There and back again: Extending optical storage lifetime by retrieving photon echoes from semiconductor spin excitations

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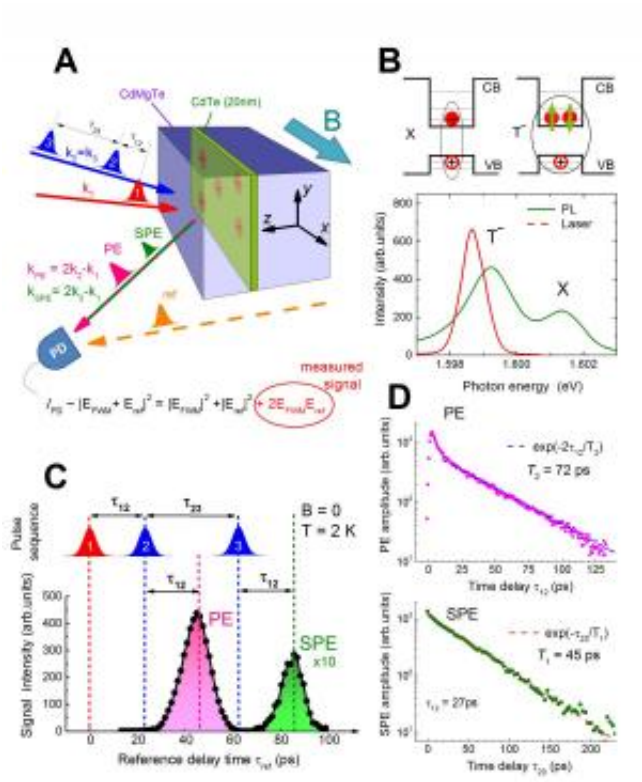


Scheme of photon echo experiment and optical properties of investigated structure. (A) The CdTe/(Cd,Mg)Te quantum well (QW) is optically excited with a sequence of three laser pulses with variable delays t12and t23 relative to each other. The resulting four-wave mixing transients |EFWM(t)| are detected in 2k2-k1 direction using heterodyne detection. All measurements are performed at temperature of 2 K. (B) Top: schematic presentation of exciton (X) and trion (T-) complexes in QW. The QW potential of conduction (CB) and valence (VB)

bands leads to spatial trapping of electrons and holes. Bottom: Photoluminescence (PL) spectrum (solid line) measured for above-barrier excitation with photon energy 2.33 eV, demonstrating X and T- emission. The laser spectrum (dashed line) used in photon echo experiment is tuned to the low energy flank of T- emission line. (C) Four-wave mixing transients for t12 = 23 ps and t23 = 39 ps. Spontaneous (PE) and stimulated (SPE) photon echo signals appear at tref =2t12 and tref =2t12 +t23, respectively. (D) Decay of PE and SPE peak amplitudes. From exponential fits (dashed lines) we evaluate T2=72 ps and T1 = 45 ps. Credit: Ilya A. Akimov.

(Phys.org) —For all of their differences, classical and quantum communication have at least one thing in common: the importance of being able to store optical information. That being said, optical storage is a complex process that depends largely on the material being used to convert, store and retrieve this information in a controllable, consistent manner – a process especially prone to short optimal memory times when implemented in certain semiconductor quantum structures. Recently, however, scientists at Technische Universität Dortmund, Germany in collaboration with Saint Petersburg State University, Russia and Institute of Physics in Warsaw, Poland demonstrated magnetic-field-induced long-lived stimulated photon echoes[1] – coherent optical phenomena in which resonant excitation of a medium by short optical pulses results in a delayed coherent optical flash response – in the electron–trion system, allowed the bidirectional coherent transfer of quantum information in a semiconductor between optical and spin excitations. (Trions are excitations comprising three charged quasiparticles – emergent phenomena that occur when a microscopically complex system, such as a solid, behaves as if it contained different weakly interacting particles in free space.)

In their study, the researchers found that picosecond optical pulses and an applied weak transverse magnetic field led to the transfer of a short-

lived optical excitation into a long-lived electron spin state. In turn, this induced stimulated photon echoes with high bandwidth on submicrosecond timescales that exceeded optical excitation lifetime by a factor exceeding three orders of magnitude. Moreover, the scientists state that the ability to address all three spin components –parallel and perpendicular to applied magnetic field – as well as the energy level structure of localized trions being identical in quantum wells and self-assembled quantum dots make their approach "highly appealing" for future memory device applications and may lead to the fabrication of semiconductor nanostructure-based optical memories.

