

TRANSPORT OF HIGH-INTENSITY RESONANT RADIATION

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A method is proposed for solving several nonlinear radiation-transport problems by reducing them to linear ones. This is attained by introducing a variable photon free path that depends on the radiation density at a point. We consider the case of resonance scattering of light in the approximation of complete frequency mixing. It is assumed that the distortion produced in the spectral lines at large radiation densities can be neglected.

1. RADIATION transport phenomena have been the subject of a large number of papers, with particular attention paid to cases when the scattering by one center is elastic.^[1, 2] Then there exists a constant mean free path, and at distances much larger than this path the particle propagation can be regarded as diffuse. This is not the case if the scattering is incoherent, i.e., is accompanied by change in the energy of the incident particle. The most typical case is resonance scattering of light. Here, if we neglect non-resonant processes, the mean free path far from resonance tends to infinity. This means that the diffusion approximation is nowhere applicable.^[2, 3] It is then necessary to solve the exact transport equation in the entire region.

Allowance for nonresonant processes leads, of course, to a limit on the mean free path, but if the cross section of these processes is small in the center of the line, then we can expect their neglect not to influence strongly the behavior of the radiation at the center of the line in the case of excited atoms at not too large distances from the boundaries or from the source. This assumption is confirmed by our calculations,^[4] if the nonresonant process is taken to be absorption of light by extraneous impurities.

The methods for accurately solving the stationary transport equation in the case of resonance scattering were developed for semi-infinite space and plane-parallel layers.^[2, 5, 6] No attempts were made, however, to solve exactly the transport equation with allowance for stimulated emission, a factor that becomes important at large light intensity. In some problems (luminescence) the need for taking stimulated emission into account arises at readily attainable light fluxes. It is easy to see that the presence of stimulated emission leads to a dependence of the photon mean free

path on the radiation density at a point, and consequently on the coordinates. This means that the inhomogeneity of the medium, which results from the fact that there are two kinds of atoms, in the ground and excited states, becomes significant.

This paper is devoted to an exposition of a method for exactly solving the stationary transport equation with allowance for stimulated emission.

2. As usual, we confine ourselves to a homogeneous medium consisting of two-level atoms, which emit light in resonance fashion. We neglect all the nonresonant processes, including elastic scattering. We assume the medium to be infinite in the X and Y directions and assume all quantities to depend on one coordinate z. Then the radiation transport equation takes the form

$$c\mu \frac{\partial I_\nu(z, \mu)}{\partial z} = -k_\nu I_\nu(z, \mu) (N - n^*) + \left(\frac{\kappa_\nu}{4\pi} + k_\nu I_\nu(z, \mu) \right) + n^* + f_\nu^*(z, \mu), \tag{1}$$

where I_ν is the density of photons of frequency ν , emitted at an angle $\vartheta = \cos^{-1} \mu$ to the z axis, k_ν is the absorption and stimulated emission coefficient, κ_ν is the spontaneous emission coefficient, N the total density of the atoms of the medium, n^* the density of the excited atoms, c the velocity of light, and f_ν^* the density of the radiation sources. Thus, if a radiating plane is located at a distance z_0 , then $f_\nu^*(z, \mu) = \varphi_\nu(\mu) \delta(z - z_0)$ (the function $\varphi_\nu(\mu)$ takes into account the dependence of the distribution of the emitted photons on the frequency and on the direction). In some cases, instead of introducing the function f_ν^* into the transport equation, an appropriate boundary condition is imposed on the photon density.^[1, 2]

Equation (1) was obtained assuming complete redistribution over the frequencies,^[2] when the probability that the atom will emit a quantum of

definite frequency does not depend on the frequency or the direction of the quantum exciting the atom. This condition is well satisfied in the case of homogeneous line broadening, for example by collisions. When the line shape is determined by the Doppler effect, a correlation exists between the emitted and absorbed photons.^[2]

The kinetic equation for the density of the excited atoms is of the form

$$\begin{aligned} & \frac{N - n^*}{2} \int_0^\infty k_\nu d\nu \int_{-1}^1 I_\nu(z, \mu) d\mu \\ &= n^* \left[\frac{1}{4\pi} \left(\int_0^\infty \kappa_\nu d\nu + \sigma \right) + \frac{1}{2} \int_0^\infty k_\nu d\nu \int_{-1}^1 I_\nu(z, \mu) d\mu \right]. \quad (2) \end{aligned}$$

The first term in (2) describes the increase in the number of excited atoms due to resonance absorption, and the second describes the loss due to spontaneous emission, collision quenching (σ), and stimulated emission.

