The project is focused on theoretical and experimental studies of the quantum coherence effects at the atomic and nuclear transitions in solids. The main results are the following.

1. Electromagnetically Induced Transparency (EIT) and all-optical writing and probing of quantum coherence in room temperature solids are demonstrated experimentally in the first time.
2. The method for coherent suppression of excited state absorption in laser crystals is suggested. It opens the prospects for realization of the variety tunable solid state lasers including UV and VUV ranges.
3. High-efficient THz generation by means of resonant three-level mixing based on EIT in solids is predicted.
4. EIT in gamma-ray range of frequencies at the nuclear transitions via the nuclear level crossing is demonstrated experimentally in the first time.
5. Slowing of the gamma-ray photons due to the absorption line doublet is demonstrated in the first time.
6. Modifications of the Mossbauer absorption spectra under the action of laser radiation demonstrated experimentally in the first time.
7. The method for suppression of the inhomogeneous line broadening of the Mossbauer spectra is suggested.
### 15. SUBJECT TERMS
Coherent nuclear phenomena, Electromagnetically induced transparency, lasing without inversion, gamma-ray laser, Mossbauer spectroscopy

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Laser Manipulation of Nuclear Transitions

Principal Investigator: Olga Kocharovskaya

Texas A&M University

College Station, TX 77843-4242

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Grant Number: FA 9550-05-1-0016, Modification P00003
Contract/Grant Title: Laser Manipulation of Nuclear Transitions
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A. Number of Papers published in referred journals: 34


B. Number of Papers submitted to referred journals, but not published: 3


C. Number of books or chapters submitted, but not yet published: 0

D. Number of books or chapters published (for each, provide a complete citation): 0

E. Number of printed technical reports/non-referred papers (for each, provide a complete citation): 0

F. Number of patents filed: 0

G. Number of patents granted: 0

H. Number of invited presentations (for each, provide a complete citation): 40


30. P.Anisimov, Y.Rostovtsev, O.Kocharovskaya, Line narrowing in Mossbauer spectroscopy, invited, 37th Winter Colloquium Physics of Quantum Electronics, Jan.3-6, 2007.
33. P.Anisimov, Y.Rostovtsev, O.Kocharovskaya, Suppression of inhomogeneous line broadening of the Mössbauer transitions, poster, Princeton-TAMU Symposium on Quantum Mechanics, Informatics and Control, Princeton, 6-7 April, 2007.

Number of Colloquia and Seminars: 5

1. O.Kocharovskaya, Distinguished Women Physicists Lecture Series, Department of Physics, University of Texas at Austin, March 2005: (i)“Laser control of nuclear transitions”;(ii)“Atomic Interference Phenomena in Solids”.
2. O.Kocharovskaya, Atomic and nuclear interference in solids, Physics Colloquium, Oklahoma State University, Department of Physics, March, 2005.
5. O.Kocharovskaya, Coherent control of the atomic optical and nuclear gamma-ray transitions in solids, April, 2008. Joint Colloquium of the Department of Physics, Harvard University and ITAMP.

J. Honors/Awards/Prizes for contract/grant employees (list attached):

   Fellow of the American Physical Society, 2005
   The Association of Former Students and Texas A&M University
   Distinguished Achievement Award in the Research, 2005
   Lecturer of the Distinguished Women Physicists Lecture series,
   Department of Physics, The University of Texas at Austin, 2005.
2. Elena Kuznetsova, graduate student, was awarded the Ethel Ashworth-Tsutsui Memorial Award for Research (the award is given to a female graduate student of College of Science of Texas A&M University). She received Ph D in 2005.

3. Petr Anisimov, graduate student, received a first prize in his topical group in the Texas A&M University Student Research Week 2005. He received his Ph D in 2008.

K. Total number of full-time equivalent graduate students and postdoctoral associates supported during this period, under this PR number: 5

Graduate students: 3
Postdoctoral Associates: 2
including the number of
Female Graduate Students: 1
Female Postdoctoral Associates: 1
the number of
Minority* Graduate Students: 0
Minority* Postdoctoral Associates: 0
including the number of
Asian Graduate Students: 0
Asian Postdoctoral Associates: 0

L. Total number of Ph D degrees obtained working on this project: 2

Elena Kuznetsova received Ph D in physics in 2005. The title of her thesis is: Atomic and nuclear interference phenomena and their applications.
Petr Anisimov received Ph D in 2008. The title of his thesis is: Quantum Coherence Effects in X-ray Optics”.

