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Finite-temperature random-phase approximation for spectroscopic properties of neon plasmas

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A finite-temperature random-phase approximation (FTRPA) is applied to calculate oscillator strengths for excitations in hot and dense plasmas. Application of the FTRPA provides a convenient, self-consistent method with which to explore coupled-channel effects of excited electrons in a dense plasma. We present FTRPA calculations that include coupled-channel effects. The inclusion of these effects is shown to cause significant differences in the oscillator strength for a prototypical case of ^1P excitation in neon when compared with single-channel and with average-atom calculations. Trends as a function of temperature and density are also discussed.

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Radiative properties of hot dense plasmas, such as those that occur in inertial confinement fusion devices, astrophysical environments, and high-power laser blowoffs, remain a subject of much current interest [1]. There are two main approaches to this problem. The first treats the atomic physics in a detailed way and incorporates the plasma effects perturbatively. The second attempts to include the plasma effects on an equal footing with the atomic processes, but its typical implementation imposes severe approximations on the atomic physics. For this second approach, one of the earliest models was the "average-atom" (AA), introduced by Rozsnyai [2] who performed a series of calculations for the radiative properties of hot, dense plasmas [3]. Although introduced more than 30 years ago, the AA model still provides one of the primary vehicles for calculating such radiative properties [4,5]. For an atom perturbed by its plasma surroundings, the AA model assumes that the quantum-mechanical states of the atom can be described by those of a selected ion and the electrons in its immediate surroundings, all within a "neutrality sphere." The effect of the other ions and electrons is taken into account through a boundary condition (ion-sphere model) or by the addition of an extra potential (ion-correlation model). The basic atomic component of the formulation proceeds through density functional theory [5] with the fractional occupation numbers of the AA orbitals becoming density- and temperature-dependent through the Fermi-Dirac distribution function.

This report describes two important ways of improving calculations of the atomic processes within the AA framework. The first includes coupling between the atomic orbitals. Previous implementations [3–5] of the AA approach have been independent-electron models. The second corrects the potential for the excited electron, which in the AA model does not have the proper form since the removal of a corresponding core electron has not been performed [6]. Use of the finite-temperature random-phase-approximation (FTRPA) can correct both of these deficiencies [7]. The FTRPA approach has been formulated as a system of coupled, partial integrodifferential equations [6]. Individual channels are defined by the removal of an electron from a (partially) occupied orbital. Thus independent-electron "excited states" can be created by the removal of electrons from different occupied orbitals and placing them in different "excited" orbitals. In the FTRPA these "individually excited states" (associated with different channels) interact through channel coupling. Previous studies [8,9] have used the single-channel approximation of FTRPA to compute spectroscopic properties of dense He and Li plasmas. In this paper, we solve a coupled-channel form of the FTRPA equations and examine spectral properties of a prototypical dense Ne plasma where coupling can be expected to be more significant than in He or Li.

In this work we shall use the ion-correlation model, which assumes a central nucleus surrounded by electrons with the electrons located in a large box. Inside the box, there is one central ion and additional ions correlated to it. Their correlated motion results in a potential that is added to that created by the central nucleus. The total charge of electrons is compensated by the charge of the central ion plus the total charge of the additional ions that are distributed inside the large box. The electrons are assumed to be in thermal equilibrium with a large heat bath of given temperature, and their motion is determined by one central potential. The starting point for the description of the FTRPA is the finite-temperature (or thermal) Hartree-Fock (FTHF) approximation, which uses the Hartree-Fock potential [8,9]

$$
\begin{aligned}
\Sigma^{\mathrm{HF}}(x,x') = & -\frac{Z}{r}\delta(x-x') + V_{\mathrm{ext}}(r)\,\delta(x-x') \\
& + \delta(x-x')\int V(\vec{r}-\vec{r}\,')\rho_{\mathrm{HF}}(x_1,x_1)dx_1 \\
& - V(\vec{r}-\vec{r}\,')\rho_{\mathrm{HF}}(x',x),
\end{aligned}
\tag{1}
$$

where $x \equiv (\vec{r},\sigma)$ refers to both the spatial ($\vec{r}$) and spin (σ) coordinates of the electrons, $r \equiv |\vec{r}|$, $V(\vec{r}-\vec{r}\,') = e^2/|\vec{r}-\vec{r}\,'|$ is the Coulomb interaction potential, Z is the nuclear charge, $V_{\mathrm{ext}}(r)$ is the potential originating from the other ions, and $\rho_{\mathrm{HF}}(x,x')$ is the HF density matrix of the electrons.

