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物理学第一分野

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# Quantum spin transport using ultracold ytterbium atoms with orbital degrees of freedom

Quantum Optics Group    Koki Ono

**Abstract** We report on quantum spin transport experiments using ytterbium atoms in the ground state  $^1S_0$  induced by the localized atoms in the metastable state  $^3P_0$ . We succeeded in the observation of the interorbital spin-exchange dynamics, and the demonstration of the novel quantum spin transport.

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Ultracold atomic gases in an optical lattice have reproduced paradigmatic models of condensed matter physics such as the Hubbard model, demonstrating the ability of quantum simulation [1]. Recently, alkaline-earth-like atoms have attracted considerable research interest due to the existence of the long-lived metastable states  $^3P_0$  and  $^3P_2$  coupled to the ground state  $^1S_0$ . This intriguing internal structure can provide the platform for the quantum simulation with orbital degrees of freedom [2].

We present two experiments with the two-orbital system consisting of the  $^1S_0$  atoms and the  $^3P_0$  atoms. The first work is motivated by the quantum simulation of the Kondo effect, which is a prominent example of the quantum many-body phenomenon highlighting a relevant role of the orbital and spin degrees of freedom. Several schemes for cold-atom quantum simulation of the Kondo effect have been proposed. In particular, the two-orbital system using the fermionic isotope  $^{171}\text{Yb}$  has received much attention as a promising candidate for the quantum simulation of the Kondo effect since our high-resolution laser spectroscopy reveals that the spin-exchange interaction between the  $^1S_0$  atom and the  $^3P_0$  atom is antiferromagnetic [3], one of the key ingredients for the emergence of the Kondo effect. In order to realize the Kondo system, we implement the two-orbital lattice system consisting of a one dimensional (1D) state-dependent optical lattice (SDL) with a wavelength of 650.7 nm and a 2D state-independent optical lattice with a wavelength of 759.4 nm, which is called the magic wavelength. The 1D SDL gives a strong confinement to the  $^3P_0$  atom and is superimposed along the axis of the 2D array of the tube traps created by the 2D magic optical lattice, realizing the quasi (0+1) D system where the  $^1S_0$  atom and the  $^3P_0$  atom are itinerant and localized, respectively. We successfully observe the interorbital spin-exchange dynamics of  $^{171}\text{Yb}$  in the two-orbital lattice system, opening the route to the quantum simulation of the Kondo effect.

Secondly, we report on the demonstration of a novel type of localized-impurity-induced quantum spin transport using the two-orbital system. So far, most studies of quantum transport in condensed matter systems have focused on a nonequilibrium steady state coupled to external incoherent reservoirs. Ultracold atomic gases allow one to focus on intrinsic transport properties because the system can be regarded as isolated from the external environment. In order to emulate a mesoscopic system with the cold atoms, however, a high degree of controllability to engineer a narrow conducting channel could be required [4]. Alternatively, the system consisting of a multi-component Fermi gas and a localized impurity provides a more feasible scheme of the quantum transport experiment, where the current can be created in the spin space by introducing the spin-dependent interaction with the impurity [5]. In this work, we observe the decoherence of the nuclear spin precession of the  $^1S_0$  atoms in the presence of the localized  $^3P_0$  atoms, exhibiting the quantum spin transport induced by the localized impurities. Our work paves the way to the quantum simulation of quantum transport, which is called atomtronics.

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# Turbulent relative dispersion in two-dimensional energy inverse-cascade turbulence

Fluid Physics Group    Tatsuro Kishi

**Abstract** Turbulence quickly disperses substances, which is one of the basic features of turbulence. We propose the scaling relation of statistics on relative separation of particle pairs via two-time Lagrangian velocity correlation function and confirm it by direct numerical simulations of two-dimensional energy inverse-cascade turbulence.

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Transport phenomena are ubiquitous in nature. Especially, turbulence efficiently transports and promptly disperses substances such as pollutants, chemical and biological agents. The turbulent diffusion is a very important phenomenon with various applications from cosmology, meteorology and biology to environmental problems, chemistry and engineering. Furthermore it is the extraordinary capability to disperse substances that characterizes turbulence itself. Therefore, it is also significant for fundamental physics to understand the nature of turbulence.

The concentration of substances exhibits complex structures over broad scales of time and space because of the nonlinearity of turbulent flows. Thus, one-particle statistics such as the root-mean-square distance is insufficient to understand the multiscale nature of turbulent diffusion. Space-time correlations of particle positions are intrinsic characteristics. The most fundamental one is the correlation between a particle pair [1].

We consider the simplest case, that is, turbulent diffusion of Lagrangian particles, which have no inertia, no back influence on the fluid velocity and no effects of molecular diffusion. Furthermore, we only deal with the statistically stationary, homogeneous and isotropic turbulence. In such a case, we can deal with the relative distance,  $r(t)$  instead of each position of particle pairs,  $\mathbf{x}_1(t)$  and  $\mathbf{x}_2(t)$ , and thus the number of variables is reduced from particle-pair statistics. We call the statistics of relative distance turbulent relative dispersion.

