Single shot x-ray diffractometry in SACLA with pulsed magnetic fields up to 16 T

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Single shot x-ray diffraction (XRD) experiments under pulsed high magnetic fields up to 16 T generated with a nondestructive minicoil have been performed with an x-ray free electron laser at SACLA. In a perovskite manganite, Pr0.6Ca0.4MnO3, the magnetic field induced phase transition from the charge-orbital ordered insulator phase to a ferromagnetic metallic phase at above ~ 8 T, which is observed in a series of single shot XRD via the accompanying lattice changes. It is discussed whether a XRD experiment under an ultrahigh magnetic field of 100 T is feasible using single-shots of the SACLA XFEL pulse and a destructive pulse magnet.

I. INTRODUCTION

As a new light source, free electron lasers (XFELs) are prominently characterized by the ultrahigh transverse coherence, an ultrashort pulse of a few femtoseconds and a high-photon flux of 1011–12 photons/pulse that realizes single shot experiments. The advent of XFEL technique has provided us with such new experimental techniques to explore new area of science as coherent diffraction from a single nano-cluster, diffraction before destruction from a living cell, femtosecond time resolution pump-probe experiments, and also experiments with single shot x-ray diffractometry (XRD) under shocked compression environment [1, 2]. As such a new experiment, we are proposing x-ray experiment at an extremely high magnetic field of above 100 T where we utilize the ultrashort temporal width and the single shot experiment of a XFEL, and a destructive pulse magnet for 100 T generation.

High field experiments have been successfully combined with synchrotron radiation (SR) based XRD and spectroscopies for condensed matter experiments with DC magnets up to 15 T [3–7] and non-destructive pulse magnets up to 50 T [8–16]. Recent advent of single shot techniques with XFEL, further made possible the observations of the weak superlattice reflections of charge density wave order appearing in a cuprate by suppressing the high-Tc superconductivity up to 32 T [17, 18]. These high-field studies with SR and XFEL have been confined below 50 T. To generate magnetic fields over 50 T is a challenge for a portable non-destructive pulse magnet. Non-destructive magnets generating above 50 T are available only in specific high magnetic field facilities over the world, where pulsed magnetic fields up to 100 T are available with sufficient safety measures [19].

For x-ray experiments at well above 50 T, we propose a use of a single turn coil (STC), a destructive pulse magnet, instead of non-destructive pulse magnets, because a STC can be portable and generates magnetic fields over 100 T [20, 21]. Generating high fields well beyond 100 T necessarily requires destructive pulse magnets where magnetic pressure destroys the coil. For condensed matter experiments, flux compression and single turn coil techniques have been implemented [22–24]. The temporal duration of the generated field by a single turn coil is only a few microseconds, which is about three orders of magnitudes smaller than a pulse of a non-destructive magnet. It is still long enough for a XFEL pulse to be used for a single shot experiment. Note also an exceptional single shot technique utilizing SR from storage rings [25], though usually the photon number of SR-based x-ray is too small for a single shot pulses of 100 T of a few microseconds.

For a 100 T experiment at an XFEL site, one needs to construct a portable 100 T generator. It is needed to test special equipments such as nonmetallic vacuum tubes and cryostats that are conventionally used with the STC experiments. Portable STCs had been implemented before [20, 21]. A portable STC to be build should be less than 200 kg. We are now constructing a portable pulse power with 30 kV charging and 4.5 kJ energy with a mono-spark gap. Conventionally, two STCs are in operation in ISSP Univ. of Tokyo, with 200 kJ at 50 kV with a total weight of 2 tons. In ISSP, specially prepared nonmetallic cryostat and vacuum system have been implemented for destructive pulse experiments at low temperatures. These equipments have been known to be proof against the explosions of the STC magnet. It is most preferable to use these equipments in the 100 T experiments in SPring-8 Angstrom Compact free electron LAser (SACL). To check this possibility, we need to test if these equipments are compatible with the x-ray experiments in SACL. Thus, it is fruitful to test the feasibility of those equipments at SACLA with a minicoil mimicking the 100 T experiment with STC.

As preceding studies for the singe shot XRD at 100 T in SACLA, we have discovered that the lattices of materials are actually responding to 100 T fields of a few microseconds pulse generated in destructive pulse magnets [26]. Recently, a high-speed strain sensor utilizing fiber Bragg grating (FBG) is devised for magnetostriiction measurements at above 100 T [27]. Solid oxygen is a candidate for structural analysis with XRD at above 100 T. A high field beyond 120 T induces a new thermodynamic phase called θ phase [28]. A cubic lattice geometry is anticipated in the θ phase in contrast to the monoclinic α phase at low field phase, which remains elusive until a

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XRD is performed at 120 T. An magnetostriction measurement do find a first order lattice change accompanying the magnetization jump, indicating a lattice change at α-β transition. A perovskite cobaltite LaCoO₃ is a candidate for investigating superlattice reflection at 100 T. A peculiar spin crossover (SCO) is induced by magnetic field beyond 100 T [29]. There are two high field phases whose origins are controversially argued to be an Bose–Einsteins condensation of excitons, a excitonic insulator, or a crystallization of the spin-state degree of freedom. This is to be determined by observing a superlattice reflection from the spin-state crystallization and also by observing the d electron states by the x-ray emission spectroscopy.

