Near-LTE linear response calculations with a collisional-radiative model for He-like Al ions

Richard M. More
Lawrence Livermore National Laboratory
Livermore, California, USA 94550

Takako Kato
National Institute for Fusion Science
Toki, Gifu, 509-52, Japan

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Richard More,
Lawrence Livermore National Laboratory
Livermore, California, 94550,

and

Takako Kato,
National Institute for Fusion Science
Toki, Gifu, 509-52, Japan

ABSTRACT

We investigate the non-equilibrium atomic kinetics using a collisional-radiative (CR) model modified to include line absorption. Steady-state emission is calculated for He-like aluminum ions immersed in a specified radiation field having fixed deviations from a Planck spectrum. The net emission is interpreted in terms of NLTE population changes. The calculation provides an NLTE response matrix, and in agreement with a general relation of non-equilibrium thermodynamics, the response matrix is symmetric. We compute the response matrix for 1% and 50% changes in the photon temperature and find linear response over a surprisingly large range.

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While the opacity of dense plasmas such as stellar interiors is usually studied using LTE (local thermodynamic equilibrium) methods, low-density plasmas such as solar corona or tokamak plasmas require NLTE (non-equilibrium) kinetic models. Intermediate plasmas including laser-produced plasmas, laser hohlraums, Z-pinchs and divertor plasmas combine aspects of high density, non-LTE and a significant radiation environment.

This paper examines non-LTE atomic kinetics using methods of nonequilibrium thermodynamics.[1,2,3] We adapt the existing collisional-radiative (CR) model of Fujimoto and Kato[5] to near-LTE conditions and verify the applicability of nonequilibrium thermodynamics by finding a symmetric response matrix.

We consider an ion in the presence of radiation which is approximately a blackbody field at the temperature of the free electrons. The difference between the actual radiation and the black-body field causes nonequilibrium populations of excited states and leads to a net difference of emission and absorption rates. The difference of emission and absorption at frequency ν is a function of the deviation from the black-body spectrum at frequency ν' and can be described by a response matrix $R_{\nu,\nu'}$.

According to nonequilibrium thermodynamics, the near-LTE steady-state is constrained by general requirements of energy conservation, by the principle of minimum entropy production, and by symmetry relations (Onsager relations) which require that the linear response function $R_{\nu,\nu'}$ should be symmetric.

The relevance of thermodynamics to atomic kinetics can easily be understood by examining a simple thought experiment, the “NLTE reaction box”, (see Figure 1) defined in analogy to well-known conceptual pictures of chemical reactions.[3] Thermodynamic formulas for the reaction box are easily derived but here we simply point out that the thought experiment invokes the usual NLTE calculation performed by an atomic kinetics model with a specified local radiation environment. The thermodynamic analysis gives an equation for the entropy production in the nonequilibrium steady state and helps identify the symmetric response function.

At densities high enough so that electron rates greatly exceed radiative rates, the deviations from LTE should be small, as shown in previous general studies of NLTE kinetics by Klapisch[5], Salzmann[6] and Fujimoto[7]. In this work we study the effect of radiation, which can force the ion to LTE or can induce various non-LTE populations.

We consider helium-like aluminum ions at $N_e = 10^{20} \text{ cm}^{-3}$. The free electrons have a fixed temperature $T_e = 150 \text{ eV}$ and the isotropic radiation field has a spectrum I_ν characterized by the number of photons per mode, n_ν . In complete equilibrium n_ν is the Bose-Einstein function,

$$n_\nu^0 = [\exp(h\nu/kT_e) - 1]^{-1} \quad (1a)$$

We study deviations from LTE induced by the difference $(n_\nu - n_\nu^0)$. In order to generate the response matrix, a nonequilibrium radiation spectrum is defined by altering n_ν for one line, using

$$n_\nu = [\exp(h\nu/k(T_e + \delta T_\nu)) - 1]^{-1} \quad (1b)$$

The CR model includes spontaneous emission, described by the rate for radiative transitions $i \rightarrow j$, (i is the upper level, j is the lower level), $(dN_i/dt)_R$. This is the rate for the transition $i \rightarrow j$, but of course initial or final states may be connected to other states (k , l , etc.). N_i , N_j are populations (cm^{-3}) of initial and final states. The radiative rates are added to collisional rates. We extend the CR model by adding stimulated emission and absorption for each transition,

$$(dN_i/dt)_R = - A_{ij} N_i (n_\nu + 1) + A_{ji} N_j n_\nu \quad (2)$$

The absorption is written using an absorption rate coefficient,

$$A_{ji} = (g_i/g_j) A_{ij} \quad (3)$$

Here g_i , g_j are the degeneracies of upper and lower levels.

