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MEASUREMENTS OF HELIUM EXCITATION IN Be^+–He AND Mg^+–He COLLISIONS

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We have observed level specific excitation of He($n = 2, 3$) in collisions of Be^+ and Mg^+ with He at projectile velocities of $\frac{1}{2}$ a.u. With both Be^+ and Mg^+ projectiles, excitation for small scattering angles (large impact parameters) is appreciable, indicating the occurrence of "direct" excitation. Angular differential excitation cross sections were measured with Mg^+ projectiles at c.m. angles up to 9.6 mrad (11.7 keV deg). "Molecular" excitation begins to dominate these cross sections at about 1.8 keV deg or $b \approx 1.5$ a.u. The total He($n = 2$) excitation cross section is $(6.86 \pm 0.38) \times 10^{-18}$ cm^2 at this velocity, as compared with a Mg^+ ($3s \rightarrow 3p$) cross section of $(8.49 \pm 2.09) \times 10^{-17}$ cm^2.

Quasi-one-electron (QOE) collision systems involve an outer valence electron and either two closed shell cores or one closed shell and a bare nucleus. The relatively simple nature of such systems makes them an obvious choice for study in order to learn about fundamental collision processes. A class of QOE systems that has been studied extensively is that involving rare gas targets and neutral alkali or alkaline-earth ion projectiles. Numerous theoretical and experimental researches involving such collisions have been reported in the literature [1]. By their very nature, these systems are generally characterized by processes involving the "active" valence electron. It has been observed however, that in particularly violent small impact parameter collisions, charge exchange and excitation processes involving closed-shell electrons occur and can actually dominate the angular differential cross sections (ADCSs) at sufficiently large scattering angles [1–5]. Such processes are interesting not only because of their paucity, but because they are the result of relatively strong interactions between the colliding species and can thus provide detailed information about the collision dynamics.

In recent extensive studies made in our laboratory of valence electron excitation in QOE systems, we have also observed significant target core (He) excitation at the highest velocities ($v = \frac{1}{2}$ a.u.) for collisions of Mg^+ and Be^+ with He [6]. A standard view of QOE collisions that has developed in the last decade [1] (primarily, but not exclusively with regard to the valence electron) is, that excitation results from either (i) violent collisions, n which excitation takes place at well localized quasi-nolecular curve crossings, or (ii) glancing collisions, where impulsive Coulomb interaction is responsible for the excitation. These mechanisms are often referred to as "molecular" and "direct" excitation, respectively. While it is in some sense artificial to make distinctions in this manner (all interactions in this context are Coulombic), the two mechanisms are conceptually useful in that they lead to qualitatively different behavior of the excitation probability P as a function of impact parameter b. Specifically, one would expect molecular excitation to be strongly impact parameter dependent and sensitive to the details of the quasimolecular structure, while direct excitation probability should be relatively insensitive to impact parameter.

In this picture, the target excitation which has been observed in low energy angular differential experiments [2–4,7] is evidently molecular in nature, in view of the sharp onset of excitation probability with decreasing b. This onset occurs as the wavefunctions of the collision partners overlap significantly, and specific curve crossings leading to core electron promotion occur. Small angle, large b, "direct" core excitation is insignificant at these energies. Direct excitation is maximal at velocities v given by the Massey criterion [1]

$$\frac{\Delta E a}{\hbar v} \simeq \pi, \tag{1}$$

where ΔE is the energy increment between the ground state and a particular excited state and a is a somewhat ambiguous interaction length, typically set equal to the combined radii of the collision partners. If we consider as an example the case of $Be^+ + He$ at 5 keV [4] and use radii of 1.97 and 0.55 a.u. for Be^+ and He respectively [8], the left hand side of eq. (1), or the "Massey parameter", is equal to 13 for He($n = 2$) excitation. Using the Massey criterion, Be^+ energies of ~ 85 keV could be expected to give maximum direct excitation of these states. Thus, it is not surprising that the low energy

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experiments observe only molecular core excitation. A few QOE experiments at higher velocities [9,10] involving alkali atoms or alkaline-earth ion projectiles have also observed core excitation, but these were optical emission experiments integrated over scattering angle and done at isolated energies, and as such provided little information about the excitation mechanisms involved. Jensen et al. [11] have observed excitation of the Mg^{2+} core as a function of energy in $Mg^+ + He$ collisions, and saw the onset of direct excitation for Mg^+ energies > 50 keV.

