Investigation of Magnetic and Charge Dynamics by combining Pulsed Neutron-X-ray sources and Pulsed High Magnetic Fields

High magnetic field scattering experiments have been conducted by using a compact pulsed field generator developed at IMR. Experiments have been successfully performed on 5f-itinerant intermetallics, multiferroic transition metal oxides and high T_c-superconductors. The determinations of the order parameters and related wave vectors as well as lattice symmetries have been conducted, which shows the usefulness of such scattering experiments in high magnetic fields.

1. Introduction

A high magnetic field induces interesting new phases of matters by its strong coupling with spin, charge and lattice degrees of freedom of correlated electrons. To investigate such exotic states, the use of neutron and X-ray scatterings are indispensable because magnetic, charge and lattice order parameters can be determined directly in these methods. The combinations with high magnetic fields, however, have been quite rare for the complexity of experimental set up and for the limited maximum field in superconducting magnets.

To breakthrough this situation, in this project, we have developed compact pulsed field generators to conduct neutron and X-ray scattering experiments in very strong magnetic fields of 20-50 T. These have been used to in experiments on various materials in oversea and domestic facilities. The developed systems have been permanently installed in two facilities, ISIS spallation neutron source in UK and X-ray free electron laser source at Stanford linear accelerator laboratory in USA. Moreover, a mobile capacitor bank which can be shared among other facilities by the international collaboration with ICC-IMR is also manufactured.

2. Outline of the generator

Figure 1 shows the mobile capacitor bank designed and assembled at IMR. It stores 16 kJ energy and can drive different mini coils tor X-ray spectroscopies and X-ray diffraction. For the X-ray diffraction, 33 T is available with a split-pair magnet. For hard X-ray absorption spectroscopy, the maximum magnetic field of 50 T is available with a solenoid coil of 3 mm bore. In neutron diffraction, 30 T and 40 T magnet inserts are developed. A compact 60 kJ capacitor bank is used for 40 T neutron insert. The photo in Fig.1 shows the 30 T magnet for neutron diffraction.

On of the most important features of the pulse magnetic field is the flexible design of magnets, which is essential to compromise the critical requirements in the highest possible level. Such ability is the important basis of the successful applications. For example, our pulsed magnet has been also used to explore unknown elementary particle called axion, which is believed to be responsible for dark matter in the space. In high magnetic fields, the high flux X-ray should converts into axion with higher efficiency. By the collaboration with Tokyo University group, we have succeeded in narrowing the area of the possible axion energy[1]. As such, the combination of strong magnetic fields and neutron and X-ray opens the new scientific area in both condensed matter and in fundamental physics. In the following, we present a few examples of scattering experiments.

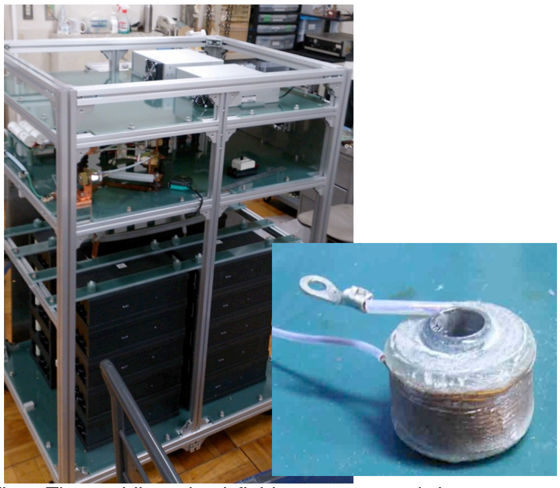

Fig. 1The mobile pulsed field generator and the magnet used for neutron diffraction.

3. Neutron diffraction

The first example is the study of magnetic field induced ordering in URu_2Si_2 with tetragonal unit cell. This compound has attracted interests in many years for the appearance of the mysterious hidden order phase, where no clear order parameter has been identified although the evidence of the phase transition is shown by the macroscopic measurement such as heat capacity. In the magnetic fields of 30-40 T, this hidden order is destroyed and some magnetic order seems to appear. For the lack of the neutron diffraction, the related magnetic structure has not been known over 30 years.

