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Molecular effects in optical beam-foil spectroscopy using H^+, H_2^+, H_3^+, D^+, D_2^+, D_3^+, HD_2^+, and HeH^+ projectiles

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Relative beam-foil level excitations in hydrogen and helium have been studied as a function of the foil thickness using incoming beams of H^+, H_2^+, H_3^+, D^+, D_2^+, D_3^+, HD_2^+, and HeH^+, thus varying the average distance between the fragments at the exit of the foil. Large changes in level excitation are observed in many cases for molecular projectiles when using thin foils. For molecular-hydrogen projectiles the same enhancement of excitation is found for all hydrogen levels studied. However, a remarkable difference is found for the heteronuclear projectile HeH^+. In that case, the uniform enhancement for different hydrogen levels vanishes. The results are discussed.

I. INTRODUCTION

Recently, molecular effects in beam-foil excitations have been studied using beams of H^+, H_2^+, H_3^+, O^+, and O_2^+ and optical detection.[1] For hydrogen it was found that all levels with principal quantum number n in the interval 3–9, showed population enhancements of 20 and 45% when H_2^+ and H_3^+ projectiles were used, instead of protons at a constant foil thickness. More irregular changes in level populations were established for oxygen. The findings indicate that for a substantial fraction of the molecular projectiles, the individual nuclei from an incoming molecular projectile ion are still fairly close together after having passed through the foil, causing a change in level excitation.

When a monoatomic projectile traverses a solid, the target atoms will respond to the projectile through coherent displacements of the electrons toward the projectile trajectory. Bohr[2] has referred to the phenomenon of coherent electron displacements as the "wake" behind the particle (for references to recent work, see, e.g., Refs. 1, 3, and 4).

When a molecular projectile enters a foil, the binding electrons of the projectile are stripped off, and the nuclei will start moving apart due to their Coulombic repulsive forces. For the low beam energy used in the present work, multiple scattering will also, to a large extent, contribute to the separation of the nuclei. The so-called Coulomb expolsion may be counteracted, partly because the electrons inside the solid will screen the nuclear charges, and partly because one of the nuclei may be caught in the wake potential of another nucleus so that it will ride along through the foil at a very short distance from the leading nucleus.[5-8] However, no matter what process governs the separation, it is clear that the distance between the fragments at the exit is short enough to influence the excitation.

The electrons of hydrogen molecular projectiles are stripped off readily when the projectile enters the foil.[9] Therefore, the foil excitation of hydrogen must result from electron pickup when the projectile leaves the back of the foil.[9] Recent data from multiply ionized projectiles indicate that the Rydberg-state excitation seen in beam-foil processes results from capture of an electron from the carbon-foil valence band into the excited state of the projectile.[10] Our earlier work[1] established molecular effects in optical beam-foil spectroscopy, but no clear conclusions were drawn. Therefore, we have continued our investigations of the excitation enhancement in search for a better, unambiguous explanation. For this purpose we have used D^+, D_2^+, and D_3^+ ions in addition to H^+, H_2^+, and H_3^+, because with the heavier-isotope nucleus d, the increase in internuclear distance when passing through the foil, caused by multiple scattering as well as by the Coulomb explosion, will be smaller than for the lighter-isotope nucleus p. We have also used the heteronuclear systems HD_2^+ and HeH^+. For heteronuclear systems an additional feature has to be taken into account, namely, that the different nuclei may have different stopping powers, and if so, the various nuclei can be split apart due to this. Our measurements with HeH^+ were done at an energy of 50 keV per nucleon, where the stopping powers for protons and helium are known to be very similar.[11] For deuterons, for which we have been unable to find previously reported data on stopping powers, we measured the energy losses and, within our errors, found them to be equal to those for protons at the same velocity. Molecular effects have previously been observed also in charge-state distributions and stopping-power measurements. For detailed references, see Refs. 1, 3, and 5–8.

II. EXPERIMENTAL

The measurements were carried out at the 400-kV heavy-ion accelerator at the Research Institute of Physics in Stockholm.[12,13] Intensities of spectral lines (200–600 nm), emitted from the projectiles immediately after having passed through the foil, were observed by means of a quantum-efficiency-calibrated optical monochromator. When scanning a spectral line, a normalization has to be carried out with respect to the beam intensity and the foil condition by using an optical monitor unit, as described in Ref. 14. However, the monitor has to be calibrated for the different beam energies and beam projectiles used in order to make comparisons between different molecular projectiles possible. This was done by intermittently measuring the beam current in a Faraday cup without a foil and the light yield in the monitor with a foil in the beam path. This calibration is what limits the accuracy in the measurements. From checks of the reproducibility we estimate the errors in the reported relative intensities to be typically 5%. The treatment of data to deduce relative level populations is described in Ref. 1. Projectile exit energy distributions were recorded by a 90° electrostatic analyzer.[15]

