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Electron scattering and dephasing rate of Rydberg atoms in a plasma

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Abstract

Rydberg atoms can be considered as mesoscopic systems at the interface between quantum and classical behaviour. The interaction with the surroundings (bath) becomes essential and leads to dephasing of the wavefunction. An important process in Rydberg plasmas is the collision with free charge carriers. Transition rates due to Coulomb interaction are considered in the Born approximation and are shown to coincide with the dephasing time according to linear response theory for mesoscopic devices. We point out that this description of the dephasing process relies on weak coupling between the Rydberg states and the bath and becomes invalid, if strong scattering is of importance.

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1. Introduction

Rydberg atoms are highly excited atomic states, where the electron is only weakly bound to the ion and its wavefunction extends over mesoscopic distances. Typical values for the principal quantum number are $n = 10, \ldots, 100$ so that the binding energy becomes lower than room temperature down to the Kelvin range. As an example, for a value of $n, l = 35$ s of a Rb Rydberg state [1], we have for the characteristic radius a value of $n^2 a_\mathrm{B} \approx 54$ nm. The ionization potential is $\approx \mathrm{Ry}/1000 \approx 0.0136$ eV, which equals a thermal energy at $T \approx 155$ K.

These weakly bound states have interesting properties. According to their mesoscopic extension, most of their properties can be obtained in a quasiclassical description. Investigating such systems helps us to understand the transition from quantum to classical behaviour, which is a general topic considered in open quantum systems [2, 3]. Different applications, in particular quantum computing [4], are under discussion [5, 6]. Mesoscopic physics is a new field of research which develops special concepts [7]. In particular, we are interested in the dephasing time which characterizes the loss of quantum coherence. Meanwhile the latter is one of the limits of applicability of such devices.

The sensitivity to perturbers is a characteristic property of Rydberg atoms. The interaction with the radiation field leads to the emission and absorption of radiation and, as a consequence, to the width of the spectral lines corresponding to the transition rate. At finite densities, the interaction with other particles (neutral as well as charged) may become more important for the transition rates. Thus in a Rydberg gas consisting of many Rydberg atoms (e.g. in a magneto-optical trap at very low temperatures $T < 1$ K [8, 9]) the dipole interaction among Rydberg atoms separated at distance R is the most perturbing contribution. The interaction potential of the latter can be introduced from resonance interaction ($\sim R^{-3}$) to van-der-Waals attraction ($\sim R^{-6}$) with high rates due to the huge atomic matrix elements [8, 10].

If no external electrical field is applied, the Rydberg gas develops into an ultracold neutral plasma (Rydberg plasma) where additionally free charge carriers (electrons, ions) are present [11–13] and the Coulomb interaction provides the strongest coupling of Rydberg energy levels which is responsible for the width of the spectral lines. The resulting broadening of stationary states limits the number of excited states and transition lines that can be clearly identified. However the line-broadening can be separated in a homogeneous part due to collective dephasing and an inhomogeneous part that rather represents line-splitting due to different levels that can be distinguished in a more detailed description. The latter can in well chosen situations even be used to suppress dephasing [5] in order to prepare qubits in mesoscopic ensembles, if the excitation laser bandwidth is small enough to resolve the splitting. The time evolution of Rydberg plasmas has been studied in [14–18] where the collision rates are mostly calculated classically. As the dominating radiation source the thermal field of the apparatus that is at room temperature has been established. Collision processes from free electrons and ions result in transitions among bound states as well as ionization and recombination.

In this paper we will focus our considerations to transitions among bound states in the presence of charged particles. For the given case, we examine the transition rates of Rydberg atoms on a many-particle background and relate it to the dephasing time. On the background of mesoscopic metal and semiconductor devices the dephasing time has been related to statistical properties of both, the reduced system and the bath [7, 19]. This approach is very attractive in a plasma environment because there the interesting statistical quantities as the dielectric function and the dynamical structure factor are well investigated and can be calculated, e.g., in linear response theory.

In this contribution we compare expressions known from plasma theory with results of the mesoscopic description of dephasing. Limits of such a treatment if strong scattering is considered are pointed out.

