Investigation of elastic and depolarizing collisions in strontium using broadband stimulated photon echoes
M. Saïdi, J.-C. Keller, J.-L. Le Gouët

To cite this version:

HAL Id: jpa-00210832
https://hal.archives-ouvertes.fr/jpa-00210832
Submitted on 1 Jan 1988

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
Investigation of elastic and depolarizing collisions in strontium using broadband stimulated photon echoes

M. Saïdi (1), J.-C. Keller (2) and J.-L. Le Gouët (2)

(1) Laboratoire de Spectronomie, Faculté des Sciences de Tunis, Tunisia
(2) Laboratoire Aimé Cotton*, CNRS II, Bât. 505, Université Paris-Sud, 91405 Orsay Cedex, France

(Reçu le 19 février 1988, accepté le 10 mai 1988)

Abstract. — Elastic and depolarizing collisions between excited strontium (5s 5p 3P1) and noble gases are investigated in a broadband stimulated photon echo (SPE) experiment. The stimulated photon echo signal produced under broadband (incoherent) excitation is calculated in the weak field regime both for correlated and for uncorrelated pulses using a simple statistical model for the light field. The relaxation of the echo signal due to elastic collisions is considered and the complete development of the corresponding propagator is derived. The collisional velocity changes concerning the total population and the alignment are compared precisely in the SPE experiment and the corresponding relaxation rates are measured. A simple long-range Van der Waals potential accounts for population scattering data; total elastic scattering cross sections are deduced from the comparison of experiment and calculation ($\sigma_{\text{el}}^{\text{el}} = 583(27) \, \text{Å}^2$; $\sigma_{\text{el}}^{\text{He}} = 190(6) \, \text{Å}^2$). The collisional model is extended for light perturbers to include both the small angle classical scattering region and the diffractive scattering region. Total rates for destruction of alignment are measured in an auxiliary photon echo experiment ($\sigma_{\text{He}}^{(2)} = 91(4) \, \text{Å}^2$; $\sigma_{\text{He}}^{(2)} = 102(14) \, \text{Å}^2$; $\sigma_{\text{He}}^{(2)} = 267(13) \, \text{Å}^2$; $\sigma_{\text{He}}^{(2)} = 329(21) \, \text{Å}^2$; $\sigma_{\text{He}}^{(2)} = 370(13) \, \text{Å}^2$). SPE data for depolarization are compared with a calculation based upon a long-range anisotropic model potential.

1. Introduction.

Several types of rephasing phenomena (photon echoes) have been observed in low pressure gases with appropriate pulsed optical excitation sequences [1]. The echo formation relies upon the creation of a specific atomic quantity that evolves during each time interval between the light pulses. The method can thus be used to measure the collisional relaxation rates of various well specified atomic quantities. These techniques also provide information on the velocity changes associated with the collisional relax-
ation of the atomic quantities of concern. The emission of the echo is due to the coincidence at some definite time $t_e$ of the Doppler phase of the oscillating dipoles that belong to the different velocity classes. Velocity changes occurring during the excitation sequence lead to imperfect rephasing and can be detected as a decrease of the echo signal. The relaxation of superposition states (optical coherences, Zeeman coherences, two-photon coherences...) as well as elastic and inelastic relaxation of the population of a single atomic state have been investigated in these ways [2-5]. In the last case the stimulated photon echo technique was used.

Stimulated photon echo (SPE) is produced by a three-pulse excitation sequence as illustrated in figure 1 [1, 5, 6]. In our case the laser pulses and the echo are copropagating. The first two pulses are resonant with the $a$-$b$ transition and are separated by a time interval $t_{12}$. The third pulse is resonant with the $b$-$c$ transition and is applied at a delay $t_{23}$ after the second pulse. Interaction between the atomic vapor and the first two pulses produces a sinusoidal modulation (Fig. 1) in the longitudinal velocity distribution of level $b$; the modulation period is $\delta t_e = \lambda / t_{12}$ where $\lambda$ is the wavelength of the $a$-$b$ transition [1]. The optical coherence $\rho_{bc}$ is built up from this modulation by the third pulse and the dipoles rephase to emit the echo at time $t_e = (\omega / \omega') t_{12}$ after this pulse. Echo formation involves three different atomic quantities during the excitation sequence (Fig. 1). The initial excitation memory is first transmitted by the optical coherence $\rho_{ab}$ between the first two pulses, then by the modulation which survives in level $b$ between the second and the third pulses, and finally by the coherence $\rho_{bc}$ after the third pulse.

During the time interval $t_{23}$, relaxation of the echo results not only from depopulation of level $b$ by spontaneous emission and by inelastic (state-changing) collisions but also from destruction of the modulation in velocity space by elastic (velocity-changing) collisions. The echo signal is due to the atoms which have not undergone velocity changes $\Delta v_e$ larger than the modulation period.

SPE are also valuable to investigate inelastic collisional processes such as atomic excitation transfer between level $b$ and a closely lying level $b'$. If the third laser pulse is resonant with the $b'$-$c$ transition, a collisional transfer $b \rightarrow b'$ results in an echo signal provided that the velocity modulation is preserved in the inelastic process. Collisional transfer between Zeeman sublevels in ytterbium [5] as well as between fine structure sublevels in calcium [7] have been previously studied using SPE.

The light pulses used for SPE experiments in low pressure gases are produced by pulsed dye lasers which are generally not transform limited. It has been demonstrated recently that the use of such a broadband (incoherent) excitation can provide improved time resolution in SPE experiment [8, 9]. Previous calculations of the SPE signal are based upon the assumption of pulsed monochromatic excitation and are not relevant for the actual experiments. In this paper we first present a perturbative calculation of the SPE signal under broadband excitation using a simple statistical model for the incoherent light field (Sect. 2). The relaxation of the echo signal due to elastic collisions is considered in section 3 and a complete development of the corresponding propagator is derived. The experimental arrangement and the measurement procedure are briefly described in section 4. The results for strontium are discussed and are compared with collisional models in section 5.

2. Stimulated photon echo produced with broadband light sources.

Calculations of the SPE signal are generally based upon the assumption of short, transform-limited pulses [1]. Relaxation measurements in gases using SPE [5, 6] generally make use of pulsed dye laser with 5-10 ns pulse duration and spectral width of several GHz. These light pulses are not transform limited and their duration $\tau_L$ is larger than the inverse inhomogeneous (Doppler) width $\Omega_D^{-1}$. They must be treated as broadband (incoherent) sources. For this purpose the optical electric field is regarded as a stochastic function of time. The laser pulses are characterized by the pulse duration $\tau_L$ and also by the coherence time $\tau_c$ which should identify with the inverse spectral width $\Omega_L^{-1}$. It has been recently...
shown that the ultimate time resolution in such coherent transient experiment is determined by this coherence time $\tau_c$ [7-9]. A statistical average, denoted by $\langle \rangle$, is needed in order to define this quantity. For a stationary stochastic function of time $f(t)$, the coherence time corresponds to the width of the autocorrelation function $\langle f(t) f^*(t - \tau) \rangle$. An ensemble average is thus needed to account for the spectral properties of the pulsed broadband source (see Sect. 2.3). As a result, the stimulated photon echo signal itself is a random quantity which fluctuates from one realization to another, i.e. from one pulse sequence to another. Averaging over a large number of realizations, i.e. a large number of pump laser shots, is thus needed in order to approach the expectation value of the echo signal. In the following, we first obtain the expression of the SPE signal under weak field excitation for a single realization of the pulse sequence. Then we present and discuss a statistical model for the broadband light source and calculate the expectation value of the signal energy.

2.1 DENSITY MATRIX EQUATIONS. — The gaseous sample (three-level atoms) is excited by a sequence of three light pulses (Fig. 1). The light beams copropagate in the gas cell and the echo signal which occurs on b-c transition is detected in the direction of the light beams (Oz axis). The time intervals between the pulses, i.e. $t_{32} = (t_3 - t_2)$ and $t_{23} = (t_3 - t_2)$, are assumed to be much larger than the pulse duration $\tau_L$ (well separated pulses). The first two pulses are either produced by the same laser source (correlated light pulses) or by two independent laser sources (uncorrelated light pulses).

