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LETTER TO THE EDITOR

The influence of experimental resolution and background upon ion-induced $v_e \simeq v_i$ electron distributions measured behind solid foils

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Abstract. Some confusion has developed with regard to the correct procedure necessary to compare charge exchange to the continuum (CEC) theories and experiment for ion-beam foil collisions where the emitted electrons have velocities $v_e \simeq v_i$. In order to make such a comparison, we emphasise that (*a*) the instrumental resolutions in angle *and* absolute value of velocity must be folded into the theoretical double differential cross section, and (*b*) that if the emission of CEC electrons occurs only because of interaction with the outermost atomic layers, those electrons originating from further inside the foil must be subtracted as background. Unfortunately, these considerations were not properly accounted for by Steckelmacher and colleagues as they compared their recent results with theory and published experiments.

In a recent paper by Steckelmacher *et al* (1978), later referred to as SSKL, entitled 'On the validity of beam–foil experiments designed to test theories of charge exchange to the continuum states of energetic ions', two points are discussed: (i) the instrumental resolution, and (ii) the treatment of the background. With regard to both of these, their paper is misleading. In the present note we wish to contribute to a better understanding of these two aspects because of their impact upon evaluation of experimental results.

Our discussion is based on the theory of charge exchange to the continuum (CEC)§ (Macek 1970, Salin 1969a, b, 1972, Band 1974, Dettmann *et al* 1974) which treats the interaction between the ion and a target atom under single collision conditions (low-density gas target). However, according to Dettmann *et al* (1974), the mechanism of CEC is also considered to be applicable for the case of ion-beam–foil interactions, if one assumes that the single-collision process of electron capture into the continuum of the emerging ion occurs effectively only in the last atomic layers of the foil (which might be *a contaminant*).

With the restriction that the ion velocity $v_i \gg Z'v_B$ and that $|v_e - v_i| < Z'v_B$, where $Z'v_B$ is the outer-shell electron velocity of the target atom, the CEC theory of Dettmann *et al* leads to a cross section

$$d\sigma/dv_e \propto |v_e - v_i|^{-1}. \tag{1}$$

§ This has also been referred to as charge transfer to the continuum (CTC).

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As explained in detail by Meckbach *et al* (1977) and in a review on the subject given by Meckbach and Baragiola (1977), when measuring the electron distribution as a function of v_e and θ, the angle with respect to the direction of the ion beam, the diverging cross section at $v_e = v_i$ (equation (1)) leads to a drastic dependence of the measured electron spectra on the instrumental acceptances on electron velocity $\delta v_e = \pm R v_e$ and solid angle $\delta\Omega$. In a velocity space generated by $\bar{v}_e$, δv_e and $\delta\Omega$ determine a resolution volume

$$v_e^2 \delta\Omega(2\delta v_e) = 2Rv_e^3\delta\Omega \tag{2}$$

where R is the relative half-width at half-maximum (HWHM) velocity resolution. The apparatus integrates over the cross section contained in this volume: consequently, when comparing theory with experiment, the cross section (equation (1)) has to be folded into the resolution volume (equation (2)).

Here we restrict our discussion to the case of longitudinal electron velocity distributions taken by scanning v_e at $\theta = 0$ in the direction of v_i.

Resolution

In order to facilitate comparison with experimental results, Dettmann *et al* ignore the resolution $|\delta v_e| = Rv_e$ in the absolute velocity and integrate equation (1) only over the surface $v_e^2\delta\Omega = \pi(v_e\theta_0)^2$ determined by an analyser acceptance cone of half-angle θ_0. For small θ_0 this integration is readily performed and leads to

$$\mathrm{d}\sigma(\theta_0)/\mathrm{d}v_e \propto (v_e/v_i)\{[(v_e - v_i)^2 + v_e v_i \theta_0^2]^{1/2} - |v_e - v_i|\}, \tag{3a}$$

which describes a cusp-shaped peak centred at v_i. Dettmann *et al* approximate equation (3*a*) by

$$\mathrm{d}\sigma(\theta_0)/\mathrm{d}v_e \propto [(v_e - v_i)^2 + v_i^2\theta_0^2]^{1/2} - |v_e - v_i|. \tag{3b}$$

Note that equation (3*b*) cannot account for the asymmetry introduced into the peak by the increase of $v_e^2\, \mathrm{d}\Omega$ with increasing v_e. The HWHM of the cusp Δv_e, described by equation (3*b*) is:

$$\Delta v_e = C_{\mathrm{app}} v_i \tag{4}$$

where $C_{\mathrm{app}} = \frac{3}{4}\theta_0$.

