Open quantum system approach in multiple atomic collisions in solids and gases

C.O. Reinhold1*, M. Seliger2, T. Minami3, S. Yoshida4, J. Burgdörfer4, J. J. Mestayer5, W Zhao5, J C Lancaster5 and F.B. Dunning5
1 Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA
2 Institute for Physics, Karl-Franzens-University Graz, A-8010 Graz, Austria, EU
3 Department of Physics, Auburn University, Auburn, Alabama 36849, USA
4 Institute for Theoretical Physics, Vienna University of Technology, A1040 Vienna, Austria, EU
5 Department of Physics and Astronomy, Rice University, Houston, Texas 77005, USA

E-mail: reinhold@ornl.gov

Abstract. A theoretical framework is briefly reviewed that treats the multiple collisions that a quantum system undergoes with particles in its environment. The dynamics is governed by the quantum Lindblad master equation for the density matrix, which can be solved using a Monte Carlo technique. Its classical limit is identified and provides an opportunity to study the role of decoherence in the quantum to classical transition. Applications are discussed related to the dynamics of ions and atoms interacting with solids and gases, and Rydberg wavepackets subject to coloured noise.

1. Introduction

Any group of particles that forms a subsystem of a larger system can only be described by its density matrix. The interaction with the environment typically leads to decoherence and reduces the subsystem to a statistical mixture of states. The main objective of open quantum system approaches is the derivation and solution of a master equation governing the dynamics of this statistical mixture of states [1,2].

Collisions represent a ubiquitous source of decoherence as atoms (or ions) interact with solids and gases. Understanding and minimizing their effect are therefore of fundamental importance for all atomic systems considered as potential carriers of quantum information or for coherent control protocols. We have developed an open quantum system approach to describe the dynamics of atoms or ions (our subsystems) undergoing a complex array of multiple collisions with particles in their environment [3-6]. The master equation governing the dynamics is a generalized Lindblad equation and can be solved using quantum Monte Carlo techniques. In this paper we briefly describe our progress with reference to the dynamics of the internal electronic states of fast ions traversing solids. We also describe the case of highly-excited (Rydberg) states, its classical limit, and possible

* To whom all correspondence should be addressed.
applications to collisional dephasing of Rydberg wavepackets as well as speculate on other applications. Unless otherwise specified, atomic units are used throughout.

2. Theoretical framework

Consider a subsystem with density matrix $\sigma(t)$ and unperturbed Hamiltonian $H_S$ interacting with a large environment. The time evolution of the density operator $\rho(t)$ of the entire system consisting of the subsystem plus the environment is given by the Liouville–von Neumann equation whose solution is usually out of reach. The focus is therefore on the reduced density matrix of the subsystem, $\sigma(t)$, defined as $\sigma(t) = \text{Tr}_E[\rho(t)]$, where all degrees of freedom of the environment (E) are traced out. Performing this operation in the Liouville–von Neumann equation and applying the Born-Markov approximation [1,5], the master equation for $\sigma(t)$ becomes

$$\frac{d}{dt} \sigma(t) = -i[H_S, \sigma(t)] + R\sigma(t) ,$$

where the coupling to the environment is treated via a relaxation operator $R$, which plays a role similar to that of a collision kernel in a Boltzmann equation. Deriving a useful and accurate expression for this operator is a difficult task even in the perturbative limit when the coupling between the system and the environment is weak. Relaxation operators that treat radiative and collisional processes on an equal footing such that equation (1) adopts the form of a generalized Lindblad master equation have recently been given [3-5]. The advantage of the Lindblad form is that equation (1) can be solved by using quantum trajectory Monte Carlo techniques [1]. In analogy to classical statistical mechanics, where the Boltzmann equation can be solved by using test particle discretization and following the trajectories of an ensemble of test particles in time according to a Langevin equation, the Lindblad form of the master equation can be solved as a Monte Carlo average over quantum trajectories

$$\sigma(t) = \lim_{N \to \infty} \frac{1}{N} \sum_{\eta} |\psi^\eta(t)\rangle \langle \psi^\eta(t)| ,$$

