様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科研費 KAKENHI

科学研究費助成事業　　研究成果報告書

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研究課題名（英文）Ultrabright x-ray harmonic comb by relativistic plasma singularity

研究代表者
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研究成果の概要（和文）：　我々が発見し、理論を構築した高強度レーザーがガスプラズマ中に作る特異点からの高輝度のコヒーレント軟X線生成に関する研究において、今回、この理論が予言する点状のX線源（特異点）を初めて計測した。また、この理論を一般化し、特異点放射のバースト的増強BISERという概念を創出した。関西研にあるレーザーの品質を大きく向上させ、この高品質化がX線の高輝度化に大きな影響を持つことを確かめた。また、プラズマ密度の調整によりX線発生点を制御した。
今後この技術を発展させていけば、小型で、高輝度・コヒーレントなアト秒X線源が実現でき、材料科学や生命科学に貢献できると考えられる。

研究成果の学術的意義や社会的意義
BISER x-ray yield enhancement and tuning open the way to a compact, ultra-bright, spatially and temporally coherent x-ray source for pumping, probing, imaging, and atto-science.
The developed diagnostics and techniques are useful for applications of high-power lasers and are adopted by the community.

研究成果の概要（英文）：Previously I experimentally discovered bright coherent soft x-rays (high harmonics) using 10-200 Terawatt [1 TW=10 W] 30-50 femtosecond [1 fs=10 5s] lasers focused to gas targets. We proposed a theory of x-ray generation by relativistic plasma singularities.
Here I tested and confirmed the theory by measuring point-like x-ray sources (the singularities). We generalized the theory and formulated a Burst Intensification by Singularity Emitting Radiation (BISER) concept. I experimentally demonstrated strong dependence of the x-ray brightness on laser quality. I significantly enhanced our J-KAREN-P laser quality. I for the first time experimentally demonstrated coherent x-ray generation tuning using plasma density tailoring.
My results open a way to compact bright coherent attosecond [1 as=10 8s] x-ray source demanded by university and industry labs for coherent x-ray imaging and other applications in material and life sciences.

研究分野：数物系科学

キーワード：コヒーレントX線放射　超高強度レーザー　レーザー光とプラズマの相互作用　相対論的レーザープラズマ　相対論的電子スパイク　レーザー光の高次高調波　BISER

１．研究開始当初の背景 Background

Bright coherent x-ray sources are necessary for fundamental research and applications in life sciences, material sciences, nanotechnology, etc. There are large-scale accelerator-based X-ray Free Electron Lasers (XFELs) and compact laser-based sources (x-ray lasers, atomic high-order harmonics, relativistic flying mirrors, etc.). Great advantages of the laser-based sources are their accessibility at university and industry laboratories and ultrashort durations down to tens of attoseconds (1 as = 10^{-18} s). However, fundamental limitations of common techniques make it overwhelmingly difficult to implement compact bright coherent x-ray source with keV photon energies.

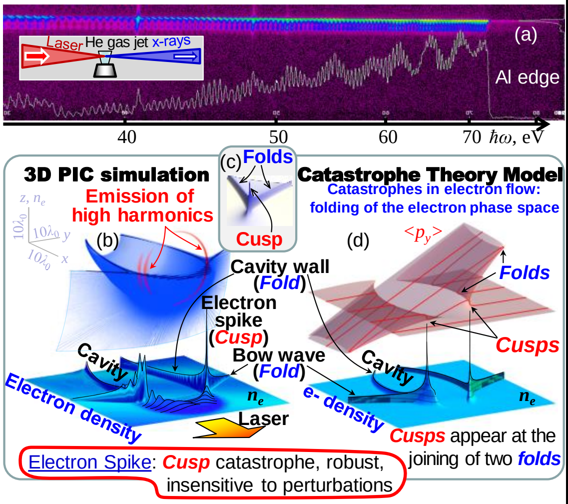

Fig. 1.(a) Typical single-shot harmonic spectrum (experiment, J-KAREN laser); (inset) experimental setup. (b) 3D PIC Simulation: Relativistic singularities (density spikes) and harmonics emission (red arcs, $\omega \geq 4\omega_0$); for higher harmonic orders, the arc length reduces resulting in *two point-like sources*. Inset (c): Discrete particle model result. (d): *Catastrophe Theory Model*: folds of the *phase space* lead to *catastrophes* (*density singularities*).

