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On mutual neutralization process in collisions of magnesium with hydrogen isotopes

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Abstract. Cross sections for mutual neutralization processes in magnesium-deuterium collisions are calculated in the collision energy range 0.001–100 eV by the probability current method. Special attention is given to the collision energies in the vicinity of E_{col} = 0.059 eV. Partial rate coefficients for mutual neutralization processes are calculated at the temperature T = 6000 K and compared with previous experimental and theoretical results. It is shown that the present data agree reasonably with the previous results.

Keywords: atomic data, atomic processes, charge transfer, magnesium, mutual neutralization

Introduction

Magnesium-hydrogen inelastic collisions have been intensively investigated theoretically (see Barklem 2016; 2017; Belyaev et al. 2012; Guitou et al. 2015; Rodionov, Belyaev 2017) but not experimentally. Recently, Grumer et. al. (Grumer et. al. 2022) have reported experimental measurements of the mutual neutralization (MN) cross sections in $Mg^+ + D^-$ collisions and theoretical estimates obtained by the Linear Combination of Atomic Orbitals (LCAO) method and the multichannel Landau-Zener (LZ) model approach. They claimed they got better agreement of their model estimates with the experimental data than the full quantum (FQ) theoretical data (Belyaev et al. 2012) for the final-state branching fractions (BF) and concluded that the model approach describes inelastic collision processes better than the FQ theory. For this reason in this paper we perform the new model calculations for $Mg^+ + H^-/D^-$ collisions, analyze MN BFs obtained by different theoretical means, and discuss some conclusions of (Grumer et al. 2022) with which we disagree.

Analysis of $Mg^+ + H^-/D^-$ branching fractions of mutual neutralization processes

We performed the nonadiabatic nuclear dynamical calculation for the mutual neutralization process in $Mg^+ + H^-/D^-$ collisions within the $^2\Sigma^+$ molecular symmetry based on the most recent *ab initio* data from (Guitou et al. 2015) by two model LZ approaches: the probability current method (PC) and the multichannel formula. BFs are investigated for the collision energy in the vicinity of E_{col} = 0.059 eV. Both methods treat 9 *ab initio* potential energy curves and one additional model potential, which corresponds to the Mg(3s4p ^{1}P) + H(1s ^{2}S) scattering channel, obtained by the asymptotic method (Belyaev 2013).

This state is added because both the simplified model (Belyaev, Yakovleva 2017a; 2017b) and the recent experiment (Grumer et al. 2022) indicate that the scattering channel Mg(3s4p ^{1}P) + H(1s ^{2}S) has non-negligible contribution into the total cross section and rate coefficient, so this state should be treated in nuclear dynamical calculations. The asymptotic energy for the Mg(3s4p ^{1}P) + H(1s ^{2}S) state is taken from NIST database (Kramida et al. 2022). In addition, the MgH fine structure is taken into account for multichannel calculations by means of the approach recently derived in (Belyaev et al. 2019; Yakovleva et al. 2019).

The present results are collected in Table 1 together with the data from (Guitou et al. 2015) and (Grumer et al. 2022). Fig. 1 shows the branching fractions for the mutual neutralization processes $Mg^+ + H^-/D^- \rightarrow Mg^*(3s\,nl) + H/D$ into the magnesium final states Mg(3s4s ^{3}S), Mg(3s4s ^{1}S), Mg(3s3d ^{1}D), and for a sum of the indistinguishable states (Mg(3s4p ^{3}P^o) + Mg(3s3d ^{3}D)).

