Nonequilibrium Equation of State in Stellar Atmospheres

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Abstract

In the stellar chromospheres, radiative energy transport is dominated by only the strongest spectral lines. For these lines, the approximation of local thermodynamic equilibrium (LTE) is known to be very inaccurate, and a state of equilibrium cannot be assumed in general. To calculate the radiative energy transport under these conditions, the population evolution equation must be evaluated explicitly, including all time-dependent terms. We develop a numerical method to solve the evolution equation for the atomic-level populations in a time-implicit way, keeping all time-dependent terms to first order. We show that the linear approximation of the time dependence of the populations can handle very large time steps without losing accuracy. We reproduce the benchmark solutions from earlier, well-established works in terms of non-LTE kinetic equilibrium solutions and typical ionization/recombination timescales in the solar chromosphere.

Unified Astronomy Thesaurus concepts: Radiative magneto-hydrodynamics (2009); Radiative transfer (1335); Radiative transfer simulations (1967); Stellar atmospheres (1584); Computational methods (1965); Solar atmosphere (1477); Stellar chromospheres (230); Ionization (2068); Solar chromosphere (1479); Recombination (2072); Spectroscopy (1558); Theoretical techniques (2093)

1. Introduction

Since the introduction of the routine application of computing power in astrophysics, it has become an integral part of the interpretative process at almost every level. In particular, the evaluation of the expected behavior of astrophysical systems, under the assumption that they obey the laws of physics, has become a powerful tool in the interpretation of observational data. In solar physics, a new level of sophistication was made possible with the groundbreaking work of Nordlund (1982) and others, with the introduction of ab initio calculation of the solar atmosphere, where it is possible to work with a strongly reduced set of assumptions and approximations. The last decade in particular has seen a sharp rise in the use of such “parameter-free” simulations in the interpretation of observations, due to the ever-increasing availability of massively parallel computing resources. To this end, several radiative magnetohydrodynamics (MHD) codes have been developed to simulate solar/stellar atmospheres, such as STAGGER (Stein & Nordlund 1998), MURaM (Vögler et al. 2005; Rempel 2017), BIFROST (Gudiksen et al. 2011), CO5BOLD (Freytag et al. 2012), MANCHA (Felipe et al. 2010), and so on.

The photosphere is dominated by a high gas density and relatively insignificant radiative losses, leaving the problem dominated by relatively local terms. In addition, the high density ensures high collision rates, resulting in timescales on which the atomic populations reach an equilibrium with their environment that is very short compared to the timescale on which that environment changes, so that equilibrium conditions may safely be assumed. This condition is known as the local thermodynamic equilibrium (LTE). Due to the low particle densities, however, this is no longer the case in the chromosphere, where the collisional rates are low and processes involving multiple particles are correspondingly rare. The evolution timescale of the magnetically dominated structures, on the other hand, is considerably shorter than in the photosphere, so that the timescales on which the atomic populations reach an equilibrium with their environment may well exceed the evolution timescale of the environment itself, so that equilibrium is never reached.

Klein et al. (1976, 1978) and Kneer (1980) demonstrated that the assumption of kinetic (statistical) equilibrium produces contrasting ionization and recombination timescales, thus proving the invalidity of that assumption in a dynamic atmosphere with shock waves. Judge (2017) proposed a fast probabilistic approach to solve the non-LTE nonequilibrium radiative transfer (RT) for dynamically evolving one-dimensional (1D) atmospheres. The RADYN code (Carlsson & Stein 1992, 1995, 2002) solves the 1D hydrodynamic equations (equation of mass, momentum, and energy conservation) together with the non-LTE RT equation using an adaptive mesh algorithm. Although RADYN takes care of the nonequilibrium and non-LTE effects consistently, because it is a hydrodynamical code, it cannot be used to study heating mechanisms that rely on the presence of magnetic fields. For these reasons, RADYN is not suitable for three-dimensional (3D) radiative MHD simulations.

The BIFROST code (Gudiksen et al. 2011) is capable of radiative MHD simulations in two dimensions (2D) and 3D that also takes nonequilibrium effects into account (Leenaarts et al. 2007, 2009). However, in BIFROST, the time dependence is neglected in the rate system, and it further approximates equation-of-state and RT calculations by using prescribed recipes for chromospheric radiative losses and hydrogen ionization in order to make the problem more tractable in 2D/3D.

Generally, in the radiation MHD simulations, chemical equilibrium is assumed for the molecular formation/dissociation in the equation of state. Although a simple nonequilibrium treatment of the H$_2$ molecule is included in Leenaarts et al. (2011), there is potential for studying the importance of other chemical reactions in the solar atmosphere.

In this paper, we focus on improving the existing methods to solve the non-LTE nonequilibrium RT problem through (a) a proper time-dependent treatment of the radiation field, (b) a
proportional nonequilibrium treatment of the molecular chemistry, and (c) development of a time-implicit numerical scheme. Furthermore, the method developed in this paper is suitable for 1D/2D/3D simulations.

In Section 2, we describe basic radiative MHD, RT, and kinetic equilibrium equations. In Section 3, we describe in detail our development of the population evolution scheme, our newly developed time-dependent short-characteristics method for the formal solution of the RT equation, and finally, our generalization of the multilevel approximate lambda iteration (MALI) scheme to solve the time-dependent nonlinear rate system. In Section 4, we describe the implementation of the method. In Section 5, we discuss the accuracy of our new method and show the benchmark solutions reproduced from our method. Finally, we present a summary in Section 6.

2. Radiation MHD

2.1. Basic Equations

The MHD equations are typically expressed as conservation equations, expressing the conservation of mass, momentum, energy, and magnetic flux along with a number of expressions to close the system.

We recall the important hydrodynamic equations here, ignoring the magnetic field for the sake of simplicity. The equations of continuity, momentum, and energy are respectively given by (see, e.g., Rempel 2014)

\[
\frac{\partial \rho}{\partial t} = \nabla \cdot (\rho \mathbf{v}),
\frac{\partial \rho \mathbf{v}}{\partial t} = -\nabla P + \rho \mathbf{g},
\frac{\partial E_{\text{HD}}}{\partial t} = -\nabla \cdot [\nu(E_{\text{HD}} + P)] + \rho \mathbf{v} \cdot \mathbf{g} + Q_{\text{rad}}.
\]

Here \(\rho\), \(P\), and \(\mathbf{v}\) denote the mass density, pressure, and velocity, respectively. We note that the atmosphere we are considering is a small box encompassing part of the convection zone, the photosphere, the chromosphere, and part of the corona. Therefore, we approximate the gravitational acceleration to be a constant taken to be \(-2.74 \times 10^7 \text{ cm}^2 \text{ s}^{-1}\). In the solar/stellar chromosphere, the dominant term in the energy equation is the loss of energy to the outer environment in the form of radiation. Here \(Q_{\text{rad}}\) is the radiative heating term given by the divergence of radiative flux \(-\nabla \cdot F\), where \(F\) is the radiative flux.

2.2. Equations of State

We assume that we have a gas mixture that consists of atomic hydrogen, denoted by H; the hydrogen molecules \(\text{H}_2\), \(\text{H}_2^+\), and \(\text{H}^+\); and several other metals. The occupation numbers of the constituents of the gas determine the pressure \(P\), and \(F\) is computed using the specific intensity obtained by solving the RT equation. When the approximation of LTE is valid, the solution of the RT equation is simple, and the pressure can be tabulated. For non-LTE conditions, such as the solar/stellar chromosphere, one cannot use tables, and explicit treatment of the elements of the gas needs to be considered.

The total energy is distributed into the kinetic and internal energies of the constituents of the gas. The internal energy of the system is distributed among its various degrees of freedom (e.g., bound and free states of atoms and molecules). The temperature can be determined by imposing the total energy to be conserved. Finally, the system is closed by imposing particle and charge conservation. The combinations of these expressions are known as the equations of state.

2.3. The RT Equation

The flux divergence term \(Q_{\text{rad}} = \nabla \cdot F\) appearing in the energy equation (see Equation (1)) requires the radiative flux \(F\), which is given by

\[
F = \int d\nu \int d\Omega \mathbf{\Omega} \cdot \mathbf{l}_{\nu,\Omega}.
\]

where \(l_{\nu,\Omega}\) is the specific intensity at frequency \(\nu\) along the ray direction \(\Omega\) (see, e.g., Mihalas 1978; Hubeny & Mihalas 2014). To obtain \(l_{\nu,\Omega}\), we need to solve the RT equation, which, in Cartesian geometry, is given by

\[
-\frac{1}{\alpha_{\text{tot}}} \Omega \cdot \nabla l_{\nu,\Omega} = l_{\nu,\Omega} - S_{\nu},
\]

where \(S_\nu = \frac{\eta_{\text{tot},\nu}}{\alpha_{\text{tot},\nu}}\) is the source function, with \(\eta_{\text{tot},\nu}\) and \(\alpha_{\text{tot},\nu}\) being the total emissivity and total opacity, taking contributions from both the lines and the continua. The emissivity and opacity depend on the atomic population density \(n\). The solution of the transfer equation, known as the formal solution, depends nonlinearly on \(n\).

We note here that we ignore the term \(\frac{dl_\nu}{d\nu}\) in the RT equation. As studied in detail in Klein et al. (1976), the ratio of the thermal relaxation time (the time needed to lose the internal energy through radiation for a heated gas) to the photon travel time is \(\sim 10^{-10}\); also, the photon travel time is much shorter when compared with the dynamic timescales (which are of the order of scale height to local sound speed \(\sim 10\)). This means that we can safely assume that the radiation field adjusts instantaneously to the state of the medium.

