Investigation of laser-driven non-equilibrium electron generation using a fast and efficient method

Henry T. Snowden[1], Markus Uehlein[3], Reinhard J. Maurer[1,2], Baerbel Rethfeld[3]

1. Department of Chemistry, University of Warwick, Coventry, UK
2. Department of Physics, University of Warwick, Coventry, UK
3. Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau

henry.snowden@warwick.ac.uk

Motivation & Introduction

Many advancing technologies rely on light-matter interactions, e.g. photocatalysis and ultrafast spectroscopy. To better understand these processes we need to be able to simulate them. The state-of-the-art for this is the Boltzmann Transport Equation (BTE).[1] This is limited primarily by it's large computational cost.

Here a new method, the athermal electron model (AthEM) is presented.[2] This model approximates the BTE without the large cost. This opens the door to fast material testing and the inclusion of higher-dimension effects such as transport or surface densities of states (DOS). The main focus will be on the relaxation to the thermal baths as well as the effect of different DOS.

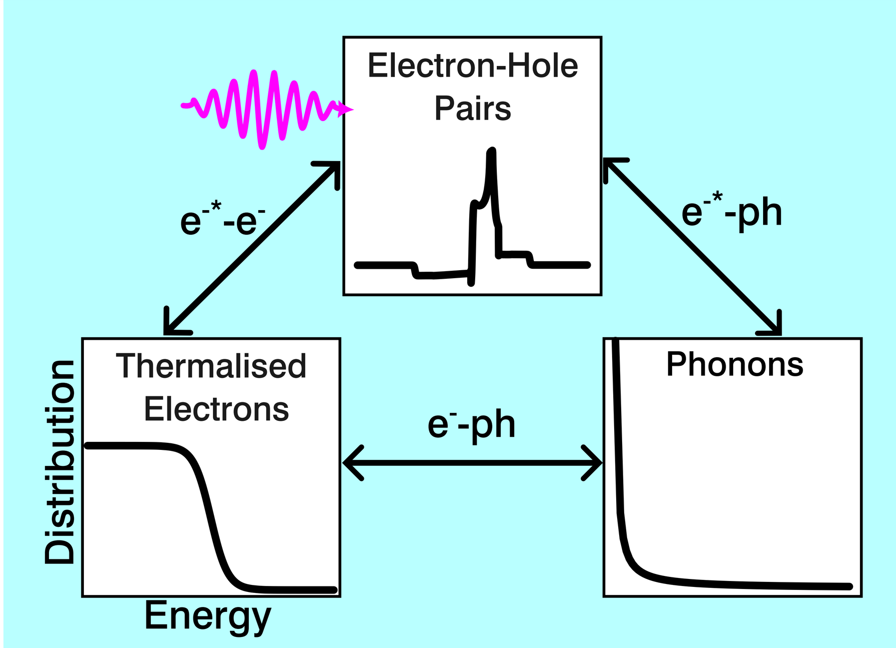
The Equations

$$\frac{\partial f_{el^*}}{\partial t} = \left.\frac{\partial f_{el^*}}{\partial t}\right|_{laser} - \left.\frac{\partial f_{el^*}}{\partial t}\right|_{el^*\text{-}el} - \left.\frac{\partial f_{el^*}}{\partial t}\right|_{el^*\text{-}ph}$$

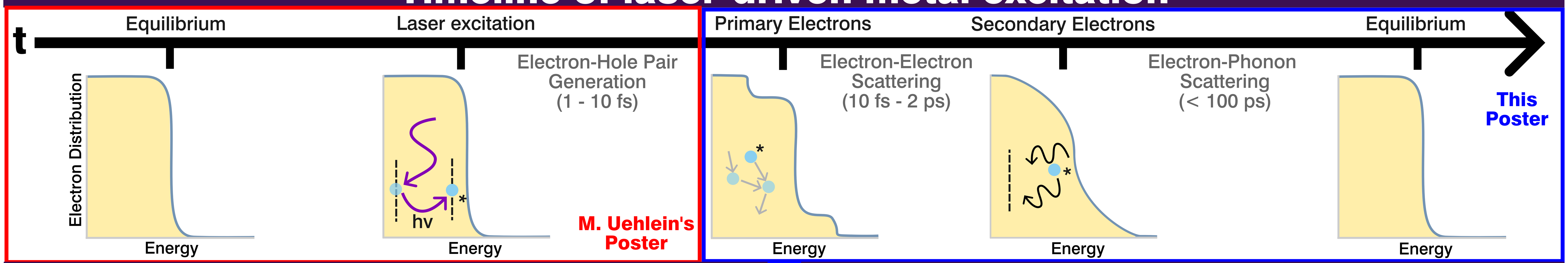
$$\frac{du_{el}}{dt} = -g(T_{el} - T_{ph}) + \left.\frac{\partial u_{el}}{\partial t}\right|_{el^*\text{-}el}$$

$$\frac{du_{ph}}{dt} = g(T_{el} - T_{ph}) + \left.\frac{\partial u_{ph}}{\partial t}\right|_{el^*\text{-}ph}$$

$$\frac{dn_{el}}{dt} = \left.\frac{\partial n_{el}}{\partial t}\right|_{el^*\text{-}el}$$