In this paper, we report an experiment of single shot XRD at SACLA with a non-destructive room-temperature-bore magnet mimicking the single turn coil. The purpose of this work is to see feasibility of an application of the ready-made explosion-proof equipments for the STC to XRD experiments at SACLA. We successfully detect the field induced phase transition of Pr₀.₆Ca₀.₄MnO₃ with a series of single shot XRD with SACLA up to 16 T. We comment on a design for a portable single turn coil system for SACLA.

## II. EXPERIMENT

The experiment was performed at a hard x-ray beam line BL2, in SACLA [30, 31]. A schematic drawing of the experimental setup is shown in Fig. 1(a). The XFEL was tuned to 16 keV with a mean pulse energy of ~ 100 µJ/pulse. Pink beam (∆E/E ~ 10⁻³) and monochromatic beam (∆E/E ~ 10⁻⁴) were used. The single shot diffraction signals were monitored with a multi-port charge-coupled device (MPCCD) image sensor [32]. The magnetic fields were generated with a mini bank system of 2.4 kJ at 2000 V (1.2 mF) [8, 33] and a room temperature coil wounded by hand. The waveforms of the pulsed magnetic fields are shown in Fig. 1(b).

The bore of the coil is 1cm diameter and 1cm long in axial direction. In the bore, a vacuum tube is suspended with a He-flow and non-metallic type cryostat located inside as shown in Fig. 1(c). The sample is put inside the cryostat, which is a position where a XFEL pulse hits and the diffracted signal escapes through Kapton windows at the back and top of the vacuum chamber. The 2θ range of 15 - 25 degree is captured with a 25.6 x 51.2 mm window of the MPCCD with a distance of 300 mm from the sample to the detector. The diffraction image is shown in Fig. 1(d).

Pr₀.₆Ca₀.₄MnO₃ was powdered from a single crystal and dispersed in a glue whose effective thickness was ~ 10 µm. Powdered samples with rough one and smoother one are called sample P and Q, which are used with the pink and the monochromatic beams, respectively. The powder was roughly powdered on purpose so that spotty Debye ring is produced in XRD, where it is expected that we observe a speckle pattern around the diffraction spot reflecting the microstrain of a particle.

![Diagram](image)

**FIG. 1.** (a) A schematic drawing of the experimental setup of the single shot XRD in SACLA with a pulse magnet. The temporal diagram of the measurement is also shown. (b) The time evolution of the magnetic field and the XFEL pulse intensity. Magnetic fields can be varied in two ways in a single shot XR. The generated magnetic field is varied with a fixed XFEL timing on top of the pulsed magnetic field. In another way, the XFEL timing is varied with a fixed waveform of the pulsed magnetic field. (c) A schematic drawing of a part of the nonmetallic cryostat used in ISSP for STC experiments, which is used this time for XRD study for verification of the compatibility. (d) An image of a single shot XRD recorded by the MPCCD image sensor with a monochromatic beam of the XFEL.

## III. RESULT AND DISCUSSION

Fig. 2 shows a series of single shot XRD at various magnetic fields up to 16.3 T at 2θ = 16.3°, where 020, 200 and 112 reflections are relevant. The spotty diffraction ring is produced from the roughly powdered sample P. One diffraction spot is coming from a single domain of a micro-particle of a sample. The magnetic field effect is visible in Fig. 2. The trend of the image is similar from 0 T to 6.6 T. The images from 9.7 T to 16.3 T have a common feature being distinct from the low field data. The images at 7.7 T and 8.7 T seem to be linear combinations of both features from low field and high filed data, indicating a transient state. Two magnifications are shown in Figs. 2(b) and 2(c). In Fig. 2(b), it is clear that the spot A appears above 7.7 T. In Fig. 2(c), on the contrary, the spot B disappears above 9.7 T. The x-ray intensities of the spot A and spot B are shown as a function of B as shown in Fig. 3. The appearance of spot A means that the θ-2θ configuration is satisfied after the field induced lattice parameter change of the particle for spot A. In contrast, for spot B which vanishes after the field induced lattice parameter change of the particle.
for spot B, the $\theta$-2$\theta$ configuration is no longer satisfied at high fields. With an energy dispersion of the pink beam, $10^{-3}$, the assignment of the diffraction peaks is inconclusive.

