Letter

Collision-induced stimulated photon echo at the transition 0–1 in ytterbium: application to depolarizing collisions

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Abstract
A new idea based on the collision-induced stimulated photon echo in the presence of weak longitudinal magnetic field is applied to the depolarizing collisions research in a gaseous mixture of ytterbium vapour with xenon. Comparison of experimental data with theoretical prediction for the collision-induced stimulated photon echo in the weak magnetic field shows that the alignment decay rate of state 3P₁ in ¹⁷⁴Yb is higher than the orientation decay rate.

Keywords: collision induced stimulated photon echo, magnetic field

(Some figures may appear in colour only in the online journal)

1. Introduction

An atom may acquire the polarization momentum as a result of anisotropic excitation, for example by electric discharge, due to collisions with an electronic beam, due to the interaction with polarized resonant radiation. The last case is the most state-selective anisotropic excitation. A lot of theoretical studies have been made since 1950 [1–3], and many experimental studies have been carried out in atomic vapours with non-laser sources.

The polarization momentum of a given atomic state with angular momentum J can be interpreted as a non-equilibrium population distribution over magnetic sub-levels m (m = −J, −J + 1, ..., 0, J − 1, J). The zero order momentum corresponds to the integral non-equilibrium population of the level. The first order momentum is equivalent to the asymmetric distribution over magnetic sub-levels and it means that an atom in this state acquires the magnetic momentum. The second order polarization momentum can be presented as the symmetric relative m = 0 non-equilibrium population distribution and it corresponds to electric quadrupole momenta acquired by the atom. Such a series of polarization momenta, up to the (2J + 1)st order, is equivalent to precise knowledge of each magnetic sub-level non-equilibrium population created due to anisotropic action.

The collision-induced decay of atomic polarization momenta is of fundamental interest because these decay rates characterize inter-atomic forces.

Historically, the first method of atomic polarization moments observation was the detection of fluorescence polarization. The photon echo is a coherent spontaneous radiation arising as a response to the action of two or more exciting radiation pulses and it is related to the reversal of inhomogeneous relaxation. Its modification, the stimulated photon echo (SPE), is generated in a gas under the action of three resonant radiation pulses separated by time delays τ₁ and τ₂. The simplified picture of SPE generation looks as follows. The first exciting pulse creates the non-equilibrium polarization in the low pressure gas which spectral line has inhomogeneous Doppler broadening. The second pulse converts this
polarization into non-equilibrium populations of magnetic sub-levels of the excited state. The populations ‘store’, during time delay $\tau_3$, the information about the interaction of an atom with two pulses, until the third pulse arrives. The third pulse converts non-equilibrium magnetic sub-level populations again into polarizations, makes a reversal of Doppler phases and, therefore, creates the conditions for SPE generation at the time moment which is approximately equal to $\tau_{spe} = \tau_3 + 2\tau_2$. During time interval $\tau_3$ the atoms are insensitive to any changes of Doppler phase. Hence, they are insensitive to velocity-changing collisions. On the other hand, they are extremely sensitive to elastic collisions intermixing magnetic sub-levels, in other words to depolarizing collisions. That is why SPE is a very convenient method for studying depolarizing collisions. Moreover, under certain experimental conditions, the SPE amplitude depends only on the three lower polarization moments: population $\gamma^{(0)}$, orientation $\gamma^{(1)}$ and alignment $\gamma^{(2)}$ decay rates.

The atomic state with low degeneracy (small angular momentum) is especially attractive for a comparison between theoretical predictions and experimental data; besides, the polarization momentum of excited level in this case has its direct physical meaning. That is why the experimental research in atomic vapour for transitions with angular momenta $J = 0$, $J = 1$ are numerous (see [4] and references therein). The stimulated photon echo (SPE) is very sensitive to depolarizing collisions, and the idea of how to measure low polarization moments decay rates at an optically allowed transition was formulated many years ago [5].

The most difficult problem for experimental research is to measure the difference between alignment and orientation decay rates because this difference is small and, therefore, such measurement requires high sensitivity and some special technique.