Timeline of laser-driven metal excitation

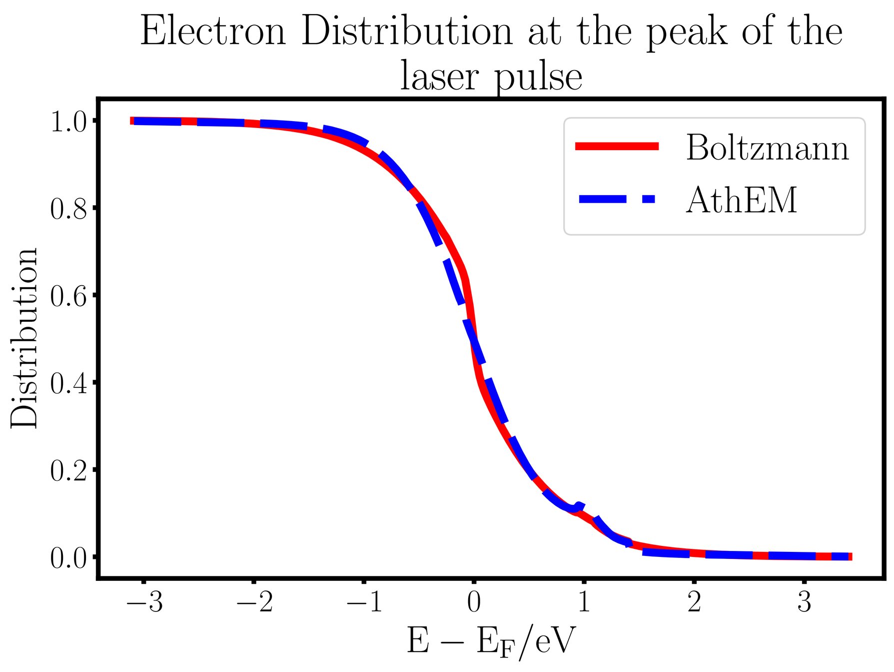


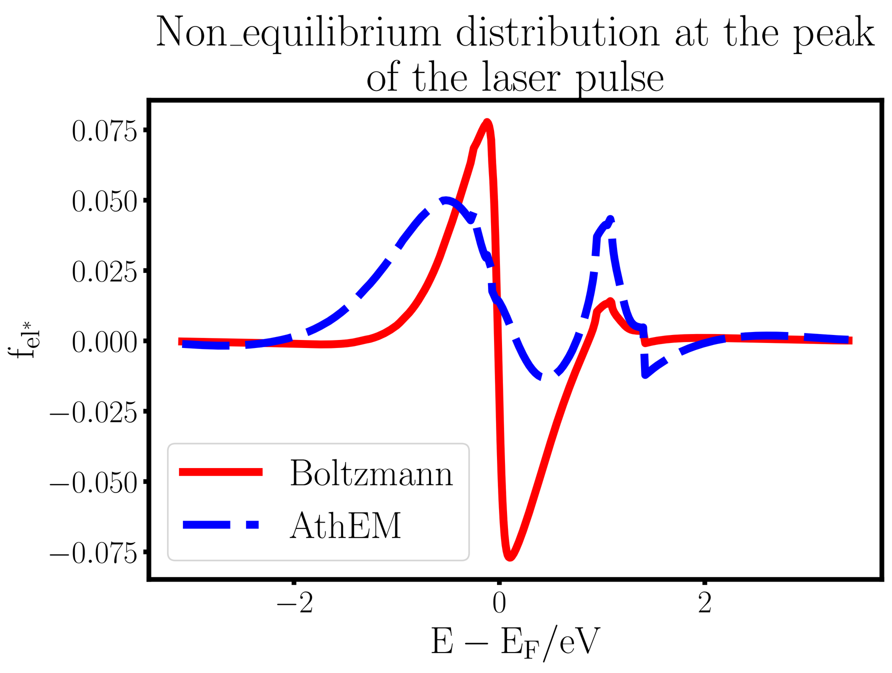
1. Electron-Electron Scattering

The relaxation to thermal baths is performed via the relaxation time approximation with a Fermi-liquid theory relaxation time. This captures the relaxation qualitatively well and allows us to investigate long-lived non-equilibrium electrons.

