Slow collisions between highly charged ions and C_{60}: Absolute ΔE values and cross sections

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We present energy-gain distributions for net one- and two-electron transfer in slow Ar^{q+}-C_{60} collisions. The ΔE-spectra for the former peak at $\Delta E_1 = 12.9 \pm 0.4$ ($nl = 7s$ and $7p$), 15.5 ± 1.0 ($n = 10$), 15.1 ± 0.5 ($n = 11$), and 13.8 ± 0.4 eV ($n = 12$) for $q = 8$, 13, 14, and 15, respectively. The total reaction cross sections are $\sigma_{tot} = 4.6 \pm 1.4$ ($q = 8$), 10.1 ± 2.8 ($q = 13$), 7.1 ± 2.0 ($q = 14$), and 10.0 ± 3.1 ($q = 15$) in units of 10^{-14} cm^2. These results are discussed within a simple qualitative model in which the transient localization of the positive charge on the C_{60}^+ ion just after transfer of one electron is a free parameter.

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I. INTRODUCTION

Whereas the electronic, rotational, and vibrational properties of free C_{60} now are fairly well established through photon spectroscopy [1], much less is known about its dynamical response to time-dependent perturbations from, e.g., moving ions, atoms or molecules [2]. Such information is crucial for understanding the fundamentals of, e.g., the behavior of C_{60} in atomic collisions and chemical reactions. In this work we present results from collisions of Ar^{q+} ions ($q = 8$, 13, 14, 15) with C_{60} at low velocities ($v \sim 0.2$ a.u.). We use our measured energy-gain (ΔE) distributions and total attenuation cross sections (σ_{tot}) to discuss the question about localization versus delocalization of charges on the C_{60} ions. This discussion is conducted within the framework of a model based on the over-the-barrier concept, which has been used extensively and with considerable success for descriptions of single- and multiple-electron transfer in slow ion-*atom* collisions for more than a decade (see, e.g., Ref. [3] and references therein).

Studies of slow collisions between highly charged ions and clusters may provide important linkage between the more established fields of ion-atom [4] and ion-surface collisions [5]. One interesting aspect in this context is that collisions on C_{60}, with its many loosely bound electrons, can be expected to lead to transfer of large numbers of electrons to highly excited states. This situation is similar to the one encountered in highly charged ion-surface collisions, where hollow ions and atoms are created [5]. One important difference, though, is that hollow ions may survive the soft encounter with a fullerene, but not the hard collision with a surface. This may lead to new possibilities for studies of this very exotic form of matter.

Walch *et al.* [6] reported C_{60}-fragmentation spectra and absolute cross sections for net transfer of up to eight electrons to slow Ar^{8+} ions. From the latter results they extracted experimental reaction radii in good agreement with those obtained from a model treating C_{60} as a conducting sphere (i.e., the positive charge appears to be smeared out and effectively acting as if positioned at the center of the fullerene) [7,8].

Recently, large cross sections ($\sim 2 \times 10^{-14}$ cm^2) for one- and two-electron transfer from neutral C_{60} to multiply charged ($q = 2,3$) fullerene projectiles was reported [9]. The results could be accounted for by assuming that the positive charges on the fullerene ions are spatially concentrated and that they are free to move on a time scale shorter than the collision time. Here, we use the experimental total reaction cross sections and ΔE-values for single-electron capture and discuss them within a simple over-the-barrier model for charge transfer, which takes the quantum nature of the projectile capture states into account. Within this model, we tentatively deduce the localization of the positive charge of C_{60}^+ immediately after transfer of one electron. It appears that a positive hole is *then* effectively positioned closest to the projectile at the distance $R_0 = 6.5 \pm 2.1 a_0$ from the center of the fullerene (the C_{60} radius is $a = 6.7 a_0$). We have extended this tentative reasoning further in order to discuss multiple-electron transfer from the target (the majority of these electrons are most probably lost by the projectile through autoionization [6]). The observed, unexpectedly large, ΔE values could possibly be explained by assuming that positive holes, close to the projectile when electron transfer occurs, rapidly move to the far side of the fullerene directly after the transfer.

