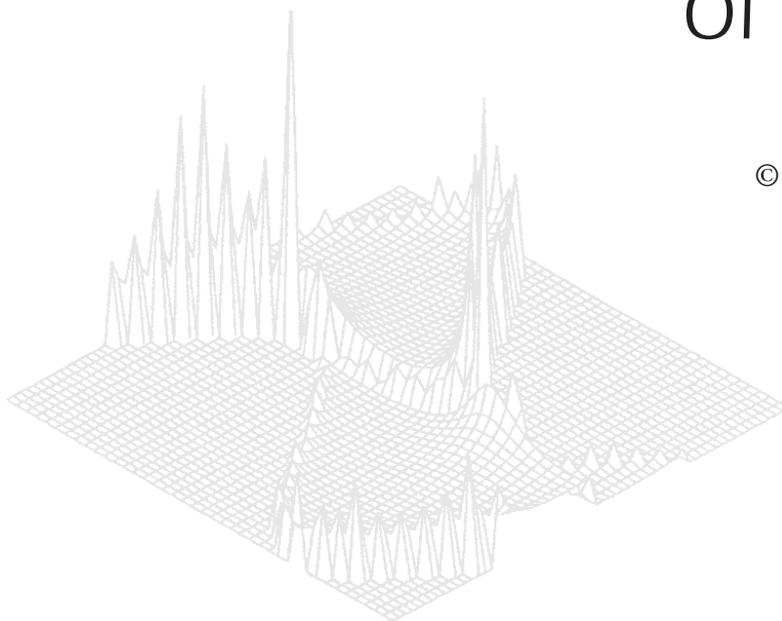

C S I R O P U B L I S H I N G

Australian Journal of Physics

Volume 52, 1999
© CSIRO Australia 1999



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Paul Trap Promotion of Electrons in Atomic Collisions*

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Abstract

Electron promotion on rotating potential saddles is proposed as an important and efficient excitation mechanism taking place in atomic collisions at intermediate energies. Measurements on the excitation of atoms by positively charged particles, in particular, excitation of He I states in p-He collisions provides experimental evidence for this ‘Paul trap’ promotion in two-centre Coulomb potentials.

1. Introduction

Atomic collisions at intermediate energies, where the projectile velocity is of the order of the velocity of bound electrons, are still not well understood theoretically. There are well-established approaches to collisions in the low-energy and high-energy limit as, for example, the molecular orbital model and the Born approximation (Briggs 1985). However, a systematic approach applicable to collisions of the intermediate energy range is not available. Various approximation methods such as semiclassical close-coupling calculations or higher-order perturbation theory have been applied, but with unsatisfactory results even for relatively simple collision systems (Fritsch and Lin 1991).

In this paper, we consider a particular class of collision systems, namely the three-body Coulomb system, where a single electron of the target atom is excited due to the interaction with the Coulomb field of a charged projectile. The collision process is assumed to be independent of the internal structure of the projectile. In this case, Paul trap promotion (von Oppen 1994) seems to be a dominant excitation mechanism, if the projectile is positively charged and if its energy is in the intermediate energy range.

Paul trap promotion can take place in three-body systems. Consider, for example, a positively charged projectile colliding with a hydrogen atom. During such a collision, the electron is moving in the two-centre Coulomb field of projectile and proton. This field has a saddle between the two positively charged centres. During a collision this saddle rotates through an angle π , if a straight-line

* Refereed paper based on a contribution to the Australia–Germany Workshop on Electron Correlations held in Fremantle, Western Australia, on 1–6 October 1998.

trajectory is assumed. If the electron reaches the saddle at about the moment of closest approach of projectile and target, its motion can be stabilised on the saddle for some time due to the rotation. Though its initial motion is directed along the molecular axis of the collision system and, therefore, unstable, it changes to a stable motion perpendicular to this axis provided the rotation is fast enough. Due to this dynamical stabilisation, a mechanism exploited also in Paul traps for ions (Paul 1990), the electron can ride on the saddle for some time, while the collision partners separate and the saddle potential is rising. When the electron finally leaves the saddle to either side, it has gained enough energy for populating excited states in the Coulomb potentials of proton or projectile. Even ionisation is possible, if the electron stays on the saddle and is finally left stranded halfway between proton and projectile in an unbound state (Pieksma and Ovchinnikov 1995).

