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RELEVANT ION TIME SCALES FOR ELECTRON IMPACT PROCESSES OF ATOMS IN DENSE PLASMAS

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INTRODUCTION

It is now well known that collisional atomic processes in dense plasmas can be considered to arise from density fluctuation perturbations^{1,2,3}. Through the fluctuation-dissipation theorem this description can be related to those of other methods⁴. In particular, it is possible to describe these processes in terms of the dynamic structure factor (DSF) $S(\mathbf{k}, \omega)$, defined by

$$S(\mathbf{k}, \omega) = \int_{-\infty}^{\infty} d\tau e^{i\omega\tau} \langle n(\mathbf{k}, \tau) n(-\mathbf{k}, 0) \rangle, \quad (1)$$

which is a measure of the power spectrum of density fluctuations. In the atomic transition problem, the DSF is evaluated at the atomic transition energy $\hbar\omega$ and, in general, the plasma density is taken as the plasma charge density and includes all species weighted by their charge. In such a Stochastic Model, interesting dynamical screening behavior arises as a result of different atomic transition energies sampling different frequency regimes of the DSF. Typically the DSF is broken up so that we may speak of electron and ion impact processes separately.

Electron impact processes, characterized by an electron one component plasma (OCP) DSF, denoted $S_{ee}(k, \omega)$, may typically be treated within the Born approximation since the electrons, which are light and typically hot, have a high average velocity. Under these conditions the rate can be written directly in terms of $S_{ee}(\mathbf{k}, \omega)$ ¹. Different dynamical screening behavior arises depending on whether the transition energy $\hbar\omega$ is greater than the electron plasma frequency, less than the plasma frequency, or near the plasma frequency.

For ion impact processes it is almost always the case that transition frequencies are well above the ion plasma frequency. The dominate behavior then arises from the high frequency limit of $S_{ii}(\mathbf{k}, \omega)$,

$$\lim_{\omega \rightarrow \infty} S_{ii}(\mathbf{k}, \omega) = \lim_{\omega \rightarrow \infty} \int_{-\infty}^{\infty} d\tau e^{i\omega\tau} \langle n_i(\mathbf{k}, \tau) n_i(-\mathbf{k}, 0) \rangle, \quad (2)$$

which has significant contributions only for small values of τ . That is, only short times are important in determining $S_{ii}(\mathbf{k}, \omega)$ in the high frequency limit. The treatment of ions is further complicated by the fact that ions can be highly charged. As such, ions cannot be treated in the same perturbative manner as the electrons. Thus, it is clear that ions represent *strong perturbors* which have *little motion* during a collision process.

In reality, of course, both electrons and ions simultaneously perturb the atoms so that the criteria governing the ions must be taken into account and included when treating electron collisions and vice versa. These arguments suggest that the ions must be treated in a non-perturbative manner with little movement during an electron collision process. Thus, the atom sees a "snapshot" of a given ion configuration throughout the duration of the transition. This prediction is in distinct contrast to previous work which either neglects the surrounding ions entirely or treats them within a thermally averaged potential, which corresponds to a long-time average. In summary, it is necessary to construct an electron collision description in which we can account for the following ion features:

- *Ion short-time behavior is important.* This follows directly from Eqn. (2) if the transition energies are large. For smaller transition energies ion motion may be important. Here large transition energy $\hbar\omega$ is defined as having frequency ω greater than the ion plasma frequency.
- *Ions are relatively slow.* Due to their large mass, ions, including protons, move with velocity $v_i < v_e/40$ if the electron and ion temperatures are the same. In general, $v_i = v_e \sqrt{T_i m_e / T_e m_i}$.
- *Ions interact strongly.* Ions may be highly charged and move slowly; simple perturbative treatments (e.g., Born approximation) are typically not valid.

A SELF-CONSISTENT MODEL

A model is now presented in which ions are included in a calculation of electron impact processes. Care is taken to treat the ions in the manner discussed above. Estimates are made of the ionization potential depression (IPD) based on this model in the next section.

The atom-plasma Hamiltonian $H = H_0 + V$ can be written as

$$\begin{aligned} H_0 &= H_a + H_i + H_e + H_{ei} \\ V &= H_{ai} + H_{ae}, \end{aligned} \quad (3)$$

where H_a is the atomic Hamiltonian, H_i is the ion Hamiltonian, H_e is the electron Hamiltonian, H_{ei} is the electron-ion interaction Hamiltonian, H_{ai} is the atom-ion interaction Hamiltonian, and H_{ae} is the atom-electron Hamiltonian. The eigenstates of H_0 are defined by

$$H_0|\text{atom}_n\rangle|\text{plasma}_m\rangle = (E_n + E_m)|\text{atom}_n\rangle|\text{plasma}_m\rangle. \quad (4)$$

The probability that there has been a transition from some initial state $|i\rangle = |\text{atom}_n\rangle|\text{plasma}_m\rangle$ to final state $|f\rangle = |\text{atom}_p\rangle|\text{plasma}_q\rangle$ at time t later is given by

$$P(t) = |\langle f|U(t)|i\rangle|^2, \quad (5)$$

where the time evolution operator is given by

$$U(t) = \frac{1}{2\pi i} \oint_c dz e^{-izt} G(z). \quad (6)$$

The contour c encloses the real axis counterclockwise and $G(z) = (z - H)^{-1}$ is the Hamiltonian resolvent. The interesting matrix elements are thus those of $G(z)$.