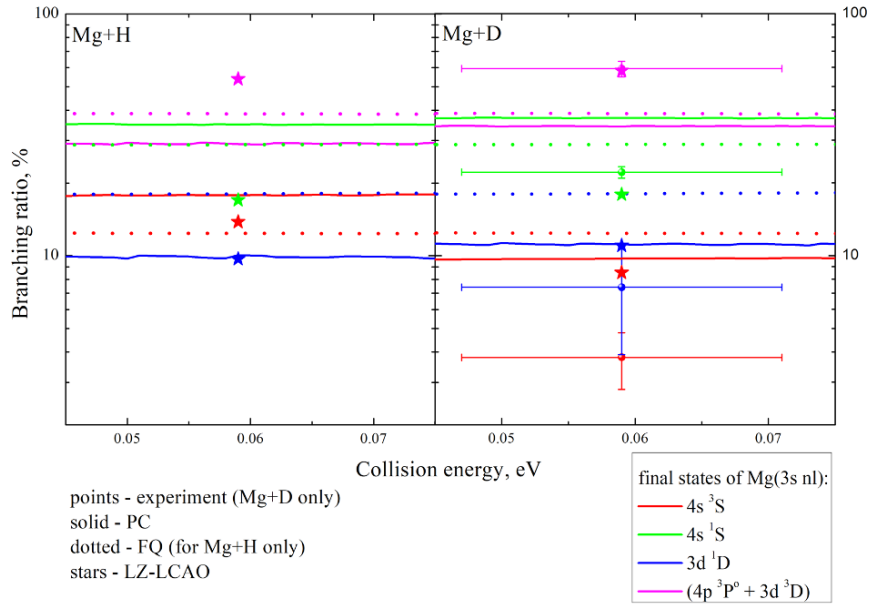

Fig. 1. Branching fractions (in per cents) of mutual neutralization processes $Mg^+ + H^-/D^- \rightarrow Mg^*(3s\,nl) + H/D$. Points with error bars correspond to the experiment (Grumer et al. 2022), stars—to the theoretical estimates of (Grumer et al. 2022), solid lines—to the Probability Current results obtained in the present work, dotted lines—to the Full Quantum results (Guitou et al. 2015)

Table 1. Mutual neutralization branching fractions (in per cent) calculated from the cross sections obtained by different means. (1) (Guitou et al. 2015), (2) (Grumer et al. 2022), (3) present calculations. Columns 2–5 correspond to $Mg^+ + H^-$ collisions, columns 6–9 correspond to $Mg^+ + D^-$ collisions

	FQ[1]	LZ-LCAO[2]	PC[3]	LZ-fine structure[3]	LZ-LCAO[2]	Experiment[2]	PC[3]	LZ-fine structure[3]
Mg final state	H^-	H^-	H^-	H^-	D^-	D^-	D^-	D^-
3s^2 ^{1}S	0.0	0.0	0.00	0.00	0.0	–	0.00	0.00
3s3p ^{3}P	0.0	0.0	0.00	0.00	0.0	–	0.00	0.00
3s3p ^{1}P	1.5	3.1	3.17	1.82	1.1	0.4 ± 0.1	1.33	0.41
3s4s ^{3}S	14.8	13.8	18.03	17.59	8.5	3.8 ± 1.0	9.86	8.91
3s4s ^{1}S	29.3	17.0	35.65	35.16	17.9	22.1 ± 1.2	37.30	36.56
3s3d ^{1}D	6.9	9.7	9.28	8.39	11.0	7.4 ± 3.5	10.82	10.01
3s4p ^{3}P	20.5	33.5	17.40	16.93	33.7	59.2 ± 4.4	20.41	19.87
3s3d ^{3}D	22.3	20.3	11.20	11.17	24.4		13.74	13.36
3s4p ^{1}P	4.4	2.5	5.27	8.93	3.4	7.1 ± 1.0	6.54	10.88
3s5s ^{3}S	0.2	0.0	–	–	0.0	–	–	–
3s5s ^{1}S	0.2	0.0	–	–	0.0	–	–	–

Let us consider magnesium-deuterium collisions. From the right panel of Fig. 1 it is seen that all considered methods provide results in very good agreement to each other for the magnesium final states Mg(3s4s ^{3}S) and Mg(3s3d ^{1}D) (red and blue lines and symbols), but for the states Mg(3s4s ^{1}S) and (Mg(3s4p ^{3}P^o) + Mg(3s3d ^{3}D)) there exists a difference between experimental and LZ-LCAO data as well as between the data obtained by the FQ and PC methods. It is clearly seen from Table 1 that the largest BFs correspond to the states Mg(3s4s ^{1}S), Mg(3s4p ^{3}P) and Mg(3s3d ^{3}D). The total BFs summed over these three states are 76% (LZ-LCAO), 81.3 ± 5.6% (experiment), 71.45% (PC) and 69.79% (LZ-fine structure). The LZ-LCAO results agree the best with the experiment, although the results of PC and LZ-fine-structure calculations are also in good agreement with the experiment. It also should be noted that the general behavior of all BFs is in good agreement with each other for all sets of data, and the experimentally observed final-state distribution is correctly reproduced by the different theoretical approaches.

