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LASERS IN CROSSED BEAM EXPERIMENTS

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Résumé

On examine les diverses utilisations possibles du laser dans des expériences de collision en jets croisés. L'exposé est limité aux collisions de faible énergie (quelques dizaines de meV) entre atomes excités et atomes ou molécules. Le premier rôle que peut jouer le laser dans ces expériences est celui de sélecteur, permettant aussi bien la préparation de l'état initial que l'analyse de l'état final, qu'il s'agisse d'un niveau atomique bien défini (radiatif ou métastable), d'une classe de vitesse, d'un sous-niveau Zeeman. Dans ces applications, le laser n'agit pas sur la collision elle-même. Il n'en va plus de même aux puissances élevées et à des fréquences décalées par rapport aux fréquences atomiques. Quelques suggestions sont faites sur les possibilités d'étudier, en jets croisés, ces collisions assistées par laser.

Abstract

Different possible uses of lasers in crossed beam experiments are examined. The scope is limited to low energy collisions (few tens of meV) between excited atoms and atoms or molecules. The first role of the laser in this kind of experiments is that of a selector, allowing either the preparation of the incoming channel or the analysis of the outgoing one, by selecting one definite atomic level (radiative or metastable), one class of velocities, one magnetic sublevel. In all these applications, the laser does not act on the collision process itself. The situation is quite different at high laser powers and frequencies detuned with respect to atomic frequencies. Some suggestions are made about the feasibility of crossed beam studies of these laser-assisted collisions.

The goal in crossed beam experiments is to prepare as well as possible the initial momenta of the collision partners, or equivalently, the relative velocity v_r and the initial direction in the center-of-mass frame ($\Theta_{CM} = 0$). According to the species under investigation, and to the financial support, one may use either mono-kinetic (supersonic) beams, or thermal beams plus velocity selector, or pulsed thermal beams plus a time-of-flight analysis. The collision energy is varied either by heating or cooling, or by selecting variable times of flight, or by changing the relative angle between the two beams. In this type of experiments the laser can be used in two different ways : (i) as a tool to complete and improve the preparation or the analysis of the collision partners, with no appreciable effect on the collision itself : (ii) as a mean to change the nature of the collision process.

I - LASERS AS SELECTORS

Except for ground state species, the internal state of the collision partners before and after the collision is generally not controlled in simple crossed beam experiments. The first possible role of the laser is to act as a selector, or an

analyzer, of states and eventually also of the velocities. These uses of laser light are summarized in fig. 1.

Fig. 1 - Lasers as selectors

The laser can be used to produce and select radiative states—the laser beam is sent directly into the collision volume —, or to depopulate selectively long lived (e.g. metastable) levels before the collision volume. These selection properties can obviously be used as well in the analysis of the final state (cf. for instance the laser induced fluorescence method). Good optical pumping conditions generally require :

- (i) a continuous tunable laser locked on an atomic line,
- (ii) a sufficiently high power, to get a stationary pumping regime, but, in some cases, not too high, to avoid undesirable saturation effects,
- (iii) a convenient choice of the excitation scheme.

I.1 - Population, or depopulation, of a specific state

a) Radiative levels of alkali atoms (or alkaline-earths) are populated directly from ground states. Many experiments of this kind have been performed to study excited atom or ground state atom collisions at thermal energies. We shall come back to this case when considering the M-selection. Other experiments have been devoted to excited alkali-on-molecule collisions, for instance :

- fine structure transition in $K^*(4P)$, or $Rb^*(5P) + H_2$ or D_2 collisions [1]. One originality of this experiment consists in that the rotational temperature is varied by seeding the $H_2(D_2)$ supersonic beam with heavier atoms, such as argon (T_{rot} is estimated from the energy balance between the different degrees of freedom).

- energy transfer (electronic to vibrational) in $\text{Na}^*(3p)$ on triatomic molecule (CO_2 , N_2O) collision [2] ; the final relative energy E'_{CM} is obtained at each scattering angle by means of a velocity selector set in front of the alkali-atom detector, and the doubly-differential cross section $d^2\sigma / (dE'_{\text{CM}} \cdot d\theta_{\text{CM}})$ is measured.
- reactive scattering experiments, using a "simple" (but differential) mass analysis, e.g. for the reactions :

(Here no information is obtained about the internal state of the product), or the laser induced fluorescence method, as in :

here relative total cross sections are obtained for each value of v , in the range 0 - 16. The same method has been used for the reactions : $\text{Cs}^*(^7\text{P}_{3/2,1/2}) + \text{H}_2 \rightarrow \text{CsH} + \text{H}$ [5] ; a first laser is used to populate Cs^* and a second one analyses the internal state of product.

