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Study of velocity-changing collisions by time resolved saturated absorption

M. Gorlicki, A. Peuriot (*) and M. Dumont

Laboratoire de Physique des Lasers, Université Paris-Nord, 93430 Villetaneuse, France

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Résumé. — L'usage de deux lasers à colorant continus, stabilisés, et de modulateurs électrooptiques permet de combiner les avantages de la spectroscopie à haute résolution et de la spectroscopie résolue en temps. Ce montage est utilisé pour l'étude de la thermalisation des vitesses d'atomes métastables de néon ($1s_3$). Le noyau de collision apparaît formé de deux composantes, l'une large et asymétrique, l'autre étroite et symétrique.

Abstract. — By use of two stabilized CW dye lasers and of electrooptic choppers we have developed an experimental set-up which combines the advantages of high frequency resolution and time resolved spectroscopy. We use this set-up to study the velocity thermalization of $1s_3$ metastable neon atoms. The collision kernel appears to be constituted of two components. The first one is broad and asymmetric, the second one is narrow and symmetric.

In gases, the knowledge of the collision kernel $A(\mathbf{v}, \mathbf{v}')$ (probability for a collision to change the atomic velocity from $\mathbf{v}$ to $\mathbf{v}'$) is of great importance. On one hand, this kernel is related to the interatomic potentials through the elastic differential cross sections. On the other hand, it has an important role in Doppler-free spectroscopy due to its contribution to the saturated absorption lineshape [1-9].

The usual saturated absorption technique, with a single CW dye laser, clearly demonstrates the existence of velocity-changing collisions but does not give enough information to determine the shape of the kernel precisely : first, the saturating beam and the counterpropagating probe beam interact with atoms of opposite velocity and it is not possible to vary v and v' independently. Secondly the observed lineshape is the result of an undetermined number of collisions occurring during the atomic lifetime. In principle, by reducing the pressure, it is possible to have no more than one collision per atom but this condition is quite difficult to fulfill with long living atoms like metastable atoms.

The first difficulty can be removed by the use of two lasers [10]. One of them is tuned to saturate atoms of velocity v , and the frequency of the second laser is scanned to probe all velocities v' . To avoid the second difficulty, Hänsch *et al.* [11] and Cahuzac *et al.* [12] used a pulsed dye laser, the probe pulse being delayed

with respect to the saturating pulse. In these experiments, the laser frequency and the pulse shape were not precisely adjustable.

To study velocity changing collisions and other transient phenomena in saturated absorption, we have designed an experimental set-up constituted of two CW dye lasers externally chopped by electrooptic devices. Each laser is frequency stabilized by means of a Fabry-Pérot interferometer (stability $\simeq 1$ MHz) which is locked to the chosen line of neon by means of an auxiliary saturated absorption Ne cell. Frequency scanning is obtained by applying a magnetic field on the neon cell (light is σ^+ polarized).

The modulating devices are AD* P crystals driven by square pulses of adjustable width and delayed with respect to the same clock (Fig. 1). The detected probe pulses are integrated in a box car, the gate of which is just broader than the pulse width. To remove the linear absorption, one saturating pulse over two is inhibited, while the sign of the probe signal is reversed for one pulse over two : the integrated signal is sensitive only to the saturated absorption.

The experiments reported in this paper have been made on the 616.3 nm line of neon ($1s_3-2p_2$). The cell was filled either with pure neon, or with a He-Ne mixture, at room temperature (pressure range : 5 to 60 mtorr). A RF discharge (155 MHz) produces a large population in the $1s_3$ metastable level ($J = 0$). Since the saturating pulses have a very low intensity (a few mW/cm²) and are much longer (2 to 300 μ s) than the $2p_2$ level lifetime (17 ns), the excited state

(*) Permanent address : CITEFA, Zufriategui and Varela, 1603 V. Martelli, Argentina.

Fig. 1. — Diagram of the pulse generation set-up. The bistable circuit (1/2 on the diagram) switches off the transmission of the saturating pulse and reverses the detection of the probe pulse, once every two clock pulses. The rise time of the saturating pulse is 5 ns and that of the probe pulse 50 ns.

population can be neglected. The probe pulse ($0.4 \mu\text{s}$) is only sensitive to the modifications of the population of the metastable level : the related experiment would preferably be called velocity selective optical pumping [13]. The beam diameters are large (saturating beam : 18 mm — probe : 7 mm) so that the diffusion of atoms out of the interaction region is not sensitive before a few tenth of microseconds.

In a first experiment, the saturated absorption signal has been recorded as a function of the probe frequency, ν_p , for several delay times after the beginning of the saturating pulse ($T_{\text{on}} < \text{saturating pulse duration}$). Figure 2 shows the curves for He-Ne mixture (46/5.5 mtorr) with the saturating frequency, ν_s , detuned by 800 MHz ($\approx$ Doppler half width) from the atomic frequency ν_0 . The signal consists in a narrow peak for $\nu_p - \nu_0 = \nu_0 - \nu_s$ and a broad background corresponding to atoms whose velocities have been changed.