The (classical) electromagnetic fields are described by:

$$E_i(t) = \mathcal{E}_i(t) \cos(\omega t - kz + \psi_i(t))$$

where $\mathcal{E}_i(t)$ and $\psi_i(t)$ are slowly varying functions of time with regard to optical oscillations. The strength of the coupling between the atoms and the fields is characterized by the (time-dependent) Rabi frequency:

$$\chi(t) = 2 \pi \mu_1 \mathcal{E}_i(t) e^{i \eta(t)}/\hbar$$

where $\mu_1 = \mu_2 = \mu_{ab}$ (respectively $\mu_3 = \mu_{bc}$) is the dipole moment of the a-b (respectively b-c) transition.

The evolution of the density matrix elements of interest is thus governed, in the frame of the rotating wave approximation, by the following equations:

$$\begin{align*}
\dot{\rho}_{bb} &= \frac{i}{2} \left[ \chi^* \hat{p}_{ab} - \chi \hat{p}_{ba} \right] - \gamma_b \rho_{bb} \\
\dot{\rho}_{ab} &= \frac{i}{2} \chi \left[ \rho_{bb} - \rho_{aa} \right] + \frac{1}{2} \Delta \hat{p}_{ab} - \gamma_{ab} \hat{p}_{ab} \\
\dot{\rho}_{bc} &= \frac{i}{2} \chi' \left[ \rho_{cc} - \rho_{bb} \right] + \frac{1}{2} \Delta' \hat{p}_{bc} - \gamma_{bc} \hat{p}_{bc}
\end{align*}$$

for $t < t_3 - \tau_L$

for $t > t_3 - \tau_L$ (3)

The complete set of equations for the density matrix of the three-level system can be found in [10].

The central frequency of light pulses 1 and 2 (respectively 3) coincides with the corresponding atomic frequency: $\omega = \omega_{ab}$ (respectively $\omega' = \omega_{bc}$) so that $\Delta = kv_z$ and $\Delta' = k' v_z$; $v_z$ is the longitudinal atomic velocity.

2.2 WEAK FIELD CALCULATION OF THE ECHO SIGNAL. — The weak field solution of equations (3) can be easily obtained in a perturbative way. The unperturbed populations are: $\rho_{bb}^{(0)} = \rho_{cc}^{(0)} = 0$ and $\rho_{aa}^{(0)} = n W(v_z)$; $n$ is the total number of active atoms per unit volume and $W(v_z)$ is the (normalized) equilibrium velocity distribution. Neglecting the relaxation terms, the level b population produced by the first two pulses is:

$$\rho_{bb}^{(2)}(v_z, t) = \frac{n}{4} W(v_z) \int_{-\infty}^{t} dt' \chi^*(t') \times$$

$$\times \int_{-\infty}^{t} \chi(t') e^{i\Delta(t-t')} + \text{c.c.}$$

When the second light pulse has died out one gets:

$$\rho_{bb}^{(2)}(v_z) = \frac{n}{4} W(v_z) \int_{-\infty}^{\infty} d\tau e^{i\Delta \tau} \times$$

$$\times \int_{-\infty}^{\infty} d\tau' \chi^*(\tau') \chi(\tau)$$

This expression can be re-arranged as follows:

$$\rho_{bb}^{(2)}(v_z) = \frac{n}{4} W(v_z) |\tilde{\chi}(\Delta)|^2$$

where

$$\tilde{\chi}(\Delta) = \int \chi(t) e^{-i \Delta t} dt.$$
After substitution of the actual expression of \( \chi(t) \) in equation (5), the contribution to \( \rho_{bb}^{(2)} \) which depends upon both light pulses (and can thus lead to a SPE signal) is obtained as:

\[
\tilde{\rho}_{bb}^{(2)}(v_z) = \frac{n}{4} W(v_z) \times e^{-i \Delta t_{12}} \hat{x}_{2}(\Delta) \hat{x}_{2}^{*}(\Delta) + \text{c.c.} \quad (8)
\]

The third laser pulse converts this population into dipoles which can rephase at time \( t_3 + (k/k') t_{12} \):

\[
\tilde{\rho}_{bc}^{(3)}(v_z,t) = - \frac{i}{\hbar} n W(v_z) \int_{-\infty}^{t} dt' \chi_{3}(t' - t_3) \times e^{i \Delta (t' - t)} \hat{x}_{2}^{*}(\Delta) \hat{x}_{2}(\Delta) e^{-i \Delta t_{12}}. \quad (9)
\]

The velocity summation of \( \tilde{\rho}_{bc}^{(3)}(v_z,t) \) is performed and a new time variable \( \tau = t - t' = \frac{k}{k'} t_{12} \) is used to derive the following expression of the macroscopic dipole of the sample \( \tilde{\rho}_{bc}^{(3)}(t) \):

\[
\tilde{\rho}_{bc}^{(3)}(t) = - \frac{i}{\hbar} n \int_{-\infty}^{\infty} \frac{d\tau}{k} \hat{x}_{3}(t - t_{c} - \tau) \times e^{i k' v_x \tau} \int dv_{z} W(v_{z}) \hat{x}_{2}^{*}(kv_{z}) \hat{x}_{2}(kv_{z}) e^{i k' v_{z} \tau}. \quad (10)
\]

We have \( t_{12} \gg \tau_{e} \) so that the integration over \( \tau \) can be extended to \(-\infty\). The echo signal \( S(t) \) emitted on the b-c transition is proportional to \( |\tilde{\rho}_{bc}^{(3)}|^2 \):

\[
S(t) = \eta \int_{-\infty}^{\infty} d\tau \hat{x}_{2}(t - t_{c} - \tau) \times \int dv_{z} W(v_{z}) \hat{x}_{2}^{*}(kv_{z}) \hat{x}_{2}(kv_{z}) e^{i k' v_{z} \tau}. \quad (11)
\]

The proportionality constant \( \eta \) is independent of times \( t \), \( \tau \) and \( \tau' \) and of velocities \( v_{x} \) and \( v_{z}' \).

In the experiments the total energy emitted in the echo pulse is usually measured; this energy is proportional to \( S = \int S(t) \; dt \).

### 2.3 Statistical Properties of the Broadband Pulses

The stochastic nature of the electromagnetic field is contained in the Rabi frequency \( \chi(t) \) (Eq. (2)) whose phase and amplitude are random functions of time. We assume that \( \chi(t) \) can be expressed as:

\[
\chi(t) = \chi^{0}(t) \; e(t) \quad (12)
\]

where \( \chi^{0}(t) \) is a slowly varying function and \( e(t) \) is a stationary random function which satisfies the following relations \([8]\):

\[
\langle e(t) \rangle = 0 \quad \langle e(t) e^{*}(t - \tau) \rangle = g(\tau) \quad (13)
\]

\[
\langle e(t) e(t - \tau) \rangle = 0 .
\]

The autocorrelation function \( g(\tau) \) is normalized so that \( g(0) = 1 \).

The correlation time can be quantitatively defined by \( \tau_{c} = \int g(\tau) \; d\tau \) and we have \( \tau_{c} \ll \tau_{L} \). The mean pulse intensity is proportional to \( \langle |\chi(t)|^{2} \rangle = \langle |\chi^{0}(t)|^{2} \rangle \). Thus \( \langle |\chi(t)|^{2} \rangle \) appears to be the envelope of the pulse intensity with temporal width \( \tau_{L} \). The spectral properties of this broadband source can be deduced from the expression of the correlation in the spectral domain:

\[
G(\Delta, \delta) = \langle \hat{x}(\Delta) \hat{x}^{*}(\Delta + \delta) \rangle = \hat{g}(\Delta) \int d\tau |\chi^{0}(\tau)|^{2} \exp i \delta \tau \quad (14)
\]

where

\[
\hat{g}(\Delta) = \int d\tau g(\tau) \exp - i \Delta \tau.
\]

\( G(\Delta, \delta) \) is a function of \( \delta \) with width \( \tau_{L}^{-1} \). The pulse spectrum may be regarded as an ensemble of elementary uncorrelated slices placed side by side. The total spectral width is \( \tau_{c}^{-1} \) as determined by the width of \( \hat{g}(\Delta) \) and the width of each slice is \( \tau_{L}^{-1} \).

