I. Project Research

Project 1

PR1   Project Research on Nuclear Spectroscopy and Condensed Matter Physics Using Short-Lived Nuclei

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Objective and Participating Research Subjects

The main objectives of this project research are the investigation of the nuclear structure of unstable neutron-rich nuclei and also the local properties of materials using short-lived nuclei.

This period is the third and last year of the project.

The research subjects reported here are as follows:

31P1-1 Technique of transferring radioactive atomic nuclei implanted in dry ice film

31P1-2 β^-–Decay spectroscopic studies of fission products using the clover detector

31P1-3 Linear polarization measurement for γ rays from ^{148}Pr

31P1-5 Measurement of the internal pressure in ultrafine bubbles by angular correlation technique

31P1-6 Temperature-dependent polaronic local structures in $La_{0.7}Ca_{0.3}MnO_3$ observed through spin relaxation of TDPAC probe nuclei

31P1-7 Local structure of In impurities doped in $SrTiO_3$ studied by TDPAC method

Main Points Described in the Reports in the Following Six pages

As a means to measure the charge distribution of an unstable nucleus, it is promising to make a muonic atom composed of the nucleus that are trapped in deuterium film and then to measure the energies of X-ray emitted from the muonic atom. However, it is unavoidable to deal with high radioactivity. One of the technical problems is how efficiently and safely long-lived radioactivity in the apparatus is removed after such an experiment is done. A. Taniguchi *et al.* (31P1-1) has attacked this problem using a beam of ^{146}LaO$^+$ radioactive ions available at KUR-ISOL and an apparatus designed for the present work. In this period, they improved the apparatus so that the removal efficiency became much higher than that obtained previously.

M. Shibata *et al.* (31P1-2) investigated the level structures of ^{153}Nd and ^{154}Nd, arising via β^- decay, respectively, from ^{153}Pr (half-life of 4.3 s) and ^{154}Pr (half-life of 2.3 s), fission products of ^{235}U + thermal neutron, available at KUR-ISOL. Because of the large Q_β values, 5752 keV for ^{153}Pr and 7790 keV for ^{154}Pr, it is possible to access to high excited levels of ^{153}Nd and ^{154}Nd. They used a clover detector consisting of 4 Ge crystals both in singles and add-back modes. For ^{153}Nd, they identified eight new excited levels and also new nineteen γ-rays. The detailed analysis is in progress for constructing the decay schemes.

Y. Kojima *et al.* had tried to measure the linear polarization of γ-rays from short-lived β decaying nuclei using a clover detector (the same detector that M. Shibata *et al.* used in 31P1-2) as a Compton polarimeter and additional coaxial Ge detectors in order to obtain the multipolarities of those γ-rays. In the last period, they confirmed the feasibility of their polarimeter system using the established 258-410 keV E2-E2 γ cascade in ^{146}Ce arising from ^{146}La via β^- decay, available at KUR-ISOL. In this period, they succeeded in obtaining the preliminary result showing the E2 character of the 1358-keV γ ray from ^{148}Pr (half-life of 2.1 m) decaying to ^{148}Nd, produced at KUR-ISOL.

Ultrafine bubbles, attracting a lot of attention in various industrial fields, are of the diameter less than one micro meter. Despite the Young-Laplace equation predicting a high internal pressure for such a small bubble in water so that it should survive only very short time, it is reported to survive more than a month. Employing the time-integral perturbed angular correlation (TIPAC) technique for ^{125}I arising from ^{125}Xe (half-life of 17 h) produced at the slant exposure tube of KUR, M. Tanigaki *et al.* successfully obtained the internal pressure of ultrafine bubbles of the average diameter about 200 nm containing Xe gas inside, the value of which is much smaller than that the Young-Laplace equation predicts. In this period, they (31P1-5) reevaluated the value of the time-integrated perturbed angular correlation.