- collision between two excited alkali-atoms [6].

b) upper radiative levels can be obtained starting from a metastable level. This is typically the case for rare gas atoms. For instance, the Hornbeck-Molnar process :

has been recently studied [7]. Excited helium atoms are produced within a metastable (2^3S)He beam by a frequency doubled CW dye laser ; about 10 mW are obtained at $\lambda = 294.5 \text{ nm}$, by using an intra-cavity ADA doubler crystal.

Of course upper levels (e.g. Rydberg states) can be also populated from a radiative level, by means of a second laser beam.

c) A specific metastable state can be depopulated before the collision volume and also eventually before the metastable atom detector. This method has been used mainly for rare gas atoms [8]. For practical reasons (spectral range), the neon atom is here the ideal one. For example, very detailed studies of $\text{Ne}^*(3s, ^3\text{P}_0) - \text{Ne}$ and $\text{Ne}^*(3s, ^3\text{P}_2) - \text{Ne}$ collisions have been performed [9]. It is noteworthy that the laser selector is isotope selective ; this gives a very easy way to separate the exchange process from the direct one, which helps to clarify the experimental test on the potential energy curves, especially in cases where many molecular states are involved (e.g. for the $^3\text{P}_2$ level) [10].

I.2 - Velocity selection

When the atomic beam is well-collimated but has a wide velocity distribution (thermal beam), the Doppler effect allows to pump selectively a narrow band of velocity. This technique is particularly useful when the second beam is monokinetic. For radiative states (alkali atoms) this laser velocity selection provides a purely optical method to measure elastic or inelastic differential cross sections [11] (see fig. 2) : a first collinear laser beam (ω_1) selects one velocity $\vec{v}_A$ within the excited atom beam ; if the initial velocity $\vec{v}_B$ of the second partner is unique, the initial relative velocity $\vec{v}_r$ is then well defined ; a second laser beam (ω_2), inducing a transition from the excited state (e_1) of A to an upper excited state (e_2), and collinear to $\vec{v}_r$, selects a definite value of the projection of $\vec{v}_r$ of the final velocity, i.e. a definite value of the CM scattering angle. An inelastic collision can be studied as well by tuning the second laser on a transition starting from another state (e'_1) of A after the collision. The observation is made via the fluorescence (ω_f) from (e_2).

Fig 2 - Optical measurements of a D.C.S

A velocity selection can be achieved also for a metastable level, by depopulating a narrow velocity band. Fig. 3 shows the time-of-flight distribution of a metastable ($3s, {}^3P_{0,2}$)Ne beam. A laser, tuned on the $1s_5 {}^3P_2 - 2p_6$ transition ($\lambda=614.3\text{nm}$) and then conveniently shifted, burns a hole in the distribution, specifically for 3P_2 level and ${}^{20}\text{Ne}$ isotope [11]. Owing to the long acquisition times (hours) required in differential measurements, the laser frequency needs to be carefully stabilized. In the present case, a CW dye laser is locked on a saturated absorption line shifted by a magnetic field [12]. Up to now, the resolution of this "negative" velocity selector is essentially limited by the angular aperture of the atomic beam ($\Delta v \approx 80$ to 100 m/s).

Fig 3 - Laser velocity selection in a Ne^* metastable beam.

1.3 - Magnetic sublevel selection

Up to now it has been implicitly supposed that no polarization effect occurs. Notice that this is not necessarily a "natural" case, because of the strong polarization of the laser light, and one has sometimes to take care to eliminate polarization effects. On another hand, a magnetic sublevel selection (M - selection) is very useful, at least when it is completely controlled.

