

RADIATIVE TRANSFER IN SPECTRAL LINES

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Excited atoms are known to play an important and sometimes decisive role in many phenomena which are the subject matter for discussion at this Symposium. The properties, kinetics, and diagnostics of a plasma are to a considerable degree determined by the concentration of excited atoms, and their distribution in space and time. The concentration of excited atoms in optically dense layers is conditioned, together with collisional processes, by radiative transfer and especially by radiation transfer in spectral lines by multiple successive acts of emission and absorption.

It is my intention to present here the main results of the modern radiative transfer theory, its experimental evidence and some applications.

BASIC PRINCIPLES OF THE THEORY OF RADIATION TRANSFER IN SPECTRAL LINES

Radiative transfer in spectral lines should be regarded as multiple successive acts of the emission and absorption of photons. As a result of both the impact and the Doppler effect photon frequency in each re-emission changes within the frequency range corresponding to a spectral line.

This fact is of great importance. We cannot divide a spectral line into several frequency ranges and analyse radiative processes independently in each range at monochromatic approximation. Frequency changes in the re-emission process require that the transfer of all photons corresponding to a given spectral line should be regarded as an entire process.

The law of attenuation of the elementary beam of particles in terms of the path is the main characteristic of any transfer process. In the case of radiative transfer in a spectral line the attenuation of the beam of photons of all frequencies corresponding to a given line should be analysed. Then, instead of the exponential law of attenuation we obtain

$$\frac{N(r)}{N_0} = f(r) = \int \epsilon_\nu e^{-K_\nu r} d\nu \quad (1)$$

where ϵ_ν is the normalized per unit frequency distribution of photons emitted by unit volume and K_ν is the absorption coefficient.

The specific form of the $f(r)$ function depends upon the shape of the absorption and emission lines; however, it is not exponential with any kind of broadening and decreases rather slowly with the distance (e.g., for collision broadening for large r , $f(r) \sim r^{-\frac{1}{2}}$).

The general relation between the frequencies of the absorbed and emitted

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photons is very complex. It may be assumed, to a fairly good approximation that, within the frequency range corresponding to a given line, no correlation exists between the frequencies of absorbed and emitted photons. Then an analysis of radiative transfer in a line can be naturally divided into two steps: the determination of the concentration of excited atoms and estimation of the radiation field. This approach seems even more justified since it is just the concentration of excited atoms that is of paramount interest for many applications.

If the distribution of excited atoms is known, the shape and the resulting intensity of the spectral line emitted by the source can be easily determined by conventional methods.

Assume, for the sake of simplicity, that the concentration of excited atoms $n_2(\bar{r}, t)$ is determined by transitions to, or from, the ground state n_1 . If no correlation exists between the frequencies of the absorbed and emitted photons the concentration of excited atoms is determined by the equation:

$$\frac{\partial n_2(\bar{r}, t)}{\partial t} = -n_2(\bar{r}, t)A_{21} - n_2(\bar{r}, t)b_{21} + n_1a_{12} + \int_V n_2(\bar{r}', t)A_{21}K(|\bar{r} - \bar{r}'|)dV' \dots (2)$$

where

$$K(|\bar{r} - \bar{r}'|) = \frac{1}{4\pi} \int_V \frac{\epsilon_\nu K_\nu e^{-K_\nu |\bar{r} - \bar{r}'|}}{|\bar{r} - \bar{r}'|^2} d\nu \quad (3)$$

A_{21} in equation (2) is the Einstein coefficient of spontaneous emission, and a_{12} and b_{21} are the corresponding number of excitation and quenching acts as a result of collisional processes. a_{12} and b_{21} are normalized per second and per unit volume and they may depend upon both the coordinates and time. If the volume under consideration is exposed to an external radiation, this can be easily taken into account by introducing a term similar to $n_1 a_{12}$ which gives the number of excitation acts normalized per unit volume by an external radiation.

A slow decrease of the $f(r)$ function with the distance manifested itself in a slow decrease of the kernel of equation (2). Due to this eq. (2) cannot be transformed into a differential one, as in case of the diffusion of particles however large the linear dimensions of the volume in which radiation transfer proceeds. The term of the diffusion coefficient is meaningless for radiation transfer in a spectral line.

