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Modulation of quantum beats signal upon photoionization of Xe isotopes in the magnetic field

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Photoionization processes involving polarized Xe atoms in the presence of an external magnetic field were analyzed in our previous studies. It was shown that the modulation of quantum beats in the observed electronic photo signals could not be explained by the Paschen-Back effect accounting for fine-structure levels only. In the current work, ionized Xe^+ isotopes are used as the output channel of photoionization. By comparing the temporal partial signals in the mass spectra of the Xe^+ isotopes, it was possible to explain the modulation of quantum beats in ion fluxes as a consequence of the presence of the hyperfine structure in the Zeeman components of the ^{129}Xe and ^{131}Xe isotopes.

Keywords: quantum beats, hyperfine structure, Zeeman effect, two-photon excitation, femtosecond ionization, „pump-probe"-experiment.

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Introduction

Studying of fluorescence after exciting atoms by polarized light allows considerable widening of both the capabilities of optical diagnostics methods for gaseous media [1] and their scope of application in various applied tasks [2]. However, instead of analyzing fluorescence signals sometimes it can be more preferable to study the photocurrent signals caused by the processes of excited states photoionization. The possibility of close to 100percent efficiency of photoelectrons recording ensures stronger response signals generated under photoionization of polarized atoms [3], thus it opens up an opportunity of observing very fine physical effects caused by alignment or/and orienting [3,4] of gaseous media due to, for example, optical pumping [5]. In this study the „pump-probe" method is used to investigate the $5p^5(^2P_{3/2})6p[3/2]_2$ ($79212.5\,\mathrm{cm}^{-1}$) state of Xe isotopes in a supersonic beam. This method was used in our previous works. Full description of the experimental setup used in these studies, including the „pump-probe"method, is given in [6]. Previously, with a similar experiment scheme, we observed quantum beat signal modulation under photoionization of Xe [7] and showed that the nature of this modulation can not be caused by the quadratic Paschen-Back effect. This method (for external magnetic field levels of an order of magnitude of 1 T) proves to be too weak on the background of such splitting of quantum states [7]. Here we demonstrate experimentally that the modulation of quantum beats signals is only observed for odd Xe isotopes and caused by the hyperfine structure of excited levels.

Photoionization of atomic states as an element of the optical diagnostics

It is important to note that in the common approach to study the ionization processes the observed photocurrent $I(t, \theta)$ is investigated as a function of angle θ between the laser beam and outgoing photoelectrons. The first study [8] that investigated the interference of e hyperfine structure of Sodium on the basis of the pattern of angular distribution of electrons was performed in 1978. A bit later authors of [9] were succeeded to record the effects of quantum beats in the angular distribution of electrons at one-photon coherent excitation of hyperfine Sodium structure levels followed by one-photon ionization with a test pulse. In [10] published in 1983 optical characteristics of Lithium are determined from the records of angular distribution of photoelectrons: the ratio of the radial matrix elements from $6p$ excited states to $l = 0$ and $l = 2$ final states, as well as phase shifts of continuum electrons.

Quantum beats in the integral intensity of photocurrent under photoionization of fine structure levels of Sodium were reported in 1979, by Leuchs and Walther [11] for highly excited ^2D-states of Sodium, with a statement of the identified dependence of beats contrast on the mutual orientation of linear polarization of two exciting pulses of the laser. In later years, a number of studies were published to investigate quantum beats in signals of photoionization of highly excited Rydberg states. An example can be the work of Wolde et al. published in 1988 [12], where

an excitation followed by ionization of Rubidium atoms by picosecond laser is implemented. Features of the observed beats in photocurrents were explained within a quasi-classic approximation with wave functions simulated as wave packets rotating around an atomic core.

The emergence of femtosecond and attosecond lasers has stimulated further interest of researchers in the studying of quantum beats physics. In one of earlier works with the use of femtosecond laser [13] a resonance excitation followed by ionization of Sodium molecules was reported. The authors succeeded to create a coherent superposition of vibrating states of the molecule and observe a manifestation of quantum interference of these states in the photocurrent of molecular ions. In a later publication in 2000 [14], the use of femtosecond laser allowed observing quantum beats with a period of 580 fs for a fine structure of $4p$-state of Potassium.