M. Training and Development:

Olga Kocharovskaya, the Principal Investigator of the project, was promoted to the Distinguished Professor position at the Department of Physics, Texas A&M University, in Spring 2007.

Roman Kolesov, who started working on the project as a postdoc, has moved to the University of Stuttgart, Germany, where he has joined the group of Prof. Wrachtrup as a Postdoctoral Research Associate. He continued his experimental work on quantum coherence and interference phenomena in solids at room temperature.

Elena Kuznetsova, who started working on the project as Ph D student received Ph D in 2005. The title of her thesis is: Atomic and nuclear interference phenomena and their applications. Then she continued working on the project as a postdoc till May, 2007. Then she moved to the University of Connecticut, where she has received a Postdoctoral Fellowship at the Department of Physics. She is also a Visiting Scientist at the Institute of Theoretical Atomic and Molecular Physics, Harvard-Smithsonian Center for Astrophysics since April 2007.
Petr Anisimov who started to work on the project as undergraduate student received Ph D in May, 2008. The title of his Ph D is: Quantum Coherence Effects in X-ray Optics”. He got an offer of the postdoc position from Prof. J. Dowling, Phys. Depart. Louisiana State University and plans to join this group in Oct.2008.

Chris O’Bryan, who started to work on the project as an undergraduate student in 2005 became a graduate student at TAMU and has continued working on the project in 2006-2008.

N. Other funding (list agency, grant title, amount received this year, total amount, period of performance)
Personnel supported by the Project:
(a) Farit Vagizov, Associate Research Scientist, 2005-2008
(b) Silviu Olariu, Assistant Research Scientist, 2005
(c) Roman Kolesov, Postdoc, 2005-2007
(d) Elena Kuznetsova, Graduate student in 2005 and then Postdoc in 2006, 2007
(e) Petr Anisimov, Graduate student, 2005-2008, graduated in May, 2008.

Collaborators
Rustem Schakhmuratov, Physical Technical Institution RAS, Kazan, Russia
Edgar Sadykov, Kazan State University, Russia
Yugeny Radeonychev, Institute of Applied Physics, RAS, Nizhny Novgorod, Russia
Rinat Akhmedzhanov, Institute of Applied Physics, RAS, Nizhny Novgorod, Russia
Yuri Roistovtsev, Department of Physics, Texas A&M University, USA
Marlan Scully, Department of Physics, Texas A&M University, USA
Jos Odeurs, Catholic Leuven University, Belgium

Part II

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   77843-4242
(b) Current Telephone Number:
   979-845-2012
(c) Cognizant AFOSR Program Officer:
   Anne Matsuura

(d) Program Objective:
This project is directed on the development of a new interdisciplinary field of basic research implying a combination and mutual fertilization of quantum optics and atomic physics with solid-state and nuclear physics. It includes experimental demonstration of the quantum coherence effects at the atomic and nuclear transitions in solids and laser manipulation of the gamma-ray transitions.

(e) Significant Results

I. Quantum Coherence Effects at Atomic Optical Transitions in Solids
I.1. Electromagnetically Induced Transparency (EIT) and quantum tomography in room temperature solids

We realized EIT and all-optical writing and reading of quantum coherence at ground-state Zeeman sublevels of Cr$^{3+}$-ion in ruby. For the first time such effect were observed in a solid at room temperature. Experiments were performed using single nanosecond optical pulses, pairs and trains of ultrashort pulses. Sharp drops in the resonantly induced fluorescence and transparency resonances are detected as the external magnetic field is varied.

At the room temperature the dominant contribution to the linewidths of the spin transitions in rare-earth and transition metal doped crystals typically originates from the Orbach relaxation. The last process can be greatly suppressed when the nearest excited electronic state is separated from the ground state by the energy essentially exceeding the typical phonon energies (of an order of a few hundred cm$^{-1}$). It results in sufficiently long decay time (of an order of microsecond) of the spin coherence allowing for realization of the atomic interference phenomena in solids at the room temperature.

Homogeneous broadening of the optical lines at the room temperature leads to simultaneous interaction of laser field with all impurity ions allowing usage of the major advantage of solids as compared to gases, the high density.

