Wake Formation by Megaelectronvolt-per-Nucleon Bare H and He Ions in Large Hydrocarbon Molecules?

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(Received 13 March 1985)

A new component is observed in $\sim 0°$ electron cusp spectra produced by megaelectronvolt-per-nucleon H^+ or He^{++} bombardment of hydrocarbon molecules with more than three C atoms. This component is narrower than, and different in skew and yield energy dependence from, electron capture or loss to the continuum processes, and has a width that is dependent on projectile Z. Of the *presently proposed* mechanisms for single forward electron production, only wake production in the larger molecules cannot be eliminated, although wake-model predictions agree just qualitatively with our experimental results.

PACS numbers: 34.70.+e

The concept of a polarization-potential "wake" trailing a charged particle as it passes through a dense medium originated with Bohr.[1] This potential wake is expected to contribute to the stopping power of the medium through which the projectile and its wake pass,[2] to affect the spatial correlation of ion clusters inside the medium,[3] to modulate the measured ejected electron yield from thin solid targets,[4] and to contribute to the cusp shape of the 0° convoy-electron spectrum.[5] The present experimental evidence supports the existence of the wake inside the medium, but is conflicting if a wake is inferred from 0° cusp spectra of electrons exiting from the medium.[5,6]

The essential requirement for a polarization wake is that there be an electron gas, as in targets with band structure, which can respond collectively to the transient Coulomb field of the projectile. This is clearly the case for a solid, but rules out atomic-gas targets. For molecular targets, however, the possibility of a polarization wake has not been examined experimentally or theoretically, even though for large hydrocarbon molecules there is theoretical[7] and experimental[8] evidence for quasicontinuum band structure. In this work we present the results of an investigation into molecular-size effects on the 0° cusp spectra of emerging electrons with velocity v_e approximately equal to the projectile velocity v_P by using C_mH_n (hydrocarbon) targets of varying size.

The electron spectra were collected with a spherical-sector electron analyzer placed either at or close to 0°.[9] The entrance half-angle of the analyzer θ_0 was 3.0° and the electrons were detected after analysis by a channel electron multiplier. The $\sim$ 1-MeV/u H^+ and $He^{+,++}$ beams were collimated and passed through a 0.38-mm-diam aperture just prior to a windowless gas target to eliminate glancing collisions from the apertures of the gas cell or electron analyzer. Typical beam currents through this aperture were a few nanoamperes; the relative precision for current integration was better than 0.1%. After setup, experimental parameters remained fixed, except for those associated with the interchange of gases. The gas pressures in the gas cell were monitored with a 0–1-mm capacitance manometer, whose output was used in a feedback loop to regulate the gas-cell pressure to within 2%. Corrections for out-of-target electron-transfer contributions to the cusp were made by bleeding gas into the target chamber to achieve the same beam-line vacuum and subtracting this contribution from the cusp spectrum.

The various contributors to 0° single-electron emission should be electron capture to the (projectile) continuum (ECC), projectile electron loss to the continuum (ELC), and wake-riding electrons. We have applied the following three general experimental criteria to aid in differentiating among the possible mechanisms for 0° electron emission: (i) differences in cusp spectra shape—including skewness and FWHM—as projectile Z (Z_P), charge state (Q_P), and energy (E_P) are varied; (ii) differences in cusp electron yields versus Z_P, Q_P, and E_P; (iii) cusp shape or yield variations dependent on C_mH_n molecular size only (hereafter labeled by m). We expect that criterion (iii) should be *the* definitive characteristic for the birth of a wake.

When bare H and He projectiles are used to bombard hydrocarbon molecules of increasing size, an abrupt change from the ECC cusp shape in the peak region is observed only when the hydrocarbon molecule has $m \geq 4$. This is shown in Fig. 1 for 0.6-MeV/u He^{++} on $m = 1$ and 7 hydrocarbon targets (similar results also pertain for H^+). The $m = 1$ cusp shape is very similar in shape to that obtained with an atomic-argon target[9] and hence is considered only ECC in origin. The $m = 7$ cusp, shown superimposed on the $m = 1$ cusp in Fig. 1, demonstrates that the $m \geq 4$ cusps are distinctly more pointed than those for $m = 1$–3.

