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Atomic excitation in strongly coupled plasmas

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Abstract. In dense plasmas atomic excitation rates arising from scattering between atoms and surrounding plasma particles are formulated on the basis of the equations of motion for density matrices in a stochastic potential. This model enables us to treat strong transitions for which the first-order perturbation theory does not apply. Within a decorrelation approximation, which enables one to break up the higher-order correlation functions of plasma density fluctuations into the products of binary correlation functions (i.e., dynamic structure factors), the interaction is effectively summed to infinite order. This method is applied to two-level atoms in hydrogen plasmas. It is thereby demonstrated that when the plasma density is sufficiently high, low-frequency ion-density fluctuations may cause coherent atomic excitation between close lying states. Such coherent excitation cannot be described by the conventional collisional rate equations based on the first-order perturbation theory.

1. STOCHASTIC PERTURBATION APPROACH

Atomic processes in high-temperature, high-density plasmas have been gaining more attention due to their relevance to the diagnostics of inertial-confinement fusion plasmas [1] or laser-produced plasmas such as dense aluminum [2]. Atoms embedded in such dense plasmas may undergo excitation due to the inelastic scattering with the surrounding plasma particles [3-5]; it is a non-thermal process, since the relevant plasma undergoes rapid time evolution.

Let us consider the excitation of an atom from state $|1\rangle \equiv |J_1, M_1\rangle$ (energy E_1) to state $|2\rangle \equiv |J_2, M_2\rangle$ (energy E_2) in a two-component plasma (TCP) consisting of electrons (charge number $Z_e = -1$, mass m_e, number density n_e) and ions (charge number Z_i, mass m_i, number density n_i) at temperature T, where $J_{1(2)}$ and $M_{1(2)}$ refer to the quantum numbers representing the total angular momentum of the atom and its projection onto a specified quantization axis, respectively. As long as the transition is weak, in the sense that first-order perturbation theory is valid, the excitation rates w_{21} can be calculated by Fermi's Golden Rule with the time-dependent perturbation field being expressed in terms of the plasma density fluctuations [3-5]:

$$w_{21} = 4(\alpha a_B c)^2 \frac{I_H}{\hbar\omega_{21}} \sum_{\sigma=e,i} \int d\boldsymbol{k}\, f(1,2;k) \frac{Z_\sigma^2}{k^2} S_{\sigma\sigma}(k,\omega_{21}) \ . \tag{1}$$

In this equation, $I_H = 13.606$ eV, α is the fine-structure constant, c is the speed of light, a_B is the Bohr radius, $\omega_{21} = (E_2 - E_1)/\hbar$ is the excitation frequency,

$$f(1,2;k) = \frac{2m_e\omega_{21}}{\hbar k^2} \frac{1}{2J_1+1} \sum_{M_1, M_2} \left| \langle J_2, M_2 | \exp(i\boldsymbol{k}\cdot\boldsymbol{r}) | J_1, M_1 \rangle \right|^2 \tag{2}$$

refers to the Bethe's generalized oscillator strength [6], and $S_{\sigma\sigma}(k,\omega)$ denotes the dynamic structure factor [7] for the plasma particles of species σ. Equation (1) implies that the excitation is induced predominantly by the plasma density fluctuations at $\omega = \omega_{21}$. In this paper we confine ourselves to the study of excitation due to electron-density fluctuations, so that only the terms with $\sigma = e$ will be retained hereafter. We further assume that the plasma temperature is sufficiently high so that $S_{ee}(k,\omega)$ can be evaluated with the random-phase approximation for classical TCP [7].

Previously, equation (1) was applied to the $2s_{1/2} \rightarrow 2p_{3/2}$ fine-structure transition of Ne^{9+} in a hydrogen TCP [4]. It was thereby shown that the excitation rates begin to increase divergently as n_e exceeds about $10^{22}cm^{-3}$. We expect that the first-order perturbation theory is not valid for such strong transitions between close-lying states at high plasma densities. It can also be shown [4] that transitions due to ion-density fluctuations are typically too strong to be treated by perturbation theories.