Although equations (3*a*, *b*) are attractive because of their simplicity, we must recognise that they are the result of an approximation where the instrumental velocity resolution has been neglected. When the resolution volume (equation (2)) is a thin disc such that

$$|\delta v_e| = v_e R \ll v_e\theta_0 \qquad \text{or} \qquad R/\theta_0 \ll 1 \tag{5}$$

then cusp-shaped peaks, as resulting from equations (3*a*, *b*), approximately describe the experimentally measured peaks.

We can represent equation (3*a*) in terms of a reduced velocity given by $x = v_e/v_i$, such that, with v_i^2 dropped,

$$\mathrm{d}\sigma(\theta_0)/\mathrm{d}x \propto x\{[(x-1)^2 + x\theta_0^2]^{1/2} - |x - 1|\}. \tag{6}$$

It is obvious that the *shapes of peaks*, obtained with ions of different velocities, *are identical* when represented in terms of x. Therefore, when represented in terms of v_e, the peak widths are proportional to v_i.

For a correct comparison with experiment, we integrate the CEC cross section equation (1) over the resolution volume equation (2). As equations (3*a*, *b*) are already the result of integration over $\delta\Omega$, only the integration over the acceptance in δv_e remains. This is equivalent to integrating equation (6) with respect to dx. For a square transmission window $[x(1-R), x(1+R)]$†, we obtain

$$\frac{\mathrm{d}\sigma(\theta_0, R)}{\mathrm{d}x} \propto \frac{1}{2xR} \int_{x(1-R)}^{x(1+R)} \mathrm{d}x'\, x'\{[(x'-1)^2 + x'\theta_0^2]^{1/2} - |x'-1|\} \tag{7}$$

where the factor $1/2xR$ normalises the cross section through the window. This integration, which is elementary, removes the singularity in the slope of the integrand at $x = 1$, and leads to a *rounded peak*.

The above analytical expression is *again* a function of $x = v_e/v_i$: hence the proportionality of the peak width Δv_e to v_i is conserved, and we can write

$$\Delta v_e = C v_i \tag{8}$$

where C is the relative peak width. This proportionality of Δv_e to v_i has been used widely as a test for the validity of CEC theory (Dettmann *et al* 1974, Meckbach *et al* 1977, Meckbach *et al* 1978, Chiu *et al* 1978, Laubert *et al* 1976, and SSKL).

It is obvious that the cusp described by equation (3*b*) cannot be considered as the ultimate result of CEC theories. However, SSKL have incorrectly stated that this cusp is the 'only acceptable looking peak shape'; that the 'CEC predicted peak widths' are only those which are derived from equation (3*b*), for example equation (4); and consequently that there are 'severe experimental constraints imposed by the CEC model' with respect to 'the resolution required of any analyser with which we may wish to examine such a peak'.

There is no reason to say that an analyser which does not cope with the condition set by equation (5) is 'running out of resolution'. Therefore, there is no reason to state that the heights and widths of peaks, which show a 'rounding at the top' are in error. The appearance of such rounded peaks is what one actually has to expect!

Figure 1 shows the derived peaks resulting from the integration of equation (7) as well as of the approximate equation (6) for the equipment of Meckbach *et al* (1977) and Chiu *et al* (1978) ($\theta_0 = 1°$; $R = 0{\cdot}0025$). Note the *slight asymmetry* of the peak.

Table 1 shows: the resolutions R and θ_0 of the equipment used in different laboratories as well as their ratios R/θ_0; the relative peak half-widths C, C_{app}; the percentage deviation $b' = (C - C_{app})/C \times 100\%$; and the corresponding peak broadening b of SSKL defined as $\mathrm{d}(\Delta v)/\Delta v \times 100\%$.

It is seen that the condition of equation (5) is closely met by most of the equipment used for investigating $v_e \simeq v_i$ electrons, except for that of Duncan and Menendez (1976) whose angular resolution is the best. Their resolution results in the narrowest $v_e \simeq v_i$ peaks obtained to date. Their peaks are rounded at the top. SSKL constructed an analyser with $R = 0{\cdot}025 \times 10^{-2}$ and $\theta_0 = 0{\cdot}024$ rad so that $R/\theta_0 \simeq 0{\cdot}011$. Consequently

† We have also carried out this integration numerically with the Gaussian velocity distribution folded in. The results are similar.