In this picture both the stretching of the collision system after the moment of closest approach and its rotation are decisive for the efficiency of electron promotion by the Paul trap mechanism. We discuss the evolution of the electron cloud in more detail in Sections 2 and 3.

Afterwards we consider several experimental results providing evidence for Paul trap promotion, in particular:

- (i) Collisional excitation by charge conjugated projectiles (Section 4).
- (ii) Resonant Paul trap excitation of He atoms by 15 keV protons (Section 5).
- (iii) Electric dipole moments of collisionally excited electron states (Section 6).

We conclude that Paul trap promotion plays an important role in intermediate-energy collisions, both for heavy projectiles and positrons. The present analysis relies mostly on classical dynamics and, therefore, allows merely some intuitive and qualitative understanding of the excitation process. Quantum physical effects are mostly disregarded here. Only for the case of resonant Paul trap promotion, the dynamical evolution of the electron state on the saddle seems to follow in-saddle sequences of molecular orbitals as described by Rost and Briggs (1991). This theory allows also some visualisation of the quantum dynamical process.

2. Electron Promotion to Parabolic States

As mentioned in the Introduction, two types of motion are relevant for Paul trap promotion of electrons to excited states: stretching and rotation of the two-centre potential. In this section, we shall consider the evolution of an electron cloud centred on the saddle of the potential of two separating positively charged centres. This evolution can be treated within the framework of saddle dynamics as introduced by Rost and Briggs (1991). Considering, in particular, a symmetrical two-centre potential as for proton impact on hydrogen, the H_2^+ -like collision system can evolve diabatically along potential curves of a series of in-saddle states. Thus, starting from the $1s\sigma_g$ molecular ground state of the collision system, the electron can be promoted along the in-saddle sequence $1s\sigma - 3d\sigma - 5g\sigma \dots \rightarrow |n; n-1, 0, 0\rangle$. This sequence leads selectively to parabolic states $|n; n_1, n_2, m\rangle$ with $m=0$ and $n_2=0$, if the electron ends up in a bound state of the atom, or $n_1=0$, if it is finally captured by the projectile. Here we refer to the direction of the projectile beam as the positive z axis, thereby fixing the meaning of the parabolic quantum numbers n_1 and n_2 (Bethe and Salpeter 1957).

Assuming an initial $1p\sigma - 2p\pi$ rotational coupling, saddle dynamics leads from the $2p\pi_u$ state along the in-saddle sequence $2p\pi - 4f\pi - 6h\pi \dots \rightarrow |n; n-2, 0, \pm 1\rangle$ to parabolic $|m|=1$ states of the target atom. These parabolic σ and π states with $n_2=0$ are those states within an electron shell, for which the electric dipole moment given by (Bethe and Salpeter 1957)

$$\langle -e \cdot z \rangle = -\frac{3}{2} n (n_1 - n_2) \quad \text{a.u.} \quad (1)$$

is maximal. This result is in accord with the intuitive visualisation of the collision process. While riding on the saddle, the electron is shifted downstream with respect to the target nucleus. This displacement gives rise to the large electric dipole moments, typical for states resulting from intermediate-energy collisions.

This intuitive visualisation of the collision process based on classical dynamics can be straightforwardly generalised with respect to asymmetric two-centre Coulomb systems. Therefore, it is also helpful for understanding excitational collisions induced by highly-charged-ion (HCI) impact. According to classical dynamics, saddle promotion is most likely for collisions where the saddle trajectory has an impact parameter $b_s \approx 1$ a.u. In this case, the saddle is lowered at the moment of closest approach to about the binding energy of the electron bound to the target atom. Therefore, the electron may reach the saddle and ride on it for some time, while the saddle potential rises again. Thus the electron gains energy and can populate excited states, when falling back to the target atom.

Two aspects of this excitation process should be noted especially: on the one hand, the displacement of the electron cloud with respect to the target nucleus mentioned before; on the other hand, its selectivity with respect to the impact parameter. Due to this selectivity, the collisionally populated states are extremely pure, that is highly coherent mixtures of $|nlm\rangle$ states with different l , even if total excitation implying integration over all impact parameters is considered. This conclusion corresponds to and generalises the result deduced above using saddle dynamics, that the parabolic σ and π states with $n_2=0$ are populated selectively.