In writing out (2) we did not take into account the processes of excitation and ionization by collisions without photon participation. The relative contribution of these processes is small at the relatively high photon densities considered in this paper. We likewise disregarded the flux term $\nu\mu\partial n^*/\partial z$, which is always permissible if there are no external forces to produce a flux of the entire medium.

We introduce the following notation:

$$x = \frac{z}{c} N, \quad n = \frac{n^*}{N}, \quad f_\nu(x, \mu) = \frac{1}{N} f_\nu^*(x, \mu). \quad (3)$$

Then Eqs. (1) and (2) take the form

$$\mu \frac{\partial I_\nu(x, \mu)}{\partial x} = -k_\nu I_\nu(x, \mu) (1 - 2n(x)) + \frac{\kappa_\nu}{4\pi} n(x) + f_\nu(x, \mu), \quad (4)$$

$$\begin{aligned} \frac{n(x)}{1 - 2n(x)} &= 2\pi \left(\int_0^\infty \kappa_\nu d\nu + \sigma \right)^{-1} \int_0^\infty k_\nu d\nu \int_{-1}^1 I_\nu(x, \mu) d\mu \\ &= \Phi(x). \quad (5) \end{aligned}$$

We shall henceforth assume that k_ν does not depend on the coordinate.

At a high degree of excitation and at high radiation densities, the properties of the medium, in general, change strongly. The line shape also changes. A study of the obtained relationships is beyond the scope of the present paper. We confine ourselves to low temperatures, at which the degree of ionization is small. We assume that the main processes leading to a broadening of the level under consideration are atomic collisions. At not too high radiation densities, the collision broadening prevails over broadening under the influence of the radiation. Under these conditions we can dis-

regard the distortion of the line shape under the influence of the radiation. We shall not present here estimates of the limits of applicability of these assumptions since they can be readily obtained.

3. Equations (4) and (5) contain nonlinear terms describing the influence of the degree of excitation of the medium on the absorption and emission processes. When $n(x) \ll 1$, they go over into the well known linear equations in which stimulated emission is not taken into account.

The equations presented differ from the corresponding linear equations by a factor $1 - 2n(x)$, which corresponds to a local increase of the mean free path of a photon of given frequency. In transport problems, a natural measure of the length is the photon mean free path at the center of the line. This suggests the following substitution:

$$dX/dx = 1 - 2n(x). \quad (6)$$

Assuming that all functions depend on the new variable X , we obtain the equation

$$\mu \frac{\partial I_\nu(X, \mu)}{\partial X} = -k_\nu I_\nu(X, \mu) + \frac{\kappa_\nu}{4\pi} \Phi + \frac{\psi_\nu(X, \mu)}{1 - 2n(X)}, \quad (7)$$

which is already linear, but has an unknown source function.

The transformation (6) is mutually unique if $1 - 2n(x) > 0$. As seen from (5), this condition is satisfied. Then

$$\delta(x - x_0) = |dX/dx| \delta(X - X_0) = [1 - 2n(X_0)] \delta(X - X_0).$$

Let us consider first a flat source of photons. Then $f_\nu(x, \mu) = \varphi_\nu(\mu) \delta(x - x_0)$ and

$$\psi_\nu(X, \mu) (1 - 2n)^{-1} = \varphi_\nu(\mu) \delta(X - X_0) = f_\nu(X, \mu). \quad (7a)$$

Substituting (7a) in (7) we obtain, in terms of the variables (X, μ) , the same equation as is derived from (4) and (5) when $n(x) \ll 1$, i.e., when stimulated emission is neglected.

