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PERSISTENCE OF VELOCITY FOLLOWING ELASTIC COLLISIONS

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Résumé. — Une expérience d'absorption saturée à deux lasers conduite sur la raie de KrI à $\lambda = 557$ nm a permis l'étude des collisions élastiques Kr*-He. Pour une classe donnée d'atomes pompés hors résonance ($v_z \neq 0$) on peut mettre en évidence la persistance de la vitesse initiale après n collisions élastiques.

Abstract. — Elastic Kr*-He collisions have been studied using a two laser saturated-absorption experiment on the $\lambda = 557$ nm line of KrI. With the pump laser detuned from the atomic resonance, one can follow the collisional relaxation of Kr* atoms from a non-thermal distribution centred at $v_z \neq 0$ back towards a thermal distribution.

In saturated-absorption experiments, a probe beam monitors the longitudinal velocity distribution of active atoms that are selectively excited by a pump beam. Velocity-changing collisions (V.C.C.) alter this longitudinal velocity distribution and produce a corresponding modification of saturated-absorption profiles. *Strong* or *thermalizing* collisions give rise to a broad Gaussian background superimposed on the narrow resonance, i.e., following a collision, on average, the active atom's velocity is independent of its initial velocity, v_z , and is determined by the thermal distribution [1-4]. *Weak* collisions give rise to a distortion of the shape of the narrow resonance; they are collisions with large impact parameters producing velocity changes smaller than the width of the hole burned in the velocity distribution [5-8]. Elastic hard sphere collisions between light perturbers and heavy active atoms lead to a non-Gaussian background signal owing to the velocity redistribution that they induce [9, 10]. The degree of thermalization following an average collision is sometimes referred to as the *persistence of velocity*.

In a previous publication [9], we studied the effect of V.C.C. on saturated-absorption profiles

($\lambda = 557$ nm of KrI) as a function of the active atom to perturber mass ratio. In that work the same laser was used to saturate and to probe the transition. During the scanning of the laser frequency, the saturator selects successive classes of velocity v_z ; V.C.C. drift and broaden the corresponding distributions of active atoms but their effect is necessarily detected by the probe at $-v_z$ only, hence symmetrical profiles centred around $v_z = 0$. To directly view the drift of active atoms, it is necessary to select them at a given $v_z \neq 0$ and to tune the probe over the velocity distribution as modified by V.C.C.

To this end, we have performed a two laser saturated-absorption experiment on the same transition of Kr($4p^5 5s[3/2]_2-4p^5 5p[1/2]_1$) with the V.C.C. provided by the perturbers. The first dye laser, at frequency ν_1 , is used to select by saturation a given class of longitudinal velocities far from zero; to accomplish this, it is detuned from the atomic resonance at ν_0 by an amount comparable to the Doppler width ($\Delta\nu_D = 720$ MHz). The second-dye laser is used to probe both saturated-absorption (counter propagating waves) and linear absorption. Fabry-Perot fringes are used to calibrate the frequency scale of the probe. A typical recording is shown on figure 1. Trace I is the linear absorption profile, centred at ν_0 . Trace II is a saturated-absorption profile obtained in pure krypton, for a detuning $\Delta = \nu_1 - \nu_0 = 600$ MHz

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FIG. 1. — Trace I : Linear-absorption profile, centred at ν_0 . Trace II : Saturated-absorption profile, obtained in pure krypton for a detuning $\Delta = 600$ MHz of the saturator. Trace III : Saturated-absorption profile, obtained in a mixture Kr-He ($P(\text{Kr}) \approx 10$ mtorr, $P(\text{He}) \sim 450$ mtorr) for a detuning $\Delta = 600$ MHz. The squares represent a profile calculated from expression (2) with the values of the parameters indicated in the text. Trace (a) is the Lorentzian centred at $-\Delta$; Trace (b) is the contribution due to thermalizing Kr^*-Kr collisions, centred at $\Delta' = 0$; Trace (c) is the contribution due to Kr^*-He V.C.C.

of the saturator; it shows a narrow resonance centred at $\Delta' = -\Delta$ and a Gaussian background centred at $\Delta' = 0$, and attributed to Kr^*-Kr collisions. Trace III shows a saturated-absorption profile obtained in a mixture Kr-He, for the same detuning. The difference in shape between trace II and III is attributed to Kr^*-He V.C.C. whose main contribution lies between $-\Delta$ and 0.