where $\eta$ labels different stochastic realizations of quantum trajectories. The time evolution of $|\psi^\eta(t)\rangle$ is given by a non-linear stochastic Schrödinger equation (NLSSE) [3-5]. Alternatively, it can be calculated from the corresponding time evolution operator, $U^\eta(t,0)$, which can be decomposed into a sequence of products of (non-linear) operators

$$U^\eta(t,0) = U^\eta(t,t_M) \prod_{j=1}^{M} U^\eta_{\text{jump}}(t_j, t_{j-1}) U^\eta_{\text{cont}}(t_j, t_{j-1}) U^\eta(0) .$$

The operators, $U^\eta_{\text{jump}}$, represent discontinuous changes of the wavefunction, i.e., stochastic jumps, at randomly chosen times $t_j$. The operators $U^\eta_{\text{cont}}(t_j, t_{j-1})$ describe the continuous change of the wavefunction during the time period $(t_{j-1}, t_j)$ between stochastic “jumps”. The form of these operators depends on the Lindblad form and the type of environment [3-5]. We have introduced improvements of the standard open quantum system approach used in quantum optics [1]. These extensions overcome several earlier limitations and provide a unified and general treatment. One important improvement is that our reduction of the master equation accounts for both the transient build-up and the destruction of coherences by stochastic processes.

In the following we provide applications of our formalism to two seemingly very different problems. The first is that of the internal electronic state of fast hydrogenic ions traversing solids. Multiple collisions in the solid excite the ions, while the interaction with the radiation field leads to their radiative decay. The sub-system, the internal state of the hydrogenic projectile, is treated
explicitly while the rest of the system, namely the solid and the radiation field, is described as an environment or reservoir. The second application deals with the internal state of a Rydberg atom subject to a source of coloured noise that resembles a stochastic sequence of impulsive collisions. Both examples feature a system Hamiltonian given approximately by

\[ H_s \approx H_{\text{free}} + zF \]  

where \( H_{\text{free}} \) is the Hamiltonian of a hydrogenic free ion or atom and \( F \) is an electric field applied along the \( z \)-axis. When the electron is initially placed in a coherent superposition of states, the resulting time-dependent wavepacket

\[ |\Psi(t)\rangle = \sum_{\alpha} a_{\alpha} e^{-i\epsilon_{\alpha} t} |\Phi_{\alpha}\rangle, \]  

consists of a coherent superposition of (Stark) eigenstates. The mean value of any observable

\[ \langle \Psi(t)|O|\Psi(t)\rangle = \sum_{\alpha,\beta} a_{\alpha}^* a_{\beta} e^{-i(\epsilon_{\alpha} - \epsilon_{\beta}) t} \langle \Phi_{\beta}|O|\Phi_{\alpha}\rangle \]  

oscillates in time with beat frequencies \( \omega_{\alpha\beta} = \epsilon_{\alpha} - \epsilon_{\beta} \). As we show below, such beating makes coherences “visible” experimentally and provides a useful tool to study the dynamics induced by stochastic processes. Because of the simultaneous presence of different beat frequencies dephasing occurs leading to damping of the beat amplitude. Such dephasing is, however, reversible as it is governed by \( H_s \). This is to be distinguished from irreversible dephasing and decoherence due to interactions with the environment. In the limiting case of highly excited (quasi-free) states, quantum trajectories obey a linear stochastic Schrödinger equation (LSSE) \[5\]. With increasing principal quantum number \( n \) radiative decay rates decrease rapidly whereas collisional rates increase due to the increased size of the excited states. Thus, in the high-\( n \) limit, decoherence can be described in terms of a stochastic field \( \vec{F}_{\text{stoc}}(t) \) that takes the form of a random sequence of momentum transfers

\[ H(t) = H_s + \vec{r} \vec{F}_{\text{stoc}}(t) = H_s - \sum_{j} \delta(t-t_j) \vec{r} \Delta \vec{p}_j \]  

where \( \vec{r} = (x,y,z) \) is the position vector of the electron. In the case of ion-solid interactions \( \vec{F}_{\text{stoc}}(t) \) represents collisions with electrons and screened ionic cores. The distributions of jump times \( t_j \) and momentum transfers \( \Delta \vec{p}_j \) are directly related to the differential collisional mean free paths for electron scattering \[5\]. In the case of Rydberg wavepackets \( \vec{F}_{\text{stoc}}(t) \) might result from randomly fluctuating external electric fields or collisions with ambient gas.