This transport equation was analyzed many times, and it is well known that the function $\Phi(X)$ satisfies the following integral equation (for an infinite medium):

$$\Phi(X) = \int_{-\infty}^{\infty} K(X - Y) \Phi(Y) dY + F(X), \quad (8)$$

where

$$K(X) = \left[2 \left(\int_0^\infty \kappa_\nu d\nu + \sigma \right) \right]^{-1} \int_0^\infty d\nu \kappa_\nu k_\nu \int_1^\infty \frac{e^{-k_\nu |X| u}}{u} du, \quad (9)$$

$$F(X) = 2\pi \left(\int_0^\infty \kappa_\nu d\nu + \sigma \right)^{-1} \int_0^\infty k_\nu d\nu \int_0^1 \frac{d\mu}{\mu} \left\{ \varphi_\nu(\mu) \right\}$$

$$\begin{aligned} & \times \exp\left[-\frac{k_v}{\mu}(X - X_0)\right] \theta(X - X_0) \\ & + \varphi_v(-\mu) \exp\left[\frac{k_v}{\mu}(X - X_0)\right] \theta(X_0 - X) \Big\}, \\ & \theta(X) = \begin{cases} 1, & X > 0 \\ 0, & X < 0 \end{cases}. \end{aligned} \quad (10)$$

For $n(x) \ll 1$ we have $X \approx x$ and $\Phi(x) \approx n(x)$. Then Eq. (8) expresses the fact that the total number of excited atoms consists of the atoms excited directly by the radiation from the source (free term) and atoms excited by scatter radiation (the integral term).

Equation (6) can be rewritten in the form

$$dx = [1 - 2n(X)]^{-1} dX = [1 + 2\Phi(X)] dX,$$

or

$$x = X + 2 \int_0^x \Phi(X) dX. \quad (11)$$

The obtained system should be solved in the following sequence. We solve first Eq. (8). Carrying out the integration in (11), we determine $x(X)$ from the known function $\Phi(X)$. We invert the obtained function and substitute $X(x)$ into $\Phi(X)$. The obtained solution contains an unknown parameter X_0 , which must be determined from the equation $x(X_0) = x_0$. The density of the excited atoms as a function of the coordinate x is determined from the formula

$$n(x) = \frac{\Phi(X(x))}{1 + 2\Phi(X(x))}. \quad (12)$$

We note that $X \leq x$ everywhere. The photon density and the excited-atom concentration change more slowly than in the linear approximation. This circumstance has a simple physical meaning, connected with the fact that the increase in the excited-atom density leads to a lowering of the photon-absorption probability. A local increase takes place in the mean free path.

We consider now two plane sources located at the points x_1 and x_2 . The transformation (6) transforms them into two sources having the same intensity as before, but situated at the points X_1 and X_2 . The superposition principle, which follows from the linearity of (8), enables us to write the solution in this case in the form of a sum of solutions for a single source. The nonlinearity of the problem is manifest in the change in the distance between the sources. As follows from (11), $|X_1 - X_2| < |x_1 - x_2|$. Thus, the simultaneous influence of the sources reduces to a decrease in the distance between them. The latter is determined

after solving the linear problem from the system of algebraic equations

$$x(X_1) = x_1, \quad x(X_2) = x_2. \quad (13)$$

The foregoing pertains to an arbitrarily distributed source. In this case it will be necessary to solve in lieu of (13) the nonlinear integral equation

$$x = X(x) + 2 \int_{-\infty}^{\infty} G(X(x), Y(y)) F(y) dy, \quad (14)$$

where $G(X, Y)$ is the Green's function of Eq. (8), connected with its resolvent $R(X, Y)$ by the relation $G(X, Y) = \delta(X - Y) + R(X, Y)$. The resolvent is the solution of (8) with an isotropic plane source at the point y , i.e.,

$$f_v^{(R)}(x, \mu) = \frac{\kappa_v}{4\pi} \delta(x - y), \quad F^{(R)}(X) = K(X - Y),$$

$F(X)$ is obtained by generalizing formula (10) to include the case of distributed sources. Equation (14) must be solved numerically.

The method described above reduces the problem with a plane source to a linear problem. As regards the linear problem itself, it has an analytic solution in the case of infinite and semi-infinite media. For layers of finite thickness, the solution can be obtained only by numerical means. With regards to the latter case, we confine ourselves to the following remark. The transformation (11) corresponds to compression of the coordinate system. Then the layer thickness in the linear problem equivalent to our nonlinear problem is smaller. Thus, the layer is more transparent with respect to radiation of higher intensity.

