Photoionization of polarized doublet states of Xenon atom

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Abstract. We studied experimentally and theoretically the ionization of a two-photon excited superposition of 4f states of Xe atom by probe femtosecond pulse laser in a supersonic beam. The revealed oscillation structure in the observed photoionization signals is associated with the interference of the coherently populated components of Xe doublet states. Our results demonstrate the high efficiency of proposed experimental scheme for registering quantum beats in the photoelectron current.

1. Introduction
Analyzing of fluorescence signals after exciting atomic states by polarized light is an important brunch of modern spectroscopy [1]. Observation of magnetic resonances in polarized gas media is the basis of construction and realization of sensitive magnetometers [2]. Diverse types of oscillating structures in detected signals in the presence of magnetic fields allow making precision measures of atomic state parameters beyond limitations due to Doppler broadening [3]. Traditional methods of spectroscopy are associated mainly with bound-bound optical transitions. In the present report, we demonstrate a possibility to examine photocurrent oscillations which occurs due to the interference between coherently excited components of doublet atomic states upon their photoionization by probe polarized photons.

2. Experimental setup
The supersonic beam of Xe atoms crosses a focused laser beam in the presence of a magnetic field, it is necessary to draw the electrons from the ionization region (figure 1). We use a "pump-probe" experiment with two-photon excitation of Xe atoms to nearby states $5p^5(^3P_{3/2})4f[3/2]_2$ and $5p^5(^3P_{3/2})4f[5/2]_2$ using a "pump" pulse. Excitation is performed by a horizontally polarized femtosecond laser pulse at a wavelength of about 220nm. Photoionization of the excited atoms in the $J=2$ states is performed by a “probe” pulse of a horizontally or circularly polarized Ti: sapphire femtosecond laser at a wavelength of 795nm, the spectral width of the ionization pulse is greater than the distance between these 4f levels.

An electron spectrometer of the “magnetic bottle” type was used to record the electron energy spectra. Analysis of the energy spectra of electrons, obtained in different delay times of the probe
pulse relative to the pump pulse, made it possible to obtain data on quantum beats. The energy spectra provide information on the levels from which ionization took place and on the intensity of ionization from these levels.

The Ti-Sapphire femtosecond laser PULSAR 10 (Amplitude Technologies, France) has the following characteristics: main wavelength - 790nm, pulse energy - 5mJ, pulse duration - 50fs, repetition rate - 10Hz. The resulting pulse was divided into two parts: (i) 70% of the pulse power pumped parametric amplifier TOPAS and was converted to UV radiation for two-photon excitation of atomic states. (ii) The rest 30% of the power was used as probe radiation in the form of the first harmonic for ionization of pump-excited atoms.

An optical system was used to control the delay time between pulses. The delay time between laser pulses was varied from 0 to 24ps. With a change in the delay time $t$, periodic oscillations of the intensity of the photoelectron current appear at the difference frequency of the levels $\Delta \omega$ (figure 3). Which equal to the frequency distance between levels $\Delta \omega=\omega_{5/2}-\omega_{3/2}$. From the experimental data shown in figure 3, we have a beat period of 0.549ps. This is in good agreement with the energy gap between these xenon levels ($Xe_{6s}$), which corresponds to a beat period of 0.55ps.

**Figure 1.** Experimental setup: OPA – optical parametric amplifier; DL – delay line; DM – dichroic mirrors; CM – concave mirror.

**Figure 2.** Researched Xe levels.

**Figure 3.** Temporal dependence of the photocurrent $I$ for one-photon ionization. The one-photon ionization occurs together with two-photon ionization due to the strong probe pulse.

3. **Theoretical analysis**

The effects of the coherent interaction between polarized quantum particles and electromagnetic fields are convenient to analyze in the frame of the irreducible tensor operators technique which deals with atomic and photon polarization moments $\rho^\chi$, $\Phi^\chi$ [2,4]. In this section, we focus on a brief overview
of our theoretical study of how the interference of the doublet components of the Xe states results in a beating structure (see figure 3) in photoionization signals upon photoionization stimulated by a single photon (single-photon photoionization).