### 3. Ion-solid interactions

Figures 1 and 2 illustrate the application of our approach to the transport of 60MeV/u Kr\(^{35+}(1s)\) ions through carbon foils. At such high energies and for thin foils the ion can be assumed to travel in a straight line with constant speed \( v_p = 47 \) a.u. The ion initially carries a single electron in its ground state. As the ion enters the solid, it induces a wake field \[7\] whose effect resembles that of an effective electric field (i.e. \( F \) in equation (4)). Simultaneously, the electron carried by the ion is excited by multiple collisions and subsequently emits light as it radiatively decays. Figure 1 illustrates the time development of the electron density matrix as the ion propagates through the foil. Specifically, it displays a density plot of the modulus, \( |C_{\alpha,\beta}| \), of the relative coherences \( C_{\alpha,\beta} = \sigma_{\alpha,\beta} / \sqrt{\sigma_{\alpha,\alpha} \sigma_{\beta,\beta}} \) in the \( \alpha,\beta \) plane of the bound states of the electron, where \( \sigma_{\alpha,\beta} = \langle \alpha | \sigma | \beta \rangle \) are the matrix elements of the
density matrix representing the internal state of the ion. $C_{\alpha,\beta}$ only provides information about the relative strength of off-diagonal coherences (since $|C_{\alpha,\alpha}|^2 = 1$). For increasing propagation length, $d = v_t \cdot t$, the system becomes nearly incoherent (i.e. $C$ is nearly diagonal), but for short propagation lengths many coherences reach their maximal amplitudes. These large coherences originate with the first collisional excitation event that occurs as the ion enters the foil. The transient build-up of coherences has a direct effect on the population dynamics, $\sigma_{\alpha,\alpha}(d)$.

Figure 1. Relative coherences $C_{\alpha,\beta}$ of the density matrix in the fine structure basis $|n, j, \ell, m_j\rangle$ for the internal state of a 60MeV/u Kr$^{35+}$ ion traversing amorphous carbon following various propagation lengths $d$.

Information on the transient populations of the $3p_{1/2}$ and $3s_{1/2}$ states and their coherence can be obtained experimentally [6,8-10] via the line emission intensities from these states as a function of foil thickness (figure 2b). For thin foils the increase in the intensity ratios as a function of foil thickness seen in figure 2b is a direct consequence of both transient collisional coherences and of state mixing induced by the wake field. This is illustrated in figure 2a by a simplified three-state model that includes only the wake field and assumes that the $3s_{1/2}$ and $3p_{1/2}$ states are initially fully coherent with either $C_{3s_{1/2},3p_{1/2}} = -i$ or $C_{3s_{1/2},3p_{1/2}} = +i$. The first case corresponds approximately to the actual value expected under single-collision conditions. The intensity ratio initially increases, reaches a maximum, and subsequently oscillates. The opposite behaviour is seen for $C_{3s_{1/2},3p_{1/2}} = +i$. (The beats observed here are a direct consequence of equation (6)). The behaviour for short times mimics that of the full multi-state transport simulations in figure 2b. Subsequent oscillations observed in the simplified three-state model are not observed for the real collision system because they are rapidly damped by collisionally-induced decoherence. Full simulations with the wake field turned off in figure 2b further illustrate the role played by wake field mixing. Also included in figure 2b are the results of simulations in which radiative transitions are neglected while the ion is inside the foil. Only when radiative and collisional processes are taken into account on an equal footing can good agreement with experiment be achieved [4-6].
Our work on ion-solid interactions has allowed us to extend open quantum system approaches. One important improvement [4] is that we properly account for the loss of probability from the finite Hilbert space used to represent the system. This is a common problem in quantum mechanics when dealing with transitions into continuum states. In our approach the complement of our finite Hilbert space becomes part of the environment. Electron loss, e.g. ionization, represents a net probability flux out of the system (a sink). Our most recent extension [3] to bare ions, which do not carry any electrons prior to entering the foil, requires that a coherent source of probability flux be taken into account. Only as the projectile captures an electron does the one-electron Hilbert space of the system become occupied. This feeding is partially coherent and requires a generalization of the Lindblad equation. We have developed an extension of the standard open quantum system approach that also accounts for probability flux into the simulated system using an inhomogeneous Lindblad master equation [3]. This generalization has enabled us to incorporate both electron capture and ionization and to identify coherences originating in the electron capture density matrix and through wake field mixing of angular momentum states.