Dr. Ilya Akimov discussed the paper that he, Doctoral Student Lukas Langer and their co-authors from Dortmund, St-Petersburg and Warsaw published in *Nature Photonics*, starting with the main challenges in devising a new experimental approach to stimulated photon echoes by transferring the information contained in the optical field into a spin system, where it is decoupled from the optical vacuum field. "A photon echo can be considered as a flash of light initiated in a medium after a sequence of two or more optical pulses," Akimov told *Phys.org*. "Properties of the photon echo pulse – that is, intensity, optical coherence and phase – are identical to the properties of the initial pulse; therefore this phenomenon can be used to store information in all-optical systems." Photon echoes because they occur in structures which rapidly dephase optical excitation in response to the first pulse, but with the second pulse reverse the dephasing process.

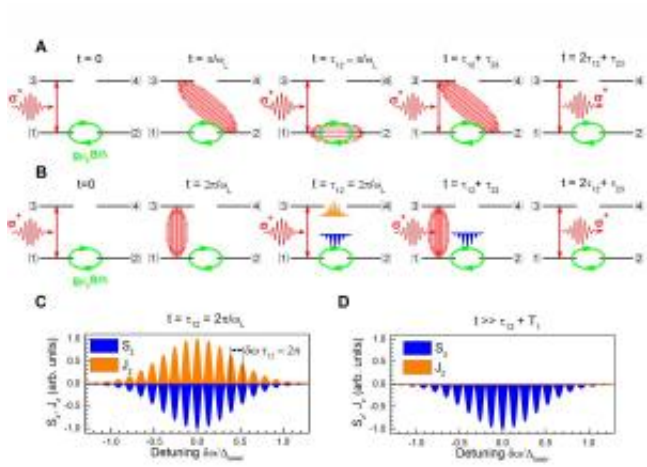
Akimov pointed out that the timescale at which photon echoes can be observed is determined by the period during which optical excitation coherence is preserved. "This is why photon echoes are coherent," he explained. "If scattering processes are suppressed, optical excitation coherence is limited by its lifetime as governed by intrinsic properties of the materials. Nevertheless there is a general rule that the more efficient light-matter interaction, the faster decay into the ground state will be

observed – for example, faster spontaneous emission due to the optical vacuum field. Here we have a dilemma," he noted. "On the one hand, we'd like to excite the medium quickly with the shortest possible pulse, which requires strong light-matter interaction. However, this limits the timescale at which the photon echoes can be observed." The opposite is also true: To observe long-lived photon echoes, materials with weak light-matter coupling are needed – and although semiconductor systems belong to materials with strong light-matter interaction, it's possible to extend photon echo decay if the optical excitation is transferred into the spin excitation and then recovered back to the optical excitation – that is precisely what Akimov and co-authors have accomplished.

"In order to achieve this goal," he said, "we've used semiconductor quantum wells with excess electrons." (A quantum well is a thin layer which can confine quasiparticles – emergent phenomena that occur when a microscopically complicated system, such as a solid, behaves as if it contains weakly interacting particles in free space – in the dimension perpendicular to the layer surface.) What's key is that the spin excitation coherence time of such electrons decays three orders of magnitude more slowly than the lifetime of optical excitations. "In our protocol the first pulse leads to optical excitation in semiconductor quantum well; next, after dephasing of the optical excitation the second pulse transfers optical excitation into the electron spin excitation; and finally – even after a long delay, which can be 1,000 times longer than the delay between the first and second pulses – we apply the third pulse." This third pulse transforms spin excitation back to optical excitation and initiates rephasing, so that resulting photon echoes are retrieved from the spin ensemble – and therefore the third pulse can be associated with the readout of optical information previously saved by the optical-to-spin transformation induced by the second pulse.

"The unique feature of photon echoes is that they contain the information about the first optical pulse," Akimov pointed out.

"Therefore, photon echoes can be used for information technology optical memory applications. Moreover, it's possible to perform not only classical light storage but to use photon echoes for realization of quantum optical memories." This is essential, he stresses, for the development of quantum information and communication systems which use the laws of quantum mechanics to significantly enhance the speed and capacity of future **computers**.