4. We consider the solution of the problem in the simplest case of a plane source in an infinite space. Equation (8) is solved with the aid of a Fourier transformation. We have

$$\Phi(p) = F(p) / (1 - K(p)), \quad (15)$$

where

$$\begin{aligned} F(p) = & \frac{2\pi\lambda}{\kappa} \int_0^{\infty} k_v dv \int_{-1}^1 \frac{d\mu}{\mu} \left[\varphi_v(\mu) \left(-ip + \frac{k_v}{\mu} \right)^{-1} + \varphi_v(-\mu) \right. \\ & \left. \times \left(ip + \frac{k_v}{\mu} \right)^{-1} \right], \end{aligned} \quad (16)$$

$$K(p) = \frac{\lambda}{\kappa p} \int_0^{\infty} k_v \kappa_v \operatorname{arctg} \frac{p}{k_v} dv, \quad \lambda = \bar{\kappa}(\bar{\kappa} + \sigma)^{-1}, \quad \bar{\kappa} = \int_0^{\infty} \kappa_v dv. \quad (17)$$

We have placed the plane source at the origin. The solution is obtained by taking the inverse Fourier transform of (15):

$$\Phi(X) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \Phi(p) e^{-ipX} dp = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{F(p)}{1-K(p)} e^{-ipX} dp. \tag{18}$$

The behavior of the function $\Phi(X)$ at large distances from the origin is determined by the region of small p . It is therefore necessary to investigate the behavior of the Fourier transforms of the kernel and of the source function at zero.

Putting $p = 0$ in (17) we get $K(0) = \lambda$. Let us consider the expression

$$[pK(p)]' - \lambda = -\frac{\lambda p^2}{\kappa} \int_0^{\infty} \frac{\kappa_\nu d\nu}{k_\nu^2 + p^2}. \tag{19}$$

For simplicity we consider the frequently encountered case when $\kappa_\nu = \beta k_\nu$, and go over from integration with respect to the frequencies to integration with respect to k_ν , assuming the latter to vary from zero to unity. The latter can always be attained by measuring the distances in terms of mean free paths at the center of the line. Then

$$[pK(p)]' - \lambda = \frac{\lambda p^2}{\bar{k}} \int_0^1 \frac{u\nu'(u)}{u^2 + p^2} du \left(\bar{k} = \int_0^{\infty} k_\nu d\nu \right),$$

where $\nu(u)$ is a function which is the inverse of k_ν . In the integral term we make the change of variable $u = tp$:

$$[pK(p)]' - \lambda = -\frac{\lambda p^2 \nu'(p)}{\bar{k}} \int_0^{1/p} \frac{t}{1+t^2} \frac{\nu'(tp)}{\nu'(p)} dt. \tag{20}$$

The integral in (20) tends to a finite limit as $p \rightarrow 0$, so that

$$\lim_{p \rightarrow 0} \frac{\nu'(tp)}{\nu'(p)}$$

decreases at large t . By integrating with respect to p we get for small values of p

$$K(p) = \lambda(1 + \alpha p^2 \nu'(p)), \quad p \ll 1. \tag{21}$$

The value of α can be calculated if one knows the concrete form of the function $\nu(p)$. These results coincide with the earlier ones^[5] when the absorption coefficients have a Doppler or Lorentz form.

We now proceed to investigate $F(p)$. It is easy to verify that $F(0) = \text{const}$. We note further that for all plane sources, except isotropic ones ($f_\nu(x, \mu) = k_\nu \delta(x - x_0)$), the function $F(x)$ decreases more rapidly than $K(x)$. The physical reason is that the number of photons arriving at the given point directly from the source is small in the case of large distances compared with the total number of photons at the given point. It is clear therefore that we obtain the correct asymptotic behavior by putting $F(p) = \text{const}$ when $p \rightarrow 0$.

Thus, the asymptotic values of $\Phi(x)$ characterize only the medium and can be obtained by using

formulas (18), (20), and (21). They turn out to be quite sensitive to the presence of quenching.

It is interesting to note that when $\sigma \ll \bar{k}(1 - \lambda) \ll 1$ there exist two regions of asymptotic values. One is obtained at not too large distances, at which the main contribution to the integral (18) is made by $p \ll 1$, such that $p^2 \nu'(p) \gg 1 - \lambda$. The second region is that of large distances, where a noticeable contribution to (18) is made only by $p \ll 1$ such that $p^2 \nu'(p) \ll 1 - \lambda$. The second region vanishes when $\lambda = 1$ ($\sigma = 0$).