It is possible to write a perturbation expansion for $G(z)$ in terms of the unperturbed resolvent $G_0(z) = (z - H_0)^{-1}$ as

$$G(z) = G_0(z) + G_0(z)V G_0(z) + G_0(z)V G_0(z)V G_0(z) + \dots \quad (7)$$

The first term vanishes if $i \neq f$ and the first order term in V describes transitions arising from the DSF between unperturbed levels². However, from the arguments given above, we must keep many higher order terms to properly treat the ion term H_{ai} which affects transitions arising from H_{ae} . Fortunately, it is possible to formally obtain the matrix elements $G_{fi}(z) \equiv \langle f|G(z)|i\rangle$ exactly thereby keeping terms to infinite order in the interaction V . The matrix element is written in terms of the level shift operator

$$R(z) = V + V \frac{Q}{Q(z - H)Q} V \quad (8)$$

as

$$G_{fi}(z) = \frac{R_{fi}}{(z - E_f - R_{ff})(z - E_i - R_{ii}) - R_{if}R_{fi}}, \quad (9)$$

where $Q = 1 - |f\rangle\langle f| - |i\rangle\langle i|$ is the projection onto states other than $|i\rangle$ and $|f\rangle$. Note that in Eqn. (8) the second term on the right contains a pole. This gives rise to an imaginary contribution to $R(z)$ in each of the matrix elements of $R(z)$ in Eqn. (9) and corresponds to a width of the state under consideration. Thus, in this description atomic states appear as resonance states near the discrete energies of the unperturbed atom. From this result we may form a modified Fermi Golden Rule for the transition rate between these resonance (shifted and broadened) states. Rather, we now estimate the IPDs based on the considerations so far, which would be used in a such a Fermi Golden Rule.

PREDICTIONS FOR IONIZATION POTENTIAL DEPRESSION

Consider now a simple model for electron impact ionization of a hydrogenic ion with nuclear charge Z . Note that the matrix element $G_{fi}(z)$ describes transitions arising from both electrons and ions since $V = H_{ai} + H_{ae}$. To isolate the electron impact contribution from the ion impact contribution, only H_{ae} is retained in $R_{fi}(z)$. This describes electron impact transitions in the presence of the electrons and ions which are kept in $R_{ff}(z)$ and $R_{ii}(z)$. (Similarly, we could keep H_{ai} in $R_{fi}(z)$ to describe ion impact processes in the presence of electrons and ions. In either case the $H_{ai} - H_{ae}$ interference term is being ignored, which is a consequence of treating the processes as distinct.) In what follows, again for simplicity, only the static term will be kept in $R_{ff}(z)$ and $R_{ii}(z)$.

A complication arises from the term H_{ei} , which couples the electron and ion subsystems. To obtain simple estimates for the level shifts, we include this coupling approximately by writing

$$\begin{aligned} H_{ae} + H_{ai} &\approx e^2 \int d^3r \frac{\delta n_e(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} - e \sum_i \Phi_i(|\mathbf{r} - \mathbf{r}_i|) \\ &\approx e^2 \int d^3r \frac{\delta n_e(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} - e\Phi(0) - e\mathbf{r} \cdot \nabla\Phi(0) - \frac{e}{2} \sum_{i,j} x_i x_j \frac{\partial^2 \Phi(0)}{\partial x_i \partial x_j} + \dots, \end{aligned} \quad (10)$$

where the $\Phi_i(|\mathbf{r} - \mathbf{r}_i|)$ are the potentials arising from statically (electron) screened ions and $\delta n_e(\mathbf{r}')$ represents electron density fluctuations. The first term will be retained in $R_{fi}(z)$ which isolates electron impact processes between levels. These levels are modified by both terms, but mostly the strong interactions of the screened ion terms, and are included in $R_{ff}(z)$ and $R_{ii}(z)$. For very slowly moving ions, H_i can be neglected and the ion configuration is the same for both the initial and final states. Since we don't know the given ion configuration, an average over ion configurations is taken at the end of the calculation. In the second equation, the total potential $\Phi(\mathbf{r})$ due to the screened ions has been expanded in a multipole expansion⁵ about small r . This approximate form is useful for *tightly bound* states and allows one to average over the microfield distribution $P(\epsilon)$. For less localized states, continuum states in particular, the full form of the interaction must be used. It is clear that, in either case, the ion microfield is largely responsible for the level shifts whereas the electrons represent a stochastic perturbation; we thus refer to this model as the Microfield Stochastic Model (MSM).