Let us consider magnesium-hydrogen collisions. For this system, to the best of our knowledge, there are no experimental data, but FQ data are available. There are four final states with the largest BFs values: Mg(3s4s ^{3}S), Mg(3s4s ^{1}S), Mg(3s4p ^{3}P) and Mg(3s3d ^{3}D), and the total BFs for these four states are: 86.9% (FQ), 84.6% (LZ-LCAO), 82.28% (PC) and 80.85% (LZ-fine structure). These results are in good agreement with each other as well, and again the general behavior of all BFs is in reasonable agreement with the FQ one, see the left panel of Fig. 1. FQ and PC methods predict that MN BF into the final state Mg(3s4s ^{1}S) is larger than the LZ-LCAO estimates by a factor of 2, but BF into the states (Mg(3s4p ^{3}P^o) + Mg(3s3d ^{3}D)) is lower than the LZ-LCAO estimates also by a factor of 2. It is worth noting that all data-sets are in reasonable agreement with each other.

Since the only difference between the $Mg^+ + H^-$ and $Mg^+ + D^-$ collisions is in masses of a hydrogen isotope, it is expected that in both cases BF distributions are similar; this has been confirmed theoretically and experimentally for LiH (Belyaev, Voronov 2021; Launoy et al. 2019; Schmidt-May et al. 2022). Theoretical calculations for the $Mg^+ + H^-$ and $Mg^+ + D^-$ collisions confirm this conclusion, see Table 1.

Analysis of $Mg^+ + H^-$ rate coefficients of mutual neutralization processes

Let us now discuss the rate coefficients obtained by different theoretical methods. Table 2 contains the rate coefficients for Mg+H collisions obtained by FQ calculations, LZ-LCAO calculations from (Barklem 2017; 2022), LZ-LCAO calculations from (Grumer et al. 2022), PC and LZ-fine structure (the present study).

Table 2. Mutual neutralization rate coefficients of $Mg^+ + H^-$ collisions at the temperature T = 6000 K calculated from the cross sections obtained by different means. (1) (Guitou et al. 2015), (2) Barklem 2022, (3) (Grumer et al. 2022), (4) present calculations. Square brackets denote the power of ten.

Mg final state	FQ[1]	LZ-LCAO[2] (.rates)	LZ-LCAO[2] (.min)	LZ-LCAO[2] (.max)	LZ-LCAO[3]	PC[4]	LZ-fine structure[4]
3s^2 ^{1}S	1.44[–12]	0	0	1.48[–14]	–	1.49[–18]	5.49[–19]
3s3p ^{3}P	1.01[–9]	5.49[–12]	6.74[–22]	1.51[–11]	–	4.11[–15]	4.82[–13]
3s3p ^{1}P	4.00[–9]	5.14[–9]	1.13[–12]	5.14[–9]	–	4.62[–9]	2.95[–9]
3s4s ^{3}S	2.00[–8]	2.23[–8]	1.27[–10]	2.23[–8]	–	2.60[–8]	2.39[–8]
3s4s ^{1}S	4.18[–8]	2.64[–8]	2.64[–8]	3.99[–8]	–	3.93[–8]	3.87[–8]
3s3d ^{1}D	2.58[–8]	1.57[–8]	9.46[–9]	5.12[–8]	–	1.20[–8]	9.40[–9]
3s4p ^{3}P	2.01[–8]	5.45[–8]	2.65[–8]	6.94[–8]	–	1.95[–8]	1.92[–8]
3s3d ^{3}D	3.16[–8]	3.36[–8]	1.36[–8]	3.36[–8]	–	1.50[–8]	1.25[–8]
3s4p ^{1}P	5.62[–9]	3.94[–9]	3.94[–9]	1.04[–8]	–	6.27[–9]	5.92[–9]
3s5s ^{3}S	3.06[–10]	8.03[–12]	4.95[–12]	1.95[–11]	–	–	–
3s5s ^{1}S	3.23[–10]	3.38[–14]	3.38[–14]	2.61[–13]	–	–	–
total	1.5[–7]	1.6[–7]	0.8[–7]	2.3[–7]	1.6[–7]	1.2[–7]	1.1[–7]

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It should be noted that in the LZ-LCAO calculations the "fluctuations" of rate coefficients can be significant, for example, the scatter of the total rate coefficient is equal to (0.8-2.3) x 10^{-7} cm^3/s. For partial processes these "fluctuations" may exceed several orders of magnitude.

