Basis Generator Method Calculations for Charge-Transfer Collisions Involving Few-Electron Systems

Tom Kirchner

York Université University
redefine THE POSSIBLE.

Few-electron dynamics

- Challenging because of nonseparability
- Interesting because of nonseparability
- Relevant for applications, but uncertainty estimates are difficult

Today's presentation

- The basis generator method (BGM) for multielectron collision problems
- A few illustrations for (low-energy) charge-transfer collisions

Theoretical Considerations

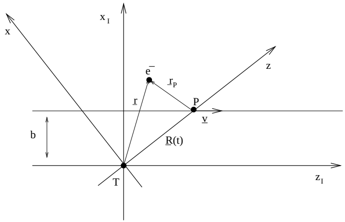
Semiclassical approximation

Justification: de Broglie wavelength of projectile
$\lambda_K = h/p = h/(\mu v) \ll a_0$ at $E_P \geq 1$ keV/amu

$\rightarrow$ (curved or) straightline projectile trajectory

$$\boldsymbol{R}(t) = (b, 0, vt)$$

b: impact parameter
v: projectile speed



electrons described fully quantum mechanically

Semiclassical approximation

Nonrelativistic theory: time-dependent Schrödinger equation (TDSE) $(\hbar = m_e = e = 1)$

$$\left(\hat{H}_e(t) - i\partial_t\right)|\psi(t)\rangle = 0, \quad |\psi(t_i)\rangle = |\phi_i\rangle$$

$$\hat{H}_e(t) = \hat{T}_e + \hat{V}_{e-n}(t) + \hat{V}_{e-e} + \frac{Z_p Z_t}{R(t)}$$

- $\hat{T}_e$: kinetic energies of electrons
- $\hat{V}_{e-n}(t)$: time-dependent electron-nucleus Coulomb interaction
- $\hat{V}_{e-e}$: electron-electron Coulomb interaction

Discussion I

- Explicit solution feasible for $N = 2$
  $\rightarrow$ bare ion – He (H_2) collisions
  - Molecular close coupling
  - Atomic close coupling
  - Time-dependent lattice (TDL) methods
  - Classical-trajectory Monte Carlo (CTMC)
  - ...
- $N \geq 3$: (mostly) beyond present capabilities

Discussion II

Approx. alternatives for $N \geq 2$ target electrons
$\longrightarrow$ model uncertainties

- Ansatz (independent electrons – IEM):

$$\hat{H}_e(t) \rightarrow \sum_{j=1}^{N} \hat{h}_j(t), \quad i\partial_t \psi_j(\boldsymbol{r},t) = \hat{h}(t)\psi_j(\boldsymbol{r},t)$$

$$\hat{h}(t) = -\frac{1}{2}\Delta - \frac{Z_T}{r} - \frac{Z_p}{|\boldsymbol{r} - \boldsymbol{R}(t)|} + v_{ee}(\boldsymbol{r},t)$$

- Choice of v_{ee} defines model
- Time-dependent density functional theory (TDDFT) provides foundation

Discussion III

Choices:

$$
\begin{aligned}
v_{ee}(\boldsymbol{r}, t) &= v_{ee}^0(r) && \text{no response} \\
v_{ee}(\boldsymbol{r}, t) &= f(t) v_{ee}^0(r) && \text{global target response}^* \\
v_{ee}(\boldsymbol{r}, t) &= v_{ee}[\psi_j](\boldsymbol{r}, t) && \text{microscopic response}
\end{aligned}
$$

Standard methods can be used within IEM

Basis Generator Method (BGM)**:

a close-coupling method based on atomic orbitals and dynamically adapted pseudo states

* Kirchner *et al.*, PRA 2000

**Lüdde et al. JPB 1996, Kroneisen et al. JPA 1999

Discussion IV

Single-particle solutions $\rightarrow$ many-electron info

- Option 1: IEM (multinomial) analysis
  e.g. single and double capture for $N = 2$:

$$P_1 = 2p_{\mathrm{cap}}(1 - p_{\mathrm{cap}})$$
$$P_2 = p_{\mathrm{cap}}^2$$

- Option 2: determinants (density matrices)*
- Further options: correlation integrals**, single-active electron model, ...