The aim of the FTRPA is to calculate the linear response function, which is obtained in the form [6]

$$L^{\mathrm{FTRPA}}(x_1,x_2,x_1';z) = \sum_n \frac{\chi_n(x_1,x_1')\chi_n^*(x_2)}{\omega_n - z}, \tag{2}$$

where $\chi_n(x) \equiv \chi_n(x,x)$. In Eq. (2) ω_n can be interpreted as the "excitation energy" of the finite-temperature system and $\chi_n(x)$ as the transition density from the initial (finite-temperature) state to the nth excited state of the system. $\chi_n(x,x')$ can be expanded on the basis of HF eigenfunctions for which the radial components are given in terms of the orbitals $P_{\nu l}$ and the channel functions $P^{LM}_{(\nu' l')nl}$. By following the developments laid out in detail in Refs. [6,8,9] we arrive at a coupled system of integrodifferential equations for these FTRPA channel functions

$$\begin{aligned}(H^{\mathrm{HF}}_{l_i} - \epsilon_{\nu_j l_j} - \omega_{nLM})P^{LM}_{(\nu_j l_j)nl_i}(r) = {} & -n_{\nu_j l_j}\Bigg\{\sum_{l,l',\lambda} B_\lambda^{l_i l_j,ll'} \sum_{\nu'} \int_0^\infty dr' v_\lambda(r,r') P^{LM}_{(\nu' l')nl}(r') P_{\nu' l'}(r') P_{\nu_j l_j}(r) \\ & - \sum_{l,l',\lambda} A_\lambda^{l_i l_j,ll'} \sum_{\nu'} \int_0^\infty dr' v_\lambda(r,r') P_{\nu_j l_j}(r') P_{\nu' l'}(r') P^{LM}_{(\nu' l')nl}(r)\Bigg\} - \sum_{\nu_i} n_{\nu_i l_i} P_{\nu_i l_i}(r) I^{nl_i}_{\nu_i l_i,\nu_j l_j},\end{aligned} \tag{3}$$

where $A_\lambda^{l_i l_j,ll'}$ and $B_\lambda^{l_i l_j,ll'}$ are angular factors defined in Ref. [8] and $I^{nl_i}_{\nu_i l_i,\nu_j l_j}$ can be interpreted as an orthogonality term that controls the overlap of the $P^{LM}_{(\nu_j l_j)nl_i}(r)$ excited orbital with those $P_{\nu_i l_i}(r)$ core electron orbitals that are incorporated in that sum [8]. The quantity $v_\lambda(r_1,r_2)$ is the multipole component in the expansion of the Coulomb potential. This equation assumes that the HF orbitals factorize into radial and angular components, since the external (density- and temperature-dependent) potential in which the electrons move is spherically symmetric. The radial functions $P_{\nu_i l_i}(r)$ are eigenfunctions of the HF operator $H^{\mathrm{HF}}_{l_i}$, which includes the usual kinetic energy operator and centrifugal terms, as well as the HF potential defined in Eq. (1).

Equation (3) presents the coupled-channel FTRPA equations in differential form. These equations are coupled in two ways, in the $\nu' l'$ and l indices. The channel indices $\nu' l'$ represent the principal and orbital angular momentum quantum numbers of the HF orbitals occupied with fractional occupation numbers from which an electron is removed and placed into an "excited orbital." The component index l represents an angular momentum component in the expansion of the excited orbital; every excited orbital (in principle) can have $l=0,1,2,\ldots$ components. In the calculations presented here for neon plasmas, we assume that two channel indices $2s,2p$ and three component indices $3p,3s,3d$ are important.