The data reported in this paper were taken using the technique of ion energy-loss spectroscopy. This technique has the advantage that data analysis does not require cascade corrections or the knowledge of detector efficiencies. The apparatus we used has been extensively described in the literature [12,13]. It is capable of measuring ADCSs with an angular resolution of < 120 μrad (lab frame), and has a typical energy resolution of one part in 10^5. $^9Be^+$ and $^{24}Mg^+$ projectiles were produced in a Colutron G-2 ion gun with energy widths between 0.5 and 1.5 eV. Following mass selection in a Wien filter, collimation, and acceleration, the ion beam traversed a static He target [13]. Scattered projectiles were magnetically analyzed and decelerated prior to passage through a spherical energy analyzer and detection by a Johnston Labs electron multiplier.

Energy-loss spectra for Mg^+ and Be^+ projectiles with velocities of $\frac{1}{2}$ a.u. (56.25 and 150 keV lab energy, respectively) are shown in figs. 1 and 2. These spectra were taken at 0° scattering angle with collimating slits providing (lab) angular resolutions of approximately ± 57 μrad for Mg^+ and ± 104 μrad for Be^+. The maximum energy–angle product (τ) contributing to the observed features is thus 0.49 keV deg for Mg^+ and 0.34 keV deg for Be^+. The "one-electron" nature of both systems is immediately evident. The first excitation of Mg^+ at 4.4 eV and Be^+ at 4.0 eV, followed by higher excitations of the ions, completely dominate these spectra. There is, however, excitation of He($n = 2$) states in both spectra, and our energy resolution allows us to observe higher He n-levels in the Be^+ spectrum. Equivalent features were absent in similar 0° spectra taken with Mg^+ projectiles at 30, 66.7 and 100 keV. Given that Olsen et al. [4] had not observed He excitation below 10 keV deg in $Be^+ + He$ collisions (albeit at low energy), we were initially surprised to see *any* target excitation with τ limited to less than 0.5 keV deg. Another interesting feature of these data is what appears to be some 2^3S excitation with Be^+ projectiles. There is no evidence for 2^3S excitation with Mg^+ projectiles at the same velocity. Our energy resolution does not allow us to distinguish between 2^1S, 2^1P and 2^3P excitation.

Angular differential cross sections were measured for He($n = 2$) excitation with Mg^+ projectiles. These were obtained by rotating the accelerator to either side of the central maximum and measuring ion count rates at several angles for the appropriate energy loss. The results are shown in fig. 3. The ADCSs decrease by an order of magnitude within the first two mrad (c.m.) and then remain roughly constant up to 4 mrad before dropping again. We note that the angular resolution of our apparatus for all the Mg^+ data was ± 0.4 mrad (c.m.). Thus, the energy-loss spectrum of fig. 1 was obtained with collisions involving ADCSs on the first, sharply descending part of the curve in fig. 3. By integrating our ADCS results, we obtain a total cross section for the He($n = 2$) excitation of $(6.86 \pm 0.38) \times$

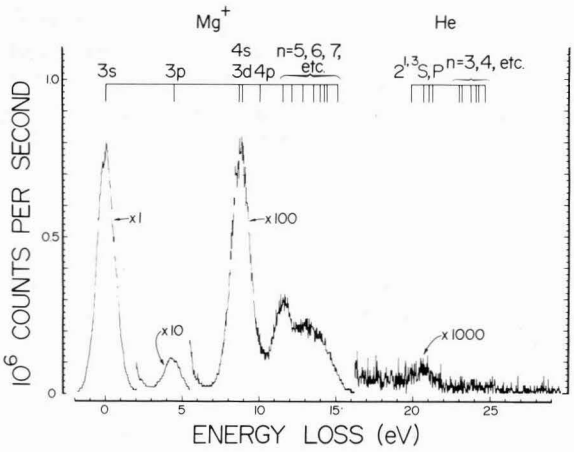

Fig. 1. $Mg^+ + He$ energy-loss spectrum; 0° scattering angle; angular resolution: ± 57 μrad (lab); energy resolution: fwhm ≈ 1.4 eV; target thickness: 4.5×10^{14} cm^{-2} (10 μ He gas pressure).

