# Use of a photon-echo technique in atmospheric sensing

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The possibility of using the photon-echo technique for laser sensing of the atmosphere is analyzed. The properties of photon-echoes are discussed using, as an example, photon echoes (PE) in the molecular iodine vapor. Possible versions of PE lidars are discussed and the ways of their optimal use for atmospheric sensing are analyzed.

### Introduction

Photon (light) echo (PE) is an optical coherent response of a resonance medium to two (and more) time-separated laser pulses with the pulse durations and the gap between the pulses being shorter than the characteristic relaxation time of the resonance medium. Spectroscopic and some technical applications of the PE technique are well known (see, for example, Refs. 1-4). A promising research field connected with this technique has arisen in nonstationary spectroscopy, and it has been called optical echo spectroscopy. 1 Thanks to the development femtosecond pulsed lasers, optical echo experiments are already being conducted in any medium (solid, liquid, and gas) and often without the use of low-temperature equipment. Such experiments give the information on fast processes. Capabilities of the PE technique in solving the problems of wave-front conversion are also well known. They gave rise to a new research field, the dynamic holography, called echo holography (see, for example, Refs. 5 and 6). With the advent of longliving PE,7 intense development of the optical-echo processors has begun (see, for example, Refs. 8 and 9).

In this paper, we undertake an attempt to demonstrate some peculiarities of the PE technique in laser sensing of the atmosphere. The monograph by Zuev and Zuev<sup>10</sup> and the reviews and papers cited below in this paper served as excellent aids for us. One of the significant points in this analysis was selection of the resonance medium for a PE lidar. First, we proceeded from the requirements that a resonance medium is most convenient for use and there is no need in cryogenic equipment. The vapor of molecular iodine is most convenient from this point of view, because its temperature in experiments is equal to the ambient temperature and the experiments are technically safe as compared, for example, with the experiments with high-temperature vapors of alkaline atoms (Na, Rb, Cs). Since we have a many-year experience in echo experiments just in the I2 vapor, all illustrations hereinafter in this paper are given just for this gaseous medium. The experiments were conducted at the

wavelengths of 570.8, 571.5, and 590 nm corresponding to three optically allowed transitions in the absorption band  $X^1 \Sigma_q^+ \to B^3 \Pi_{0u}^-$  of the I<sub>2</sub> vapor. The pressure of I<sub>2</sub> vapor varied from 15 to 70 mTorr. Under these conditions, the time of longitudinal relaxation  $T_1$ ranged from 0.2 to  $0.95~\mu s$ , and the time of irreversible transverse relaxation  $T_2$  ranged from 40 to 70 ns. The duration of the exciting pulses (10 ns) and the intervals  $\tau$  between the pulses (20-70 ns) were shorter than the relaxation times, and therefore the condition of coherent interaction with the resonance medium was fulfilled. The PE intensity achieved 10% of the intensity of exciting pulses. Note that in Ref. 11 the continental and maritime aerosol was studied experimentally at the wavelengths from 426 to 920 nm, including the wavelength of 572 nm. This allows us to hope for the utility of the PE technique in studying spectral and dynamic characteristics of the aerosol and of the processes of its formation, internal kinetics, and disintegration.

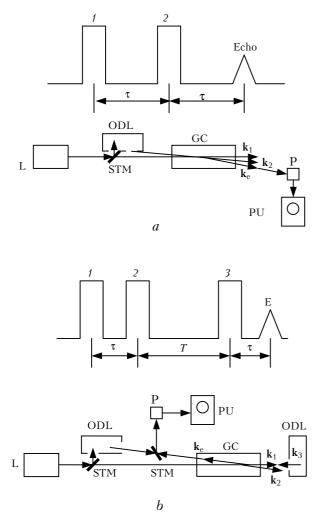
Impurity crystals, especially, those from the class of Van Fleck paramagnetic crystals, can be used as a resonance medium of the PE lidar. According to Ref. 10, the Ti:sapphire laser is widely used in laser sensing of the atmosphere. This laser operating in the femtosecond range of pulse duration is also used in echo spectroscopy of Van Fleck paramagnetic crystals and, in particular, trivalent thulium doped crystals at the wavelength near 780 nm. It is well-known<sup>8,9</sup> that Van Fleck paramagnetic crystals are the promising medium for optical echo processors with the time of the optical phase memory achieving ten hours. This allows the PE lidar to combine the study of received laser signals with their long-time storage and processing.