Let us analyze the data from the columns 2, 3, 6–8 in detail. One can see from Table 2 that the total MN rate coefficient obtained by different methods has values in the range (1.1-1.6) x 10^{-7} cm^3/s and agrees within the uncertainty ≈30% with the FQ data (1.5 x 10^{-7} cm^3/s).

The MN processes into the final magnesium states Mg(3s4s ^{3}S), Mg(3s4s ^{1}S), Mg(3s3d ^{1}D), Mg(3s4p ^{3}P), Mg(3s3d ^{3}D), Mg(3s4p ^{1}P) are of particular interest because these processes belong to the so-called "optimal window" according to the simplified model (Belyaev, Yakovleva 2017a; 2017b). The optimal window for Mg is located at the excitation energy range of 5.1–6.1 eV. Table 2 shows that the main deviations of our present results from those of the FQ and LZ-LCAO results are for the processes $Mg^+ + H^- \rightarrow$ Mg(3s4s ^{1}S) + H(1s ^{2}S), Mg(3s4p ^{3}P) + H(1s ^{2}S), Mg(3s3d ^{3}D) + H(1s ^{2}S). These differences do not exceed the factor of 3, which is less than almost all "fluctuation" ranges for these processes obtained by LZ-LCAO method (columns 4–5). In general, we conclude that the MN rate coefficients with the values exceeding 10^{-8} cm^3/s and obtained by different theoretical methods agree reasonably well with each other.

Discussion

Let us discuss some conclusions from (Grumer et al. 2022). First of all, it should be noted that Grumer et.al. (Grumer et al. 2022) compared their results with FQ results from (Belyaev et al. 2012), which is not the most recent work and does not include the most complete FQ data. The most recent MgH treatment was reported in the (Guitou et al. 2015) paper for 13 MgH states, while in the (Belyaev et al. 2012) paper only eight states of the MgH molecule were considered: seven covalent and one ionic states. The comparison of the truncated data from the (Belyaev et al. 2012) paper instead of the complete data (Guitou et al. 2015) with the experimental and theoretical data (Grumer et al. 2022) misleaded Grumer et al. to the wrong conclusions. Namely:

1) (Grumer et al. 2022) wrote, "The FQ for Mg+H calculation, with Mg(4s ^{1}S) as the predicted dominating channel with a branching fraction of 61.9%, reproduces the experimental data poorly. The current state-of-the-art theoretical data, based on detailed quantum-chemistry calculations, do not correctly reproduce the observed final-state distributions."

Grumer et al. (Grumer et al. 2022) made such conclusion because they compared their theoretical and experimental data for 11 scattering final channels in Mg+H and Mg+D collisions with the data calculated for seven final scattering channels (Belyaev et al. 2012) in Mg+H collisions. In this case, one or two of the most populated final channels were not included into the consideration. Grumer et al. (Grumer et al. 2022) obtained the BF of 61.9% for the Mg(3s4s ^{1}S) + H final state by taking into calculation only the truncated data set of seven final covalent channels from (Belyaev et al. 2012) while for their calculations of the BFs by the LZ-LCAO method Grumer et. al. (Grumer et al. 2022) took into account 11 final covalent channels. This leads to the BFs of 17.0% (Mg+H) and 17.9% (Mg+D) as well as to the experimental value of 22.1%±1.2%. Obviously, the BF distribution depends on a number of scattering channels taken into calculations, and the most accurate state-of-the-art theoretical FQ data were presented in (Guitou et al. 2015) for 13 molecular states. It is shown in the previous section that FQ as well as PC and LZ-fine structure methods based on the *ab initio* potentials provide the results that are in reasonable agreement with the experimental data: in Mg+H collisions BFs for magnesium state Mg(3s4s ^{1}S) is 29.3% for FQ, 35.65% for PC and 35.16% for LZ-fine structure method. Thus, the conclusion (1) is incorrect. The correct calculations of the complete FQ data lead to a good agreement with the experimental data.