2.4. Kinetic Equilibrium Equation

In general, we will assume that the rate of change of the population densities \(n\) of atomic excited states in a medium is given by the coupled system of equations

\[
\frac{\partial n}{\partial t} + (\nu \cdot \nabla) n = A \cdot n,
\]

where the rate matrix \(A\) typically contains the radiative and collisional transition rates between the different atomic energy levels. The radiative rate coefficients are themselves nonlinearly dependent on the instantaneous populations, so that the system of equations does not have an analytical solution in general. In many situations of interest, stationary or quasi-stationary conditions can be assumed, implying that

\[
\frac{\partial n}{\partial t} = 0, \quad (\nu \cdot \nabla) n = 0,
\]

and thus the populations must satisfy the condition

\[
A \cdot n = 0,
\]

or, in a component form,

\[
\sum_{\ell \neq 1} n_{\ell} C_{\ell,1} + \sum_{\ell > 1} n_{\ell} R_{\ell,\ell'} - \sum_{\ell < 1} n_{\ell} C_{\ell,\ell'} - \sum_{\ell < 1} n_{\ell} R_{\ell,\ell'} = 0,
\]
where $l$ is the level index of the population densities. Besides
the locally determined collisional rates $C_{l'j}$, these equations
contain radiative rates $R_{l'j}$ that depend on the angular and
transition-averaged intensity $I_{l'j}$, which is highly nonlocal in
optically thin conditions. Equation (7) is known as the kinetic
equilibrium equation or statistical equilibrium equation. The
solution of this system of equations is typically found by
iterative evaluation of the RT equation in order to improve an
initial estimate of $n$, a process that is usually referred to as non-
LTE RT.

2.5. Timescales

An important aspect of the non-LTE radiation field in the
solar/stellar chromosphere is that the timescale of the radiative
recombination process can vary from $\sim 50$ up to $10^5$ s (Carlsson
& Stein 1992, 1995, 2002), which is much longer than the
dynamic timescale of $\sim 10$ s (Klein et al. 1976). Therefore, the
assumption of instantaneous kinetic (statistical) equilibrium
(Equation (7)) is not always valid in the chromosphere. There-fore, we need to treat the atomic populations in nonequilibrium. Further, due to the interdependence of the non-LTE radiation field and the atomic populations, we need to solve the non-LTE RT equation and the nonequilibrium rate system, consisting of collisional and radiative rates, to obtain the occupation numbers of all of the constituent atomic species simultaneously.

3. Evolution of State

3.1. Population

We have seen that under the kinetic equilibrium conditions, we have Equation (7), the solution of which, together with the
simultaneous solution of the RT equation, provides the
population density $n$. As discussed in Section 2.5, we cannot
assume kinetic equilibrium in general. When equilibrium
cannot be assumed, we are left with no choice but to evaluate
Equation (4), which has the form

$$A \cdot n = b,$$

or, in component form with level index $l$,

$$\frac{\partial n_l}{\partial t} + (\mathbf{v} \cdot \nabla)n_l = \sum_{l'j} n_{l'} C_{l'j} + \sum_{l'j} n_{l'} R_{l'j} - \sum_{l'j} n_{l'} C_{l'j} - \sum_{l'j} n_{l'} R_{l'j}.$$  

We note here that Equation (9) has the same form as that of
Equation (7), which is the non-LTE kinetic equilibrium equation. However, we now have a nonzero vector $b$ on the
right-hand side. We solve this system by integrating
Equation (4) from time $t_n$ to time $t$, resulting in the formal solution

$$n(t) = n(t_n) + \int_{t_n}^{t} A(t') \cdot (n(t') - b) \, dt'$$

$$- \int_{t_n}^{t} (\mathbf{v}(t') \cdot \nabla)n(t') \, dt'.$$  

To find a solution to this system, we follow the ideas that lead
to the short-characteristics method of Kunasz & Auer (1988),
which was originally developed for an efficient evaluation of
the formal solution of the non-LTE RT equation in a

multidimensional geometry. Along each ray of the angle
quadrature direction, instead of traversing the entire space, the
intensity is evaluated locally at the central point of a three-point
stencil, using the known intensity in the upwind direction. The
nonlinear spatial dependence of the source function is approximated using a polynomial in terms of the known
source function values at the spatial grid points on the local
stencil. The polynomial form of the source function allows a
direct evaluation of the formal integral. This process is repeated
down the ray to cover the entire space (see also Auer &
Paletou 1994).

We now apply the same ideas to the time variable $t$. Denoting $t$ at two successive time steps as $t_n$ and $t_{n+1}$, we define $\delta t = t_{n+1} - t_n$ as the time interval between $t_n$ and $t_{n+1}$. We are interested in evaluating $n(t)$ at $\delta t$ for each time step. For
this purpose, all of the time-dependent quantities are expressed
as linear polynomials in $t$ that use the known values of these
quantities at a previous time step and ignore all of the cross-
terms of second and higher order. This allows a direct
evaluation of the time integration in Equation (10), yielding a
linear system of equations.

We start with the linear polynomial form of the populations,

$$n(t) = n_0 + \int_{0}^{\delta t} A(t') \cdot (n(t') + n_0) \, dt'$$

and substitute it in Equation (10), which yields

$$\dot{n}_n = \int_{0}^{\delta t} A(t') \cdot (\dot{n}_n t' + n_0) \, dt'$$

$$- \int_{0}^{\delta t} (\mathbf{v}(t') \cdot \nabla)(n_n t' + n_0) \, dt'.$$  

At $\delta t$, we have

$$\dot{n}_n \delta t = \int_{0}^{\delta t} A(t') \cdot (\dot{n}_n t' + n_0) \, dt'$$

$$- \int_{0}^{\delta t} (\mathbf{v}(t') \cdot \nabla)(n_n t' + n_0) \, dt'.$$  

which can be solved for $\dot{n}_n$. The main difficulty in solving this
system lies in the implicit nonlocal and nonlinear dependence
of the first term on the right-hand side on $\dot{n}_n$. This dependence is similar to that found in equilibrium non-LTE problems, for
which it is known that it is very stiff and converges very slowly
when solved by iterative means only. A more successful
approach is to linearize and localize Equation (13) in $\dot{n}_n$ and
solve the linear system while solving for the remaining nonlinearity iteratively, a process that is usually referred to as “acceleration” (see Cannon 1973a, 1973b; Scharmer 1981; Olson et al. 1986; Rybicki & Hummer 1991, 1992; Hubeny 2003, and references therein).

We therefore focus our attention on the nonlinear term and
expand it,

$$\int_{0}^{\delta t} A(t') \cdot (\dot{n}_n t' + n_0) \, dt' = \int_{0}^{\delta t} A(t') \cdot t' \cdot \dot{n}_n$$

$$+ \int_{0}^{\delta t} A(t') \cdot t' \cdot n_0.$$  

In light of Equation (9), this expression contains three types of integrals over time that must be calculated. The integrals over
the constant spontaneous emission terms are trivial and not
discussed. The collisional rates, however, are a complex
function of the temperature \( T_n(t) \) of the form
\[
C_{l',l} = C_0 n_e(t) \sqrt{T(t)} e^{-\frac{E_{l'}}{kT(t)}} \Gamma[T(t)].
\] (15)

Assuming the time dependence of the temperature, the electron density, and the coefficients \( \Gamma[T(t)] \) in the time interval \([t_n, t_{n+1}]\) to be linear,
\[
T(t) = \tilde{T}_n t + T_n,
\] (16)
\[
n_e(t) = \tilde{n}_e t + n_{e,c},
\] (17)
\[
\Gamma(t) = \tilde{\Gamma}_n t + \Gamma_n,
\] (18)
we can express the collisional rate coefficients as linear polynomials in \( t \) and write them as
\[
C_{l',l} = \tilde{C}_{l',l,n} t + \bar{C}_{l',l,n}.
\] (19)

The time-integrated radiative rates are given by
\[
\int_0^{\delta t} R_{l',l}(t')dt' = \oint \frac{dV}{4\pi} \Omega \frac{d
u}{h\nu} U_{l',l}(t') \left[ n_e(t') U_{l',l}(t') + (V_{l',l} + n_e(t') V_{l',l}(t'))I(t') \right],
\] (20)
where the quantities \( U_{l',l}, U_{l',l}', V_{l',l}, \) and \( V_{l',l}' \) are defined in Appendix A. Evaluation of this expression presents a challenge, however, since the intensity is a highly nonlinear, nonlocal function of the populations and, in addition, a complicated function of time. Upon substituting the time-linear expression for all of the time-dependent quantities (see Appendix A for details) and neglecting all terms of second or higher order in time in the above equation, we have
\[
\int_0^{\delta t} R_{l',l}(t')dt' = \oint \frac{dV}{4\pi} \Omega \frac{d
u}{h\nu} \left[ (U_{l',l} + n_e U_{l',l}') \delta t \right. \\
+ (V_{l',l} + n_e V_{l',l}')I + \left. (V_{l',l} + n_e V_{l',l}')I \right],
\] (21)
where
\[
\mathcal{I} = \int_0^{\delta t} I(t')dt' \quad \text{and} \quad \tilde{I} = \int_0^{\delta t} I(t')t'dt'
\] (22)
were introduced.

### 3.2. Radiative Quantities

To obtain the intensity at a given frequency and angle in every point on the grid, we must first be able to calculate the opacity and emissivity. The opacity at a given frequency \( \nu \) is the result of the sum over all lines, added to the continuum opacity \( \alpha_c \),
\[
\alpha_\nu = \alpha_{c,\nu} + \frac{\nu \hbar}{4\pi} \sum_{l \neq l'} \phi_{l',l}(t') \left[ B_{l',l} n_{l'} - B_{l',l} n_l \right].
\] (23)