Consider now an ionization process from an $n = 2$ level to a plane wave state $|\mathbf{q}\rangle$ in an approximation where $R \approx V$. Ignoring the monopole term $-e\Phi(0)$, the first order energy correction to the bound state arises from the total electric field ϵ of the perturbing screened ions. Since the $n = 2$ level has angular momentum degeneracy, care must be taken in evaluating the energy level shift. This will, of course, give rise to the well-known Stark shifts⁶ of the bound state levels. Thus we see that the $n = 2$ sublevels *will not be shifted equally*: some levels will move toward the continuum (become less tightly bound) and others move away from the continuum (become more

tightly bound). Thus, for the $n = 2$ bound states,

$$E_i + R_{ii} \rightarrow -\frac{Z^2}{2n^2} \pm \frac{3n(n-1)}{2Z} \epsilon \quad (11)$$

$$\approx -\frac{Z^2}{8} \pm \frac{3}{Z} \left(\frac{4\pi}{3}\right)^{2/3} n_e^{2/3}, \quad (12)$$

where $\epsilon = -\nabla\Phi(0)$. (Atomic units will now be used.) In the second step a typical field strength of $\epsilon = 1/r_e^2$ has been used (r_e is the electron sphere radius). The $\pm$ energies refer to the upper and lower Stark states: the other two $n = 2$ levels are unshifted. For the final state we have

$$\begin{aligned} E_f + R_{ff} &\approx \frac{q^2}{2} - \langle \mathbf{q} | \sum_i \Phi_i(|\mathbf{r} - \mathbf{r}_i|) | \mathbf{q} \rangle \\ &\approx \frac{q^2}{2} - \frac{2\pi}{5} \left(\frac{3\bar{z}}{4\pi}\right)^{2/3} n_e^{1/3}, \end{aligned} \quad (13)$$

where ion-sphere potentials have been used as a model screened ion; the multipole expansion has not been used for the continuum state. (In general the potentials will also depend on temperature.) From these estimates, we find that the ionization potential I is given by

$$\begin{aligned} I &= E_f(q=0) - E_i \\ &= -\frac{2\pi}{5} \left(\frac{3\bar{z}}{4\pi}\right)^{2/3} n_e^{1/3} - \left[-\frac{Z^2}{8} \pm \frac{3}{Z} \left(\frac{4\pi}{3}\right)^{2/3} n_e^{2/3} \right]. \end{aligned} \quad (14)$$

Here $\bar{z}$ refers to the mean ion charge surrounding the target atom. This result can now be compared with the shift predicted by simply placing the atom in an ion-sphere cell. This gives an ionization potential I_{IS} , again for $n = 2$, of

$$I_{IS} = \frac{Z^2}{8} - \frac{(Z-1)^{2/3}}{2} \left(\frac{4\pi}{3}\right)^{1/3} n_e^{1/3}. \quad (15)$$

Comparisons of I and I_{IS} are given in Fig. 1 and Fig. 2 for argon and carbon, respectfully. In the argon calculation $Z = 18$ and $\bar{z} = 1$, which roughly corresponds to ICF conditions. It is clear that the simple ion-sphere screening model predicts a much larger IPD than the MSM model. In particular, note that the lower Stark state is nearly unshifted as a result of a near cancellation of the increased Stark binding and the lowering of the continuum. For such a state, we would not expect a large increase in the ionization rate. In the carbon calculation $Z = 6$ and $\bar{z} = 6$, which roughly corresponds to short-pulse laser experimental conditions. Again we see that the lower Stark state is mostly unshifted, even out to unrealistically large densities. Interestingly, the upper Stark state follows fairly closely to the simple ion-sphere prediction. It is important to note however, that it is only the upper Stark state which follows that prediction, which appears to be fortuitous since the argon case did not show this behavior.

In summary, a new model for treating collisional atomic processes has been presented. This model simultaneously and self-consistently treats electron and ion processes. For transitions frequencies above the ion plasma frequency, it is argued that little

ion motion occurs although the interaction is still strong; the ion microfield perturbs the atom. The electron impact processes may still be described by a DSF between levels of the perturbed atoms, suggesting the name Microfield Stochastic Model (MSM) for this method. Future work will be directed towards refining some of the approximations used here for application to realistic systems.

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Figure 1. Argon.

Figure 2. Carbon.