

Quantum-kinetics of charge-transferring atom-surface collisions

Franz Xaver Bronold, Johannes Marbach, and Holger Fehske

Ernst-Moritz-Arndt-University Greifswald, Felix-Hausdorff-Str. 6, 17489 Greifswald, Germany

Charge-exchange between an atomic projectile and a surface plays a central role in applied surface science. Many advanced surface diagnostics utilize surface-based charge-transfer processes. This type of collision is however also of fundamental interest since it couples a system with a finite number of discrete states—the atomic projectile—to a large electron reservoir with a continuum of states—the target surface. Irrespective of the coupling between the two, either due to tunneling or due to Auger-type Coulomb interaction, charge-transferring atom-surface collisions are thus perfect realizations of time-dependent quantum impurity systems. By a judicious choice of the projectile-target combination as well as the collision parameters Kondo-type features due to Coulomb-blockades can be realized as in any other quantum impurity system, with the advantage, however, of being able to control the time scale on which the impurity couples to the reservoir. We will be concerned with the theoretical description of this type of collisional system beyond the saddle-point (semiclassical) approximation. Using an Anderson-Newns-type model in the pseudo-particle representation [1] which allows us to treat strong intra-projectile Coulomb interactions we will show how the time development of the occupancies of the projectile states can be obtained from Keldysh's integral equations for contour-ordered Green functions. Our approach utilizes an exponential resummation to iteratively solve for the double-time retarded Green function [2]. Inserting cumulant-type expressions for the retarded (advanced) Green function into the integral equation for the double-time less-than (larger-than) Green function provides a framework for calculating corrections to the saddle-point approximation, which is usually the method of choice for reducing the double-time quantum-kinetics of Green functions to the single-time quantum-kinetics of occupancies. The saddle-point approximation misses however important non-adiabatic effects, in particular those which drive the Kondo-type features of the collision process. Having a method at hand for going beyond the saddle-point approximation without the necessity of solving the full two-time quantum-kinetic equations [3] is thus of great practical value. We present results for the infinite-U Anderson-Newns model describing resonant charge-transfer but the approach is not restricted to this particular case.

[1] J. Marbach, F.X. Bronold, and H. Fehske, Phys. Rev. B **86**, 115417 (2012)

[2] J. Marbach, F.X. Bronold, and H. Fehske, Phys. Rev. B **84**, 085443 (2011)

[3] H. Shao, D. C. Langreth, and P. Nordlander, Phys. Rev. B **49**, 13929 (1994)