Kinetics of saturation of the Doppler spectrum

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Measurement of the kinetics of saturation of a Doppler-broadened resonant transition by monochromatic light provides an experimental answer to the question of how strongly the collisions change the velocity and what is the effective collision cross section.

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1. The nature of the elastic collisions that change the velocity have practially no effect on the luminescence or nonlinear-absorption spectra.[1] Attempts were therefore made in recent years to investigate such collisions by methods of nonlinear spectroscopy.[2–5]

We show in our paper that the kinetics of saturation of an inhomogeneously Doppler-broadened two-level system by means of a monochromatic field contains information concerning the type and frequency of the collisions that change the velocity. As a measure of the population saturation we can use the integral intensity of the luminescence (or) of the absorption (beam) of a weak field on a transition that is adjacent to the one acted upon by the saturating field.

Because of its simplicity, the saturation method, which in contrast to the method described in[2–5] does not call for measurement of the spectral characteristics, is extensively used to study migration in magnetic-resonance spectra[6] and in solid-state laser materials.[7] Its use, however, is hindered by the difficulties of the theoretical interpretation of the experiment. Thus, the influence of the spectral migration on the kinetics of the saturation could be correctly taken into account so far only within the framework of the model of an uncorrelated Markov process,[6–8] which corresponds in terms of velocity changing collisions to the strong-collision model.[1] However, as indicated in[2], the latter model, generally speaking, describes real collisions poorly. We present below the results of a calculation of the saturation kinetics in the case of a Gauss-Markov frequency migration (weak collision model).[1]

2. Proceeding as in[9] before, we introduce the function $m(t)$ defined by

$$n(t) = n(0)\, m(t)\exp\left(-\frac{t}{T}\right) + \frac{n_0}{T}\int_0^t m(t')\exp\left(-\frac{t'}{T}\right)dt', \tag{1}$$

where T is the longitudinal-relaxation time, $n(t) = \rho_{11}(t) - \rho_{22}(t)$ is the running population difference between levels 1 and 2 of the saturated transition ($t = 0$ is the instant when the field is turned on). If level 1 is the ground level, then n_0 is the equilibrium population difference. If, however, both levels are excited, then n_0/T has the meaning of the rate of pumping to the level 1.

A contribution to the time variation of m is made only by the optically-induced relaxation modulated by the migration of the frequency over the Doppler contour. When the conditions $\Gamma \gg (k^2 d\nu)^{1/3}$, $2V$, T^{-1}, t^{-1} are satisfied, this problem is described by a balance equation that takes the following form[9] in the weak-collision model[1]:

$$\frac{\partial m(v,t)}{\partial t} = \left[-2w(v) + \nu\left(1 + v\frac{\partial}{\partial v} + d\frac{\partial^2}{\partial v^2}\right)\right] m(v,t) \tag{2}$$

with initial condition

$$m(v,0) = \phi(v) = \frac{1}{\sqrt{2\pi d}}\exp\left(-\frac{v^2}{2d}\right).$$

Here Γ is the homogeneous width of the 1—2 transition, ν is the frequency of the velocity-changing collisions, $\langle 1|\hat{F}|2\rangle = DEe^{i\omega t}$ is matrix element of the interaction of the atom with the saturating field, D is the corresponding element of the dipole moment, E is the complex amplitude of the field, $V = |DE|/\hbar$,

$$w(v) = \frac{2V^2\Gamma}{(kv)^2 + \Gamma^2} \tag{3}$$

is the probability of the 1—2 transition, $k = \omega_{21}/c$, ω_{21} is the frequency of this transition, c is the speed of light, and $\phi(v)$ is a Maxwellian distribution with respect to the velocities v with a variance d. It is assumed that the collisions that change the velocity and cause loss of phase coherence are statistically independent, and also that $\omega = \omega_{21}$ and $\Gamma \ll k\sqrt{d}$. The function $m(t)$ is determined by starting from a solution of Eqs. (2), in accordance with the equation

$$m(t) = \int_{-\infty}^{\infty} m(v,t)\,dv.$$

3. It can be shown that under the condition $2\bar{w} \gg \nu$, where

$$\bar{w} = \int_{-\infty}^{\infty} w(v)\,\phi(v)\,dv \approx \sqrt{\frac{2\pi}{d}}\,\frac{V^2}{k},$$

the saturating field manages to saturate only the central component of the spectrum after a certain time interval t_Γ following its application, so that $m(0,t_\Gamma) \approx 0$, even though we still have $m(t_\Gamma) \approx m(0) = 1$. This should make it possible to neglect, at $t > t_\Gamma$, the term Γ^2 in the demonimator of (3). Solving the resultant simplified equation by separating the variables (the eigenfunction and eigenvalues of this equation have been obtained in[10]), we obtain

$$m(t) = \frac{[\Gamma(s+1)]^2}{\sqrt{\pi}\,\Gamma\left(2s+\frac{3}{2}\right)} \exp[-(2s+1)\nu t] F\left(s+\frac{1}{2},\; s+\frac{1}{2};\; 2s+\frac{3}{2};\; e^{-2\nu t}\right), \qquad (4)$$

where

$$= \frac{1}{4}\left\{\left[\left(\frac{4V}{k}\right)^2 \frac{\Gamma}{\nu d} + 1\right]^{1/2} - 1\right\},$$

$\Gamma(z)$ is the Euler gamma function and $F(a,b;\,c;\,z)$ is a hypergeometric function.

Under the condition

$$2w(\sqrt{d}) \gg \nu \quad \left(\text{i.e., } s \approx \frac{V}{k}\sqrt{\frac{\Gamma}{\nu d}} \sim \sqrt{\frac{2\bar{w}\Gamma}{k\sqrt{d}\,\nu}} \gg 1\right)$$

almost the entire kinetics of $m(t)$ is described by the formula

$$m(t) \approx \exp\left(-V\sqrt{\frac{2\pi\Gamma\nu}{k^2 d}\,t}\right),$$

which is obtained from (2) at $\nu = 0$. In this case the field strength is so large that it manages to saturate the entire spectrum even before the collisions of the atoms begin to influence the process.

At intermediate powers, $\nu \ll 2\bar{w} \ll k\sqrt{d}\nu/\Gamma$, we can set in (4) $s = 0$, whence

$$m(t) = \frac{2}{\pi}\arcsin e^{-\nu t} \approx \begin{cases} 1 - \dfrac{2\sqrt{2\nu t}}{\pi}, & \nu t \ll 1 \\[2ex] \dfrac{2}{\pi}\, e^{-\nu t}, & e^{2\nu t} \gg 1 \end{cases} . \qquad (5)$$

The presence of an explicit dependence of this expression on ν, and its square-root dependence on the time during the initial stage, are due to the fact that in this case the saturation of the atoms is controlled by their diffusion [with a coefficient equal to νd, cf. (2)] in velocity space in the vicinity of the point $v = 0$, where the interaction with the field is strongest.

Finally, at $2\bar{w} \ll \nu$ the spectral migration becomes so rapid that the rates of saturation of all the atoms become equal to the average transition probability w, as a result of which $m(t) = \exp(-2\bar{w}t)$.

Of particular interest is the kinetics of the saturation in the intermediate range of powers, it differs radically from the analogous results[11] obtained in the strong-collision model: $m(t) \approx \exp(-V(2\pi\Gamma\nu/k^2 d)^{1/2} t)$. In contrast to this result, no field dependences is seen in (5), and the relaxation is determined entirely by the frequency of the velocity-changing collisions. These attributes make it possible to establish experimentally whether the velocity-changing collisions are weak or strong, and to determine their frequency.

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