INTERPRETATION OF PROFILES. — To calculate the resulting profile in the presence of elastic collisions, one considers their effect on the phase of active dipoles (introduced *via* classical broadening constants) and their effect on the velocity associated with the atomic state populations (introduced *via* a *collision propagator* [11]). The propagator G which propagates a population density $n(v_z, 0)$ ($v_z =$ longitudinal velocity) to the value $n(v'_z, t)$ *via*

$$n(v'_z, t) = \int G(v_z \rightarrow v'_z, t) n(v_z, 0) dv_z$$

satisfies the equation :

$$\begin{aligned} \frac{\partial G}{\partial t}(v_z \rightarrow v'_z, t) = & -\Gamma G(v_z \rightarrow v'_z, t) + \\ & + \Gamma_1 \int W_1(v''_z \rightarrow v'_z) G(v_z \rightarrow v''_z, t) dv''_z \\ & + \Gamma_2 \int W_2(v''_z \rightarrow v'_z) G(v_z \rightarrow v''_z, t) dv''_z \quad (1) \end{aligned}$$

with

$$G(v_z \rightarrow v'_z, 0) = \delta(v_z - v'_z).$$

Both thermalizing and non thermalizing collisions are assumed to occur with rates Γ_1 and Γ_2 respecti-

vely. The quantity Γ is the total rate of population density loss, $\Gamma = \Gamma_0 + \Gamma_1 + \Gamma_2$ where Γ_0 is the inverse lifetime of the state population under consideration, which is here the time during which the atoms stay in the fields [9]. $W_1(v'_z \rightarrow v_z)$ is the probability density for a thermalizing event to occur per unit time;

$$W_1(v''_z \rightarrow v'_z) = W_1(v'_z) = \frac{1}{\sqrt{\pi}u} \exp - (v'_z/u)^2$$

where u is the most probable speed of the thermal distribution. $W_2(v''_z \rightarrow v'_z)$ is the probability density for non-thermalizing events; we choose the model of Keilson and Storer [12] to express :

$$W_2(v''_z \rightarrow v'_z) = \frac{1}{\sqrt{\pi}\Delta u} \exp - [(v'_z - \alpha v''_z)/(\Delta u)]^2$$

where α is the mean *strength* of V.C.C. related to the r.m.s. change of velocity Δu by $(\Delta u)^2 = (1 - \alpha^2) u^2$. In this model Γ_2 can be fairly-well approximated by the rate of collisions in a hard sphere model and for a small ratio of perturber to active atom mass $m/M \ll 1$, $\alpha \approx 1 - \frac{4}{3} \frac{m}{M}$ [11].

Equation (1) can be solved by iteration and one obtains [10] :

$$\begin{aligned} \tilde{G}(v_z \rightarrow v'_z) = & \frac{\delta(v_z - v'_z)}{\Gamma} + \frac{\Gamma_1 W_1(v'_z)}{\Gamma_0(\Gamma_0 + \Gamma_1)} + \\ & + \frac{1}{\Gamma} \sum_n \left(\frac{\Gamma_2}{\Gamma} \right)^n \frac{1}{\sqrt{\pi} \Delta u_n} \exp - \left[\frac{v'_z - \alpha^n v_z}{\Delta u_n} \right]^2 \end{aligned}$$

where

$$(\Delta u_n)^2 = (1 - \alpha^{2n}) u^2$$

and

$$\tilde{G}(v_z \rightarrow v'_z) = \int_0^\infty G(v_z \rightarrow v'_z, t) dt.$$

Saturated-absorption profiles are given, in a two laser experiment (counter propagating beams) by the following expression (at very low saturation) [10]

$$\begin{aligned} I = I_0 \int dv_z \int dv'_z \frac{\tilde{\gamma}}{\tilde{\gamma}^2 + (\Delta - kv_z)^2} \tilde{G}(v_z \rightarrow v'_z) \times \\ \times \frac{\tilde{\gamma}}{\tilde{\gamma}^2 + (\Delta' + kv'_z)^2} W_1(v_z) \end{aligned}$$

where $\tilde{\gamma}$ is the homogeneous width (H.W.H.M.), Δ the detuning of the saturator and Δ' the detuning of the probe.