II. EXPERIMENT

The Ar-ion beams for the present experiment were provided by means of the cryogenic electron beam ion source in the Manne Siegbahn Laboratory at Stockholm University. Ar gas was introduced in the ionization region of the source,

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where a 12-keV electron beam of about 150 mA ionized the atoms to the desired charge states through sequences of electron-ion collisions. The ions were extracted slowly from the source in ~100 ms beam pulses at energies of $\sim 3.3q$ keV and analyzed with a 90-degree bending magnet. This magnet limited the energy spread to $\sim 0.4q$ eV. The energies of the primary beams and the ΔE-distributions for net one- and two-electron transfer from C_{60} were analyzed by means of a retardation lens, a 180° hemispherical energy analyzer and a two-dimensional position-sensitive detector. The retardation lens reduces the kinetic energy by a factor of ~20, the analyzer separates different energy- and charge-state components of the beams and these separations are displayed as position distributions on the detector.

We calibrated the energy-gain scales on an absolute level, by means of a method that give accuracies in the range 0.3–1.0 eV. The method, which recently has been described in more detail elsewhere [10,11], relies on very precise measurements (1 part in 10^5) of the energies for the primary and the various charge-reduced beams. Typically, the positions of the primary beam on the detector were measured as a function of the retardation voltage for a fixed voltage (ΔV) between the analyzer spheres. Then we recorded (at least) one spectrum for net one-electron transfer with the same value of ΔV and retardation voltage $V_{\rm ret}^{1e}$. In the next step, we recorded a few more positions for the primary beam (still with the same ΔV) in order to check that no shift in the position had occurred. Finally, primary-peak positions were recorded for a new setting of the analyzer voltage $\Delta V'$. The primary beam energies, E_0, were determined through

$$E_0 = q\frac{V_{\rm ret}^{\rm pri}(N) - (\Delta V/\Delta V')V_{\rm ret}^{\rm pri'}(N)}{1-(\Delta V/\Delta V')}, \qquad (1)$$

where $V_{\rm ret}^{\rm pri}(N)$ and $V_{\rm ret}^{\rm pri'}(N)$ were the retardation voltages for the primary beam in position N (channel number) on the detector. Equation (1) and the corresponding expression for ΔE were derived in [10]. The ΔE-value for position N is

$$\Delta E(N) = (q-1)[V_{\rm ret}^{1e}(N) - V_{\rm ret}^{\rm pri}(N)] - E_0/q, \qquad (2)$$

while the ΔE-value for a peak in channel N' was obtained through $\Delta E(N') = \Delta E(N) + \phi(q-1)(N-N')$ with ϕ measured as $\phi = dV_{\rm ret}^{\rm pri}/dN$ [10].

The C_{60} target was provided by means of sublimation of 99.9% pure C_{60} powder in the temperature range 400 °C–500 °C. The powder was contained in the central part (30 mm) of a 60-mm-long stainless steel cylinder. The entrance and exit apertures, made in Cu disks that were mounted inside the tube, had diameters of 0.5 and 1 mm, respectively. Outside these apertures heat shields with 2-mm central holes were mounted in order to prevent condensation of C_{60} on the Cu disks. The cylinder was wound tightly with a double-folded heat wire and a thermocouple gauge was inserted in a drilled channel. We recorded the number of projectiles hitting the position-sensitive detector as a function of the temperature for a preset amount of charge hitting the exit slits of the separator magnet. Using this normalization and the pressure-temperature dependence of Abrefah *et al.* [12], we thus deduced total charge-exchange cross sections.