A perovskite manganese oxide $La_{0.7}Ca_{0.3}MnO_3$ (for short, LCMO) is well known to exhibit colossal magnetoresistance. It is considered that polaron dynamics plays a significant role in this phenomenon. Because of the large variation in physical properties of $La_{1-x}Ca_xMnO_3$ with x, the A-site (La or Ca) information is especially important. In order to obtain this local information for LCMO, W. Sato *et al.* (31P1-6) synthesized 2 at. % nonradioactive Cd-doped LCMO containig a tracer amount of radioactive ^{111m}Cd (half-life of 49 m) produced in the pneumatic tubes of KUR and took ^{111}Cd($\leftarrow^{111m}$Cd) TDPAC (time-differential perturbed angular correlation) spectra at temperatures above and below the Curie temperature of $\sim$ 250 K. They succeeded in observing the static and dynamic polarons around the probe nuclei at the A site of LCMO.

$SrTiO_3$ is a cubic perovskite oxide. Doped with metal impurity ions, it exhibits a wide variety of electronic properties. In order to obtain local information on the impurity site, S. Komatsuda *et al.* (31P1-7) synthesized $SrTiO_3$ containing a tracer amount of commercially available ^{111}In (half-life of 2.8 d) and took the room-temperature TDPAC spectrum of ^{111}Cd arising from ^{111}In. They found three kinds of quadrupole frequencies in the spectrum: one is zero frequency indicating ^{111}Cd at the defect-free substitutional site and the other two non-zero frequencies implying ^{111}Cd at the substitutional site associated with oxygen vacancy.

PR1-1   Technique of Transferring Radioactive Atomic Nuclei Implanted in Dry Ice Film

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INTRODUCTION: Measurement of the emitted X-ray energy from muonic atoms is a powerful method for the investigation of the nuclear charge radius [1]. Recently, a new method using solid hydrogen film has been developed to produce muonic atoms. In this method, negative muons are injected to solid deuterium film in which nuclei of interest are implanted beforehand, thereby muonic atoms being formed through the highly-efficient muon transfer reaction. The feasibility of this method was demonstrated and promising results were obtained [2]. In this study, one of the problems in applying this method to unstable nuclei, concerned with handling of residual radioactivities in deuterium film after experiments, in particular their highly efficient recovery, was approached using a radioactive-isotope beam from KUR-ISOL and dry ice film instead of solid hydrogen film.

EXPERIMENTS: An apparatus capable of implanting radioactivities to dry ice film was installed at the beam line of KUR-ISOL [3]. This apparatus has two coolable copper blocks and one CO_2 gas diffuser in the vacuum chamber. Dry ice film is formed on the surface of one cooled block (catcher) by sprayed CO_2 gas through the diffuser and radioactivities are implanted into it. In a series of recent experiments, even if films of the same appearance were formed on the catcher, large variations were observed in the amounts of radioactivities transferred to the other block (trap). It is considered that the condition of dry ice film is important and the forming parameters should be controlled more finely in order to improve the reproducibility of the transfer efficiency. In this work, film was formed while recording the vacuum degree in the chamber with a data logger. In addition, by attaching "standing collars" on the four sides of the film formation surface, more CO_2 gas from the diffuser is guided to the surface during film formation, furthermore during transfer, the collars cover the catcher-trap gap so that sublimation components become less likely to escape into the chamber. About 10^6 ions of $^{146}LaO^+$ were implanted into dry ice film on the catcher in every run. After the implantation, the other pre-cooled trap was moved to the frontal vicinity of the catcher, and then the catcher was warmed by stopping LN_2 flow. With this procedure, the atoms of ^{146}Ce and ^{146}Pr were released from the catcher and were re-trapped on the trap together with CO_2. The transfer efficiency was measured by detecting the γ-rays emitted from each of the blocks. In each run, it took about 2 hours from the implantation to the completion of the transfer.

RESULTS AND DISCUSSION: The transfer efficiencies were measured by changing the amount of CO_2 while confirming the vacuum degree when dry ice films were formed. Although the detail data analysis including the absolute trap efficiency for the implanted radioactivities is in progress, the higher transfer efficiency result of about 880% was obtained (see Fig.1). The efficiency is defined as the ratio of the radioactivities on the trap to those remaining on the catcher, which was deduced from the peak counts of 454 keV γ-ray of ^{146}Pr by taken into account of the detection efficiency of each Ge detector in its actual arrangement. This result is still poorly reproducible and the appropriate parameters for film formation have not yet been fixed, but much higher than the previous results [4]. Therefore, it can be said that the control of the vacuum degree is one of the important parameters.

