COLLISIONAL REDISTRIBUTION OF RADIATION IN THE NON-IMPACT REGION OF SPECTRAL LINES*

By J. Cooper**, R. J. Ballagh

Joint Institute for Laboratory Astrophysics, University of Colorado, and National Bureau of Standards, Boulder***

and E. W. Smith

Time and Frequency Division, National Bureau of Standards, Boulder

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The theory for collisional redistribution of scattered radiation is outlined starting from its origins in early line shape theories for emission and absorption. Recent developments are discussed including redistribution from the impact region to the far line wings, the effect of m-degeneracy, the influence of a strong driving field, and a consistent treatment of radiative damping.

The study of the interaction of light with atoms subjected to collisions has a long history. Early theories on line broadening introduced the ideas associated with the impact theory (due to Lorentz [1] in 1906) and the quasi-static approximation (due to Holtsmark [2]). The quasi-static theory has been on firm theoretical ground since the pioneering work of Jabłoński [3] and considerable progress has been made on the impact theory in recent years (see Griem [4]). Further, theories which can bridge the gap between the impact region (near line center) and the quasi-static theory (in the line wings) have become available [5, 6]. The transition occurs at a frequency separation $\Delta\omega$ from line center given by $\Delta\omega \sim \tau_c^{-1}$ where τ_c is the duration (or correlation time) of a collision.

The scattering (or redistribution) of radiation by atoms in the presence of collisions has been the subject of a number of recent theoretical studies (see for example Refs. [7–11]). For the case of weak incident radiation, Huber [7] has studied scattering from a two-state atom in both the impact and non-impact region while Omont et al. [8] have

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** John Simon Guggenheim Memorial Fellow, 1977–1978. On leave (1977–78 AY) at Physics Department, University of Otago, Dunedin, New Zealand.

*** Address: Boulder, Colorado 80309, USA.

investigated the ieal atom effects associated with m-degeneracy for scattering in the impact region. At the present time, only the work of Mollow [9, 10] appears to have considered the radiation interaction to all orders, but his work is restricted to a two-state atom in the impact region.

Recently, however, interesting experiments [12] on collisional redistribution have been performed in the region $\Delta\omega > \tau_c^{-1}$ (i. e., outside the impact region) and Cooper [11] has extended the earlier work of Omont et al. [8] to include the situation where absorption or emission can occur in the line wings. In that work [11], the atom is allowed to have three levels — initial, excited and final — each of which may be degenerate; the incident light (assumed monochromatic with frequency ω_1) is absorbed (or scattered) in a real or virtual transition between the initial and excited levels that have a frequency difference of ω_{ei} (see Fig. 1). The "unified" theory [5, 6] is used to derive expressions for the intensity

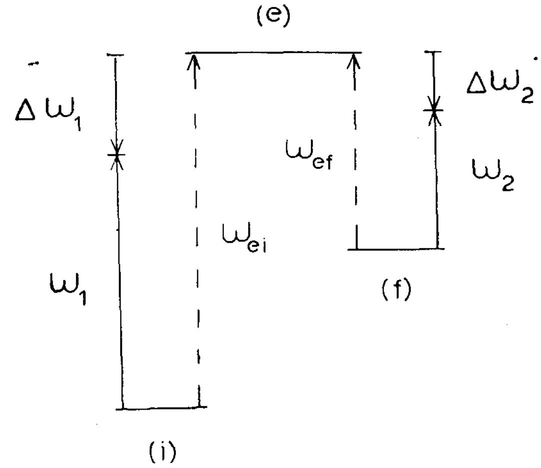

Fig. 1. Frequency separations involved in scattering from a three-level atom

spectrum of the scattered light (frequency ω_2) emitted in a transition from the excited level to the final level (transition frequency ω_{ef}). As in the work of Omont et al. [8], elastic and inelastic (or quenching) collisions are clearly distinguished. The expressions for the scattered intensity are valid provided the time between collisions is long compared to the duration of a collision, and the incident field is weak.

Defining $\Delta\omega_1$ as the detuning of the incident radiation ($\Delta\omega_1 = \omega_1 - \omega_{ei}$) and $\Delta\omega_2$ as the difference between the scattered frequency and the transition frequency ω_{ef} (i. e., $\Delta\omega_2 = \omega_2 - \omega_{ef}$), we find that convenient simplifications occur in the regimes

(a) $|\Delta\omega_1| - |\Delta\omega_2| < \tau_c^{-1}$
(b) $|\Delta\omega_1| < \tau_c^{-1}$, $|\Delta\omega_2|$ unrestricted (i. e., absorption within the impact region)
(c) $|\Delta\omega_2| < \tau_c^{-1}$, $|\Delta\omega_1|$ unrestricted (i. e., emission within the impact region).

These regimes encompass almost all cases of physical interest, and in fact by making the additional assumption of no lower state interaction (a common approximation) we recover forms for the angle averaged scattered intensity similar to those of the impact theory, except that the Lorentzian line shapes (and their attendant constant line width parameters) of the impact theory must be replaced by the more general absorption (or emission) profiles of the unified theory with frequency dependent line width parameters. Raman scattering, which occurs when $\Delta\omega_1 = \Delta\omega_2$ (and is termed Rayleigh scattering when the initial and final levels are the same), is clearly distinguished from collisionally redistributed radiation (fluorescence).

The inclusion of atomic spatial degeneracies allows an explicit calculation of the polarization and angular dependence of the scattered radiation, which may be different than for the non-collisional case. For example, besides redistributing the radiation in frequency, collisions may also cause transitions between the degenerate states in the excited level thus allowing the atom to emit radiation of different angular and polarization character than in the non-collisional case. The theory accounts for such "angular redistribution", in contrast to the case of a two-state atom where this information is not available.

