Experimental Evidence of the Asymmetry of the Soft Electron Peak in Ion-Atom Ionization

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We have measured the doubly differential energy and angular distribution of low-energy electrons emitted in collisions of H^+ and $^3He^{2+}$ on Ne at 106 keV/u. In this way, we are able to obtain information about the shape of the soft electron ionization peak. Against current belief, but in accordance with a two-Coulomb-center interaction of the emitted electron, we find it to be strongly asymmetric in the forward-backward direction. We discuss the shape of the cross section and introduce a parametric expression for fitting the data.

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There is no doubt about the existence of a strong soft electron (SE) peak in the spectra of electrons emitted in ion-atom collisions. The corresponding cross section $d\sigma/d\mathbf{v}$, doubly differential in the direction and modulus of the electron velocity ($\mathbf{v}$), is expected to have a $1/v$ divergence in the limit $v \to 0$. Consequently, the energy distribution $d\sigma/dE\,d\Omega$ should be a smooth function for small electron energies (E). Until now, no measurements have been presented that permit a discussion of the shape of the SE peak, mainly because of the well-known experimental difficulties for the accurate analysis of electrons propagating with small velocities.

The SE peak is one of the three important features of the spectra which give direct evidence of the electron-ion Coulomb interaction, in ion-atom and atom-atom ionization. Meanwhile much attention has been paid to the two remaining reactions, i.e., electron capture to the continuum (ECC) and electron loss to the continuum (ELC). This paper is the first attempt to study the shape of the SE peak.

Fast collisions accompanied with small momentum transfer are usually treated with the first Born approximation (FBA), in which the triply differential cross section (TDCS), differential in the velocity ($\mathbf{v}$) of the emitted electron and momentum transfer ($\mathbf{K}$) of the scattered bare ionic projectile, is given (in atomic units) by [1]

$$\frac{d\sigma}{d\mathbf{v}\,d\Omega_{\mathbf{K}}} = \frac{4Z^2}{V}\frac{1}{K^2(V^2-2E_i)} f_{\mathbf{v},i}(\mathbf{K}) , \quad (1)$$

where $f_{\mathbf{v},i}(\mathbf{K})$ is the generalized oscillator strength. Here Z and $\mathbf{V}$ are the charge and initial velocity of the projectile, $\mathbf{K}$ the momentum transfer during the collision, and E_i the energy of the initial bound target state, with quantum numbers $i=n,l,m$.

The oscillator strength can be expanded in spherical harmonics, giving [2]

$$f_{\mathbf{v},i} = \sum_J A_J(i,v,K) P_J(\cos\theta_{\mathbf{K}}) , \quad (2)$$

where $\theta_{\mathbf{K}}$ is the angle between $\mathbf{v}$ and $\mathbf{K}$.

The doubly differential cross section (DDCS) in the velocity $\mathbf{v}$ is obtained by integration on $\mathbf{K}$ and gives [2]

$$d\sigma/d\mathbf{v} = (1/v)\sum_L C_L(i,V,v) P_L(\cos\theta) , \quad (3)$$

where θ is the angle between $\mathbf{V}$ and $\mathbf{v}$. We write explicitly the $1/v$ divergence associated with the asymptotic Coulomb potential of the residual target ion.

For a $1s$ initial state and a Coulomb final wave for the electron, Eq. (3) can be written as a double series expansion [3,4],

$$d\sigma/d\mathbf{v} = (1/v)\sum_{L,k} B_L^{(k)}(V) v^k P_L(\cos\theta) , \quad (4)$$

which, within the considered FBA, has the constraint $B_L^{(k)}=0$ for $k+L$ odd and for $L>k+2$. For a generic initial state Burgdörfer *et al.* [5] studied the limit $v\to 0$ of Eq. (3) and obtained a finite expansion

$$d\sigma/d\mathbf{v} = (1/v)\sum_{L(\text{even})}^{2n} C_L(i,V,v=0) P_L(\cos\theta) , \quad (5)$$

where $C_L(i,V,v=0)=B_L^{(0)}$. For a $1s$ initial state only two multipoles contribute, as found earlier by Briggs and Day [6], who apply Eq. (5) to obtain a soft electron peak of forward-backward symmetry. In connection with a discussion of ELC these calculations were performed in a reference frame moving with V, assuming an effective charge for the target atom which in that frame acts as a projectile of velocity $-V$. However, this target atom is neutral and will not play the role of a second Coulomb center which distorts the electron cloud following the emerging ion. Consequently a verification of symmetric ELC could not be considered as a proof for the applicability of a first-order perturbative treatment. On the other hand, there is evidence for some asymmetry of measured ELC peaks which, however, cannot be considered as consistent with respect to magnitude and direction [7].

