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Electron Transfer to Continuum States

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Gene Rudd's analysis of doubly differential cross sections for the ionization of He atoms by proton impact suggested that electrons were being carried along by the proton for a short period of time after being ejected from the target region. Normally, this would represent an electron capture event in which an excited state of atomic hydrogen is formed. Because the electron ends up ionized it was recognized that these states of the proton must be continuum states. This insight was confirmed by observations of the continuum electron capture (CEC) cusp when the electron velocity equals the proton velocity in the final state. The impact of this idea upon the theory of ionization at high energies is reviewed.

I. INTRODUCTION

It is a great honor for me to speak at this symposium commemorating Gene Rudd's accomplishments in physics. These accomplishments are many and are amply recounted by other speakers at this symposium. My talk concentrates on the discovery of what is now known as the continuum capture cusp. Probably we should call it the Rudd peak in honor of its discoverer. In any event, the occasion recalls the great fun Gene, his students, and I had trying to understand ionization in ion-atom collisions. It is a pleasure to recount the discovery of the continuum capture mechanism, not the least because Gene was my patient mentor during my first ventures into this field. I am forever indebted to him for bringing me into his research program to see doubly differential cross sections through his eyes. Atomic units are used in this manuscript although Gene always advocated SI units.

II. DISCOVERY OF THE CONTINUUM CAPTURE CUSP

Gene's work first came to the attention of the general physics community when he observed (1) Auger electrons produced in $Ar^+ + Ar$ collisions, thereby

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confirming the Fano-Lichten (2) model that accounted for structure in the energy loss of Ar^+ ions observed by Kessel and Everhart (3). The research that Gene did on this process is well known. That process represents a mechanism for ionization quite different than the one of interest here. There is a connection, however, in that one of Gene's early discoveries was that Auger states of both the target ion and projectile ion were excited (4). These were distinguished experimentally by the Doppler shift of the projectile electrons. While this is a simple observation, it was quite important at the time in that it confirmed another aspect of the Fano-Lichten model, namely, electron promotion occurred in a region of space where electrons from the projectile and from the target participated equally. The Physical Review Letter on projectile Auger electrons excited much interest in Professor Fano's group at the National Bureau of Standards. It represented elegant, convincing, evidence for the model, so much so that Professor Fano remarked at the time, "I wish I had thought of that!" I suspect that the Doppler effect set Gene to thinking along the lines that eventually led to the theory of continuum electron capture.

During one of our many discussions about doubly differential cross sections in my first summer at the University of Nebraska, Gene showed me his analysis of the data that he had published together with Sautter and Baily (5). He had developed a First Born Approximation program to compute the theoretical doubly differential cross section (DDCS) in order to interpret the measurements. The computed and measured electron distributions generally agreed fairly well except for those electrons ejected at small and large angles. There was an excess electrons at small angles for almost all electron velocities, but the relative excess was most pronounced at electron velocities equal to the projectile velocity. This led Oldham (6) to suggest that the electron-proton interaction was playing a role not well represented in the first order calculations. To analyse the role of the projectile, Gene plotted the position of a peak in the electron velocity distribution vs the variable $v \cos \theta$ where v is the projectile velocity, and θ is the angle of ejection of the electron relative to the beam direction. Such a plot is shown in Fig.(1) where you can see that the peak position is linearly related to $\cos \theta$. This is not the plot that Gene actually showed me, since atomic units are used whereas Gene always employed SI units.

This figure was the key to the puzzle; the extra electrons appeared to come from the projectile just like Doppler shifted Auger electrons. Gene therefore posed the question, "Are there states of the hydrogen atom that would produce this Doppler shifted feature?" We discussed this idea at length and exhausted several possibilities; high Rydberg states subsequently stripped by secondary collisions would not appear at angles as large as 30° and a substantial neutral component of the primary beam was unlikely. But Gene's analysis suggested that the basic assumption of the First Born Approximation, namely that the electrons in the final state are target eigenstates, was incorrect. Perhaps one should try using projectile eigenstates in the First Born Approximation. Gene encouraged me to work out this idea, and it turned out

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FIG. 1. Plot of the peak position of excess electrons vs $v \cos \theta$.

to give the right shape to the electron angular distribution for electrons whose velocity matched the primary beam velocity of the protons.

Fig.(2) shows our comparison of the data of Ref. (5) with computations based on this picture. Because the angular distribution agreed well in shape, we were encouraged that we were on the right track. My first paper using this idea was rejected since the use of projectile eigenstates was, in the words of the referee, "purely ad hoc." Fortunately, Francisco Pratts had taught me about Faddeev's equations during my stay at the National Bureau of Standards so it was easy to use these equations to justify projectile eigenstates and the computations were eventually published (7). Meanwhile Gene noted that the calculations must predict an infinite cross section when the electron velocity exactly matches the proton velocity in the final state. He immediately saw that this provided a way to probe the continuum capture mechanism experimentally.