Physical peculiarities of radiation transfer in spectral lines have been discussed elsewhere^{1, 2}. In one of these papers² a steady-state variant of eq. (2) was obtained ($\partial n_2/\partial t = 0$) and several numerical solutions given. Somewhat later Holstein³ analysed a non-steady state variant of the theory assuming that $a_{12} = b_{21} = 0$ and obtained

$$n_2(\bar{r}, t) = \sum_K n_2^K(r) e^{-\lambda_K t}$$

where $n_2^K(r)$ and λ_K are the eigenfunctions and the eigenvalues of a homogeneous integral equation with the kernel (3) respectively. Since λ_K increases

with increasing K , after a sufficiently large time it takes the form $n_2(r, t) = n_2(r)e^{-\lambda_1 t}$. Holstein calculated λ_1 for a layer enclosed in two parallel planes³ and later for a cylindrical volume⁴. Characteristic time proved to increase with increase in optical density of the layer and to be able to exceed $t = 1/A_{21}$ by several orders of magnitude.

Some achievements of the mathematical theory of radiative transfer in spectral lines have been due to the contributions of Sobolev⁵ and Ivanov⁶.

A new method for the solution of the steady-state variant of the equation (2) was advanced and its strict solution obtained for the case when radiation transfer occurs in a semispace restricted by a plane, while broadening is determined by the Doppler-effect. The solution was so cumbersome, that the distribution of excited atoms could be more easily found by numerical methods. However, investigation of the analytical solution allowed the establishment of several interesting asymptotic regularities and expressions.

APPROXIMATE METHODS

In order to solve eq. (2) well known approximate methods can be applied. Thus, Holstein when analysing the non-steady-state condition at $a_{12} = 0$ found λ_1 , the first eigen-value of the corresponding homogeneous integral equation by the Ritz method. To solve the steady-state variant of eq. (2), the method of successive approximations can be used whose convergence, as a rule, proves to be slow. In addition we can use some other methods of approximate solution of integral equations².

Let us consider in detail the method proposed in an earlier paper⁷. Although it is not sufficiently well established, yet many comparisons with the solutions obtained numerically show the possibility of its utilization. The method is simple and universal.

The kernel of eq. (2) is characterized by its logarithmic divergence at $\vec{r}' \rightarrow \vec{r}$ and its slow decrease with the increase of $|\vec{r} - \vec{r}'|$. This leads to the fact that in the steady-state, approximately

$$n_2(\vec{r}) = \frac{n_1 a_{12}}{b_{21} + A_{21} [1 - \int_V K(|\vec{r} - \vec{r}'|) dV']} \quad (4)$$

$[1 - \int_V K(|\vec{r} - \vec{r}'|) dV']$ is the probability of the photon escape from a given point beyond the volume V without absorption. By designating $A_{21}^* = A_{21} [1 - \int_V K(|\vec{r} - \vec{r}'|) dV']$ i.e. by introducing an effective coefficient of spontaneous emission, we obtain

$$n_2(\vec{r}) = \frac{n_1 a_{12}}{b_{21} + A_{21}^*} \quad (5)$$

To calculate A_{21}^* approximate expressions for $\int_V K(|\vec{r} - \vec{r}'|) dV'$, available in the literature can be used.

Thus, for example, if the volume V is limited by two parallel planes, then for Doppler broadening

$$\int_0^x K(|x - x'|) dx = 0.5 - \frac{1}{4K_0 \times \sqrt{\pi \ln K_0 x}} \quad (6a)$$

and for collisional broadening

$$\int_0^x K(|x - x'|) dx = 0.5 - \frac{1}{3\sqrt{\pi K_0 x}} \quad (6b)$$

where K_0 is an absorption coefficient in the centre of the line. The expressions presented are satisfactorily accurate at $K_0 x > 3$. If necessary, an evaluation of the error in eq. (5) can be made⁸.

It is of importance that eq. (5) yields accurate results in two extreme cases: viz. at large and small optical densities. In the latter case $A_{21}^* \rightarrow A_{21}$, while eq. (5) degenerates into the Stern-Volmer formula.

LIMITS OF APPLICABILITY AND SOME GENERALIZATIONS OF THE THEORY

Applicability limits of a theory are usually determined by the assumptions made during its development.

The basic assumption of the theory of radiative transfer in spectral lines is that of the absence of correlation between the frequencies of the emitted and absorbed photons. This assumption is strictly satisfied when broadening is determined by the impact. If, on the other hand, the shape of the line is essentially the Dopplerian one, the assumption is no longer strict. Nevertheless, qualitative considerations allowed us² and later Holstein³ to accept this assumption.