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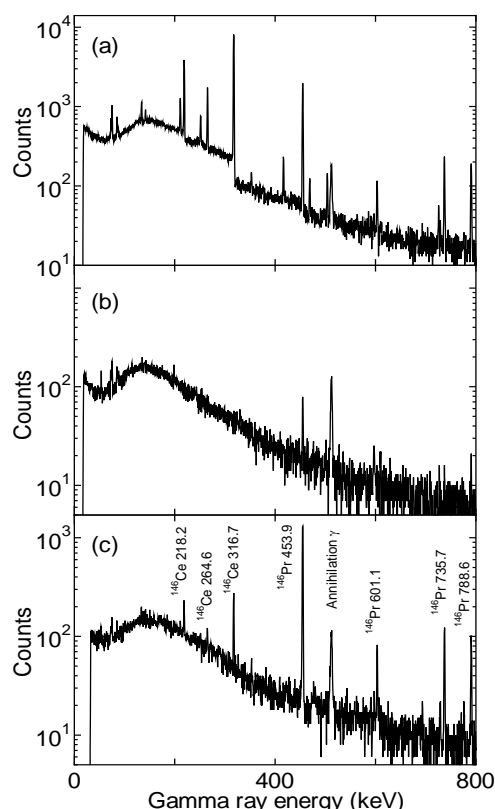[4] A. Taniguchi *et al.*, KURNS Prog. Rep. 2018, p28.

Fig.1 Transfer of radioactivities using a cold trap type recovery apparatus. (a)Gamma-ray spectrum of the catcher immediately after ^{146}La implantation. ^{146}La ($T_{1/2}$ = 10 s and 6 s) decayed out and γ-rays of ^{146}Ce ($T_{1/2}$ = 14 m) and ^{146}Pr ($T_{1/2}$ = 24 m) were observed. Gamma-ray spectra of (b) the catcher and (c) the trap after the transfer.

PR1-2   β-Decay Spectroscopic Studies of Fission Products Using a Clover Detector

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INTRODUCTION: Precise decay scheme of fission products are very important to study nuclear structure and also decay heat evaluation in nuclear reactor. The neutron-rich and short-lived nuclei around mass number 150 have low yields then their decay data are scarcely studied. In particular, ^{153}Pr ($T_{1/2}$ = 4.3 s) and ^{154}Pr ($T_{1/2}$ = 2.3 s) were studied by separating from the fission products of ^{235}U with isotope separator on-lines (ISOL) and was reported few γ-rays[1, 2], nevertheless, the $Q_β$-value of them are reported to be 5762 and 7790 keV [3], respectively. It means that the both daughter nuclei are expected to have much higher excited levels and also isomeric states having μs order of half-lives. To identify higher levels and high energy γ-rays and to propose detailed decay scheme, the γ-rays associated with the β-decay of ^{153}Pr and ^{154}Pr were measured with a clover detector at KUR-ISOL.

The clover detector has 4 large Ge crystals of 80 mm in diameter and 90 mm in length, they are arranged as the shape of a four-leaf clover, and it has through-hole which diameter is 15 mm in the detector. The solid angle is 98% in center of the hole, so γ rays can be measured with high efficiency. The list mode including time information, so-called a time-stamped list mode, data acquisition (DAQ) system was adapted to identify the γ-γ coincidence relation. With this system, two types γ-ray spectra, those are "singles" and "add-back" spectra can be obtained by off-line analyses. In the add-back spectra compared with singles spectra, sum peaks are strongly observed, on the other hand, the cascading γ rays are observed weakly by coincidence summing out. The off-line analyses, the location of γ rays in the decay scheme can be made clear.

To deduce the half-lives of isomers with the DAQ system, β-γ time difference method was applied. In this method, the time difference between the γ-ray of interest and a certain continuum part of spectrum which corresponds β-rays are extracted from the list mode data. In this method, the counting rate during the measurement is an important parameter for obtain reliable results. The isomer ($T_{1/2}$ = 52.6 μs) at 1088 keV in ^{95}Y of daughter ^{95}Sr ($T_{1/2}$ = 23.9 s) is proper nucleus to check the effect of the counting rate on the obtained value.