$\longrightarrow$ source of model uncertainties

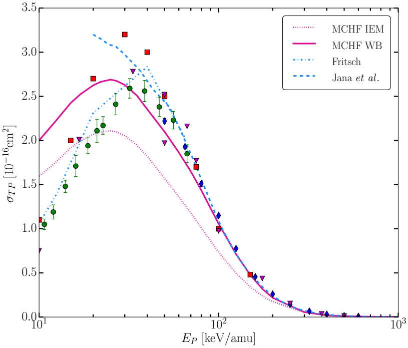
*Lüdde and Dreizler JPB 1985

** Baxter and Kirchner PRA 2016

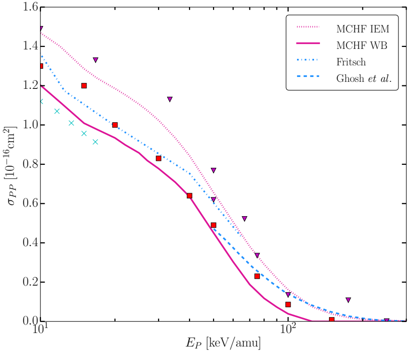
Illustrations

He^{2+}- He: single and double capture

single capture



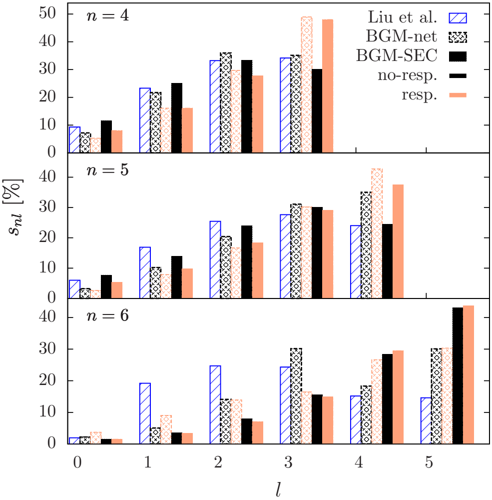
double capture



——: two-electron ——: DFT-based

Baxter and Kirchner, PRA **93**, 012502 (2016)

Ne^{10+}- Ne: single capture



$$s_{nl} = \sigma_{nl} / \sum_{nl} \sigma_{nl}$$

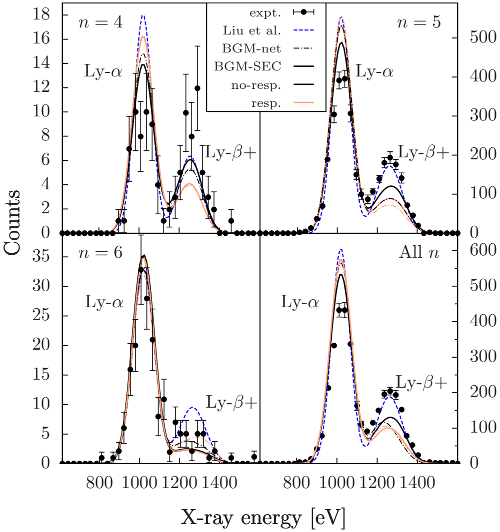
IEM level

Liu et al., PRA **89**, 012710

BGM: PRA **92**, 032712

$E_P = 4.54$ keV/amu

Ne^{10+}- Ne: x-ray spectra



Expt.: Ali et al., Astrophys. J. Lett. **716**, L95

Liu et al., PRA **89**, 012710

BGM: PRA **92**, 032712

$E_P = 4.54$ keV/amu

Molecules

- More difficult than atoms because of multi-center structure
- Electron dynamics can be described in similar way if “small“ (e.g. H_2O, CH_4)*
- Relevant for cometary x-ray emission