3.1. Partial photoionization signals
The authors of the book [2] obtained an explicit expression for the photoionization cross section (Equation 2.3-2.5) for a polarized degenerate quantum J-state with Zeeman sublevels \( M = -J, ..., J \). In our recent work [5], we applied the formalism from [2] for the case when external magnetic fields split the corresponding Zeeman M-components. A fairly direct reformulation of the previous results makes it possible to describe the photosignal \( I(t) \) upon photoionization of the studied doublet state \( \Lambda = \{\Lambda_1, \Lambda_2\} \) of Xe, consisting of two sub-states (see figure 2)

\[
\Lambda_i = \frac{5s^2 5P^2(\gamma P)}{2\Pi_i} n[K_i] , \quad i = 1, 2 \quad K_1 = 3/2; K_2 = 5/2
\]

with the same total atomic orbital moment \( J \) (j-coupling scheme [4]). The corresponding photo signal looks like this:

\[
I_{\Lambda,\Lambda'}(t) / J_{ion} = |C_1|^2 \Pi_{11} + |C_2|^2 \Pi_{22} + 2Re(C_1^* C_2 \Pi_{12} \exp(i\Delta\omega)),
\]

\[\Pi_{ij} = \sum_{\nu=0}^{2} D_{\nu}^\mu M_{\mu}^\nu \Phi_{\nu}^\mu \rho_{\nu}^\nu \cdot \]

Here \( J_{ion} \) is the intensity of the luminous flux of the ionizing laser. The indexes in \( I(t) \) indicate the quantum numbers \( \Lambda = \{\Lambda_1, \Lambda_2\} \) of the polarized bound doublet atomic state in the initial channel and \( \Lambda' = \frac{3s^2 3P^2(\gamma P)}{2\Pi} e^\epsilon [K']_p \) that’s in the final (ionization) channel with the notation \( e^\epsilon \) for the free photoelectron energy. It is assumed that the quantum numbers \((P, \ell)\) of atomic core remain unchanged (the absence of intercombinaion transitions).

The first two terms on the right-hand side of equation (2) determine the contributions to the ionization of the doublet \( \ell \)-sub-states, while the third term is responsible for the effect of their interference. Coefficients \( C_i \) correspond to sub-states \( \Lambda_i \) probability amplitudes normalized to the total sub-states population over all their Zeeman sublevels: \( n_i = |C_i|^2 = \sum_{\nu=0}^{2} |C_{i\nu}|^2 \). It is important to note that although the amplitudes \( C_i \) may differ, the atomic polarization moments \( \rho_{\nu}^\nu \) with \( \kappa = 0 \) (population), \( \kappa = 1 \) (orientation) and \( \kappa = 2 \) (alignment) must be the same, since both components \( \Lambda_i \) of the doublet have the same quantum number \( J \) and they are excited with the same pump photons. The presence of the atomic moments \( \rho_{\nu=0}^\nu \) in equation (3) is due to the pure polarization (linear or circular) of the probe laser that implies that the corresponding photon moments \( \Phi_{\nu}^\nu \equiv 0 \) if \( \nu \neq 0 \) [2].

The factors \( M_{\mu}^\nu, D_{\nu}^\mu \) take into account the influence of the mutual orientation of the orbital moments of atoms and ionizing photons on the photoionization processes efficiency. The value of \( M_{\mu}^\nu \) turns out to be equal to one (\( M_{\mu}^\nu = 1 \)) while factors \( D_{\nu}^\mu \) are expressed through 6j-symbols

\[
D_{\nu}^\mu = (-1)^{I+J+1} \begin{bmatrix} I & 1 & \kappa \\ J & J & J' \end{bmatrix} (\Lambda_l || d || \Lambda')(\Lambda_l || d || \Lambda')
\]

and the reduced dipole matrix elements for bound-free optical transitions [2,4].

Expressions (2)-(4) provide information on the partial photoionization cross-section, when the quantum numbers \((P, \ell, K', J')\) in the final channel turn out to be fixed (i.e. measured in an experiment).
3.2. Aggregate photoionization signal

In our experiment, we do not separate photoelectrons over its quantum numbers \( l', K', J' \). In other words, all partial signals contribute to the final aggregate photocurrent signal \( I(t) = \sum_{\lambda \lambda'} I_{\lambda \lambda'}(t) \).

Importantly, the sum rule (see in [4] equation (4.85)) for the product of three 6j-symbols entering the factor \( D^x \) (4) makes it possible to perform explicit summation: \( I(t) = \sum_{\lambda \lambda'} |(nl \parallel d \parallel e'')|^2 I_{\lambda \lambda'} \), where the remaining partial signals \( I_{\lambda \lambda'} \) relate to the output of a free photoelectron with two possible values \( l' = l \pm 1 \) of its orbital quantum number. The ratio \( I_{\lambda'} / I_{\lambda \lambda} \) is expressed through a formula similar to equation (1) with a somewhat modified coefficient

\[
D^x = \begin{bmatrix} 1 & 1 & \kappa \\ l & l & l' \end{bmatrix}
\]

and a nontrivial factor

\[
M^x_{\lambda \lambda'} = (2J + 1)\sqrt{(2K + 1)} \sqrt{(2K' + 1)} \begin{bmatrix} K_i & K_i & \kappa \\ J & J & 1/2 \end{bmatrix} \begin{bmatrix} 1 & 1 & \kappa \\ K_i & K_i & j \end{bmatrix} (-1)^\Phi
\]

with a phase \( \Phi = K_j + K_i - 0.5 + J - l - j \).