The reliably stated difference between orientation $\gamma^{(1)}$ and alignment $\gamma^{(2)}$ decay rates of excited state ($6s6p)^3P_1$ of $^{174}$Yb, and estimate for magnitude $|\gamma^{(0)} - \gamma^{(2)}|$ were made in our experimental work [6] for Yb + Kr collisions according to the ideas [5].

As it follows from the theoretical analysis of SPE induced by collisions of active atoms with buffer atoms, this phenomenon appears exclusively due to the difference between decay rates of alignment and orientation [7]. So, it seems natural to use just the collision-induced SPE for a depolarizing collisions research.

Here, a new approach proposed in [8] and allowing to find unambiguously the sign of difference between decay rates of orientation and alignment is applied to the depolarizing collisions research.

2. Experimental technique

The resonant radiation for coherent responses generation was provided by three Rhodamine 110 lasers pumped by the second harmonic of Nd$^{3+}$:YAG lasers. The details of dye lasers construction can be found in [9, 10]. Each dye laser was arranged by fine frequency tuning. In this experiment, all three pulses of exciting radiation operated at the wavelength resonant to inter-combination transition ($6s^2)^1S_0$–($6s6p)^3P_1$ of $^{174}$Yb. Two first radiation pulses were of 10 ns duration, the third one was of 5 ns duration. The line width of the third exciting pulse (HWHM) was about 300 MHz. The radiation of the first two pulses consisted of two or three longitudinal modes separated by $\sim 1.5$ GHz (cavity length of the master oscillator in each dye laser was about 10 cm). An average over 100–120 pulse realizations produced an estimate for the line width of about 2 GHz for the first and second pulses.

To make the most homogeneous transverse distribution for each beam, we used the Kepler telescope at the output of the dye laser followed by the diaphragm. After that, the set of flat dielectric mirrors directed all three exciting pulses into the working cell at a small angle (not over $2 \times 10^{-3}$ rad) to each other along the edges of a trihedral pyramid (such geometry guaranteed the spacial separation of the collision induced SPE signal from the exciting pulses to avoid the photomultiplier bleaching). After the reflection from these mirrors, the beams were expanded till the diameter of about 20–30 mm and then passed through the diaphragm (10 mm in diameter), limiting the radial size of the beams and cutting their central parts.

The exciting radiation absorption by the gas mixture in the working cell was less than 10% so, we could consider the sample optical density as low.

The influence of the exciting pulses power fluctuations was minimized by discarding those pulses which fluctuations exceeded 10% of the average power. Each experimental point was obtained as a result of averaging over 100–120 realizations.

In our experiment, only a small part of the Doppler contour of $^{174}$Yb was excited. Therefore, it was necessary to increase all the exciting beams intensity and to fit each beam intensity to optimize the collision-induced SPE signal. Usually, this signal is several orders of magnitude less than the conventional SPE which is remarkably lower than the exciting pulses intensity.

At the working temperature of the vacuum cell filled with Yb vapour and the buffer gas of about 800 K, the first two pulses spectra covered the whole Doppler contour of $^{174}$Yb, practically without the excitation of odd Yb isotopes [11]. The narrower spectral width of the third pulse (without which SPE can not be generated) provided even more guarantee that only $^{174}$Yb atoms were involved into coherent response generation.

3. Results and comparison with theory

The collision induced SPE was generated by three exciting pulses with linear specially chosen polarizations. Each exciting pulse polarization was linear with the accuracy not less than $10^{-3}$ in power, and their relative orientation was also chosen optimal for the collision-induced SPE generation without the background of conventional SPE: the 1st pulse polarization was crossed with other two exciting pulses linear polarizations (the above-mentioned sequence of three exciting polarizations can not generate a conventional SPE in pure Yb vapour).
This collision-induced SPE in zero magnetic field is polarized as the first exciting pulse [7]. Therefore, just the component of collision induced SPE that coincides with the first pulse polarization was measured in this experiment as a function of the longitudinal magnetic field. This dependence is shown in figure 1; the vertical bars shown for three points indicate the measurement errors. The initial part of the curve in figure 1 for the weak magnetic field (not over 0.5 G) shows clearly the ascending behaviour and, according to the predictions of [8], it demonstrates the domination of alignment decay rate \( \gamma(2) \) over the orientation decay rate \( \gamma(1) \) for state \( ^3P_1 \) of \(^{172}\)Yb. The time delays between the exciting pulses were chosen as \( \tau_2 = \tau_3 = 30 \) ns. The pressure of active atoms was 30 mTorr, and the Xe buffer pressure of 195 mTorr was close to the optimal buffer pressure for the collision-induced SPE generation [7].