## 1. Photon echoes and their properties

Note that, among the variety of optical echo signals, the signals of primary and stimulated PE have gained the widest acceptance. The primary PE is the result of coherent effect of two time-separated exciting pulses onto the resonance medium, and stimulated PE

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results from the effect of three pulses. The order of their excitation is explained in Fig. 1.



**Fig. 1.** The order of excitation of signals of primary (a) and stimulated (b) PE: laser L, optical delay line ODL, semitransparent mirror STM, gas cell GC, photodetector P, and unit for processing echo signals PU;  $\mathbf{k}_1$ ,  $\mathbf{k}_2$ , and  $\mathbf{k}_3$  are wave vectors of exciting pulses,  $\mathbf{k}_e$  is the echo wave vector ( $\mathbf{k}_e = \mathbf{k}_1 - \mathbf{k}_2 + \mathbf{k}_3$ ; at  $\mathbf{k}_1 = -\mathbf{k}_3$  we have  $\mathbf{k}_e = -\mathbf{k}_2$ ).

Unlike the radio wave analogs, photon echoes are emitted by the resonance medium in certain directions defined by the conditions known as the conditions of spatial synchronism. Thus, for the signal of primary PE emitted by the medium at the time  $2\tau$  (where  $\tau$  is the interval between the exciting pulses), this condition can be written as

$$\mathbf{k}_{\mathrm{e}} = 2\mathbf{k}_2 - \mathbf{k}_1,$$

where  $\mathbf{k}_1$  and  $\mathbf{k}_2$  are the wave vectors of the exciting pulses, and  $\mathbf{k}_e$  is the echo wave vector. In the PE experiments in gases, the angles between the wave vectors  $\mathbf{k}_1$  and  $\mathbf{k}_2$  are small because the zone of interaction of both pulses with the gas is to be maximally enlarged. However, if the second pulse is a

standing wave, then the calculations show that the echo called the reversed signal propagates along the direction  $-\mathbf{k}_1$  regardless of the direction of the standing-wave pulse. The wave front (i.e., the surface of constant phase  $\phi(\mathbf{r})$ ) of certain pulses can bear information, i.e., be nonplanar. The calculations show<sup>3</sup> that if the first pulse has a nonplanar wave front and the second one has a plane wave front, then the wave front of the echo signal is phase-conjugated with respect to the wave front of the first pulse:

$$\varphi_{e}(\mathbf{r}) = -\varphi_{1}(\mathbf{r}),$$

i.e., in this case the mode of an active PE mirror occurs. In the regime of stimulated PE, both the phase conjugation of the echo-signal wave front and retrace (with a delay) of the code-pulse wave front can occur. Note that some problems of echo holography in degenerated and multilevel systems (characteristic of gaseous media) have been already discussed by us in Ref. 12, but that discussion was not aimed at the use of echo holography for atmospheric sensing. In this paper, this gap is partially compensated for with the use, as an example, of the active PE mirror.

For simplicity, the further discussion deals with the signals of primary PE. If the resonance medium is exposed to a time-coded series of coherent pulses, whose summed duration is shorter than the time of phase relaxation, then this medium responds, with some delay, by emitting the time-reversed series of coded echo signals. 13 In this case, the amplitude of the stored signals in the series should not be very high, so that  $\sin\theta_{\eta} \approx \theta_{\eta}$ , where  $\theta_{\eta}$  is the "area" of the corresponding signal. Information can be entered not only into the wave front, but also into the temporal shape of the exciting signals and reproduced with a delay in the temporal shape of echo signals. In Refs. 14 and 4 we have described in detail the corresponding equipment. As an illustration, Fig. 2 shows the oscillogram<sup>15</sup> of the first pulse with the coded temporal shape, the second pulse (that is often referred to as readout beam), and, finally, the primary PE signal, whose temporal shape is reversed with respect to the temporal shape of the first pulse (Fig. 2).