Electron-impact (collisional) rates for excitation and de-excitation obey detailed balance and we do not change these

collisional rates. Electron-impact ionization to and 3-body recombination to/from the hydrogen-like ion are included, but recombination to the Li-like ion is omitted. The population density of ground-state He-like ions is held fixed. Solution of the collisional-radiative model determines steady-state populations of He-like excited states. The original CR model also includes radiative and dielectronic recombination but for simplicity these processes have been suppressed.

To construct a 5x5 response matrix we perform 5 different NLTE calculations. For each calculation we vary $n_{\nu'}$ for one specific line ν' (Table I). The variation is induced by the photon temperature change $\delta T_{\nu'}$. All other lines interact with the black-body spectrum n^0_{ν} . For each line, in NLTE there is a net radiated power,

$$dE_{ij}/dt = h\nu [A_{ij} N_i (n_{\nu} + 1) - A_{ji} N_j n_{\nu}] \quad (4)$$

This rate can be positive (emission) or negative (absorption). The quantity in Eq. (4) is zero in LTE and should be proportional to $\delta T_{\nu'}$ for small perturbations. In NLTE there is emission or absorption for every line, but we examine only the same 5 lines to make a 5x5 response matrix. The linear response matrix is defined by

$$R_{\nu,\nu'} = h\nu [A_{ij} N_i (n_{\nu} + 1) - A_{ji} N_j n_{\nu}]/\delta T_{\nu'} \quad (5)$$

Then $R_{\nu,\nu'}$ is the net emission from line ν ($i \rightarrow j$) induced by a temperature change $\delta T_{\nu'}$ applied to the line ($i' \rightarrow j'$) denoted ν' . According to nonequilibrium thermodynamics $R_{\nu,\nu'}$ should be a symmetric matrix.

An important question concerns the range of linear response. For large perturbations $dT_{\nu'}$, the photon population $n_{\nu'}$ changes in a nonlinear way, especially for lines with $h\nu' \gg kT_e$. We find a larger range of "linear response" for a modified matrix,

$$RL_{\nu,\nu'} = R_{\nu,\nu'} ([\partial n_{\nu'}^0 / \partial T] \cdot \delta T_{\nu'} / [n_{\nu'} - n_{\nu'}^0]) \quad (6)$$

In Eq. (6) the extra factor involves the perturbed frequency. The extra factor is unity for small perturbations so the modified response matrix is still symmetric in that case. However $RL_{\nu,\nu'}$ remains constant for larger perturbations.

Because hydrogenlike ions are coupled through collisional ionization and recombination, it is necessary to use the appropriate ratio of helium-like and hydrogen-like ions. (Coupling to lithium-like ions is neglected.) This is done by finding the steady state solution of the rate equations, i.e., by using excited-state populations to determine effective ionization and recombination rate coefficients which determine the ratio of He-like and H-like ions.

Table I defines the transitions studied here. These transitions are in the singlet spectrum but the calculation includes couplings to triplet states. Table II gives the matrix $RL_{\nu,\nu'}$ calculated as the average of results for small temperature changes of +1% and -1%. Table III gives the response matrix $RL_{\nu,\nu'}$ for radiation temperature perturbations of $\pm 50\%$.

The linear-response matrix in Table II is accurately symmetric. Table III shows the range of linear response. For the He β transition a 50% increase in photon temperature raises the photon flux by a factor sixty, while a 50% decrease reduces n_{ν} by a factor 10^5 . These large changes only produce small changes in the response function $RL_{\nu,\nu'}$. The range of linear response is surprisingly large.

To interpret the response function, we consider the first row of the matrix in Table II. For this row $n_{\nu'}$ is increased relative to the black-body function for the transition from the $1s^2$ groundstate to the $1s2p^1P$ excited state. Absorption of the extra radiation produces a negative diagonal matrix-element. This absorption decreases the groundstate population and increases the $(1s2p^1P)$ excited-state population.