### 2.4 Expectation Value of the Signal Energy

The description of the light pulses is complemented by two additional assumptions:

i) the following inequalities holds for the correlation time \( \tau_{c} \), the pulse duration \( \tau_{L} \) and the inhomogeneous (Doppler) width \( \Omega_{D} \):

\[
\tau_{c}^{-1} \gg \Omega_{D} \gg \tau_{L}^{-1};
\]

ii) the random process is supposed to be Gaussian and we can use the moment factorization procedure \([8, 11]\).

Finally the third pulse is supposed to be uncorrelated with the first two ones. The latter point corresponds to the actual experimental situation where a separated laser source produces the third \( \omega' \) pulse (see paragraph 4).

The expectation value \( \langle S \rangle \) is deduced from equation (11)
The correlation time $T_c$ is much smaller than all other relevant characteristic times so that equation (15) can be written as follows:

$$\langle S \rangle = \frac{\eta n^2}{64 \tau_c} \left[ \int |x_3^q(t)|^2 dt \right]$$

$$\times \int d\tau \int d\tau' \int d\tau'' \int d\tau''' W(\tau) W(\tau') \phi(\Delta, \Delta') \exp ik' (\tau' - \tau) \tau .$$

Making use of the moment factorization procedure for a Gaussian process, one gets for the expectation value $\phi(\Delta, \Delta')$:

$$\phi(\Delta, \Delta') = \langle \hat{x}_1(\Delta) \hat{x}_1^*(\Delta') \rangle \langle \hat{x}_2(\Delta) \hat{x}_2^*(\Delta') \rangle + \langle \hat{x}_1(\Delta) \hat{x}_2^*(\Delta') \rangle \langle \hat{x}_2(\Delta) \hat{x}_1^*(\Delta') \rangle + \langle \hat{x}_1(\Delta) \hat{x}_1^*(\Delta') \rangle \langle \hat{x}_2(\Delta) \hat{x}_2^*(\Delta') \rangle .$$

According to equations (7) and (13), the last term is zero, the first one is non-zero only for correlated pulses and the second one contributes both for correlated and for uncorrelated pulses. When correlated pulses are used we have:

$$\chi_2(t - t_2) = \alpha \chi_1(t - t_1) \quad \chi_2(\Delta) = \alpha \chi_1(\Delta) \times \exp i \Delta (t_2 - t_1).$$

This corresponds to the experimental situation (see paragraph 4) where the first two pulses are obtained from a single beam by splitting, differential delay and recombination. The echo signal is obtained from equations (14), (16) and (17), when we consider that $f_2 D = c_1$ (i.e. $g(\alpha) = g(\theta) = r$, for the spectral range of interest).

The time scale for the $\theta$ dependence of the echo signal is $\Omega_D^{-1}$ for the first term (coherent signal) and $\tau_L$, as given by $h(\theta)$, for the second term (incoherent signal). However, since $\int_{-\infty}^{\infty} h(\theta) d\theta = 1$ [12], both contributions to the integrated signal are identical according to the Parseval-Plancherel theorem. When uncorrelated pulses are used we only have the incoherent signal (i.e. the second contribution in (18)).

We can define a mean Rabi frequency $\bar{\chi}_i$ and an effective pulse area as follows:

$$\bar{\chi}_i^2 = \frac{1}{\tau_L} \int |x_1^q(t)|^2 dt ; \quad \theta_i = \sqrt{\tau_c \tau_L} \bar{\chi}_i .$$

The S.P.E. signal corresponding to uncorrelated pulses (incoherent signal) is now expressed as:

$$\langle S \rangle_i = \frac{2 \pi n^2}{64 k'} \theta_1^2 \theta_2^2 \int (W(v))^2 dr .$$

The corresponding coherent signal obtained with correlated pulses is:

$$\langle S \rangle_c = \frac{\eta n^2}{64 k'} \theta_1^2 \theta_2^2 \int (W(v))^2 dr .$$

The above expression is identical to that obtained for short, transform-limited pulses ($\tau_c = \tau_L \ll \Omega_D^{-1}$) under small pulse area conditions ($\theta, \ll 1$).

2.5 VELOCITY MODULATED POPULATION. — During the time interval between the second and the third light pulse, the quantity of interest for the build up of the S.P.E is given by equation (8) for a definite realization of the pulse sequence. The memory of the successive interactions with the first two pulses is contained in the oscillating factor $\exp -ikv z t_12$ with a longitudinal velocity period $1/k l_12$. In the case of short monochromatic pulses, this results in an actual modulation in the velocity distribution of level b [1]:

$$\rho_{bb}(v_2) = \frac{n}{2} \theta_1 \theta_2 W(v_2) \cos (kv z t_12 + \phi_{12}) .$$

The same expression holds for the excitation value $\langle \rho_{bb}(v_2) \rangle$ of the contribution to the level b population obtained with correlated broadband pulses satisfying $\tau_L^{-1} \ll \Omega_D \ll \tau_c^{-1}$, provided that we use the effective pulse areas defined in equation (20). For uncorrelated broadband pulses, the expectation value $\langle \rho_{bb}(v_2) \rangle$ is zero. This is due to the random variations of the phase factor $\phi_{12}$ from one pulse sequence to the other and to the corresponding shift of the modulation extrema.

The expectation value of the signal $\langle S \rangle$ is a function of the second order momentum of $\rho_{bb}(v_2)$ i.e. of $\phi(\Delta, \Delta')$ (Eq. (17)). According to equations (13), (14) and (17) this quantity can be expressed as:

$$\Phi(\Delta, \Delta') = \theta_1^2 \theta_2^2 / \tau_c^2 \hat{g}(\Delta) \hat{g}(\Delta') \times [1 + \psi(\delta) \psi(\delta)] .$$
where $\delta = (\Delta - \Delta')$ and
\[
\psi_i(\delta) = \int dt \left| \chi_i^0(t) \right|^2 \exp i \delta t / \int dt \left| \chi_i^0(t) \right|^2.
\] (25)

The first contribution in equation (24) is obtained with correlated pulses only while the second contribution is obtained either with correlated pulses or with independent pulses. The spectral width of $\tilde{g}(\Delta)$ is $\Delta_{D}^{-1} \gg \Omega_D$ while the spectral width of $\psi(\delta)$ is $\Delta_{L}^{-1} \ll \Omega_D$. The effective modulation is thus produced either over the whole Doppler width (coherent term) or over narrow independent portions with width $\Delta_{L}^{-1}$ placed side by side in the Doppler profile (incoherent term). When the delay $t_{12}$ is smaller than the pulse duration $\tau_1$, the effective modulation (period $(t_{12})^{-1} < \tau_{D}^{-1}$) can be obtained with correlated pulses only.

3. Relaxation of the echo signal due to elastic collisions.

In order to describe relaxation, one has to introduce a multiplying factor $\Lambda(v_z)$ in the expression of $\tilde{\rho}_{bc}^{(3)}(v_z, t)$ (Eq. (9)) before performing the summation over longitudinal velocity. This results in the change of $W(v_z)$ into $\Lambda(v_z)W(v_z)$ in the expression of the echo signal $S(t_{23})$, with:
\[
\Lambda(v_z) = \exp \left\{ -\gamma_{ab}(v_z) t_{12} - \gamma_{bb}(v_z, t_{12}) t_{23} - \gamma_{bc}(v_z) \frac{k}{k'} t_{12} \right\}. \tag{26}
\]

Radiative and collisional relaxations during the light pulses are neglected since $\tau_1$ is supposed to be much shorter than the time intervals $t_{12}$ and $t_{23}$. Each relaxation rate $\gamma_{ij}(v_z)$ is the sum of a natural relaxation rate $\gamma_{ii}$ and of a collisional relaxation rate $\Gamma_{ij}(v_z)$. Only velocity averaged relaxation rates $\bar{\Gamma}_{bb}(t_{12})$ and $\bar{\Gamma}_{ab} + (k/k') \bar{\Gamma}_{bc}$, as defined in paragraph 3.4, can be measured in our experiment.

The velocity dependent relaxation rates $\gamma_{ij}(v_z)$ introduced (Eq. (26)) in the expression of the echo signal can account for the effect of binary collisions between the active atoms and foreign perturbers admixed in the vapor cell.