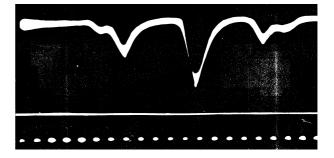


Fig. 2. Oscillogram illustrating the effect of reversal of the temporal shape of the primary PE (the first from the right) in  $\rm I_2$  vapor with respect to the temporal shape of the first pulse. <sup>15</sup> Marks on the scale are separated by 10 ns.

It was shown in papers 16 and 17 devoted to experiments, that if the code pulse (or pulse series) is a series of coherent coded signals with mutually perpendicular linear polarizations, then, using the linear polarization of the second pulse as a key (as a primary PE signal), the parts of the information borne by the code pulse (or by the code pulse series) can be read out step-by-step. This mode may prove important for laser transfer of coded information in space.

With a large number of polarization channels, a specialized device is needed for correct identification of the channel number. An example of such a device with four information channels has been discussed in Ref. 5. The set of readouts at the output of these channels at every instant of time is a 4D vector with the interdependent coordinates, and every such vector can be considered in the algebra of quaternions. 18,19 Changing the conditions of PE excitation, we can change the content of operation of processing the numbers in this algebra. This is important for information protection against illegal access during laser transfer in space.

The PE signals are characterized by certain polarization regularities, the use of which for gas studies has led to the advent of the polarization-echo spectroscopy. Depending on the type of the energy transition, the angular momentum (J), and the angle  $\psi$ between the linearly polarized exciting pulses, the angle  $\varphi_e$  between the linear polarizations of the primary PE and the second pulse in the case of large angular momenta  $(J \gg 1)$  can be determined as:

- (a) for the transition  $J \to J$ :  $\tan \varphi_e = \frac{1}{3} \tan \psi$ ,
- (b) for the transition  $J \leftrightarrow J + 1$ :  $\tan \varphi_e = -\frac{1}{2} \tan \psi$ .

Since the sensing pulse with the polarization "wandering" in time plays the role of the first pulse in a PE lidar, the angle  $\varphi$  of the echo signal varies in time. However, the PE signal is emitted with a delay, during which it can be stored and numerically processed. Note that the non-Faraday rotation of the polarization vector of the echo signal takes place in the longitudinal magnetic field<sup>20–22</sup>; in some cases, this can help in polarization identification of echo signals.

# 2. Possible schemes of a lidar based on photon echo (PE lidar)

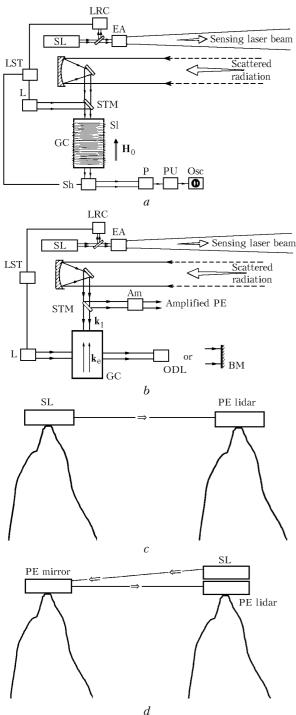
The PE lidar differs from the traditional lidar 10 by the optical phase memory that allows

- (a) studying and processing the received sensing signals with a controlled delay, protective signal coding for further laser transportation in the atmosphere;
- (b) using femtosecond multipulse series in place of a single sensing pulse and then studying fast atmospheric processes in the atmosphere in the mode of femtosecond accumulated PE (or other modes, see Ref. 23);

- (c) implementing and using various modes of dynamic echo holography for visual study of objects, sensing pulses meet on their propagation path;
- (d) implementing the phase conjugation regime (whose advantages are well-known<sup>24</sup>); essentially, PE lidar has some properties of the active mirror with the phase memory and controlled delay;
- (e) studying and converting the spectra of received signals, because the PE spectrum depends directly on the spectrum of exciting pulses:  $S_e \sim S_1^* S_2^2$ (Ref. 1);
- (f) along with the study of "wandering" polarization of sensing signals, PE lidar allows conversion and processing of polarization (for example, in longitudinal magnetic fields).