$\tilde{G}(v_z \rightarrow v'_z)$ is the sum of three terms, hence the total profile is also a sum of three terms; it can be expressed in the Doppler limit ($\tilde{\gamma} \ll ku$) as :

$$I = I_a + I_b + I_c = I_0 \frac{\sqrt{\pi}}{\Gamma} \frac{e^{-\Delta^2/k^2 u^2}}{k u \tilde{\gamma}} \left\{ \frac{2 \tilde{\gamma}^2}{4 \tilde{\gamma}^2 + (\Delta + \Delta')^2} + \frac{\sqrt{\pi}}{k u} \tilde{\gamma} \frac{\Gamma_1 \Gamma}{\Gamma_0(\Gamma_0 + \Gamma_1)} e^{-(\Delta'/k u)^2} + \right. \\ \left. + \frac{1}{\sqrt{\pi}} \sum_n \left(\frac{\Gamma_2}{\Gamma} \right)^n \frac{\tilde{\gamma}}{k \Delta u_n} \int_{-\infty}^{+\infty} \frac{2 \tilde{\gamma}_n}{4 \tilde{\gamma}_n^2 + (\Delta' + \alpha^n \Delta + x)^2} \exp - \left(\frac{x}{k \Delta u_n} \right)^2 dx \right\} \quad (2)$$

where $2 \tilde{\gamma}_n = \tilde{\gamma}(1 + \alpha^n)$.

The first term in (2) is a Lorentzian corresponding to atoms which have not undergone velocity changes; it is centred at $-\Delta$ with total width (F.W.H.M.) of $4 \tilde{\gamma}$. The second term is a Gaussian corresponding to atoms which have undergone at least one thermalizing collision; it is centred at $\Delta' = 0$ and has the Doppler width. The third term is a sum of Voigt integrals and corresponds to atoms which have undergone velocity changes but no thermalizing event; it is asymmetrical and expresses the progressive loss of memory of the initial velocity following n collisions.

The profile shown on figure 1 has been compared to a theoretical one (expression (2)) by use of a least square fit. α has been chosen equal to 0.94 corresponding to the predicted value [11]; $\tilde{\gamma}$ has been evaluated at 18 MHz according to the experimental situation. Thus, profiles depend on two parameters only, Γ_2/Γ and $\Gamma_1 \Gamma/\Gamma_0(\Gamma_0 + \Gamma_1)$ which determine the intensities of the two components due to V.C.C. compared to the intensity of the Lorentzian. Here we have chosen to fix Γ_2/Γ and we have looked for a value of $\Gamma_1 \Gamma/\Gamma_0(\Gamma_0 + \Gamma_1)$ which gives account of the shape of the profile; the same procedure has been used for several different values of Γ_2/Γ , n being limited to sixty. The best fit, determined by the minimum value of the mean square deviation, has been obtained for a well-defined couple of parameters; in particular, the shape of the profile is very sensitive to the value of Γ_2/Γ .

An example of the fit is shown on figure 1; on trace III, dots represent the calculated profile, sum of the three terms of equation (2), each of them being represented by a *thin* curve (trace a, b, c). The asymmetry of the component due to Kr*-He V.C.C. shows the non-thermalizing nature of these collisions; this is better shown on figure 2, where different Voigt profiles related to different values of n are represented.

A profile associated to n represents the contribution of atoms which have undergone a mean velocity change Δu_n after n collisions.

FIG. 2. — Contribution of different Voigt profiles with different n values to the component associated to Kr*-He V.C.C. (Trace (c) of figure 1). Positions of these different components are marked by dashes. Note that the profile for $n = 1$ is already shifted of an amount $k \Delta u$.

The fit yields the following values for the parameters : $\Gamma_1/\Gamma_2 = 0.04$ and $\Gamma_0/\Gamma_2 = 1.6$ in close agreement with those obtained in [9] on Kr saturated-absorption with one laser. However we can notice that the fit of the asymmetrical profile is much more sensitive to certain parameters; in particular, the spread of α values around 0.94 indicates that, actually, α is defined in a narrow range : $\alpha = 0.94 \pm 0.01$. Thus, our results indicate that a simple model of hard sphere collisions can consistently explain the effects of V.C.C. on saturated-absorption profiles.

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