Elastic collisions contribute to the decay of the echo signal along two ways. They destroy the quantum mechanical coherence between states $a$ and $b$ (a and c) and this results in the decay of $\rho_{ab}(\rho_{bc})$. They also produce velocity changes of atoms whether they are in a superposition state or in a pure state. The consequence of these effects is threefold. Collisions which destroy coherence between states add their phase-interruption rate $\Gamma_{ij}^{ph}$ to the natural decay rate $\gamma_{ij}$ of the coherence $\rho_{ij}$. Collisional velocity changes perturb the building of the Doppler phase shifts $k v_z t_{ij}$ and they hamper the perfect rephasing of the dipoles at the echo time. Finally, collisional velocity changes wash out the periodic structure which is stored in the velocity distribution of level populations and which preserves the excitation memory in the stimulated photon echo scheme.

When collisions occur in a superposition state they perturb both the internal coherence and the atomic velocity [2, 13]. The superposition state of concern in the photon echo scheme combines states which are separated by optical distances. These states are different enough so that close encounters destroy the coherence between them. Thus short distance, large scattering angle collisions are completely accounted for by their contribution to $\Gamma_{ij}^{ph}$ while long distance, small scattering angle collisions which preserve optical coherences are described by velocity changes of atoms in a superposition state. In other words, as long as classical trajectories can be used (classical collision region), they are so different for the two states that the coherent superposition is destroyed. This is no longer the case in the diffractive scattering region where velocity change can occur while preserving the coherent superposition. If the accumulated velocity change during the time interval $\tau = \delta u$, the latter effect is negligible as long as $k \tau < \delta u << 1$, otherwise it results in a time-dependent relaxation rate $\Gamma_{ij}(\tau)$ for optical coherence with [2]:
\[
\Gamma_{ij}(\tau) - \Gamma_{ij}(0) = a \tau^2 + \cdots. \tag{27}
\]

Except for a few measurements with He foreign gas (see paragraph 5.1), this correction is negligible in our experiments due to the rather low values for $t_{12}$.

Collisions which occur in a pure state obliterate the periodic structure stored in the corresponding velocity distribution. An atom no longer contributes to this structure as soon as its accumulated velocity change between $t_2$ and $t_3$ is larger than the modulation period $2 \pi / k t_{12}$. Thus a SPE signal collects the contributions from the only atoms for which the collisional velocity change during the $t_{23}$ interval does not exceed $2 \pi / k t_{12} [6]$. In present experiments where $t_{12}$ is varied typically between 10 ns and 1 $\mu$s, this upper limit is contained between 50 m/s and 0.5 m/s. It should be compared with the thermal velocity which amounts several hundred m/s. Thus only atoms which have undergone small velocity changes can contribute to the echo signal. After its building at time $t_2$, the periodic structure in velocity space decays at a rate $\Gamma_{bb}(t_{12})$ which depends upon the period $2 \pi / k t_{12}$ of the structure. In the present section we derive an expression for this relaxation rate involving quantities of interest for collisional problems such as differential cross section or collisional kernel.
3.1 TRANSPORT EQUATION. — Let us consider a homogeneous sample of active atoms interacting with a bath of perturbators with density $N$ and velocity distribution function $W_p(v_p)$. The probability density for the diffusion of an active atom from velocity $v$ to velocity $v'$ is given by:

$$N \, d^3v' \int d^3v'_p \, d^3v_p \, W(v_p) \, u^{-1} \left( \frac{d\tau}{dt} \right) \times$$

$$\times \delta(u - u') \, \delta(V - V') = W(v, v') \, d^3v'$$  \(28\)

$u$ is the relative velocity with modulus $u$, $V$ is the centre of mass velocity thus $v = u + V$, non-primed or primed symbols are used respectively for initial and final velocities. $W(v, v')$ is known as the collision kernel.

The evolution of the number of active atoms $P_{aa}(v)$ $d^3v$ in level $\alpha$ within the velocity space volume $d^3v$ is governed by the balance between atoms arriving in the volume and those escaping from it

$$\frac{d}{dt} \rho_{aa}(v) = -\Gamma_{aa}(v) \rho_{aa}(v) + \int d^3v' \, W_{aa}(v', v) \rho_{aa}(v')$$  \(29\)

The formal solution of equation (29) can be obtained using the propagator $G_{aa}(v', v, t - t')$:

$$\rho_{aa}(v, t) = \int \rho_{aa}(v', v, t - t') \, W_{aa}(v', v) \, d^3v'.$$  \(30\)

This propagator obeys the following equations [14]:

$$\Gamma_{aa}(v, v, t - t') = -\Gamma_{aa}(v) \, G_{aa}(v', v, t - t') + \int d^3v'' \, W_{aa}(v'', v) \, G_{aa}(v', v, t - t').$$  \(32\)

The propagator $G_{aa}$ represents the fraction of the atomic population transferred from velocity class $v'$ towards velocity class $v$ during the time interval $(t - t')$. When the velocity changes of interest are much smaller than the mean atomic velocity and provided the time interval $(t - t')$ is much smaller than the thermalization time, this propagator is a slowly varying function of $v$ for a fixed value of $(v - v')$ and a rapidly varying function of $(v - v')$ for a fixed value of $v$. The interactions with the laser only select the velocity component along the beam direction (longitudinal velocity). The transverse atomic motion is not directly modified and we neglect the influence of the non equilibrium longitudinal velocity distribution on this transverse movement. It results that the level-population velocity-distribution takes on the form:

$$\rho_{aa}(v, t) = \rho_{aa}(v, v') \, W(v).$$  \(34\)

This allows us to define, and to use, a unidirectional propagator as:

$$G_{aa}(v', v, t - t') = \int d^3v''_+ \, d^3v'_+ \, W(v'_+) \, G_{aa}(v', v, t - t').$$  \(35\)

where $W(v'_+)$ is the equilibrium transverse velocity distribution of the active atoms. We also define other unidirectional functions:

$$W_{aa}(v'_+, v) = \int d^3v' \, W(v') \, W_{aa}(v', v).$$  \(36\)

$$\Gamma_{aa}(v) = \int d^3v'_+ \, W(v'_+) \, G_{aa}(v, v').$$  \(37\)

The unidirectional propagator obeys the equations obtained from equation (32) after substitution of the involved quantities by their unidirectional counterpart. The evolution of the density matrix $\rho_{aa}(v, t)$ is given by:

$$\rho_{aa}(v, t) = \int d^3v''_+ \, G_{aa}(v', v, t - t') \, \rho_{aa}(v', t').$$  \(38\)

3.2 EVOLUTION OF THE VELOCITY MODULATED POPULATION. — After the extinction of the second pulse the population $\bar{\rho}_{bb}^{(2)}(v, t)$ is given by:

$$\bar{\rho}_{bb}^{(2)}(v, t) = \frac{n}{4} \int d^3v'_+ \, G_{bb}(v'_+, v, t - t_2) \, W(v'_+),$$

$$x \, \mathcal{X}_{l}^{*}(kv'_+) \, \mathcal{X}_{2}(kv'_+) \, \exp -i \, k \, v'_+ \, t_{12} + \text{c.c.}$$  \(39\)

From the value of this population at time $t = t_3$ one can deduce the expression of the atomic dipole $\bar{\rho}_{bb}^{(2)}(v, t)$ after the third pulse and that of the echo signal $S(t)$. The expectation value of the integrated signal is then obtained as (cf. paragraph 2.2):

$$\langle S \rangle = \frac{n^2}{64} \int dr \int d\tau \int d\tau' \left( \chi_{1}^{*}(t - t_c - \tau) \chi_{3}(t - t_c - \tau') \right) \int d^3v \, d^3v' \, \exp i \, k \, (v'_{c} - v_{c} \tau' - v_{z} \tau) \times$$

$$\times \int d^3v' \, d^3v'' \, G_{bb}(v''_{z}, v'_{z}, t_{23}) \, G_{bb}(v''_{z}, v'_{z}, t_{23}) \, W(v'_{z}) \, W(v''_{z}) \, \Phi(kv'_+, kv''_+).$$

$$\times \exp -i \, k \, (v'_{z} - v''_{z}) \times t_{12} \times \exp i \, k \, (v'_{z} - v''_{z}) \times t_{12}.$$

\(40\)
Let us define a new propagator \( \hat{G} \):
\[
\hat{G}(kt_{12}, v_z, t - t_2) = \int d\xi \ G_{bb}(v_z + \xi, v_z, t - t_2) e^{-i\xi kt_{12}}. \tag{41}
\]

If we assume that \( G_{bb}(v_z + \xi, v_z, t - t_2) \) is an even function of \( \xi \), then the propagator \( \hat{G}(kt_{12}, v_z, t - t_2) \) is a real quantity.