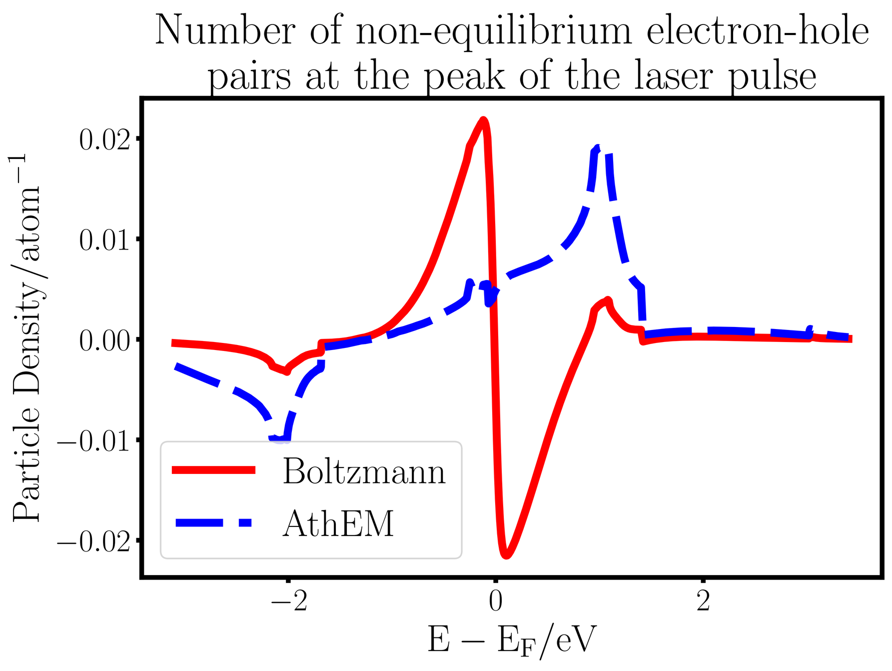
$$\left.\frac{\partial f_{el^*}(E)}{\partial t}\right|_{el^*\text{-}el} = -\frac{f_{el^*}(E) + (f^{rel}(E) - f_{el}(E))}{\tau_{el^*\text{-}el}(E)}$$

$$\tau_{el^*\text{-}el}(E) = \tau_0 \frac{\mu^2}{(E-\mu)^2 + (\pi k_B T_{el})^2} \qquad \tau_0 = \frac{128}{\sqrt{3}\pi^2 \omega_p}$$





Coupling to molecular dynamics simulations uses the electron-hole pair density rather than the distribution.

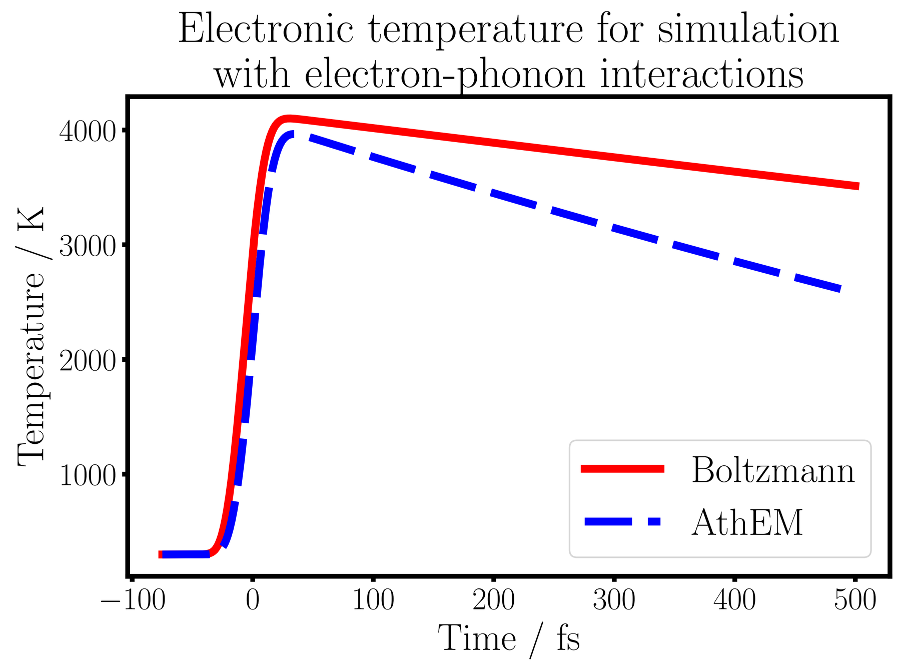


2. Electron-Phonon Scattering

Electron-Phonon relaxation provides the timescale of the cooling of the hot-electron distribution and therefore the timescale that high-energy electrons can be utilised.