The turbulent relative dispersion can be dimensionally analyzed by Kolmogorov's phenomenology, which successfully describes the Eulerian velocity statistics of turbulence, though they are Lagrangian statistics [2]. The square mean separation of particle pairs is given by  $\langle r^2(t) \rangle \simeq g\varepsilon t^3$ , exhibiting super-diffusivity. Here  $\varepsilon$  is the energy dissipation rate of turbulence and  $g$  is a universal constant called the Richardson constant. However, there is no consistent theoretical derivation from the Navier-Stokes equations, and also there is no clear observation by both laboratory and numerical experiments [3].

We discuss the turbulent relative dispersion in terms of the initial separation dependence via the two-time Lagrangian velocity correlation function  $C^L(t_1, t_2)$  as shown in Fig.1, which is directly related to the mean square separation of particle pairs as follows:

$$\langle r^2(t) \rangle \simeq r_0^2 + \int_0^t \int_0^t C^L(t_1, t_2) dt_1 dt_2, \quad (1)$$

where  $r_0$  is an initial separation of particle pairs. Here  $C^L(t_1, t_2) \equiv \langle \delta \mathbf{v}(t_1) \cdot \delta \mathbf{v}(t_2) \rangle$ , where  $\delta \mathbf{v}(t)$  is a relative velocity between a particle pair. We propose that the two-time Lagrangian velocity correlation function has a self-similar form with scaling exponents which can not be determined by the dimensional analysis. Furthermore, we confirm the proposed scaling relation by direct numerical simulation of two-dimensional energy inverse-cascade turbulence. We discuss finite Reynold number effects on the scaling exponents and show the improved scaling law for the turbulence relative dispersion.

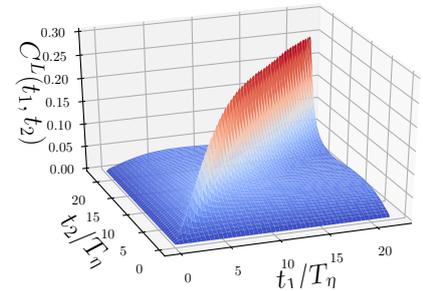


Fig. 1: Two-time Lagrangian velocity correlation function,  $C^L(t_1, t_2)$ .  $T_\eta$  is a time scale of energy dissipation.

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# Transport properties of photoexcited carriers and excitons in ultrapure diamond

Solid State Spectroscopy Group      Kazuki Konishi

**Abstract** We have investigated transport properties of photoexcited carriers and excitons in a semiconductor diamond of extremely high purity. A clear correlation between the exciton diffusion coefficient, lifetime, and residual strain was found from 4 to 300 K for the first time. We propose an extended surface recombination model, which successfully explains the temperature dependence of the carrier and exciton lifetimes and luminescence quantum efficiency of excitons.

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An electron-hole system in a photoexcited semiconductor exhibits various complexes of carriers, such as excitons, polyexcitons, and electron-hole droplets [1]. Even in a simple mixture of free carriers and excitons, the fraction of exciton density to the total carrier density largely affects the transport properties. In the recent study using a monolayer semiconductor [2], the density dependence of the diffusion coefficient was investigated, revealing intriguing phenomena that occur due to many-body interactions. However, measurements at low temperatures have not been performed because trapping at defects becomes prominent. In order to elucidate changes in the transport properties as a function of temperature, we focus on diamond, in which the excitons are stabilized owing to the large exciton binding energy. Thanks to the high-quality diamond samples available today, relationships between diffusion, lifetime, and residual strain have been systematically elucidated without impurity trapping.

In this study, we used three single crystals of ultrapure diamond (Element Six, Ltd.) to ensure that the exciton decay was not affected by trapping at impurities. Figure 1 shows the temperature dependence of the exciton lifetime measured at different excitation spots by the time-resolved photoluminescence method [3]. By combining information gained from the diffusion coefficient measurements, exciton fraction determined by terahertz time-domain spectroscopy [4], and strain distribution evaluated by birefringence imaging, we revealed that excitonic diffusion has a significant impact on the lifetime in diamond. We proposed a surface recombination model extended for nonradiative recombination at dislocations after diffusion (see the Inset), and the temperature dependence was successfully explained from 4 to 300 K (solid lines). We also showed that this model enables the prediction of internal quantum efficiency of exciton luminescence, which is a crucial parameter for the optoelectronic applications of diamond toward realization of efficient devices working in the deep ultraviolet region useful for denaturation of viruses.

Furthermore, we evaluated the carrier lifetime and mobility by the time-resolved cyclotron resonance and time-of-flight (ToF) methods on the same samples from 2 to 300 K [5]. We demonstrated that the temperature dependent electron lifetime is also explained by the simple model proposed above. Therefore, our findings are generally applicable to a wide range of excitonic materials in which excitons and free carriers coexist.