The statistical properties of the light pulses (Eqs. (13, 14, 17)), as well as the properties of the propagators \( G_{bb} \) (cf. paragraph 3.1) and \( \hat{G}_{bb} \), are then taken into account to derive the following expression for the S.P.E. signal, under the assumption that \( \tau_L < t_{12} \):
\[
\langle S \rangle = \frac{n n^2}{64 k} \theta^2 \int d\theta \times \left[ \left| \int d v_z e^{i k v_z \theta} W(v_z) \hat{G}(kt_{12}, v_z, t_2) \right|^2 - 2 \pi \hbar \theta \int d v_z |W(v_z)\hat{G}(kt_{12}, v_z, t_2)|^2 \right]. \tag{42}
\]

Within our model, the plain effect of elastic collisions during the time interval \( t_{23} \) is to change \( W(v_z) \) into \( \hat{G}(kt_{12}, v_z, t_2) W(v_z) \) in the expression of the echo signal (Eq. (18)). Despite of the existence of two different contributions to the echo signal, the effect of elastic collisions can be taken into account by only one propagator \( \hat{G}(kt_{12}, v_z, t_{23}) \). This result, obtained for well-separated pulses \( (t_{12} \gg \tau_L) \), would no longer hold for overlapping pulses \( (t_{12} < \tau_L) \) [7]. In the latter situation the collisional relaxation of the incoherent signal should be different from that of the coherent one.

3.3 DETERMINATION OF THE PROPAGATOR. — The propagator \( \hat{G} \) must satisfy the following equation, deduced from equations (32, 33) and definitions (34-37) and (41):
\[
\frac{\partial}{\partial t} \hat{G}_{bb}(x, v_z, t - t_2) = -\Gamma_{bb}(v_z) \hat{G}_{bb}(x, v_z, t - t_2) + \int d\xi W_{bb}(v_z + \xi, v_z) e^{i\xi t} \hat{G}_{bb}(x, v_z + \xi, t - t_2) \tag{43}
\]

3.3.1 Approximate expression for \( \hat{G} \). — \( \hat{G}(x, v_z + \xi, t - t_2) \) is a slowly varying function of \( \xi \) which can be identified with \( \hat{G}(x, v_z, t - t_2) \) and factorized out of the integral in equation (43) to give the following approximation for the propagator:
\[
\hat{G}_{bb}(kt_{12}, v_z, t - t_2) = \exp -\Gamma_{bb}(v_z, t_{12})(t - t_2) \tag{45}
\]
\[
\Gamma_{bb}(v_z, t_{12}) = \Gamma_{bb}(v_z) - \int d\xi W_{bb}(v_z + \xi, v_z) e^{i\xi t_{12}}. \tag{46}
\]

Let us consider two limiting cases for the velocity change \( \Delta v_z \) during the time interval \( t_{23} \):

(a) \( k \Delta v_z t_{12} \ll 1 \); the velocity changes do not participate in the decay rate of the echo signal during this time interval \( (\Gamma_{bb}(v_z, t_{12}) = 0) \) since, as \( \exp(k \Delta v_z t_{12}) \approx 1 \), the departure term \( \Gamma_{bb}(v_z) \) and the restitution term balance each other (Eq. (30));

(b) \( k \Delta v_z t_{12} \gg 1 \); the contributions to the restitution term average to zero and the relaxation rate for the destruction of the periodic structure in the velocity space identifies with the total elastic rate \( \Gamma^{el}(v_z) \).

Except for the additional constant \( \Gamma^{el}(v_z) \), \( \Gamma_{bb}(v_z, t_{12}) \) is the Fourier transform of the collision kernel.

3.3.2 Complete development of the propagator. — The slow variation of \( \hat{G}(kt_{12}, v_z, t) \) with \( v_z \) suggest that equation (43) can be solved by an iterative procedure using a development for the propagator:
\[
\hat{G}_{bb}(kt_{12}, v_z, t) = \sum_{n=0}^{\infty} \hat{G}^{(n)}_{bb}(kt_{12}, v_z, t). \tag{47}
\]

The different terms in the expansion are obtained as solutions of the following system of coupled equations:
\[
\frac{\partial}{\partial t} \hat{G}^{(0)}_{bb}(kt_{12}, v_z, t) = -\Gamma_{bb}(v_z) \hat{G}^{(0)}_{bb}(kt_{12}, v_z, t) + \int d\xi W_{bb}(v_z + \xi, v_z) e^{i\xi t_{12}} \hat{G}^{(0)}_{bb}(kt_{12}, v_z + \xi, t) \tag{48}
\]
\[
\hat{G}^{(n)}_{bb}(kt_{12}, v_z, t) = -\left[ \Gamma_{bb}(v_z) - \int d\xi W_{bb}(v_z + \xi, v_z) e^{i\xi t_{12}} \right] \hat{G}^{(n-1)}_{bb}(kt_{12}, v_z, t) + \int d\xi W_{bb}(v_z + \xi, v_z) e^{i\xi t_{12}} \left[ \hat{G}^{(n-1)}_{bb}(kt_{12}, v_z + \xi, t) - \hat{G}^{(n-1)}_{bb}(kt_{12}, v_z, t) \right] \tag{49}
\]
\[
\hat{G}_{bb}(kt_{12}, v_z, 0) = \delta_{m0}. \tag{50}
\]
The approximate expression derived in paragraph 3.3.1 identifies with the first term of the expansion $G^{(0)}(kt_{12}, v_z, t)$. For small $t$ values, far from going back to thermodynamical equilibrium, $G^{(0)}$ is a good approximation for $\hat{G}$. The difference between $\hat{G}$ and $G^{(0)}$ increases when we approach this equilibrium. For large $t$ values, the population is thermalized and the propagator is $\hat{G}(kt_{12}, v_z, t) = \delta (t_{12}) W(v_z)$. This propagator does not satisfy the equation for $G^{(0)}$.

**3.4 VELOCITY AVERAGED RELAXATION RATES.**

In our experiments the relaxation rates are obtained by measuring the ratio $R(p)$ of the echo signal at a given (low) perturber pressure $p$ and of the same signal at zero perturber pressure. According to the expression of the echo signal (paragraph 2) and of the relaxation terms (paragraph 3), this ratio is:

$$R(p) = \int dv_z(W(v_z))^2 \exp -2 \Gamma(v_z, t_{12})/ \int dv_z(W(v_z))^2$$

where:

$$\Gamma(v_z, t_{12}) = \left[ \Gamma_{ab}(v_z) + \frac{k}{\hbar} \Gamma_{bc}(v_z) \right] \times t_{12} \left[ \Gamma_{bb}(v_z, t_{12}) \right] t_{23}.$$

Let us define the velocity averaged quantities:

$$\Gamma_{ij} = \int dv_z(W(v_z))^2 \Gamma_{ij}(v_z)/ \int dv_z(W(v_z))^2$$

$$\Gamma_{bb}(t_{12}) = \int dv_z(W(v_z))^2 \Gamma_{bb}(v_z, t_{12})/ \int dv_z(W(v_z))^2$$

$$\Gamma(t_{12}) = \int dv_z(W(v_z))^2 \Gamma(v_z, t_{12})/ \int dv_z(W(v_z))^2$$

$$= \left[ \Gamma_{ab} + \frac{k}{\hbar} \Gamma_{bc} \right] t_{12} + \left[ \Gamma_{bb}(t_{12}) \right] t_{23}.$$

$$\bar{R}(p) = \exp -2 \Gamma(t_{12}).$$

As long as the ratio of $R(p)$ and $\bar{R}(p)$ is close to unity, one gets an exponential decay law for the measured ratio (Eq. (56)) which allows to deduce the velocity averaged relaxation rates given by equations (53), (55).