The narrow peak rises rapidly during the first microseconds and reaches an equilibrium when the pumping rate is balanced by relaxation processes, essentially by velocity-changing collisions. The background increases slowly and its shape strongly varies

with the delay. After $5 \mu\text{s}$, each atom has experienced a few collisions only : the background is restricted to the neighbourhood of the initial peak, mainly on the low velocity side. This background gives a qualitative picture of the collision kernel. As the delay increases, the number of collisions per atom increases : the background tends to be centred at $\nu = 0$ and its shape tends towards the gaussian thermal equilibrium. Nevertheless an asymmetry remains even after a delay as long as $240 \mu\text{s}$. By furthermore increasing the delay we would obtain the CW limit like in reference [10].

This experiment suggests a collision kernel broader than the saturation peak, but much narrower than the thermal distribution. With pure neon the behaviour is essentially the same but the thermalization is faster and more complete : the collision kernel is broader for Ne^*-Ne collisions than for Ne^*-He collisions, in agreement with the mass ratio argument. Furthermore, in Ne^*-Ne collisions, exchange of metastability may occur and produces a faster thermalization of velocities. Nevertheless the Ne^*-Ne collisions are not *strong collisions* (thermalization after one collision) like it has often been assumed.

A second experiment has been made to study with more details the evolution of the narrow peak during and after the saturating pulse. This pulse was chosen short enough to keep the background as small as possible. The first feature is that the peak remains remarkably lorentzian and keeps the same centre within the accuracy of the experiment (2 MHz). Figure 3 shows a set of curves recorded with 25 mtorr of pure neon, and figure 4 shows the time variations of the height and the width of the lorentzian curves. The small flat background is disregarded (¹).

(¹) The study of the background build up after a short saturating pulse would be the most meaningful experiment for the determination of the broad kernel, but when the few pumped atoms are spread over a large velocity range the signal is very small. We hope to increase enough the sensitivity of the experiment to carry out this study.

Fig. 2. — Build up of the saturated absorption signal in He-Ne mixture (46/5.5 mtorr), for successive delay times after the beginning of the saturating pulse ($\nu_0 - \nu_s = 800 \text{ MHz}$). All delay times are shorter than the saturating pulse duration ($300 \mu\text{s}$) : All curves are recorded during the saturating pulse with the same sensitivity of the detection. The small peak at $+800 \text{ MHz}$ is due to the reflexion of the saturating beam on the window of the cell. The zero signal is obtained without saturating beam (darker line), it is slightly distorted by the linear absorption signal which is not completely eliminated due to small imperfections in the electronical set-up.

Fig. 3. — Build up and relaxation of the saturated absorption peak. Pure neon : 25 mtorr. Saturating pulse width : 4 μ s.

Fig. 4. — Time dependence of the width and of the height of the saturated absorption peak in the conditions of figure 3. The theoretical lower limit width is 19 MHz (determined by the $2p_2$ level radiative lifetime).

The small increase of the width during the saturating pulse may be due either to velocity-changing collisions or to power-broadening by the pumping beam. After the end of the saturating pulse, the fast decrease of the peak height is mainly due to velocity-changing collisions, which produce the broad background (as observed in the previous experiment), but the broadening of the peak can be produced only by very weak velocity-changing collisions. This process corre-

sponds to a kernel which is narrower than the saturated absorption peak and symmetric in $(v - v')$. In He-Ne mixtures we found the same behaviour except for a small dissymmetry of the peak, which seems to appear after a long time, but which is smaller than the present accuracy of our measurements.

In conclusion these preliminary experiments suggest that the collision kernel is made of two very different parts. The first one is a broad asymmetrical kernel very efficient to thermalize velocities (broader for Ne*-Ne than for Ne*-He collisions). This kernel is the one observed in the largest number of publications, when the background is studied on a broad bandwidth; it looks very similar to the Keilson and Storer (K. S.) kernel [14] used by Le Gouët [5] and other authors to fit the background in static experiments ⁽²⁾. The second contribution to the kernel is a narrow peak, centred at the initial velocity, which affects only precise measurements of the saturated absorption peak. Many authors have studied this line broadening by weak elastic collisions. Among them, S. Avrillier [8] have used a symmetrical kernel calculated for a R^{-6} potential and Cahuzac *et al.* [6] have fitted a narrow K. S. kernel (which is not symmetric in $v - v'$). The stability of the peak position as the delay increases is not in favour of the narrow K. S. kernel. Nevertheless, in our conditions (detuning $\approx$ Doppler half width $\approx$ 800 MHz), a K. S. kernel with, for instance, $1 - \alpha = 1.25 \times 10^{-3}$ would be a gaussian curve of 40 MHz half width, shifted by only 1 MHz. The resulting broadening would be important, but the shift would be lower than our present sensitivity.

After some experimental improvements, to increase accuracy ⁽³⁾ and sensitivity, we will be able to do a numerical analysis of results for different values of all the parameters (pressure, discharge, laser intensity...). We hope to be able to determine the collision kernels very precisely and to find their physical interpretation.

Our experimental technique appears to be very sensitive for the study of collision kernels. With faster pulses our set-up will also permit the study of transient processes due to the upper level and to optical coherence.

⁽²⁾ It has not yet been possible to do a numerical fit of a K.S. kernel or of a hard sphere Kol'chenko kernel (see Ref. [5]), but assuming that the background after a few microseconds (Fig. 2) is due to only one collision per atom, we find that the width and the shift of this background are consistent with an α Keilson and Storer parameter between 0.95 and 0.98 for He-Ne* collisions.

⁽³⁾ In these preliminary experiments we had no precise measurement of time : the time calibration of our oscilloscope appeared to be out of work. Measured times are overestimated by a factor 1.5 to 2.

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