2. Electron scattering at Rydberg atoms

We now consider the Rydberg atoms in a cold, dilute electron–ion plasma that has been studied in several experiments [11]. In the following we choose a typical setup of the plasma with electron temperature $T = 20$ K and density $\rho_e = 10^9$ cm^{-3}. A further characterizing property of such a plasma is the plasma frequency, here $\omega_{pl} = \sqrt{\rho_e e^2/(\varepsilon_0 m_e)} = 8.6 \times 10^9$ s^{-1}. The Coulomb coupling parameter is $\Gamma = \frac{e^2}{4\pi\varepsilon_0 k_B T}(4\pi\rho_e/3)^{1/3} = 0.135$, and the degeneracy parameter $\Theta = 2m_e k_B T/\hbar^2(3\pi^2\rho_e)^{-2/3} = 5 \times 10^5$. Such a plasma can clearly be described as non-degenerate.

Averaging over l and summing over l', the transition rates for weak scattering can be related to the rate coefficient $k_{nn'}(T)$, obtained by averaging the total cross section $\sigma_{nn'}(E_e)$ of an incident electron with energy E_e over the Maxwell–Boltzmann distribution, which holds

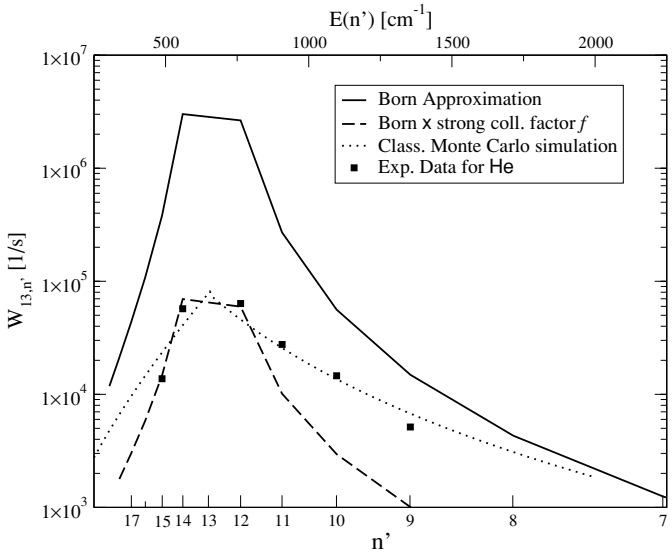

Figure 1. Transition rates of $n = 13$ to near n' states by $T = 300$ K electron plasma with density $\rho_e = 10^9$ cm^{-3} for the BA with and without effect of strong collisions [20] comparing to He data from [21] and classical trajectory MC calculation [22].

approximately for an ultracold plasma as long as one neglects the utmost regions [18],

$$W_{nn'} = \rho_e k_{nn'}(T) = \rho_e \langle \sqrt{(2E_e/m_e)}\, \sigma_{nn'}(E_e) \rangle_T . \tag{1}$$

The total cross section can be expressed in terms of the Born approximation (BA),

$$\sigma^{(\mathrm{BA})}_{nn'}(E_e) = \frac{8\pi \mathrm{Ry}}{E_e} \int_{\sqrt{2m_e/h^2}|\sqrt{E_e}-\sqrt{E_e-\hbar\omega_{nn'}}|}^{\sqrt{2m_e/h^2}(\sqrt{E_e}+\sqrt{E_e-\hbar\omega_{nn'}})} \frac{M^2_{nn'}(q)\,\mathrm{d}q}{q^3}, \tag{2}$$

with transition frequency $\hbar\omega_{nn'} = E_{n'} - E_n = \mathrm{Ry}\,(1/n^2 - 1/(n')^2)$ and the atomic matrix element $M^2_{nn'}(q) = n^{-2} \sum_{lm,l'm'} |\langle n'l'm'| \exp(\mathrm{i}\mathbf{q}\cdot\mathbf{r})|nlm\rangle|^2$. The modification due to strong collision is taken into account by a reduction factor $f(n, \triangle n; \theta)$ obtained in semiclassical approximation [20],