Examination of cusp shapes measured slightly off 0° provides additional insight into these shape variations.

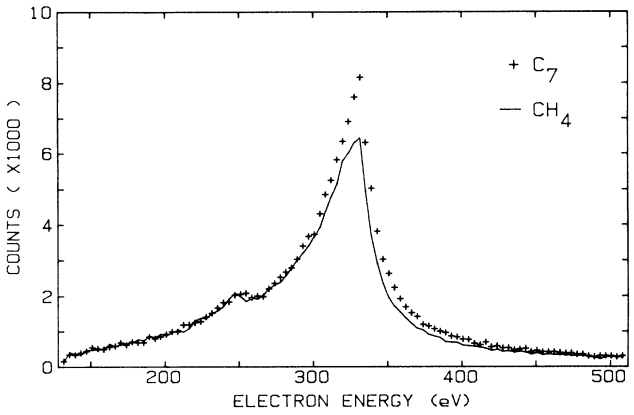

FIG. 1. Cusp spectra for 0.6-MeV/u He^{++} on $m=1$ and 7 hydrocarbon gases. The $m=1$ cusp is shown superimposed on the $m=7$ cusps to emphasize the abrupt increase in cusp pointedness for $m \geq 4$. The C K Auger electrons appear at ~ 250 eV.

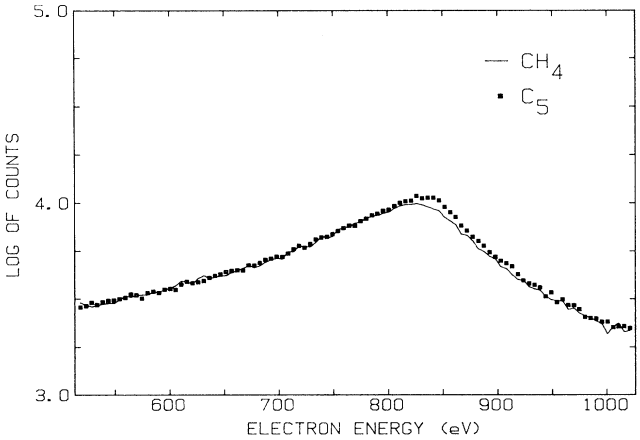

FIG. 2. Cusp spectra at $\sim 0.7°$ for 1.5-MeV H^+ on CH_4 (solid curve) overlaid on C_5H_{12} (squares). The W peak is observed slightly above the ECC cusp peak.

In Fig. 2 the $m=1$ and 5 hydrocarbon cusp shapes obtained with 1.5-MeV H^+ and the electron analyzer at an angle of $\theta \sim 0.7°$ are overlaid in a log(counts) versus electron-energy plot. Except in the region just above the peak in the ECC cusp (for $m=1$), the overall cusp shape observed for $m=5$ is just that measured for $m=1$. There is an additional bump in the $m=5$ cusp shape, slightly above the ECC peak. This result, combined with the $\theta=0°$ data in Fig. 1, suggests that this new structure arises from an independent mechanism adding to the ECC contribution *only* for $m \geq 4$. To examine this new structure (hereafter called the W peak) an obvious approach would be to subtract the ECC cusp shape obtained for $m=1$ from the "W + ECC" cusp shape obtained with $m \geq 4$, with use of fractional stripping. Fractional stripping was used routinely to check cusp-shape reproducibility over a run and also to examine differences between atomic and molecular cusp shapes. Alternative procedures employed previously to demonstrate changes in cusp shapes used ratios of electron yields (to compare cusps obtained with isotachic H^+ and H_2^+ [10]), or the essentially visual observation that the cusp appeared to have a narrow component superimposed on a broader, underlying distribution.[5] Direct subtraction, moreover, permits straightforward comparison of relative yields of W and ECC cusp contributions and determination of relative energy dependences.