2. DENSITY-MATRIX FORMALISM

We present a non-perturbative theory of atomic excitation in dense plasmas based on the density-matrix formalism [8,9]. Let us consider a hydrogen-like atom which has nuclear charge Ze and two non-degenerate atomic levels $|1\rangle$ and $|2\rangle$ specified by the wave functions $\varphi_1(\boldsymbol{r})$ and $\varphi_2(\boldsymbol{r})$, respectively. The density matrix $\rho = \{\rho_{ij}\}$ (with $i, j = 1,2$ denoting the level indices) obeys the equation of motion, $d\rho/dt = (i/\hbar)(\rho H - H\rho)$, where the total Hamiltonian H is the sum of the atomic part H_{atom} and the fluctuating potential $V(\boldsymbol{r},t) = \sum_j e^2/|\boldsymbol{r} - \boldsymbol{r}_j(t)|$ produced by all the plasma electrons ($j = 1,2,...,N$) at positions $\boldsymbol{r}_j(t)$. We assume that initially ($t = 0$) the atoms are in the lower states $|1\rangle$, that is, $\rho_{11}(0) = 1$ and $\rho_{22}(0) = 0$. The equation of motion can then be rewritten in the form of an integral equation,

$$\langle\rho_{22}(t)\rangle = \frac{1}{\hbar^2}\int d\boldsymbol{r}\,\varphi_1^*(\boldsymbol{r})\,\varphi_2(\boldsymbol{r})\int d\boldsymbol{r}'\,\varphi_2^*(\boldsymbol{r}')\,\varphi_1(\boldsymbol{r}') \times \int_0^t dt' \int_0^{t'} dt''\,\left\langle V(\boldsymbol{r},t')\,V(\boldsymbol{r}',t'')\,[1-2\rho_{22}(t'')]\right\rangle \exp[i\omega_{21}(t''-t')] + \text{c.c.}, \qquad (3)$$

which governs the average time evolution of the excited-level populations $\langle\rho_{22}(t)\rangle$. Here, $\langle\cdots\rangle$ represents the statistical average over the plasma states and "c.c." stands for the complex conjugate. The term $\langle V(\boldsymbol{r},t')V(\boldsymbol{r}',t'')\rho_{22}(t'')\rangle$ on the right-hand side physically describes the effect of back reaction which is neglected in the first-order perturbation theory. To evaluate this term, we adopt a decorrelation approximation [10], $\langle V(\boldsymbol{r},t')V(\boldsymbol{r}',t'')\rho_{22}(t'')\rangle \approx \langle V(\boldsymbol{r},t')V(\boldsymbol{r}',t'')\rangle\langle\rho_{22}(t'')\rangle$, which breaks up the many-body correlation function into the products of binary correlation functions, i.e., $S_{ee}(k,\omega)$. The integral equation (3) can then be solved analytically with the aid of the Laplace transform,

$$\langle\rho_{22}(t)\rangle = \frac{1}{2\pi i}\int_{\varepsilon-i\infty}^{\varepsilon+i\infty} ds\,\exp(st)\,\frac{1}{s}\,\frac{X(s)}{1+2X(s)}, \qquad (4)$$

where ε denotes a positive infinitesimal; the quantity

$$X(s) = \frac{2Z\omega_e}{\pi}\,\frac{I_H}{\hbar\omega_{21}}\int_{-\infty}^{\infty} d\omega\,D(\omega)\,\frac{2}{s^2+(\omega-\omega_{21})^2} \qquad (5)$$

measures the strength of the transition with $\omega_e = \sqrt{4\pi n_e e^2/m_e}$ being the electron plasma frequency, and

$$D(\omega) = \frac{a_B\omega_e}{Zn_e}\int_0^{\infty} dk\,f(1,2;k)\,S_{ee}(k,\omega) \qquad (6)$$

describes the frequency spectrum of electron-density fluctuations.

For a weak transition such that $X(s) \ll 1$, we may set $X(s)/[1+2X(s)] \approx X(s)$; equation (4) then reproduces the result (1) of the first-order perturbation theory, $\lim_{t\to\infty} d\langle\rho_{22}(t)\rangle/dt = \lim_{s\to 0} sX(s) = w_{21}$. Equation (4) applies also to a strong coherent field such that $D(\omega) = \lim_{\Delta\to 0} D_0\Delta/[(\omega-\omega_0)^2+\Delta^2]$; in this

case, the Rabi oscillation [8,9], $\langle\rho_{22}(t)\rangle = (\omega_{\text{Rabi}}^2/2\Omega^2)[1-\cos(\Omega t)]$, is correctly reproduced, with $\Omega = [(\omega_{21}-\omega_0)^2 + \omega_{\text{Rabi}}^2]^{1/2}$ and $\omega_{\text{Rabi}} = [8ZD_0(I_H/\hbar\omega_{21})]^{1/2}\omega_e$ denoting the flopping frequency and the Rabi frequency, respectively. We therefore expect that the decorrelation approximation may offer a reasonable description of the atomic excitation of arbitrary strength.