EXPERIMENTS: The nuclei of interests were mass-separated by the KUR-ISOL from the fission products of ^{235}U. A 72 mg of 93% enriched UF$_4$ was irradiated with a thermal neutron flux of 3×10^{12} *n*/cm^2/s. The radioactive sources were collected on a thin Mylar tape and were periodically transported to the center of the detector with computer-controlled tape moving system. The tape moved periodically with two times of each half-life. In the through-hole of the detector, the two semicircle-formed 6mm thick acrylic sticks were set on the both side of Mylar tape as β-ray absorbers. The detector was shielded with 10 cm thick lead bricks and 5 cm thick boron-doped polyethene bricks. The data were recorded in the DAQ system of APV8008 and APV8016 made by Techno AP Corporation. The measurements of ^{153}Pr, ^{154}Pr and ^{95}Sr were performed for 8, 11 and 8 hours, respectively.

RESULTS: Here, the preliminarily result of each experiment was briefly described. Concerning ^{154}Pr, a typical coincidence spectrum gated by the 162.4 keV γ-ray, which were previously reported in ref [1], is shown in Fig.1. Ten γ-rays including previously proposed the 70.8, 794.4 and 895.6 keV γ-rays [1] and Nd Kx-rays were in coincidence with the 162.4 keV γ-ray. In addition, the sum peak of 162.4 + 169 keV was observed in the add-back spectrum, it means they are cascade relation each other.

Concerning ^{153}Pr, eight excited levels and nineteen γ-rays were newly identified in addition to the previous result [2]. The half-life value of isomeric state of 191.7 keV was determined to be 1.13(10) μs with the β-γ time difference method. This value is in agreement with the previous value of 1.06(5) μs.

From the results of the half-life of the 1088 keV level in ^{95m}Y through the β-decay of ^{95}Sr by the β-γ time difference is method, the higher counting rate is, the shorter the result became. It was found desirable to keep the count rate below 5 kcps in this method.

Further analyses for construction of the decay schemes of ^{153}Pr and ^{154}Pr are in progress.
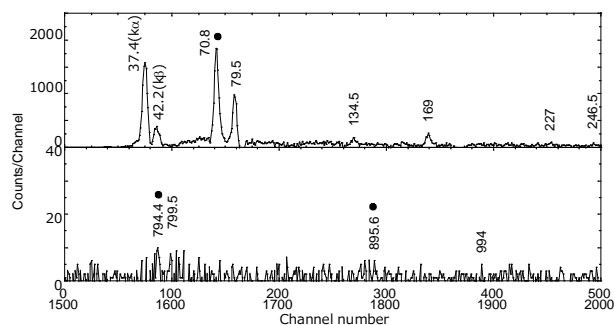

Fig. 1. Coincidence spectrum gated by the 162.4 keV γ-ray in the decay of ^{154}Pr. The closed circles indicate the reported γ-rays [1].

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Linear Polarization Measurement for γ Rays from [148]Pr

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INTRODUCTION: Gamma-ray multipolarities are one of the most important properties for nuclei because they are necessary in discussion of nuclear structures. For determination of the γ-ray multipolarities, the linear polarization is useful, and is usually measured using an asymmetry of Compton scattering events. In recent years, we are trying the linear polarization measurements for short-lived β-decaying nuclei using a clover HPGe detector [1]. The clover detector consists of four large Ge crystals packed closely, and the size of each crystal is 80 mm in diameter and 90mm in length. In this technique, a Compton scattering asymmetry A in orthogonal directions is determined by means of coincidence measurements. After correcting the A-value using the polarimeter sensitivity Q, the value of A/Q is compared with the degree of polarization which has been calculated for various multipolarities.

In AY2018, we tried linear polarization measurements for [146]La produced by on-line isotope separator KUR-ISOL [2], and succeeded in observation of the Compton scattering asymmetry for 258-410 keV E2-E2 γ cascade. In this report, we present the preliminary results obtained for [148]Pr.

EXPERIMENTS: The detection system consisted of the clover detector as a polarimeter, a coaxial 60% (relative efficiency) HPGe detector as a directional detector, and a coaxial 38% HPGe detector to deduce correction factors of detection efficiencies. The directional detector was placed perpendicular to the clover detector, and was used to define a *reference plane*. A detector-to-source distance was 10 cm for all detectors. The preamplifier signals from the detectors were processed by a VME-based data acquisition system, and the data on the pulse height and the detection time were recorded in event-by-event mode.