Recently Hearn⁹ carried out an extensive computing work and compared solutions of a transfer equation for Dopplerian shape of the line, obtained both when taking into consideration frequency correlation and when neglecting it. It turned out that the assumption of the absence of correlation gives an error of less than 10 per cent, therefore, it does not practically restrict the applicability of the theory.

When deducing eq. (2), the diffusion of excited atoms was neglected. This assumption restricts the applicability of the theory. An equation can be put forward taking into consideration both the mechanisms of excitation transfer. This equation is difficult to solve. However, a criterion allowing the determination of the conditions of the prevalence of either mechanism can easily be obtained by various means⁸. The diffusion of atoms can be neglected provided that $A_{21}^* t_a \gg 1$, where t_a is time of the atom diffusion at a distance of the order of linear dimensions of the volume examined. It is indicative that the diffusion coefficient of excited atoms is determined not by a gas-kinetic free path, l , but by an effective path, with

$$\frac{1}{l_{ef}} = \frac{1}{l} + \frac{A_{21} + b_{21}}{v}$$

The term $A_{21} + b_{21}/v$, where v —the mean velocity of the atom—allows for a free path decrease due to emission and quenching. A similar criterion

follows from the work of Veklenko¹⁰. Actual evaluations show that excitation transfer due to the motion of atoms can be neglected if the transfer 2-1 is optically allowed. If, on the contrary, the probability of A_{21} is small, the motion of atoms is decisive. Fowler¹¹ replacing the spectral line by a rectangle, i.e. neglecting large free paths of photons corresponding to the wings of the line, has drawn the conclusion that it is the motion of atoms which is decisive. This viewpoint disagrees with experimental data²⁰.

There are conditions, however, when the motion of excited atoms cannot in any case be neglected, for example, when radiation is transferred in spectral lines in a moving medium. In an earlier work¹² the author considered the radiation transfer of a shock wave in a cool gas flow moving against the shock front. Radiation absorption inducing excitation of atoms or molecules was taken into consideration as well as the subsequent process of the non-steady-state radiative transfer, and the displacement of atoms with the gas flux. The equation written for the plane shock wave was solved:

$$\frac{\partial n_2(x, t)}{\partial t} = v \frac{\partial n_2(x, t)}{\partial x} + A_{21} \int_0^{\infty} n_2(x', t) K(|x - x'|) dx' - \quad (7)$$

$$n_2(x, t)[A_{21} + b_{21}] + B(x)$$

where $B(x)$ accounted for the excitation induced by the wave radiation.

The approximate solution (eq. 7) shows that before the front of a shock wave there moves a wave of excited atoms whose concentration in a cool gas may reach values close to Boltzmannian at the equilibrium temperature established behind the shock front. At the same time photo-ionization of excited atoms proceeds before the shock front. As a result, a noticeable concentration of electrons appears at considerable distances ahead of the shock front. Considerable concentrations of excited atoms and electrons at large distances from the shock front were observed in several experimental studies¹³ carried out in shock tubes. The results of measurements are in agreement with the calculations performed by Lagarkov and Yakubov¹⁴ which supplemented the work of Biberman and Veklenko¹² by the consideration of specific conditions of a shock tube. The appearance of excited atoms at large distances from the wave front is closely related to the peculiarities of radiation transfer in spectral lines. If this process was similar to usual diffusion, excited atoms could be observed only in the immediate neighbourhood of the front, since the cool gas flow would drive them behind the wave front. However, due to a slow decrease of the $f(r)$ function (cf. eq. 1) which determined the non-diffuse character of radiation transfer in spectral lines, excited atoms are found far from the wave front at flux velocities exceeding heat ones by some orders of magnitude.

If the mean distance between the atoms is small compared with the wavelength, then they cannot be regarded as a number of independent emitters. Under these conditions collective effects are expected to be realized which can be exemplified by mirror reflection of the resonance radiation from vapours observed by R. Wood. These effects restrict the applicability of the transfer theory in case of high densities. When taking into consideration the disturbance of coherence due to heat motion, it can be evaluated that coherent phenomena will begin when the concentration of atoms satisfies the inequality:

$$n \gg 10^5 \lambda^{-2} \frac{1}{f} \sqrt{\frac{T}{M}}$$

where f is the oscillator strength; T , the gas temperature; and M , the molecular weight.

The criterion (eq. 8) is in agreement with experimental data on the reflection of the resonance radiation from the boundary of a metal vapour layer.