To obtain a time-dependent opacity in the interval \( t = t^n \) to \( t^{n+1} \) that we can work with, we must approximate the time dependence of the line profile \( \phi_{l',l}(t) \), as was done earlier for the populations \( n(t) \). Under the assumption that the acceleration over each time interval is small compared to the Doppler width of the line, we can make a linear expansion of \( \phi_{l',l}(t) \) and the bulk velocity \( v(t) \) around \( t = t^n \),
\[
\phi_{l',l}(t) \approx \phi_{l',l}(t^n) + \frac{\nu}{c} \frac{\partial \phi_{l',l}(t^n)}{\partial t^n} \left[ t^n \right] \equiv \phi_{l',l}(t^n) + \frac{\nu}{c} \frac{\partial \phi_{l',l}(t^n)}{\partial t^n}.
\] (24)

and substitute it in Equation (23). The resulting opacity for the transition \( l \rightarrow l' \),
\[
\alpha_{l,l'}(t) = \frac{\nu^2}{4\pi} \left[ \phi_{l',l}(t^n) + \frac{\nu}{c} \frac{\partial \phi_{l',l}(t^n)}{\partial t^n} \right] \left( B_{l',l} n_{l'} - B_{l',l} n_l \right) t \\
+ B_{l',l} n_{l'} - B_{l',l} n_l,
\] (25)
is a quadratic function of time, due to the interplay between the time dependence of the population densities and the Doppler-shifted line profile. However, since we neglected all time dependence of second and higher order in the populations and line profile, we may, without loss of accuracy, proceed by dropping all terms of second and higher order in time from Equation (25), yielding
\[
\alpha_\nu(t) \approx \alpha_{c,\nu}(t) + \nu^2 \sum_{l \neq l'} \phi_{l',l}(t^n) \left[ B_{l',l} n_{l'} - B_{l',l} n_l \right] + \phi_{l',l} \frac{\nu}{c} (B_{l',l} n_{l'} - B_{l',l} n_l) \right]
\] (26)
Similarly, we approximate the emissivity
\[
\eta(t) \approx \eta_{c,\nu}(t) + \nu^2 \sum_{l \neq l'} \phi_{l',l}(t^n) \left[ B_{l',l} n_{l'} - B_{l',l} n_l \right] + \phi_{l',l} \frac{\nu}{c} (B_{l',l} n_{l'} - B_{l',l} n_l).
\] (27)

For strong spectral lines, the contribution of continuum sources to the emissivity and opacity is generally small, so that the order of the contribution to the time dependence of them can safely be assumed to be linear. Clearly, this assumption reduces the time dependence of the opacity and emissivity to linear,
\[
\eta_\nu(t) \approx \eta_{c,\nu}(t) + \nu^2 \sum_{l \neq l'} \phi_{l',l}(t^n) \left[ B_{l',l} n_{l'} - B_{l',l} n_l \right] + \phi_{l',l} \frac{\nu}{c} (B_{l',l} n_{l'} - B_{l',l} n_l),
\] (28)
where
\[
\eta_{c,\nu} = \eta_{c,\nu} + \frac{\nu^2}{4\pi} \sum_{l \neq l'} B_{l',l} \phi_{l',l}(t^n) n_{l'},
\]
\[
\eta_{c,\nu} = \eta_{c,\nu} + \frac{\nu^2}{4\pi} \sum_{l \neq l'} B_{l',l} \phi_{l',l}(t^n) n_l,
\]
\[
\alpha_{c,\nu} = \alpha_{c,\nu} + \frac{\nu^2}{4\pi} \sum_{l \neq l'} \phi_{l',l} (B_{l',l} n_{l'} - B_{l',l} n_l),
\] (29)
The source function now assumes the simple rational form
\[
S_\nu(t) = \frac{\eta(t)}{\alpha(t)} = \frac{\eta_{c,\nu}(t) + \eta_\nu(t)}{\alpha_{c,\nu}(t) + \alpha_\nu(t)}.
\] (30)
3.3. Time-dependent Short Characteristics

The method of short characteristics (Kunasz & Auer 1988; Auer & Paletou 1994) solves the RT equation along the characteristics of the equation (rays) that are limited to individual grid cells. While this method generates the problem of required knowledge of the intensity on the boundary of each grid cell, an especially complicating factor when the calculations need to be parallelized, it involves numerical work that is constant for each grid cell and thus scales linearly with the number of grid cells, a property that, despite this drawback, has made it the method of choice for multidimensional problems in the last two decades.

We thus proceed by calculating the intensity on the grid using this method and formulate the integration over the optical depth in each cell using quadratic interpolation for the atmospheric properties as a function of optical depth (see also Kunasz & Auer 1988; Auer & Paletou 1994). We note that although the formulation provided here is for quadratic polynomials, in practice, we use linear polynomials, as they are more stable and faster to compute. Alternatively, Bezier or Hermite polynomials could be used to improve the stability and accuracy of the method (see, e.g., Auer 2003).

We adopt the method of the locally comoving laboratory frame (see, e.g., van Noort et al. 2002; Leenaarts et al. 2009), which assumes for each grid point that it is at rest. This method provides a convenient mix of the observer’s frame and the comoving frame formalisms but requires all local quantities (opacity, emissivity) to be interpolated to the locally comoving frame. A key advantage of this method is that the angle-dependent frequency redistribution of the radiation field induced by spatial gradients in the bulk velocity of the gas is automatically taken into account. In the current work, we will limit ourselves to situations where the velocity gradients are sufficiently small that the Doppler shift within one grid cell does not significantly exceed the frequency grid spacing, so that additional subgridding of the characteristics can be omitted.

To calculate the comoving observer’s frame optical depth at frequency \( \nu \), we need to integrate the opacity, given by Equation (23). To integrate this expression, the spatial dependence of \( \Phi_{j}^{\mu} \) and \( n_{i} \) must be known analytically. To obtain this dependence, we interpolate the relevant physical quantities from the grid to the end points of the characteristic at distances \( \pm \Delta s \) from the center. To guarantee positivity, we require the analytic continuation along the characteristic to be linear in the spatial coordinate, \( s \),

\[
\alpha_{\nu}(s, t) \approx \alpha_{\nu}^{c}(t) + \frac{1}{\Delta s} \alpha'_{\nu}(t) s, \tag{31}
\]

where \( \alpha_{\nu}^{c}(t) \) is the opacity in the central grid point, and \( \alpha'_{\nu}(t) \) is the spatial derivative of the opacity, obtained by means of spatial interpolation at the intersection of the characteristic and the upwind cell boundary,

\[
\alpha'_{\nu}(t) = \frac{\sum_{j} C_{j} \alpha_{\nu}^{d}_{j}(t)}{\sum_{j} C_{j}} - \alpha_{\nu}^{c}(t), \tag{32}
\]

where \( \alpha_{\nu}^{c}(t) \) and \( \alpha_{\nu}^{d}_{j}(t) \) are the opacity values in the current grid point and the upwind grid points with interpolation index \( j \) and at the Doppler-shifted frequency,

\[
\nu'(t) = \nu + \frac{\nu}{c} \left( \sum C_{j} \alpha_{\nu}^{d}_{j}(t) - \alpha_{\nu}^{c}(t) \right), \tag{33}
\]

at the upwind limit of the characteristic, respectively. The number of contributions to the sum depends on the order chosen for the interpolation and is four for the linear scheme illustrated in Figure 1. The trivial integration along the characteristic now results in the optical depth

\[
\tau_{\nu}^{u}(t) = \int_{0}^{\Delta s_{u}} \alpha_{\nu}(s, t) d s \approx \left[ \alpha_{\nu}^{c}(t) + \frac{1}{2} \alpha'_{\nu}(t) \right] \Delta s, \tag{34}
\]

which is readily calculated using Equation (28) to have the form

\[
\tau^{u}(t) = \tau^{u}_{\nu} + \tau^{u}, \tag{35}
\]

where

\[
\tau^{u}_{\nu} = \left[ \frac{\Delta s_{u}}{2} \left( \sum C_{j} \alpha_{\nu}^{d}_{j} - \alpha_{\nu}^{c} \right) \right] \Delta s, \tag{36}
\]

Similarly, the downwind optical depth \( \tau^{d}(t) \) can be calculated from

\[
\tau^{d}(t) = \tau^{d}_{\nu} + \tau^{d}, \tag{37}
\]

where

\[
\tau^{d}_{\nu} = \left[ \frac{\Delta s_{d}}{2} \left( \sum C_{j} \alpha_{\nu}^{d}_{j} - \alpha_{\nu}^{c} \right) \right] \Delta s, \tag{38}
\]

Figure 1. Short characteristics on a 3D grid.
We are now ready to evaluate the formal integral along the short characteristic. Using the method from Auer & Paletou (1994), we write the source function as a parabolic function of the optical depth,

\[ S(\tau, t) = c_0(t) + c_1(t)\tau(t) + c_q(t)\tau^2(t), \]  

and evaluate the formal integral,

\[ I(t) = I_0(t)e^{-\tau(t)} + \int_0^{\tau(t)} (c_0(t) + c_1(t)s + c_q(t)s^2)e^{-s}ds, \]  

along the characteristic, which has the general solution

\[ I(t) = [I_0(t)e^{-\tau(t)} + c_0(t)(1 - e^{-\tau(t)}) + c_1(t)(1 - (\tau(t) + 1)e^{-\tau(t)}) + c_q(t)(2 - 2(1 + \tau(t)) + \tau(t)^2)e^{-\tau(t)})], \]  

where the coefficients of \( S \) are given by

\[ c_0(t) = S'(t), \]

\[ c_1(t) = \frac{[S'(t) - S(t)]\tau(t)^2 - [S'(t) - S(t)]\tau^2(t)^2}{\tau(t)^2[\tau(t) + \tau^2(t)]}, \]

\[ c_q(t) = \frac{[S^2(t) - S(t)^2][\tau(t)^2 - \tau^2(t)^2]}{\tau(t)^2[\tau(t) + \tau^2(t)]} \]

To obtain the time-integrated rates, we still need to integrate over frequency, angle, and time. The unappealing prospect of carrying the analytic form of Equation (41) through to the end suggests that it might be advantageous to carry out the time integration of Equation (41) over the time step before the angular and frequency integration.

Substitution for \( S \) in the coefficients yields

\[ c_0 = \frac{\eta_e}{\alpha_e}, \]

\[ c_1 = \frac{\alpha_u\alpha_d\eta_e - \alpha_u\alpha_d\eta_k - \alpha_u\alpha_d\eta_e - \alpha_u\alpha_d\eta_k\tau_d^2}{(\tau_u + \tau_d)^2\tau_u\alpha_u\alpha_d}, \]

\[ c_q = \frac{(\alpha_u\alpha_d\eta_g - \alpha_u\alpha_d\eta_k\tau_u + (\alpha_e\alpha_d\eta_k - \alpha_e\alpha_d\eta_g)\tau_d}{(\tau_u + \tau_d)^2\tau_u\alpha_u\alpha_d}, \]

in which the explicit time dependence was dropped for notational brevity. Despite the substantial simplifications made earlier, the result has clearly regained a complicated time dependence through the high-order rational form of the coefficients.