The foregoing is illustrated in the table, which lists the asymptotic solutions of $\Phi(x)$ in the case when the spectral lines have Lorentz or Doppler shapes.

In the table, $\omega = (\nu - \nu_0)/\Gamma$, where ν_0 is the resonance frequency and Γ is the line half-width. We note that in the absence of quenching ($\lambda = 1$) we have $1 - K(p) \rightarrow 0$ as $p \rightarrow 0$, and therefore the form of $F(p)$ ($F(p) \rightarrow \text{const}$ as $p \rightarrow 0$) is of no importance whatever for the determination of the asymptotic values.

As seen from (11), $x - X \approx \text{const}$ when x and $X \gg 1$. This means that the asymptotic values of $n(x)$ coincide in the nonlinear case with the asymptotic solutions of the linear equation. The difference reduces to a shift of the coordinates.

It is easy to investigate the solution of Eq. (8) for small values of X . It is obvious that it depends strongly on the type of source. By way of an example we consider an isotropic source with $\varphi_\nu(\mu) = \kappa_\nu/4\pi$. Then $F(X) = K(X)$. As follows from (9),

$$K(X) \propto |\ln X|^{-1} \text{ for } X \ll 1.$$

A similar behavior holds also for $\Phi(X)$, as can be seen directly from Eq. (8). The divergence of this expression at small X is due to the presence of photons which move practically parallel to the source plane.

Using (11), we can easily express X in terms of x when $X \ll 1$. An approximate solution of (21) is of the form $X \approx x/|\ln x|$. Substituting this into the expression for $\Phi(X)$ we get

$$\Phi(x) \propto \ln \frac{|\ln x|}{x}$$

and ultimately

$$n(x) = \frac{\Phi(x)}{1 + 2\Phi(x)} \approx \frac{1}{2} - \text{const} / \ln \frac{|\ln x|}{x}. \tag{22}$$

Thus, in this case the presence of nonlinearity changes the result very strongly. It is generally evident that nonlinearity can have a strong effect only at sufficiently small distances from the source.

k_v	$K(p) (p \ll 1)$	$\Phi(X)$	Region of applicability
$\frac{1}{1 + \omega^2}$	$\lambda \left(1 - \frac{\sqrt{2 p }}{3}\right)$	$\frac{\lambda F(0)}{6 \sqrt{\pi} (1 - \lambda)^2} X^{-3/2}$	$X \gg \frac{1}{\sigma^2}$
do.	do.	$\frac{3F(0)}{\sqrt{2\pi}} X^{-1/2}$	$\frac{1}{\sigma^2} \gg X \gg 1$
$e^{-\omega^2}$	$\lambda \left(1 - \frac{\sqrt{\pi}}{4} \frac{ p }{\sqrt{ \ln p ^{-1}}}\right)$	$\frac{\lambda F(0)}{4 \sqrt{\pi} (1 - \lambda)^2 X^2 \sqrt{\ln X}}$	$X \gg \frac{1}{\sigma}$
do.	do.	$\frac{3/8 \pi^{-3/2} F(0) [(\ln \sigma^{-1})^{3/2} - (\ln X)^{3/2}] }{1/5 \gg X \gg 1}$	$1/5 \gg X \gg 1$

5. We now proceed to consider the problem of nonlinear transport in a half-space. The corresponding linear problems were considered by a number of authors.^[2, 4, 7] It is simplest to obtain the answer by the Wiener-Hopf method.^[1, 4, 8]

We shall not describe here the method of solving the linear problems, and will use only certain asymptotic properties. We note first that, just as in infinite space, the decrease in the concentration of the excited atoms at large distances from the source does not depend on the type of source. The physical reason is that the photons that reach large distances are essentially those which are multiply reflected. It is also obvious that near the source the concentration will vary in the same manner as near the source in an infinite space.

Since $\Phi(X)$ decreases at large distances from the source, the conclusion that the asymptotic values coincide (apart from a shift of the coordinate) in the linear and nonlinear cases remains in force. In order to show the character of the influence of the nonlinearity of the change in the excited-atom density, we present plots of $n(x)/n(0)$ in the case of normal incidence of light on the medium. For comparison, we present a similar plot for the linear problem (curve a in the figure).