Equation (2) was obtained when induced emission was neglected. The consideration of this effect leads to a non-linear integro-differential equation which is very difficult to solve.

The present theory can be easily generalized for the case when the spectral line overlaps the continuous background and when the gas volume within which radiation transfer occurs is restricted by partially reflecting planes^{8, 15, 16}.

It is appropriate to notice that the basic principles of radiative transfer in spectral lines have been applied to the theory of radiation transfer in the continuous spectrum (e.g., ionization continuum). In this case the frequency of the photon that induced ionization and that of the photon resulting from recombination do not coincide either. This makes it necessary to consider radiation transfer in all frequencies of the continuum as an entire process¹⁷.

EXPERIMENTAL TEST OF THE THEORY

In an earlier paper¹⁸ a mercury vapour layer was illuminated by resonance radiation of 2537 Å. The ratio of the intensity of radiation that passed through the layer to the incident radiation was measured at various concentrations of absorbing atoms. In another study²⁰ the non-steady-state variant of the theory was tested. The constant characterizing the emission rate of resonance radiation was measured. The study was carried out in Hg¹⁹⁸ vapour which eliminated possible errors connected with ultrafine structure.

Similar work was performed by Phelps and McCoubrey¹⁹ who varied the linear dimensions of the gas volume. This allowed the authors to show that the results obtained by Fowler were in quantitative contradiction with the experimental ones whereas the results reported by Phelps and McCoubrey¹⁹ as well as by other workers^{18, 20} are in good agreement with present theory.

The above mentioned works differ in that values characterizing integral properties of the volume in which radiation transfer occurs were measured. Recently Tkachuk²¹ measured radiation field in different gas volumes. His results are also in agreement with the theory.

APPLICATIONS OF THE THEORY OF RADIATION TRANSFER IN SPECTRAL LINES

1. Deviation from the thermodynamic equilibrium

The escape of radiation from a gas volume may be the cause of a deviation from thermodynamic equilibrium. To make it concrete, assume that

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impact excitation and quenching are realized by collisions with electrons. Even though the concentration and temperature of electrons do not depend upon the coordinates, the concentration of excited atoms due to radiation escape will decrease towards the boundaries of the volume taken by the gas. Thus Boltzmann's law is not fulfilled. The deviation from the equilibrium concentration can be analysed when introducing $y(\bar{r})$ is a reduced concentration (ratio actual to equilibrium concentration). Then eq. (2) for the steady-state variant can be written:

$$y(\bar{r}) = \frac{1}{1 + \beta} \cdot \int_V y(\bar{r}') K(|\bar{r} - \bar{r}'|) dV + \frac{\beta}{1 + \beta} \quad (9)$$

where

$$\beta = \frac{b_{21}}{A_{21}^*}$$

The equilibrium is set up [$y(\bar{r}) = 1$] if the volume V is infinite in any direction or irradiated with black radiation whose temperature equals that of electrons. Otherwise $y(\bar{r}) < 1$. An approximate expression for $y(\bar{r})$ can be obtained using eq. (4). Assuming that there is no external radiation and using the relation of probabilities of the 1st and 2nd kinds we obtain:

$$y(\bar{r}) = \frac{\beta^*(r)}{1 + \beta^*(r)} \quad (10)$$

where

$$\beta^* = \frac{b_{21}}{A_{21}^*}$$

According to eq. (10), $y(\bar{r})$ decreases towards the boundaries of the gas volume, attaining the highest values in its central part. The system proves to be close to equilibrium in the points for which $\beta^* > 1$. That is, in inner regions of the gas volume the equilibrium may be attained even at a low probability of collisional processes.

Ivanov⁶ analysed a case when radiation transfer occurred in semispace, the line was of Doppler's shape, and b_{21} was constant. The problem proved to be analysable when

$$\beta = \frac{b_{21}}{A_{21}^*} \ll 1.$$

It is rigorously shown in this case that $y(\bar{r})$ is determined by the value of β^* . Approximate expressions were obtained for $y(\bar{r})$ that are valid at small and large β^* . A strict solution may be obtained for all β^* . It should be noted that eq. (10) gives values very close to accurate ones over the whole region, excluding the points located in the immediate neighbourhood of the boundaries.