Figure 3 shows two possible schemes of including the PE lidar (and PE mirror) in laser sensing of the atmosphere. When developing these schemes, we tried to keep maximally the units of a traditional lidar. 10 Along with the units of the traditional lidar, the PE lidar (Fig. 3a, upper panel) includes a cell GC with a resonance gaseous medium (for example, I2 vapor) placed in a solenoid for generation of a longitudinal magnetic field  $H_0$ , the unit for synchronous triggering of the laser LST, frequency tunable laser (optical layout of this laser with the associated units is described in detail in Refs. 14 and 4), photodetector (ELU-FTS), signal processing unit, and fast oscilloscope. Laser sensing of the atmosphere proceeds in the same way, as in a traditional lidar. The sensing radiation is assumed pulsed. A laser signal passed through the atmosphere plays the role of the first pulse at excitation of the primary PE. A portion of the sensing radiation is directed through the synchronous triggering unit to the entrance of the tunable dye laser and initiates generation of the second pulse needed for PE excitation. The exciting signals passed through the medium are cut off by a shutter, which passes only the PE signal to the photodetector input. From the photodetector output, the electric analog of the PE signal comes to the processing unit. PE signals can be also observed visually on the screen of a fast oscilloscope. During operation in the regime of echo holography, a CCD camera should be used instead of the photodetector.

Note that optical echo spectroscopy employs also continuous-wave optical sources with the very low degree of coherence. In this case, the mode of incoherent PE takes place (see, for example, Ref. 25). As known, the radiation of any non-laser source is nevertheless characterized by coherence during ultrashort time intervals (of femtosecond duration). Because of the existence of such ultrashort intervals of coherence, accumulated femtosecond PE can be excited.<sup>26</sup> The mode of incoherent PE can be useful in the cases when sounding radiation loses its coherence in the atmosphere to a great degree. Figure 3b shows the optical arrangement of a lidar with an active PE mirror.



**Fig. 3.** Inclusion of PE lidar and PE mirror in the process of laser sensing of the atmosphere: (a) a possible scheme of PE lidar: laser radiation control unit LRC, sensing laser SL, emitting antenna EA, unit of laser synchronous triggering and shutter control LST, laser L for excitation of PE, semitransparent mirror STM, solenoid Sl, gas cell GC, shutter Sh, photodetector P, unit for processing of echo signals PU, oscilloscope Osc; (b) possible scheme of active PE mirror: optical delay line ODL, back mirror BM, amplifier Am, all other designations are the same as in the upper panel; (c) scheme involving a PE lidar in the process of single-pass atmospheric sensing; (d) scheme of using a PE lidar in double-pass atmospheric sensing.

Certainly, the effect of stimulated Brillouin scattering (SBS) can be used for the wave front phase conjugation.<sup>24</sup> But PE is convenient, because it gives rise to optical phase memory, and the echo signal itself is characterized by some delay. As in the previous case (Fig. 3a), the pulse of the sensing radiation is directed onto the cell filled with a gas, and there it plays the role of the first pulse at excitation of the reversed PE. A part of the sensing radiation passes through the synchronous triggering unit and turns on the tunable laser, whose pulses propagate in the direction normal to the direction of the sensing pulse. Having passed through the cell, the second pulse comes to the optical delay line (in the simplest case, reflects from the back totally reflecting mirror) and then propagates in the opposite direction. In the case that the mirror plays the role of the delay line, the medium is exposed to the standing-wave pulse. It is known that under these conditions the wave vector  $\mathbf{k}_2$  of the second pulse does not meet the condition of spatial synchronism of the echo signal:  $\mathbf{k}_e$  = -  $\mathbf{k}_1$ , i.e., the echo signal propagates in the direction opposite to the direction of the sensing pulse, and its wave front (as was shown in Section 1) is phase-conjugated with respect to the wave front of the sensing pulse. Then the echo signal is directed to the amplifier by a semitransparent plate, and then the amplified echo signal plays the role of the sensing pulse. Figures 3c and d explain the modes of single-pass and two-pass sensing of the atmosphere.

Note that the effect of thermal blooming on laser beams propagating in the atmosphere<sup>27</sup> is studied in nonlinear optics for a long time (see, for example, Ref. 28, which studies soliton modes of optical signal propagation).

We understand, of course, that this paper mostly formulates the problems of using coherent phenomena (like PE) for laser sensing of the atmosphere and transportation of coded signals through it. The results of solution of these problems will be likely the subject of future publications.

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