In low energy collisions rather complex processes may occur at large inter-nuclear distances ($R \gtrsim 10 a_0$): coupling terms, negligible at small R , such as coupling due to the rotation of the molecular axis $\vec{R}$, spin-orbit coupling, etc..., become more and more efficient at large R . The problem here comes from the fact that these terms may have comparable magnitudes, which makes difficult to construct the "good" molecular basis. The M - selection of initial and final states offers an interesting way to investigate experimentally these intricate long range effects.

To understand how the laser M - selection operates, one has to consider with attention the reference quantization axis. For sake of clarify we shall consider molecular states dissociating at infinite R into $\text{A}^*(^1\text{P}) + \text{B}(^1\text{S})$ (see fig. 4).

When AB is an heteronuclear system, three molecular states have this dissociation limit: π_+ , π_- , Σ , for which the component on $\hat{R}$ of the electronic angular momentum is $+1$, -1 , 0 respectively. Three quantization axes need to be considered, namely: $\hat{z}_i$ for incoming state, $\hat{R}$ for the collision itself and $\hat{z}_f$ for the outgoing state. A pure state prepared along $\hat{z}_i$ (e.g. $M(z_i) = +1$) is generally a coherent superposition of states $|\pm 1\rangle$, $|0\rangle$, along $\hat{R}$. The outgoing combination along $\hat{R}$ is different from the incoming one, i.e it is not globally shifted, because the potentials are different for $\Lambda = 1$ and $\Lambda = 0$ and also because long range static or dynamical (according to the chosen molecular basis) couplings occur. Finally one gets a definite superposition of outgoing amplitudes f_0 , $f_{\pm 1}$, referred to $\hat{z}_f$. A similar conclusion holds when fine structure, and hyperfine structure exist.

Fig 4 - M-selection in a $A(^1P) + B(^1S)$ collision

a) examples for radiative levels

- By choosing different excitation schemes (directions of atom and laser beams, polarization, external magnetic field) it is possible to study a given process (elastic or inelastic) with different superpositions of incoming channels. The relative contributions of different incoming molecular states (and consequently of different types of coupling) to the apparent total cross section for the fine structure transition :

have been studied by this way [13], choosing different excitation schemes, such as : (i) a wide-band laser ($0,1\text{\AA}$) linearly polarized, with a magnetic field perpendicular to the collision plane, (ii) a narrow-band laser ($\leq 1\text{MHz}$) circularly polarized. This allows a measurement of two quantities :

$$A(1/2) = \sum_m \sigma(3/2, 1/2 \rightarrow 1/2, m) ; A(3/2) = \sum_m \sigma(3/2, 3/2 \rightarrow 1/2, m)$$

as functions of energy (0.07 to 0.4 eV), from which the relative contributions of states $A^2 \pi_{3/2}$ and $B^2 \Sigma_{1/2}$ are derived.

- From the differential view point, a spectacular effect of M-selection has been found by Dören et al [14] : when excited potassium atoms $K^*(4^2P)$ are prepared by a σ^+ laser light (referred to $\hat{z}_i$ perpendicular to the initial velocity plane) and then collide with e.g. K_r atoms, a right-left asymmetry is observed in the differential cross section, and the difference between $+\theta$ and $-\theta$ scattered intensities oscillate with θ .

- In atom-diatom collisions, the initial angular momentum of the excited atom, and a part of the relative angular momentum can be transferred to the molecule. A M-selection with respect to the quantization axis $\hat{z}_i$ perpendicular to the collision plane, well defined in a differential experiment, allows a direct study of these angular momentum transfers, for instance in $\text{Na}^*(3P) + \text{N}_2$ or H_2 collisions [15].

b) examples for metastable levels

- A laser M-selection is possible also with metastable levels by a suitable choice of the optical transition, polarization and external magnetic field. For instance, for $\text{Ne}^*(^3P_2)$, one can use the Zeeman splittings between $3s\ ^3P_2 - 3p\ ^3P_2$ lines to separate different values of M_J .