3.3. Results

We have derived that the observed photocurrent signal \( I(t) \) during single-photon photoionization of an atomic doublet state with polarized components is calculated as a sum

\[
I(t) = \sum_{\lambda \lambda'} \chi_{\lambda'} I_{\lambda \lambda'}(t); \quad \chi_{\lambda'} = (nl \parallel d \parallel e')^2
\]

is calculated as a sum of two partial signals \( I_{\lambda'}(t) \)

\[
I_{\lambda'}(t) / \tilde{Y} = C_1 C_1^\dagger \Pi_{11} + C_2 C_2^\dagger \Pi_{22} + 2 \text{Re}(C_1 C_2^\dagger \Pi_{12} \exp(i\Delta \omega)),
\]

whose statistical weights \( \chi_{\lambda'} \) are given by the reduced dipole matrix elements (7) of bound-free optical transitions of the valence electron. The multiplier \( \tilde{Y} \) depends on intensities of applied lasers and experimental set-up, while the probability amplitudes \( C_i \) of the initial doublet sub-states (1) depend on the parameters of the pump laser, in particular, on its polarization. The factors \( \Pi_{\lambda \lambda'} \) (3) are expressed via 6j-symbols as it is seen from equations (5), (6) and via the polarization moments connected with Wigner 3j symbols [2]

\[
\Phi_0^x = \sqrt{2\kappa + 1}(-1)^{m} \begin{bmatrix} 1 & 1 & \kappa \\ m & m & 0 \end{bmatrix}; \quad \rho_0^x = \sqrt{2\kappa + 1}(-1)^{J-M} \begin{bmatrix} J & J & \kappa \\ M & M & 0 \end{bmatrix}
\]

which values are presented in table 1.

It is noteworthy that our theoretical results can be extended to the case of two-photon ionization of polarized atomic states. The corresponding formulas are similar to expressions (7), (8), however the factors \( \Pi_{\lambda \lambda'} \) (3) contains five atomic polarization moments \( \rho_0^x \) with \( \kappa = 0,...,4 \) and 9j Wigner symbols. Taking into account the presence of a control external magnetic field is reduced to introducing the time dependence of the moments \( \rho_0^x \), as described in detail in [5].

4. Conclusion

The polarization atomic moments (PAM) strongly affect the magnitude of the long-range dipole-dipole interaction between the excited states of the atoms [6]. This interaction plays an important role
in modern applied physical problems associated with quantum computing based on an ensemble of cold atoms in magneto-optical traps [7]. For this reason, real-time control and diagnostics of PAM are an urgent problem in the implementation of entangled atomic states (Q-bits) and in the performance of basic quantum operations [8]. Traditional optical diagnostics of PAM is associated with photon-stimulated bound-bound transitions and registration of accompanying absorption [9] or fluorescence [2, 3] spectra. The use of bound-free transitions for diagnostic purposes has a number of advantages due to almost 100% registration of photoelectrons, which significantly increases the possibilities of recording subtle physical effects [10, 11]. The presented here and in our previous work [5] results on the study and description of the oscillatory structure of the photocurrent with their subsequent spectral analysis make it possible to find PAM necessary for calculating the dipole-dipole interaction between excited atoms in cold media.

Table 1. Photon $\Phi_0^\kappa$ and $\rho_0^\kappa$ atomic polarization moments. $\Phi_0^\kappa$-moments are shown for circular ($m=1$) and linear polarizations ($m=0$) of the probe laser, while $\rho_0^\kappa$-moments corresponds to the excitation states with $J=2$ and Zeeman component $M=2$ [2].

| $\kappa$ | $\Phi_0^\kappa$; linear | $\Phi_0^\kappa$; circular | $\rho_0^\kappa$ |
|---------|-------------------------|---------------------------|-----------------|
| 0       | 23.56                   | 34.64                     | 23.76           |
| 1       | 0                       | 1/2                       | $\sqrt{2}/5$   |
| 2       | $-\sqrt{2}/3$           | 1/6                       | $\sqrt{2}/7$   |

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