$$\begin{aligned} k_{nn'}(T) &= \langle \sqrt{2E_e/m_e}\, \sigma_{nn'}(E_e) \rangle^{(\mathrm{BA})}_T\, f(n, \triangle n; \theta), \\ f(n, \triangle n; \theta) &= \frac{\ln\left[1 + \frac{1}{\triangle n\theta(1+2.5n/(\triangle n\theta))}\right]}{\ln\left[1 + \frac{1}{\triangle n\theta}\right]}, \qquad \theta = \sqrt{\frac{|E_n|}{k_B T}}. \end{aligned} \tag{3}$$

Equation (3) is valid for excitation $n' - n = \triangle n > 0$. A corresponding formula holds for deexcitation.

In figure 1 the transition rates from $n = 13$ to near n' of the BA (H wavefunction) with and without consideration of strong collisions are compared to an experiment on He atoms [21] and classical trajectory Monte Carlo (CTMC) simulations [22]. The principle difference between H and He atoms, the quantum defect of the He $l = \mathrm{s}, \mathrm{p}$ levels, does not play a considerable role for the $n \to n'$ rates, because they have a low statistical weight due to small degeneracy when summing over l' (averaging over l).

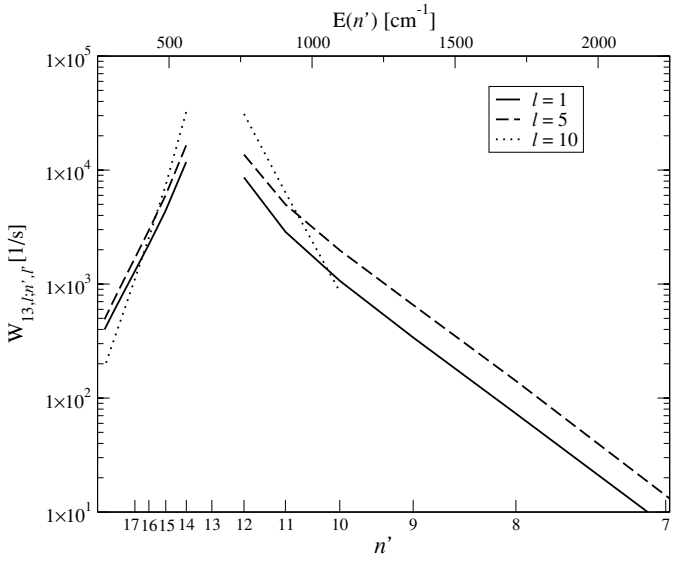

Figure 2. Radiative dipole transitions of $n = 13, l = 1, 5, 10$ to near $n', l' = l \pm 1$ states at $T = 300$ K thermal BBR.

As is well known, the BA strongly overestimates the part from strong collisions with $E_e < |E_n|$. The comparison with the CTMC calculations shows that the measured rates can be reasonably explained with classical calculations. Comparing classical and quantum calculations, one should note that the quantum correspondence of the classical treatment are wave packets describing localization in space. These can be represented by superpositions of a set of atomic energy eigenstates with adequate quantum numbers n, l, m; see [23]. In the present work we do not deal with wave packets as we consider atomic energy eigenstates with given quantum numbers n, l, m, and sum or average over l', m' or l, m, respectively. A mixing (superposition) of atomic eigenstates with different quantum numbers n, l, m occurs choosing the appropriate initial and final states induced by the necessary treatment of strong collisions and the introduction of the corresponding reduced density-matrix as discussed in section 4.