Since the groundstate population is reduced with respect to LTE, there is net emission for the line $1s3p^1P \rightarrow 1s^2$. However the

population of $1s2p\ ^1P$ is higher than LTE, and so there is net absorption for transitions $1s2p\ ^1S \rightarrow 1s3s\ ^1S$ and $1s2p\ ^1P \rightarrow 1s3d\ ^1D$. Finally, there is evidently enough collisional transfer $1s2p\ ^1P \rightarrow 1s2s\ ^1S$ to slightly overpopulate the $1s2s\ ^1S$ state and for that reason there is also weak absorption for the transition $1s2s\ ^1S \rightarrow 1s3p\ ^1P$. This simple reasoning explains the first row of the response matrix.

In the second row the signs are reversed. When the applied radiation field is enhanced in the He β ($1s^2\ ^1S \rightarrow 1s3p\ ^1P$) line, the groundstate population is reduced and the $1s3p\ ^1P$ population is raised relative to LTE. There is weak emission on the $3 \rightarrow 2$ transitions, principally because of collisional transfer between the $n=3$ levels, which distributes the enhanced population of the $1s3p\ ^1P$ state. The $1s2p\ ^1P$ population increases by cascade and there is net emission on the resonance transition ($1s2p\ ^1P \rightarrow 1s^2\ ^1S$). The rest of the matrix is easily understood by similar reasoning.

One should not speak of emission or absorption lines because $R_{\nu,\nu'}$ involves one-atom rates. It is a separate question of radiative transfer to decide whether a line would appear light or dark against its background as seen from outside the plasma.

We have performed calculations with a NLTE screened-hydrogenic average-atom model[8] for the same density-temperature case. The calculations will be reported in detail elsewhere, but give qualitatively similar results for the response matrix. (Fig. 2a,b)

The CR model used here includes levels up to $n=20$ (with n,l,S -splitting for n up to 4 and n,S -splitting up to $n=8$) but does not include autoionizing doubly-excited levels. Li-like ions, and Li-like satellite levels are omitted, and dielectronic processes are omitted. There is ionization from excited states. If the set of ion charge-states and excited states is not complete the LTE solution of the model cannot have the exact partition function. However we have verified that when $n_{\nu} = n^0_{\nu}$ for all transitions, the population ratios are Boltzmann ratios, so the solution of the model differs from true LTE only in the overall normalization of the populations.

Symmetry of the response matrix is a test of detailed balance. The values of the response coefficients test the entire set of rates in the calculation and give an excellent way to compare different atomic models.

The linear-response method considered here is limited to a special class of NLTE states. The system must be in a steady state and must be "near" LTE, although in these calculations we have found a remarkably large range of linear response. We believe this response function can be used to simplify calculation of NLTE atomic kinetics in plasma simulation calculations.

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Table I. He-like aluminum transitions analyzed in Tables II, III.

transition 1	:	$1s^2\ ^1S - 1s2p\ ^1P$	1.598 keV
transition 2	:	$1s^2\ ^1S - 1s3p\ ^1P$	1.869 keV
transition 3	:	$1s2s\ ^1S - 1s3p\ ^1P$	0.289 keV
transition 4	:	$1s2p\ ^1P - 1s3s\ ^1S$	0.268 keV
transition 5	:	$1s2p\ ^1P - 1s3d\ ^1D$	0.273 keV

TABLE II

Linear-response coefficients $RL_{\nu,\nu'}$ (Watts/atom-eV) for He-like aluminum plasma at electron density $n_e = 10^{20}\text{ cm}^{-3}$ and temperature $T_e = 150\text{ eV}$. The matrix is obtained by averaging NLTE calculations with +1% and -1% perturbations in lines identified in Table I. The matrix is accurately symmetric.

	1	2	3	4	5
1	$-5.234 \cdot 10^{-10}$	$3.564 \cdot 10^{-10}$	$-9.317 \cdot 10^{-12}$	$-1.662 \cdot 10^{-12}$	$-4.333 \cdot 10^{-11}$
2	$3.564 \cdot 10^{-10}$	$-6.221 \cdot 10^{-10}$	$1.068 \cdot 10^{-11}$	$1.590 \cdot 10^{-12}$	$3.538 \cdot 10^{-11}$
3	$-9.317 \cdot 10^{-12}$	$1.068 \cdot 10^{-11}$	$-2.344 \cdot 10^{-12}$	$1.178 \cdot 10^{-14}$	$2.449 \cdot 10^{-13}$
4	$-1.662 \cdot 10^{-12}$	$1.590 \cdot 10^{-12}$	$1.177 \cdot 10^{-14}$	$-3.601 \cdot 10^{-13}$	$4.507 \cdot 10^{-14}$
5	$-4.333 \cdot 10^{-11}$	$3.538 \cdot 10^{-11}$	$2.449 \cdot 10^{-13}$	$4.506 \cdot 10^{-14}$	$-8.827 \cdot 10^{-12}$

Table IIIa

Response coefficient $RL_{\nu,\nu'}$ (Watts/atom-eV) for He-like aluminum plasma at electron density $n_e = 10^{20} \text{ cm}^{-3}$, temperature $T_e = 150 \text{ eV}$. The perturbations are 50% of T_e . While the matrix is not much different from Table II the symmetry is no longer perfect.