We have discovered a new regime of high-order harmonic generation by high-power (10-200 TW, 1 TW = 10^{12} W) relativistic-irradiance ($>10^{18}$ W/cm^2) femtosecond lasers (~30-50 fs, 1 fs = 10^{-15} s) focused to gas jet targets, Fig. 1 (a) [Pirozhkov, invited presentation at CLEO:2013 QW1A.3 (San Jose, USA); Pirozhkov *et al. PRL* **108**, 135004 (2012), 朝日新聞平成24年3月15日 p25, Pirozhkov 若手研究(B) 23740413 平成23-25]. Comb-like spectra with hundreds of even & odd harmonic orders, reaching the 'water window' spectral range with the photon energy of up to 500 eV, are generated by either linearly or circularly polarized pulses. The photon number is scalable and at 120 TW laser power is ~10^2 times greater than at 10 TW. A 120 eV harmonic contains up to 4×10^9 photons (90 nJ energy), which is very difficult to achieve with other techniques. Using Particle-In-Cell (PIC) simulations and mathematical catastrophe theory, we have introduced a new mechanism of harmonic generation by extremely sharp, structurally stable, oscillating electron spikes (density singularities) situated off-axis, at the joints of the wake wave and bow wave boundaries, Fig. 1 (b).

２．研究の目的 Purpose

Because the harmonics are coherently emitted by the oscillating electron spikes, it is of paramount importance to understand how to control the formation of the spikes, their shape, and particle number in them.

The purpose of this study was to find out how to control the spike properties and enhance the harmonic generation process using the following two methods:

(C) *via plasma density profile **C**ontrol,*

(S) *via shaping of the laser focal **S**pot.*

The control of the harmonics generation process had not been previously demonstrated. The parameters of the XUV and x-ray pulses were expected to exceed those which can be demonstrated using previously known accessible compact coherent x-ray sources.

３．研究の方法 Methods

Laser-plasma interaction regime

The Project was based on the new harmonic generation regime discovered by us earlier. The most important parameters are: the dimensionless laser amplitude $a_0 = eE_0/m_e c\omega_0$, irradiance $I_0 = a_0^2\times1.37\times10^{18}$ W/cm^2/(λ_0([μm])^2, ratio of plasma density n_e to critical density $n_{cr} = m_e\omega_0^2/4\pi e^2 \approx 1.1\times10^{21}$ cm^{-3}/(λ_0[μm])^2, and ratio of the laser power P_0 to relativistic self-focusing threshold $P_{SF} = P_c n_{cr}/n_e$. Here $P_c = 2m_e^2c^5/e^2 \approx 17$ GW, e and m_e are the electron charge and mass, c is the speed of light, and λ_0, ω_0, and E_0 are the laser wavelength, frequency, and peak electric field.

In our harmonics generation regime $a_0 >> 1$, i.e. $I_0 >> 10^{18}$ W/cm^2 and $n_e \sim 10^{19}$ to 10^{20} cm^{-3}, i.e. $n_e/n_{cr} \sim 10^{-2}$ to 10^{-1}. In this case $P_{SF} \sim (10$ to $100)\times P_c = (0.2$ to $2)$ TW. For our multi-terawatt laser pulses, $P_0 >> P_{SF}$, which leads to relativistic self-focusing and increases a_0.

The coherently emitted photon number is proportional to the number of electrons squared: $N_{ph} \sim N_e^2$, while the maximum harmonic order scales as $n_H \sim a_0^3 \sim I_0^{3/2}$, according to classical electrodynamics [J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1998)].