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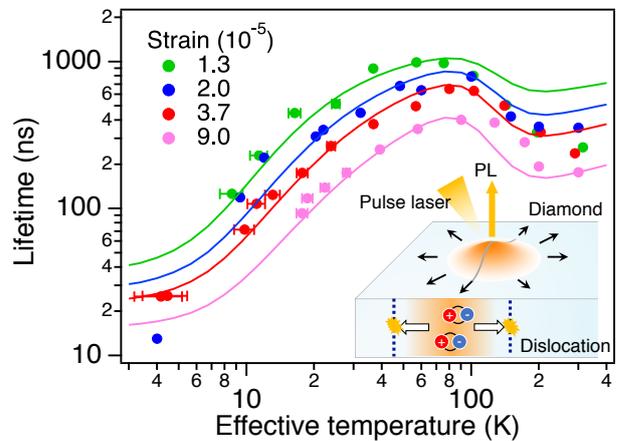


Fig. 1 Exciton lifetimes measured at different spot positions (circles) in comparison with values calculated according to the surface recombination model (solid lines). Inset: Schematic of the exciton diffusion measurements and recombination at dislocations. Adapted from Ref. [3].

# Unconventional quantum oscillations and thermal metallic state of charge-neutral fermions in Kondo insulator YbB<sub>12</sub>

Quantum Condensed Matter Group      Yuki Sato

**Abstract** The presence of Fermi surface, which is an unambiguous signature of metals, is manifested by quantum oscillations, and finite linear  $T$ -dependent terms in specific heat and thermal conductivity. Here, we report notable observations of those “metallic” behaviors in Kondo insulator YbB<sub>12</sub>.

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Strong electronic correlation in many-body systems often results in a wide variety of ground states, such as heavy Fermi liquid, unconventional superconductivity, and strongly correlated insulator. Understanding of those exotic electronic phases and excitations (i.e. emergent quasiparticles) from these ground states is the most fundamental and important subject in condensed matter physics. Kondo insulator (KI) is a typical example of such strongly correlated insulators, where hybridization between conduction  $c$ -electron and localized  $f$ -electron opens up charge gap across Fermi level. KIs have recently attracted much interest due to the observation of quantum oscillations (QOs) in magnetization of SmB<sub>6</sub> [1], which is a conflicting evidence of the Fermi surface of metals.

Here we present high field torque and transport measurements of another KI YbB<sub>12</sub>. Remarkably, YbB<sub>12</sub> exhibits distinct QOs not only in magnetization but also in resistivity (Shubnikov-de Haas effect) even in the insulating phase (Fig. a). The angular dependence of the oscillatory frequency captures 3-dimensional geometry, indicating that the signals derive from the insulating bulk. The temperature dependence of the oscillatory amplitude follows the conventional Fermi liquid theory of metals with a large effective mass, suggesting the presence of an unconventional Fermi surface in an insulator [2].

We also show low temperature heat-transport measurements to discuss low energy excitations in the ground state of YbB<sub>12</sub>. At zero field, sizeable linear  $T$ -dependent terms in specific heat and thermal conductivity are clearly resolved (Figs. b, c), indicating the presence of gap-less fermionic excitations with an itinerant character. Remarkably, linear  $T$ -dependent thermal conductivity leads to a spectacular violation of the Wiedemann–Franz law: the Lorenz ratio is  $10^4$ – $10^5$  times larger than that expected in conventional metals, indicating that YbB<sub>12</sub> is electrically insulating but thermally metallic. Moreover, we find these fermions couple to magnetic fields, despite their charge neutrality. Our findings expose novel quasiparticles in this unconventional quantum state [3].

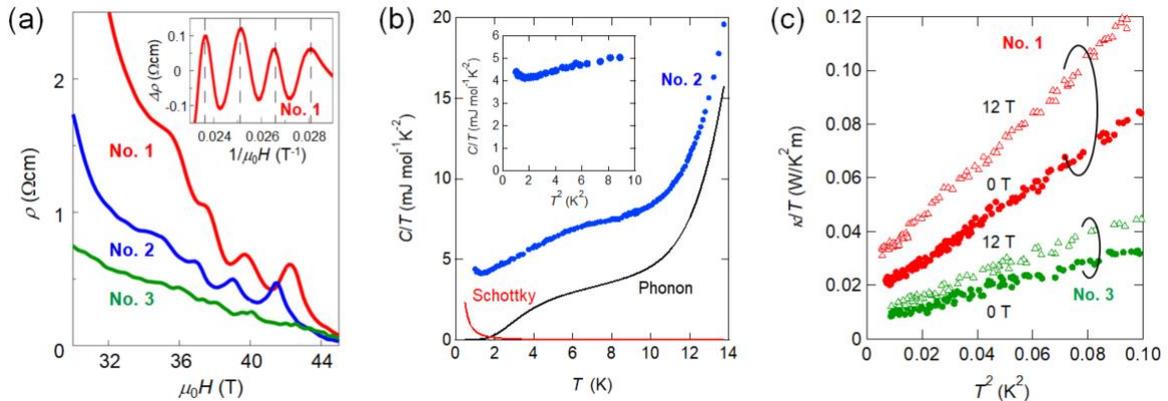


Fig. (a) Observed Shubnikov-de Haas effect. (b) Specific heat and (c) thermal conductivity data at low temperatures.