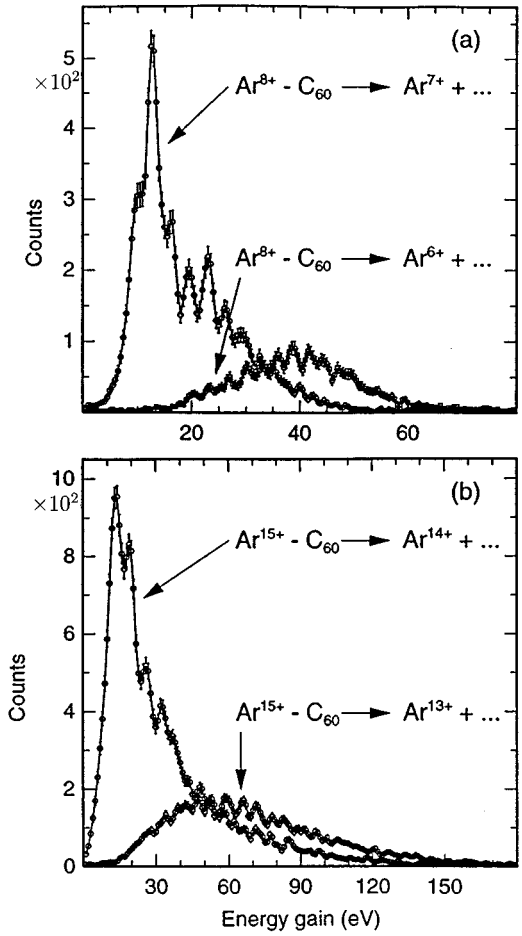

FIG. 1. Energy-gain spectra for net one- and two-electron transfer in $\sim 3.3q$ keV Ar^{q+}-C_{60} collisions. Note the difference in ΔE scales for (a) $q=8$ and (b) $q=15$. The error bars show statistical uncertainties.

III. RESULTS

The present experimental results are summarized in Figs. 1 and 2. In Fig. 1, we show the energy-gain spectra for net one- and two-electron transfer to the projectile for $q=8$ and $q=15$. The spectra are most likely due to:

$$Ar^{q+} + C_{60} \rightarrow Ar^{(q-s)+} + {C_{60}}^{s+} \rightarrow Ar^{(q-r)+} + {C_{60}}^{s+} + (s-r)e^-, \qquad (3)$$

where $r=1$ or 2 and $s \geq 1$ or $s \geq 2$, respectively. Fragmentation of ${C_{60}}^{s+}$ would only influence the data mildly, since the fragmentation time is expected to be much longer than the collision time (10^{-15}–10^{-14} s) [6].

In Fig. 2, we show the energy-gain spectra for $q=8$, 13, 14, and 15 where the upper curves are the added intensities for net one- and two-electron transfer. The lower curves, which show two-electron transfer alone, are affected by double collisions. For $q=15$ this contribution was deduced from the measured $(q-1)$ spectra for $q=15$ and $q=14$ and the corresponding absolute cross sections. Double collisions contribute with ~25–30% of the total $(q-2)$ spectra and are most important at the lower ΔE values. It is worthwhile to note that the addition of spectra relies on precise absolute