Recently it has been shown that the expansion given by Eq. (3) has a validity more general than that given by the Born approximation [8], and contributions from odd Legendre polynomials are possible. The coefficients C_L have been expressed in terms of the state multipoles for

H^+-atom collisions and their values discussed when they are extrapolated below threshold [8].

For ECC the first-order Brinkman-Kramers approximation gives also a symmetric cusp; however, for bare projectiles, experiments show consistently a strong asymmetry, which is due to the long-range Coulomb attraction by the residual target ion. In this connection a large amount of work has been devoted to the introduction of second-order effects.

Photoionization studies provide another source of theoretical information about the possible angular shape of the SE emission. The electric field of the photon imparts to the electron an impulsive force equivalent to that produced by a glancing collision of an ion, with large impact parameter and small momentum transfer [9]. Therefore the angular distribution of the soft electrons emitted from a target by ion impact would be similar to that of photoelectrons. However, this should be valid when small K dominate, i.e., for slow electrons ejected by very-high-energy projectiles.

An expression formally equivalent to Eq. (2) was obtained for the electron distribution resulting from photoionization [10]. In this case $\theta_{\mathbf{K}}$ must be replaced by the angle $\theta_{\mathbf{j}}$ between $\mathbf{v}$ and the polarization vector $\mathbf{j}$ of the incident photon, and v is determined by the energy of the photon. The coefficients are evaluated in the dipole approximation, neglecting retardation, using the optical oscillator strength:

$$f_{\mathbf{v},i}(0)=f[1+b(i;v)P_2(\cos\theta_{\mathbf{j}})]\,. \quad (6)$$

In that case we have contributions only from the two first even-order terms in the expansion (3). This equation is valid in the electric dipole approximation and the coefficient b gives an approximate value to the ratio C_2/C_0 in Eq. (5), which results from neglecting high-order multipoles. The anisotropy parameter $b(i;v)$ has been evaluated for the photoionization of electrons initially in different shells of noble gases and the outgoing electron was described by different Hartree-Fock wave-function formalisms [11]. The value of $b(i;v)$ depends on the target species, the state from where ionization takes place and the final electron velocity. Calculations show that the value of $b(i;v)$ depends strongly on these conditions, in particular for very small v [10]. The point that we would like to emphasize from this comparison is that the sensitive dependence of $b(i;v)$ on the electron velocity suggests that the coefficients C_L could have a strong variation on the wings of the SE cusp. Consequently cross sections derived in the $v\rightarrow 0$ limit could only be reliable at the very top of the SE peak. This possible strong v dependence of the C_L has been already suggested by Burgdörfer [8]. As in the ELC case, in photoionization, we have no postcollisional electron-projectile interaction and the first-order perturbative approach gives an appropriate description.

We have extended previous measurements [12] by determining the spectra of low-energy (E) electrons emitted in the collision of 106 keV/u H^+ and $^3He^{2+}$ ions with neon gas. The experimental array and details have been described previously [13] and, with reference to the measurements of the soft electrons, will be discussed in a forthcoming paper. For fixed emission angle θ the electrons are energy analyzed with a cylindrical mirror electrostatic analyzer, which has an energy resolution $\delta E/E \approx 0.4\%$ and angular acceptance $\theta_0=2°$. The angle θ can be varied continuously in the full range from 0° to $\pm 180°$. This also allows for a control of the angular accuracy by comparison of θ and $-\theta$ spectra; the resulting error amounts to 15% for $E<10$ eV and decreases for larger E. The spectra were normalized relative to each other according to the pressure in the collision chamber and to the collected beam charge. We estimate that uncertainties in the determination of the pressure in the collision area introduce an error of $\approx 2\%$. The spectra for different angles are relatively normalized by a control measurement of the angular distribution at a fixed value of E. Absolute values have been derived by integration of the spectra, over energy and angle, and normalization to recommended values of the total cross section [14]. However, they are not essential in the context of the present study. Statistical counting errors, relevant for low-energy electrons, have been estimated to be about 12% for $E=2$ eV.