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# Controlling the polarization of high-order harmonics in solids by manipulating the trajectory of Bloch electrons

Nanophotonics Group      Yasuyuki Sanari

**Abstract** The mechanism of high-order harmonic generation (HHG) in solids was investigated by using a two-color excitation scheme. We manipulated the intraband and interband components of nonlinear currents in GaSe and found that the polarization selection rules for HHG can be well explained by considering the two-dimensional trajectory of Bloch electrons.

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Recent developments in the generation of intense and ultrashort laser pulses have enabled us to study extremely nonlinear optical phenomena, which cannot be treated in a perturbative manner [1]. One technically important example of such extremely nonlinear light–matter interactions is high-order harmonic generation (HHG), where a material emits a broad spectrum consisting of many harmonics of the excitation frequency [2]. While the HHG in atomic gas systems can be accurately explained by the so-called three-step model [3], the mechanisms of HHG in solids are still under debate. In the case of solids, it is considered that high-order harmonics (HHs) are generated by the nonlinear currents induced by an incident laser field, that is, the current due to the motion of Bloch electrons in a band (intraband current) and the current that describes the nonlinear polarization between different bands (interband current) [4]. Therefore, it can be expected that the manipulation of the trajectory of Bloch electrons allows us to control the HH polarization. Since a two-color excitation scheme using orthogonally polarized laser fields enables a flexible manipulation of the trajectory of Bloch electrons, such two-color excitation experiments should be very useful in clarifying the details of HHG in solids. For the polarization control, it is also important to understand the relation between the HH polarization state and the crystal orientation, because the nonlinear interband polarization is dominated by the symmetry of the system. In this work, we studied the mechanism of the HHG in a GaSe crystal under excitation with orthogonally polarized laser fields at different wavelengths, and discussed the relation between the HH polarization state and the crystal orientation.

In our experiments, a GaSe crystal was simultaneously excited by two orthogonally polarized optical pulses with photon energies of  $\hbar\omega_1 = 0.517$  eV (electric field amplitude  $|\mathbf{E}_1| \sim 10$  MV/cm) and  $\hbar\omega_2 = 0.954$  eV ( $|\mathbf{E}_2| \sim 1$  MV/cm). We observed a broadband emission of HHs; the observed HH spectra exhibited multiple peaks at photon energies equal to  $m\hbar\omega_1 + n\hbar\omega_2$  where  $m$  and  $n$  are integers that define the harmonic order. Since  $\hbar\omega_2$  was not an integral multiple of  $\hbar\omega_1$ , it was possible to investigate the different orders independently. We found that the harmonic orders that obey the restriction  $m + n = \text{odd}$  are governed by the intraband contribution and those obeying  $m + n = \text{even}$  are governed by the interband contribution. The orthogonally polarized components of the HHs generated by the interband current exhibited a strong crystal angular dependence, and thus the polarization direction of these harmonic orders can be controlled by the crystal angle. On the other hand, we also observed orders whose polarization components along the direction of the weak incident electric field  $\mathbf{E}_2$  exhibited high intensities at any crystal orientation, and these were assigned to components generated by the intraband current. Because the angle dependence of the intraband-related components cannot be explained by conventional nonlinear optics, we considered the electron trajectory in  $k$ -space that is manipulated by the weak electric field  $\mathbf{E}_2$ . Our calculation results reproduced the experimental data well, suggesting that two-dimensionally driven Bloch electrons in the first conduction band generate a strong nonlinear current in the direction of  $\mathbf{E}_2$ . These results show that a manipulation of the intraband and interband currents by controlling the laser fields and the crystal orientation, enables the control of the polarization of HHs [5,6].

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# Study on the characteristic motion of swimming droplets

Laboratory of Dissipative and Biological Physics Saori Suda

**Abstract** The transition from linear to curvilinear motion of a spherical swimming microdroplet was studied experimentally and theoretically. The experimental results and the model's analysis show that the motion transition depends on the droplet's sensitivity to external perturbations in the direction of motion.

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Over the last few decades, there has been a growing interest in active matters. The knowledge of physical phenomena specific to self-propelled objects has potential applications in various fields such as statistical physics, chemical technology, and life sciences [1]. Swimming droplets have received much attention due to their relative ease of chemical control. Furthermore, various life-like motions have been observed in individual droplets as well. However, the mechanism of their motion transitions is not yet clearly understood.

There exist several reports of motion transitions in swimming droplets. In liquid crystal droplets, the extraction of specific modes of motion is possible via control of the internal symmetry of the liquid crystal phase [2]. For two-dimensional droplets, studies have experimentally and theoretically investigated spontaneous symmetry breaking and straight-to-curvilinear motion transitions [3]. Recently, it has been reported by experimental research that an increased concentration of external surfactants initiates persistent random motion of swimming spherical droplets [4]. Although the straight-to-curvilinear motion transition is one of the most fundamental spontaneous symmetry breaking phenomena observed in a self-propelled system, the mechanism behind the transition has not been explicated for a three-dimensional system of droplets.