The process of electron scattering generally competes with radiative transitions due to black-body radiation (BBR) of the environment, in particular the experimental apparatus. This leads in dipole approximation to induced emission and absorption transition rates given by [20, 24],

$$W_{nl,n'l'} = \frac{4\pi^2}{3}\frac{e^2}{4\pi\varepsilon_0\hbar^2 c} I(\omega_{nn'}) \frac{\max\{l, l'\}}{2l+1} |\langle n'l'|r|nl\rangle|^2, \tag{4}$$

that represent the interaction of Rydberg levels with the thermal radiation (photon) field. The intensity distribution $I(\omega_{nn'})$ for BBR is given by Planck's formula [24] and the dipole matrix elements can be calculated analytically for hydrogen. For high n alkali atoms the dipole matrix elements can be calculated numerically [1]. The transition rates from $n = 13$ to near n' for $l = 1, 5, 10, l' = l \pm 1$ at room temperature $T = 300$ K for hydrogen are shown in figure 2. It clearly shows that the largest rates arise at small $\triangle n$. This is even more pronounced for the higher l states though the effect is smaller. The summed rates give an overall lifetime that lies in the order of several microseconds, which equals the $n \to n'$ electron scattering transitions at an electron density $\rho_e \sim 10^7$ cm^{-3}.

Another process that needs to be mentioned here is the resonant dipole interaction, which acts like a hopping process [10]. The interaction shows R^{-3} dependence and is proportional to the density of Rydberg atoms in the involved states. Due to the huge dipole matrix element, its strength compares to the processes discussed above. In a Rydberg gas, the disorder of the Rydberg atom's distances can be considered as a source of dephasing for a two-atom superposition state, if at least one of the states is exposed to resonant dipole interaction. Its contribution to the linewidth has been found to agree with the predicted $\Delta\omega \approx (2\pi)^{-1} V_\mu$ for the potential $V_\mu = (4\pi\varepsilon_0)^{-1}\mu^2 R^{-3}$ and dipole matrix elements μ [8].

In general, all three processes mentioned are important for the time evolution of Rydberg states in a many-particle environment. The situation we are especially interested in occurs for parameter values, when the dominating process is due to collisions with charge carriers. This is the case, e.g., for the electron densities $\rho_\mathrm{e} \sim 10^9\ \mathrm{cm}^{-3}$ given above. In the following sections, we restrict ourselves to transition rates by collisions with charged particles as can be calculated by the cross section, equations (2), and (3). In particular we are interested in the way how to take into account strong collisions appropriately and extend the BA.

3. Many-particle picture of bath scattering

To describe the interaction of the Rydberg atoms with a many-particle environment the expressions of the last section are valid only, if one restricts oneself to the case of asymptotically free scatterers. Generally it is necessary also to include the collective properties of the plasma (e.g. plasmons). A possible approach offers the transversal dielectrical function which is related to the polarisation function of the medium [25]. In its general form also effects of bound states in the medium can be included [26].

Considering a probe atom within the medium, the total state can be assumed to factorize to the form $|\Psi\rangle = |nlm\rangle \otimes |\phi_\mathrm{e}\rangle$. The transition rates in lowest order perturbation theory can then be obtained from Fermi's golden rule,

$$W_{nn'} = \frac{4e^4}{\hbar^2(4\pi\varepsilon_0)^2\Omega_0}\int\frac{\mathrm{d}\mathbf{q}}{q^4}\int_{-\infty}^{\infty}\mathrm{d}\omega\, S_\mathrm{e}(\mathbf{q},\omega)\, M^2_{nn'}(q)\,\delta(\omega-\omega_{nn'}), \tag{5}$$

with the definition of the dynamical structure factor (DSF) of the plasma electrons,

$$S_\mathrm{e}(\mathbf{q},\omega) = \sum_{\phi_\mathrm{e},\phi'_\mathrm{e}} |\langle\phi'_\mathrm{e}|n_\mathbf{q}|\phi_\mathrm{e}\rangle|^2\delta(\omega+\omega_{\phi_\mathrm{e}\phi'_\mathrm{e}})P_{\phi_\mathrm{e}}. \tag{6}$$

The DSF is a well-known quantity in linear response theory (LRT) containing the particle density $n_\mathbf{q}(\mathbf{r})$, the many-particle states $|\phi_\mathrm{e}\rangle$, and their statistical weight P_{ϕ_e}. From the fluctuation–dissipation relation,

$$S_\mathrm{e}(\mathbf{q},\omega) = \frac{\hbar\Omega_0}{\pi}\frac{1}{\mathrm{e}^{\beta\hbar\omega}-1}\frac{\varepsilon_0 q^2}{e^2}\mathrm{Im}\left\{\varepsilon_\mathrm{e}^{-1}(\mathbf{q},\omega)\right\}, \tag{7}$$

the DSF can be calculated from the dielectric function, $\varepsilon_\mathrm{e}(\mathbf{q},\omega)$ [27].