The problem of experimentally measuring electrons ejected at zero degrees was posed to a very capable graduate student, Jeff Crooks. He was given a two week timetable to check out the zero degree electron velocity distribution experimentally. He just completed it in the two week time frame, and his results (8) are shown in Fig.(3). They confirmed the infinite cross section at $k = v$, where k is the electron velocity in the laboratory frame, and firmly established the "continuum electron capture" mechanism.

After this seminal discovery, Gene's research focused on more global questions related to ionization by charged particle impact. However, the con-

FIG. 2. Plot of measured and calculated DDCS for the ionization of He by 300 keV proton impact. The measurements are from Ref. (5) and the calculations are from Ref. (7).

tinuum capture cusp was part of that global problem. We both sought a description of the cusp that did not depend upon detailed calculations involving all sorts of approximations. To this end Gene recalled a key formula from his Doppler shift studies, namely the transformation from the lab to the projectile frame. This transformation is implicit in the replacement of target electron wave functions by projectile electron wave functions, but does not imply a particular approximation for the cross section.

The transformation of the DDCS from the lab(unprimed) to the projectile(primed) frame is

$$\frac{d^3\sigma}{dE_k d\Omega_k} = \frac{k}{k'} \frac{d^3\sigma'}{dE_{k'} d\Omega_{k'}} \quad (1)$$

where the electron wave vector k' in the projectile frame relates to the wave vector k in the target or lab frame according to

$$k' = k - v \quad (2)$$

If for some reason the projectile frame DDCS is non-zero when $k' = 0$ it immediately follows from Eq.(1) that the lab frame cross section is infinite when $k = v$. Now an essential feature of charged particles is that they bind electrons in states of any principal quantum number n' and such states are always populated to some extent by electron capture reactions. As n' becomes

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FIG. 3. Energy distribution of electrons ejected from He at 0° by proton impact showing the continuum capture cusp. From (8).

infinite this capture process extrapolates smoothly across the threshold, which separates states of high n' from continuum states of vanishingly small E_k . It follows from continuity that the cross section for ejecting electrons moving with zero velocity in the *projectile* frame is non-zero. Eq.(1) then predicts a $1/|k - v|$ singularity in the lab frame cross section, as verified experimentally in Ref. (8). A similar cusp is predicted for projectiles which carry electrons that are stripped off in collisions. The cusp was actually observed by Lucas and co-workers (9) for electrons ejected when protons passed through thin carbon foils. The cusp observed in this case could have been caused by a two step process where the proton captures an electron which is stripped off in a subsequent collision. In retrospect the two step process is probably unlikely, but there is no way to distinguish the two possibilities for foil targets. In any event, this model-independent prediction of the cusp seemed to satisfy Gene that the process was well understood, at least when considered as an isolated feature (10). But this cusp posed great conceptual problems for theory, so in the rest of this talk I will describe the status of efforts to build a comprehensive theory of ionization which naturally incorporates the continuum cusp.

III. FURTHER DEVELOPMENTS

A. The Bethe ridge and the elastic scattering model

As background to a comprehensive theory it is useful to look at the Galilean invariant cross section, defined as

$$\frac{d^3\sigma}{dk^3} = \frac{1}{k} \frac{d^3\sigma}{dE_k d\Omega_k}, \quad (3)$$

plotted vs the projection $k_{\parallel}$ of $\mathbf{k}$ on the $\mathbf{v}$ direction and the magnitude of $\mathbf{k}$ perpendicular to this direction, namely $k_{\perp} = [k^2 - k_{\parallel}^2]^{1/2}$, as given by the First Born Approximation (Fig.4).

Notice that the only prominent feature is the semicircular ridge centered at $\mathbf{k} = \mathbf{v}$ with radius v . This is just the binary encounter or Bethe ridge characterized by the absence of target ion recoil in the final state, i.e. all of the momentum lost by the projectile is transferred to the electron. To a first approximation there are no other features.

The Bethe ridge feature is aptly described by a simple model called the elastic scattering model (ESM). In this model the target is regarded as a source of electrons moving with a velocity $-\mathbf{v}$ in the projectile frame which scatter quasi-elastically from the projectile into some other direction with $k' \approx v$. This picture qualitatively describes the surface in Fig.4 and is quantitatively more accurate than the Born approximation when $k_{\parallel} > v/2$. The elastic scattering model misses the cusp which tends to fill in the valley in the vicinity of $k' = 0$ or $\mathbf{k} = \mathbf{v}$.