3. Paul Trap Stabilisation

The classical approach to saddle dynamics is most helpful for giving some intuitive arguments explaining the surprisingly high efficiency of saddle dynamics with respect to excitation by intermediate-energy projectiles. According to this approach, the efficiency results from a stabilisation of the electron's motion on the saddle accomplished by the rotation of the saddle during the collision. Due to this rotation, the initial unstable motion of the electron along the symmetry axis of the saddle is turned into a stable oscillating motion perpendicular to the symmetry axis, provided the rotation is fast enough. If the angular velocity $\omega_{\text{rot}} \approx v_s/b_s$ of the rotation is at least of the order of the frequency ω_{osc} of the oscillating motion,

$$\omega_{\text{rot}} \geq \omega_{\text{osc}}, \quad (2)$$

the electron's motion cannot follow the rotation adiabatically. Taking into account that $b_s \approx 1$ a.u. and also that $\omega_{\text{osc}} \approx 1$ a.u. at the moment of closest approach, one finds that stabilisation takes place only if the saddle velocity v_s is also at least of the order of 1 a.u.

This stabilising mechanism due to rotation is basic for Paul traps, where particles are captured on a permanently rotating (or oscillating) saddle potential. Regarding collision processes, both ω_{rot} and ω_{osc} depend on time. They scale with R_s^{-2} and $R_s^{-3/2}$, respectively, where $\mathbf{R}_s(t)$ is the (straight-line) saddle trajectory (von Oppen 1994). For the sake of simplicity, we shall here consider a saddle rotating with a fixed frequency (Paul 1990). The motion of charged particles in such Paul traps has also been analysed quantum dynamically (Brown 1991). However, a classical description suffices here. In spite of these simplifications, the model can help to gain some insight into the stabilisation mechanism and allow some important conclusions.

In a body-fixed frame with cartesian coordinates (χ, η, ς) and the saddle in its centre, the potential near the saddle point is given by

$$V(\chi, \eta, \varsigma) \approx \frac{1}{2} \gamma (\chi^2 + \eta^2) - \gamma \varsigma^2, \quad (3)$$

where $\gamma = (\sqrt{q} + 1)/(\sqrt{q} R_s^3)$. Transforming this potential function to a space-fixed coordinate system (x, y, z) , one obtains the following differential equation for the electron's motion on the saddle in the $x-z$ (scattering) plane:

$$\begin{aligned} \ddot{x} + \gamma \left\{ x \left[\frac{3}{2} \cos(2\omega t) - \frac{1}{2} \right] - \frac{3}{2} z \sin(2\omega t) \right\} &= 0, \\ \ddot{z} + \gamma \left\{ \frac{3}{2} x \sin(2\omega t) - z \left[\frac{3}{2} \cos(2\omega t) + \frac{1}{2} \right] \right\} &= 0. \end{aligned} \quad (4)$$

Disregarding the tangential components of force, which in first order do not affect the stabilisation, the electron's motion follows the Mathieu equations (Paul 1990):

$$\ddot{x} + (a - 2p \cos 2\tau) x = 0, \quad \ddot{z} + (a + 2p \cos 2\tau) z = 0, \quad (5)$$

where $\tau = \omega t$, $p = \frac{3}{4}(\gamma/\omega^2)$ and $a = -\frac{1}{2}(\gamma/\omega^2)$.

For $a = 0$, the Mathieu equation has stable solutions if $p < 0.9$, that is $\omega^2 > 0.9 \times \frac{3}{4}\gamma$. With $\omega = \omega_{\text{rot}}$ and $\gamma = \omega_{\text{osc}}^2$, one finds the stability condition $\omega_{\text{rot}} > 0.8 \omega_{\text{osc}}$ in accord with the estimate (2).

Actually one has to take into account the offset a with $a/p = -\frac{2}{3}$, which results from the fact that the destabilising force in the ς direction is twice as large as the stabilising force in the χ direction. Therefore, stabilisation is more exceptional than expected according to (2). However, here we are not interested in a permanent stabilisation on a constantly rotating saddle, but in a transient stabilisation on the potential saddle during a collision. The foregoing consideration suggests that such a transient stabilisation is most likely according to the stability diagram of Mathieu's equation (Paul 1990), if

$$\omega_{\text{rot}} \approx 0.8 \omega_{\text{osc}}, \quad (6)$$

but it may be also effective to some extent if $\omega_{\text{rot}} > 0.8 \omega_{\text{osc}}$. These conclusions are confirmed by the experimental results discussed in the following sections.