Using $\mathrm{Im}\left\{\varepsilon_\mathrm{e}^{-1}(\mathbf{q},\omega)\right\} \approx -\mathrm{Im}\{\varepsilon_\mathrm{e}(\mathbf{q},\omega)\}$, which is valid far off the plasmon pole $\omega = \omega_\mathrm{pl}$, and the dielectric function in its well-known random phase approximation within equation (7), equation (5) reduces to exactly the same rates of the BA in equations (1), and (3).

In equation (5), however, additional many-particle effects originating from inter-electron collisions can be included into the rates [28]. One obtains the rates for weak scattering of a non-ideal electron gas at a Rydberg atom. The restriction to weak scattering that follows from Fermi's golden rule as a starting point for equation (5) ignores the effect of strong collisions.

For comparison, in the similar situation of the theory of spectral line-shapes this restriction belongs to the impact approximation in the BA. Strong collisions, on the other hand, are dealt with by the ionic microfield concept and cut-off procedures [26, 29]. On the other hand, for transitions among low H states the approach within LRT has been extended to the treatment of strong collisions [30].

4. Dephasing rate and life time

The many-particle formulation of the last section is a special case of a system-background picture known from the studies of open quantum systems [3]. Within these, one separates the system of interest (S) that is coupled by an interaction potential V_I to a heat bath (B) whose microscopic states cannot be specified. The total Hamiltonian of such a system reads

$$H = H_\mathrm{S} + H_\mathrm{B} + V_\mathrm{I}. \tag{8}$$

The time evolution of the system coupled to the bath is described by a statistical operator $\rho(t)$ according to the von-Neumann equation. Assuming that it can be decomposed as $\rho(t) = \rho_\mathrm{S}(t) \otimes \rho_\mathrm{B}$ so that the correlations between the system ($\rho_\mathrm{S}(t)$) and the bath (ρ_B, in thermal equilibrium) can be neglected, the variables of the bath can be averaged out, so that in lowest order with respect to the interaction V_I a quantum-mechanical (Lindblad) master equation [25, 31] is obtained. The general form of the dynamics can be written as

$$\mathrm{i}\hbar \frac{\partial}{\partial t}\rho_\mathrm{S}(t) = [H_\mathrm{S}, \rho_\mathrm{S}(t)] - \frac{\mathrm{i}}{2}\sum_k \left(L_k^+ L_k \rho_\mathrm{S} + \rho_\mathrm{S} L_k^+ L_k - 2L_k \rho_\mathrm{S} L_k^+\right), \tag{9}$$

with the Lindblad operators L_k acting in the Hilbert space of the system's states.

The properties of the master equation, in particular the relation to the Pauli master equation, has been discussed in the literature for the harmonic oscillator coupled to a bath of harmonic oscillators. Especially the phenomenon of decoherence (i.e. the suppression of wavefunction interferences) is investigated. In the picture of equation (9) decoherence is described by the damping of the non-diagonal elements of the reduced density operator of the system ($\rho_\mathrm{S}(t)$) represented in a basis where the interaction potential operator is diagonal [31]. The application of a master equation of Lindblad type to Rydberg atoms is discussed recently in the treatment of superradiance [32] and was derived for atomic properties in a optical dense medium in [33].