This method has been applied recently by H. Haberland et al [09] to $\text{Ne}^* - \text{Ne}$ collisions. An important advantage of metastable atoms is that this preparation is made before the collision volume. Therefore it is possible, by means of an appropriate magnetic field ($\vec{B}$) configuration, to rotate adiabatically the quantization axis, for instance from $\hat{z}_i$ to the initial direction $\hat{R}_i$ of the molecular axis ; "adiabatically" means here that the time evolution of $\vec{B}$ seen by the moving atom is slow with respect to the precession period, so that the projection of the angular momentum along $\vec{B}$ is a constant. The same technique may be used to transform adiabatically $\hat{R}_i$ into $\hat{z}_f$. Finally preparation and analysis are referred to the molecular axis, which gives a very direct view of the rotational coupling effects : for an infinitely slow relative motion, the electronic angular momentum would be "attached" to $\hat{R}$ and a pure M-state would be observed along $\hat{R}_f$; in fact the rotation of $\hat{R}$ is not adiabatic and a combination of final M-states is observed. It may be noticed that such a measurement is extremely sensitive : strong depolarizing effects are observed even for very small couplings ; hence they do not necessarily imply that molecular potentials obtained via non M-selective measurements are wrong.

- Another way to achieve the M-selection for metastable atoms consists in using the Doppler velocity selection described in paragraph I.2. If the optical pumping takes place within a magnetic field, with a fixed laser frequency, each Zeeman component gives a hole in the velocity distribution, at a definite value of the velocity. Fig. 5 shows the hole profile obtained in neon, with a π polarization with respect to the magnetic field ($B \approx 480$ G) ; the angle between Ne^* beam and laser beam is 30° ; the solid lines correspond to the theoretical Zeeman spectrum. For a demonstrative purpose the experimental conditions have been arranged in such a way that the whole spectrum lies within the velocity distribution. Obviously it is preferable to increase the magnitude of B so that just one component remains in the distribution.

Fig 5 - Laser M-selection in a Ne^* metastable beam

II - LASER ASSISTED COLLISIONS

When the collision volume is irradiated by a high power laser beam, which frequency is more or less detuned with respect to atomic frequencies, the collision process is no longer independent of the interaction with the electromagnetic (EM) field. As shown in fig. 6 the "main" laser, characterized by its power (or electric field amplitude $\mathcal{E}_0$) and detuning (δ), acts during the collision itself. As indicated another laser can be used as a selector.

Fig 6 - Laser-assisted collision

To get a correct formulation of the problem, one has to consider as a whole both collision partners A,B interacting by V , the EM field and the interaction v between A(or B) and the EM field, i.e. to consider the hamiltonian :

$$H = h_A + h_B + h_R + V + v$$

$h_{A,B}$ are the internal hamiltonians of A,B, h_R is that of the EM field. At given collision energy and impact parameter the characteristic time for V is the collision time τ_c . The characteristic time for v is Ω'^{-1} , where Ω' is the Rabi frequency :

$$\Omega' = \left[\left(\frac{v \mathcal{E}_0}{\hbar} \right)^2 + \delta^2 \right]^{1/2}$$

The hamiltonian $h_{A(\text{or } B)} + h_R + v$ is that of the dressed atom, whereas in the Born-Oppenheimer approximation, $h_A + h_B + V$ describes the quasimolecule AB. The main theoretical problem here is to establish some preeminence among the various terms contained in V, and v, in other words to construct what we might call "dressed Hund's cases". Obviously the problem becomes serious — and interesting — for laser powers large enough to give $\Omega' \sim \tau_c^{-1}$.

Let us turn now to the experiment view point, and more specifically to crossed beam experiments. One knows the difficulties encountered in studying laser-assisted collisions in cell experiments, and the high powers generally required to attain the high intensity limit. Therefore to imagine crossed beam experiments in this field might seem rather utopic since collisional events in this type of experiment come from rare encounters between atoms belonging to two density media. However crossed beams are not necessarily a pure inconvenience : state-selections which are possible in such experiments can allow a separation of the investigated process from many other unuseful ones. Nevertheless one has to choose carefully the studied case and to respect some general requirements :

- (i) use low energies (i.e. large collision times and large distances)
- (ii) use excited species (potential curves are close, coupling more efficient), with long lifetimes.
- (iii) study long-range inelastic processes, involving dipole transitions,
- (iv) choose a laser assisted process easy to be characterized,
- (v) use a sufficiently high intensity to get $\Omega' \gtrsim \tau_c^{-1}$

A sensitive method to detect even small collisional laser effects consists in using interferences between coherent scattering amplitudes. As an example, let us consider a collision between two equal Z atoms A, A' ; A' is initially in its ground-state (0) whereas A is in a long-lived excited state (1) (metastable or Rydberg). The laser frequency is slightly detuned with respect to an allowed transition, from state (1) to an upper excited state (2), the lifetime of which is large compared to Ω'^{-1} and to the transit time T of A across the laser beam.