Since the cusps obtained with the small hydrocarbon targets are used to define the shape of the ECC contribution, clearly this experiment is most sensitive to *changes* in cusp shape as the size of the molecule increases. The most reasonable method to eliminate the ECC contribution to the overall cusp shape is to scale the cusp shape of CH_4 so that in the difference spectrum, $\Delta_m = S_{C_mH_n} - c \times S_{CH_4}$ (c is the scaling factor), the low- and high-energy wings of the cusp, where the distribution is entirely ECC, are subtracted out.

In Fig. 3 Δ_m spectra for 0.6-MeV/u H^+ and He^{++} on the $m=7$ hydrocarbons are shown. The W peak shapes seen in Fig. 3 are clearly different from the CH_4 ECC cusp shape seen in Fig. 1. The W-peak FWHM in Fig. 3 is observed to depend on Z_P, and its shape is different from the ELC cusp shape obtained with isotachic He^+.[11] The ECC cusps obtained with isotachic H^+ and He^{++} on CH_4 are indistinguishable in shape,[11] i.e., no Z_P dependence is observed, consistent with previous work.[12] On the other hand the ECC cusp shape for CH_4 differs considerably from the ELC cusp shape obtained with isotachic He^+.[11] Other experiments have shown that the ELC cusp, which arises when the projectile carries electrons into the col-

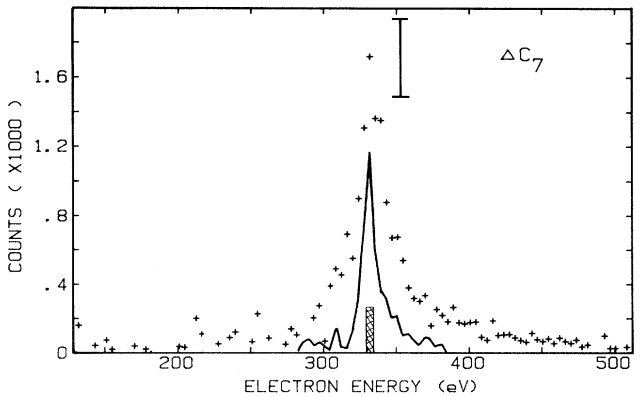

FIG. 3. Difference spectra for 0.6-MeV/u H^+ (solid curve) and He^{++} (pluses) on $m=7$ hydrocarbon gases. The statistical error shown is 2σ. (Experimental resolution of ~ 5 eV is shown crosshatched.)

lision, is also quite insensitive to Z_P and Q_P.[12] Since all these various peaks have different skews and FWHM's, by criterion (i) we conclude that the W peak is *not* due to ECC or ELC processes.

The steplike trend observed for the W-peak yields of the bare projectiles versus m signify a new process contributing to the cusp. Is this contribution evident in the overall cusp yield, too? To emphasize any such contribution the hydrocarbon cusp yields, $Y(C_mH_n)$, were used to compute the quantity $R = Y(C_mH_n)/mY(CH_4)$. The number of H atoms in the molecule has little effect on R for the $\sim$ 1-MeV/u projectiles used here because the ECC cross sections for the H atoms in the hydrocarbons are no more than a few percent of those for the C atoms.[13] If strict atomic cross-section additivity held for ECC processes, then $R \cong 1$ for any of the hydrocarbons.