Closely related to this general treatment of open quantum systems, the dephasing of a quantum subsystem has been studied in mesoscopic devices that are of the scale like Rydberg atoms [7]. For a statistical system that has a very high quantum uncertainty it was shown [19] that any transition due to the dynamical system–bath interaction randomizes the phase of the system so that the total transition rate can be taken as a measure of dephasing. In this case the dephasing time τ_ϕ can be estimated by the formula obtained in using LRT [7],

$$\frac{1}{\tau_\phi} = \frac{1}{\hbar^2 (2\pi)^2 \Omega_0} \int \mathrm{d}\mathbf{q} \int_{-\infty}^{\infty} \mathrm{d}\omega \, |V_q|^2 S_\mathrm{S}(-\mathbf{q}, -\omega) S_\mathrm{B}(\mathbf{q}, \omega). \tag{10}$$

Both the system and the bath enter equation (10) by their DSF. If we assume the system to be a single, (with respect to the bath) isolated Rydberg atom, the DSF of the system can be expressed in terms of the atomic matrix elements, $S_\mathrm{S}(-\mathbf{q}, -\omega) = \sum_{\alpha n'} |M_{\alpha n'}(q)|^2 \delta(\omega_{\alpha n'} - \omega) P_\alpha$. With the sum over α and P_α the initial preparation of the quantum-mechanical state (which has to be assumed metastable in this statistical formulation) enters into the DSF. It generally arises from the experimental setup. Using $S_\mathrm{B}(\mathbf{q}, \omega) = S_\mathrm{e}(\mathbf{q}, \omega)$ and setting $P_\alpha = \delta_{n\alpha}$, however, we arrive again at equation (5).

Therefore, the determination of the dephasing time according to equation (10) is consistent with the evaluation of the total transition rate, $\tau_\phi^{-1} = \sum_{nn'} W_{nn'}$, as calculated in equation (5). Also it is compatible with the evaluation of the transition rates in BA according to equation (1) if we follow the approximation discussed in the last section. We conclude that the calculation of the dephasing time τ_ϕ and the rate of collisions with the medium are compatible in the BA, leading to identical results.

This relation shows, however, that equation (10) is of restricted validity. It applies to the limit of weak coupling of the system and the bath that is appropriate as long as the BA gives the correct transition rates. But, on the other hand, we saw in figure 1 that strong collisions significantly modify the rates obtained in the BA. It is evident that in this case also equation (10) becomes questionable. The same holds for the LRT approach of section 3. As is well known from the theory of spectral line-shapes, for strong collisions the result of the BA will be reduced, as can be done using a cut-off procedure [29] or more sophisticated approaches treating the atomic self-energy in T-matrix approximation [30]. Therefore equation (10) has to be improved, if strong collisions become of importance.

A possible solution of this problem being present, if the Rydberg atom is strongly coupled to the plasma, is achieved going beyond the BA. In particular, the perturbing charged particle (within the Weisskopf radius [29] separating strong scattering processes) can be included into the system under consideration, which corresponds to the full solution (T-matrix) of the three-particle system given by the Rydberg atom and the charged particles from the plasma. With respect to low H lines, such an approach has been performed in the theory of spectral line-broadening [30]. Incorporating the strongly perturbing charged particles into the system S, equation (10) after this new decomposition of system and bath may become valid again. Then, of course, the matrix elements based on the isolated atomic (H) states have to be replaced by the corresponding matrix elements of the extended system consisting of the Rydberg atom an the strongly coupled charge carriers.

5. Conclusion

Transitions to neighboured levels by collision with charged particles become the dominating process for Rydberg atoms in an ultracold plasma at increasing charge carrier density. The rates in the BA, that are valid for weak scattering, can be obtained on the basis of a kinetic as well as a statistical way in the framework of LRT. They are consistent with the dephasing rate following an approach of mesoscopic devices.

The restriction to weak collisions overestimates the scattering rates for the considered transitions, if strong collisions play an important role. If we deduce from the scattering rate the decoherence time according to the mesoscopic procedure ignoring strong collisions we arrive for typical conditions at a dephasing time of 10–100 ns for a $n = 13$ level. If one applies a renormalization factor as common for spectral linewidth, the dephasing time will increase by a factor of about 50. Thus an improvement of expression (10) for the dephasing time is necessary, which goes beyond the BA. This may be achieved including strong perturbers in the definition of the reduced system, so that the original separation into a reduced system strongly coupled to the bath is avoided.

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