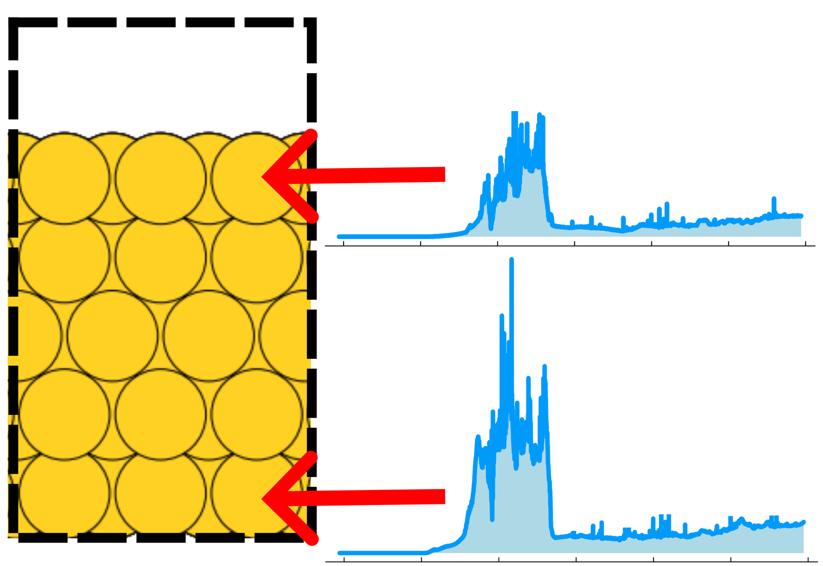
$$\left.\frac{\partial f_{el^*}}{\partial t}\right|_{el^*\text{-}ph} = -\frac{f_{el^*}}{\tau_{el^*\text{-}ph}} \qquad el \leftrightarrow ph = g(T_{el} - T_{ph})$$

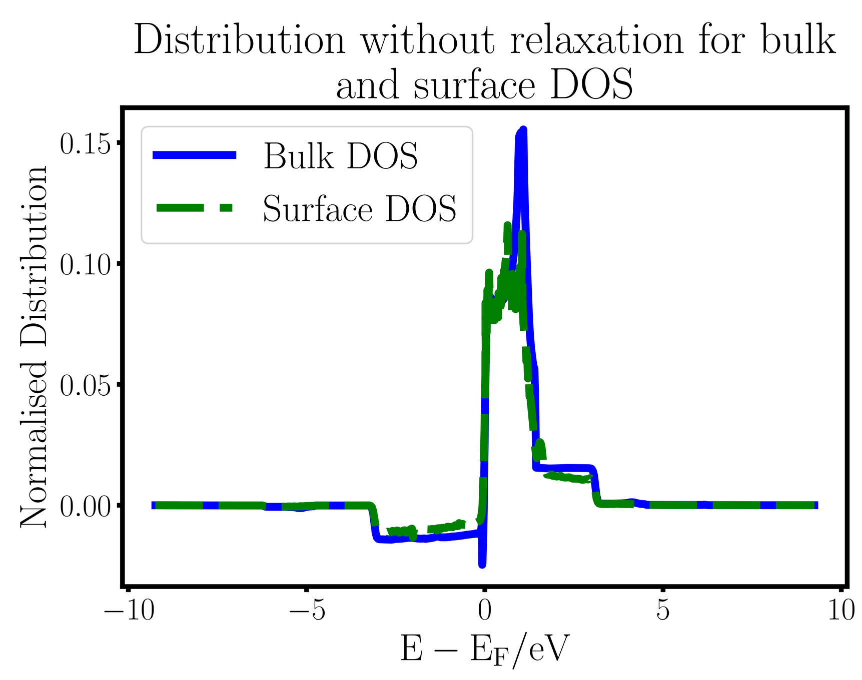
$$\tau_{el^*\text{-}ph} = \tau_{mfp}\frac{\hbar\omega}{k_B\theta_D} \qquad g = \frac{\pi k_B \lambda\langle\omega^2\rangle}{\hbar D(E_F)}\int_{-\infty}^{\infty} D^2(E)\left(-\frac{\partial f}{\partial E}\right)dE$$



3. Surface DOS

We can utilise the speed and flexibility of the code to model a slab with a height-dependent DOS.





Conclusion

Here we present AthEM and show how it compares to the state-of-the-art BTE for both electron-electron and electron-phonon scattering. The result is qualitative agreement with improvements to be made in the electron-electron lifetime. We also present how the model can go beyond the BTE to perform the investigation in higher dimensions using variable DOS'.

References & Acknowledgements

1. B. Y. Mueller and B. Rethfeld, Phys. Rev. B 87, 035139 (2013)
2. M. Uehlein, H.Snowden et al., in preparation

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