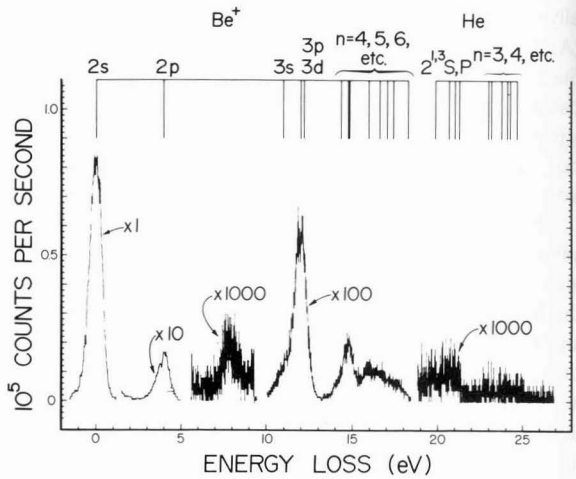

Fig. 2. $Be^+ + He$ energy-loss spectrum; 0° scattering angle; angular resolution: ± 104 μrad (lab); energy resolution: fwhm ≈ 0.8 eV; target thickness as in fig. 1. Note the energy-loss feature at 8 eV: this corresponds to double collision events involving two first excitations of the valence electron.

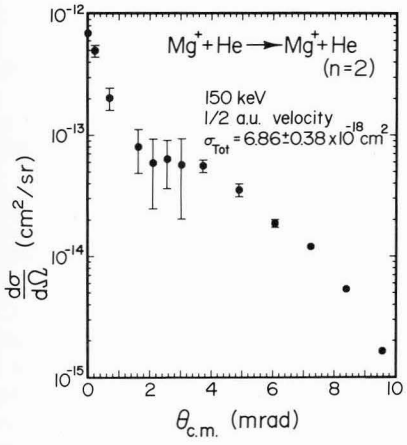

Fig. 3. He($n = 2$) angular differential cross sections. The error bars represent the standard deviation of the mean of all runs.

10^{-18} cm^2. This is only an order of magnitude less than the total cross section for the Mg^+ (3s → 3p) excitation ($(8.49 \pm 2.09) \times 10^{-17}$ cm^2) that we have measured, as compared to the [He($n = 2$)]/[Mg^+(3p)] peak height ratio in the 0° energy-loss spectrum of $\sim 5 \times 10^{-3}$. As in the low energy experiments, small angle collisions contribute little, if anything, to the total core excitation cross sections.

To further elucidate the collision dynamics of the Mg^+ + He system, we have transformed our smoothed ADCS results to yield a P vs b plot [4,13]. An exponential (Born–Meyer) scattering potential of the form

$$V(R) = Ae^{-BR} \tag{2}$$

was used, with the strength and range parameters A and B given by Sondergaard and Mason [14]. Several other physically reasonable parameter values were tried

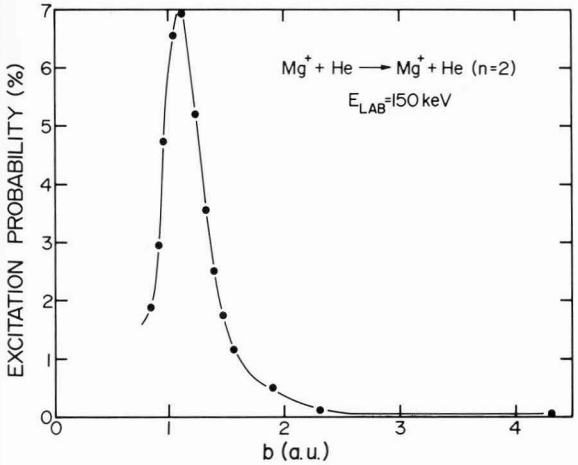

Fig. 4. Probability of He($n = 2$) excitation vs impact parameter. These results are obtained from the data of fig. 3, transformed using an exponential potential (see text).