4. Collisional Excitation by Charge Conjugated Projectiles

The first experimental evidence favouring the Paul trap model comes from a comparison of stopping powers (Schmidt *et al.* 1998) and ionisation cross sections (Knudsen and Reading 1992) measured for collisions with charge conjugated projectiles. These cross sections are charge independent in the high-energy limit where Born's approximation applies, but are, however, significantly larger for positive projectiles than for negative ones in the intermediate-energy range. Regarding heavy particles as protons and antiprotons or positive and negative muons and pions, there is the well known Barkas effect (Linhard 1976). The stopping power in gases and solid matter is substantially larger for positive than for negative particles, if their velocity is of the order of 1 a.u. Similarly, also for the charge conjugated light particles, electrons and positrons, one finds that the ionisation and also the excitation cross section for positron impact are larger than for electron impact in a limited ($E \leq 100$ keV) energy region above the excitation threshold (Charlton and Laricchia 1990).

Usually these differences in the excitation cross sections of charge conjugated particles are explained by referring simply to the polarising influence of the charged projectile on the electron distribution of the target atom. At intermediate energies, the electron density seen by the projectile rises for positive and is diminished for negative projectiles due to the polarisation of the electron cloud. In the light of the idea of Paul trap promotion, this explanation seems incomplete. A positive projectile can shift the electron on the saddle of the two-centre potential, where it is efficiently promoted to higher energies. However, such a saddle is missing in the case of singly-charged negative-particle impact. Therefore, Paul trap promotion does not contribute to excitation and ionisation in collisions with negative projectiles.

5. Resonant Paul Trap Promotion

Due to the non-zero offset a in the equations of motion (5), Paul trap promotion is likely to have a resonance-like efficiency, where $\omega_{\text{rot}} \approx 0.8 \omega_{\text{osc}}$. Such a resonance of excitation cross sections has indeed been measured for proton impact on He (van den Bos *et al.* 1968). Close to the effective excitation threshold, the emission cross sections $\sigma(n^1\text{D})$ of the $\lambda(1snd\ ^1\text{D} - 1s2p\ ^1\text{P})$ spectral lines ($n = 3$ to 6) rise to a narrow maximum at a proton energy $E_p \approx 15$ keV, and only at $E_p \approx 50$ keV is a broad and lower second maximum reached, which roughly corresponds to the cross section maximum expected according to Born's approximation (Massey and Gilbody 1974). Corresponding structures at $E_p \approx 15$ keV are also found in the excitation functions of the $1snp\ ^1\text{P}$ ($n = 3$ to 5) (van den Bos *et al.* 1968) and $1s4f\ ^1F$ levels (Aynacioglu *et al.* 1987).

These pronounced cross section maxima close to the effective threshold can be explained by referring to resonant Paul trap promotion where $\omega_{\text{rot}} \approx 0.8 \omega_{\text{osc}}$. The $(\text{H-He})^+$ collision system is similar to H_2^+ with respect to single-electron excitation. During the collision, the active electron moves in the two-centre potential of the proton and the He^+ core consisting of the He nucleus and the passive (spectator) electron. Therefore, it is reasonable to apply the theory of saddle dynamics as outlined for the H_2^+ system in Section 2.

Since the proton velocity $v_p \approx 0.8$ a.u. is still below 1 a.u. at the resonant maximum, a quasi-molecular evolution along an in-saddle sequence is likely. Assuming a close correspondence between resonant Paul trap promotion and promotion along in-saddle sequences, one is led to the conclusion that parabolic Stark states with $n_2 = 0$ are mainly populated by these collisions. This conclusion is strongly supported by measurements of Buettrich and von Oppen (1993) who investigated the charge distributions of He atoms excited by 12.5 keV proton impact.