2) Grumer et al. wrote, "Asymptotic model calculations are shown to describe the process much better (than FQ)" (Grumer et al. 2022).

The LCAO method should be compared with the *ab initio* Multi-Reference Configuration Interaction (MRCI) method used in (Guitou et al. 2015). Calculations performed by Guitou et al. (Guitou et al. 2015) were based on 57 basis orbitals for Mg plus 23 basis orbitals for H plus 5 diffuse orbitals, that is, 85 basis wavefunctions as a starting point in total. The LCAO method used a single undisturbed orbital for H^- and a few orbitals for covalent states. Obviously, the MRCI calculations are more accurate than those by the LCAO used in (Grumer et al. 2022). So, the conclusion (2) is also wrong.

3) Grumer et al. wrote, "A simpler asymptotic-model approach based on multichannel Landau-Zener dynamics combined with a LCAO approach for the coupling strengths describes the MN process and the resulting excitations of the neutral products much better (than FQ)" (Grumer et al. 2022).

There are several arguments to contradict this statement:

a) when the electronic structure, potentials and couplings are known with high accuracy over the whole range of internuclear configurations, the accurate non-adiabatic nuclear dynamics can be calculated by solving the Coupled Channel Equations (CCEs) taken into account not only long-range but also intermediate- and short-range non-adiabatic regions as well as all non-adiabatic couplings. This is done in (Guitou et al. 2015) and also in the present investigation by the PC method within the LZ model. Alternatively, the multichannel analytical formula can be used for estimate state-to-state transition probabilities, when non-adiabatic regions are localized in a particular sequence, usually for long- and intermediate-range regions; short-range regions are usually not taken into account in this kind of estimates. Grumer et al. (Grumer et al. 2022) used the multichannel formula. Obviously, solutions of the CCEs have higher accuracy than the multichannel formula, though the latter has some advantages.

b) In the collision theory there is the fundamental problem, the so-called Electron Translation (ET) problem (sometimes called the Molecular-state problem), related to non-zero asymptotic non-adiabatic radial couplings. It is shown in (Belyaev 2010) that this might lead to divergence of a partial cross section. Guitou et al. (Guitou et al. 2015) solved the ET problem by using the reprojection method (Belyaev 2010; Grosser et al. 1999), while Grumer et al. (Grumer et al. 2022) did not take this problem into account. Thus, FQ (full quantum) non-adiabatic nuclear dynamical treatment is more complete and more accurate than the multichannel approach, and the conclusion (3) of the paper (Grumer et al. 2022) is also not correct.

Finally, we come to the general conclusion that the FQ study of inelastic collision processes, in particular, the FQ of MN in $Mg^+ + H^-/D^-$ collisions, is more accurate than the model estimates of (Grumer et al. 2022). However, we wish to point out that FQ calculations are time-consuming and expensive, and hence model approaches are useful and efficient.

In the case of magnesium-hydrogen collisions, we believe that new accurate quantum-chemical calculations of the adiabatic potentials and couplings are desired, in particular, taking into account more excited MgH molecular states, at least $Mg(3s4p\ ^1P^o) + H(1s\ ^2S)$.

Conclusions

In this work, we investigated the mutual neutralization processes in collisions of $Mg^+ + H^-/D^-$ at the vicinity of the collision energy E_{col} = 0.059 eV by the probability current method and by the multichannel formula (with accounting for fine structure). It is shown that the results based on the ab initio potential energy curves from (Guitou et al. 2015) have reasonable agreement with the full quantum nuclear dynamical calculations (Guitou et al. 2015), with the LCAO-multichannel-LZ estimates (Grumer et al. 2022), and with the experimental data (Grumer et al. 2022). We emphasize that new quantum-chemical calculations of the adiabatic potentials and couplings taking into account higher-lying excited MgH molecular states, at least $Mg(3s4p\ ^1P^o) + H(1s\ ^2S)$, are highly desired.

Conflict of Interest

The authors declare that there is no conflict of interest, either existing or potential.

Author Contributions

The authors have made an equal contribution to the paper.

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