In this study, the motion and internal flow of spherical swimming water microdroplets are measured, and the transition from straight to curved motion, including randomly steered one, is quantitatively investigated in a three-dimensional system. A three-dimensional theoretical model is constructed to clarify the motion transition mechanism, using the advection-diffusion equation, the source and removal terms, and the Stokes equation (Eq. (1)).

$$\frac{\partial c}{\partial t} + \mathbf{u} \cdot \nabla c = \frac{1}{R^2} D \Delta_{\text{sphere}} c - \alpha c + \alpha \beta \delta \left( \theta - \frac{\pi}{2}, \phi - \phi_0(\mathbf{v}) \right), \quad (1)$$

The model was validated quantitatively by comparing it with the measured internal convection of the droplet. The model's analysis reveals that the motion transition depends on the perturbation-sensitivity, which varies with the size of the droplet.

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# Nonequilibrium quantum phenomena and topological superconductivity in atomic layer materials

Condensed Matter Theory Group      Hiroomi Chono

**Abstract** We proposed a way to realize topological *s*-wave superconductivity with the application of circularly polarized laser light in transition metal dichalcogenides bilayer. We believe that our study is an important first step to search for topological superconductivity in various atomic layer materials with a weak spin-orbit coupling.

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In the last two decades, motivated by the breakthrough of the laser experimental techniques, controlling of physical properties of materials by terahertz wave and intense short-pulse lasers have been attracting attention. The photo-induced phase transition has been also significantly garnered attention. As a further development, photo-induced topological phase transitions, where the topology of an electronic system is changed by laser light, are studied intensively. Floquet theory is a method for theoretically describing such nonequilibrium steady-state, and the attempts are called Floquet engineering. Floquet engineering has attracted interests as one of the strategies for realizing topological phases of matter which is difficult to be stabilized in equilibrium. For example, photo-induced anomalous quantum Hall states were theoretically proposed in graphene [1], and the observation of this phenomenon was experimentally confirmed recently [2]. It was also proposed that laser light can induce the topological superconducting phase by opening the band gap in thin films of cuprates, which are nodal *d*-wave superconductors [3]. Such nonequilibrium steady-state and relaxation processes induced by laser light are the stage for the realization of physical properties that cannot be realized in the equilibrium state. Furthermore, these attempts have been extended to the superconducting phases.

On the other hand, the recent inauguration of nanotechnology and atomic layer materials research has created an experimental environment for two-dimensional superconductivity with atomically thin materials. Their low-dimensionality has the merit of enabling us to control the superconducting properties. For making insulating materials superconductive, a high-density electric carrier is required. The solution is to use an electric double-layer transistor (EDLT), where ionic liquids and electrolytes are used as the gate electrodes. With this method, two-dimensional electron systems can be transferred to the superconducting phase by electric field induction. In the previous experiments, transition metal dichalcogenides (TMD) such as MoS<sub>2</sub> [4] and NbSe<sub>2</sub>, and iron-based superconductors FeSe have been focused.

Motivated by these situations, we have studied the nonequilibrium quantum phenomena and proposed possible schemes to realize topological superconductivity (TSC) with the application of circularly polarized laser light in two-dimensional TMD layered materials [5]. In contrast to a previous proposal of laser-induced TSC, the TSC in this study does not need SOC and it is robust against the SOC with a realistic magnitude.

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# Anisotropic response of spin susceptibility on a spin-triplet superconductor UTe<sub>2</sub>

Quantum Materials Laboratory      Genki NAKAMINE

**Abstract** UTe<sub>2</sub> is a promising candidate for a spin-triplet superconductor due to its extremely high upper critical field. To investigate the superconducting (SC) properties, we performed <sup>125</sup>Te-NMR measurements. Slight decrease of the Knight shift ( $\Delta K$ ) in the SC state, and anisotropic magnetic field response of  $\Delta K$  support the spin-triplet scenario.

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There are two types of superconducting (SC) spin states: spin-singlet ( $S = 0$ ) and spin-triplet ( $S = 1$ ). Since the SC order parameter of the spin-triplet pairing state has the spin degrees of freedom, unusual behaviors such as multiple SC phases, and magnetic field ( $H$ ) boosted superconductivity are expected. However, there are few examples of spin-triplet superconductors, and thus the physical properties of the spin-triplet superconductivity have not been well understood.

Uranium (U) -based ferromagnetic (FM) superconductors UGe<sub>2</sub>, URhGe and UCoGe are promising candidates for a spin-triplet superconductor, as their SC phases are inside or overlapped with the FM phase, and their upper critical field  $H_{c2}$  is far beyond the Pauli-limiting field ( $H_P$ )[1]. Unfortunately, the investigation of pure spin-triplet SC properties is difficult in these systems because the spin-triplet SC state is affected with the FM ordered state.

In 2018, S. Ran *et al.* discovered superconductivity in UTe<sub>2</sub> with  $T_c = 1.6$  K[2]. UTe<sub>2</sub> also exhibits very large and anisotropic  $H_{c2}$  exceeding  $H_P$  along the three crystalline principal axes[2, 3] as observed in U-based FM superconductors[1]. Particularly, in  $H \parallel b$ , the  $T_c$  shows an upturn around 15 T as shown in Fig.1 and the superconductivity remains up to 35 T until it is terminated by a metamagnetic transition[4]. UTe<sub>2</sub> has also been considered to be a promising candidate for a spin-triplet superconductor as the spin-singlet SC state could not survive in such a high- $H$  region. An important feature of UTe<sub>2</sub> is the absence of any magnetic order at ambient pressure, in contrast to U-based FM superconductors. Therefore, we can investigate pure SC properties in UTe<sub>2</sub>.