4. Dynamics of highly-excited (Rydberg) states

The high-$n$ limit, where the dynamics is governed by a stochastic Hamiltonian (equation (7)), is of interest because it has a classical interpretation. In the classical limit the state of the system is described by a classical phase space probability whose dynamics is governed by a classical Liouville equation with a collision (relaxation) kernel. This equation can be solved using Monte Carlo techniques by following the trajectories of a discrete set of test particles in time according to the Langevin equation for the stochastic Hamiltonian, equation (7) [11]. The LSSE limit provides the opportunity to study the role played by the environment in the classical limit of quantum mechanics. For transport of fast ions/atoms through amorphous solids, where collisional decoherence rates are very large, classical and quantum simulations for weakly bound states yield very similar results even when the electrons are initially close to their ground states [12]. However, in the case of Rydberg atoms one can envision applications where coherence can be maintained for extended periods. Rydberg wavepackets with values of $n \sim 400$ can be routinely produced in the laboratory [13] and can...
be engineered and probed by means of carefully tailored electromagnetic pulses. Such atoms have classical orbital periods $T_n = \frac{2\pi}{n^3}$ of $\sim 10\text{ns}$ and can be manipulated with conventional electronics. At the same time, because of their large physical size and weak binding, they are very fragile and interact strongly with stray fields and ambient particles. These characteristics make them a unique laboratory to explore fundamental notions related to coherent control and decoherence.

Rydberg atoms provide a means to address questions related to the problem discussed in the previous section. Consider a stationary $K(351p)$ state produced by laser excitation. Sudden application of a “strong” dc field ($F=F_{dc}$ in equation (4)) couples the initial state to other angular momentum states in the initial and neighbouring $n$-manifolds leading to a wavepacket like that described by equation (5). The dominant beat frequencies can be observed by applying, after a variable delay time $t_D$, a probe half-cycle pulse (HCP) of duration $T_{probe} \ll T_n$. In this limit, the probe HCP delivers an impulsive momentum transfer or kick $\Delta p_{probe} = \int_{-\infty}^{\infty} dt F_{probe}(t)$ to the excited electron and the resulting survival probability reflects the temporal state of the electron. Use of the pump-probe scheme represented by the time-dependent field shown at the top of figure 3 leads to periodic variations in the survival probability, i.e., in the fraction of Rydberg atoms not ionized by the probe pulse, as the time delay $t_D$ is varied. Analysis of the frequency spectrum of these beats [14] indicates that the field step creates a wavepacket centred at $n \approx 350$ with dominant frequencies consisting of the (shifted) Kepler frequencies $\omega_n^* = n^{-3} \pm (3/2)nF_{dc}$ and the Stark frequency $\omega_{\text{Stark}} = 3F_{dc}n$ (equal to the nearest neighbour splitting in the Stark manifold). The rapid oscillations in the simulations shown in figure 3(a) are associated with the shifted Kepler frequencies and have a period $\sim 6\text{ ns}$. The slower oscillations at the Stark period $T_{\text{Stark}} \sim 40\text{ ns}$ provide the dominant peak in the Fourier transform of the survival probability displayed in figure 3(b).