The error which results of the substitution of $\bar{R}(p)$ to $R(p)$ is given by:

$$[R(p)/\bar{R}(p)] = 1 + 2 (\bar{\Gamma}^2 - \Gamma^2) + \ldots$$

where

$$\bar{\Gamma}^2 = \int dv_z(W(v_z))^2 \Gamma^2(v_z, t_{12})/ \int dv_z(W(v_z))^2.$$

**3.5 ELASTIC SCATTERING OF $k$ MULTIPOLES.**

Up to now, the formation and the relaxation of the echo signal have been discussed without considering the Zeeman degeneracy of levels and the laser beam polarizations, so that we were concerned with population in level $b$ only. In a real situation, we create a population $\rho^b$ in level $b$ but, possibly also, orientation $\rho^s$, alignment $\rho^l$ and higher order multipoles according to $J$ values and to beam polarizations. The expansion of the density matrix on an irreducible tensorial basis greatly simplifies the analysis of the problem [15, 16]. The transport equation which determines the collisional evolution of the density matrix elements $\rho^k_{\phi, \phi'}$ of active atoms immersed in a perturber bath is given by [17]:

$$\frac{\partial}{\partial t} \rho^k_{\phi, \phi'}(v) = - \sum_{\phi''} \Gamma_{\phi \phi''}^k(v) \rho^k_{\phi'', \phi'}(v) + \int W^k_{\phi \phi''}(v'') \rho^k_{\phi'', \phi'}(v'') d^3 v'.$$

As discussed with many details in [5], under various assumptions one can neglect the collisional coupling between multipoles as well as between the different components of a given multipole and thus obtain for unidirectional quantities:

$$\Gamma_{\phi \phi''}^k(v_z) = \delta_{kk} \delta_{\phi \phi''} \Gamma^k(v_z)$$

$$W^k_{\phi \phi''}(v_z, v_z') = W^k(v_z', v_z)$$

$$\Gamma^k(v_z) = \int W^k(v_z', v_z) dv_z'.$$

In the same way as discussed in paragraph 3.3 and 3.4, velocity averaged relaxation rates can be defined for the $k$ multipole modulated in the velocity space as obtained in SPE experiments:

$$\Gamma^k_{bb}(t_{12}) = \int dv_z \Gamma^k_{bb}(v_z, t_{12})(W(v_z))^2/ \int dv_z(W(v_z))^2$$

$$\Gamma^k_{bb}(v_z, t_{12}) = \Gamma^k(v_z) -$$

$$\int W^k(v_z + \xi, v_z) e^{ik\xi t_{12}} d\xi.$$
The relaxation rate of the velocity modulated total population $R_g(t_{12})$ goes to zero for small values of $t_{12}$ because, in this case, the restitution term balances the departure term in equation (64). For large values of $t_{12}$, the oscillating term is responsible for a cancellation of the restitution term and the relaxation rate equals the total elastic rate for all the $k$-multipoles: $\lim_{t_{12} \to \infty} R_b^e(t_{12}) = R_{el}^{d}$ [18, 19].

4. Stimulated photon echo in strontium vapor.

Our experiment has been performed with Sr atoms ($Z = 38$) using level $5s^2 \, ^1S_0$, $5s \, 5p \, ^3P_1$ (lifetime 21 $\mu$s) and $5s \, 5d \, ^3D_1$ (lifetime 16.7 ns) as level a, b and c respectively. The corresponding wavelengths are indicated in figure 2. The laser beams are linearly polarized and the echo is detected in the same linear polarization as the third laser pulse. The polarization direction of the first two pulses is either perpendicular (Fig. 2b) or parallel (Fig. 2c) to that of the third pulse. In the latter case, the signal only appears when depolarizing ($m$ state changing) collisions occur. Due to the particular $J$ values and polarizations, we can only produce population $\rho^0$ and alignment $\rho^2$ in level b. Denoting respectively $I_\perp (p)$ and $I_\parallel (p)$, the echo signals detected with perpendicular (Fig. 2b) or parallel (Fig. 2c) polarization setting at a given perturber pressure $p$, we obtain [5]:

$$\frac{I_\perp (p)}{I_\perp (0)} = \exp - \left[ R_{ab} + \frac{k}{k} R_{bc} \right] t_{12} \times \exp - \left[ R_b^{(0)}(t_{12}) \right] t_{23} \times \left\{ 2 + \exp - \left[ R_b^{(2)}(t_{12}) - R_b^{(0)}(t_{12}) \right] t_{23} \right\}$$

$$\frac{I_\parallel (p)}{I_\parallel (0)} = \left\{ 2 - 2 \exp - \left[ R_b^{(2)}(t_{12}) - R_b^{(0)}(t_{12}) \right] t_{23} \right\} / \left\{ 2 + \exp - \left[ R_b^{(2)}(t_{12}) - R_b^{(0)}(t_{12}) \right] t_{23} \right\}$$

The relaxation rates are measured as follows:

a) we first set $t_{23} = 0$ and measure $I_\perp (p)/I_\perp (0)$ for different perturber pressure to determine $\frac{d}{dp} \left[ R_{ab} + \frac{k}{k} R_{bc} \right]$;

b) for $t_{23} \neq 0$, we measure $I_\parallel (p)/I_\perp (p)$ as a function of $p$ and obtain $\frac{d}{dp} \left[ R_b^{(2)}(t_{12}) - R_b^{(0)}(t_{12}) \right]$;

c) for $t_{23} \neq 0$, we measure $I_\perp (p)/I_\perp (0)$ as a function of $p$ and, taking into account the results of a) and b), we deduce $\frac{d}{dp} R_b^{(0)}(t_{12})$ for different values of $t_{12}$.

The measurement of the total rate of destruction of alignment is performed in an auxiliary standard photon echo experiment on the b-c transition [5]. A first linearly-polarized laser pulse, resonant with the a-b transition, populates the sublevel $m = 0$ in level b at time $t = 0$. Two subsequent laser pulses, resonant with the b-c transition, irradiate the sample at times $T$ and $(T + \tau)$. They produce a regular photon echo signal at time $(T + 2 \tau)$ from the population which survives in level b at time $T$. According to whether their polarization is crossed with or parallel to the first pulse polarization, the last two pulses probe either the $m = 0$ or the $m = \pm 1$ sublevels. In the latter case, the signal in zero in the absence of collisional transfer between Zeeman sublevels. Since no velocity selection operates on level b multipoles, the ratio of echo signals in

Fig. 2. — Stimulated photon echo in strontium. (a) Three level scheme. (b) Crossed polarization excitation. (c) Parallel polarization excitation.
crossed and parallel configurations is related to \( \gamma_b^{(2)} \) by:

\[
\left[ \frac{I_1(p)}{I_\perp(p)} \right]^{1/2} = \frac{(2 - 2 \exp - \gamma_b^{(2)} T)/}{(2 + \exp - \gamma_b^{(2)} T)}.
\]  

From the measurement of this ratio at different perturber gas pressure we obtain \( \frac{d}{dp} \gamma_b^{(2)} \). The corresponding cross-sections \( \Sigma^{(2)} \) for destruction of alignment are given in table I.

Table I. — Experimental and calculated cross-sections for depolarization in Sr-noble gas collisions.

<table>
<thead>
<tr>
<th></th>
<th>( \Sigma^{(2)} )</th>
<th>( \frac{d}{dp} \gamma_b^{(2)} )</th>
<th>( \Sigma^{(2)} )</th>
<th>( \frac{d}{dp} \gamma_b^{(2)} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>91 (4)</td>
<td>1.4</td>
<td>16</td>
<td></td>
</tr>
<tr>
<td>Ne</td>
<td>102 (14)</td>
<td>2.7</td>
<td>28</td>
<td></td>
</tr>
<tr>
<td>Ar</td>
<td>267 (13)</td>
<td>11.1</td>
<td>55</td>
<td></td>
</tr>
<tr>
<td>Kr</td>
<td>329 (21)</td>
<td>16.7</td>
<td>70</td>
<td></td>
</tr>
<tr>
<td>Xe</td>
<td>370 (13)</td>
<td>27.8</td>
<td>90</td>
<td></td>
</tr>
</tbody>
</table>

The experimental set-up is shown in figure 3. The pumping source is a Nd-Yag laser (Quantel) operating at 15 Hz repetition rate. The frequency doubled beam (\( \lambda = 532 \text{ nm} \)) is used to pump the second dye laser (operating at 488 nm (coumarin 480 + methanol)). A set of beam splitters is used to separate the necessary beams and recombine them before entering the oven. The delay \( t_{12} \) can be varied with 8 ns increments using a first white cell delay line while large values for the delay \( t_{23} \) are given by a second delay line (increment 34 ns). The oven (\( \theta = 400^\circ \text{C} \)), the electro-optical shutter and the detection scheme are the same as in [5].