Formula (4) may further be extended to degenerate two-level systems as

$$\langle\rho_{22}(t)\rangle = \frac{1}{2\pi i}\int_{\varepsilon-i\infty}^{\varepsilon+i\infty} ds\, \exp(st)\,\frac{1}{s}\,\frac{X(s)}{1+(\eta+1)\,X(s)}, \tag{7}$$

where the phenomenological parameter $\eta = (2J_1+1)(2J_2+1)^{-1}\exp(\hbar\omega_{21}/k_BT)$ ensures that $\langle\rho_{22}(t)\rangle$ eventually approaches the steady-state population at thermal equilibrium [11].

3. NUMERICAL EXAMPLES

For a concrete example, we consider the $2s_{1/2} \rightarrow 2p_{3/2}$ transition of Ne^{9+} ($\hbar\omega_{21}$ = 0.45 eV) and Ar^{17+} ($\hbar\omega_{21}$ = 4.75 eV) in a hydrogen TCP of n_e (= n_i) = 10^{23} cm^{-3} and k_BT = 340 eV. The density-fluctuation spectrum $D(\omega)$ (equation (6)) is illustrated in Figure 1a. We find that the spectrum exhibits a relatively flat distribution for $\omega/\omega_e > 0.1$, which implies that the plasma field in this frequency regime is incoherent. The origin of this incoherent field is the fluctuations due to random thermal motions of the electrons. On the other hand, since $m_i \gg m_e$, ions perform thermal motions at frequencies much smaller than those for the electrons. Due to the electron-ion correlation, the electrons follow the low-frequency ion motions and produce a coherent field as indicated by the sharp peak in the spectrum at $\omega = 0$. Since the excitation frequency of Ne^{9+} ions, $\omega_{21}/\omega_e = 0.038$, lies within the width of this peak, their excitation is strongly influenced by the coherent field. On the other hand, the excitation frequency of Ar^{17+} ions, $\omega_{21}/\omega_e = 0.405$, is relatively large, hence their excitation is predominantly caused by the incoherent field due to electron thermal fluctuations.

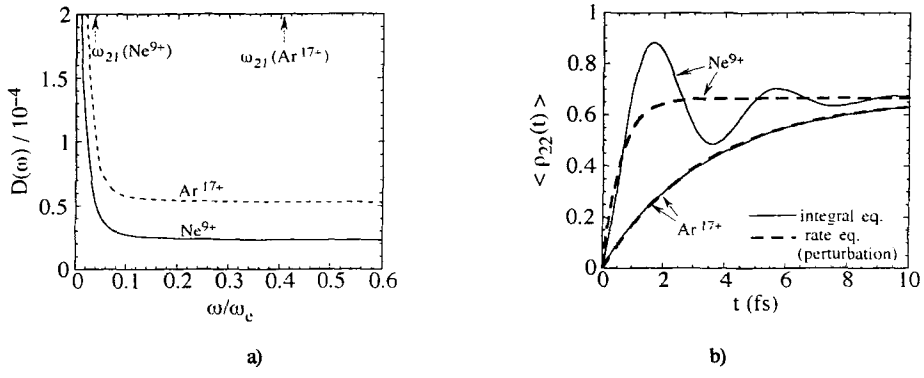

Figure 1. a) Frequency spectrum of the electron-density fluctuations. b) Time evolution of the excited-level populations.

In Figure 1b, the resultant time evolution of the excited-level populations, computed by equation (7), are plotted and are compared with the predictions of the collisional rate equations [11], $\langle\rho_{22}(t)\rangle = (\eta+1)^{-1}\{1-\exp[-(\eta+1)w_{21}t]\}$, where the rate coefficient w_{21} is evaluated in accordance with equation (1) based on first-order perturbation theory. For Ne^{9+} ions, we predict a transient oscillation of the population caused by coherent excitation, as indicated by the solid curve. This is a phenomenon similar to the Rabi oscillation of an atom interacting with a laser field [8,9]. Such a coherent excitation cannot be correctly described by the conventional rate equations, which lead to the result shown by the dashed

curve. On the other hand, we observe excellent agreement between the two sets of calculations for the case of Ar^{17+}, which means that the first-order perturbation theory is valid for such a weak, incoherent excitation. Good agreement has also been confirmed for Ne^{9+} at lower density, $n_e = 10^{20}cm^{-3}$, where coherent excitation does not occur.

4. ISSUES FOR FUTURE STUDY

As we see in Figure 1b, the population floppings may be observed during a very short time (typically a few femtoseconds) before the atom reaches thermal equilibrium with the surrounding plasma. It remains to be seen whether such transient oscillations can actually be detected through spectroscopic experiments.

In this study, we have considered a transition between two particular levels and have neglected all the other levels, including the continuum. Density-matrix analysis of the transitions involving many-level atoms [12] is one of the outstanding issues. Also we want to improve the theory by incorporating the modification of atomic energy levels due to the screening by surrounding plasma [5], and the Stark splitting produced by the ion microfields [11].

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