Gamma rays from the $β^-$ decay of [148]Pr (half-life of 2.1 min) were measured at KUR-ISOL. The [148]Pr nuclei were produced by the thermal-neutron-induced fission of [235]U. The fission products thermalized in the target chamber were transported by a He-N_2 mixture gas jet stream to a surface-ionization-type ion source. After ionization to the chemical form of $[148]Pr[16]O^+$, the nuclei were extracted, accelerated to 30 keV, and mass-separated. The mass-separated beams were implanted into an aluminized Mylar tape in a tape transport system. The source was periodically moved to a detector port with time intervals of 220 s. The measuring times were 73 h and 11 h under a 1-MW and 5-MW operation condition, respectively.

RESULTS: The list data were analyzed using an off-line sorting program. In this analysis, we considered data recorded within 460 ns as a coincident event, and focused on the 302-451 keV and 302-1358 keV γ-ray cascades. The former cascade is known as an E2-E2 transition, while the multipolarity of the 1358 keV γ ray in the latter cascade is unknown. The 1358-keV γ ray is a transition from a 2^+ to a 2^+ level, so that the multipolarity is expected to be M1 or E2.

From coincidence spectra gating on the 302-keV γ ray, we obtained the asymmetry value A of 0.045(17) for the 451-keV γ ray. Because the theoretical value of polarization P is 0.1667 for the E2-E2 cascade in the 4^+-2^+-0^+ spin-parity sequence, the sensitivity Q was found to be $Q = A/P = 0.27(10)$. As shown in Fig.1, this Q-value agrees with those obtained in the previous experiments. For the 1358-keV γ ray, the asymmetry value of $A = -0.008(15)$ was observed. Because the sensitivity is about 0.16 for 1358 keV (Fig.1), the degree of polarization was deduced to be $-0.05(10)$. Here, the theoretical P-values are 0.43 and 0.08 if the 1358-keV γ ray is a M1 and E2 transition, respectively. Our result favors the multipolarity of the 1358-keV γ ray is E2, while the experimental uncertainty is large.

CONCLUSIONS: Compton scattering asymmetries were observed for γ rays from [148]Pr. The result suggests the 1358-keV γ-ray is an E2 transition.
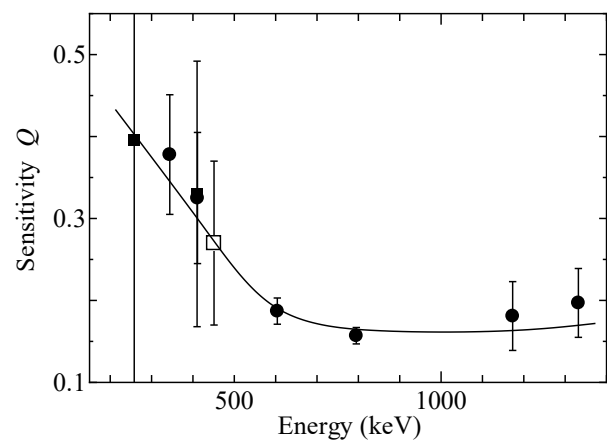
Fig. 1. Energy dependence of the polarization sensitivity. Open and filled marks show data obtained in this experiment and previous measurements using ^{60}Co, [134]Cs, [152]Eu and [146]La [3], respectively. The curve is only a guide to the eye.

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Measurement of the Internal Pressure in Ultrafine Bubbles
by Angular Correlation Technique

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INTRODUCTION: Ultrafine bubbles, the gaseous cavities with diameters less than one micrometer [1], recently attracts a lot of attention because of their multifunctionalities. While applications of ultrafine bubbles are extended in a wide variety of fields, fundamental studies on ultrafine bubbles itself are not well extended because of its small size, smaller than the wavelength of radiant rays.

In nuclear physics, the measurements of angular correlations of iodine isotopes in Xe gas reportedly show large dependences on the pressure of Xe gas [2][3][4], thus suitable as the probe for the pressure inside the gaseous cavity.

We have reported the first measurement of the angular correlation of ^{125}I induced inside the Xe ultrafine bubbles induced by the neutron irradiation at Kyoto University Research Reactor (KUR) for the determination of the internal pressure [5].

In this paper, the progress in the analysis of the previous measurement and the current status of the study are described.