The dependence of the collision induced SPE intensity (the polarization component collinear with the first exciting pulse polarization) versus the external longitudinal magnetic field is determined by the factor [8]:

\[
I(\varepsilon) = \cos^4(\varepsilon \tau_2)|1 - e^{-(\gamma(1) - \gamma(2) \tau_2)} \cos(2\varepsilon \tau_2)|^2 \tag{1}
\]

where

\[
\varepsilon = \frac{g_\mu_B B}{\hbar},
\]

\( \mu_B \) is the Bohr magneton, \( g_\mu = 1.5 \) is the g-factor of the upper level, \( B \) is the magnetic field induction, and \( \hbar \) is the Plank constant. This dependence is sensitive to the sign of difference \( \gamma(1) - \gamma(2) \). If \( \gamma(1) > \gamma(2) \) then the echo intensity is expected to decrease in the weak magnetic field, while, in the alternative case \( \gamma(2) > \gamma(1) \), it is expected to increase.

It should be mentioned, however, that equation (1) is valid under two conditions. First, the spectral line must be narrow for the exciting pulses, i.e. \( k u T \ll 1, k = 2\pi /\lambda, \lambda \) is the wavelength of the resonant transition, \( u \) is the atomic thermal velocity, \( T \) is the pulse duration. Second, the areas \( \theta_n \) of the exciting pulses must be as follows:

\[
\theta_1 = \pi/2(2n_1 - 1), \quad \theta_2 = \pi(2n_2 - 1), \quad \theta_3 = \pi(2n_3 - 1) \tag{2}
\]

where \( n_1, n_2, n_3 \) are integer numbers.

In the present experiment, \( k u T \approx 16 \) for the 5 ns pulse duration; so, the spectral line appears to be broad for all the exciting pulses. The optimal areas of the exciting pulses were estimated as \((10 \div 11)\pi\). With such large values of the areas, the narrow spectral line approximation may be good enough due to the suppression of the Doppler dephasing during the pulse by its strong driving field. Though we cannot attribute the accurate value to each of the exciting pulse, the exciting pulses power were varied around the above mentioned \((10 \div 11)\pi\) to get the best signal intensity. Since the maximum echo intensity, in the case of the collision-induced SPE, is obtained at the values defined by (2), we may assume that the condition (2) is satisfied. With all above-said in mind, we apply formula (1) to describe the experimental curve presented in figure 1. The ascending of this curve in the weak magnetic field indicates unambiguously that \( \gamma(2) > \gamma(1) \).

According to experimental curve—\( I_{in}/I_0 = 10 \), where \( I_{in} \) is the echo intensity at its first maximum, while \( I_0 \) is the echo intensity in the zero magnetic field. So, we may evaluate the magnitude of difference \( \gamma(2) - \gamma(1) \) in order to fit this ratio \( I_{in}/I_0 = 10 \) in the theoretical curve described by (1). It appears to be: \( \gamma(2) - \gamma(1) = 5.77 \times 10^6 \) s\(^{-1}\). The theoretical curve, determined by (1) and normalized in such a way that both theoretical and experimental intensity values coincide in the zero magnetic field, is presented in figure 2.