**Figure 3.** Top: Profile of applied electric field sequence. (a) Time dependence of the survival probability for $K(351p)$ atoms and $F_{dc}=18.8\text{ mV/cm}$, and $\Delta F_{probe}=-0.532/n\text{ a.u.}$ (b) Fourier transform of the survival probability which peaks near $\nu = \nu_{\text{Stark}} = \frac{2\pi}{\omega_{\text{Stark}}} = \frac{2\pi}{(3F_{dc}n)}$. (c) and (d) same as (a) and (b) respectively but with a stochastic sequence of short electric field pulses with amplitude $\Delta F=0.1F_{dc}$ superposed on $F_{dc}$ (see text). Experimental data (symbols) are compared with the results of classical trajectory simulations (solid lines).

The damping of the beats evident in figure 3(a) is due to the finite spread of the frequency spectrum of the wavepacket which lead to coherent “reversible” dephasing. Stark quantum beats provide a valuable tool to probe decoherence, i.e., “irreversible dephasing,” because the coherent dephasing time is very long (hundreds of nanoseconds). Decoherence can be observed through accelerated damping of the Stark beats due to interactions with the environment. Figures 3(c) and (d) show proof of principle experiments and calculations using “controllable” noise provided by superposing on the dc field $F_{dc}$ the output of a pulse generator that produces stochastic variations,
$\Delta F = \pm 0.1 F_{dc}$, in its magnitude. This generator divides time into a series of bins of width $T_f$ and in each randomly outputs a voltage of zero or $V$. Noise accelerates the dephasing and the increased decoherence rate can be clearly seen from the broadening of the peak of the Fourier transform. Insights into the decoherence mechanisms can be gained by changing the amplitude ($\Delta F$) or the colour of the noise ($\nu T \equiv 1/T_f$) [14].

The Heisenberg time (or quantum break time), $\tau_H$, beyond which classical and quantum dynamics diverge increases rapidly with $n$ ($\tau_H \propto n^4$) and at high $n$ becomes large (> 1 μs at $n \sim 400$). Thus, our analysis of figure 3 was based on considerations of classical dynamics. At lower $n$ a number of quantum phenomena such as revivals [15] should be within reach of experiment and might also be used to study the role of decoherence in the classical limit of quantum mechanics.

5. Outlook

A theoretical framework for studying the dynamics of quantum systems in contact with a large environment based on a stochastic non-linear Schrödinger equation has been discussed together with its application to the dynamics of the internal states of fast ions passing through solids and undergoing multiple collisions. Coherent beating induced by the wake field and its subsequent collisional destruction was addressed. It was also shown that similar behaviour can be seen for Rydberg wavepackets in a dc field and subject to external stochastic noise.

Applications in the study of decoherence of Rydberg wavepackets due to collisions with ambient gas can be envisioned. For sufficiently large values of $n$ the Rydberg electron and core ion act as independent scatterers. Previous work involving Rydberg atoms in stationary states [16] has shown that studies of collision processes dominated by the electron-target interaction can provide valuable information on inelastic electron-molecule scattering cross sections at energies extending down to a few μeVs. Since even at these energies the velocity of the electron is typically greater than that of the target neutral, the collision velocity can be controlled by initially exciting the electron to different $n$-levels. A variety of inelastic electron-molecule collision processes that lead to ionization of the Rydberg atom have been examined in this manner. The damping of Stark beats might provide a sensitive tool for examining quasi-elastic electron-atom (or molecule) collisions at ultra-low electron energies [17]. The enhanced damping rate should be proportional to the scattering cross section, which could be extracted by fitting experimental data to calculations including collisional decoherence and a few free parameters determining the cross-sections. The development of more sophisticated protocols to separate different sources of dephasing by exploiting echo phenomena induced by reversing the external dc field [18] can also be imagined.

Another extension involves the interaction of the wavepackets associated with free particles, in particular neutral atoms, with crystalline surfaces under grazing incidence. Because of the very different parallel and perpendicular momenta, the wave nature of the wavepacket becomes important even for beam energies of several keV permitting observation of diffraction patterns [19,20]. Kilo-electronvolt beams, however, undergo inelastic collisions with the thermally excited lattice providing a source of decoherence that should influence the contrast of the diffraction patterns. Theoretical work employing the present approach to analyze the role of decoherence in diffractive scattering is underway [21].