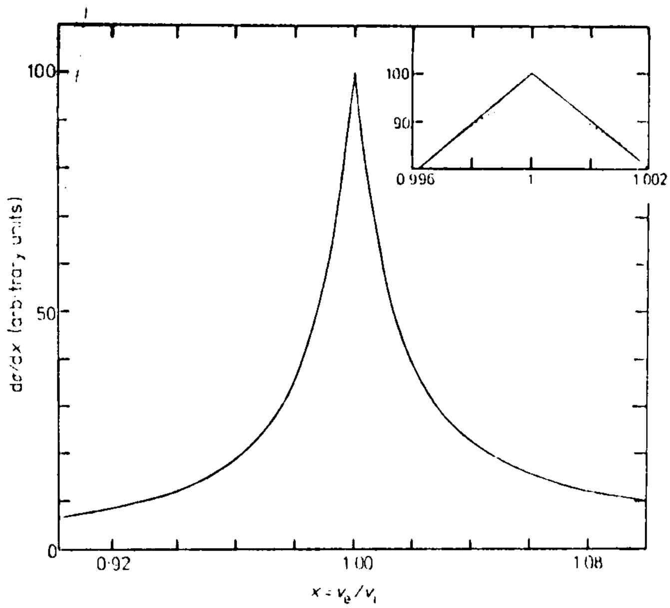

Figure 1. Longitudinal differential cross section plotted as a function of reduced velocity $x = v_e/v_i$. The full curve represents equation (6) which has the angular acceptance $\delta\Omega = \pi\theta_0^2$ folded in; the broken curve represents equation (7) which has both angular acceptance $\delta\Omega$ and velocity acceptance $\delta x = Rx$ folded in. The inset shows the expansion of the cusp. The resolution acceptances used are $\theta_0 = 0{\cdot}018$ rad and $R = 0{\cdot}0025$ (Meckbach *et al* 1977).

Table 1. Transverse θ_0 and longitudinal R instrumental resolutions and their ratio; longitudinal relative HWHM, $C = \Delta v_e/v_0$ obtained by folding these resolutions into the CEC cross section, equation (7); and approximate relative HWHM, $C_{app} = \frac{3}{4}\theta_0$, obtained from equation (4). The percentage deviation b' (%) is compared with b (%) estimated by Steckelmacher *et al.*

Reference	θ_0 degrees	θ_0 radians	R	R/θ_0	$C = (\Delta v_e/v_i)$	$C_{app} = \frac{3}{4}\theta_0$	b' (%)	b (%)
Dettmann *et al* (1974)	4·6	$8{\cdot}0\times10^{-2}$	$0{\cdot}5\times10^{-2}$	0·06	0·063	0·060	5·0	10
Duncan and Menendez (1976)	0·36	0·63	0·5	0·8	0·0086	0·0048	44·0	133
Lauberfet *et al* (1978)	1·3	2·3	0·35	0·15	0·0195	0·0170	12·8	—
Meckbach *et al* (1977)	1·0	1·8	0·25	0·14	0·0148	0·0130	12·2	25
Steckelmacher *et al* (1978)	1·35	2·4	0·025	0·011	0·0180	0·0177	1·67	1·8

SSKL-measured peaks come closest to the cusp-shaped peaks given by the approximation contained in equations (3*a*, *b*).

Our percentage deviation at HWHM, b', disagrees with the values of b given by SSKL. Their results are derived from equation (3*b*) by using a crude and questionable procedure in which 'an error in the total height' is determined by the level at which the instrumental acceptance in velocity δv_e is equal to the width of the cusp. In essence, their derivation follows from an improper use of differential error analysis.

Background

If, as was done by SSKL, the background is included when measuring the peak width at a certain fraction of its maximum height, is it possible to obtain a *unique* determination of the characteristic peak width dependence as a function of the ion velocity? No! Of course not.

If the relative contribution of the background to the total peak height were a function of v_i, the resulting dependence of the peak width would change according to the particular fraction of the peak height chosen. As a matter of fact, the authors state that the shape of the lower-velocity wing would lead to an infinite width 'if the half-width were computed at half the peak height with the background included at these high projectile velocities'. This implies that the background is more than half the peak height, a situation which is *not* at all compatible with equations (3*b*) and (4) on which their discussion is based.