The mass analyzing magnet of the accelerator has only moderate mass resolution. Thus, e.g., D^+ cannot be separated from H_2^+, and therefore it was essential that the deuterium was purified to a high degree, since the ion source forms molecular-hydrogen ions with much larger concentrations than monoatomic ions. However, when deuterium was admitted to the ion source, and the analyzing magnet was set at mass number 2, no protons from H_2^+ dissociated in the foil could be observed in the energy analyzer. Of all molecular combinations of H and D, only HD_2^+ is uncontaminated by others and, since HeD^+ could not be separated from D_3^+, only HeH^+ and not HeD^+ was used.

After the foil chamber there is a 90° electrostatic analyzer[15] of radius 0.5 m, allowing energy distributions of particles having passed essentially undeflected through the foil to be studied. In this way, additional information, namely, about energy loss and straggling, was obtained. The precision of the energy analyzer is better than 0.5%.[15]

The particle-energy spectra were scanned also with no foil in the beam path. From the small size of the peaks, originating from particles which had undergone dissociation in collisions with the residual gas, it could be seen that the vacuum was so good that such dissociation processes could be ignored.

The foil thicknesses were determined from measuring the energy loss of 200-keV protons in the different foils and by applying the stopping graph for hydrogen in carbon given in Ref. 11. Four different target thicknesses were used. By using a mass density of 1.65 g/cm^3 for carbon,[16] the foils turned out to be around 19, 41, 59, and 70 nm. We must, however, mention that the mass density of 1.65 g/cm^3 (Ref. 16) differs from that given in Ref. 11, which is 2.266 g/cm^3.

III. RESULTS AND DISCUSSION

A. Particle-energy spectra

When monoatomic incoming projectiles were used, the particle-energy spectrum always contained one single, fairly narrow peak with no structure (except for He where He^+ as well as He^{2+} was observed), for an example, cf. the top of Fig. 1. With incoming molecular projectiles, molecular ions as well as monoatomic ions might appear after the foil. For the thinnest foils used and with molecular incoming projectiles, the peak of monoatomic projectiles after the foil clearly showed some structure which could not be resolved into two or more groups, cf., e.g., Fig. 1 (bottom). We also detected a small fraction of the molecular beam which had survived the passage through the foil. This peak did not exhibit

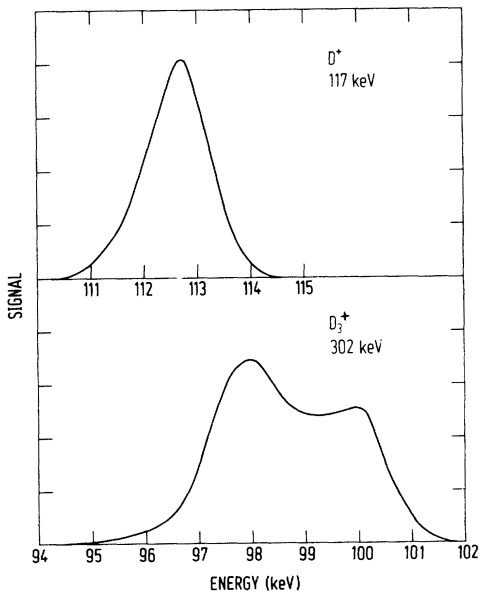

FIG. 1. Signal from the particle-energy analyzer versus projectile energy after the foil. The upper part of the figure is for incoming D^+ projectiles at 117 keV, and the lower part is the energy spectrum for outgoing D^+ from incoming D_3^+ at 302 keV.

any apparent asymmetric structure.

Structure like that presented in Fig. 1, (the bottom part), has been seen previously and been interpreted[5-8] in terms of the wake potential generated by the particles as they traverse the solid. Thus, our particle-energy spectra indicate that wake-riding effects are active, even at our fairly low projectile velocities. The structure in the energy peak (Fig. 1, bottom part) disappeared gradually when the foil thickness was increased, indicating that multiple scattering, combined with the repulsive Coulomb force, gradually counteracts the binding effect of the wake.