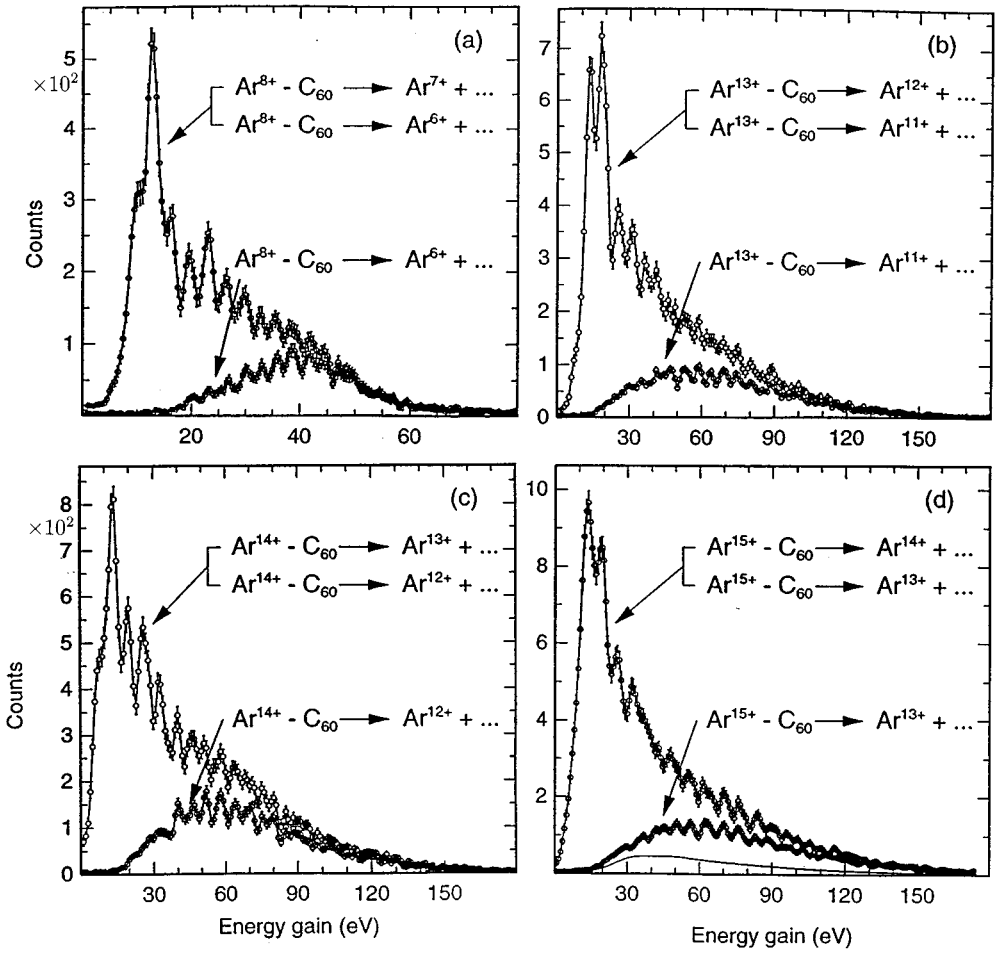

FIG. 2. Energy-gain spectra for $3.3q$ keV Ar^{q+}-C_{60} collisions for (a) $q=8$, (b) $q=13$, (c) $q=14$, and (d) $q=15$. The upper curves are the *summed* spectra for net one- and two-electron transfer to the projectiles, while the lower curves are the spectra for net two-electron transfer alone. The peaks at high ΔE values are most likely due to initial transfer of many electrons followed by electron emission from the projectile (cf. text). The error bars show statistical uncertainties.

calibrations of the energy-gain scales for the $(q-1)$ and the $(q-2)$ spectra. The peaks lowest in ΔE are measured to be $\Delta E_1=12.9\pm0.4$ eV, $\Delta E_1=15.5\pm1.0$ eV, $\Delta E_1=15.1\pm0.5$ eV, and $\Delta E_1=13.8\pm0.4$ eV, which we assign to single-electron capture to $7s$ and $7p$, $n=10$, $n=11$, and $n=12$ in $(q-1)=7$, 12, 13, and 14, respectively. The result for $q=8$ is consistent with the theoretical prediction of Thumm [7].

There is virtually no intensity at ΔE values lower than these, indicating that single-electron capture occurs without accompanying electronic or vibrational excitation of C_{60}. Similarly, close inspections of the kinetic energy distributions in the vicinities of the "primary" $(q+)$ beams with and without C_{60} gas in the cell indicated that pure C_{60} excitations with energies larger than $\sim 0.4q$ eV (the experimental resolution) are unimportant. At this point, we cannot make a definite assignment of the peaks closest to the ΔE_1 peaks, but we note that single-electron capture to $n=9$ $(q=13)$ and $7d-7i$, $6d-6h$, and $6s-6p$ $(q=8$: the left and right shoulders of $7sp$ and the peak close to $\Delta E=20$ eV) fit rather well with the measured peak positions.

Another important feature of the data is that the summed spectra (upper curves in Fig. 2) extend to large ΔE values. The intensities at high ΔE are mostly due to contributions from $(q-2)$ spectra, which have their maxima at much higher energies than the $(q-1)$ spectra. It is somewhat tempting to try to assign the sequences of nearly equidistant peaks in the summed spectra to initial transfer of progressively larger number of electrons from C_{60} in the fashion indicated by Eq. (3). It is also clear, however, that such an assignment must be built on a successful comparison with a physically sound model, since we do not measure the charge of C_{60}^{s+} after the collision. Such comparisons are very difficult due to, e.g., the close separations between adjacent peaks and the possibility of emission of energetic electrons which could distort the spectra. We thus refrain from attempting such an identification here. However, we believe that the large ΔE values are due to multiple-electron transfer, since the peaks in the $(q-2)$ spectra coincide with those in the summed spectra of Fig. 2 and become relatively more intense as ΔE increases. This suggests that a higher-lying ΔE peak can be associated with a larger tendency for net two-electron transfer and therefore it is plausible to assume that they are associated with an initial transfer of a larger number of electrons. Such a picture is consistent with the general experience drawn from a huge body of data on highly charged ion-atom collisions where many-electron transfer leading to emission of all but one or two of these electrons are known to be important processes [4]. Further, Walch *et al.* [6] found evidence for production of C_{60}^{s+} with s ranging at least up to $s=6$ in Ar^{8+}-C_{60} collisions.