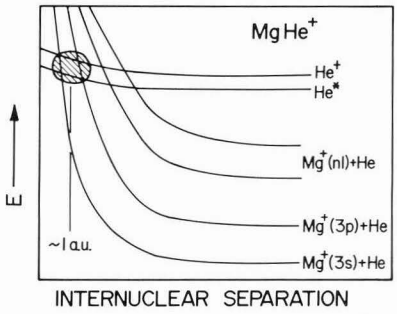

Fig. 5. Schematic representation of the $MgHe^+$ quasimolecule. The value of ~1 a.u. for the crossing of the ground state with higher-lying channels has been calculated in ref. [15] for the isoelectronic NaHe system.

and yielded $P(b)$ values which were essentially indistinguishable from values given by the Sondergaard–Mason parameters. The results are shown in fig. 4. The P vs b curve is typical of molecular excitation, with the excitation probability rising sharply over a fairly narrow range of impact parameters between about 1.1 and 1.6 a.u. The probability at larger b, corresponding to the small angle ADCSs before the "plateau" at θ(c.m.) = 2 mrad, is attributed to direct excitation. This conclusion is supported by comparing fig. 4 with the data of Tuan et al. [7] for the isoelectronic Na + He collision system at lab energies of 10 keV. They observed a sharp decrease to zero in excitation probability at $b = 1.4$ a.u., without the attendant "tail", characteristic of direct excitation, at higher b.

The quasimolecular curves responsible for this behavior are shown schematically in fig. 5. At large internuclear separation R the large energy difference between the incoming molecular states corresponding to Mg^+(3s) + He and Mg^+(3s) + He($n = 2$) precludes any molecular excitation at low collision velocities. However, as R approaches 1 a.u., the Mg^+(3s) level is strongly promoted through levels correlating with He core excited states (He* or He^+). This is a result of the promotion of the He(1s) 3dσ single electron orbital through the Mg^+ 3pσ and 3sσ valence electron orbitals [15]. The position of these strongly avoided crossings in the isoelectronic NaHe system has been calculated by Courbin-Gaussorgues and co-workers [15,16] to occur near 1.2 a.u. As this value of R is reached, strong radial and rotational coupling at the crossings populate the He* and He^+ channels, and can lead to higher $Mg^+(nl)$ Rydberg levels through subsequent couplings at larger R. Our P vs b results peak near 1.2 a.u., in accord with ref. [15], and with Tuan et al.'s data. As the collision velocity increases towards the Massey peak value, direct excitation for larger b collisions becomes appreciable.

Why Be^+ projectiles appear to be exciting the 2^3S states while the Mg^+ projectiles do not, remains a puzzle. One would not expect direct excitation to result

in triplet population because of the Wigner spin rule; triplets would have to be excited in hard collisions with appreciable amplitude for spin exchange processes. This would explain why no triplet excitation is seen with Mg^+ ions (excluding the possibility that unresolved 2^3P states are being produced). The Mg^+ 0° energy-loss spectrum is limited to c.m. scattering angles of 0.4 mrad (corresponding to $b \geq 2.1$ a.u.) where only direct excitation occurs. Indeed, one might expect triplet excitation in the "plateau" region of the $MgHe^+$ ADCSs. The Be^+ energy-loss spectrum centered at 0° includes c.m. angles as large as ± 0.34 mrad. Transforming angles using a Born–Meyer potential for the $Be^+ + He$ system, this angular spread corresponds to the range $b \geq 1.8$ a.u. While this is a somewhat smaller limiting b than that for the Mg^+ spectra, the Be^+ radius is also considerably smaller (1.97 vs 2.42 a.u. [8]), so simple geometric core penetration arguments do not solve the problem. In limited $Be^+ + He$ charge transfer ADCS data that we have taken, there is no indication of significant "molecular" charge transfer (as might be indicated by a decrease in slope, a plateau, or even a rise in the ADCSs with angle) below 1 mrad c.m. In addition, the $BeHe^+$ quasimolecular curves exhibit no strongly avoided crossings in the region between 1 and 2 a.u. equivalent to those in the $MgHe^+$ case [17]. All these factors argue against the observed triplet excitation in the Be^+ energy-loss spectrum. A measurement of $He(n = 2)$ ADCSs with Be^+ projectiles is clearly needed.

In summary, while it is clear from our energy-loss spectra that Be^+–He and Mg^+–He are collision systems which essentially have only one active electron, the deviation from this picture represented by He excitation provides valuable insight into the "molecular" aspects of the collision process. Future studies of small b valence electron processes in these QOE systems would undoubtedly clarify the dynamics of these collisions.

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