A second question—are we allowed to include the background when measuring peak widths? No! Particularly in the case of ion-beam–foil collisions where we are dealing with ion-induced secondary electron emission from a solid foil. This process has been described by Meckbach (1976)—energetic electrons are produced through collisional-energy transfer inside the solid. On travelling to the surface these electrons lose energy and are scattered from their initial direction. It is therefore obvious that CEC electrons cannot originate from inside the foil. For this reason, Dettmann *et al* introduced their 'last-layer model': CEC electrons, if emitted from a foil, must have been captured into the continuum of the ion in its last atomic layers. Therefore, if we wish to study this mechanism, we must subtract the contribution from the independent secondary-electron emission process which has to be considered as a background. It would appear from SSKL (p 2716, first paragraph) that when the secondary-electron emission background is subtracted, they also demonstrate the independence of the peak widths observed by Meckbach *et al* (1977) which were further discussed by Brandt and Ritchie (1977), Meckbach *et al* (1978) and recently confirmed for the case of heavy-ion-beam–foil interactions by Laubert *et al* (1978).

The entire discussion of SSKL has been based upon the validity of the theory of Dettmann *et al* which depends upon single-particle collisions. With respect to this point, further confusion developed when they wrote: 'In the solid, single-collision conditions are absent so that the measured quantity 'yield' is influenced by any energy dependence of the effective target thickness as well as that of the cross section'. This statement negates the very applicability of CEC theories to the ion-beam–foil electron emission process, which they actually wanted to defend in their paper.

Finally, we reconfirm that the conclusion of Meckbach *et al* (1977) that charge transfer to the continuum theories do not correctly describe the emission of $v_e = v_i$ electrons in ion-beam–foil collisions, is not invalidated by the report of SSKL. The mechanism for the production of these electrons remains an open question.

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Reply to *Chiu et al*

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We agree that there has been some confusion in the literature regarding the correct procedure when comparing charge exchange to the continuum (CEC) theories and experiment for ion-beam-foil collisions, and that this concerns both the correct treatment of instrumental resolution and ionisation background. In Steckelmacher *et al* (1978, to be referred to as SSKL) we had demonstrated that by careful experimental design, we had overcome possible errors due to insufficiently low values in the ratio of velocity resolution to angular acceptance. We are grateful for the correction to our differential error analysis, which does not affect our own results and somewhat reduces the possible errors in the literature. However, as we discuss below, we find that our main point of disagreement concerns the treatment of direct ionisation background.

(*a*) *Resolution*

Let us first be clear that the differential cross section $d\sigma/dv_e$ proposed by Dettmann *et al* (1974, to be referred to as DHL) and given as equation (1) in the above letter only diverges if both R and θ_0 are equal to zero§. As soon as one incorporates a finite θ_0 for the measuring spectrometer $d\sigma/dv_e$ becomes finite everywhere, though if R were, in principle, again zero the slope of $d\sigma/dv_e$ would still exhibit a discontinuity at $v_e = v_i$ as in figure 9 of DHL. The incorporation of a finite R removes this last discontinuity so of

§ This is a somewhat academic point since the measured signal must then drop to zero.

course we agree that one must in practice expect to see 'some rounding at the top'. The questions to be asked are: 'how much rounding?' and 'under what conditions is it severe enough to cause a given error in peak height and hence peak width at any chosen height?'. For example, in figure 1 of Chiu *et al* (1979) the yield curve has been correctly normalised to the cusp peak (corresponding to zero R) and not to the approximately 10% lower rounded peak as in Meckbach *et al* (1977), to which this refers. This introduces an error in HWHM.

One purpose of our article (SSKL) was to show that because the function describing the peak is vectorial the important quantity to be considered when designing a spectrometer is not R alone but R/θ_0. That constraints are severe is seen from table 1 of the article by Chiu *et al* (1979). If R/θ_0 is reduced to 0·14 or 0·15 the systematic broadening of the peak as defined by their b' is more than 12%. Now if HWHM are to be plotted against v_i equation (4.18) of DHL will predict a slope of $\frac{3}{4}\theta_0$ but only if $R/\theta_0 \ll 1$. The measured slope C (in Chiu *et al*'s notation) should always be greater than this by an amount depending on R/θ_0. The amount by which C differs from $\frac{3}{4}\theta_0$ is the amount by which we meant the analyser to have 'run out of resolution'. Notice we did not say the cusp is the 'only acceptable peak shape' but that even when the peak has been substantially smeared it will still *look* acceptable to the casual observer.