B. A free-free transition model and the continuum capture cusp

To reconcile the intuitively attractive ESM with the apparently contradictory cusp, consider an alternative interpretation of the quasi-elastic scattering. Fig.5 illustrates schematically an electron of wave vector $\mathbf{s} = m\mathbf{v}/\hbar$ initially bound in a target eigenstate. The electron's momentum distribution about the value $-\mathbf{v}$ is represented by $\mathbf{s}$, which is to be integrated over. The electron wave function in the field of the projectile is the continuum function $\psi_{\mathbf{s}-\mathbf{v}}^+$. In the final state the electron wave function is $\psi_{\mathbf{k}'}^-$. The ESM considers that this transition is a quasielastic scattering. If we use that picture no cusp structure is predicted.

Fig.(5) suggests a more general interpretation in terms of free-free transitions between projectile eigenstates

$$E_{\mathbf{s}-\mathbf{v}} \rightarrow E_{\mathbf{k}'} \quad (4)$$

under the action of some transition operator. This operator emerges from Briggs' (11) expansion of capture amplitudes in powers of the projectile charge

FIG. 4. Plot of components of velocity for 1.5

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FIG. 4. Plot of the First Born Approximation Galilean invariant cross section vs components of electron momentum parallel and perpendicular to the incident proton velocity for 1.5 MeV proton impact on atomic hydrogen.

Z_T . Using this expansion one finds that the appropriate transition operator is

$$\tilde{V}_T(\mathbf{s} + \mathbf{J}) \exp[i(\mathbf{s} + \mathbf{J}) \cdot \mathbf{r}] \quad (5)$$

where $\mathbf{J}$ is the recoil momentum of the ionized target in the final state and $\tilde{V}_T(\mathbf{p})$ is the Fourier transform of the electron-target nucleus interaction. The corresponding cross section in this approximation, known as the Distorted-wave Strong Potential Born (DSPB) approximation, is

$$\frac{d^3\sigma}{dE_{\mathbf{k}} d\Omega_{\mathbf{k}}} = (2\pi)^4 \mu K_i \int d\Omega_{\mathbf{K}} \left| \int d^3s \langle \psi_{\mathbf{k}'}^- | \exp[-i(\mathbf{J} + \mathbf{s}) \cdot \mathbf{r}] | \psi_{\mathbf{s}-\mathbf{v}}^+ \rangle \right. \\ \left. \times \tilde{V}_T(\mathbf{J} + \mathbf{s}) \tilde{\varphi}_i(\mathbf{s}) \gamma_{\mathbf{k}}(\mathbf{s}) \right|^2, \quad (6)$$

where $\gamma_{\mathbf{k}}(\mathbf{s})$ is a quantity of nearly unit magnitude for $s \approx 0$ but is rapidly varying for large values of s . To a first approximation this quantity is omitted in calculations of the cross section.

Fig.(6) shows the Galilean invariant cross section calculated using Eq.(6). Note that the continuum capture cusp appears as well as the more pronounced Bethe ridge. The accuracy of Eq.(6) is better illustrated in Fig.(7), which compares the computed and measured (12) DDCS at zero degrees for the ionization of He by 1.5 MeV proton impact. The calculations use Eq.(6) with

FIG. 5. Schematic illustration of an initially bound electron (a) scattering quasi-elastically from a projectile and (b) making a free-free transition to a final continuum state of the projectile.

an approximate He initial state. Since unobserved quantum numbers are summed over approximately using unitarity, the calculations refer to inclusive cross sections. The agreement is quite good over the electron energy range from 100 eV up to energies well above the position of the binary encounter peak at . Contrary to the ESM, both the cusp and the binary encounter peak are accurately reproduced.

That the cross section of Eq.(6) is infinite at the cusp follows from the use of the projectile eigenstates $\psi_{\mathbf{k}}^-$ which incorporate a normalization factor proportional to $1/\sqrt{k'}$ for small k' . It is known that this normalization factor must not be present near the binary encounter peak, otherwise the cross section is much too large. This is the conceptual difficulty alluded to earlier—it seems that the cusp, which requires projectile eigenstates, is incompatible with the Bethe ridge, which appears to require target eigenstates. This apparent contradiction is resolved by the free-free transition picture.