5. Results and discussion.

5.1 RELAXATION OF OPTICAL COHERENCES. — The measured quantity is \( \beta_1 = \frac{d}{dp} \left[ \frac{\Gamma_{ab} + k}{k} \Gamma_{bc} \right] \). As discussed at the beginning of paragraph 3, \( \beta_1 \) includes two contributions; the first one is the usual dephasing term and the second one is due to velocity changing collisions occurring in the diffractive scattering region which can preserve the superposition states. The photon echo experiment is sensitive to the latter contribution when \( k \Delta v^d \cdot t_{12} > 1 \); \( \Delta v^d \) is the longitudinal velocity change associated with diffractive scattering [20]: \( \Delta v^d \sim \frac{h}{m} (\sigma^d)^{1/2} \); \( \sigma^d \) is the total elastic scattering cross-section and \( m \) is the mass of the active atoms. Owing to the values of \( \sigma^d \) (see Tab. II) and to the range of variation of \( t_{12} \) in our measurements (10 ns \( \leq t_{12} \leq 114 \text{ ns} \) with He perturber; 10 ns \( \leq t_{12} \leq 66 \text{ ns} \) with Ar perturber), diffractive collisions only contribute with He perturber for the largest values of \( t_{12} \). This results in a variation of the relaxation rate compatible with equation (27). We thus obtained for strontium-ar-
Table II. — Experimental and calculated cross-sections for elastic scattering in Sr*-noble gas collisions.

<table>
<thead>
<tr>
<th></th>
<th>A(β)</th>
<th>σ_{exp} (Å)²</th>
<th>σ_{cal} (Å)²</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ar</td>
<td>0.77</td>
<td>583 (27)</td>
<td>737</td>
</tr>
<tr>
<td>He</td>
<td>0.82</td>
<td>190 (6)</td>
<td>217</td>
</tr>
</tbody>
</table>

gon collisions : β₁ = (64 × 10^6 s⁻¹ 1 Torr⁻¹), and for strontium-helium collisions :

β₁ = (58 × 10^6 s⁻¹ 1 Torr⁻¹) +
+ (8 × 10^20 s⁻² 1 Torr⁻¹)(t₁₂)².

These values have been used in the determination of the relaxation rates for the velocity modulated population and alignment as discussed in paragraph 4.

5.2 ELASTIC SCATTERING IN Sr (5s 5p ³P₁)-NOBLE GAS COLLISIONS. — With the exception of a work concerned with alignment and orientation in the ³P₁ level of Yb [21], the photon echo experiments have not made possible the direct inversion of the Fourier transform in equation (46) to provide an experimental collision kernel. SPE data have thus been discussed along two different approaches. One can assume a particular empirical shape for the collisional kernel (Lorentzian, Gaussian ...) and use two parameters, namely the width of the kernel and the total elastic rate, to be fitted to the experimental results [3, 6]. One can also start with the interatomic potential to evaluate the differential cross-sections, the collisional kernel and the relaxation rate. Such a potential has been computed for excited strontium (1P₁) interacting with ground state argon perturber [22].

When the active atom is in a J = 1 level and interacts with a structureless perturber, the multipolar expansion of the interatomic potential is composed of two terms only :

V(r, j) = V(0)(r) + \left( j_j^2 - \frac{j_j^2}{3} \right) V^(2)(r) (69)

V(0)(r) and V^(2)(r) are the isotropic and the anisotropic components respectively ; r is the interatomic distance and j_j is the component of the kinetic momentum j along the internuclear axis. We shall assume that the long range part of the potential is predominant and corresponds to a dipole-dipole interaction :

V(0)(r) = - C(0)/r⁶; \quad C(0) ≈ 0
V^(2)(r) = - C(2)/r²; \quad C(2) positive or negative.

In a previous experiment on Yb, we used such a potential to derive total elastic cross-sections from the data [5]. Our value of the Yb*-He cross-section was confirmed in a subsequent measurement performed by other authors [3].

An analytical expression has been derived previously for \( \Gamma_b^{(0)}(t₁₂) \), assuming a C₄/r⁴ potential and small angle classical elastic scattering [5]. In this case, the adjustable parameter is the potential coefficient C₄ or the related total elastic scattering cross-section σ_el. The corresponding rate for the relaxation of the velocity modulated population is :

\[ \Gamma_b^{(0)}(t₁₂) = Nσ_{el}(\bar{u}, θ₁₂) \bar{u}, A(β) \] (70)

where N is the perturber density, \( \bar{u} \) is the mean relative velocity, \( \bar{u} \) is the most probable relative velocity and θ₁₂ = (m + mₚ)/m₂ kᵣ t₁₂. A(β) is a kinematic factor depending upon the ratio β = m/m₂ of the masses of the active atom and of the perturber.

\[ A(β) = \int dv₂ A(v₂) (W(v₂))^2 / \int dv_v (W(v_v))^2. \] (71)

The expressions for A(v₂) and σ_{el}(\( \bar{u} \), θ₁₂) are given in appendix B of [5] :

\[ σ_{el}(\bar{u}, θ₁₂) = \int_{θ₁₂}^{π} \left( \frac{dσ_{el}}{dθ} \right) dθ = \pi (s - 1)^{1/2} \times \]

\[ \times \left[ \frac{1}{π²} \sin \left( \frac{π}{s - 1} \right) \Gamma \left( \frac{2}{s - 1} \right) \right]^{(s - 1)/2} \times \]

\[ \times \left( \frac{μ₂}{h} \right)^{(s - 2)/2} (θ₁₂)^{-2/s} × (σ_{el})^{(s - 1)/2} \] (72)

\( \Gamma_b^{(0)}(t₁₂) \) may be considered as the rate of collisions which occur at a scattering angle larger than θ₁₂. At this scattering angle in the center of mass frame the longitudinal velocity change of the active atoms equals the modulation period (k₁₂)⁻¹. Although the S.P.E. experiment cannot provide us with the differential cross-section, some angular information can be obtained by varying the lower bound θ₁₂ in the experiment (Eqs. (70), (72)).

For a Van der Waals long range potential (s = 6) equations (70)-(72) lead to a (t₁₂)⁻⁶ dependence for the relaxation rate \( \Gamma_b^{(0)}(t₁₂) \). We have plotted (Figs. 4 and 5) the measured values for collisions with Ar and He as a function of (t₁₂)⁻¹. As in our previous experiment on ytterbium [5], the measurements are found to be compatible with the hypothesis of a long range 1/r⁶ potential. The slope of the adjusted straight lines in figures 4 and 5 allows us to determine the elastic cross-sections, provided provision is made for the kinematic factor A(β) (Tab. II). In the case of He collisional partner and for the longest time delays t₁₂, one could possibly...
Fig. 4. - Elastic collision between Sr (5s 5p 3P₁) and argon. Vertical bars represent experimental data. The fitted calculated rate is represented by a straight line issuing from origin.

Fig. 5. - Elastic collisions between Sr (5s 5p 3P₁) and helium. Vertical bars represent experimental data. The fitted straight line corresponds to classical small angle scattering. The crosses correspond to the universal curve for light perturbers as discussed in section 5.2.

We have \( q(6) = 1.53 \) and \( C(6) = 2.06 \). Using equation (72) to describe the classical scattering region and assuming that equation (73) conveniently describes the shape of the differential cross-section in the diffractive region, one can derive an exponential representation for the differential cross-section in a 1/r³ potential [23, 24]:

\[
d\sigma/d\Omega = \varepsilon \left( \frac{K}{4\pi} \sigma_{el} \right)^2 q(s) \exp \left( \frac{C(s)}{8\pi} K^2 \sigma_{el} \theta^2 \right)
\]  

(73)

\( K = 2\pi \mu u/h \) is the wavevector associated with the active atom movement.

\[
q(s) = 1 + \left[ \tan(\pi/(s-1)) \right]^2
\]

(74)

\[
C(s) = (1/2\pi) \tan \left[ (\pi/s - 1) \right] \times \left[ \Gamma(2/(s-1)) \right]^2/\Gamma(4/(s-1)).
\]

(75)

We have \( q(6) = 1.53 \) and \( C(6) = 2.06 \). Using equation (72) to describe the classical scattering region and assuming that equation (73) conveniently describes the shape of the differential cross-section in the diffractive region, one can derive an exponential representation for the differential cross-section in a 1/r³ potential [23, 24].