In the publication in 2010 by Mauritsson [15], a novel technique was proposed to study the interference processes of bound states of Helium atom. The ground state of Helium was excited by an attosecond pulse with a wavelength that corresponds to the ionization energy of Helium atom. Attosecond pulses have abnormally high spectral width due to energy-time uncertainty relation $\Delta t \cdot \Delta E \geq \hbar$. This allows simultaneously coherently populate both bound states and continuum states of a Helium atom. Further ionization of bound Rydberg states by an infrared femtosecond pulse synchronized with an attosecond pulse results in an interference between the primary and secondary excited continuum states, which is manifested in the form of quantum beats in time curves of the energy spectrum of photoelectrons.

In the publication in 2018 by Forbes et al. [16], it is proposed to investigate the effects of atomic states interference within the quantum beat spectroscopy (QBS) technique, where the beats themselves are observed in a photoelectronic image. The rationale for applicability of the QBS technique to studying the fine splitting in atoms of noble gases and atoms of alkali metals is based on the coincidence of nanosecond and picosecond time scales of the experiment with lifetimes and inverse frequency intervals typical for the fine structure. In this study we have built photoelectron images, so called photoelectron angular distribution (PAD) images for various time delays. The obtained experimental data was used to derive various constants of fine and hyperfine splitting, magnetic dipole constant A and electric quadrupole constant B for the $(^2P^0_{1/2})6s^2[1/2]_0$ state of Xenon.

In our previous works we reported about the oscillations of the electron photocurrent, which are caused by the interference of Zeeman components of $(^2P_{3/2})6p[3/2]_2$ quantum sublevel in the fine structure of Xenon. In this study we are focused on the ionic photocurrent oscillations resulted from coherent excitation of Zeeman sublevels of hyperfine structure of $(^2P_{3/2})6p[3/2]_2$ isotopes of Xenon.

Experimental setup

A beam of monokinetic Xenon atoms with natural isotope composition is generated in a supersonic jet. These atoms are two-photon excited to the $5p^5(^2P_{3/2})6p[3/2]_2$ ($79212.5\,\mathrm{cm}^{-1}$) state by a short radiation pulse ($\tau = 60$ fs, $\lambda = 252.5$ nm). The direction of magnetic moments ($J = 2$) of excited atoms is defined by polarization of the pumping radiation. The excitation takes place in the interaction zone of the electronic time-of-flight spectrometer of „magnetic bottle“type, i. e. in a vertical magnetic field ~ 0.8 T. From the moment of excitation the magnetic moment precession of excited atoms starts in the magnetic field **B** with a circular frequency of $\omega_L = g\gamma_0\mathbf{B}$, where g is Lande factor, and $\gamma_0 = 0.88 \cdot 10^{11}\,\mathrm{rad{\cdot}s^{-1}T^{-1}}$. The excited atoms are two-photon ionized by a radiation pulse ($\tau = 50$ fs, $\lambda = 795$ nm) as well, with the use of an adjustable delay. The electronic spectrometer records electrons with an energy of $6536\,\mathrm{cm}^{-1}$, that correspond to the two-photon ionization of excited atoms, as well as electrons with an energy of $20985\,\mathrm{cm}^{-1}$, that correspond to the three-photon ionization of Xenon atoms by the pumping radiation. These electronic peaks have nearly equal intensity. On the curve of excited atoms? ionization signal as a function of delay quantum beats are observed, which are related to the Zeeman splitting of level in magnetic field. A modulation is observed on this periodic structure, and the following experiment was carried out to find out the nature of this modulation. Instead of recording electrons, Xe^+ positive ions were recorded. They were pushed out by the electric field from the interaction zone into the short time-of-flight mass spectrometer, that allowed distinguishing of Xenon isotopes. The magnetic field was not switched off at the same time. The mass-spectrum as a function of delay is shown in Fig. 1, *a*. It is worth to note that the ionic signal is a sum of both ionization processes, therefore the depth of quantum beats signal modulation is lower than that of electron registration.

In this experiment a horizontal polarization of radiation pulses was used, and quantum beats took place at a double Larmor frequency. For the $5p^5(^2P_{3/2})6p[3/2]_2$ state of Xenon the quantum beats frequency is $\nu_b = 2\omega_L/2\pi \approx 32\,\mathrm{ns}^{-1}$ (period $T_b = 1/\nu_b \approx 31.42$ ps) (Fig. 1) [17].