The total charge-exchange cross sections are found to be very large $(\sim 10^{-13}$ cm$^2)$. In Table I, we list measured ΔE-values for single-electron capture, ΔE_1, along with identifications of the states involved, and total reaction cross sections $\sigma_{\rm tot}$. The total cross section for electron capture by 80-keV Ar^{8+} from C_{60}, was measured by Walch *et al.* [6] to

TABLE I. Energy-gain values for the first peak (the one lowest in energy; ΔE_1) in the spectra for net one-electron transfer and absolute, total, electron-capture cross sections, σ_{tot}, in units of 10^{-14} cm^2 for slow Ar^{q+}-C_{60} collisions. The capture states (nl) and the corresponding expected Q_1-values, $Q_1 = E_B(nl) - I_1$, are also shown. The error bars show one standard deviation.

q	ΔE_1(eV)	σ_{tot}	nl	Q_1(eV)
8	12.9±0.4	4.6±1.4	$7s,7p$	13.1,12.2
13	15.5±1.0	10.1±2.8	10	15.4
14	15.1±0.5	7.1±2.0	11	14.5
15	13.8±0.4	10.0±3.1	12	13.7

be $(4.4\pm1.8)\times10^{-14}$ cm^2, which is in agreement with the present result $(4.6\pm1.4)\times10^{-14}$ cm^2.

IV. DISCUSSION

As mentioned above, it is not quite clear how to describe the dynamical electronic properties of neutral and charged C_{60}. Is it appropriate to assume that an effect of charging C_{60} is that, from outside, it looks as if the charge was residing at the center of C_{60}? This point of view was advocated by, e.g., Walch *et al.* [6] and Thumm [7,13]. Alternatively, we may describe a charged fullerene as having its charges spatially concentrated on its ''surface'' as done by, e.g., Petrie *et al.* [14–16] and Shen *et al.* [9]. This issue is closely linked to the aromaticity of C_{60} and the mobility of its electrons. From the literature on chemical physics it appears that the latter issues are not yet settled [17]. Here, we will, in the hope of shedding some light on such problems, compare our data (ΔE_1, σ_{tot}) with a simple classical over-the-barrier model in which the apparent location of the positive charge of C_{60}^+ is a free parameter R_0. In the usual fashion of such models we assume that all impact parameters smaller than a critical, q-dependent, internuclear distance R_1 (measured from the ion $q+$ to the center of C_{60}) contribute to the cross section as $\sigma_{tot} = \pi R_1^2$. We take R_1 as the crossing of the quasimolecular potentials

$$U_{in}(R) = -(1/2)\alpha_0(q^2/R^4) \tag{4}$$

and

$$U_{out}(R) = (q-1)/(R-R_0) - (1/2)\alpha_1(q-1)^2/R^4 - Q_1(n). \tag{5}$$

The quantum nature of the projectile capture states is introduced through the term $Q_1(n)$ and the polarizabilities of C_{60} and C_{60}^+ are denoted by α_0 and α_1, respectively. The electrostatic interaction between the assumed positive hole at position R_0 from the center of C_{60} and the projectile is expressed by the term $(q-1)/(R-R_0)$ in (5). The attractive interaction between the remaining electrons on C_{60}^+ and the projectile is taken to be $-(1/2)\alpha_1(q-1)^2/R^4$, which includes (the major part of) the interaction of the projectile with the image charge. U_{in} and U_{out} can also be derived from the more general potentials of Bárány and Setterlind [8] by setting $\epsilon = 1$ in their model and introducing experimental and estimated values for $\alpha_0 = 540a_0^3$ [18] and $\alpha_1 = 390a_0^3$, respectively. We estimate α_1 as $\alpha_1 = \alpha_0(I_1^{ave}/I_2^{ave})^{3/2}$ with $I_1^{ave} = 17$ eV (from Thumm *et al.* [19]) and $I_2^{ave} = I_1^{ave} + 4 = 21$ eV taken as the estimated average binding energies of the 240 and 239 valence electrons of C_{60} and C_{60}^+. Note that a negative value of R_0 would correspond to a situation in which the positively charged hole resulting from transfer of the first electron would appear to be further away from the projectile than the center of C_{60}^+, while a positive value of R_0 would indicate a hole located on the projectile side of the singly charged fullerene ion. A R_0 value consistent with zero would indicate that the C_{60}^+ behaves as a metal sphere in line with the assumption made by Walch *et al.* [6] and Thumm [7].