Schematic presentation of the main mechanisms responsible for magnetic-field-induced stimulated photon echoes (SPE). The whole process comprises three steps: 1. pulse 1 creates the optical excitation (initialization—conversion of the optical field into a material excitation); 2. pulse 2 performs a transformation of the optical excitation into the spin system (storage); 3. pulse 3 stimulates the photon echo (readout). Optical pulses are circularly polarized. (A) Transfer of optical coherence into electron spin coherence (Sx and Sy components). The efficiency is maximum for t12=p/wL. (B) Creation of spectral spin fringes for

electrons and trions (Sz and Jz components). This mechanism is most efficient for t12=2p/wL. The spectral spin gratings for electrons and trions are shown in (C) at the moment of creation by the second pulse (t=t12=2p/wL) and in (D) after trion recombination and before arrival of pulse 3 (t>>t12+T1). Credit: Ilya A. Akimov.

"For this reason, research on quantum optical memories attracted a lot of attention," Akimov told *Phys.org*. "Current investigations of photon echoes have concentrated primarily on atomic vapors and rare earth crystals with long storage times, which are crucial for implementation of robust light-matter interfaces. However," he noted, "light-matter coupling is weaker in these systems, so operation speed is not as fast as it could be in semiconductors. For example," he illustrated, "efficient optical excitation in atomic systems is possible with optical pulses longer than one nanosecond, which slow down the operation speed by three orders of magnitude as compared to our protocol – and for rare earth crystals the pulse duration should be even longer."

In contrast to classical storage, quantum memory forbids measurement of the optical field during saving and retrieving processes. "In other words," Akimov said, "storage of non-classical quantum light – such as squeezed light or a single photon – should occur without knowing which optical fields have been stored and retrieved, because otherwise the quantum state would be irreversibly destroyed during the measurement procedure. However, our protocol allows quantum storage since transfer between optical excitation and spin excitation does not require state measurement." In other words, the new protocol transfers a quantum superposition between optically coupled states (optical excitation) and the other pair of states coupled by a magnetic field (spin excitation). In this process no measurement takes place – just the transformation between different excitations.

Regarding the quantum well, the researchers specifically concentrated on an n-doped CdTe/(Cd,Mg)Te quantum well where storage time increased from picoseconds to tens of nanoseconds. The structures were grown by Prof. Grzegorz Karczewski and Prof. Tomasz Wojtowiczin the Institute of Physics, Polish Academy of Sciences in Warsaw using molecular beam epitaxy. (CdTe/(Cd,Mg)Te is a cadmium telluride compound in which some of the cadmium is replaced by magnesium.) "The cadmium telluride semiconductor quantum well structure is a model proof-of-principle system for extending the photon echo delay," Akimov told *Phys.org*. "In such two-dimensional structures, the carriers are confined in one direction; this results in well-defined spin-level system and clean selection rules for optical transitions. Secondly, n-type doping of barriers with donors provides excess electrons in the quantum well which, again, are responsible for long-lived spin excitations."

That said, while using cadmium telluride quantum wells enabled very clean experiments on the ensemble of trions to be performed because their optical transitions are well isolated spectrally, the researchers had to maintain weak optical pulse intensity to prevent interactions between weakly localized trions. "In order to increase the efficiency and to achieve longer delays for photon echoes it is necessary to try different type of semiconductor nanostructures which can be also based on other compounds.,' Akimov explained. "One of such candidates is the ensemble of quantum dots where the electrons and holes are localized much more strongly in all three dimensions. This is in contrast to quantum wells where strong confinement is present only along one direction."

Finally, Akimov noted that in semiconductors there are two types of fundamental optical excitations: *excitons* (electron-hole pairs bound by Coulomb interactions) and *trions* – charged excitons consisting of an exciton bound with an excess electron or hole. "A trion is a three-particle complex, and after its decay there's always an excess carrier left," he

explained. "In our case, we deal with excess electrons which possess spin 1/2. Therefore, in contrast to excitons, it is possible to save information about optical excitation in the spin of the excess electrons left after trion recombination. This transformation is only possible when an external magnetic field is applied, since it allows us to mix the electronic states in the proper way." The most salient advantage of quantum well structures is that exciton and trion resonances are spectrally well separated – meaning that picosecond laser pulses let the researchers address only the optical transition from excess electron to trion.