In these measurements, the transient states, in which the He atom is found immediately after a collision, were determined by recording the fluorescence light intensity of He I spectral lines as a function of an electric field F_z applied parallel ($F_z > 0$) and antiparallel ($F_z < 0$) to the proton beam (Fig. 1). Since primarily only He I singlet states are excited by proton impact (Aynacioglu *et al.* 1987), usually only singlet lines are emitted. However, when scanning the electric field, a series of singlet–triplet anticrossings is passed, where singlet and triplet states are strongly mixed (Kaiser *et al.* 1993). These anticrossings give rise to a resonance-like appearance of triplet lines (and corresponding resonance-like dips in the singlet-line intensities). Both the measured intensity functions $I_1^n(F_z)$ of singlet transitions $1snl - 1s2p$ 1P and the intensity functions $I_3^n(F_z)$ of triplet transitions $1snl - 1s2p$ 3P are extremely asymmetric with respect to the sign of F_z , thereby reflecting the extreme asymmetry of the collisionally excited state. A thorough evaluation (Buettrich *et al.* 1998) confirmed that indeed the parabolic $^1\Sigma$ and $^1\Pi$ states with $n_2 = 0$ are predominantly populated as suggested by Fig. 1.

6. Coulomb Excitation at Intermediate Energies

Though Paul trap promotion is most likely, if the resonance condition $\omega_{\text{rot}} \approx 0.8 \omega_{\text{osc}}$ is fulfilled, it contributes to excitation also at higher intermediate energies. This conclusion is supported by investigations on the collisional excitation of He I states by proton impact up to energies $E_p = 500$ keV (Drozdowski *et al.* 1998) and by HCI impact (von Oppen *et al.* 1998). In these experiments, again the intensity functions $I(F_z)$ of the He I spectral lines were recorded, but for an electric-field range ($-30 < F_z < +30$ kV cm $^{-1}$) considerably larger than that scanned in the 12.5 keV experiments (see Fig. 2). The analysis of these recordings revealed that, even at energies up to about $E_p \approx 300$ keV, proton impact leads to highly coherent collisionally excited He I states. However, these states are different from the parabolic states populated by 12.5 keV-proton impact. With increasing proton energy, the high-angular momentum components with $l \geq 3$, which are strongly present in all parabolic states with $n \geq 4$, disappear. At proton energies $E_p > 100$ keV the collisionally excited $n^1\Lambda$ states of He I are well represented as completely coherent superpositions of the low-angular momentum states with $l \leq 2$ (see Figs 2b and 2c). In the high-energy limit ($E_p > 300$ keV) excitation of the $1snp$ 1P levels is dominant by at least two orders of magnitude as expected according to Born's approximation. Even at the relatively high proton energies up to $E_p \approx 300$ keV, the measured intensity functions are still strongly asymmetric (Drozdowski *et al.* 1998). This asymmetry, which is particularly pronounced for the pattern of anticrossing resonances, indicates that the coherent superpositions of low- l states also have large electric dipole moments directed upstream, that is the electron cloud is shifted downstream with respect to the He nucleus as for 12.5 keV-proton impact.