Two alternative methods are available to deal with the resolution problem expressed by the finite nature of R/θ_0. One can feed the finite R into the theory as is done in equation (7) of Chiu *et al* (1979) or one can make a spectrometer with R/θ_0 so much smaller than 0·14 that the errors so introduced approach the 1% level where we feel they can reasonably be ignored. Since we reduced the resolution problems to around this level by careful attention to experimental design our data presented in SSKL are not subject to any inconsistency or correction from this point of view. They stand as they are.

We agree that it is instructive to look at the effect of finite velocity resolution with a rectangular window as is done by Chiu *et al.* Taking their equation (3a), which is obtained directly from DHL, we can compare the cross section at the peak σ^{peak} expected for $R \to 0$ with that for finite R. Since we are at the top of the peak we have $v_e = v_i$ and so drop the factor in front of the curly brackets of (3a) and perform the integration exactly as in equation (7). For the case of $R/\theta_0 \ll 1$ a simple expansion in powers of R/θ_0 gives the successive corrections which must be applied†.

$$\sigma^{\text{peak}}_{R/\theta_0 \ll 1} = \sigma^{\text{peak}}_{R\to 0}(1 - \tfrac{1}{2}R/\theta_0 + \ldots \text{order } R^2/\theta_0^2).$$

The mistake in our 'improper differential error analysis' was to forget the $\frac{1}{2}$ and for that we unreservedly apologise. This half should enter equations (5) and (8) of SSKL and will reduce the 'Δr' and 'b' values of our table 1 so that the latter are not too different from those of table 1 of Chiu *et al.* One must still emphasise however that the correction for finite instrumental resolution has a greater than 12% effect on half-width for much of the data reported if those data are to be compared with equation (4·18) of DHL. If the data are to be compared with wake riding or other theories in the manner of Brandt and Ritchie (1977) or Meckback *et al* (1978) an expression appropriate to those theories must be used in an equation analogous to (7) of Chiu *et al* to make a similar correction.

We note however that without the resolution correction, data such as those of Meckbach *et al* (1977) would give slopes C (Chiu *et al*'s notation) greater than $\frac{3}{4}\theta_0$

† In the case of Duncan and Menendez (1976) where R and θ are comparable a different expansion is required. It gives $b' = 44\%$ as quoted by Chiu *et al.*

rather than the HWHM almost independent of v_1 reported by them. We must therefore look for another source of disagreement and we believe this lies in their treatment of the direct ionisation background.

(*b*) *Background*

It is difficult to better the discussion provided by Macek (1970) for gas targets and we recommend his paragraphs 2, 3 and 4 on page 236 as the starting point.

We have to find the scattering amplitude for an electron in the field of two Coulomb centres, one stationary, one moving, for all velocities of the electron relative to each centre. Once we have this amplitude the derivation of the cross section follows rigorously from the resultant T matrix. The central difficulty is that we cannot write down a single wavefunction which will represent the electron for all possible situations. We have to construct separate amplitudes for the situations (*a*) when the interaction between the electron and target is strong and (*b*) when the interaction between the electron and projectile is strong.

Having done this we have counted twice—there is only one electron—so we must introduce a counter term so arranged as to cancel (*a*) when (*b*) is dominant and vice versa. These three terms are respectively a_{23}, a_{12} and a of Macek's equation (16). Since the Born approximation may be more familiar we write them out below in the notation of DHL, figure 6, restricting outselves to first order since that is sufficient to make the point.

$$T=\langle e^{iK_f\cdot R}\varphi_k^-(r)|V_i|\phi_{i,K_i}\rangle+\langle e^{iK_f'\cdot R'}\varphi_{k'}(r')|V_i|\phi_{i,K_i}\rangle-\langle e^{i(k\cdot r+K_f\cdot R)}|V_i|\phi_{i,K_i}\rangle.$$