We have \( q(6) = 1.53 \) and \( C(6) = 2.06 \). Using equation (72) to describe the classical scattering region and assuming that equation (73) conveniently describes the shape of the differential cross-section in the diffractive region, one can derive an exponential representation for the differential cross-section in a 1/r³ potential [23, 24]:

\[
d\sigma/d\Omega = \varepsilon \left( \frac{K}{4\pi} \sigma_{el} \right)^2 q(s) \exp \left( \frac{C(s)}{8\pi} K^2 \sigma_{el} \theta^2 \right)
\]  

(73)

\( K = 2\pi \mu u/h \) is the wavevector associated with the active atom movement.

\[
q(s) = 1 + \left[ \tan(\pi/(s-1)) \right]^2
\]

(74)

\[
C(s) = (1/2\pi) \tan \left[ (\pi/s - 1) \right] \times \left[ \Gamma(2/(s-1)) \right]^2/\Gamma(4/(s-1)).
\]

(75)

With this procedure, we can derive, for light perturbers (\( \beta^2 \ll 1 \)), an expression of the relaxation rate \( r_b(0)(t_{12}) \) which can be used both in the classical and in the diffractive small angle scattering regions (i.e. both for small and for large values of \( t_{12} \)). Using the variable

\[ y = k \Delta \omega^0 t_{12} = (2h\kappa/m) t_{12}/\sqrt{2\pi\sigma_{el} C(s)} \]

we get for \( (\Gamma^0_b(0)/N\sigma_{el} \overline{\nu_b}) \) a universal function which only depends upon the coefficient s of the potential [24]. This is illustrated in figure 5 for Sr*-He collisions. In the case of Yf*-He collisions, this universal shape was used to successfully reproduce both our SPE data (population elastic scattering, small \( t_{12} \) values) and those of Yodth et al. [3] (Zeeman coherence scattering, large \( t_{12} \) values) [19].

5.3 Depolarization in (5s 5p 3P₁)-Noble Gas Collisions. — SPE data provide us with \( \beta(t_{12}) = [\Gamma^0_b(t_{12}) - \Gamma^{(2)}_b(t_{12})] \) which, according to equations (63)-(65), involves the difference between the restitution terms for the velocity modulated alignment and population respectively. Our experimental procedure enables us to subtract the velocity changing effects concerning the total population (elastic scattering) and to isolate the specific velocity-changing effects associated with depolarizing collisions. In connection with the discussion on elastic scattering, \( \beta(t_{12}) \) may be considered as the rate of collisions which destroy atomic alignment while occurring at a scattering angle smaller than \( \theta_{12} \). The \( t_{12} = 0 \) limit for \( \beta(t_{12}) \) is nothing but the total rate for destruction of alignment \( \gamma^0_b \).

The available experimental results are, on the one hand, the values of \( \gamma^0_b \) (tab. I) and, on the other hand, a few values of \( \beta(t_{12}) \) corresponding to the small \( t_{12} \) region. As in the case of elastic scattering
we would like to compare our data with a calculated expression. This can be obtained along the following semi-classical picture for the depolarizing collisions. We consider that the trajectories are governed by the isotropic part of the potential $V^{(0)}$ as determined by elastic scattering data and we consider $k$-multipole decay probabilities along these trajectories [5, 24]. The latter probabilities are estimated for a $1/r^4$ anisotropic part of the potential in the frame of Anderson’s model for depolarizing collisions [25]. Defining a dimensionless variable $x$, proportional to $t_{12}$, one gets for $R(x) = [\Gamma_0^{(2)}(t_{12}) - \Gamma_0^{(0)}(t_{12})]/\gamma_0^{(2)}$ an universal expression which only depends upon $s$ and upon the measured quantities $\sigma_{el}$ and $\Sigma^{(2)}$ (see appendix C in [5]). The calculated curve $R(x)$ is compared with experimental data in figure 6. The curve corresponding to a hard sphere potential would be well below the SPE experimental points [5, 24].

Fig. 6. — Depolarization in Sr*-helium and Sr*-argon collisions. The dashed areas correspond to experimental data. The full line is the calculated « universal » curve of section 5.3.

5.4 CROSS-SECTION CALCULATION. — Within the above considered models, the total elastic cross-section $\sigma_{el}$ and the alignment destruction cross-section $\Sigma^{(2)}$ can be expressed as a function of $C^{(0)}$ and $C^{(2)}$ respectively [15, 23]. For a Van der Waals potential one gets:

$$\sigma_{el} = 8.083 \left( \frac{2 \pi}{\hbar} \right)^{25} C^{(0)}$$
$$\Sigma^{(2)} = 2.7 \left( \frac{2 \pi}{\hbar} \right)^{25} C^{(2)}$$

with $\bar{u} = (2kT/\mu)^{1/2}$.

The dipole-dipole effective potential between the active atom and the rare gas, ground state, perturber can be estimated from the values of the perturber polarizability and that of radial integrals for the active atoms [25]:

$$C^{(0)} = \alpha_B \left[ \langle r^2 \rangle_p + \langle r^2 \rangle_s - \frac{2}{3} \langle r^2 \rangle_{sp} \right]$$
$$C^{(2)} = \frac{\alpha_B}{2} \left[ \frac{3}{5} \langle r^2 \rangle_p - \langle r^2 \rangle_{sp} \right].$$

Using tabulated values for $\alpha_B$ [26] and radial integrals computed with a parametric central potential [27] one could derived the calculated values of table I and table II.

5.5 DISCUSSION. — As in the case of ytterbium [5], the data for population elastic scattering are compatible with our assumption on the potential shape. This is illustrated both by the $t_{12}$ dependence of $\Gamma_0^{(0)}(t_{12})$ (Figs. 4, 5) as well as by the comparison between calculated and experimental elastic cross-sections (Tab. II). Elastic cross-sections derived in a similar way for Yb* have been confirmed by independent measurements from other authors [3].

Concerning depolarizing collisions, the measured values of the cross-section for the $^3P_1$ level are very close to the corresponding values previously obtained by Alford et al. [28] for the $^1P_1$ (Tab. I). The calculated anisotropy is much smaller than the real one (Tab. I) as in the case of ytterbium. In addition, for strontium, the behaviour of $\beta(t_{12})$ does not seem to be compatible with our assumed anisotropic potential and/or with our depolarization model. This questions our initial assumption of a $1/r^4$ dependence for the anisotropic part of the potential. The calculated anisotropy due to polarization forces only is much too low. This suggests that important contributions of short range interaction to the depolarization process should be effective. More realistic potentials can be obtained from ab-initio calculations [22] but are not yet available for the strontium $^3P_1$ level.


We have discussed the formation and the relaxation of Stimulated Photon Echoes (SPE) produced by broadband light pulses in atomic vapor. The echo signal has been calculated in the weak field regime using an appropriate statistical model for the chaotic light field. The light pulses are characterized by their temporal envelope and by the autocorrelation function of the light field. The echo signal appears as the sum of a coherent and of an incoherent signal; the former one is absent when non correlated pulses are used. Both contributions to the time integrated signal are identical when $t_{12} \gg \tau_1$ and appear as the intensity summation of the elementary signal corresponding to the different velocity classes.

The relaxation of the SPE signal due to elastic collisions has been considered and a complete develop
opment of the corresponding propagator has been derived. A more tractable approximate expression for the relaxation rate has been obtained and used for the interpretation of the experimental results.

The corresponding experiment has been performed in strontium vapor using Nd-Yag pumped dye laser. Both elastic and depolarizing Sr*-noble gases collisions have been investigated and the corresponding cross-sections have been measured. The SPE data have been interpreted with a simple $1/r^6$ long range potential. Depolarization results gives evidence that a more realistic potential is needed to account for the anisotropy of the interaction.

References