We performed <sup>125</sup>Te-nuclear magnetic resonance (NMR) measurements to investigate the SC state of UTe<sub>2</sub>. The decrease in the spin part of the Knight shifts ( $\Delta K_{\text{spin}}$ ) is much smaller than the decrease expected in the spin-singlet state[5]. Furthermore, the  $H$  response of  $\Delta K_{\text{spin}}$  for  $H \parallel b$  and  $\parallel c$  is anisotropic as shown in Fig.2.;  $\Delta K_{\text{spin}}$  for  $H \parallel b$  is almost  $H$  independent up to 6.5 T while  $|\Delta K_{\text{spin}}|$  for  $H \parallel c$  decreases with increasing  $H$  and is almost zero at 5.5 T. The anisotropic  $H$  response and the unchanged Knight shift in  $H \parallel c$  strongly support the spin-triplet scenario. In addition, from the estimation of the critical field  $H_{\text{cr}}$  with  $\Delta K_{\text{spin}}$ , we suggest that the SC properties are changed at  $H_{\text{cr}} \sim 13$  T as shown in Fig.1[6].

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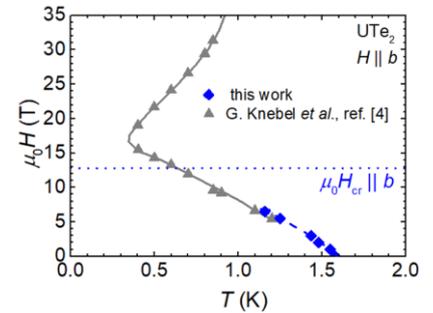


Fig.1.  $H$ - $T$  phase diagram of UTe<sub>2</sub> in  $H \parallel b$ . The dotted line represents the critical field where the SC properties are expected to be changed.

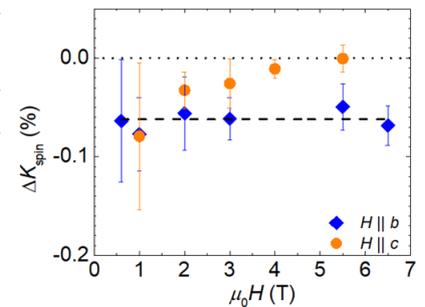


Fig.2.  $H$  dependence of the decrease in the spin part of Knight shift at the lowest temperature.

# Structure of single Xe nanoparticles upon crystallization

Physics of Disordered Systems Group

Akinobu Niozu

**Abstract** We investigated the structure of single Xe nanoparticles crystallized in adiabatically cooled Xe gas jet by single-shot X-ray diffraction using an X-ray free-electron laser. The single-particle structure of Xe nanoparticles indicates that crystallization kinetics play an essential role in determining the structure.

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Crystallization is one of the most ubiquitous physical phenomena in nature. Nevertheless, atomic- and nanoscale structural dynamics upon crystallization is still a subject of controversy. Classical nucleation theory assumes that crystallization starts from a small spherical nucleus having the structure of the stable phase in bulk. On the other hand, more than a hundred years ago, Ostwald [1] proposed his famous step rule, stating that phase transition can proceed via intermediate metastable phases. Recently, theoretical and computational studies have provided novel insights into the structural dynamics of crystallization [2]. In contrast, experimental observations of the crystallization dynamics have been so far mostly restricted to slow dynamics, such as the crystallization of colloidal systems [3].

In this work, we overcome the restriction with the recently available structure analysis method using X-ray free-electron lasers (XFELs). We investigated the structure of single Xe nanoparticles crystallized in a supercooled Xe gas jet. The experiments were carried out at SACLA [4] beamline 3. The single-particle structure of unsupported Xe nanoparticles was probed by XFEL pulses several hundred microseconds after the growth. The diffraction signals were recorded on a shot-by-shot basis with a two-dimensional X-ray detector.

The experimental powder X-ray diffraction pattern indicates that the Xe nanoparticles form not only the face-centered cubic (fcc) structure (the bulk stable phase) but also a structure composed of randomly stacked close-packed layers, the random hexagonal close-packed (rhcp) structure (Fig. 1). The existence of the rhcp structure was further supported by a newly developed analysis method for the single-shot diffraction patterns [5], where the three-dimensional structure factor was deduced from positional correlations of two Bragg spots in single-shot diffraction patterns. Furthermore, through an analysis of the single-particle diffraction patterns, we revealed that the fcc and rhcp phases coexist in single Xe nanoparticles. The experimental results are consistent with the scenario of structure aging, in which crystallization initially occurs at the metastable rhcp phase, and the structure later transforms into the stable fcc structure. The observations are quite analogous to those in the colloidal hard-sphere system [3] and also supported by a recent computational study on ice nucleation [2]. The scenario is potentially applicable to the crystallization of a wide variety of materials. The present work paves the way for the real-time observation of various non-equilibrium dynamics in atomic systems using XFELs.