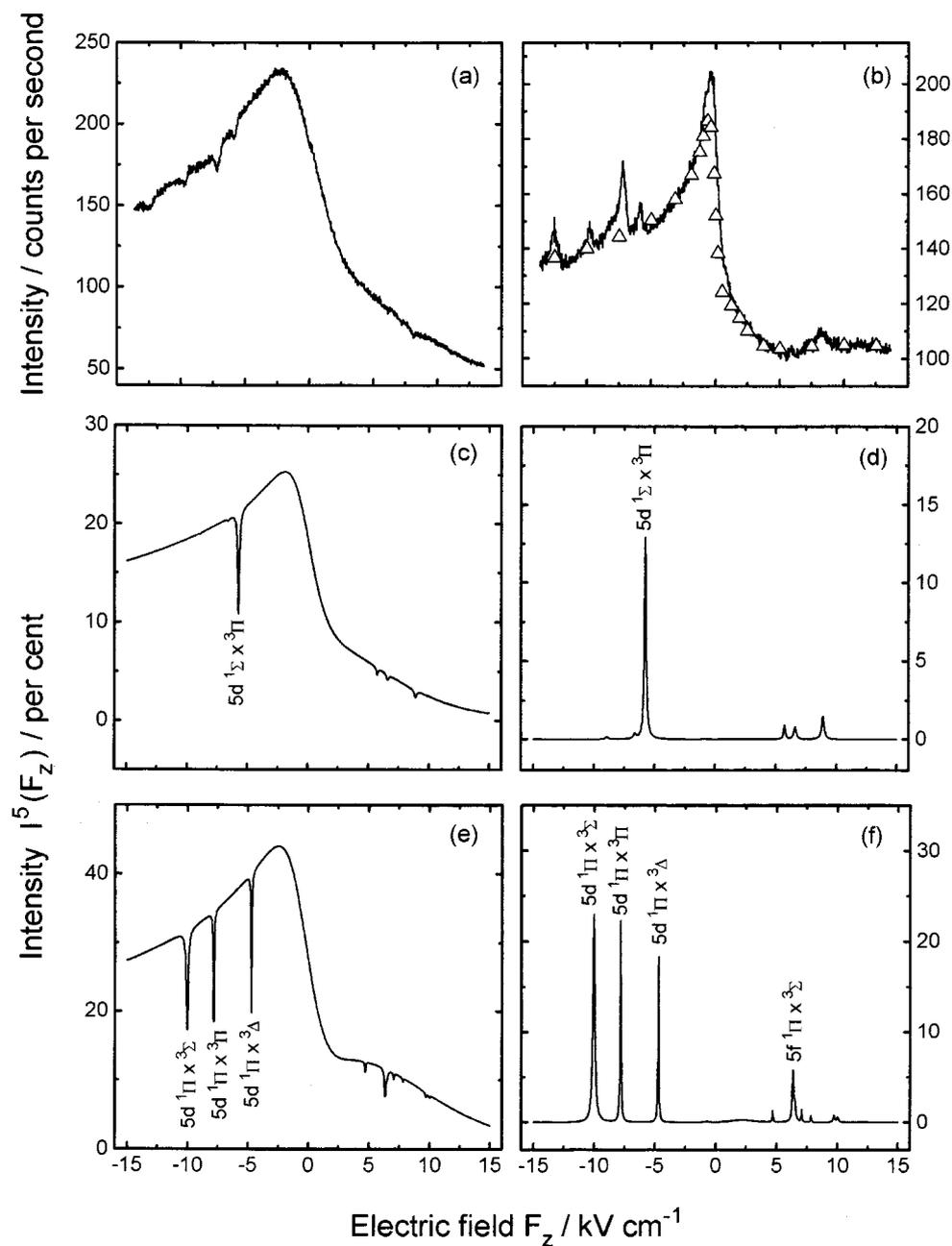


Fig. 1. Measured (a,b) and calculated (c-f) intensity functions $I_\lambda(F_z)$: (a) $\lambda(1s5l - 1s2p^1P) = 439$ nm and (b) $\lambda(1s5l - 1s2p^3P; 1s7l - 1s2p^1P) = 401$ nm, both for 12.5 keV proton impact on helium. (Triangles give the calculated singlet component of the 401 nm recording.) The theoretical curves were calculated assuming selective excitation of the parabolic $^1\Sigma$ state $|5; 4, 0, 0\rangle$ (c,d) and $^1\Pi$ states $|5; 3, 0, 1\pm\rangle$ (e,f) and decay to the $1s2p^1P$ level (c,e) and $1s2p^3P$ level (d,f). The singlet-triplet transitions are due to anticrossings as indicated (from Buettrich *et al.* 1998).

The high coherence of the collisionally excited states together with the asymmetry of the recorded intensity functions strongly supports the assumption that Paul trap promotion is also important at these higher intermediate energies and that a narrow range of impact parameters is decisive for collisional excitation. These impact parameters can be assumed somewhat larger than 1 a.u. to account for the fact that only low- l states are populated, a circumstance indicating that the target atom is less strongly disturbed by the Coulomb field of the projectile and a second-order perturbation approach is applicable.

In conclusion, these measurements on the excitation of He atoms by proton impact have led to an intuitive visualisation stressing some global features of these intermediate-energy collisions. The Paul trap model bridges the gap between the molecular orbital model valid in the low-energy range and the Born approximation applicable to high-energy collisions. Resonant Paul trap promotion can still be understood as an MO-like process. But with increasing impact energies, the impact parameter of collisions leading most effectively to excitation becomes larger. Therefore, a second-order and finally a first-order approximation is justified in the high-energy limit.