* Kirchner *et al*, Adv. Quant. Chem. **65**, 315 (2013)

O^{6+}- H_2O, CH_4: S,D,T capture

O^{6+}-H_2O

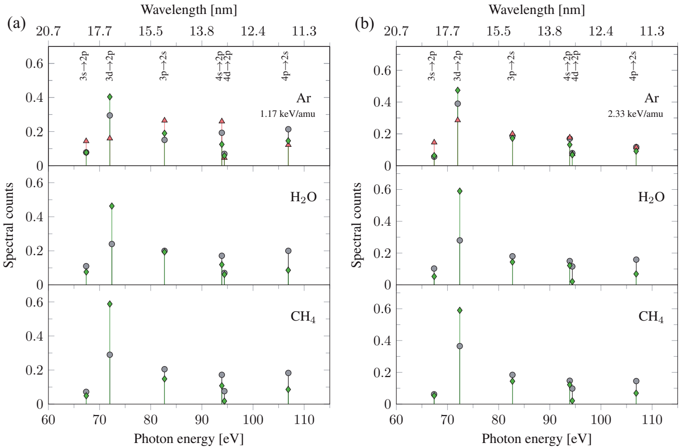
E_P (keV/amu)		BGM	Expt. [15]	CTMC [15]
1.17	SEC	59.7	47.3 ± 3.2	55.0
	DEC	15.5	8.3 ± 0.6	6.30
	TEC	4.57	3.7 ± 0.3	0.580
2.33	SEC	58.1	45.9 ± 3.1	57.4
	DEC	12.9	7.4 ± 0.5	6.53
	TEC	3.18	2.6 ± 0.2	0.586

O^{6+}-CH_4

E_P (keV/amu)		BGM	Expt. [15]	CTMC [15]
1.17	SEC	52.7	42.9 ± 2.9	54.2
	DEC	26.6	17.8 ± 1.3	6.76
	TEC	4.94	2.7 ± 0.2	0.659
2.33	SEC	50.9	50.2 ± 3.4	56.7
	DEC	23.2	16.3 ± 1.2	6.94
	TEC	4.30	2.3 ± 0.2	0.634

Expt & CTMC: Machacek et al., Astrophys. J. **809**, 75 (2015)

O^{6+}- H_2O, CH_4: x-ray spectra (SC)



CTMC (◇): Machacek et al., Astrophys. J. **809**, 75 (2015)
BGM no response (○); BGM response (△)

Conclusions: multielectron systems

Model uncertainties (IEM):

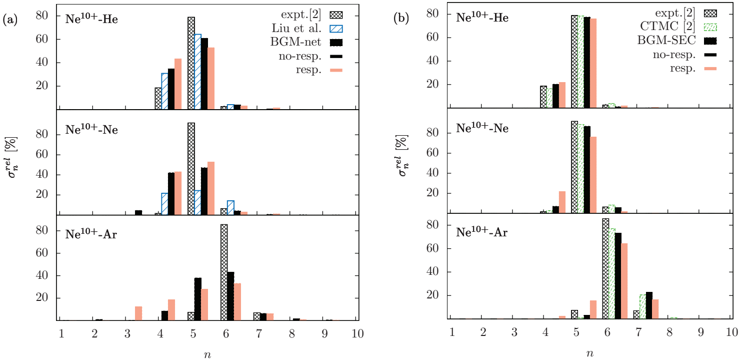
- Effective potential
- Multielectron analysis
- Subsequent processes (autoionization, radiative cascades, ...)

Uncertainty estimates currently based on:

- Comparisons of different model (variants)
- Comparisons with experimental data

Need benchmark data for benchmark systems!

Ne^{10+}- He,Ne,Ar: single *n*-capture



$E_P = 4.54$ keV/amu

Liu et al., PRA **89**, 012710
BGM: PRA **92**, 032712
CTMC and Expt: Ali et al., Astrophys. J. Lett. **716**, L95

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