The resulting spectra for $0.5 \le E \le 20$ eV are shown in Figs. 1 and 2, for H^+ and $^3He^{2+}$, respectively. We represent $d\sigma/dE\,d\Omega$ for different θ and we have included estimated error bars. The corresponding velocity range for these spectra is $0.19 \le v \le 1.21$ a.u. We observe that the energy distributions have a behavior which can be considered as flat in the low electron energy range, within the limits of the experimental accuracy. They decrease as the angle grows up to 120° and then increase for larger angles. Our main evidence is the large forward-backward difference of $d\sigma/dE\,d\Omega$, which we also display

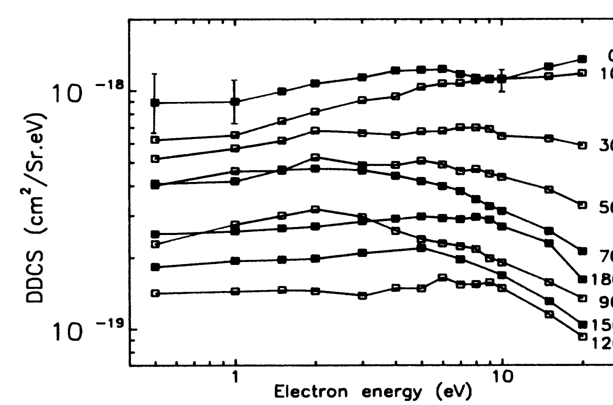

FIG. 1. Double differential energy distributions of electrons emitted at fixed angles θ when H^+ of 106 keV/u interacts with Ne.

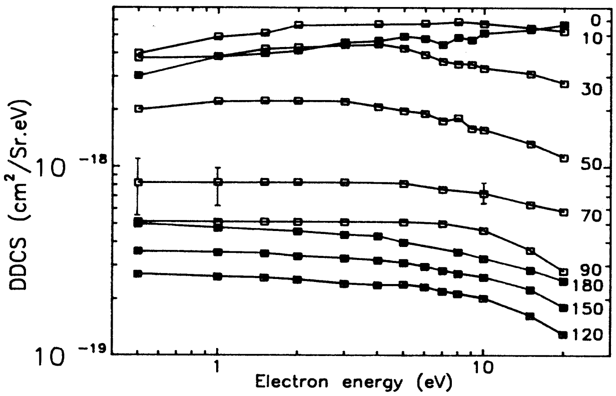

FIG. 2. Same as Fig. 1, for $^3He^{2+}$ projectiles.

in the angular spectra seen in Figs. 3 and 4. This corresponds to a strong asymmetry in the wings of the SE peak that is clear down to the lower electron velocity of $v=0.19$ a.u. covered by our measurements. An extrapolation towards $v \approx 0$ shows that the asymmetry should remain in this limit.

Now we try to express our data as in Eq. (3). From the observed forward-backward differences we note that an expansion containing even-order Legendre polynomials is not enough for the description of the experimental data. The post-collisional interaction between the emitted electron and projectile must be considered as the cause for appearance of the odd-order coefficients in Eq. (3) [8,15]. However, theoretical values for these coefficients are not yet available for ionization.

A choice must be made as to the number of terms to consider in Eq. (3). We will try to fit the data with the simpler expression that results from considering two terms. We write

$$d\sigma/dE\,d\Omega = A[1+\beta_1 P_1(\cos\theta)+\beta_2 P_2(\cos\theta)]\,. \qquad (7)$$

Here we have three coefficients available for each of the fits to the H^+ and the $^3He^{2+}$ data. We can reduce this number by applying the rough assumption that A and $A\beta_2$ come mainly from the first perturbative order and therefore will depend on the charge of the projectile as Z^2, whereas $A\beta_1$ is due to the second-order contributions and will vary as Z^3. This requires that higher-order terms in Z are neglected in the cross section. This hypothesis leaves us with three parameters for a simultaneous fitting of the H^+ and $^3He^{2+}$ data. The factor A only depends on the absolute normalization of the data and, as we say above, is not relevant for the study of the cusp shape.