The form of Equation (41) suggests that it is advantageous to rewrite it to

\[ I(t) = I_0(t)e^{-\tau(t)} - [c_0(t) + c_1(t)(1 + \tau(t)) + c_q(t)(2(1 + \tau(t)) + \tau(t)^2)]e^{-\tau(t)} + c_0(t) + c_1(t) + 2c_q(t), \]  

then expand and add up the coefficients \( c_0, c_1, \) and \( c_q \). The resulting expression is a high-order polynomial of time, containing cross products of the time derivative of all of the quantities in the numerator and denominator of the \( c(t) \). Without loss of accuracy, we retain only the linear terms in both the numerator and the denominator, yielding

\[ [c_0(t) + c_1(t)(1 + \tau(t)) + c_q(t)(2(1 + \tau(t)) + \tau(t)^2)]e^{-\tau(t)} \approx \frac{c_{\tau1} + c_{\tau2}e^{-\tau(t)}}{c_{\tau3} + c_{\tau4}}, \]

where the \( c \) are extensive combinations of the average and time derivative of optical depth, opacity, and emissivity terms in nearby grid points.

Similarly, we can reduce the final term in Equation (48) to a simple rational form,

\[ c_0(t) + c_1(t) + 2c_q(t) \approx \frac{c_{\tau1} + c_{\tau2}e^{-\tau(t)}}{c_{\tau3} + c_{\tau4}}, \]

We note that Equations (49) and (50) can be rewritten as expressions resulting from a Padé approximation. Now the function to evaluate becomes

\[ I(t) \approx I_0(t)e^{-\tau(t)} - \frac{c_{\tau1} + c_{\tau2}e^{-\tau(t)}}{c_{\tau3} + c_{\tau4}} + \frac{c_{\tau1} + c_{\tau2}e^{-\tau(t)}}{c_{\tau3} + c_{\tau4}} + \frac{c_{\tau1} + c_{\tau2}e^{-\tau(t)}}{c_{\tau3} + c_{\tau4}} \]

which is readily solved using

\[ \int_{\xi - \tau(t)}^{\xi - \tau(t)} e^{-\xi}d\xi = e^\xi - \tau(t)) - \tau(t)) \]

and

\[ \int_{\xi - \tau(t)}^{\xi - \tau(t)} e^{-\xi}d\xi = \xi + (p_{\tau1} - \tau(t)\ln(-p_{\tau1} + \xi) \]

to yield

\[ T(t) = \int_0^{\tau(t)} I_0(t)e^{-\tau(t)}d\tau + [K_0\phi_0\phi_0(\xi + \xi) + K_0\phi_0\phi_0(\xi + \xi) + K_0\phi_0\phi_0(\xi + \xi)], \]

with

\[ Q_{\tau, 0} = e^{-\xi(t)} - (z_{\tau, 1} - p_{\tau, 1})e^{-\xi(t)} + \xi e^{-\xi(t)} = \xi + (z_{\tau, 1} - p_{\tau, 1})\ln(\xi + \tau(t)) \]

Similarly,

\[ \int_{0}^{\tau(t)} I_0(t)e^{-\tau(t)}d\tau + [K_0\phi_0\phi_0(\xi + \xi) + K_0\phi_0\phi_0(\xi + \xi) + K_0\phi_0\phi_0(\xi + \xi)], \]

where

\[ Q_{\tau, 1} = -\frac{1}{\tau_u} \times [(1 - z_{\tau, 1} + p_{\tau, 1})e^{-\xi(t)} + \xi e^{-\xi(t)} - \tau(t)(z_{\tau, 1} - p_{\tau, 1})e^{-\xi(t)} + \xi e^{-\xi(t)} + \frac{1}{2}\xi(\xi + 2z_{\tau, 1} - 2p_{\tau, 1}) - \tau(t)(z_{\tau, 1} - p_{\tau, 1})\ln(\xi + \tau(t))], \]
with

\[ K_{e,0} = K_{r,1} = - \frac{c_{e,1}}{c_{e,3}}, \]
\[ K_{r,0} = K_{r,1} = \frac{c_{r,1}}{c_{r,3}}, \]

\[ z_{e,1} = - \frac{c_{e,2}}{c_{e,1}}, \quad p_{e,1} = - \frac{c_{e,4}}{c_{e,3}}, \]
\[ z_{r,1} = \frac{c_{r,2}}{c_{r,1}}, \quad p_{r,1} = \frac{c_{r,4}}{c_{r,3}}, \]
\[ \xi_0 = \delta t, \quad \xi_1 = \delta t. \]  

(61)

This completes the calculation of the time-integrated formal solution.

### 3.4. Propagation of the Intensity

All we need now to complete the calculation of $\bar{T}$ and $\hat{I}$ is to evaluate the first term on the right-hand side,

\[ \int_0^{\bar{t}} I_0(t) e^{-\tau_i(t)} dt \quad \text{and} \quad \int_0^{\bar{t}} t I_0(t) e^{-\tau_i(t)} dt. \]  

(62)

Unfortunately, $I_0(t)$ is the result of a spatial and frequency interpolation of the upwind intensity and does not have a simple form. In fact, it is straightforward to show that to retain the exact time dependence of the radiation field, for every propagation step across the grid, new terms in a sum of increasing length are introduced, quickly leading to an intractable expression.

To overcome this complexity, we approximate the intensity with a polynomial form, since this form will not grow in complexity when interpolated spatially. Since the weighting that this procedure will give over the interval of integration is not clear in general, we choose not to apply any weight and to simply fit a polynomial of order $n$ with equal weights for all deviations from the true intensity across the time interval. In this simple case, the optimal coefficients are given by those that minimize the distance between the polynomial form and the true intensity over the time step.

We define the distance as the integral of the square of the difference between the two functions on the time interval $[0, \delta t]$,

\[ \chi^2 = \int_0^{\bar{t}} (I(t) - \bar{P}_n(t))^2 dt, \]  

the minimum of which is located where the partial derivatives to the coefficients $c_n$ of the polynomial $P_n(t)$ vanish,

\[ \frac{\partial}{\partial c_n} \chi^2 = 0. \]  

(64)

We substitute for $\chi^2$ and use the linearity of integration and differentiation,

\[ \frac{\partial}{\partial c_n} \int_0^{\bar{t}} (I(t) - \bar{P}_n(t))^2 dt = \int_0^{\bar{t}} \frac{\partial}{\partial c_n} (I(t) - \bar{P}_n(t))^2 dt, \]  

(65)

to obtain

\[ \int_0^{\bar{t}} -2[I(t) - \bar{P}_n(t)]t^\alpha dt = -2 \left[ \int_0^{\bar{t}} I(t)t^\alpha dt - \int_0^{\bar{t}} \bar{P}_n(t)t^\alpha dt \right] = 0. \]  

(66)

Interestingly, if we assume a linear form, $I(t) = P_1(t) = c_0 t + c_1$, and we minimize $\chi^2$, we obtain

\[ c_1 \frac{1}{3} \delta t^3 + c_0 \frac{1}{2} \delta t^2 = \int_0^{\bar{t}} \bar{I}(t) dt \equiv \bar{I}(t) \]  

(67)

and

\[ c_1 \frac{1}{2} \delta t^2 + c_0 \delta t = \int_0^{\bar{t}} I(t) dt \equiv \bar{I}(t), \]  

(68)

the right-hand side of which contains only moments of the time-integrated intensity, $\bar{I}(t)$ and $\bar{I}(t)$, both of which were already computed in all upstream grid points and can thus be obtained for free. In terms of these quantities, the coefficients of $P_1(t)$ are given by

\[ c_0 = \frac{2(2\bar{I}(t)\delta t - 3\bar{I}(t))}{\delta t^2}, \]  

(69)

\[ c_1 = \frac{6(2\bar{I}(t) - I(t)\delta t)}{\delta t^3}, \]  

(70)

which can now be used to spatially interpolate the intensity and calculate the last missing term.

### 3.5. Solving the Nonlinear Rate System

In this section, we describe the nonlinear rate system and its solution. For ease of discussion, we first fix the values of temperature and electron density, and then we describe how we include them in the nonlinear rate system.

As discussed in the previous sections for a given population density, the formal solution provides the radiation field quantities $\bar{T}$ and $\hat{I}$; by integrating these, we can obtain the radiative rates. Using the fixed temperature and the electron density, we can obtain the collisional rates. For a given set of collisional and radiative rates, solving the system in Equation (8) provides the population density. Since the population densities and radiation field are nonlinearly coupled to each other, we need to iterate between the formal solution and the nonlinear rate system in Equation (8). The most effective way to solve this nonlinear problem is by linearizing the rate system and using iterative methods. Here this linearization is done by evaluating the derivatives of $\bar{T}(t)$ and $\hat{I}(t)$ with respect to $\bar{n}_e$. In general, the derivatives of the radiation field to solve the nonlinear non-LTE problems are known as the approximate lambda operators, and the resulting iterative methods are known as approximate lambda iteration (ALI) methods (see Cannon 1973a, 1973b; Scharmer 1981; Olson et al. 1986; Hubeny 2003, and references therein). Particularly for the multilevel case, various approaches have been described in the literature for obtaining the approximate lambda operators. The most important of them are the "complete linearization" technique (see Auer & Mihalas 1969; Scharmer & Carlsson 1985; Carlsson 1991) and the one that is described as the MALI formalism by Rybicki & Hummer (1991, 1992). The former method leads to a slightly faster
convergence; the latter method has recently found the most widespread use, as it is relatively simple to implement. Although direct evaluation of the derivatives appears simple enough, even if we only consider the dependence on local populations, the expressions become quite extensive. We therefore draw on the basic assumption of the MALI formalism and determine the derivative with respect to the local emissivity only. The resulting dependence describes the intensity variations adequately and produces a much simpler expression for the derivative than when all other dependencies are considered. For the acceleration, we follow the preconditioned MALI scheme. Here the main idea is to perform an operator splitting of the nonlinear radiation field term in the rate equation and use the local operator \( \hat{\Psi} \) for the acceleration (Rybicki & Hummer 1991, 1992).