Macek and Taulbjerg (13) have shown that Eq.(6) actually gives the elastic scattering model in the region where the momentum of the recoil ion $\mathbf{J}$ tends to zero. Specifically, they show that when $\mathbf{J} \approx 0$ one has $k' \approx v$ and

$$\frac{d^3\sigma}{dE_k d\Omega_{\mathbf{k}}} = \frac{(2\pi)^4}{v^2} 2k \int_0^\infty \int_0^{2\pi} |T^{elas}(\mathbf{k}', -\mathbf{J} - \mathbf{v})|^2 |\tilde{\varphi}_i(-\mathbf{J})|^2 d\phi_J J_\perp dJ_\perp \quad (7)$$

where $\mathbf{J}$ is resolved into components parallel $J_\parallel$ and perpendicular $J_\perp$ to the incident velocity vector $\mathbf{v}$;

$$\mathbf{J} = J_\parallel + J_\perp \quad (8)$$

and T^{elas} is the T-matrix for elastic scattering of an electron by the projectile. The magnitude of the parallel component is given by

$$J_\parallel = \frac{1}{v} \left(\frac{1}{2} k'^2 - \frac{1}{2} v^2 + |\epsilon_i| \right) \quad (9)$$

while all values of $J_\perp$ are integrated over. The free-free transition picture yields the elastic scattering model when $J \approx 0$; thus accurate descriptions of

FIG. 6. Plot of momentum p of proton impact

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FIG. 6. Plot of the DSPB Galilean invariant cross section vs components of electron momentum parallel and perpendicular to the incident proton velocity for 1.5 MeV proton impact on atomic hydrogen.

both the continuum capture cusp and the binary encounter peak emerge from Eq.(6). It must be emphasized that the standard Born approximation is still needed to describe electrons that have low velocity in the target frame. Even so, the theory of ionization by fast charged particles is in a fairly satisfactory state since we have available two good approximations which give reasonably accurate cross sections in their domains of applicability. The computations needed to evaluate Eq.(6) are rather complicated, but Madsen and Taulbjerg (14) have been able to obtain a simpler expression valid in the vicinity of the binary encounter peak. They show that Eq.(7) actually incorporates a multiplicative factor $\zeta_{k'}(-\mathbf{J} - \mathbf{v})$ which equals unity near the binary encounter peak where $k' \approx v$ but equals the singular normalization factor $2\pi Z_P/k'$ near $k' = 0$ which is responsible for the cusp. In essence they show that the continuum capture mechanism even modifies the binary encounter peak slightly through the factor $\zeta_{k'}$. This demonstrates rather directly that ionization by charged particle impact cannot be fully understood without incorporating Gene's insight that electrons are carried along by the positively charged projectile in the ionization process.

FIG. 7. The doubly differential cross section at 0° for the ionization of He by 1.5 keV proton impact. The solid curve is the DSPB theory and the dots are the measurements of Ref. (12).

IV. OTHER DEVELOPMENTS

While the story I have just related shows how Gene's work on the continuum capture mechanism was essential for the development of a comprehensive theory of ionization by charged particle impact, it by no means does justice to the wide impact this idea has had. Let me conclude by mentioning a few investigations that looked at different aspects of "continuum electron" capture. The unexpected connection between capture to high Rydberg states and ionization was investigated by Rødbro (15) for proton-Helium collisions. His work substantiated the basic premise of continuum capture. On the theoretical side Dettman (16) made the first attempt to include the Thomas double scattering mechanism in the theory of the cusp. This mechanism was much discussed in connection with capture to bound states, although firm experimental evidence for it was lacking. Shakeshaft and Spruch (17) recognized that the cusp could be used to test the double scattering theory and might provide the first experimental evidence for second Born terms in the theory of electron capture generally. They pointed out that the double scattering terms gave a cusp which had a step function discontinuity at $k = v$ for electrons ejected in the forward direction. Brenig et al. (18) found this discontinuity experimentally, thereby obtaining the first evidence for multiple scattering effects in electron capture. The cusp anisotropy then became a subject of

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much interest. Joachim Burgdorfer (19) developed expressions that connect the cusp anisotropy with the density matrix of states of high principal quantum number produced by electron capture. Meckbach (20), among others, made extensive measurements of the cusp angular distribution to probe various theories (21) of ionization. Harrison and Lucas (9) and others noted that two electrons could be captured into autoionizing states. These states often decayed by emitting very slow electrons in the projectile frame giving rise to subsidiary peaks on either side of the cusp. To investigate these, Stolterfoht (22) devised a tandem parallel plate analyser and was able to do high resolution spectroscopy of these very weakly bound autoionizing states.

Measurements specifically designed to exploit the Doppler shift of electrons in the projectile frame are now quite common, and it is difficult for a mere theorist to do justice to the ingenuity of experimentalists (23) working in this field. The one sentence summaries of measurements and theory given above represent some of the developments that followed the seminal discovery of the CEC cusp. Undoubtedly I have omitted many important studies in my brief account, since there are now so many observations. That's one aspect of the cusp—it's always there. If you make measurements of electrons ejected in the forward direction you cannot miss it. Indeed, the cusp feature has entered into the standard nomenclature of ionization by charged particle impact. For that reason this aspect of Gene's work will be recalled whenever the ionization process is invoked. That is perhaps a fitting characterization all of Gene's work—the innovative interpretation of his carefully measured data stands the test of time.

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