Effect of the anion orbital momentum on mutual neutralization processes

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Recent improvement of the merged beam setups allowed to study the partial cross section for a wide range of atom-atom mutual neutralization (MN) reactions. This range is mainly limited by two factors: The ability to produce the ion beams (ideally in a single electronic state) and the mass ratio between the anion and the cation. This mass ratio limit is around 10 for the UCLouvain single pass setup and 20 at the DESIREE facility. Therefore, it does not allow studying reactions with higher mass ratios, such as Fe⁺⁺⁺⁺H⁻. To circumvent this experimental limit, it would be useful to replace the H⁻ anion by another anion with similar properties in order to indirectly measure this reaction.

Using the UCLouvain merged beam setup, we recently measured the branching ratio for the MN between C⁺⁺/N⁺⁺/O⁻⁻ and C⁺⁺/Si⁺⁺/O⁻⁻/P⁻⁻/D⁻⁻/S⁻⁻ [1]. This systematic study allowed us to make a comparative study, e.g., the effect of the anion's binding energy on the outcome of MN reactions. But this analysis in terms of electron affinity was guided by previous studies which mainly focused on reactions involving anions in an S state. While the anion binding energy dependency was effectively observed, the branching ratios differ largely for systems whose anion has a similar binding energy but a different orbital momentum, e.g., C⁺⁺/O⁻⁻ and D⁺⁺/P⁻⁻.

Using an anion centered asymptotic model combined with a multi-channel Landau-Zener model, we have found that higher anion orbital momentum increases the number of Λ molecular symmetries in which the collision happens and that the interaction potential between ionic (A⁺⁻B⁻) and covalent (A⁰⁻B⁰) states decreases for higher Λ. This allows the production of lower excited states of the neutralized cation, but also lowers the partial cross section for all other states, by distributing the reaction probability among several symmetries.

References

[1] A. Dochain, Systematic study of mutual neutralization reactions between atomic species using the merged beam method and an asymptotic model, PhD. Thesis, Université catholique de Louvain, Louvain-la-Neuve, Belgium (2022)