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New Version Announcement

MQCT 2024: A program for calculations of inelastic scattering of two molecules (new version announcement) ☆

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ABSTRACT

This is a revised and updated version of the package MQCT. The package includes routines for the calculations of ro-vibrational state-to-state transition cross sections for molecule + molecule collisions, using the mixed quantum/classical theory approach. One important addition to the package is an efficient and accurate adiabatic-trajectory approximation method (AT-MQCT), which enables calculations for larger and more complex molecules. An efficient parallel IO is implemented for state-to-state transition matrix, which now permits to handle very large matrixes, but also helps to truncate these matrixes by neglecting insignificant matrix elements and to balance the load on different CPUs during the matrix calculations and during the trajectory propagation. A stable recursive approach is added for computation of Wigner 3j-symbols, which expands the range of rotational states of molecules up to $j \sim 100$. Examples of input files are provided for all ten system types covered by the package, along with updated user manual.

New version program summary

Program title: MQCT 2024
CPC Library link to program files: https://doi.org/10.17632/sg36r35njz.2
Developer's repository link: https://github.com/MarquetteQuantum/MQCT
Code Ocean capsule: https://codeocean.com/capsule/1260072/tree/v1
Licensing provisions: GNU GPL v3.0
Programming language: FORTRAN
Journal reference of previous version: Comput. Phys. Commun. 252 (2020) 107155
Does the new version supersede the previous version?: Yes
Reasons for the new version:

1) Recently, we developed and tested the adiabatic-trajectory method [1–3] within MQCT framework, which appears to be both efficient and accurate, and is likely to become a "workhorse" for inelastic scattering calculations in molecule + molecule systems. In the new version of the package this method is made available as an option, AT-MQCT.
2) Since MQCT trajectory calculations became more affordable with AT-MQCT, we started doing calculations with larger and larger rotational basis sets and quickly encountered several new problems. The major one was an extremely slow serial IO of the state-to-state transition matrix to a single file, which is now replaced by a parallel IO to multiple files written to a separate directory. Very large matrixes (up to 10^{10} non-zero matrix elements) are now handled efficiently.
3) Another problem was an incorrect calculation of Wigner 3j-symbols for the rotational states around $j \sim 40$ and above. In the revised version a new stable subroutine is employed that uses a recursive method and works properly at least up to $j = 100$.
4) Several "bugs" were caught and fixed, with one major in the Monte-Carlo sampling of MQCT trajectories. Now users can add a batch of new trajectories to the previously sampled batch. In addition, users can monitor the convergence of Monte-Carlo sampling in detail. These options work for all levels of theory, including AT-MQCT. Several errors were found in the equations for calculations of state-to-state transition matrix using the expansion of potential energy surface, and in symmetrization of matrix elements for the case of identical collision partners. All these have been corrected.

☆ The review of this paper was arranged by Prof. J. Ballantyne.

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Summary of revisions: Several technical revisions concern the treatment of diatomic molecules in the code: A bug was found and fixed in the PES interface that converts the units of vibrational coordinate (the diatomic bond distance). Now the PES can be expressed in either Angstrom or Bohr, as specified in the input file. Integration of matrix elements over the vibrational coordinate is modified to use an equidistant grid by default (Gauss-Legendre method is still available as option). And the same for integration over Euler angles α and γ (for all system types). Integration over β uses Gauss-Legendre quadrature only. The format of ro-vibrational wavefunctions pre-computed externally and supplied as input is made more intuitive, as described in the manual. Finally, a new method for the sampling of orbital quantum number ℓ is implemented, which skips some values of ℓ and obtains results by interpolation of the data at the retained values of ℓ (for all system types, as described in the manual). Last version of the code is always available at https://github.com/MarquetteQuantum/MQCT.
Supplementary material: Updated and expanded MQCT user guide
Nature of problem: Calculations of rotationally and vibrationally inelastic scattering of two molecules, with possible applications in astrophysics and atmospheric chemistry
Solution method: Mixed quantum/classical theory (MQCT) approach

References

[1] B. Mandal, A. Semenov and D. Babikov, J. Phys. Chem. A 124 (2020) 9877-9888
[2] B. Mandal, C. Joy, A. Semenov and D. Babikov, ACS Earth Space Chem. 6 (2022) 521-529
[3] B. Mandal, C. Joy, D. Bostan, A. Eng and D. Babikov, J. Phys. Chem. Lett. 14 (2023) 817–824.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Dmitri Babikov reports financial support was provided by National Science Foundation. Dmitri Babikov reports financial support was provided by NASA. Dmitri Babikov reports equipment, drugs, or supplies was provided by National Energy Research Scientific Computing Center. Dmitri Babikov reports a relationship with National Science Foundation that includes: funding grants.

Data availability

No data was used for the research described in the article.

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Appendix A. Supplementary material

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.cpc.2023.108938.