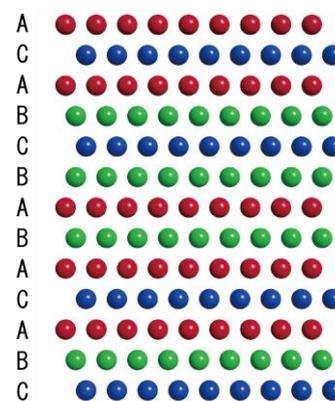


Fig. 1. Schematic of the random hexagonal close-packed (rhcp) structure.

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# Injection Locking of Resonant-Tunneling-Diode Terahertz Oscillator

Solid State Spectroscopy Group

Tomoki Hiraoka

**Abstract** We investigated injection-locking properties of the resonant-tunneling-diode terahertz oscillator in the small-signal injection regime. The locking range was proportional to the injection amplitude and consistent with Adler's model. The noise-reduction behavior due to injection locking was qualitatively explained by Maffezzoni's model of the noise reduction in general limit-cycle oscillators.

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Resonant tunneling diode (RTD) oscillator is a semiconductor device which can oscillate in terahertz (THz) frequency region at room temperature [1]. RTD works as a gain component due to a negative differential conductance originating from the resonant tunneling effect. The dynamics of RTD THz oscillator is not obvious because the conduction properties of RTD are affected by several quantum effects: Under strong THz field, a photon-assisted tunneling occurs and a current-voltage curve of an RTD changes [2]. It is also known that the shot-noise suppression or enhancement occurs in an RTD depending on a bias voltage [3].

One of the major concerns of the RTD THz oscillator is its large linewidth, which is typically 10 MHz for an oscillator operating around several hundred GHz. To stabilize the oscillation frequency, we focused on the injection locking [4, 5], where a noisy oscillator is synchronized to a narrow-band injection signal. To understand the injection-locking mechanism, it is important to clarify the injection-locking properties in a small-signal injection regime. However, experiments in the small-signal injection regime have not been conducted so far. This is because even the slightest return light from surrounding objects acts as an optical feedback, hiding the intrinsic properties of injection locking in the small signal regime.

In this study, we studied the injection-locking properties in the small-signal injection regime without an optical feedback effect. We developed an isolator for THz wave [6] to eliminate minute return light from the detection system. We succeed in the injection locking and dramatically reduced the line width of the emission spectrum from 4.4MHz in the free-running state to less than 120mHz. Figure 1 shows emission spectrum for various injection frequencies. The locking range was proportional to the injection amplitude and consistent with Adler's model [4]. We also observed the noise suppression under injection signal of various amplitudes at the free-running frequency. As increasing the injection amplitude, the noise component in the power spectrum decreased, and the injection-locked component alternatively increased. The noise-reduction behavior was qualitatively consistent with Maffezzoni's model [5] for general limit-cycle oscillators.

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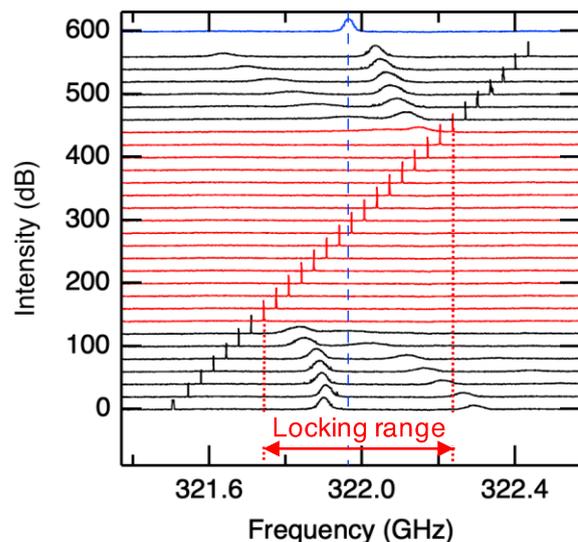


Fig. 1. Emission power spectrum of the RTD THz oscillator under various injection frequencies. The top blue trace is a free-running spectrum. The narrow peaks in other traces show the injection frequencies. The red traces show injection-locked state.

# Identifying exogenous and endogenous activity in social media

Nonlinear Dynamics Group      Kazuki Fujita

**Abstract** Existing time series data are full of non-stationarity. This fluctuation may have been caused by external stimulation, or/and internal self-excitation. We developed a statistical model to make the inference for the occurrence of events, and apply it to a real time series of online social network data.

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Highly clustered event sequences are observed in certain types of real data, such as earthquakes, financial transaction, crimes, neuron activity, and tweets[1]. The occurrence of new events in a system is typically driven by external causes and by previous events taking place inside the system. For instance, Hawkes processes are linear self-reinforced processes, where the occurrence of an event increases the likelihood of future events[2][3].

Distinguishing between exogenous and endogenous forces is critical for understanding the mechanisms that drive dynamics of systems and has important practical applications, such as the quantification of marketing or external factors that may manipulate the social system[3], however, this assumption has not been examined in any detail. This is a general statement, applying to a range of situations including, more recently, to the activity of users in online social networks (OSNs).