Problems to be solved

Particle-In-Cell (PIC) simulations [Esirkepov 基盤研究(C)25390135 平成 25-27] showed that a tight focal spot and a relatively dense plasma ($n_e \sim 10^{19} - 10^{20}$ cm^{-3}) are advantageous, since the electron spikes form closer to the laser axis and oscillate stronger while their density is higher compared to low-density case. However, typical gas jets have sub-mm gradual density rise, i.e. much longer than the plasma wavelength $\lambda_p \approx \lambda_0(n_{cr}/n_e)^{1/2} \sim 10$ μm. The laser evolution in such long plasma results in filamentation, Fig. 2, and inefficient harmonic generation.

Further, the filamentation develops much faster if the laser focal spot quality is not high enough.

Methods to solve the problems

In this Project, I controlled the laser irradiance and electron spike formation and position via the laser focal spot shaping and plasma profiling. This allowed enhancement of the harmonic x-ray yield and significant stability improvement.

In particular, I implemented:

(C) A gas jet target with tailored (**C**ontrolled) density profile and density up to $\sim 10^{20}$ cm^{-3}.

(S) A tight, high-quality laser **S**pot.

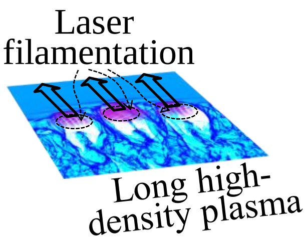

Fig. 2. Laser filamentation in a usual high-density gas jet. The blue scale shows the electron density.

４．研究成果 Results

BISER concept [⑤]

Our original theory and model included electron plasma flow and plasma waves, and catastrophe theory stating that these waves produced electron density singularities, which in turn coherently emitted high-frequency electromagnetic radiation. During this Project, we recognized that this mechanism is much more general. We formulated it in the most general way, where *elementary emitters* of any *media* exhibiting *multi-stream flow* form *structurally stable singularities* which produce bright *traveling waves*. The singularity emission is much brighter than the surrounding medium due to high concentration and synchronism bringing constructive interference. If the momentum distribution of elementary emitters smoothly depends on their coordinates, this emission is *spatially* and *temporally* coherent and scales as number of emitters *squared*. In addition, the brightness increases if the singularity moves at a relativistic speed. We called this process

Burst Intensification by Singularity Emitting Radiation (BISER) [⑤], Fig. 3.

Our experiments corresponded to a particular implementation of BISER where the medium was relativistic plasma, elementary emitters were electrons, and the traveling wave was electromagnetic. We also predicted [⑤] that similar process should happen in space plasmas; in particular, the BISER sources can be progenitors of Gamma Ray Bursts (GRBs) and Fast Radio Bursts (FRBs). In addition, strong gravitational waves can also be emitted via the BISER mechanism.

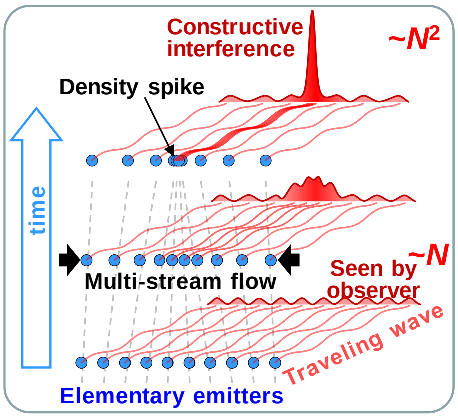

Fig. 3. Formation of a *singularity* (density spike) and corresponding *burst intensification* of emitted radiation: Multi-stream converging flow consisting of elementary emitters eventually forms a *density spike* capable of generating a burst of coherent emission [⑤].

BISER theory test and confirmation [⑤]

The BISER theory explained previous results; in addition, as all good theories do, it also predicted several particular properties, which were not observed experimentally earlier:

(i) The source of coherent x-ray emission is tiny.

(ii) Due to diffraction, there is off-axis emission.