According to this model, for excitational collisions the impact parameter of the saddle trajectory is larger or at least as large as 1 a.u. Therefore, one expects that the dipole approximation is valid, where the perturbation is essentially determined by the electric field $\mathbf{F}(0,t)$ induced by the projectile at the site of the He nucleus at $\mathbf{r} = 0$. Under these conditions, the Janev-Presnyakov (JP) (1980) scaling law should apply. That is, impact excitation by ions with charge $q > 1$ and mass number A is expected to induce the same collisionally excited states as proton impact provided collisions with the same scaled energy

$$E_0 = E_{\text{ion}}/Aq \quad (7)$$

are considered. Strong evidence for the validity of the JP scaling law in the energy range $20 \leq E_0 \leq 1000$ keV was provided already by Anton *et al.* (1993), who measured the relative population of 1s4d ^1D Zeeman sublevels after proton and HCI impact excitation.

By analysing the charge distribution of states collisionally excited by HCI impact in the energy range $125 \leq E_0 \leq 333$ keV, we showed that the JP scaling law applies to the coherence parameters as well (Tschersich 1998; von Oppen *et al.* 1998). They measured the intensity functions $I(F_z)$ for various He I spectral lines induced by $^{40}\text{Ar}^{q+}$ impact with $q=6, 13$ and 14 . Well resolved anticrossing signals with a high signal-to-noise ratio were measured, in particular, for the 1s4l – 1s2p ^3P transition (Fig. 2a). This intensity function is surprisingly similar to the intensity function of Fig. 2b measured for proton impact at an approximately equal scaled energy. Both recordings are well reproduced theoretically (Fig. 2d) by assuming that the completely coherent superposition states

$$\begin{aligned} |4\ ^1\Sigma\rangle_{\text{coll}} &= -0.932 |4\ ^1\text{P}, 0^+\rangle + 0.362 |4\ ^1\text{D}, 0^+\rangle, \\ |4\ ^1\Pi^\pm\rangle_{\text{coll}} &= -0.964 |4\ ^1\text{P}, 1^\pm\rangle + 0.266 |4\ ^1\text{D}, 1^\pm\rangle \end{aligned} \quad (8)$$

are populated in the ratio $\sigma(4\ ^1\Sigma)/\sigma(4\ ^1\Pi) = 0.536$.

The similarity is even more evident when the signal broadening due to the inhomogeneity of the electric field is taken into account (Fig. 2c). The only discrepancies obvious for some anticrossing signals at fields $F_z > 0$ can be related to cascade feeding neglected in the calculation.