The diagonal parts of the first two summations on the right-hand side of Eq. (3) (i.e., the terms for which $\nu' l' = \nu_j l_j$ and $l = l_i$) can be interpreted as representing the exchange and direct contributions of the $P_{\nu_j l_j}$ orbital to the HF potential $\Sigma^{\mathrm{HF}}_{l_i}$, respectively. [The potential $\Sigma^{\mathrm{HF}}_{l_i}$ is the radial component in the expansion of the HF potential $\Sigma^{\mathrm{HF}}(x,x')$ defined in Eq. (1).] Moving these two terms to the left-hand side of Eq. (3) results in a subtraction that creates exactly the proper potential for the excited electron denoted as $\Sigma^{N-n_{\nu_j l_j}}_{l_i}$, with N designating the number of bound electrons and $n_{\nu_j l_j}$ representing the (fractional) occupation number of the initial orbital to be removed. In the zero-temperature case, this subtraction produces the well-known V^{N-1} potential that is suitable for calculating excited orbitals. The off-diagonal terms contained within the first two summations on the right-hand side of Eq. (3) (for which $\nu' l' \neq \nu_j l_j$ and/or $l \neq l_i$) represent the coupling between the various channel and component functions. In order to be able to compare with orbitals and energies from the AA model [10], we introduce on the left-hand side of Eq. (3) the local-exchange approximation, which results in replacement of the $\Sigma^{\mathrm{HF}}_{l_i}$ by the appropriate AA potential $\Sigma^{\mathrm{AA}}_{l_i}$; the $P_{\nu_j l_j}$ orbitals then represent AA, rather than HF, eigenfunctions.

In this work, we use the linear algebraic method [11] to solve the set of coupled differential equations in Eq. (3). This method has been used with much success in electron-atom and electron-molecule scattering [12] and has been adapted here for handling coupled-channel eigenvalue problems. A detailed description of the computational techniques applied in the linear algebraic method has already been given [12]. The linear algebraic method converts the set of coupled differential equations into coupled integral equations and proceeds by introducing a quadrature to the integrals and a discrete mesh to the functions and solving the equations on a numerical grid. By further rewriting the equations in matrix form, standard numerical methods can be applied to solve the set of equations in an efficient manner.

We calculate optical oscillator strengths for a neon plasma as this is a convenient quantity for examining the effects of coupling between the channels. Following Csanak and Meneses [8], and after angular and spin analyses, we can obtain the dipole matrix element as

$$\begin{aligned}D_{nLM} = {} & \sum_{l,l'} \begin{pmatrix} 1 & l & l' \\ 0 & 0 & 0 \end{pmatrix} \left[\frac{2(2l+1)(2l'+1)}{3}\right]^{1/2} (-1)^{l-l'} \delta_{L1}\delta_{M0} \\ & \times \int_0^\infty dr_1 r_1 \sum_{\nu'} P^{L=1,M=0}_{(\nu' l')nl}(r_1) P_{\nu' l'}(r_1),\end{aligned} \tag{4}$$