For fixed values of temperature and electron density, we can rewrite Equation (8) so that the system we actually solve has the form

\[
A^* \cdot \mathbf{x} = b^*,
\]

(71)

where the solution we look for is \( \mathbf{x} = \{ n_1, \ldots, n_N \} \), where \( N \) is the total number of atomic energy levels considered. Following the preconditioned MALI scheme, we obtain the nonlinear terms of the form \( n_1 n_m \) in our system. They are linearized by using the “previous iteration solution,” or “old” \( \hat{n} \), for one of the \( \hat{n} \)s and solve the linear system for the “new” \( \hat{n} \). The choice of the particular \( \hat{n} \) that is “old” or “new” determines the speed of the acceleration.

When the temperature and electron densities are not fixed, the nonlinear system needs to be rewritten to include \( \hat{n}_1 \) and \( \hat{T} \).

The modified nonlinear system has the same form as Equation (71), but now we solve for \( \mathbf{x} = \{ \hat{n}_1, \hat{T}, \hat{n}_1, \ldots, \hat{n}_N \} \). This general case leads to additional nonlinear products of the form \( n_1 n_e, n_1 n_m, n_1 T, \) and \( \hat{T} \hat{n}_e \). We use the same technique of linearizing these products by using the previous iteration solution, or old values, for one of the time derivatives in the product.

The form of the nonlinear rate system written in order to solve \( \mathbf{n} \) as in Equation (8) (or rewritten in order to solve \( \mathbf{x} \) as in Equation (71)) is similar to the non-LTE multilevel kinetic equilibrium equation (see Equation (7)) but with a generalized form to take care of the time dependence. The important difference between the kinetic equilibrium equation and the nonequilibrium rate system is that the rate matrix \( A \) and the vector \( b \) both nonlinearly depend on the solution \( \mathbf{n} \) through the radiation field terms of the form \( \hat{J}_{l,l'} \) and \( \hat{J}_{l,l'} \), with \( l \) and \( l' \) being labels of energy level (see below). A detailed description of the time-dependent MALI scheme with the acceleration is given in Appendices A and B. The final expression of the time-dependent rate system is given by

\[
\begin{align*}
\partial_t \mathbf{n} &+ \left\{ \sum_{l'} \hat{n}_{l'} n_e \hat{D}_{l' l} - \sum_{l'} \hat{n}_l n_e \hat{D}_{l l'} \right\} \\
&- \left\{ \sum_{l'} \hat{n}_{l'} (\hat{U}_{l' l} + n_e \hat{U}_{l' l'} + \hat{J}_{l' l} + n_e \hat{J}_{l' l'}) \right\} \\
&- \left\{ \sum_{l'} \hat{n}_l (\hat{U}_{l l'} + n_e \hat{U}_{l l'} + \hat{J}_{l l'} + n_e \hat{J}_{l l'}) \right\} \\
&- \hat{n}_e \left\{ \sum_{l'} (n_e \hat{D}_{l' l} - n_l \hat{D}_{l l'}) + \sum_{l'} n_e (\hat{U}_{l' l} + \hat{J}_{l' l'}) \right\}
\end{align*}
\]

(72)

where \( \hat{n}_{l,n-1} \) is the solution from the previous time step with time index \( n-1 \), and the various other terms are described in Appendices A and B.

In the steady-state case, the time evolution of the solution will settle toward the equilibrium solution. As the solution settles, the remaining difference between the current solution and the equilibrium solution decreases, and the time-step control gradually allows the time steps to become larger. Since the scattering rate is not dependent on the time step, as the time steps increase, more scatterings take place in each time step, resulting in a numerically “stiffer” problem. In the dynamic case, the time step is never allowed to become larger than the dynamic timescale; hence, the number of scatterings remains roughly constant. As a result, in the dynamic case, the system is expected to be less stiff.

The solution \( \hat{n}_l \) of the rate system in Equation (72) is used to update the opacity and emissivity, from which the updated \( \hat{I} \) and \( \hat{I} \) are then computed. Integrating them, the rates can be updated, and Equation (72) can be solved. These two steps are iterated until convergence is obtained. In this way, the time dependence in our formalism is implicit, as the nonlinear time dependence appears through iteration between the formal solver and the rate system.

When the temperature is not fixed, it also becomes part of the solution of the nonlinear rate system (see above). To this end, we combine the atomic rate system with the molecular rate equations (see Section 3.6 below), the energy balance equation (see Section 2.2), and finally the Saha–Boltzmann equations to treat the metal equations. The nonequilibrium treatment in the rate system is always applied to the hydrogen atom and molecules. For all other elements, a provision is made for them to be treated either in LTE or in nonequilibrium similar to the treatment of the hydrogen atom itself.

### 3.6. Nonequilibrium Molecular Rates

Molecular formation/destruction takes place through multiple paths, which together are known as a chemical reaction network. This reaction network for each molecule results in an
additional equation in our rate system that has the general form

\[
\frac{\partial N_{\text{mol,i}}}{\partial t} + (\mathbf{v} \cdot \nabla) N_{\text{mol,i}} = \sum_{l} N_{\text{mol,i}}N_{\text{mol,j}}K_{l}^+ - N_{\text{mol,i}}N_{\text{mol,j}}K_{l}^-, \tag{73}
\]

where \(K_{l}^+\) and \(K_{l}^-\) are the reaction rates for the \(l\)th chemical reaction, and \(N_{\text{mol,i}}\), \(N_{\text{mol,j}}\), \(N_{\text{mol,r}}\), and \(N_{\text{mol,s}}\) are the reactants. We have also treated molecular formation and dissociation of \(\text{H}_2\), \(\text{H}_3^+\), and \(\text{H}^-\) in nonequilibrium by considering several collisional and radiative chemical reactions and their rates. However, in this paper, we restrict ourselves to stationary solutions for fixed molecular densities, metal densities, and temperature. A detailed study of the role of nonequilibrium treatment of these molecules is deferred to a subsequent paper.

### 3.7. Coupling to Hydrodynamics

The coupling of the RT and hydrodynamics is through the mass density \(\rho\), velocity \(\mathbf{v}\), gas pressure \(P\), and radiative flux divergence \(F\). At the beginning of each step, the mass density \(\rho\) per time step is converted into number densities of the total hydrogen in atomic and molecular form and the number densities of other elements assuming their abundance to be fixed. This total hydrogen in all forms is used for the particle conservation equation. From the solution of the RT equation, we have \(I\), which can be readily used to calculate the radiation flux \(F\). From the solution of the rate system, we have \(n_i\), \(\dot{n}_i\), and \(T\); using these, the population \(n_e\), electron density \(n_e\), and temperature \(T\) per time step are updated, and their partial pressures are computed, the total of which forms the total gas pressure \(P\). The velocity \(\mathbf{v}\) per time step is used for advecting the \(\dot{n}_i\) and for the Doppler shifts of the spectral lines. For the MHD part, we use the original Max Planck Institute for Solar System Research, University of Chicago Radiation MHD (MURaM) solver. In the dynamic case, for each time step, the RT and the MHD solvers are iterated until converged. This iteration is the outermost iteration loop in addition to those involving RT quantities. Since none of these are relevant for the stationary case, further details and testing will be provided in a forthcoming paper dedicated to the dynamic case.

### 4. Implementation

The scheme described above is implemented as a module in the MHD simulation code MURaM. The code is written in C++. The module is written in 1D/2D/3D but currently tested only in 1D. The results from the future extension of our work to 2D/3D will be presented in subsequent papers. We stress here that for the current paper, we restrict ourselves to a study of steady-state solutions with the aim of verifying that our RT module works and provides correct results. The RT module iteratively solves the formal solution of the RT equation using a short-characteristics formal solver with a choice of 1D/2D/3D Cartesian geometry, self-consistently and iteratively with the time-generalized Rybicki–Hummer MALI scheme (see Section 3.5, Appendices A and B) that solves the rate system, for a multilevel atom system. The formal solution involves evaluation of the local and propagation parts of \(\bar{I}\) and \(\hat{I}\) in different optical depth regimes. The RT solver is also implemented to handle velocity fields. A flow diagram of the important steps of our RT scheme is presented in Figure 2.

![Flow diagram showing the RT iterative scheme for a given time step.](image)

### 4.1. Numerical Details

An important step in the implementation is that we approximate the time integrals of the time-dependent intensity to obtain \(\bar{I}\) and \(\hat{I}\). As already described in Section 3, this is analogous to the integration of the spatially dependent source function in the original short-characteristics formal solution method. The integrated expressions thus obtained contain exponential integrals that need to be evaluated numerically. Further, for different optical depth \(\tau\) values (e.g., for small \(\tau\)), we expand the polynomial form of \(I(t)\) using various series expansions to avoid numerical cancellations. In these cases, the exponential integrals simplify to become polynomial expressions.
4.2. Time-step Criterion

In this section, we discuss the time-stepping criterion we adapted in our RT scheme. Since we solve the rate system for $n_i$ and $\dot{n}_i$, and we assume a linear time dependence for the populations, we impose a criterion of the form

$$\delta t_{i+1} = f \times \min \{(n_i + \delta t \dot{n}_i)/\dot{n}_i\}, \quad (74)$$

where $i$ and $i + 1$ are the indices of two successive time steps, and a minimum is taken over all of the levels and the spatial dimensions. Here $f$ is a fraction taken to be a free parameter less than unity that determines how crude or fine the time resolution is. For example, $f = 0.01$ means that the change in the population is 1% of the population itself. Since we solve for $\dot{n}_i$ at each time step, we do not know the value of $n_{i+1}$ at the beginning of the $(i + 1)$th time step. Therefore, $\dot{n}_i$ itself is used for estimating $\delta t_{i+1}$. Further, to ensure a smooth evolution, we take a linear combination of two successive $\delta t$ estimates (steps $i - 1$ and $i$) as the actual $\delta t$ for the $i$th step. The time stepping so defined ensures that the change $\delta n_i = \delta t \dot{n}_i$ is small enough compared to $n_i$, which is important for a stable evolution of the system.

At every time step, the value of $\delta t$ is set by the largest $\dot{n}$ over all levels and spatial points. In other words, $\delta t$ is set by the largest change in the system. Once the population $n$ at a given level and spatial point reaches its equilibrium value, the corresponding $\dot{n} \to 0$, and consequently, at this level and spatial point, $\delta t \to \infty$. However, the criterion searches for the smallest $\delta t$, which is now set by the new largest $\dot{n}$ that corresponds to a different level and/or spatial point. In this way, $\delta t$ automatically takes larger and larger values as the $\dot{n}$ at different levels and spatial points reaches its equilibrium values.