Acknowledgments

This research is supported by the OBES, U.S. DoE to ORNL managed by the UT-Batelle LLC under Contract DE-AC05-00OR22725, the FWF-SFB-P016 and FWF-P173591 HPRI-CT-2005-026015 (Austria), the NSF, the Robert A. Welch Foundation, and the ONR.

References

[1] C.W. Gardiner and P. Zoller 1999 Quantum Noise (Springer, New York).
[2] K. Blum 1981 *Density Matrix Theory and Applications* (Plenum, New York).

[3] M. Seliger, C.O. Reinhold, T. Minami, D.R. Schultz, M. S. Pindzola, S. Yoshida, J. Burgdörfer, E. Lamour, J.-P. Rozet, and D. Vernhet 2007 *Phys. Rev. A* 75 032714.

[4] M. Seliger, C.O. Reinhold, T. Minami, and J. Burgdörfer 2005 *Phys. Rev. A* 71 062901.

[5] T. Minami, C. O. Reinhold, and J. Burgdörfer 2003 *Phys. Rev. A* 67 022902.

[6] T. Minami, C. O. Reinhold, M. Seliger, J. Burgdörfer, C. Fourment, E. Lamour, J.-P. Rozet, D. Vernhet, and B. Gervais 2002 *Phys. Rev. A* 65 032901.

[7] P.M. Echenique, W. Brandt, and R. H. Ritchie 1986 *Phys. Rev. B* 33 43.

[8] E. Lamour, B. Gervais, J.-P. Rozet, and D. Vernhet 2006 *Phys. Rev. A* 73 042715.

[9] D. Vernhet, C. Fourment, E. Lamour, J-P. Rozet, B. Gervais, L. J. Dubé, F. Martin, T. Minami, C. O. Reinhold, M. Seliger, and J. Burgdörfer 2001 *Phys. Ser. T92* 233.

[10] D. Vernhet, J-P. Rozet, I. Bailly-Despiney, C. Stéphan, A. Cassimi, J-P. Grandin, and L. J. Dubé 1998 J. Phys. B 31 117.

[11] J. Burgdörfer and J. Gibbons 1990 *Phys. Rev. A* 42 1206.

[12] P. Kürpick, C O Reinhold, J Burgdörfer, and B. Gervais 1998 *Phys Rev A* 58 2183.

[13] F.B. Dunning, J.C. Lancaster, C.O. Reinhold, S. Yoshida, and J. Burgdörfer 2006 *Adv. At. Mol. Phys.* 52 49.

[14] S. Yoshida, C.O. Reinhold, J. Burgdörfer, W. Zhao, J.J. Mestayer, J.C. Lancaster, and F.B. Dunning 2007 *Phys. Rev. A* 75 013414.

[15] J.A. Yeazell, M. Mallalieu, and C.R. Stroud 1990 *Phys. Rev. Lett.* 64, 2007.

[16] S.B. Hill, M.T. Frey, F.B. Dunning, I.I. Fabrikant 1996 *Phys. Rev. A* 53 3348.

[17] C.O. Reinhold, J. Burgdörfer, and F. B. Dunning 2005 *Nuc Instr Meth B* 233 48.

[18] S. Yoshida, C. O. Reinhold, J. Burgdörfer, W. Zhao, J.J. Mestayer, J.C. Lancaster, and F. B. Dunning 2007 *Phys. Rev. Lett.* 98, 203004; J. J. Mestayer et al 2007 ICPEAC proceedings of invited papers.

[19] P. Rousseau, H. Khemliche, A.G. Borisov, and P. Roncin 2007 *Phys. Rev. Lett.* 98, 016104.

[20] A. Schüller, S. Wethekam, and H. Winter 2007 *Phys. Rev. Lett.* 98, 016103.

[21] N. Simonovic, F. Aigner, B. Solleder, and J. Burgdörfer 2007 ICPEAC contributed abstract Mo31.