B. Photon intensity versus foil thickness

Figures 2–4 plot photon intensity enhancements as functions of the foil thickness for incoming projectiles of H_2^+, H_3^+, D_2^+, D_3^+, and HeH^+. The measurements were all carried out at the same projectile velocity after the foil, corresponding to 50 keV per nucleon. Here the enhancement is the photon yield per incoming nucleus using molecular projectiles, divided by the corresponding photon yield per nucleus when monatomic species are accel-

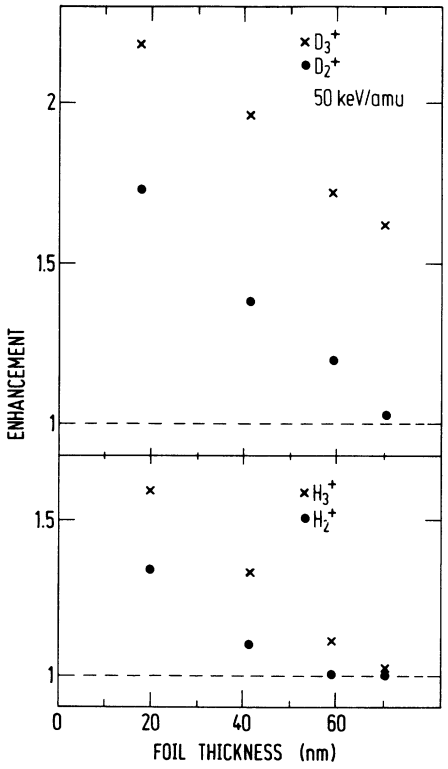

FIG. 2. Average photon intensity enhancements for Balmer lines (cf. Table I) versus foil thickness for D_2^+, D_3^+ (top), and H_2^+, H_3^+ (bottom) projectiles at 50 keV per nucleon.

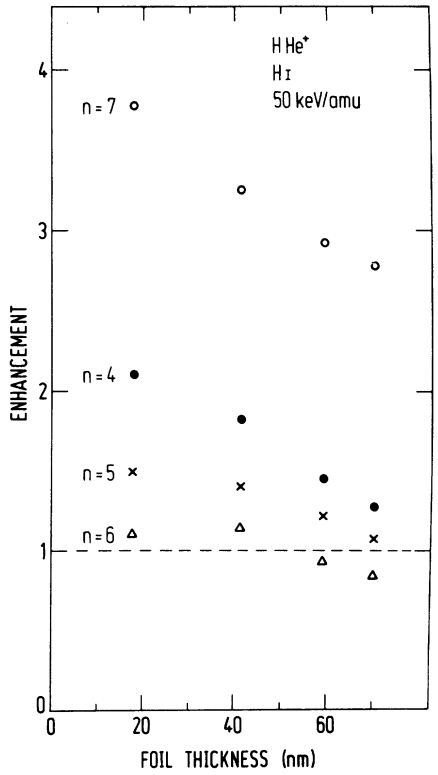

FIG. 3. Photon intensity enhancements for Balmer lines versus foil thickness for HeH^+ projectiles at 50 keV per nucleon.

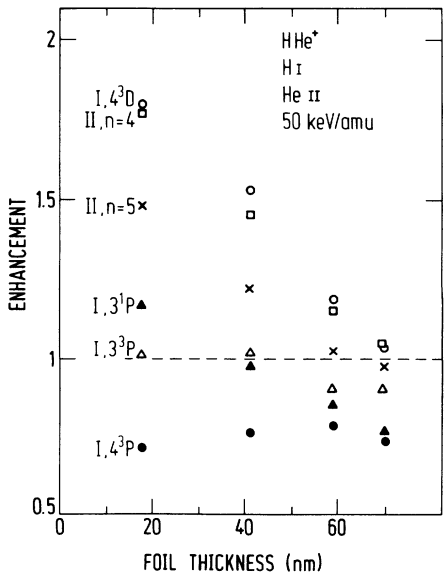

FIG. 4. Photon intensity enhancements for some helium lines versus foil thickness for HeH^+ projectiles at 50 keV per nucleon.

erated.

With H_2^+, H_3^+, D_2^+, and D_3^+ we can see in all cases (Fig. 2) that the photon yield per nucleus is greatly enhanced for the thinnest foils, and also that the enhancement almost disappears for the thickest foils, except with D_3^+ projectiles. Note especially with the thinnest foils and D_3^+ projectiles that the light per particle has increased by more than a factor of 2 above that induced by D^+. Also, for all foil thicknesses (or dwell times in the foil), the enhancement is larger for deuteron-containing species than for proton-containing ones. This is not unexpected, because for deuterium and proton molecules at the same velocity and foil thickness (i.e., the same dwell time in the foil), the Coulomb repulsion will be the same but, due to their larger mass, the deuteron clusters are scattered less. (We mention that we naturally observed equal excitation probabilities for H and *D* when the monoatomic projectiles H^+ and D^+ were accelerated.) It is not clear whether the difference in yields between proton- and deuteron-containing species is wholly due to the slower separation of the deuterons; i.e., if the abscissa scale in Fig. 2 could be correctly converted to average distances between the nuclei at the exit of the foil, equal yields would follow.