Intramolecular "outscattering" attenuation processes[13,14] are expected to produce an exponential falloff in R as m increases, yielding a linear dependence in a log$R$ versus molecular-size plot. In Fig. 4, log$R$ values for 0.6- and 0.8-MeV/u H^+ on C_mH_n gases have been plotted versus m. Isotachic He^{++} results (not shown) gave very similar variations of R vs m. As in the case of electron capture to bound states (ECB) of these same projectiles,[14] the deviations from $R = 1$ for the bare projectiles are most noticeable at the lowest projectile energies. There is a noticeable break in the R vs m plot around $m = 3$–4, at the position where the onset of the W-peak structure occurs.

When R for incident He^+ ions is plotted in Fig. 4 also, we observe much larger deviations from $R = 1$. All R values for 0.15–0.8-MeV/u He^+ ions fell within error of one another, and hence only the averaged R values are shown in Fig. 4. The break in the R vs m plot around $m = 3$–4 is much more noticeable for the He^+ projectiles, as are the departures from $R = 1$.

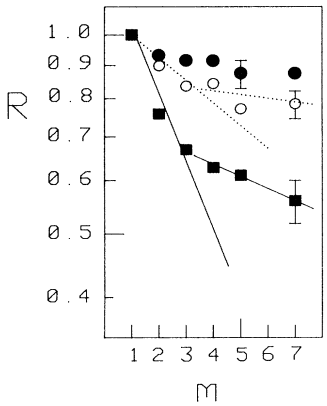

FIG. 4. Log$R$ vs m for H^+ (open circles, 0.6 MeV; solid circles, 0.8 MeV) and He^+ (solid squares, averaged over all energies) on $m = 1$–7 hydrocarbons. Lines are drawn to guide the eye only.

The sudden changes in cusp shape seen with bare projectiles when $m \geq 4$ *and* the break in the R vs m plots between $m = 3$ and 4 signal the advent of a new process, according to criterion (iii).

When the ECC- and W-peak yields were fitted with a power-law energy dependence, it was found that the H^+ and He^{++} ECC cusp yields fell off as $\sim E_P^{-2.7}$ and the W-peak yield (H^+) fell off as $\sim E_P^{-1.3}$. By criterion (ii) these considerably different projectile-energy dependencies again demonstrate that the W peak has an origin other than ECC processes. Similarly it is not due to ELC processes, such as could occur if there were ECB followed by ELC, i.e., a two-step sequence. The ELC cusp shapes, skews, and yield energy dependence are all different from the ECC- or W-peak results.[11]

There is one additional possibility to consider: instrumental electrostatic field ionization of Rydberg states (FIRS) of the projectile, first mentioned prominently by Vager *et al.*[15] The FIRS contribution was observed as part of the cusp (convoy) electron spectrum obtained with solid targets, and arose from electrons originally captured into bound states being pumped into Rydberg states as the projectile traversed the remainder of the solid target. This might appear to be a facile explanation of the W-peak appearance; nevertheless it does not explain the *abrupt* appearance of the W peak observed with bare projectiles, the significantly narrower shape of the W peak, or the lack of this W peak with He^+ projectiles. Nor would the FIRS mechanism explain the Z_P variation of the W-peak FWHM, since it is fundamentally an ELC mechanism and these processes show very little dependence on Z_P, Q_P, and E_P.[12] In conclusion, the elimination of ECC, ELC, and FIRS processes leaves only wake production, of the *presently proposed* single forward electron producing mechanisms, as the origin of the W peak.

Are the properties of the W peak consistent with wake formation? The abrupt onset is certainly suggestive of the initiation of wakes for $m \geq 4$ molecules. The valence-electron energy-level calculations by Hoffman[7] for alkanes (CH_4, C_2H_6, C_3H_8, C_5H_{12}, and C_7H_{16} are among some of the alkane gases employed in this experiment) show that the *spread* in valence-electron binding energies tends to grow with increasing m until $m = 4$, while for $m \geq 4$ the level density increases, indicating the onset of solidlike band structure.