Knowing the concentration of the excited atoms, we can easily find the spectral density of the light flux radiated by a half-plane. It turns out that the spectral and angular distribution of the emitted light is proportional, in the case when the sources are outside the medium, to the source power, and its form does not depend on the intensity of the light incident on the medium. This can be seen most readily from the relation which coincides with the expression for the intensity of the emitted light in the linear case.

$$I_v(0, \mu) = \frac{\kappa_v}{4\pi|\mu|} \int_0^\infty \exp\left[-\frac{k_v}{|\mu|} \int_0^x (1 - 2n(t)) dt\right] n(x) dx$$

$$= \frac{\kappa_v}{4\pi|\mu|} \int_0^\infty \exp\left(-\frac{k_v}{|\mu|} X\right) \Phi(X) dX \quad (\mu < 0),$$

When a plane source is located at the point x_0 the expression for the intensity of the outgoing light coincides with that obtained in the linear case with a source at the point $X_0 = X(x_0)$. Thus, the influence of the stimulated emission reduces to an apparent shift of the source towards the surface.

We now consider the case when the source is located at infinity (the Milne problem). It can be shown that in this case the nonlinear problem reduces to a linear one with a source at infinity. The solution of the latter was obtained in^[4, 5].

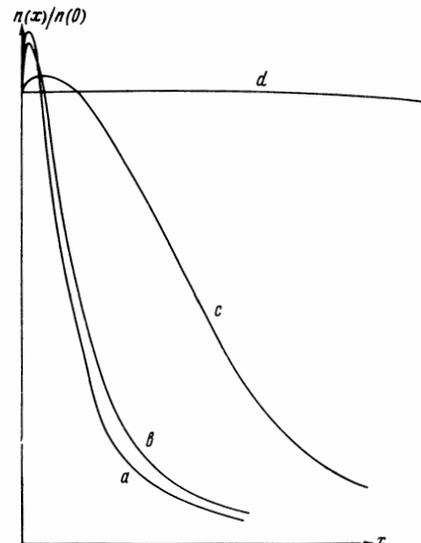
The asymptotic behavior of the solution depends essentially on the line shape and does not depend on the type of the source located at infinity. For example, for a Lorentz line shape ($k_p = 1/(1 + \omega^2)$) we have

$$\Phi(X) \propto X^{1/4} \quad (\lambda = 1).$$

From (11) we obtain $x \propto X^{5/4}$ ($X \gg 1$) and $X \propto x^{4/5}$. Consequently, $\Phi(x) \propto x^{1/5}$ and

$$n(x) \approx \frac{1}{2} - \frac{C}{x^{1/5}}. \tag{23}$$

We can obtain analogously the asymptotic behavior for $\lambda < 1$ and for different absorption-line shapes.



Plots of $n(x)/n(0)$. The intensities of the incident radiation in cases a, b, c, and d are related like 0.1 : 1 : 10 : 100.

The proposed method makes it possible to obtain relatively easily the solution of the nonlinear transport equation with allowance for the degree of excitation and stimulated emission, provided the solution of the corresponding linear problem is known. We note, however, that such a reduction to the linear problem cannot be effected if account is taken of the light absorption outside the line, or an attempt is made to include into consideration the "additional illumination"—excitation of the atom by an extraneous agent. The nonstationary problem is likewise not reduced to a linear one. Thus, the proposed method is not universal and leads to the solution of only several problems in the theory of nonlinear transport. We note that the method is applicable directly to the problem of diffusion of radiation without redistribution over the frequencies. The solutions of the corresponding linear problems for this case have been known for a long time,^[1,2] so that allowance for stimulated emission entails no difficulty whatever.

6. Let us consider, for example, how the Milne problem is modified in this case. As is well known,^[1,2] at large distances from the boundary ($x \gg 1$) the solution of the transport equation coincides asymptotically with the solution of the one-dimensional diffusion equation, so that we have $\Phi(X) \propto X$ for $X \gg 1$. As follows from (11), $X \propto x^{1/2}$. Then the concentration of the excited atoms is determined from the formula (12):

$$n(x) = 1/2 - \text{const}/\sqrt{x}. \quad (24)$$

The described method can be used so long as the distortions of the spectral lines, occurring at

large radiation density, remain insignificant.

After this paper was written, the authors have learned that the described method was recently proposed by Ambartsumyan for a solution of nonlinear transport problem.^[9] He called it the "method of self-consistent optical depth." However, he did not use this method for a direct solution of the transport equation.

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