Assuming a pure Coulomb explosion from an initial separation of 1.17 Å (the value corresponding to the maximum of the initial distribution given in Kanter *et al.*[17], neglecting screening and multiple scattering) we have calculated nuclear exit distances for H_2^+ and D_2^+. Plotting the yields against this distance, instead of the foil thickness brings the curves for H_2^+ and D_2^+ closer to each other, but the D_2^+ values are still 10–20% higher. The neglect of multiple scattering is probably not justified (cf. Gay and Berry[18] who claim that at this energy this is the dominating process). However, it is not clear whether the curves would be brought together if multiple scattering is included.

A possible isotope effect can result from differences in the incoming beam species as well as from an isotopical differentiation in the beam-foil interaction. In the ion source, various rotational and vibrational levels of the same nuclear species will be excited. The lifetimes of such levels are large compared with the time it takes the ions to reach the foil. For deuteron-containing molecular ions, the population of rotational and vibrational levels will show a broader distribution than that for proton-containing molecular ions,[19] and this may cause the apparent differences between the top and bottom sections of Fig. 2. In addition, isotopical differences may occur during the molecular dissociation processes at the emergence of the foil, taking place at the same time as the beam-foil electronic excitations are created.

There have been some attempts to visualize molecular beam-foil phenomena as electron capture at the foil exit into some quasimolecular levels combined with electron sharing or excitation during the nuclear dissociation taking place downstream from the foil.[18,20] Therefore, we shall here compare our results to excitation in dissociation.

Möhlmann *et al.*[21] found in a dissociation study, in which they bombarded H_2 and D_2 with 0–2000-eV electrons, that the H Balmer cross sections are larger than the D Balmer cross sections for all projectile energies. Also, the isotope effect changes with n. We observe in the present work no dependence on n, and also that the Balmer light from H is enhanced less than that from D, cf. Fig. 2, both findings being different from the dissociation results of Möhlmann *et al.*[21]

Khayrallah[22] has estimated a power-law level population in electron-induced dissociation of H_2 and D_2. He finds that the relative level population can be approximated by n^p, with a value of p between −5 and −6.5. This differs rather much from what we find, namely $p=-3.7$. Also, we find the same power law for H^+, H_2^+, H_3^+, D^+, D_2^+, and D_3^+, whereas in collisions of electrons with H_2 and D_2, H and D have different powers.[22] Again this makes us believe that the assumption of dissociation from a high-lying state, and the potential-energy curve diagram do not explain our beam-foil excitation results. Our above-mentioned result for the power $p=-3.7$ is, on the other hand, equal to what is found in ion-atom collisions.[23,24]

The difference in nuclear scattering was also apparent when using the heteronuclear projectile HD_2^+. The Balmer spectral lines H_β and D_β are separated by 0.13 nm due to different reduced masses. Thus, using narrow slits (30 μm) for the monochromator, a clear asymmetry was revealed, originating from the shift in wavelength between H_β and D_β. A D_3^+ beam was used to determine the width of the spectral line at 486.1 nm. The intensities for D_β and H_β when using a HD_2^+ beam were obtained by a peak-fitting program. In this analysis the linewidth, determined earlier, was kept fixed as well as the distance between the peaks (0.13 nm), thus just fitting the intensities of the lines and their locations. The result with a foil thickness of about 50 nm was that the light intensity per incoming deuteron was 2.6 ± 0.5 times higher than that per incoming proton (i.e., in the spectral scan the line intensity for D_β was 5.2 times higher than that for H_β).

The total line intensities, induced by HD_2^+ and

D_3^+ (normalized to equal currents) were almost equal. This means that, in addition to the above result for the I_{D_β}/I_{H_β} intensity ratio from HD_2^+ projectiles, there is with HD_2^+ an enhancement of the D light per D by $(20\pm5)\%$ compared to the D intensity using D_3^+, and also a reduction of the H_β light intensity by $(15\pm5)\%$ compared to the case with a pure proton beam.

HD_2^+ is an asymmetric top. More energy levels are therefore permitted for this molecule than for H_3^+ and D_3^+, because the latter ones have higher symmetry. Thus, some of the incoming HD_2^+ clusters can well have been with some internuclear separations which do not occur for H_3^+ and D_3^+. Also, the post-foil molecular dissociations will be different for the asymmetric HD_2^+. Most likely, the two deuterons will be closer together than the proton and any of the deuterons. Such an asymmetric charge distribution may result in an overall increase in electron capture by either of the deuterons at the expense of electron capture by the proton.