In Figure 3, we show the time evolution of the proton density $N_p$ at a fixed column mass using very fine (black line; $f = 0.01$) and intermediate (green line and stars; $f = 0.05$) time stepping. These two curves are visually indistinguishable, as the difference between them is very small. An example of a very large $\delta t$ leading to negative proton density is also shown (blue line and stars). The two vertical lines are drawn to indicate the initial and final time values of this large $\delta t$ (red lines). The zero line is also drawn for reference (purple line). For further details, see Section 4.2.

Figure 3. Illustrative time evolution of the proton density $N_p$ at a fixed column mass using very fine (black line; $f = 0.01$) and intermediate (green line and stars; $f = 0.05$) time stepping. These two curves are visually indistinguishable, as the difference between them is very small. An example of a very large $\delta t$ leading to negative proton density is also shown (blue line and stars). The two vertical lines are drawn to indicate the initial and final time values of this large $\delta t$ (red lines). The zero line is also drawn for reference (purple line).

4.3. Computational Demands

We stress here that the purpose of this paper is to introduce our formalism and show that it provides the correct results; consequently, the code has not yet been extensively optimized. As discussed below, the computational demands of the newly developed nonoptimized RT module are much higher when compared to that of the original MURaM. The increase in the computational costs has two main causes: (1) the increased wavelength dependence of the radiation field and related quantities and (2) the requirement of the use of the long double data type for a precise evaluation of the exponential integrals and solving the resulting rate system. While the former cannot really be optimized extensively, the latter can possibly be dealt with by means of a more appropriate formulation of the relevant expressions.

For the convenience of the discussion, we define CPU time for convergence as the computing time required to complete the convergence cycle and reach a fixed level of accuracy. For this purpose, we define the maximum relative error as

$$R_c = \max_{i,k} \left| \frac{N^{(m,i,k)} - N^{(m-1,i,k)}}{N^{(m,i,k)}} \right|, \quad (75)$$

where $N^{(m,i,k)}$ is the population in the $m$th iteration for the $i$th energy level and $k$th grid point in a given time step. We then impose at each time step the maximum relative error $R_c$ to be less than a given parameter $\epsilon$, which is generally known as the convergence criterion. We used $\epsilon = 10^{-4}$ for the results presented in this paper.

For example, for a 1D atmosphere with an extent of 2.8 Mm, represented using 151 spatial points, using a three-level hydrogen atom with a total of 90 wavelength points (including the line and continuum), the CPU time required by the nonoptimized code is 2 s per iteration per time step on a single core of an Intel Xeon Gold 6150 CPU running at 2.70 GHz. The total time per time step depends on the total number of iterations needed per time step, which is larger when $\delta t$ is larger. Here we chose a fine time resolution of $f = 0.01$. Typically, in the above example, when $\delta t \sim 10^{-3}$, a total
30–40 iterations per time step are needed to reach convergence, resulting in an execution time of about 2 minutes per step.

5. Verification

In this paper, we focus on the stationary solutions. We have thoroughly tested our code for the accuracy of the method and validated it by reproducing the benchmark results from earlier papers. For all of the results presented in this paper, we use the average BIFROST (Gudiksen et al. 2011) atmosphere from one of the publicly available snapshots and keep the density and energy fixed, but we evolve populations. For a fixed mass density and energy at every depth point, we solve the LTE chemical equilibrium equation and also obtain a temperature and pressure consistent with this energy and mass density (solving for the initial equation of state). Other than atomic hydrogen, H, we have hydrogen molecules H$_2$, H$_2^+$, and H$^-$ and several other elements while solving for the initial LTE equation of state. The initial energy, mass density, velocity from the MHD part of the MURaM, temperature, molecular densities, and metal densities from the initial LTE equation-of-state solution are kept fixed for all studies in this paper. A study of the molecular rates and dynamic solution is reserved for separate forthcoming papers.

In Section 5.1, we show that our nonequilibrium solver evolves the populations to an equilibrium solution, which closely resembles the non-LTE kinetic equilibrium solution computed by the well-tested RH code (Uitenbroek 2001). In Section 5.2, the accuracy of the linear approximation of the time dependence of the populations is explored. Finally, in Section 5.3, we reproduce the timescales from previous studies that used a different numerical method (Carlsson & Stein 2002).

5.1. Benchmark Test

In Figure 4, we show the evolution of populations in a three-level H atom setup. We initialize the evolution of populations with their LTE values. When they eventually reach equilibrium, their values correspond to the instantaneous kinetic equilibrium solution of a non-LTE RT problem. To show that we indeed get a correct equilibrium solution, we overplot the solution from the non-LTE spectral synthesis code RH (Uitenbroek 2001), with the same atmospheric structure, atomic data, and background opacity. In addition to the fixed atmospheric structure, in both codes, we fix the electron density computed from an initial LTE equation-of-state solver. We obtain a very good match between the RH code solution and ours for all bound (ground and first excited) and free (ionized) states.

5.2. Accuracy of the Linear Approximation

Left to themselves, atomic populations away from the equilibrium state evolve to reach a steady state (or equilibrium state), which can be mathematically shown to follow an exponential law as a function of time, namely,

$$n(t) \sim a_0 \exp(-a_1 t) + a_2,$$

where $a_0$, $a_1$, and $a_2$ are constants. Using the above form in an RT equation is not practical to solve. Instead, as described in Section 3.1, we use a piecewise linear function to describe the time dependence of the populations. Therefore, the accuracy of this description depends on the time resolution in our numerical setup (in other words, the value of $f$).

Now we analyze the accuracy of the linear time dependence of the populations. We use a two-level atom model for this study,
with only ground and ionized states. We compare the $N_p$ that used time resolutions that were very fine ($f = 0.01$ in Equation (74)) and very crude ($f = 0.5$ and 0.8 in Equation (74)). In other words, $f = 0.01$ corresponds to $\delta N_p = (\delta t)N_p$; namely, the change in $N_p$ is 1% of $N_p$, and $f = 0.5$ ($f = 0.8$) corresponds to the case where $\delta N_p = \delta tN_p$ is 50% (80%) of $N_p$.

These $N_p$ are shown as a function of column mass in Figure 5 (top panels). In the bottom panels, we show the relative errors between the two $N_p$ curves in the top panels. All of the plots correspond to a fixed time where the relative error is maximum. The maximum relative error is $\sim 3\%$ in the left panels and $\sim 4\%$ in the right panels. Thus, even with such extremely crude time resolutions, the method remains stable, producing relative errors of only a few percent.

In Figure 6 (top panels), we plot $N_p$ for the same fine and crude time-stepping criteria as in Figure 5 but as a function of time. In the bottom panels, we plot the relative error in percent between the two $N_p$ curves shown in the top panels. All plots correspond to a fixed column mass where the relative error is maximum. We observe here that, once $N_p$ reaches its equilibrium value, $N_p$ computed using the crude time-stepping criterion oscillates around the value of $N_p$ computed using the fine time-stepping criterion at alternate time steps. As a consequence, as seen in the bottom panels, the relative error
also oscillates, which never settles to zero. This is in contrast to a general expectation for the amplitude of the oscillations and the corresponding error to become smaller and smaller with time. We attribute this behavior of the error to the implicit interdependence of the solution $\dot{n}$ and the time step $\delta t$ as defined by the time-stepping criteria (Equation (74)).

To understand this behavior of the error, in Figure 7, we plot the evolution of the relative errors between fine ($f = 0.01$) and crude ($f = 0.5$ and 0.8) for the top and bottom panels, respectively) time stepping with both cases having an upper limit on the time step of $\delta t_{\text{max}} = 100$ (panel (a)) and $\delta t_{\text{max}} = 500$ (panel (b)). We observe here that when the time step is fixed to a given value, the relative error continues to oscillate but with an amplitude that gradually decreases with time, and it saturates to a smaller value. This saturation value of the error is smallest for $f = 0.5$, $\delta t_{\text{max}} = 100$, which corresponds to the finest time stepping among all four crude time-stepping criteria shown in this figure. Thus, the saturation value of the error determines the accuracy of the time stepping.

Thus, we show here that the assumption of the linear approximation of the time dependence of populations in our method produces solutions that are accurate and robust against changes in the time resolution.

5.3. Ionization/Recombination Time Scales

In Figure 8, we show different temperature structures and the corresponding timescale of ionization/recombination as a function of column mass. We follow the same approach as Carlsson & Stein (2002). We consider a two-level atom model with only a ground state and an ionized state. We start with an LTE solution and let our nonequilibrium solver reach equilibrium for a fixed atmospheric structure that defines an initial equilibrium proton density $N_p(0)$. We perturb the temperature by increasing its value by 1% throughout the atmosphere. We then let the populations settle to this new temperature structure. This equilibrium proton density is denoted as $N_p(\infty)$. The time evolution of the proton density from the initial state to the final state is represented by $N_p(t)$. We recall that the physical relaxation timescale, denoted here as $t_s$, of any system is defined as the time it takes to change by a value of $e$. In particular, for computing a relaxation timescale of $N_p$, we perform a least-squares linear fit to the function

$$\frac{N_p(t) - N_p(\infty)}{N_p(0) - N_p(\infty)} = \exp(q_0 + q_1 t),$$

where $q_0$ and $q_1$ are coefficients of the least-squares fitting with the relaxation timescale defined as $t_s = -1/q_1$ (see Carlsson & Stein 2002).

We find that for various atmospheric structures that we have chosen (density, energy, and corresponding temperature), the maximum timescales can vary by orders of magnitude, from $\sim 100$ to $\sim 10^4.5$ in the mid-chromosphere and higher. This dependence of the timescales on the atmospheric structure is caused by the strong density dependence of the collision rates that determine the timescales (see Carlsson & Stein 2002, for a detailed discussion). The maximum timescale that we obtain is $\sim 10^5$ (shown as dashed blue lines in Figure 8), which is quite close to the long timescales of $\sim 10^7$ obtained by Carlsson & Stein (2002, Figure 6). However, the timescales are small when we are in the photosphere due to equilibrium conditions.