In Figs. 3 and 4 we observe a subtle but systematic variation of the angular distribution with v. However, the experimental errors obscure an accurate analysis of that variation, i.e., a study of the dependence of the coefficients on the velocity. Therefore, for each θ we have evaluated an average of the data in the low electron energy range $0.5 < E < 5$ eV, and we tried to fit these mean values with Eq. (7). The resulting value for the asymmetry and anisotropy parameters are $\beta_1=0.72$ and $\beta_2=0.61$, respectively. (The normalization factor results in $A=0.35\times10^{-18}$ cm²/eV sr). We have verified that these values for the coefficients are numerically stable. The fit curves are represented by solid lines in Figs. 3 and 4. We note that the values obtained for β_1 and β_2 are of the same order as those derived from theoretical studies of the excitation cross sections [8].

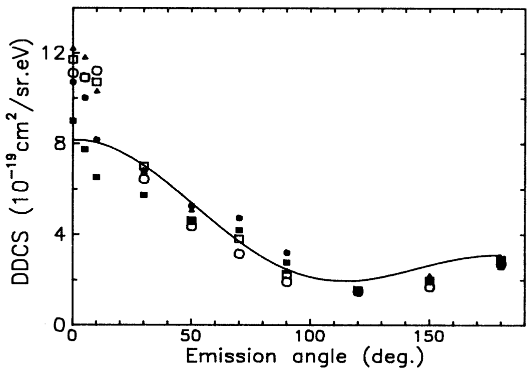

FIG. 3. Angular distributions for electrons emitted at fixed energy when H^+ of 106 keV/u interact with Ne. Electron energies: (■) 1 eV; (●) 2 eV; (▲) 5 eV; (□) 7 eV, and (○) 10 eV. The solid line results from fitting data with Eq. (7).

An improvement of the fitting could be obtained by introducing additional Legendre polynomials in the expansion. It is most probable that the emitted electron comes from an external $2p$ shell of Ne, and Eq. (5) indicates that terms up to P_4 could give contributions to Eq. (3). An additional possibility is to release the scaling with Z, which is not a secure premise at the 106 keV/u impact energy of our measurements. Under these conditions we would stay with four asymmetry coefficients for each projectile, which would allow for a more precise fitting.

In spite of the fact that until now emission of soft electrons has been believed to be symmetric, the now ob-

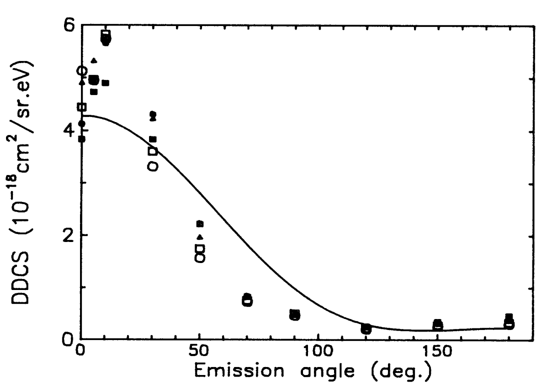

FIG. 4. Same as Fig. 3, for $^3He^{2+}$ projectiles.

served strong asymmetry is in accordance with the fact that there is a postcollisional attraction of the emitted electron, not only with the residual target ion but also with the receding projectile. The ridge-shaped structure joining the SE and ECC peak continues, with increasing v, into the well-known negative skewness of the ECC cusp [16], and with decreasing v, into the now discovered positive skewness of the SE peak. It is then not surprising that both these asymmetries are of the same order of magnitude.

The principal conclusion of this work is that our experiment shows the first evidence that the SE peak is strongly asymmetric. Furthermore we can expect that this asymmetry (a) further increases with Z, (b) decreases for larger V, and (c) increases when the effective residual target charge decreases. In view of the divergence of the SE emission for $v \rightarrow 0$ we can say that these findings may give rise to a renewed cuspology.

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