	1	2	3	4	5
1	$-5.230 \cdot 10^{-10}$	$3.561 \cdot 10^{-10}$	$-9.309 \cdot 10^{-12}$	$-1.661 \cdot 10^{-12}$	$-4.329 \cdot 10^{-11}$
2	$3.563 \cdot 10^{-10}$	$-6.220 \cdot 10^{-10}$	$1.068 \cdot 10^{-11}$	$1.589 \cdot 10^{-12}$	$3.537 \cdot 10^{-11}$
3	$-7.335 \cdot 10^{-12}$	$8.407 \cdot 10^{-12}$	$-1.845 \cdot 10^{-12}$	$9.269 \cdot 10^{-15}$	$1.928 \cdot 10^{-13}$
4	$-1.651 \cdot 10^{-12}$	$1.579 \cdot 10^{-12}$	$1.170 \cdot 10^{-14}$	$-3.577 \cdot 10^{-13}$	$4.477 \cdot 10^{-14}$
5	$-3.993 \cdot 10^{-11}$	$3.260 \cdot 10^{-11}$	$2.257 \cdot 10^{-13}$	$4.153 \cdot 10^{-14}$	$-8.135 \cdot 10^{-12}$

Table III

Response coefficient $RL_{\nu,\nu'}$ (Watts/atom-eV) for He-like aluminum plasma at electron density $n_e = 10^{20} \text{ cm}^{-3}$ and temperature $T_e = 150 \text{ eV}$. The perturbation is -50% of T_e . It is emphasized in the text that this is a large perturbation. The matrix is not much different from Table II.

	1	2	3	4	5
1	$-5.234 \cdot 10^{-10}$	$3.564 \cdot 10^{-10}$	$-9.317 \cdot 10^{-12}$	$-1.662 \cdot 10^{-12}$	$-4.333 \cdot 10^{-11}$
2	$3.564 \cdot 10^{-10}$	$-6.222 \cdot 10^{-10}$	$1.068 \cdot 10^{-11}$	$1.590 \cdot 10^{-12}$	$3.538 \cdot 10^{-11}$
3	$-1.156 \cdot 10^{-11}$	$1.326 \cdot 10^{-11}$	$-2.908 \cdot 10^{-12}$	$1.461 \cdot 10^{-14}$	$3.040 \cdot 10^{-13}$
4	$-1.670 \cdot 10^{-12}$	$1.597 \cdot 10^{-12}$	$1.183 \cdot 10^{-14}$	$-3.618 \cdot 10^{-13}$	$4.528 \cdot 10^{-14}$
5	$-4.619 \cdot 10^{-11}$	$3.772 \cdot 10^{-11}$	$2.611 \cdot 10^{-13}$	$4.804 \cdot 10^{-14}$	$-9.410 \cdot 10^{-12}$

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Figure 1.) The connection of nonequilibrium thermodynamics to atomic kinetics can be understood simply by considering an active plasma medium immersed in a controlled radiation field coming from an array of hohlraums, each coupled over one band of photon frequencies. As is usual for such "thought experiments" the x-ray mirrors and pipes are idealized and provide perfect reflection and transmission respectively. Blackbody radiation (denoted B_V) comes from each hohlraum and the sample returns a modified flux I_V . Control of the hohlraum temperatures permits exploring LTE and near-LTE plasmas in a systematic manner.

Figure 2.) Calculations from the NLTE screened-hydrogenic average-atom model for aluminum plasma at the conditions described in Table II. The response matrix shown here has units of Watts/[gram-keV³] because it is normalized by frequency group-widths for emitted and perturbed portions of the radiation spectrum. The signs for response to perturbations of He_α (Fig 2a) and He_β (Fig 2b) lines are equivalent to results from the CR model given in Table II.

Figure 1

Figure 2a

Figure 2b

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