With H_2^+, H_3^+, D_2^+, and D_3^+ projectiles we observed, for the same projectile and with a given foil thickness, that all Balmer lines ($n=4$–11) showed the same enhancement within errors (cf. Table I) or, in other words, that the change in level excitation is independent of the excited level, in agreement with our earlier result for protons and hydrogen molecules.[1] This is worth observing because the internuclear separations at the foil exit are comparable to the mean radii of the excited states studied. As mentioned above, it is difficult to estimate the internuclear separations in a molecular cluster upon emergence from the foil. However, pure Coulomb explosion may be assumed (cf. also Ref. 16) to get a rough estimate of the separation to compare with the sizes of different hydrogenic states. The beam energy (50 keV/amu) used in this experiment corresponds to a velocity of 3 nm/fs, which gives the dwell times in the foils of our experiment as approximately 6, 14, 20, and 23 fs. The mean radii of the $n=4$ states in hydrogen are ~0.8 nm. Our calculations yield an explosion time of around 10 fs for H_2^+ and 14 fs for D_2^+ to reach a separation of 0.8 nm. Thus, we conclude that we have separations between the fragments both smaller and larger than the mean radii of different hydrogenic states. We find it remarkable that it does not seem to matter whether the internuclear distance is smaller or larger than the mean radius of the excited atom.

TABLE I. Population enhancement, determined from line intensities in the Balmer series, for atomic levels n in deuterium when using molecular projectiles. The foil thickness in this particular case was 30 nm.

n	$I_{D_2^+}/I_{D^+}$	$I_{D_3^+}/I_{D^+}$
4	1.45	1.89
5	1.39	1.76
6	1.35	1.76
7	1.42	1.80
8	1.34	1.75
9	1.50	1.88
10	1.65	1.93
11	1.38	1.69

The uniform enhancement of all levels studied implies that the molecular effect for homonuclear projectiles will also appear as a charge-state distribution phenomenon. The enhancement of the neutral fraction when using a molecular-hydrogen beam has been studied by Gaillard *et al.*[16] They also gave a tentative explanation which predicts the enhancement when using H_3^+ to be twice as large as for H_2^+, and also the enhancement for "zero" foil thickness to be 50 and 100% for H_2^+ and H_3^+, respectively. These predictions are not contradicted by the results for H_2^+ and H_3^+ shown in Fig. 2, taking the uncertainties of the points into account, but they disagree for D_2^+ and D_3^+; see also Fig. 2. Whether or not the isotope effect in this work only results from different separation rates, it is concluded that the predictions in Ref. 16 do not cover the whole process.

In addition to projectiles containing only hydrogen isotopes, we also studied light intensities using a HeH^+ beam. Gay and Berry[18] have recently observed that the level population, as well as the resultant atomic level alignment, can be changed appreciably when HeH^+ is accelerated rather than He^+. These authors suggested that their results could be explained by considering a combination of two effects, namely, (i) a reduction in nonradiative deexcitation processes at the foil exit which will lead to a total increase in level excitations with molecular projectiles, and (ii) quasimolecular curve crossings. However, with HeH^+ we observe up to a 30% reduction in level population for the He I, $4p\,^3P$ term, see Fig. 4, and this disagrees with (i) above.

We observed no change in relative intensity for the He I 388.9-nm transition using HeH^+, in agreement with Gay and Berry.[18] They, however, as well as Groeneveld *et al.*,[25] observe a change in alignment for that transition. Thus, molecular effects may well be present though no relative intensity change is observed.

From Figs. 3 and 4 we observe that with HeH^+ projectiles the spectral line intensities change with foil thickness, but different levels change in different ways, in contrast to the case with pure hydro-

genic species (Fig. 2). A similar complicated picture was found for oxygen previously.[1] It is remarkable that the picture is completely different for HeH^+ compared to H_2^+. For HeH^+ the enhancement is not uniform any more. The population is reduced for some levels and enhanced for others, for helium as well as for hydrogen, without showing an obvious pattern. In particular we note the dramatic enhancement for the hydrogen $n=7$ line, quite at odds with the trend for lower n values. The observation of molecular effects both as reduction and as enhancement is, however, consistent with the effect on I_{H_β}/I_{D_β} light from HD_2^+ projectiles discussed above. It would be desirable to check different levels when using HD_2^+ in the future to see whether reduction or enhancement follows the uniform behavior found for monoisotopic projectiles, or whether it exhibits a more complicated picture like that of HeH^+

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