6. Summary

In this paper, we focus our attention on developing a numerical method to solve the non-LTE nonequilibrium RT problem through (a) a proper time-dependent treatment of the radiation field, (b) a proper nonequilibrium treatment of the molecular chemistry, and (c) the development of a time-implicit numerical scheme. The method is based on an integral equation approach to the RT equation that involves a generalization to the time dimension of (i) the short-characteristics technique for the formal solution of the RT equation and the (ii) MALI technique to solve the nonlinear rate system.
We validate our newly developed method with two important benchmark tests: (i) we start with LTE populations on a fixed atmospheric structure, allow them to evolve to the equilibrium solution, and verify that this agrees with the kinetic equilibrium solution obtained from the RH code (Uitenbroek 2001); and (ii) we show that the physical timescales required to reach equilibrium are similar to those obtained by Carlsson & Stein (2002), who used a different numerical method. We also show that the solver remains stable and the solution is robust against changes in the time resolution.

The final aim of this work is to integrate this module with the radiation MHD code MURaM to carry out fully dynamic evolution of the MHD and RT quantities. This work is in progress, and the studies of the dynamic solution will be presented in a subsequent paper.

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Appendix

Here we present some important equations for solving the time-dependent rate system. For simplicity of notation, we omit the dependence of all of the quantities on $\Omega$ and $\nu$ in general and write it only when necessary. We start by presenting the rate equations in Appendix A, the solution of which must generally be obtained by means of an iterative solver, after which, in Appendix B, we derive the expressions needed for the acceleration of the convergence of the iterative solver by means of operator splitting, a method commonly referred to as ALI (see Cannon 1973a, 1973b; Scharmer 1981; Olson et al. 1986; Hubeny 2003, and references therein). Specifically, we implement here a variation on the MALI scheme (Rybicki & Hummer 1991, 1992). For a given time step, we iteratively solve for $\dot{n}$, $\dot{n}_{\text{e}}$, and $\dot{T}$, where we note that the nonlinear dependence of the radiation field $I(t)$ on the temperature is treated implicitly, by using “old” temperature derivatives $\dot{T}$ (which are solved for, iteratively, in the previous iteration) while solving for the current $\dot{n}$.

Appendix A

Time-dependent Rate Equations

We can write the time-dependent rate system as

$$
\sum_l n_l(t) n_e(t) C(T(t)) + \int d\Omega \sum_l \rho \frac{d\Omega}{d\nu} \left[ n_l(t) U_{l,t}^\nu (T(t)) + n_l(t) n_e(t) U_{l,t}^{\nu \nu}(T(t)) + n_l(t) V_{l,t}^\nu (T(t)) I(t) + n_l(t) n_e(t) V_{l,t}^{\nu \nu}(T(t)) I(t) \right] - \sum_l n_l(t) n_e(t) C_{l,t}(T(t)) - \sum_l \int d\Omega \sum_l \rho \frac{d\Omega}{d\nu} \left[ n_l(t) U_{l,t}^\nu (T(t)) + n_l(t) n_e(t) U_{l,t}^{\nu \nu}(T(t)) + n_l(t) V_{l,t}^\nu (T(t)) I(t) + n_l(t) n_e(t) V_{l,t}^{\nu \nu}(T(t)) I(t) \right] = \dot{n}_l \delta t.
$$

(A1)

Figure 8. Temperature stratification (top) and equilibrium timescale $t_e$ (bottom) for a number of different atmospheric structures. The case with the maximum timescale and corresponding temperature structure is shown as a blue dashed line.
on \( n(t) \) and \( T(t) \). The function \( C_{\ell l}(T(t)) \) is the temperature-dependent collision rate coefficient, and the functions \( U_{\ell l}(t), \ U^*_{\ell l}(t), \ V_{\ell l}(t), \) and \( V^*_{\ell l}(t) \) are time-generalized versions of the \( U \) and \( V \) functions from Rybicki & Hummer (1992), defined as follows. For line transitions between \( l \) and \( l' \), we have

\[
U_{\ell l}(\Omega, \nu, t) = \begin{cases} \frac{h \nu}{4 \pi} A_{\ell l}(\nu, t), & l > l', \\ 0, & l < l', \end{cases}
\]

\[
U^*_{\ell l}(\Omega, \nu, t) = 0,
\]

\[
V_{\ell l}(\Omega, \nu, t) = \frac{h \nu}{4 \pi} B_{\ell l}(\nu, t),
\]

\[
V^*_{\ell l}(\Omega, \nu, t) = 0,
\]

where \( A_{\ell l} \) and \( B_{\ell l} \) are the Einstein coefficients and \( \varphi_{\ell l} = \varphi_{\ell l} \) is the line profile function. Here \( h \) and \( k_b \) are Planck’s constant and Boltzmann’s constant, respectively. Here \( V_{\ell l} \) has the same form for both \( l > l' \) and \( l < l' \), although depending on whether it is an upward or a downward transition, we need to use the appropriate Einstein \( B \) coefficient.

Similarly for a continuum transition between \( l \) and \( l' \), we have

\[
U_{\ell l}(\nu, t) = 0,
\]

\[
U^*_{\ell l}(\nu, t) = \begin{cases} \Phi_{\ell l}(T(t)) \frac{2 h \nu^3}{C^2} e^{-\frac{E_i}{k_b T(t)}}, & l > l', \\ 0, & l < l', \end{cases}
\]

\[
V_{\ell l}(\nu, t) = \begin{cases} 0, & l > l', \\ a_{\ell l}(\nu), & l < l', \end{cases}
\]

\[
V^*_{\ell l}(\nu, t) = \begin{cases} \Phi_{\ell l}(T(t)) e^{-\frac{E_i}{k_b T(t)}} a_{\ell l}(\nu), & l > l', \\ 0, & l < l', \end{cases}
\]

where \( C \) is the speed of light, \( a_{\ell l}(\nu) \) is the photoionization cross section, and \( \Phi(T(t)) \) is the Saha–Boltzmann factor, given by

\[
\Phi(T(t)) = \frac{g_{\ell l}}{2 \pi} \left( \frac{h^2}{2 \pi m_e k_b T(t)} \right)^{\frac{3}{2}} \exp \left[ \frac{(E_i - E_\ell)}{k_b T(t)} \right].
\]

Thus, the time-dependent radiative rate coefficient \( R_{\ell l} \) is defined as

\[
R_{\ell l} = \int d \nu \int d \Omega \frac{d \nu}{h \nu} \left[ (U_{\ell l}(\Omega, \nu, t) + n_\ell(t) U^*_{\ell l}(\Omega, \nu, t)) + (V_{\ell l}(\Omega, \nu, t) + n_\ell(t) V^*_{\ell l}(\Omega, \nu, t)) \right] O_{\ell l}(\nu),
\]

defined for both lines and continua and for transitions involving both \( l > l' \) and \( l < l' \).

First, we express all of the time-dependent quantities except \( I(t) \) in the form of linear polynomials as follows,

\[
\begin{align*}
n_\ell(t) &= n_e + t \, \delta n_e, \\
n_\ell(t) &= n_\ell(t) + \delta n_\ell, \\
\varphi(t) &= \varphi + \delta \varphi, \\
T(t) &= T + t \, \delta T,
\end{align*}
\]

and so on. For simplicity, the terms corresponding to zeroth order of \( t \) (namely, \( \delta^0 \)) are denoted by the same notation as the original time-dependent function. For example, the \( \delta^0 \) term for the temperature function \( T(t) \) is denoted as \( T \) itself. By substituting the above expansion in collisional and radiative rate integrals, we can separate the terms containing \( \delta^0 \) and \( \delta^1 \) in these rate equations. We can rewrite the rate system as

\[
\begin{align*}
\sum_{\ell l} & \left\{ (n_\ell \delta n_e + (\delta n_\ell n_e + \delta n_\ell \delta n_e)) C_{\ell l} \\
& + (n_\ell n_e + t (\delta n_\ell n_e + \delta n_\ell \delta n_e)) T \frac{\partial C_{\ell l}}{\partial T} \right\} \\
& + \sum_{\ell l} \delta t \left[ \int d\Omega \frac{d\nu}{h \nu} \left( n_\ell U_{\ell l} + t (\delta n_\ell U_{\ell l} + n_\ell \frac{\partial U_{\ell l}}{\partial T} \right) \\
& + (n_\ell n_e + t (\delta n_\ell n_e + \delta n_\ell \delta n_e)) U_{\ell l} \\
& + (n_\ell n_e + t (\delta n_\ell n_e + \delta n_\ell \delta n_e)) T \frac{\partial U_{\ell l}}{\partial T} \right] I(t) \\
& - \sum_{\ell l} \left( n_\ell \delta n_e + (\delta n_\ell n_e + \delta n_\ell \delta n_e)) C_{\ell l} \\
& - \sum_{\ell l} \left( n_\ell n_e + t (\delta n_\ell n_e + \delta n_\ell \delta n_e)) T \frac{\partial C_{\ell l}}{\partial T} \right) \\
& - \sum_{\ell l} \delta t \left[ \int d\Omega \frac{d\nu}{h \nu} \left( n_\ell U_{\ell l} + t (\delta n_\ell U_{\ell l} + n_\ell \frac{\partial U_{\ell l}}{\partial T} \right) \\
& + (n_\ell n_e + t (\delta n_\ell n_e + \delta n_\ell \delta n_e)) U_{\ell l} \\
& + (n_\ell n_e + t (\delta n_\ell n_e + \delta n_\ell \delta n_e)) T \frac{\partial U_{\ell l}}{\partial T} \right] I(t)
\end{align*}
\]

\[
A3
\]
Ignoring all terms that are second order or higher in time, we have
\[
\sum_{l'} \left\{ (n_{l'} n_{e} + t(n_{l'} n_{e} + n_{e} n_{l'})) C_{l' l} \\
+ (n_{l'} n_{e}) \frac{\partial C_{l' l}}{\partial t} \right\} \\
+ \sum_{l'} \oint d\Omega \frac{d\nu}{\nu} \left\{ n_{l'} U_{l' l} + t \left( \frac{\partial U_{l' l}}{\partial t} + n_{l'} \frac{\partial U_{l' l}}{\partial t} \right) \right\} \\
+ (n_{l'} n_{e} + t(n_{l'} n_{e} + n_{e} n_{l'})) U_{l' l}^* + (n_{l'} n_{e}) \frac{\partial U_{l' l}^*}{\partial t} \right\} \\
= \sum_{l'} \left\{ (n_{l'} n_{e} + t(n_{l'} n_{e} + n_{e} n_{l'})) C_{l' l} \\
+ (n_{l'} n_{e}) \frac{\partial C_{l' l}}{\partial t} \right\} \\
= n_{e} \delta t.
\]