When the optical linewidth exceeds the hyperfine structure the coherent effects become the most pronounced under the action of the ultrashort optical pulses with the time interval between the pulses multiple to the period of the spin oscillations. An analytic solution of the density matrix equations for three-level system of A configuration interacting with an arbitrary number of pulses is obtained. Two subsequent ultrashort pulses lead to harmonic modulation of the EIT resonance produced by a single pulse. Such Ramsey interference was observed experimentally in ruby. It allows one to read out the complete state of atomic coherence including its amplitude and phase, i.e. to retrieve the state of a Q-bit. With increase of the number of ultrashort pulses, the periodic modulation develops into a sequence of narrow CPT resonances within the original broad level-crossing resonance.

Experimental demonstration of room temperature EIT and Ramsey fringes in solids opens the prospects for many practical applications of EIT including controllable optical delay lines, quantum information storage and processing, high sensitive magnetometry, interferometry, metrology, etc.

I.2. Experimental observation of EIT in new solids at the cryogenic temperature

In collaboration with a group of Dr. Rinat Akhmedzhanov from the Institute of Applied Physics, Nizhny Novgorod, Russia we have observed for the first time the electromagnetically induced transparency resonances in Nd$^{3+}$:LaF$_3$ and Pr$^{3+}$:LaF$_3$. So far most of the solid-state EIT experiments were done in Pr$^{3+}$:Y$_2$SiO$_5$ and nitrogen vacancy centers in diamond. The material studied in our work look promising for optical pulse storage based on EIT, if the NMR technique of hyperfine decoherence suppression is applied.

The dependence of the depth and width of the transparency “window” on the intensity of the drive field is analyzed. Our experimental results make it possible to measure the magnetic dipole-dipole interaction between pairs of Nd$^{3+}$ ions, separated by ~4 Å in LaF$_3$ and observe pair splitting in $^4G_{5/2}$ excited state and $^4I_{9/2}$ ground state.

I.3. Coherent suppression of excited state absorption in laser crystals and its application for building of tunable solid state lasers

A method of suppression of losses due to excited-state absorption (ESA) in laser crystals is proposed, based on the well-known phenomena of electromagnetically induced transparency (EIT) and coherent population trapping (CPT). Typical situations in which ESA prevents from lasing are considered. Theoretical analysis shows that using an additional driving laser field one can inhibit ESA and make lasing possible. On the basis of available spectroscopic information estimates for rare-earth ion doped laser materials are presented. They show feasibility of obtaining optical gain at parity-allowed 4f$^n$ -4f$^{n-1}$5d transitions of rare-earth ions in the ultraviolet
(UV) spectral region if ESA at an emission wavelength into the conduction band of a host material is suppressed.

We theoretically studied also the coherent population trapping effect induced by a train of short pulses in a three-level Λ-system with an upper state replaced by a continuum. Population transfer to the continuum can be totally suppressed provided that the splitting between the bound states is a multiple of the pulse repetition rate. Compared to the traditional case with only two laser fields coupling the bound states to the continuum, CPT with a pulse train allows one to avoid incoherent population losses due to interaction of the fields with "wrong" transitions, which results in only partial suppression of ionization. A method of suppression of excited-state absorption of pumping radiation in laser crystals, with pumping in the form of the pulse train, is proposed. As the particular examples the possibilities of ESA suppression in Ti$^{3+}$:YAlO$_3$ and Ce$^{3+}$:YAG using this technique are studied.

Coherent suppression of ESA opens the prospects for realization of the variety tunable solid state lasers including UV and VUV ranges

**I.4. Photonic Band Crystal Laser in UV range of frequencies**

A new type of photonic crystal (PC) laser, namely photonic band gap (PBG) laser, which generates radiation at the frequency falling to the PBG, is suggested. Propagation of generated radiation is provided either by the waveguide defect or by the natural waveguide formed in PC in the case of pseudogap. The basic idea is to suppress a radiative decay of the upper laser level at the lasing frequency due to PBG. Great potential of PBG lasers for the development of widely tunable continues wave (cw) and/or mode-locked femtosecond UV and VUV all-solid-state lasers is shown.

Suppression of radiative decay decreases the threshold pump intensity and provides favorable conditions for coherent control of the excited state absorption (ESA). It resolves also the laser gain dilemma, allowing for unique combination of the high emission cross-section and large population inversion. Different designs of 2D and 3D PBG laser are proposed.