(iii) For linear laser polarization, the x-ray source consists of two singularities (double source).

To test these theoretical predictions, I performed dedicated experiment with the J-KAREN laser (KPSI, QST). The experimental schematic is shown in Fig. 4. Linearly polarized pulses from the J-KAREN laser (20 TW, 35 fs, 10^{19} W/cm^2) irradiated a helium gas jet ($n_e \approx 3\times10^{19}$ cm^{-3}) and produced *density singularities* (the *Cusps*) situated on both sides of the axis, in the laser polarization plane. These *singularities* generated bright coherent x-rays via the BISER mechanism into a wide range of angles. The soft x-ray pulse energy was up to 100 nJ in the 60 to 100 eV spectral range, corresponding to ~10^{10} photons. I used a normal-incidence multilayer x-ray mirror with aperiodic coating providing broadband reflectivity of ~11% in the 60 to 100 eV spectral range [⑧] to image the BISER x-ray source onto a detector with the magnification M = 5.5. To avoid resolution limitation caused by a CCD pixel size, I employed a LiF crystal detector [Faenov *et al.* *Opt. Lett.* **34**, 941 (2009).]

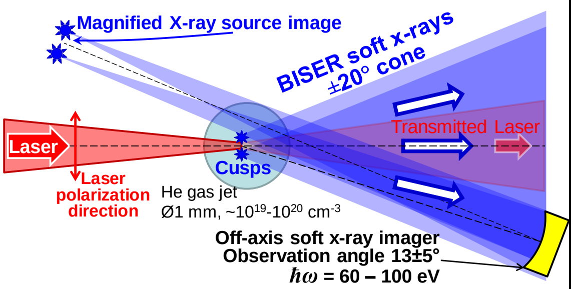

Fig. 4. Experiment schematic. Laser pulse irradiates helium gas jet producing *density singularities (Cusps)* emitting bright coherent x-rays via the BISER mechanism with a wide angular distribution. An x-ray imaging mirror intercepts off-axis radiation (8° to 18° off-axis) and images with magnification the source onto a LiF crystal detector [⑤].

The resulting single-shot image is shown in Fig. 5 (a) [⑤]. It confirms all three predictions of the theory: indeed, the x-rays were generated by tiny (sub-μm) sources *(i)*; the imaging mirror was situated off-axis and accepted the angles from 8 to 18° *(ii)*; the source consisted of two points situated approximately in the laser polarization plane *(iii)*. With imaging spectrograph, I also obtained double x-ray source; its spectrum is shown in Fig. 5 (b) [⑤].

Importantly, the modeling using physical optics propagation and analysis of fringes visible in Fig. 5 (a) showed that the individual sources were much smaller, probably not exceeding 100 nm, while the sub-μm images resulted from aberrations of the spherical mirror used for the imaging. A sub-100 nm source is consistent with PIC simulations, which show 14 nm full-width at half-maximum (FWHM) electron density spike size [⑤]. In addition, the simulations showed that the x-rays were emitted as an attosecond pulse train with the individual pulse duration of ~170 as, which was close to the transform limit of 150 as. Thus, my experimental measurements and simulations allowed to estimate the peak spectral brightness. At the wavelength of 18 nm it was 10^{27} photons/mm^2 ·mrad^2·s in 0.1% bandwidth, which is *5-6 orders of magnitude brighter than synchrotrons* and 2-3 orders of magnitude lower than that provided by the brightest available soft x-ray sources, free-electron lasers, operating at similar wavelengths [Ackermann *et al.* *Nat. Photon.* **1**, 336 (2007)]; the advantage of the BISER source is that it is much more compact.

BISER dependence on laser spot quality [③]

In accordance with the Project Purpose, I performed experimental study of the dependence of the BISER process on the laser focal spot. I found a very strong dependence on the spot quality, Fig. 6 [Refs. ③,④]. In fact, even moderate improvement of the spot quality resulted in dramatic enhancement of the BISER x-ray yield. This was especially important for achieving higher photon energies and reaching the "water window" spectral range, where BISER x-rays were only generated with high quality spots.