We now integrate with respect to time, frequency, and angle and reorganize to obtain
\[
\dot{n}_{e} \delta t - \left\{ \sum_{l'} \hat{n}_{l'} n_{e} \hat{D}_{l' l} - \sum_{l'} \hat{n}_{l'} n_{l'} \hat{D}_{l' l} \right\} \\
- \left\{ \sum_{l'} \hat{n}_{l'} (\hat{U}_{l' l} + n_{l'} \hat{U}_{l' l}^* + \hat{J}_{l' l} + n_{e} \hat{J}_{l' l}^*) \\
- \sum_{l'} \hat{n}_{l'} (\hat{U}_{l' l} + n_{l'} \hat{U}_{l' l}^* + \hat{J}_{l' l} + n_{e} \hat{J}_{l' l}^*) \right\} \\
- \sum_{l'} \hat{n}_{l'} \left\{ \hat{U}_{l' l} - \hat{U}_{l' l}^* - \hat{J}_{l' l} - \hat{J}_{l' l}^* \right\} \\
\dot{\hat{D}}_{l' l} = \int_{0}^{\delta t} d t \ C_{l' l},
\]
\[
\dot{\hat{U}}_{l' l} = \int_{0}^{\delta t} d t \ U_{l' l},
\]
\[
\dot{d} \hat{D}_{l' l} = \int_{0}^{\delta t} d t \ \frac{\partial C_{l' l}}{\partial t}, \quad (A6)
\]
\[
\dot{\hat{U}}_{l' l} = \int_{0}^{\delta t} d t \ U_{l' l},
\]
\[
\dot{d} \hat{U}_{l' l} = \int_{0}^{\delta t} d t \ \frac{\partial U_{l' l}}{\partial t}, \quad (A7)
\]
\[
\dot{\hat{J}}_{l' l} = \int_{0}^{\delta t} d t \ V_{l' l} I(t),
\]
\[
\dot{\hat{J}}_{l' l} = \int_{0}^{\delta t} d t \ \frac{\partial V_{l' l}}{\partial t} I(t), \quad (A8)
\]
\[
\dot{\hat{J}}_{l' l} = \int_{0}^{\delta t} d t \ V_{l' l} I(t),
\]
\[
\dot{d} \hat{J}_{l' l} = \int_{0}^{\delta t} d t \ \frac{\partial V_{l' l}}{\partial t} I(t). \quad (A9)
\]
\[
\dot{\hat{J}}_{l' l} = \int_{0}^{\delta t} d t \ V_{l' l} I(t),
\]
\[
\dot{d} \hat{J}_{l' l} = \int_{0}^{\delta t} d t \ \frac{\partial V_{l' l}}{\partial t} I(t). \quad (A10)
\]

Appendix B
Acceleration: Time-dependent Preconditioned MALI Scheme

We follow the preconditioning approach by Rybicki & Hummer (1992) to treat nonlinearities in the rate system and generalize the method for the time-dependent case. Applying operator splitting on the radiation field leads to
\[
I_{\Omega, \nu, l} = \Psi_{\Omega, \nu, l}[t \ \hat{n}_{\Omega, \nu}] + (\Psi_{\Omega, \nu, l} - \Psi_{\Omega, \nu, l})[t \ \hat{n}_{\Omega, \nu}]^\dagger. \quad (B1)
\]
Integrating over time, we have
\[
I_{\Omega, \nu} = \Psi_{\Omega, \nu}[\hat{n}_{\Omega, \nu}] + (\Psi_{\Omega, \nu} - \Psi_{\Omega, \nu})[\hat{n}_{\Omega, \nu}]^\dagger. \quad (B2)
\]
and

\[ \dot{h}_{\omega \nu} = \hat{\psi}^*_{\omega \nu} [\dot{h}_{\omega \nu}] + (\hat{\psi}^*_{\omega \nu} - \hat{\psi}^*_{\omega \nu}) [\dot{h}_{\omega \nu}], \]  
(B3)

where

\[ \dot{h}_{\omega \nu} = \sum_{l' \neq l} \dot{n}_l (U_{l', \omega} + n_e U_{l', \nu} - T \sum_{l' \neq l} n_l \frac{\partial U_{l', \omega}}{\partial T} + n_e \frac{\partial U_{l', \nu}}{\partial T} + \dot{n}_e \sum_{l} U_{l', \nu}). \]  
(B4)

Substituting these expressions in Equation (A5) and simplifying, we obtain

\[ \dot{n}_i \delta t = - \left\{ \sum_l (\dot{n}_i n_e B_{l, i} - \sum_{l'} \dot{n}_l n_e B_{l', i}) \right\} - \left\{ \sum_l (\dot{n}_i (U_{l, i} + n_e U_{l, \nu} + J_{l, i} + n_e J_{l, \nu} - \sum_l \dot{n}_i (U_{l, \nu} + n_e U_{l, \nu} + J_{l, \nu} + n_e J_{l, \nu}) \right\} - \dot{\hat{\psi}} \left\{ \sum_l (n_l \dot{n}_e \dot{D}_{l, i} - n_l \dot{D}_{l, \nu}) \right\} + \sum_l \dot{n}_i (dU_{l, i} + n_e dU_{l, \nu} + dJ_{l, i} + n_e dJ_{l, \nu}) - n_l (dU_{l, \nu} + n_e dU_{l, \nu} + dJ_{l, \nu} + n_e dJ_{l, \nu}) + \sum_l \dot{n}_i \hat{\psi}_{l, i} + (\hat{\psi}_{\text{Vla}, i} + \hat{\psi}_{\text{Vla}, \nu}) + \hat{\psi}_{\text{Vla}, \nu} \right\} \]

\[ \right\} = \sum_l (n_l \dot{n}_e \dot{D}_{l, i} - n_l \dot{D}_{l, \nu}) + \sum_l (n_l \dot{U}_{l, i} + n_l \dot{U}_{l, \nu} + n_l \dot{J}_{l, i} + n_l \dot{J}_{l, \nu}) - (n_l \dot{U}_{l, \nu} + n_l \dot{U}_{l, \nu} + n_l \dot{J}_{l, \nu} + n_l \dot{J}_{l, \nu}) + \hat{\psi}_{\text{Vla}, i} + \hat{\psi}_{\text{Vla}, \nu} \right\} \]  
(B5)

where

\[ \hat{\psi}_{l, i} = \left\{ (V_{l, i} + n_e V_{l, \nu}^*) \hat{\psi}^* \right\} \]

\[ + \left\{ \sum_{m, m'} n_m (V_{l, m} + n_e V_{l, m}^*) - n_l (V_{l, m} + n_e V_{l, m}^*) \right\} \hat{\psi}^* \]

\[ \times \left\{ \sum_{m, m'} U_{l, m}^* + n_e U_{l, m}^* \right\} \]

\[ - \left\{ \sum_{m, m'} n_m (V_{l, m} + n_e V_{l, m}^*) - n_l (V_{l, m} + n_e V_{l, m}^*) \right\} \hat{\psi}^* \]

\[ \times \left\{ \sum_{m, m'} U_{l, m}^* + n_e U_{l, m}^* \right\}. \]  
(B6)

\[ \hat{\psi}_{\text{Vla}, i} = \dot{n}_i \left\{ \sum_{m} n_l V_{l, m} - n_m V_{m, i} \right\} \hat{\psi}^* \left\{ \sum_{m} U_{l, m}^* + n_e U_{l, m}^* \right\}. \]  
(B7)

\[ \hat{\psi}_{\text{Vla}, \nu} = \dot{n}_i \left\{ \sum_{m} n_l V_{l, m} - n_m V_{m, \nu} \right\} \hat{\psi}^* \left\{ \sum_{m} U_{l, m}^* + n_e U_{l, m}^* \right\}. \]  
(B8)

\[ \hat{\psi}_{\text{Vla}, \nu} = \dot{n}_i \left\{ \sum_{m} n_l V_{l, m} - n_m V_{m, \nu} \right\} \hat{\psi}^* \left\{ \sum_{m} U_{l, m}^* + n_e U_{l, m}^* \right\}. \]  
(B9)

\[ \hat{\psi}_{\text{Vla}, \nu} = \dot{n}_i \left\{ \sum_{m} n_l V_{l, m} - n_m V_{m, \nu} \right\} \hat{\psi}^* \left\{ \sum_{m} U_{l, m}^* + n_e U_{l, m}^* \right\}. \]  
(B10)

\[ \hat{\psi}_{\text{Vla}, \nu} = \dot{n}_i \left\{ \sum_{m} n_l V_{l, m} - n_m V_{m, \nu} \right\} \hat{\psi}^* \left\{ \sum_{m} U_{l, m}^* + n_e U_{l, m}^* \right\}. \]  
(B11)
\[ \hat{\Psi}_{\text{He},l,l'} = \left\{ \sum_{\ell} \left( n_l \left( \frac{\partial V_{\ell l'}}{\partial T} + n_e \frac{\partial V_{\ell l'}}{\partial T} \right) + n_e \phi \frac{\partial V_{\ell l'}}{\partial T} \right) \right\} \]

\[ - n_e \left( \frac{\partial V_{\ell l'}}{\partial T} + n_e \phi \frac{\partial V_{\ell l'}}{\partial T} \right) \]

\[ \times \Psi^* \left( \sum_{m,m'} n_m \left( \frac{\partial U_{m,m'}}{\partial T} + n_e \phi \frac{\partial U_{m,m'}}{\partial T} \right) \right) \]

\[ \times \left( \sum_{m,m'} n_m \left( \frac{\partial U_{m,m'}}{\partial T} + n_e \phi \frac{\partial U_{m,m'}}{\partial T} \right) \right) \]

\[ \times \left( \sum_{m,m'} n_m \left( \frac{\partial U_{m,m'}}{\partial T} + n_e \phi \frac{\partial U_{m,m'}}{\partial T} \right) \right) \]