Fig. 4(a) shows a result of a series of single shot XRD at pulsed high magnetic fields with a monochromated XFEL beam. Fig. 4(b) shows integrated intensities of x-ray in vertical direction of the pink colored area of Fig. 4(a). The less spotty picture than Fig. 2 indicates the sample is a bit smoother in the sample Q. It is clear that the 112 reflection increases at above 8 T, while the 020, 200 reflections decreases. This behavior is in good agreement with the previous result [8]. It is reported that, in Pr$_{0.6}$Ca$_{0.4}$MnO$_3$, the charge and orbital ordered insulator (COOI) phase appears at low temperatures [34]. When COOI phase appears, the $a$ and $b$ axis elongates and the $c$ axis shrinks [35, 36]. When COOI phase is collapsed as a result of an external magnetic field, the $a$ and $b$ axis shrinks, and the $c$ axis elongates, as observed in the previous study using SR and a mini pulse magnet. According to this picture, the 020 and 200 diffractions are supposed to shift to higher 2$\theta$ overlapping the peak of 112, and the 112 diffraction stays. As a result, the diffraction peak at 16.4° decreases and the diffraction at 16.5° increases. The observation in Fig. 4(b) is in good agreement with the above expectation, indicating that the field induced
COOI with a single shot XRDs in SACLA is successfully observed, with a slight discrepancy that the 020 reflection does not clearly disappear at high fields. This discrepancy may arise from the fact that the Debye ring is not smooth enough to obtain a good statistics, which is evident from the XRD picture in Fig. 4(a). A smooth data will be obtained with a use of adequately finely powdered sample. So far it is shown that the single shot XRD is successfully conducted with a 1.4 ms-pulsed magnetic field and the non-metallic cryostat and the vacuum tube that are compatible with the STC experiments. Also, the lattice change of Pr$_{0.6}$Ca$_{0.4}$MnO$_3$ accompanying the field induced phase transition is observed in a series of single shot XRD experiments. The observed changes of XRD are consistent with the shrinkage of $a$ and $b$ axis and an elongation of $c$ axis at high fields. Here, we propose and discuss the feasibility of a single shot powder XRD measurement in SACLA up to 100 T for a structural analysis. In the present study, a rough powdered sample was employed with the motivation to observe speckle patterns from a micrograin under stress in the two phase coexistence state during the phase transition. However, such speckle patterns are not observed around the diffraction spots in Fig. 2. The 100 T XRD experiment will focuses on the structural analysis by means of the smooth powder diffraction using a well powdered sample. In the present study, a limited range of $2\theta < 25^\circ$ is covered, where a small number of low indexed diffraction peaks are obtained, being limited by the opening angles of the fiber-reinforced plastic (FRP) based cryostat and the minicoil. Dimension of the present minicoil is $d = 10$ mm in diameter and $h = 10$ mm in axial length. For structural analysis, it is favorable to cover a larger angle up to $2\theta \sim 60^\circ$. A larger coverage of $2\theta$ becomes possible with a use of all-Kapton cryostat and a vacuum tube with a Kapton window as shown in Fig. 5. In the 100 T XRD experiment, a double turn coil is to be used which has a larger aspect ratio of $h/d = \sim 0.5$. This allows us to cover $2\theta$ up to $60^\circ$ as schematically inspected in Fig. 5.

Presently, a pink beam does not show an appreciable resolution of XRD for the structural analysis. A use of narrower band beams such as monochromatic beam or the seeded beam [37] is practical in the 100 T XRD experiment. Besides, use of a photon energy of 10 keV increases the photon number by a factor of $\sim 6$ in stead of a photon energy of 16 keV used in the present study. Presently, $h\nu = 16$ keV is used due to the limitation of the small $2\theta$ range. With the larger $2\theta$ as proposed above, the use of $h\nu = 10$ keV is allowed. A magnetic field of 80 T is estimated to be obtained with the double turn coil with a diameter of 6 mm and the portable bank system of ours currently under construction, which is rated at 30 kV with an energy of 4.5 kJ, generating 200 kA. Instead of the MPCCD detector, we plan to use an imaging plate (IP) because IP allows a robust detection even with a fragmentation of the coil, which is not feasible with the MPCCD detectors.

The single crystalline experiment aiming a weak structure like superlattice reflection is at this moment not feasible due to the limited volume of the magnetic field generated in STC, where sample rotation mechanism should be somehow installed. Evaluation of this method will be a future task.

IV. SUMMARY

We performed single shot XRD experiments in SACLA with a pulsed magnetic field up to 16 T. We have successfully observed field induced lattice change in Pr$_{0.6}$Ca$_{0.4}$MnO$_3$, which is originated in a collapse of COOI phase and appearance of a ferromagnetic metal. Based on the result, a methodology of a single shot XRD experiment up to 100 T range in SACLA using a portable 100 T generator is discussed.

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