EVALUATION OF INTERNAL PRESSURE OF ULTRAFINE BUBBLE: In the previous report, $A_{22}\overline{G_{22}}(\infty)$ for 55-188 keV cascade in ^{125}I in Xe- ultrafine bubble was determined to be +0.097 ± 0.037 by taking into account the corrections of detector solid angles, size of the source and the contribution of ^{125}I existing as the solute in the water [5]. This time, the contribution of ^{125}I existing as the solute in the water was evaluated in more detail by using the event numbers of 55-188 keV cascade in ^{125}I.

The counting number for the saturated water irradiated at 5 MW was only 18.25 ± 0.64% of that obtained from the ultrafine bubble sample irradiated at the same time, therefore Xe was eventually maintained inside the ultrafine bubbles at the time of the angular correlation measurement for samples irradiated at 5 MW.

It should be noted that the counting number for the Xe saturated water is only 14.53±0.50% of that irradiated at 1 MW. The difference in the counting numbers observed between these Xe saturated water is the result of the difference in the elapsed time after the first repacking. The samples irradiated at 5 MW spent two days more before irradiation and Xe existed as the solute went out of the water and was accumulated at the vacant 0.5 cm^3 space in the PP cylinder for this additional two days. Even though Xe
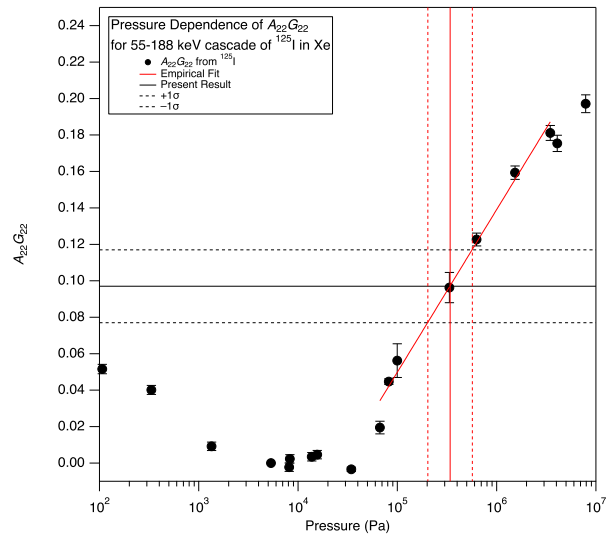

Fig. 1 Pressure dependence of $A_{22}\overline{G_{22}}(\infty)$ of ^{125}I in Xe gas.

nuclei at the vacant space were also activated at the reactor, they went away during the repacking process for the angular correlation measurement. By taking the ratio of counting numbers between the saturated water and ultrafine bubble water, $A_{22}\overline{G_{22}}(\infty)$ was re-evaluated to be +0.097 ± 0.020. The internal pressure of the Xe-ultrafine bubble was obtained to be $3.4^{+3.3}_{-2.3} \times 10^5$ Pa (Fig. 1).

PRESSURE DEPENDENCE OF $A_{22}\overline{G_{22}}(\infty)$ OF ^{125}I IN Xe-ULTRAFINE BUBBLE: Since the internal pressure of ultrafine bubbles should be equal to the pressure from the outside including the water pressure, $A_{22}\overline{G_{22}}(\infty)$ should be changed once the outside pressure is changed. This should also give a clue to the inside temperature of ultrafine bubbles by combining the size of ultrafine bubbles. The first trial of the angular correlation measurement for the sample that was pressurized by Xe-gas was performed, and the analysis is on the way.

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Temperature-Dependent Polaronic Local Structures in $La_{0.7}Ca_{0.3}MnO_3$ Observed through Spin Relaxation of TDPAC Probe Nuclei

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INTRODUCTION: It is well known that a perovskite manganese oxide $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) exhibits the effect of colossal magnetoresistance, and it has received high expectations for application to a variety of functional magnetic materials. For its practical application, it is essential to investigate the magnetotranport phenomenon of the oxide by obtaining information on the local field at each site on an atomic scale in addition to its bulk properties. Especially, the *A* site information is very important because magnetoresistance of $R_{1-x}A_xMnO_3$ is governed largely by doping of *A*-site ions. For a detailed investigation of the *A*-site field, we employed time-differential perturbed angular correlation (TDPAC) spectroscopy with the $^{111m}Cd(\rightarrow^{111}Cd)$ probe [1]. Because the ionic radius of the Cd ion is close to those of La and Ca, the probe nucleus can selectively occupy the *A* site, which allows us to observe the change of the local fields at temperatures above and below T_C (~ 250 K). In the present work, we report a successful observation of dynamic motion and freezing of Jahn-Teller (JT) polarons formed by e_g electrons on Mn ions.