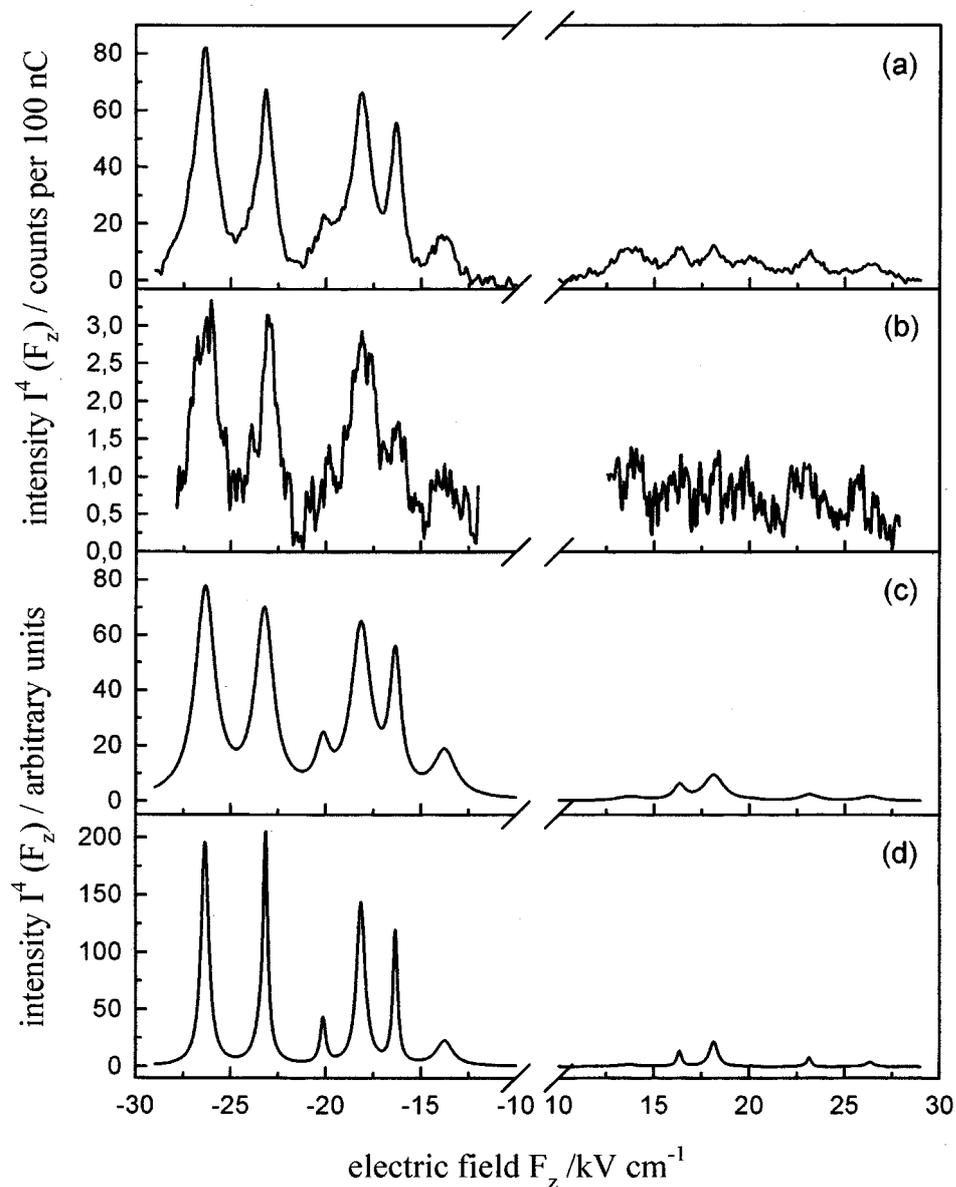


Fig. 2. Measured (a,b) and calculated (c,d) intensity functions $I_\lambda(F_z)$ for $\lambda(1s4l - 1s2p \ ^3P) = 447$ nm: (a) for 65 MeV $^{40}\text{Ar}^{13+}$ impact on helium ($E_0 = 125$ keV); (b) for 200 keV proton impact on helium; (c) pattern of anticrossing resonances calculated assuming the state vectors given by (8) as collisionally excited states and taking into account the broadening of the signals due to the inhomogeneity of the electric field; and (d) same as (c), but without inhomogeneity broadening.

7. Conclusions

We have tried to draw attention to some common features of atomic collisions at intermediate energies, which can be qualitatively understood within the framework of the Paul trap model. In particular, experiments on collisional excitation of He atoms by proton and HCl impact reveal a characteristic property and a universal energy dependence of the collisionally excited states irrespective of their principal quantum number n . These transient states have large electric dipole moments and are surprisingly coherent. This coherence is considered as strong evidence for the conclusion that a narrow range of impact parameters is decisive for excitational collisions. Furthermore, the universal energy dependence of the collisionally excited states suggests that these excitational collisions proceed via a particular excitation mechanism.

Referring to classical dynamics, we propose the Paul trap model for providing an appropriate description of excitational collisions at intermediate energies. According to this model, excitation proceeds via electron promotion on the saddle of a two-centre Coulomb potential, where the electron's motion is stabilised due to the rotation of the molecular axis of the collision system. Using this simple model, various prominent features found experimentally for p-He and HCl-He collisions could qualitatively be explained.

The Paul trap model sheds new light also on a striking, but well-known difference between the excitation cross sections of collisions using charge conjugated projectiles. Paul trap promotion can contribute only to excitation by positively charged projectiles. Since different excitation cross sections are measured not only for charge conjugated heavy particles, but also for positrons and electrons, we conclude that Paul trap promotion takes place not only in heavy-particle collisions, which can be described semiclassically, but also for positron-impact excitation, where the projectile cannot be localised and its motion should be represented by a wave.

Therefore, the classical description of the Paul trap mechanism presented in this paper should be considered only as a guide allowing a first orientation in the sense of the correspondence principle. Theoretical approaches to the Paul trap mechanism, where quantum dynamical effects are accounted for, and which can, in particular, be applied also to positron impact, have still to be developed.

Acknowledgment

We are grateful to Dr H. Kerkow for drawing our attention to the publications on the Barkas effect.

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Manuscript received 2 October 1998, accepted 1 February 1999