Wake-electron cusp shapes can be readily compared to the W-peak shape. Brandt and Ritchie[16] derived an expression for $\Delta E/E$ (where ΔE is the FWHM of the cusp) which is given here as

$$\frac{\Delta E}{E} = \frac{4\omega_p^{3/4}}{v_P^{7/4}}(\ln 2)^{1/2}\left[Z_P^{\mathrm{eff}} C\eta \ln\left[\frac{8Z_P^{\mathrm{eff}} C\eta v_P}{\Gamma\omega_p}\right]\right]^{1/4}. \quad (1)$$

The plasma frequency is ω_p, C is the wake damping constant, $\Gamma = 1.781$, η is the wake-wake bound-electron correlation constant, and Z_P^{eff} is the effective charge number. Equation (1) predicts that a He^{++}/H^+ wake-electron peak FWHM ratio should be 1.32 for 0.6-MeV/u projectiles, whereas the experimental ratio for the W peak is 1.77 ± 0.32. The predicted wake FWHM values (solid C) of 34 eV for 0.6-MeV/u H^+ (W-peak experimental value, 9.5 ± 1.5 eV) and 46 eV for 0.6-MeV/u He^{++} (W-peak experimental value, 17 ± 1.5 eV) are considerably larger than those observed for the W peak. This is consistent with the results of Ref. 5, using thin Al targets, where the width of the narrow component observed in the convoy-electron cusps, and identified as being wake associated, was considerably below the wake-model prediction. By way of contrast, for ECC processes, $\Delta E/E = 3\theta_0$ (angle in radians), independent of E_P and Z_P.[17] We measured a FWHM of 41 ± 2 eV for our H^+ and He^{++} cusps at 0.6 MeV compared with the ECC prediction of 50 eV, whereas the He^+ ELC cusp at 0.6 MeV/u had a FWHM of 35 ± 2 eV.

It has been observed that the peak in the ECC- and ELC-cusp electron-energy distributions drops off in energy as the electron analyzer moves off 0°.[18] The wake model of Brandt and Ritchie, however, predicts that the wake-electron velocity distribution maintains its peak at v_e at or slightly above v_P. In Fig. 2, just this behavior can be seen between the underlying ECC cusp and the W peak.

With Eq. (1) it is possible to estimate the plasma frequency of a hydrocarbon molecule from the FWHM of the H^+ W peak. The plasma frequency obtained in this way is an order of magnitude smaller than the value obtained for solid C.[3] Surprisingly, a similar situation pertains for a plasma frequency for solid Al derived from the Al thin-target measurements of Gladieux and Chateau-Thierry.[5] If the application of Eq. (1) in this manner is a reliable way to estimate the plasma frequency of a *single* molecule (or even of a solid material), the predicted wake dimensions ($\sim \omega_p^{-1}$) then far exceed the dimensions of any of the molecules used in this experiment. This incongruity appears to be the fundamental difficulty in the application of the wake model to molecules, since the other wake-model predictions are at least in qualitative agreement with our experimental results. The paucity of experimental data on forward-directed electron production, whether with solid or molecular targets, that are in good quantitative agreement with the predictions of the wake model might, alternatively, signal a weakness in the present model.

In summary, measurements of cusp electron spectra produced by ~ 1-MeV/u H^+, He^+, and He^{++} ions bombarding $m = 1$–7 hydrocarbon molecules indicate the appearance of a new component in the cusp for $m \geq 4$ molecules whose shape, skew, Z_P, Q_P, and energy-yield dependences, as well as the sudden appearance for $m \geq 4$ molecules, signal the onset of another forward-directed electron-production mechanism besides ECC, ELC, or FIRS. The properties of this component are generally in agreement with those expected for the advent of wake production in these larger molecules, although considerable difficulties still remain in this interpretation and warrant continued study.

We wish to thank the National Science Foundation for supporting the acquisition of a magnetic field controller unit (National Science Foundation Grant No. TFI-8023771). We also would like to acknowledge useful discussions with Rufus Ritchie.

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