No quantum beats signal modulation (QBSM) of Xe isotopes is observed for isotopes with even masses of 132, 134, 136. In contrast to the isotopes with odd masses, isotopes with even masses have unsplit frequency component of the quantum beat of Fourier spectrum (Fig. 1, *b*). Fourier spectra of partial signals from each isotope allow determining the circular frequency of quantum beats ω_b together with circular frequencies of their modulation $\omega_m = 2\pi\Delta\upsilon$ for different isotopes of Xenon (Fig. 1, *b*). Clearly observed modulations of quantum beat signals are attributed to odd isotopes ^{129}Xe and ^{131}Xe. Cores of these odd isotopes have magnetic moments of 1/2 and 3/2,

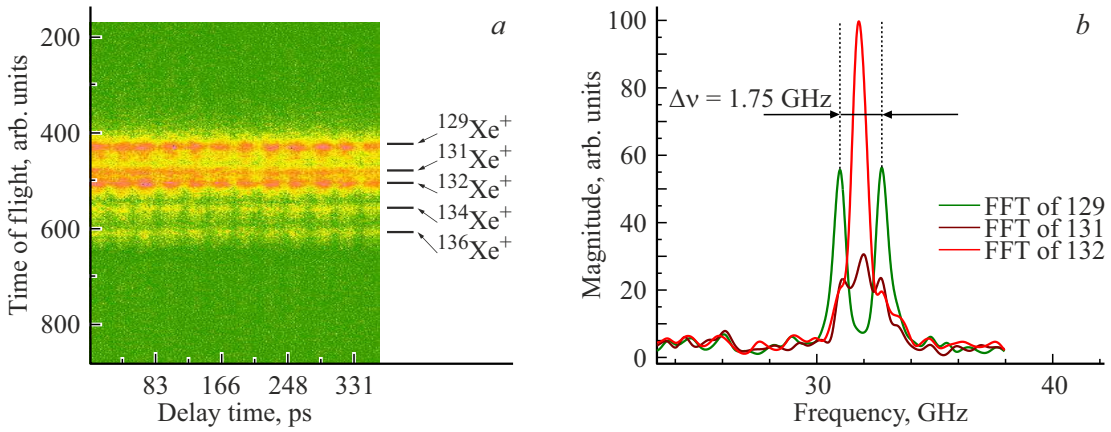

Figure 1. *a* — mass-spectrum of Xe^+ isotopes as a function of delay time; *b* — Fourier spectrum of ionization signals of Xe^+ isotopes.

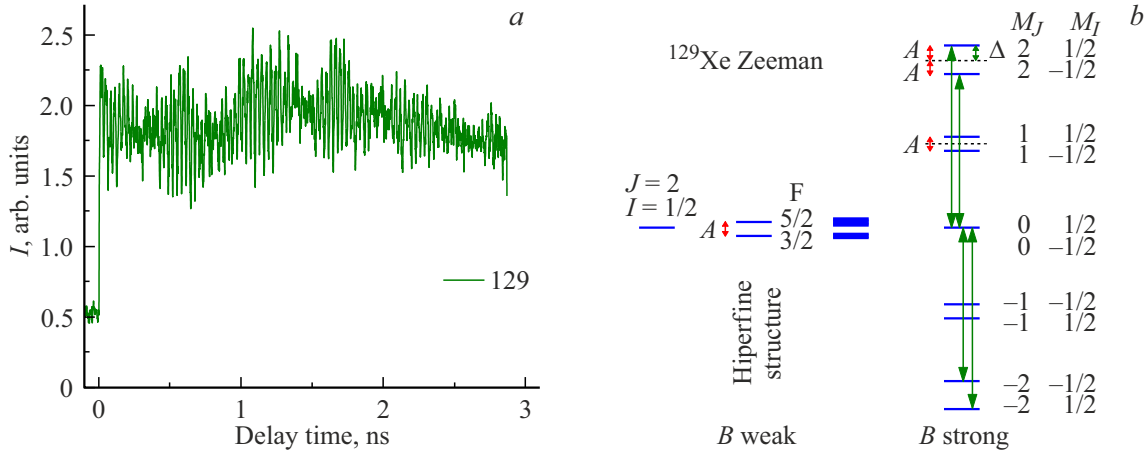

Figure 2. *a* — ^{129}Xe isotope ionization signal as a function of delay; *b* — hyperfine structure of ^{129}Xe isotope in weak and strong magnetic fields.

respectively, in contrast to even isotopes that have zero moment of their cores.