**I.5. High-efficient resonant three-level mixing for THz generation**

We have theoretically shown that a coherently driven solid state medium can produce short intense coherent pulses of THz radiation. Optical crystals such as ruby and alexandrite can generate THz pulses with energies hundreds pJ - nJ at room temperature with ps pulse durations, and μJ at liquid helium temperature with ns pulse durations.

**II. Quantum Coherence Effects at Gamma-ray Nuclear Transitions**

**II.1. EIT with the single gamma quanta at the nuclear transitions**

We theoretically demonstrate that the interference effects (similar to electromagnetically induced transparency, which was widely studied earlier at electronic transitions in optics) may appear in transmission gamma-ray spectra at nuclear transitions under the condition of the nuclear level anti-crossing. dc coupling of the degenerate sublevels plays a role of an effective driving of the adjacent transition in the corresponding Λ system. An advantage of the gamma-ray nuclear transitions as compared to the optical atomic transitions is the large energy of the quanta which allows for EIT observation at the level of single photons. We have observed EIT at the nuclear transitions experimentally both in the crystal samples and in the powder of FeCO$_3$. This result is important from the fundamental point of view since it demonstrates an interference in absorption of a single photons due to the nuclear coherence. It opens also the prospects for realization of the concept of lasing without inversion at the nuclear gamma-ray Mossbauer transitions. Full suppression of the resonant absorption (if realized) would reduce the threshold incoherent pump by 3 orders of magnitude (defined by the typical ratio of the resonant to off-resonant cross-sections) reducing also heating of the sample and providing the possibility for pumping of active nuclei in the host lattice without destroying the conditions of Mossbauer effect, i.e. resolving a famous gamma-ray laser dilemma.

**II.2. Acoustically induced transparency in optically dense resonant medium**
It is shown that mechanical vibration (acoustic oscillation) of a solid medium along the propagation of multifrequency laser radiation enables one to control the resonant absorption. There exists an optimal spectral structure of the incident field dependent on vibration amplitude as well as the number and intensity of the frequency components that provides the full resonant transparency. Transparency of this kind is shown to appear also via adiabatic modulation of the atomic transition frequency by an external microwave field. A novel method of manipulation of Mosaic radiation (based on compression of frequency modulated radiation) which would result in generation of a train of pulses of coherent gamma radiation much shorter than the nucleus excited-state lifetime. is suggested.

II.3. Modifications of the Mossbauer absorption spectra under the action of laser radiation

The double optical-gamma resonance experiments (with driving laser field resonant to the electronic transition in an ion and a gamma-ray probe field resonant to the Mossbauer transition) was performed for the first time in Fe:MgO, Fe:Y₃Al₅O₁₂, Eu:CaF₂, Eu:Y₃SC₂Al₅O₁₂ and Eu₃Ga₅O₁₂.

Dramatic modifications of the Mossbauer spectra (such as 30% suppression of the resonant absorption and appearance of the prominent sidebands) were observed in the case of pulsed lasers excitation in Fe:MgO and Eu₃Ga₅O₁₂. In order to clarify the origin of this effect the new Mossbauer technique with microsecond time-resolution was developed for the first time. Namely, a series of time-domain experiments was performed, in which the Mossbauer spectra were collected only during 2.5 μs gate interval. This gate interval was swept from 5μs to 190μs over the time interval between the two laser pulses. It allowed us to observe oscillations of the center-of-mass position of the Mossbauer spectra. These oscillations were attributed to the vibrations of the sample induced by the laser pulse. This is the first observation of a laser-produced vibrations with the Mossbauer technique.

Small modifications of the Mossbauer spectra (such as a few % decrease of an amplitude of the resonant absorption, shift of line and increase of the absorption area) were observed in Fe:Y₃Al₅O₁₂, Eu:CaF₂, Eu:Y₃SC₂Al₅O₁₂ and Eu₃Ga₅O₁₂ in the case of cw laser excitation.

These effects were attributed to the electronic excitation leading to the nuclear transitions to the excited nuclear-electron state which has typically broader HF structure. Since the effects are small it is difficult to distinguish them from the spectra modification caused by laser heating of sample. Careful comparison of the results with those obtained under thermal heating of the samples was given.

These results open the prospects for development of the laser-Mossbauer spectroscopy.