Now the first term can represent target ionisation: that is the electron 'm' moving slowly away from the target M, and M' moving rapidly away from M and m. Hence if $\varphi_k(r)$ is a Coulomb wave centred on M this term is the usual Born amplitude and we call it 'direct ionisation'. The second term is CEC with $\varphi_{k'}(r')$ the Coulomb wave about M', which represents M' and m moving slowly away from one another, but both moving rapidly away from M. We could call this 'rearranged ionisation' though strictly such a term implies a two-step process and the use of a second-order approximation. The third term is the counter term in the limit of all three particles separating quickly. Notice how it works. If CEC dominates (second term) the electron has large k with respect to the target. The Coulomb wave $\varphi_k(r)$ becomes $e^{ik\cdot r}$ and the first and third terms cancel. There is no other source of electrons to provide a 'background' which has to be called 'direct ionisation' and subtracted. As we move into the wings the cancellation from the counter term becomes progressively imperfect so that it is necessary to calculate all three terms to predict the whole velocity spectrum. This of course is what Macek has done. We see here the great similarity between the Dettmann and Macek approaches. Macek has only a first-order approximation but can compute the whole spectrum, Dettmann retains an analytic approach and to second order but can only describe the neighbourhood of the peak in the spectrum. Notice that if exact computations were applied to Dettmann's three terms, at the cost of a loss of analytic form, interference effects would arise just as they do in Macek's approach. To say that the 'situation where the background is more than half the peak height is not at all compatible with Dettmann's differential cross section' is to exhibit a serious misunderstanding of the way the T matrix is constructed and of the nature of the direct ionisation.

We now have to make the connection with the solid. To do this one must understand that secondary electron emission from a solid *is just the result of ionisation* of the atoms of that solid (Sternglass 1957). Further, as Macek says, 'there is no physical distinction between ionisation and charge exchange to a continuum state, indeed the latter is only one mechanism for ionisation'. Hence the $v_e \approx v_i$ peaks from a solid, and the direct ionisation too, are intimately connected with ionisation in a gas. To include the direct ionisation in the gas case and subtract it for the solid is totally inconsistent. The sole complicating factor is the ability of the electrons, once produced by ionisation, to leave the solid where scattering and absorption are very strong.

We believe, as does Cross (1977) for proper charge exchange, that the process of capture to the continuum state and subsequent loss, as well as direct ionisation, continues throughout the passage of the projectile through the solid. Most of the electrons so produced never reach the surface of the solid with sufficient energy to escape. The vast majority of those that do escape have energies of less than 10 eV (Krebs 1968). For an electron captured in a continuum state inside the solid to be subsequently detected outside the solid in a spectrometer set for the forward direction it must preserve both its speed and direction in spite of the strong scattering to be expected. We therefore turned (DHL) to the data and calculations which are known as the 'escape depths' of electrons from solids (Duke *et al* 1970, Powell 1974, Carlson 1974). These inelastic mean free paths λ_i are obtained from Auger electron spectroscopy and x-ray photoelectron spectroscopy of solids. Where data are available the latter technique is particularly apposite since the x-rays generate electrons at all depths in the solid just as we believe our projectiles do. However both techniques give similar results. For 262 eV electrons in carbon, i.e. those having the same speed as 481 keV protons, $\lambda_i = 7{\cdot}5$ Å (Jacobi and Hölzl 1971). Both theory and experiment show that in the energy range 100—1500 eV, λ_i is approximately proportional to $E^{1/2}$ (Carlson 1974, Carter *et al* 1974) so that for an electron of 123 eV (225 keV proton) we expect $\lambda_i \approx 5{\cdot}0$ Å. Hence our choice of one atomic layer as a rough estimate of the effective target density in the DHL paper.

Let us now apply these arguments to the relative ion energy dependence of the yield as measured by Meckbach *et al* (1977). We recall that the yield (electron/proton) is given by the product of the cross section and target density. They observe a yield proportional to $E^{-3\cdot1}$ from carbon foils in an energy range where the effective target density is increasing as $E^{1/2}$ because of the energy dependence of λ_i. This therefore implies an energy dependence for the cross section of $E^{-(3\cdot1+0\cdot5)}$ in exact agreement with the value $E^{-3\cdot5\pm0\cdot1}$ reported by the same group (Chiu *et al* 1978) using the same apparatus but a helium *gas* target under single collision conditions!

It is really so clear that there is some fundamental difference between gas and solid targets?

We should like to thank Robin Shakeshaft for discussing the importance of the direct ionisation with us.

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