If the laser intensity is low (Ω'^{-1} is much larger than the duration τ_c of a collision), the collision process is completely decoupled from the laser interaction: a linear time dependent superposition of states (1) and (2) evolves within the laser beam up to a time t_c^- "just" before the collision time t_c (but in fact $\sim -\infty$ for the collision itself). In the time interval t_c^-, t_c^+ (just after collision), the adequate description is the molecular one. The evolution of the system is governed by potentials $1_{u,g}, 2_{u,g}$ (see fig. 7) and the laser has no appreciable effect during that time.

The outgoing collisional state is then a linear combination of atomic states A'_0 , $A_{1,2}$ and $A'_{1,2}$, A_0 (quasi-resonant excitation transfer). As the laser induces transition 1-2 for A only, the final ($t = T$) population in A_1 takes the following from :

$$|a_1(T)|^2 = C[1 + \rho \cos(2\Omega'T + \phi)]$$

where real quantities C, ρ and ϕ depend on moduli and relative phaseshift of the direct scattering amplitudes :

$$\frac{1}{2} [f_g^{(1)} + f_u^{(1)}] , \frac{1}{2} [f_g^{(2)} + f_u^{(2)}]$$

Fig 7 - Symmetric collision in a laser beam

At high laser intensity ($\tau_c \approx \Omega'^{-1}$), the laser effect can no longer be neglected within the time interval t_c^-, t_c^+ , for atoms A and for atoms A'. One is dealing with a 4-state problem (4 standard molecular states $1_g, 2_u; 1_u, 2_g$, coupled by the radiation field, or 4 dressed molecular states). The final result on $|a_1(T)|^2$ consists in an additional interference between direct and exchange scattering amplitudes.

Another example of a laser-modified interference effect is given by an asymmetric collision $A + B$, in which one of the two atoms has an internal angular momentum, for instance : $A(1, {}^1S) + B({}^1S)$, combined with $A(2, {}^1P) + B({}^1S)$, by means of a laser slightly detuned with respect to the $1, {}^1S - 2, {}^1P$ transition. At low laser intensity and for a linear polarization $\hat{e}$ parallel to the initial direction $\hat{R}_1$ of the inter-nuclear axis, the incoming collisional state ($t = t_c^-$) is a linear combination of states $A(1, {}^1S)$, $B({}^1S)$ and $A(2, {}^1P_0)$, $B({}^1S)$. From the molecular viewpoint, the atomic groundstate is connected to a Σ state whereas the excited one is connected to $\Sigma({}^1P_0)$

and $\Pi(^1P_{\pm 1})$ states. Therefore, if a long-range Π - Σ rotational coupling is present, the outcoming ($t = t_c^+$) collisional state takes the form :

$a_1(t_c) f_1 |^1S\rangle + a_2(t_c) \sum_{\lambda=0,\pm 1} f_2^{(\lambda)} |^1P_{\lambda}\rangle$, where the f - s are scattering amplitudes. For small scattering angles, the laser acts after the collision ($t_c^+ \leq t \leq T$) only on the projection : $a_1(t_c) f_1 |^1S\rangle + a_2(t_c) f_2^{(0)} |^1P_0\rangle$. (at large scattering angles, because of the rotation of $\hat{R}$, any component $^1P_{\lambda}$ would have a projection onto $M = 0$ along $\hat{e}$). Consequently the final groundstate population contains an interference term depending on $|f_1|$, $|f_2^{(0)}|$ and their relative phaseshift. As before, this interference term will be modified at large intensities, for which the laser acts on the system during the collision itself.

To conclude about the feasibility of such assisted crossed beam experiments, let us point out a promising experimental method developed recently by Pillet et al [16]. The basic idea is to use transitions between high n ($n \approx 20$) Rydberg levels, e.g. of Na, induced by a microwave field ; for instance :

Because of the very large values taken by the dipole moments, the so-called high-intensity regime is accessible with quite reasonable microwave powers (few W/cm^2) ; the long life time of such states is also an obvious advantage of the method.

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