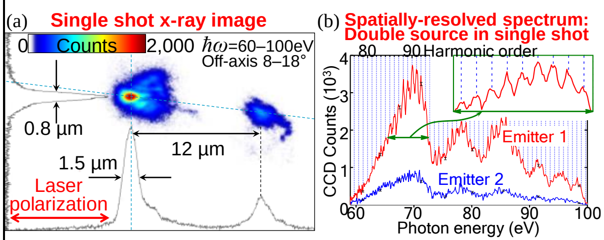

Fig. 5. Single-shot image (a) and spatially-resolved spectrum (b) of the BISER x-ray source (Experiment) [⑤].

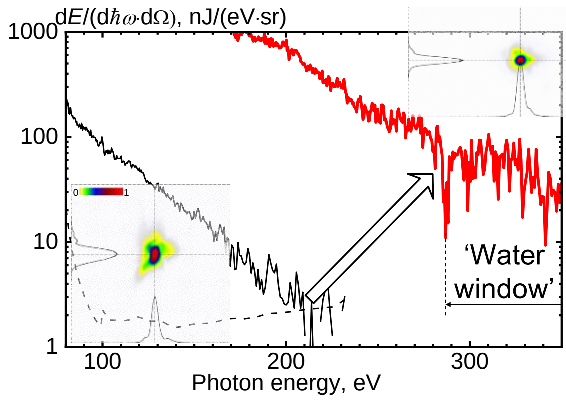

Fig. 6. Optimum BISER spectra corresponding to the two different focal spot shapes (Experiment) [③].

Laser requirements for BISER x-ray generation [③]

In order to quantify laser requirements for the efficient BISER x-ray generation, I studied the dependence of laser spot shape on various imperfections, including the wavefront distortions and spatiotemporal couplings. Summarizing [③,④], the rms wavefront error should be smaller than ~100 nm (~λ/8), which is close to the Maréchal criterion of a diffraction-limited beam. Further, the angular dispersion should be kept considerably smaller than the diffraction divergence, i.e., μrad level for 100 to 300-mm beam diameters. The corresponding angular chirp should not exceed 10^{-2} μrad/nm for a 40-nm bandwidth. For typical high-power laser compressors, this requires ~10 μrad grating alignment accuracy.

(S) Enhancement of the J-KAREN-P laser [⑥]

In order to achieve the strict requirements formulated above, I designed and constructed a very precise beamline alignment system, performed careful tuning of the beam wavefront with a recently installed deformable mirror, made compressor alignment with the accuracy of ~10 μrad for all 12 grating angles (4 gratings, 3 angles each), and tuned the spectral phase to achieve clean pulse compression. As a result, I achieved very high focal spot quality with the irradiance about 50% of the diffraction limited one, Fig. 7 (a); I note that the *focal spot measurement was performed with the full bandwidth and nearly full-power (300 TW) beam* attenuated by 10 high-quality wedges; the surface quality of all the wedges in their holders was tested with an interferometer. I also achieved nearly bandwidth-limited pulse compression, with the peak power ~90% of bandwidth-limited pulse, Fig. 7 (b).

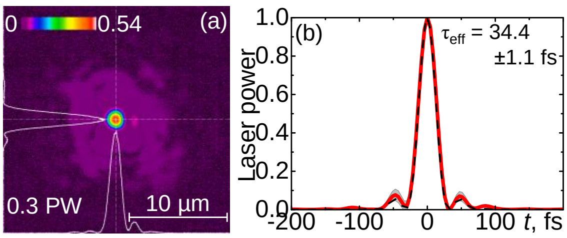

Fig. 7. J-KAREN-P laser spot (a) and pulse (b) [⑥]; the dashed line in (b) is the bandwidth-limited pulse.

(C) Plasma density tailoring

In accordance with the Project Purpose and method proposed in the Application, I performed a successful experiment on the BISER generation, where I made a plasma density bump, Fig. 8. By moving a translation stage, the position of this bump moved accordingly. The source position and properties, in turn, depended on the bump position, Fig. 9.