When the incident light is detuned into the far line wings ($|\Delta\omega_1| \gg 0$) the final expressions of Ref. [11] show an increase in fluorescence relative to the Rayleigh component and the cross section for the latter becomes independent of collisions and falls as $(\Delta\omega_1)^{-2}$. The integrated fluorescence intensity in this case becomes directly proportional to the generalized absorption profile.

In a recent development [13], the authors have derived a master equation which describes the density matrix ϱ_A of a degenerate atom which is subjected both to collisions and intense incident radiation (referred to as the laser radiation). The atom is assumed to have two participating levels (both of which may be degenerate) which are coupled by a dipole operator to the radiation field, while the laser radiation may occupy some small group of modes in a given direction. Using the projection operator technique of Zwanzig [15], which treats the interactions to all orders, we first examine the interaction of the atom with the spontaneous (i. e., non-laser) modes of the field and find that it is correlated on a much smaller time scale than those associated with collisions or the laser radiation. In other words, spontaneous emission proceeds unaffected by the collisions or the laser, and the expected radiation damping operator is recovered under the usual approximations.

Next, the collisions are treated by the projection operator technique and by using the fact that collisions occur with a duration which is shorter in time than the radiative decay of the atom, we eventually find the following master equation for ϱ_A ($\hbar = 1$)

$$i\dot{\varrho}_A(t) = \{L_A + L_{LR}(t) - i\gamma\}\varrho_A(t) - i\int_0^t \sigma_p(0)\,\mathrm{Tr}_p\,\{\tilde{L}_c(\tau)\mathcal{U}_c(\tau, 0)\tilde{L}_c(0)\varrho_A(t-\tau)\}d\tau. \tag{1}$$

Here, each operator L_x is a Liouville operator defined for an arbitrary matrix M by

$$L_x M = [H_x, M], \tag{2}$$

where H_x is the part of the total Hamiltonian referring to the isolated atom ($x = A$), the laser-atom interaction ($x = LR$), the laser field Hamiltonian ($x = l$), the total perturbers'

Hamiltonian ($x = \mathrm{p}$) or the collisional interaction between perturbers and atom ($x = \mathrm{c}$). The Liouville operators with time arguments are defined as follows

$$L_{\mathrm{LR}}(t) = e^{itL_{\mathrm{I}}} L_{\mathrm{LR}} e^{-itL_{\mathrm{I}}}, \qquad L_{\mathrm{c}}(t) = e^{it(L_{\mathrm{A}}+L_{\mathrm{p}})} L_{\mathrm{c}} e^{-it(L_{\mathrm{A}}+L_{\mathrm{p}})} \tag{3}$$

and

$$\mathcal{U}_{\mathrm{c}}(\tau, 0) = \text{Time Ordered } \exp\{-\int_0^\tau [\tilde{L}_{\mathrm{LR}}(s)+\tilde{L}_{\mathrm{c}}(s)]ds\}. \tag{4}$$

Finally, γ is the expected operator [14] governing spontaneous decay of a degenerate level, $\sigma_{\mathrm{p}}(0)$ is the initial density matrix of the perturbers, and Tr_{p} indicates a trace over all perturber states.

The master equation (1) for the atom density matrix is a non-Markovian equation including the effect of radiative damping, a strong laser field, and collisional damping. The collisional aspects of the problem are contained in the integral term of equation (1) which is similar to a form obtained and discussed at length in the unified theory [5, 6]. However, in the present case the effect of laser radiation on the collisional processes is also present, via the $\mathcal{U}_{\mathrm{c}}(\tau, 0)$ operator, but if the condition $\langle L_{\mathrm{LR}}\rangle \tau_{\mathrm{c}} \ll 1$ (which corresponds to the Rabi Period being large compared to τ_{c}) is satisfied, this effect is negligible. The spectrum of the scattered radiation is obtained from the dipole autocorrelation function of the atom [16], and it is possible to show that under certain circumstances (corresponding to neglect of initial correlations) the two-time correlation function obeys a master equation *similar* in form to equation (1). It is the integral term, of course, that is responsible for the non-Markovian form of equation (1), and it is necessary to keep this form to have a proper description of "incomplete" collisions (i. e., $t < \tau_{\mathrm{c}}$) which dominate the formation of the line wing [$\Delta\omega > (1/\tau_{\mathrm{c}})$]. The net effect of the non-Markovian behaviour is then to introduce frequency dependent relaxation parameters.

If one is only interested in the line center (i. e., the impact region), which is dominated by large times $t \gg \tau_{\mathrm{c}}$, one may make a Markov approximation and factor $\varrho_{\mathrm{A}}(t-\tau)$ out of the integral as $\varrho_{\mathrm{A}}(t)$ and extend the upper limit of integration to infinity. Under this approximation we regain the familiar impact theory, and the master equation for $\varrho_{\mathrm{A}}(t)$ is the same as that for the correlation function (as required by the quantum fluctuation--regression theorem [16]). Outside the impact region, although the master equations for $\varrho_{\mathrm{A}}(t)$ and the correlation function are similar, the frequency dependence of their relaxation parameters is not necessarily the same (since there is a different frequency dependence for emission and absorption). For a weak laser field the results of [11] can be obtained from this master equation approach (under conditions where it is valid to neglect initial correlations, so that absorption and emission processes are uncorrelated).

In summary one may say that many of the currently interesting problems in light scattering involve frequency redistribution from the line center to the far wings. The pioneering work of Jabłoński on the description of line wing radiation has made it possible to approach this complicated problem with a high degree of physical insight and understanding.

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