It?s worth noting that a two-time reduction of the magnetic field results in reduction of the beats frequency for Xe isotopes: from $32\,ns^{-1}$ down to $16\,ns^{-1}$. No changes are observed in frequencies of isotope modulation signals. For example, modulation frequencies of the ^{129}Xe isotope for two-fold different magnetic fields are 1.75 and 1.82 GHz, which is within the ± 0.05 GHz uncertainty of the experiment. The error of frequency determination is defined by the limited range of delays (3 ns) with a factor of 1/10. This additionally confirms our previous observations [7] regarding the lack of a connection between the nature of quantum beats signal modulation and the Paschen-Back effect for fine structure levels. The realization of this effect gives rise to a typical nonlinear dependence of ω_m on the magnetic field strength [1,5,18] with prevailing quadratic term such that with a decrease of ν_b by two times almost four-times decrease of ω_m should be expected.

The QBSM is observed in the integral signal of Xe^+ isotopes as a result of QBSM of Xe isotopes with add masses of 129, 131. It?s worth noting that the natural Xenon contains almost 48% of odd isotopes [19].

Discussion of experimental results

The emergence of quantum beat signal modulation can be explained by the hyperfine (HF) structure of atomic levels. The pattern of energy splitting of Zeeman components in a magnetic field is dependent to a significant extent on the I spin of the core, which is $I = 1/2$ for the ^{129}Xe isotope. With the presence of I spin in atom, it is necessary to take into consideration the additional interaction of magnetic moment μ_I of the core with the moment μ_J of the electron shell, as well as the interaction with the external magnetic field **B**. In case of weak field, the total moment $\mathbf{F} = \mathbf{I} + \mathbf{J}$ of atom is $F = 3/2, 5/2$, while the energy of Zeeman sublevels M_F is linearly dependent on B [2,18]:

$$\varepsilon_{m_F} = \mu_0 g_F M_F B, \quad M_F = -F, \ldots, F. \tag{1}$$

Atomic states for F equal to 3/2 and 5/2 are split into $2F+1$ sublevels, i.e. into four and six components, respectively (the case of weak field in Fig. 2, *b*).

For the case of strong external field, which energy of interaction with the electronic moment μ_J is greater than the HF-interaction between **I** and **J**, each M_J sublevel of the Zeeman electronic multiplet is split into $2I+1$ HF-components [2] (the case strong field in Fig. 2, *b*). Existing values of hyperfine splitting are defined by relation (VIII.13) from [18]:

$$\delta W = 2\Delta, \quad \Delta = AM_JM_I, \tag{2}$$

where $A = 0.02976\,\mathrm{cm}^{-1} = 0.893\,\mathrm{GHz}$ — constant of magnetic dipole interaction for ^{129}Xe isotope [20], M_J, M_I — projection of electronic shell moment and core on the quantization axis oriented along the magnetic field.

In this experiment the two-photon excitation of horizontally polarized radiation is used to transit the ^{129}Xe isotope to a superposition of two states of Zeeman components with projections of $M_J = \pm 2$ (green lines in Fig. 2, *b*). The correspondent HF-splitting according to formula (2) is $2A = 1.786\,\mathrm{GHz}$. This value is approximately equal to the above-presented experimentally measured beats modulation frequencies for the ^{129}Xe isotope.

The HF-splitting for the ^{131}Xe isotope is somewhat more complex and is beyond the scope of this study.

Conclusion

The experimentally observed modulation of quantum beats signals in recorded photocurrents is manifested in partial signals of Xe^+ isotopes with odd masses 129, 131; at the same time there is no similar signal modulation of isotopes with even masses 132, 134, 136. Modulation structure of the ^{129}Xe isotope is considered in details in this study. Our analysis of obtained laws shows that for isotopes with non-zero spins of their cores the modulation of observed signals takes place at a frequency that corresponds to the hyperfine splitting. The emergence of the modulation itself is caused by the nonlinear Zeeman effect with involvement of sublevels of the HF structure when external magnetic fields cause nonequidistant energy shifts of Zeeman components. Therefore, when it comes to the behavior of the second and further harmonics in the oscillating signals of photocurrent, their amplitudes nonlinearly track the growth of magnetic fields. This sort of nonlinearity makes it easier to record the polarizing moments of high-order atoms in the techniques sought to design quantum magnetometers (FM NMOE) [21] and based on the nonlinear magneto-optic effect.

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Conflict of interest

The authors declare that they have no conflict of interest.

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