where we restrict ourselves to dipole transitions from the ^1S

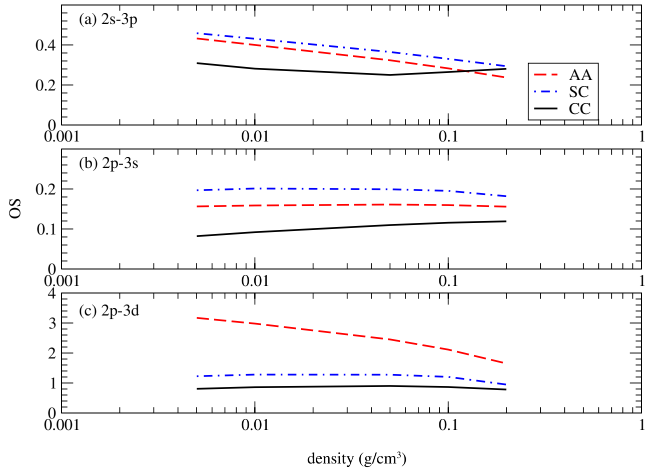

FIG. 1. (Color online) Oscillator strengths for neon ${}^1P$ excitations as a function of density, at a temperature of 25 eV, using our coupled-channel approach. We show results for three transitions: (a) the case where the $3p$ component is dominant, (b) the case where the $3s$ component is dominant, and (c) the case where the $3d$ component is dominant. We compare AA results (dashed lines) with SC FTRPA results (dot-dashed lines) and with CC FTRPA results (solid lines).

ground state to singlet excited states, i.e., to ${}^1P$ states. We define the optical oscillator strength (OS) as

$$\mathrm{OS} = 2\omega_{nLM}|D_{nLM}|^2, \tag{5}$$

where ω_{nLM} is the transition energy in atomic units from the ground state of a neon atom.

We now turn to an exploration of the solution of the FTRPA equations for systems at finite temperatures and densities. Here, we examine the effects of channel and component coupling in neon plasmas ($Z=10$) at various temperatures and densities. The APATHY code [10] was used to generate AA orbitals and potentials for different density/temperature pairs. The APATHY code solves the AA equations fully quantum mechanically for both the bound and continuum wavefunctions used in constructing the electron density. These equations are solved self-consistently, with the correlation between the ions computed using hypernetted chain theory [10].

We present oscillator strengths for the solutions of the FTRPA equations for ${}^1P$ excited states where the $3p$, $3s$, and $3d$ components are dominant, for a neon plasma at various densities for a temperature of 25 eV, in Fig. 1. We compare the single-channel (SC) FTRPA results with the coupled-channel (CC) FTRPA results and with the oscillator strength computed using the AA "excited" orbitals. In the CC FTRPA equations discussed here, the "hole" $2s$ and $2p$ channels are coupled (i.e., $\nu_j l_j \equiv 2s,2p$) and the "particle" $3p$, $3s$, and $3d$ components are also coupled (i.e., $nl_i \equiv 3p,3s,3d$) through the right-hand side of Eq. (3). In the SC FTRPA equations, the channel-coupling terms are omitted from the right-hand side of Eq. (3). The AA "excited" orbitals are obtained directly from APATHY and, even though as previously discussed, these "excited" orbitals are computed in an incorrect potential, we show the resulting oscillator strengths here for comparison purposes.

First of all we remark that the absolute energies of the CC $3l$ excited orbitals (not shown) are significantly lower than the absolute energies of the $3l$ AA "excited" orbitals. We recall that the FTRPA excited orbitals are computed in the proper $\Sigma^{N-n_{\nu_j l_j}}$ potential for the excited electron, whereas the AA "excited" orbitals are computed in the unphysical Σ^N potential. Consequently, the FTRPA excited states are more tightly bound than the AA "excited" orbitals. This difference results in transition energies for the AA calculation which are typically higher than the FTRPA transition energies by around 5–10 %.

The oscillator strengths from the AA calculations fall between those from the SC and CC FTRPA calculations for the transitions where the $3p$ and $3s$ components dominate [Figs. 1(a) and 1(b)]. The CC calculations differ from the SC calculations by between 20–60 % for these cases. For the transition where the $3d$ component dominates, the AA calculations are significantly higher than the SC and CC FTRPA calculations. The SC and CC calculations differ by around 50% over the density range considered.

As the density is increased for a given temperature, the excited-state energy increases due to pressure ionization. The resulting transition energy between the two bound states also decreases as the density is increased; this behavior is a result of the bound states becoming squeezed closer together at higher densities. The oscillator strength for the CC solution in which the $3p$ component is dominant trends slightly downwards as the density is increased until around 0.05 g/cm^3, and then increases. The SC and AA solutions trend uniformly downwards. For the case in which the $3s$ component is dominant, the CC calculations for the oscillator strength increase linearly with density. For the solution in which the $3d$ component is dominant, the oscillator strengths obtained from both the SC and CC solutions again increase slowly with density, until about 0.05 g/cc, where the oscillator strength starts to decrease somewhat. The AA solution strongly decreases with density. As the density is further increased, the excited state will eventually move into the continuum, and the oscillator strength will then drop to zero.