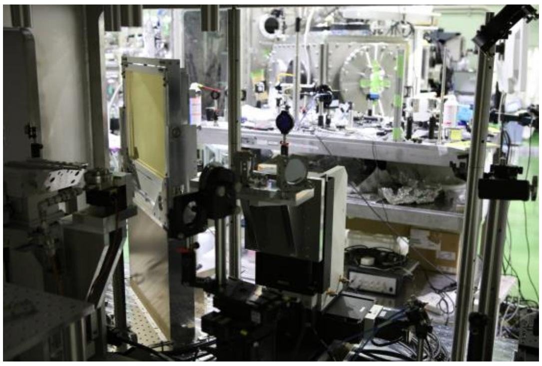

Fig. 8. Target chamber photo.

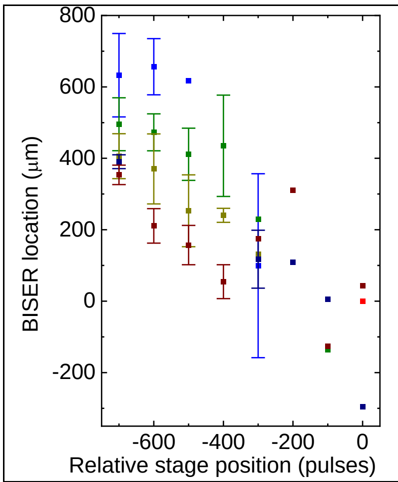

Fig. 9. BISER location vs. stage position at different plasma densities (experiment with the J-KAREN-P laser).

The results of this experiment have been presented at domestic and international conferences, including invited presentation at OPIC-HEDS (2018) (No. 1 in the presentation list below). The paper is now under preparation.

Significance and Future outlook

Multi-terawatt lasers necessary for BISER x-ray source are now commercially available and installed at many universities and even industrial labs. The gas jet targets are easily accessible, self-replenishable, debris-free, and suitable for repetitive operation. My demonstration of BISER x-ray yield enhancement with the laser quality and x-ray source properties tuning using the plasma density tailoring opens the way to a *compact, ultra-bright, spatially* and *temporally coherent* x-ray or XUV source. This will impact many areas requiring a bright compact x-ray or XUV source for pumping, probing, imaging, or attosecond science.

The diagnostics and experimental techniques developed by me during this Project are very useful for experiments with and applications of high-power lasers and will be adopted by future university and industry labs employing such lasers.

Apart from x-ray source applications, the BISER concept is expected to become a cornerstone of new generation of *Laboratory Astrophysics* studies, where ultra-bright cosmic emitters, such as GRBs and FRBs, can be studied in limited similarity experiments with relativistic laser plasma.

５．主な発表論文等 Main publications

〔雑誌論文〕（計　10　件） **Journal papers**

①. H. Kiriyama, A. S. Pirozhkov (+13 authors), "High-contrast high-intensity repetitive petawatt laser," *Opt. Lett.* **43** (11), 2595-2598 (2018) (Refereed). DOI: 10.1364/OL.43.002595.

②. M.Kando*, T.Esirkepov*, J.Koga*, A.Pirozhkov*, S.Bulanov*(all authors contributed equally), "Coherent, Short-Pulse X-ray Generation via Relativistic Flying Mirrors," *Quantum Beam Sci.* **2** (2), 9 (24 Apr 2018) (Refereed). DOI: 10.3390/qubs2020009.

③. A.S.Pirozhkov (+21 author), "Laser Requirements for High-Order Harmonic Generation by Relativistic Plasma Singularities," *Quantum Beam Science* **2** (1), 7 (20 Mar 2018) (Refereed). DOI: 10.3390/qubs2010007.