For all of these seemingly daunting challenges, the researchers' key insight was to study photon echoes emitted by trions in semiconductor nanostructures subject to an external magnetic field – and by then using a transient four-wave mixing (FWM) technique to measure magnetic-field-induced long-term photon echoes, they were able to show that photon echoes can be retrieved from excess electron spin ensembles. (Transient four-wave mixing belongs to time-resolved coherent spectroscopy based on non-linear optics, whereby interactions between two or three optical pulses in medium produce fourth optical field in the signal) "We used ultrashort optical pulses with duration of about one picosecond," Akimov explained, "because efficient optical excitation in semiconductors is possible on the order of 0.1-1ps." In addition, he said, the experiments had to be performed at extremely low temperatures – about two degrees above absolute zero – in order to keep the system robust against interactions with phonons (collective excitations, similar to quasiparticles, in a periodic, elastic arrangement of atoms or molecules in condensed matter, such as solids and some liquids), as well as to suppress other relaxation mechanisms which could lead to irreversible dephasing of optical and spin excitations and thereby loss of coherence. "From an experimental point of view," he added, "our primary challenge was combining four-wave mixing with ultrashort picosecond pulses and external magnetic fields at low temperatures."

The current study demonstrates that photon echoes can be retrieved from the spin system on the timescale of 10-100 ns. "However," Akimov said, "this time delay is still too short for practical applications. In order to solve this problem we need to extend the decay time of spin excitations." There are two possible reasons for decay of spin excitations: dephasing of spins and irreversible spin relaxation through decoherence – that is, due to interaction with the environment. "The first point can be addressed by means of spin resonance techniques using dynamic decoupling," he explained, "which is an approach largely the same as photon echo but based on periodic excitation of the spin ensemble with microwave pulses which lead to spin echoes. In that way it will be possible to keep the spin ensemble of excess electrons free of dephasing, and timescales up to tens or even hundreds of microseconds may be achieved. However, irreversible spin relaxation is more difficult to solve – but there are several attempts to reduce hyperfine interaction between nuclear and electron spins. One of the solutions would be to use compounds with isotopes carrying zero nuclear spin. In this case storage times in the milliseconds can be available."

In fact, Akimov added that the scientists plan to investigate extending the timescale of photon echoes further into the microsecond and millisecond range. "We'll test other nanostructures, such as quantum dots with strong trion localization, and will search for new materials with suppressed spin excitation decay. In addition," he said, "we'll use spin resonance techniques in order to eliminate spin dephasing in the ensemble of excess electrons."

Akimov also mentioned applications beyond optical memory. "While most applications are related to optical memories where the optical information should be saved and released on demand," he said, "there's another fundamental aspect: Our studies combine optical and spin phenomena, and in this sense it's very interesting to explore our approach for monitoring the time evolution of combined optical and spin

excitations."

A unique feature of photon echo experiments is the dephasing which already occurs at the initial stage directly after excitation with the first pulse, where the sequence of two linearly polarized pulses create comprehensive spin distribution for excess electrons without net spin polarization. While each of the electrons has a certain well-defined spin, the ensemble spin polarization, or average spin, is zero – and the information about the optical pulses, such as polarization and interpulse delay, is encoded in the spins of excess electrons. "This differs from conventional techniques," Akimov pointed out. "For example, in well-established pump-probe experiments the non-zero spin polarization in the system is first induced by a circularly polarized pump pulse, and *then* the evolution of the spin in time is probed." The scientists therefore believe that their approach based on photon echoes in a magnetic field constitutes an interesting platform for fundamental spin studies.

Along these exploratory lines, *Phys.org* asked Akimov if, given that storage times of seconds or longer might be possible by further exploiting the hyperfine interaction between electrons and nuclei in quantum dots, quantum wells and self-assembling quantum dots might at some point be combined in a single quantum system that emulates human short- and long-term memory. "I think we are still far from that," he replied. "In order to achieve this goal it would be necessary to establish a net of such quantum dot ensembles, analogous to cells, which would communicate between each other." He added that while he does not exclude such possibility, he emphasizes that such a quantum system would very complex and would contain and integrate far more than a simple set of quantum nanostructures. "Several challenging issues such as communication between different ensembles have to be addressed, and for that it is necessary to accomplish directed and selective coupling of light at the nanoscale in and out of the cells. Accordingly, realization of such a network would need integration of photonic crystals or

waveguide layers which can be based on semiconductors. Nevertheless," he concluded, "this is a special area of research which deserves a lot of attention."

More information: Access to long-term optical memories using photon echoes retrieved from semiconductor spins, *Nature Photonics* (Published online 28 September 2014), doi:10.1038/nphoton.2014.219

Related:

[1]Magnetic-Field Control of Photon Echo from the Electron-Trion System in a CdTe Quantum Well: Shuffling Coherence between Optically Accessible and Inaccessible States, *Physical Review Letters* **109**, 157403 (2 October 2012), doi:10.1103/PhysRevLett.109.157403