In Fig. 2 we show the oscillator strengths as a function of temperature, and at a constant density of 0.01 g/cc. We again compare the oscillator strengths calculated from the SC FTRPA equations with those from the full CC FTRPA equations and with those using the AA "excited" orbitals. As the temperature is increased, the excited-state orbitals become more tightly bound. This behavior is probably due to the increase in the number of free electrons as the temperature is increased, so that the remaining bound electrons feel more of the attractive nuclear potential. The transition energy also increases with temperature since the core ("ground state") electrons will be more tightly bound than the excited electrons. Again, the oscillator strengths for all possible $3l$ solutions display different trends. The oscillator strength for the dominant $3p$ component transition generally increases with temperature, for the SC, CC, and AA solutions. The oscillator strengths corresponding to the solutions in which the $3s$ component is dominant and in which the $3d$ component is dominant decrease with increasing temperature, for all these

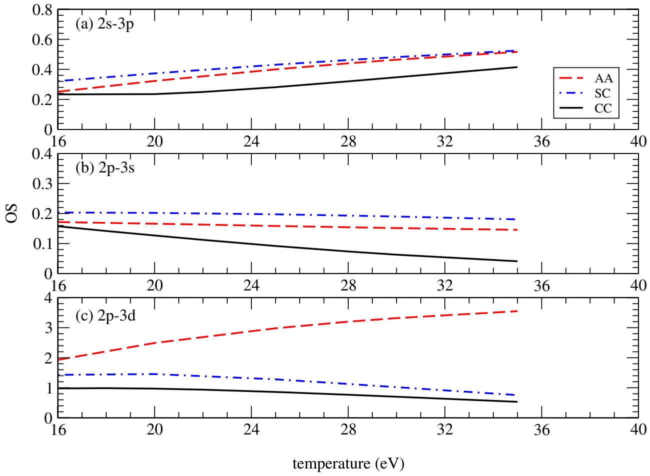

FIG. 2. (Color online) Oscillator strengths for neon ^1P excitations as a function of temperature, at a density of 0.01 g/cm^3, using our coupled-channel approach. The calculations and transitions shown are for the same cases as in Fig. 1.

calculations, except for the AA, $3d$ results, where the trend is strongly increasing. For the solution in which the $3s$ component is dominant, the difference between the SC and CC results increases as the temperature is raised, until the difference at the highest temperature considered (35 eV) is almost 70%. These differences further underscore the importance of including channel coupling in the calculations. The differences between the SC and CC solutions for the other cases are smaller in magnitude, around 20 or 30 %, and are fairly constant over the range of temperatures.

Finally we note that the SC transition energies are very similar to the CC transition energies, for all temperature and density cases, although the oscillator strengths can be quite different, due to differences in the component functions. This behavior implies that CC effects on any spectra will be limited to the magnitude of spectral lines, rather than to their position. However, it is clear that FTRPA calculations will result in differing positions of spectral lines when compared with AA calculations. We have also shown that AA calculations can result in oscillator strengths which can be markedly different from oscillator strengths from FTRPA calculations, and which also can display different trends as functions of plasma density and/or temperature.

In this paper we have extended previous work [6,8,9] on the finite-temperature random-phase approximation (FTRPA) by including channel-coupling effects in solving the FTRPA equations. We have used the linear-algebraic method [11] for the solution of the coupled equations, which relies upon the AA code APATHY [10] to generate the temperature- and density-dependent AA orbitals. We have presented coupled-channel oscillator strengths for neon plasmas at various temperatures and densities. The effect of channel coupling on these quantities was observed to be significant. In future work we intend to explore the possibility of using a fully nonlocal Hartree-Fock exchange potential in the finite-temperature formalism instead of the local-exchange approximation used currently in the AA model.

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