The deviation from equilibrium has been thus far considered in two levels approximation. The situation is more complex for a real atom when both the excitation of all levels and ionization occur since it is necessary to

take into consideration transitions between excited states as well as between discrete states and continuous energy spectrum. This problem in the case of an optically thin layer has been analysed recently^{22, 23}. The transition to an optically dense layer is realized by substituting A_{ik}^* for A_{ik} , a similar procedure being used for the ionization continuum¹⁷. As a result, it is possible to evaluate the degree of deviation from Boltzmann's law and from the formula of Saha for a given kinetic temperature.

Table 1. Values of electron concentration normalized to equilibrium concentration

Pressure (p) atm \diagdown l_{cm}	Thin layer	1 cm	10 cm
10^{-2}	1.2×10^{-3}	0.43	0.66
10^{-1}	3.6×10^{-3}	0.87	0.98
1	1.2×10^{-2}	1.0	1.0

The effect of radiation escape upon the deviation from the equilibrium is illustrated by the data given in Table 1; calculations were made for hydrogen at $T = 8000^\circ\text{K}$ and pressures of $p = 10^{-2}$, 10^{-1} and 1 atm. Table 1 presents values of electron concentration normalized to the equilibrium concentration calculated for an optically thin layer and for inner regions of layers of 1 and 10 cm.

It is quite natural that with the increase of pressure and layer thickness the concentration of atoms should approach equilibrium. Obviously the escape of radiation cannot be the only cause of inequilibrium. However, the above methods of considering the effect of radiation escape can be applied to more complicated problems.

2. Errors associated with the methods of reversal of spectral lines

The possibilities of measuring temperature by spectral line reversal are closely related to the problem of the deviation from equilibrium. The method of reversal is known to give the actual temperature of the gas if Kirchoff's law is obeyed or, if the concentration of excited atoms is Boltzmannian. Otherwise the averaged temperature of excitation of a given level is measured. The degree of approximation to Kirchoff's law is determined by the equation

$$\delta = \frac{\phi}{\phi^0} = \frac{\int_0^L \gamma(x) A_{21}^*(x) dx}{\int_0^L A_{21}^*(x) dx} \quad (11)$$

By evaluating eq. (11), the degree of approach to equilibrium can be checked and measurements can be interpreted by the reversal method. It can be easily shown that the relative error in temperature measurement associated with the deviation from equilibrium equals⁸:

$$\frac{\Delta T}{T} = \frac{KT}{h\nu} \ln \delta$$

for collisional broadening:

$$\delta \approx \frac{\beta \sqrt{K_0} l}{0.75 + \beta \sqrt{K_0} l}$$

3. Photosensitized reactions

Experiments on quenching resonance radiation by admixtures are one of the methods of measuring cross-sections of energy transfer from excited atoms of one gas to the atoms or molecules of another. These experiments measure the dependence of the secondary emission of the gas volume exposed to resonance radiation upon the admixture concentration. The treatment of measurement results and the finding of unknown sections are usually based on the above presented radiation transfer theory.

Closely connected with the problem are some others associated with the theory of photosensitized chemical reactions. The methods described above allow the choice of optimal conditions for such reactions, namely the pressure of the optically excited gas, the dimensions of the vessel, etc.

4. Excited atoms in kinetics

Excited atoms play an important role in physical kinetics. This is particularly due to the fact that cross-sections of atom ionization by electron impact increase sharply with the increase of the principal quantum number²⁴. Due to this in many cases the kinetics of ionization is in fact that of generation of excited atoms. This is especially manifested when analysing the establishment of the degree of equilibrium ionization behind the shock front. Excited atoms, as well as electrons, created before the front as a result of ionization of excited atoms by the radiation of the wave pass, together with the gas current, a density jump and give rise to ionization relaxation. Consideration of this effect together with the generation of excited atoms in the relaxation region as a result of the absorption of radiation from the equilibrium zone made it possible to explain the observed times of ionization relaxation behind the shock front in inert gases²⁵. An important qualitative result should be noted. It can be rigorously shown that when taking no account of radiation processes, the mechanism of ionization must be a binary one, *i.e.* at a given velocity of the wave the product of the pressure before its front and the relaxation time remains constant. If radiative transfer is taken into consideration, the regularity is no longer valid, which completely agrees with the results of observations.

CONCLUSION

The theory of radiation transfer in spectral lines is attracting the attention of physicists and, especially, of astrophysicists whose works were almost unmentioned here. This theory has been checked experimentally and has found many scientific and technical applications. In the very near future the theory will certainly be improved, and approximate methods developed, facilitating its further application.

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