By the series of experiments at ILL and at ISIS, we have found that the magnetic structures of the high field phases are sensitive to the doping.

Namely, a commensurate up-up-down type order appears in the tetragonal plane when 4 % of Ru is replaced with Rh. This commensurate order disappears when Rh doping is reduced to 2 % and then an incommensurate density wave like magnetic phase is found in pure compound[2]. This observation indicates that the magnetic field induced transition is closely related with the Fermi surface structure and its nesting. Such unique information cannot be obtained without neutron diffraction.

The second example is the magnetoelectric Lithium orthophosphate $LiNiPO_4$. The application of a magnetic field along the crystallographic *c*-axis causes the electrical polarization $\boldsymbol{P}=\alpha\boldsymbol{H}$ along the *a*-axis. The magnetoelectric coefficient is among the largest in transition metal compounds. Motivated by pulsed-field magnetization and electric polarization measurements up to 55 T, we have engaged in a campaign to determine the characteristic propagation vectors of the magnetic phases for high magnetic fields[3].

One of the present highlight is the neutron diffraction at 41.2 T with which we could determine that the phase VII appearing above 39 T is commensurate and the phase VI appearing in 37-39 T is incommensurate. To the best of our knowledge, this is the highest magnetic field ever employed in neutron diffraction experiment. With the present results, we confirmed that all magnetoeelectric phases are commensurate, while all non-polar phase is incommensurate in $LiNiPO_4$. Such universal behavior can be only known by the neutron diffractions in the wide range of magnetic field.

In our experiment, we successfully employed the pulsed field Laue diffraction technique and managed to determine the magnetic propagation vectors of all magnetic phases of interest. It seems certain that pulsed field Laue diffraction will become an increasingly important part of the neutron scattering toolbox, and that many exciting high-field magnetic phases in a diverse range of material classes are now within reach. From a technical perspective, increases of the maximum field are feasible, and further improvements of neutron instrumentation and beam power will guarantee the future success of pulsed-field Laue diffraction.

4. X-ray Diffraction

One of the recent highlights of X-ray diffraction is the observation of unidirectional and three -dimensionally correlated charge density wave(CDW) in magnetic fields above the upper critical fields. The CDW states appearing in Cu-oxide superconductors have been intensively studied. However, the direct observation of the CDW super-lattice peak had been made only in magnetic fields had been desired for long time.

We combined two pulsed tools, the pulsed magnet and the pulsed X-ray source to accomplish this difficult task. The magnetism division of IMR has developed an extremely compact split-pair magnet generating magnetic fields over 30 T. The bore of the coil is only 3 mm and the length of the coil is as short as 25 mm. The Stanford Laboratory offers the most intense pulsed laser source on the earth. When these two tools are combined, weak CDW peaks in $YBa_2Cu_3O_{6.67}$ can be detected clearly in a single shot pulsed magnetic field.

In the present experiments, a new CDW state with strong three-dimensional correlation is discovered. The index of this CDW state is given by (δ, 0, l), where δ is related to the localized charge modulation in the *c*-plane. Moreover the index l is found to be an integer. This unique feature shows a sharp contrast to the two-dimensional CDW peak found in lower magnetic fields at (δ, 0, $l/2$) position, where the modulation pitch along the *c*-axis is doubled. The intensity of l=integer peak grows rapidly in high magnetic fields, while that of the half-integer peak saturates in lower fields. The appearance of a state with localized electrons with strong three-dimensional correlation shows the essential role of the strong electron correlation in the normal state of $YBa_2Cu_3O_{6.67}$[4, 5]. The observation of the weak super lattice peak shows the possible detection of the Fermi surface nesting anomaly by high field X-ray diffraction.

5. Summary

Through the combined efforts of materials discovery and specialized high-field laboratories worldwide, a steady progress of novel materials displaying exotic phases and phase transitions at high magnetic fields are emerging. Typical material classes are superconductors, multiferroics, magnetoelectrics and quantum magnets. In all such cases, a significant step towards a full understanding the microscopic physical processes responsible for the occurrence of these phases is the identification of magnetic, electric order parameters and of the lattice symmetry. The present project established the usefulness of pulsed magnetic fields for scattering experiments.

Reference

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