④. A.S.Pirozhkov (+21 authors), "High-Order Harmonic Generation by Relativistic Plasma Singularities: the Driving Laser Requirements," *Springer Proc. Phys.* **202**, 85-92 (24 Feb 2018) (Refereed). DOI: 10.1007/978-3-319-73025-7_13.

⑤. A.Pirozhkov*, T.Esirkepov*(contributed equally) (+21 authors), "Burst intensification by singularity emitting radiation in multi-stream flows," *Scientific Reports* 7, 17968 (21 Dec 2017) (Refereed). DOI: 10.1038/s41598-017-17498-5.

⑥. M. Alkhimova, A. Faenov, I. Skobelev, T. Pikuz, M. Nishiuchi, H. Sakaki, A. S. Pirozhkov (+13 authors), "High resolution X-ray spectra of stainless steel foils irradiated by femtosecond laser pulses with ultra-relativistic intensities," *Opt. Express* **25** (23), 29501-29511 (13 Nov 2017) (Refereed). DOI: 10.1364/OE.25.029501.

⑦. A.S.Pirozhkov (+15 authors), "Approaching the diffraction-limited, bandwidth-limited Petawatt," *Opt. Express* **25** (17), 20486-20501 (21 Aug 2017) (Refereed). DOI: 10.1364/OE.25.020486.

⑧. A.S.Pirozhkov, E.N.Ragozin, "Aperiodic multilayer structures in soft X-ray optics," *Physics - Uspekhi* **58** (11), 1095-1105 (Nov 2015) (Refereed). DOI: 10.3367/UFNe.0185.201511e.1203.

⑨. H. Kiriyama, M. Mori, A. S. Pirozhkov (+21 authors), "High-Contrast, High-Intensity Petawatt-Class Laser and Applications," *IEEE J. Sel. Top. Quantum Electron.* **21** (1), 1601118 (2015). doi: 10.1109/JSTQE.2014.2336774.

⑩. A.S.Pirozhkov (+39 authors), "High order harmonics from relativistic electron spikes," *New J. Phys.* **16**, 093003 (4 Sep 2014) (Refereed). DOI: 10.1088/1367-2630/16/9/093003.

〔学会発表〕（計　25　件） **Presentations**

1. A. S. Pirozhkov *et al* [Invited] "Control of Burst Intensification by Singularity Emitting Radiation (BISER) with density jump," Optics & Photonics Int. Congress, Int. Conf. High-Energy Density Sciences, 23-27 Apr 2018, Yokohama, Japan.

2. A. S. Pirozhkov *et al.*, [Invited] "Point-like coherent soft x-ray sources and challenges for optics," 14th Symp. X-ray Imaging Optics, 29-30 Nov 2017, Tsukuba University, Tsukuba, Japan.

3. A. S. Pirozhkov *et al* [Invited] "Point-like x-ray sources," XXI Int. Symp. "Nanophysics and Nanoelectronics" 13-16 Mar 2017, Nizhny Novgorod, Russia.

4. A. S. Pirozhkov *et al* [Invited] "High-order harmonic generation by relativistic plasma singularities," 15th Int. Conf. X-Ray Lasers, 22-27 May 2016, Nara Kasugano International Forum, Nara, Japan.

5. A. S. Pirozhkov *et al* [Invited] "High-order harmonics from singularities of relativistic plasma," Int. Symp. "Topical Problems of Nonlinear Wave Physics" NWP-2014, 17-23 July 2014, Volga River, Russia.

〔その他〕 **Others**

1. QST Press release on paper [⑤] "プラズマの特異点からの強力な軟Ｘ線バーストを発見 ―宇宙でも起こり得る普遍的な新しい放射機構の提案―" http://www.qst.go.jp/information/itemid034-003332.html

2. APS-TV film about KPSI, including my short talk about the discovery of the high-order harmonic generation by relativistic electron spikes: https://www.youtube.com/watch?v=UBK86zX_LCk

６．研究組織 Research organization

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(2)研究分担者 Co-Investigators: No

(3)連携研究者 Collaborating researchers: No

(4)研究協力者 Research collaborators: No