Using our measured ΔE_1 values ($\Delta E_1 \sim Q_1$), we get $R_0 = +6.9\pm2.2$, $+12.2\pm3.9$, $+2.3\pm3.6$, and $+5.0\pm3.3$ for $q = 8$, 13, 14, and 15 by setting πR_1^2 [with R_1 from the crossing of (4) and (5)] equal to the experimental cross section σ_{tot}. The errors in R_0 (one standard deviation) are dominated by the relative errors in the cross section measurements, while the errors in the ΔE values give smaller contributions. The relative errors are obtained from the observed reproducibilities in the measurements of σ_{tot} (15–20 %) and are due to, e.g., uncertainties in the normalization procedure. The weighted average of the four values given above is $R_0 = 6.5\pm1.5a_0$. Although the scatter between the individual results may appear large it is fully consistent with a purely statistical analysis since three out of four measurements are within one standard deviation from the weighted mean. We include the absolute uncertainty in the target pressure, which is mainly due to the uncertainty in the vapor-pressure curve by Abrefah *et al.* [12], and arrive at the final result $R_0 = 6.5\pm2.1a_0$. This is in agreement with the mean radius of the (nuclear) structure of the C_{60}^+ molecule. We have tested the sensitivity of the result for R_0 on various model assumptions and we note, e.g., that neglect of the polarization terms in (4) and (5) would give only a slight modification of the result (to $7.7\pm2.2a_0$).

There are thus some indications favoring a single-electron charge-transfer model with the positive charge of C_{60}^+ positioned close to the fullerene surface on the side closest to the projectile just after electron transfer. We also note that the potential energy between a positively charged hole and the projectile becomes smaller if the hole is able to move to the far side of the singly charged fullerene ion on a time scale that is short in comparison with the collision time (10^{-15}–10^{-14} s). In order to discuss multiple-electron transfer we introduce the assumption that ''positive holes'' that are created through sequential electron transfer will be (sequentially) repelled to the far side of the fullerene. We stress here, though, that the assumptions about the motion of holes in C_{60} ions have *no* influence on the results for the parameter R_0.

In an approach including the classical over-the-barrier condition [8], the first electron can only be transferred when the barrier between the $1+$ hole and the $q+$ ion is sufficiently low *and* when there is a resonance between the potentials (4) and (5). The experiment actually yields the ΔE_1-values that are associated with the outermost potential curve crossings of (4) and (5) fulfilling both criteria, with the exception for $q = 13$ where population of $n = 10$ was observed while $n = 11$ was expected. The second electron can be trans-