4. Discussion

There is an evident difference between figures 1 and 2 for the ‘strong’ magnetic field area (over 5 G). The theoretical curve in figure 2 shows a regular oscillation in magnetic field from zero to maximum, while the experimental curve never comes to zero. We have already obtained such kind of behaviour both for the photon echo [12] and for the stimulated photon echo [13] for the ‘strong’ magnetic field (over 5 G up to 40 G) both leading to the generation of ‘unpolarized’ echo [14]. Such discrepancy between the theoretical and experimental curves may be attributed to the fact that, in theoretical formula (1), the action of the magnetic field during the excitation pulse was neglected. However, in the magnetic field \( B > 5 \) G and for pulse duration \( T = 10 \) ns, polarization rotation angle \( \varepsilon T > 0.2\pi \) is large enough to be neglected. The theoretical dependence (1) of the echo intensity on the magnetic field induction is a periodic function with a single period determined by time interval \( \tau_2 = \tau_3 \) while, in reality, such dependence contains some superposition of oscillations with a number of various periods determined not only by \( \tau_2 \), but also by its combinations with pulse durations \( T \). So, less regular oscillations in figure 1 are not surprising. On the other hand, the calculations in [8] accounted for depolarizing collisions—the most important factor for the collision-induced photon
In fact, the difference is small, as compared to the atomic transition $T_{\text{O}}$ and as a result of active atoms 1 and 29.59 $10^2$ Torr and the measurement of the magnitude this difference as a function of the buffer gas pressure (Kr was used as a buffer in [6]). This work shows a principal possibility to find the sign of this difference.

The value $(\gamma^{(2)} - \gamma^{(1)})/p_{\text{Xe}} = (29.59) \cdot 10^6$ s$^{-1}$. Torr$^{-1}$ obtained from the estimate in section 3 of this work and Xe pressure seems reasonable in comparison with the magnitude of polarization moment decay rate difference $|\gamma^{(2)} - \gamma^{(1)}|/p_{\text{Kr}} = (6.8 \pm 0.6) \cdot 10^6$ s$^{-1}$. Torr$^{-1}$ measured in [6] if we keep in mind that $d\gamma^{(i)}/dp$ increases usually with the mass of the buffer (see, e.g. [4]).

It is worth noting that we used the theoretical formula obtained under the conditions of the narrow spectral line, while our experiment was performed on a broad spectral line; so, we do not expect a perfect quantitative agreement, but rather a qualitative agreement and rather rough estimations of the difference in the alignment and orientation relaxation rates of the upper level.

There is a possibility [5] to get orientation and alignment decay rates separately, at least for atomic transition 0–1 and, for this purpose, it is necessary to analyze SPE as a function of time delay $\tau_{23}$ between the second and the third exciting pulses for specially chosen angles $\psi_1$ and $\psi_2$ made by polarization vectors of the first and of the second exciting pulses with the polarization vector of the third exciting pulse. Such experimental set up is now in preparation in our laboratory. It should be stressed, however, that the measurement of $\gamma^{(2)}$ and $\gamma^{(1)}$ separately should be made accurately as high as possible. The high measurement accuracy was reached just in our experiments on collision-induced phenomena: for the collision-induced two-pulse photon echo generated due to the velocity dependent relaxation (relaxation anisotropy) [12] and the magnetic field effect on this signal [9], and for the collision-induced SPE [7] generated at atomic transition 0–1 exclusively because of the difference in $\gamma^{(2)}$ and $\gamma^{(1)}$ as a result of active atoms collisions with the buffer atoms. The technique proposed in a given work may prove advantageous due to sensitivity to the difference between two quantities of a comparable magnitude.

5. Summary

The method proposed in [8], based on the collision-induced stimulated photon echo behaviour in the weak magnetic field, proved reliable and a relatively simple way to find the sign of the difference between $\gamma^{(2)}$ and $\gamma^{(1)}$ for atomic transition 0–1. Moreover, the comparison between the theoretical and experimental curves gives a reasonable estimate for the magnitude of this difference. In this experiment in the Yb + Xe mixture, from the comparison of the theoretical prediction with the experimental data on the collision-induced SPE in the weak magnetic field, we found that the alignment decay rate of state $^{3}P_1$ in $^{174}$Yb is higher than the orientation decay rate.

Acknowledgments

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