Here we develop a method for extracting from a series of posting times the relative contributions that are exogenous, e.g., news media, and endogenous, e.g., information cascade. The method is based on the fitting of a generalized linear model (GLM) equipped with a self-excitation mechanism[5]. We assume the rate of event occurrence  $\lambda(t) = \exp(\gamma(t) + \alpha \sum_k h(t - t_k))$ , where  $\gamma(t)$  and  $\alpha$  represent the time-varying external environment and the degree of internal self-excitation, respectively.  $h(t)$  represents the time course of internal excitation. As a prior distribution of  $\lambda(t)$ , we assume that external modulation  $\gamma(t)$  is slow, by penalizing the large gradient,  $|d\gamma/dt|$ . With the maximum *a posteriori* (MAP) estimate, we can estimate external modulation and the degree of internal self-excitation.

We test the method with synthetic data generated by a nonlinear Hawkes process, and apply it to a real time series of tweets with a given hashtag. In the empirical dataset, the estimated contributions of exogenous and endogenous volumes are close to the amounts of original tweets and retweets respectively.

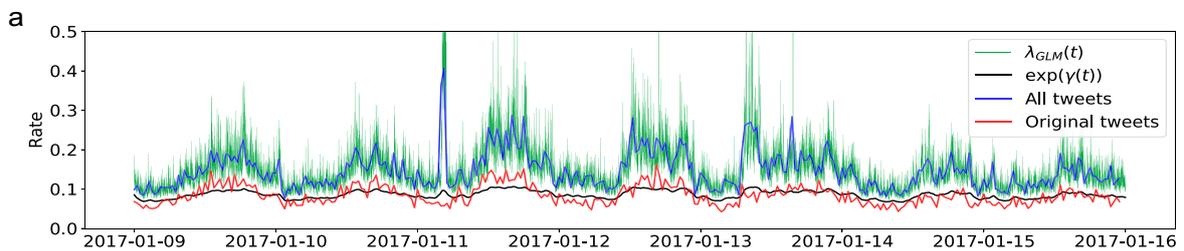


Fig. 1. Original tweeting rate estimation for the one week samples of tweets between Jan 9 and Jan 16, 2017. The solid blue line shows the rate of all tweets, red line below shows the rate of original tweets, black curve represents the inferred exogenous activity and green is the total rate given by the GLM.

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# Theoretical study of nonlinear transport in parity-violating magnets

Hikaru Watanabe (Condensed Matter Theory Group)

**Abstract** This work investigates the nonlinear current generation induced by AC/DC electric field by utilizing the magnetic parity-violation, by which both of the parity and time-reversal symmetries are broken. Combining the symmetry analysis and microscopic calculations, we demonstrate the unique property of these responses.

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It is well known that the parity violation in solids gives rise to various emergent responses and exotic quantum phases. For instance, the coupling between spin and charge degrees of freedom has been used to manipulate the microscopic magnets. The importance of parity breaking has now been widely shared in fields of condensed matter physics. Usually, the parity-violation is satisfied by the asymmetry in crystal structure and by the external electric field. On the other hand, we can notice another type of parity-violation, that is magnetic parity-violation. The magnetic parity-violation breaks not only the parity (P) but time-reversal (T) symmetries while the crystal parity-violation preserves the T-symmetry. The fundamental differences between the magnetic and crystal parity-violations are clarified by the group-theoretical classification making use of multipole degree of freedom in solids [1]. Interestingly, the magnetic parity-violation is highly tunable since it is induced by the magnetic order. Such high controllability is contrasting to the fact that the crystal asymmetry is less controllable.

We theoretically investigate the magnetic parity-violation by working on non-reciprocal current generations via AC and DC electric field, which are photocurrent response and (non-reciprocal) nonlinear conductivity, respectively [2,3]. Although these phenomena can be implemented by the crystal parity-violation as well as magnetic parity-violation, they should be distinguished by the preserved symmetry. The T and PT-symmetries (PT operation is the combination of the P and T-operations) preserved in the crystalline and magnetic parity-violating system play important roles in classifying the emergent responses [1,4]. Comparing with the non-reciprocal responses in the T-symmetric systems, we systematically study the striking properties arising from the magnetic parity-violation as follows.

## 1. Field-free non-reciprocal Hall effect in metallic antiferromagnets [2]

Making use of the symmetry analysis and microscopic calculations, we demonstrate the field-free Hall effect in a metallic antiferromagnet, doped BaMn<sub>2</sub>As<sub>2</sub>, and propose a potential application to antiferromagnetic spintronics [5].

## 2. Giant chiral photocurrent generation in a topological antiferromagnet [3]

Following the systematic classification with the T and PT-symmetries, we discover the current generation response induced by circularly-polarized-lights. This response is explained by the shift-current mechanism and closely related to the quantum geometry in the electronic band structure. Moreover, the strongly-enhanced photocurrent response is demonstrated by the microscopic analysis of the antiferromagnetic compounds hosting the topologically-nontrivial electronic structure.

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