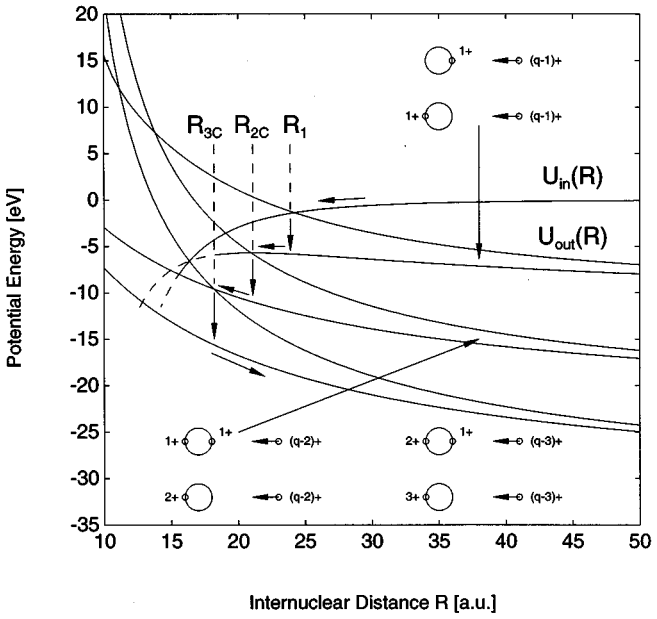

FIG. 3. Schematic potential energy curves for transfer of one, two, and three electrons to Ar^{8+} in slow collisions with C_{60}. The initial potential curve is given by (4) in the text and the upper single-capture potential is given by (5) with $R_0=6.5a_0$ and Q_1 taken to be the mean value for $7s$ and $7p$. The lower single-capture potential differs from (5) in that the positive hole is then localized on the far side of the fullerene. The internuclear distances R_{2c} and R_{3c} for transfer of the second and the third electrons are taken to be given directly by the barrier conditions. Also here relaxation of the potential energies occurs when the 1+ hole moves to the far side as indicated by the vertical arrows. A path leading to three-electron transfer is shown. For the two- and three-electron transfer curves, we have made the simplifying assumption that the 1+ holes merge on the far side of the multiply charged C_{60}^{s+} ions (cf. text).

ferred when the barrier due to the potentials from three point charges [1+ holes on each side of C_{60} plus the $(q-1)+$ ion] is low enough (cf. Fig. 3). Once the second electron has been transferred a second 1+ hole moves to the far side of C_{60}. For the moment we make the simplifying assumption that the two holes merge, since this has only a very small effect on the potential energy curves shown in the figure. Assuming that there are quasi continua of capture states for electron n:o 2,3,..., we arrive at model Q values and cross sections for initial removal of m electrons from the target. Initial, tentative, estimates built on this reasoning are able to explain the extents to large energies for the ΔE spectra at least in a qualitative way. For example, the ΔE values for Ar^{8+}-C_{60} would then reach a maximum of ~ 60 eV for transfer of six electrons (assuming full screening of the projectile charge by earlier transferred electrons), which is in agreement with the present results (cf. Fig. 2). There are, however, uncertainties present in the higher-order ionization potentials of C_{60} [we used $I_i(\text{eV})=7.58+4.0(i-1)$ as extrapolated from the ones given by Javahery *et al.* [20], cf. also Steger *et al.* [21]], in the amount of screening of the projectile by the transferred electrons, and in the positions of the positive charges on C_{60}^{s+}. The part of the model that deals with multiple-electron transfer is thus more speculative than the part concerned with single-electron capture.

V. CONCLUSIONS

We have presented experimental results on translational energy-gain and absolute and total charge-exchange cross sections for slow collisions between highly charged ions and C_{60}. These measurements show structured energy-gain spectra and large total reaction cross sections. Comparisons with a simple model seem to *indicate* that the charges of singly and multiply charged C_{60} can be treated as being spatially concentrated and mobile on the fullerene surface during the collision. By means of our simple qualitative model and the present experimental data, we have inferred a separation between the positive charge of C_{60}^{+} and its center of $R_0=6.5\pm 2.1a_0$ just after transfer of one electron to the slowly moving projectile. However, we do not consider the issue about charge localization during the collision to be settled due to the simplicity of the model and the relatively large uncertainties in the individual measurements of R_0. Still, many aspects of our experimental results can be rationalized if one assumes that a 1+ hole first localizes on the projectile side of C_{60}^{+} during electron transfer and then moves to the far side of the fullerene after the transfer. Treating this as a sequential process in our model would qualitatively explain the observation of very large Q values for multiple-electron transfer. The interpretation of the present data is consistent with the one given by Shen *et al.* [9] [slow multiply charged C_{60} (C_{70}) on C_{60}] and by Petrie and coworkers [14–16] (multiply charged C_{60} on various atomic gases). Also these authors concluded that their respective measurements of large cross sections and rate coefficients could be accounted for by assuming that the charges on positive C_{60} ions appear as positive holes that are movable on the surface.

ACKNOWLEDGMENTS

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