II.4. Suppression of the inhomogeneous line broadening of the Mossbauer spectra

A new technique for narrowing of Mossbauer resonances in crystals is suggested. Similar to high resolution nuclear magnetic resonance spectroscopy, it uses a combined action of a continuous wave radio-frequency and dc magnetic field under a "magic-angle" condition. However, the condition itself is essentially different from the one known previously. Moreover, this technique suppresses the contribution of the dipole-dipole interaction to the energy of Mossbauer transition only (it does not suppress the contribution of the dipole-dipole interaction to the energy of individual levels). It works rather well even in the case of relatively strong dipole-dipole interaction.

This method opens the prospects for spectral resolution improvement in Mossbauer spectroscopy, for spectroscopic studies of the long lived nuclear transitions and for realization of lasing without inversion on the basis of such long-lived transitions which may be efficiently pumped.

II.5. Slowing down the single gamma-ray photons

Slowing down of the classical light pulses due to the doublet structure of the absorption light was observed recently in atomic vapors (Camacho R.M., et al., Phys.Rev.Lett., 98,043902,2007). We observed similar effect of slowing down of the single gamma-ray photons using the coincidence technique.

Co-57 Mössbauer source is a single-photon source which emits two successive single photons (122keV and 14.4keV) after the formation of an excited state (I=5/2). The 14.4 keV state (I=3/2) of Fe⁵⁷ provides a convenient source for observation of time evolution of a single photon propagating through a medium with doublet absorption lines. The formation of 14.4 keV state is indicated by a 122 keV gamma quanta in the decay of Co⁵⁷ excited state (I=5/2). So, in the experiment, the zero of time was determined by detecting with a counter the 122 keV precursor of the 14.4 keV quanta coming from a Co-57 source. A second, thinner scintillation
counter measured the time of detection of the 14.4 keV photon which had passed though a doublet line absorber. The schematic diagram of a time-of-flight experiment by the Mössbauer coincidence technique is shown below.

The probability amplitude of 14.4 keV photon has a sharply rising leading edge and an exponentially decaying tail (Fig.1.). The sharp leading edge is a result of causality: no photon can be emitted before the formation of an excited state. It has maximum probability when the excited state (3/2) is formed. Then the probability amplitude decays exponentially with time. The exponential decay of $^{57}$Co is presented in Fig.1. However an exponential decay is greatly modified when such radiation passes through the sample with the doublet absorption line structure (see Fig.2) due to interference of two absorption passes for the single photon. The estimates show that the gamma-ray photon slows down in this experiment about 1000 times.

These results are under the preparation for publication (F. Vagizov et. al., PRL, to be submitted). (All the results briefly outlined in the previous paragraphs have been published in the refereed journals. The references are given in paragraph A of this report).
Impact:

1. Electromagnetically Induced Transparency (EIT) and all-optical writing and reading of quantum coherence in room temperature solids demonstrated experimentally in the first time. It opens the prospects for many practical applications of EIT including controllable optical delay lines, quantum information storage and processing, high sensitive magnetometry, interferometry, metrology, etc.

2. The method for coherent suppression of excited state absorption in laser crystals based on EIT in the upper laser state was suggested. It opens the prospects for realization of the variety tunable solid state lasers including UV and VUV ranges.

3. High-efficient pulsed THz generation via resonant three-level mixing based on EIT in solids is predicted. The estimated efficiency is comparable to the existing methods at the room temperature and is essentially higher at the cryogenic temperatures.

4. EIT in gamma-ray range of frequencies at the nuclear transitions via the nuclear level crossing is demonstrated experimentally in the first time.

5. Modifications of the Mossbauer absorption spectra under the action of laser radiation demonstrated experimentally in the first time. It opens the prospects for development of the laser-Mossabuer spectroscopy.

6. Acoustically induced transparency is predicted. It makes possible generation of a train of pulses of coherent gamma radiation with the pulse duration much shorter than the nucleus excited-state lifetime.

7. The method for suppression of the inhomogeneous line broadening of the Mossbauer spectra is suggested. It opens the prospects for spectral resolution improvement in Mossbauer spectroscopy, for studies of the long lived nuclear transitions and realization of inversionles amplification at such long-lived transitions which may be efficiently pumped.

8. Slowing of the gamma-ray photons due to the absorption line doublet is demonstrated in the first time.