EXPERIMENTS: Stoichiometric amounts of La_2O_3, MnO_2, and $CaCO_3$ were mixed well in a mortar, and the powdery mixture was calcined at 1273 K for 12 h. The sample was again ground to uniformity, and it was pressed into a disk. The disk was then sintered at 1473 K for 96 h. Successful synthesis of LCMO was confirmed by powder X-ray diffraction patterns and magnetization measurements.

Neutron irradiation was performed for cadmium oxide (CdO) enriched with ^{110}Cd in Kyoto University Reactor to produce radioactive ^{111m}Cd by a neutron capture reaction. The radioactive $Cd(^{111m}Cd)O$ powder was mixed well with the LCMO prepared in advance, and the pressed disk was sintered in vacuum at 1373 K for 45 min. TDPAC measurements were carried out for the $^{111m}Cd(\rightarrow^{111}Cd)$ probe on the 151-245 keV cascade γ rays with the intermediate state of $I = 5/2$ having a half-life of 85.0 ns. In the present work, we obtained the perturbed angular correlation as a function of the time interval of the cascade γ-ray emissions by the following expression:

$$A_{22}G_{22}(t) = \frac{2[N(\pi,t) - N(\pi/2,t)]}{N(\pi,t) + 2N(\pi/2,t)}, \qquad (1)$$

where A_{22} denotes the angular correlation coefficient, $G_{22}(t)$ the time-differential perturbation factor as a function of the time interval t between the cascade γ-ray emissions, and $N(\theta, t)$ the number of the delayed coincidence events observed at an angle θ. The measurements were performed at different temperatures.

RESULTS: The TDPAC spectra of $^{111m}Cd(\rightarrow^{111}Cd)$ embedded in LCMO are shown in Fig. 1. We found that the room-temperature spectrum consists of two different components reflecting a distorted and a less distorted sites having the fractional ratio of 7 : 3. The ratio agrees with that of Mn^{3+} and Mn^{4+} abundances, suggesting that the former local distortion arises from JT polarons formed by localized e_g electrons on Mn^{3+} ions. This inference is supported by the observation that the oscillatory structure of the distorted component completely disappeared from the spectra at 201 K and 77 K ($< T_C$). The disappearance of the oscillation of the distorted component below T_C can be explained by the fast relaxation of the probe nucleus caused by fluctuation of the polaronic local lattice dragged by the conduction electrons in the ferromagnetic metal phase. At helium temperature, however, the TDPAC spectrum consists of a single component with an averaged quadrupole frequency, indicating freezing of the fluctuation. This observation suggests that the lattice can no longer follow the electron movement at this low temperature. The present work shows significance of nonmagnetic probes in the study of local fields in magnetic materials.
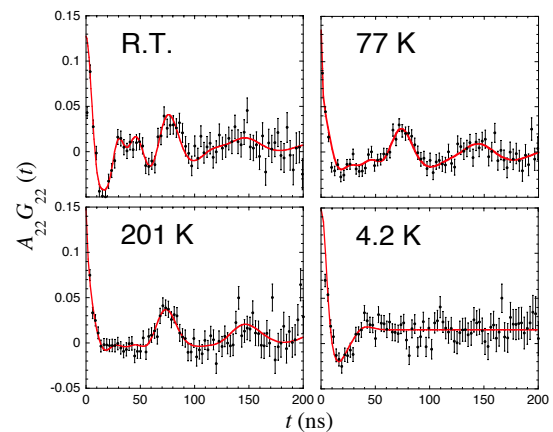

Fig. 1. TDPAC spectra of $^{111m}Cd(\rightarrow^{111}Cd)$ in $La_{0.7}Ca_{0.3}MnO_3$ measured at temperatures indicated.

REFERENCE:
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PR1-6   Local Structure of In Impurities Doped in SrTiO$_3$ Studied by TDPAC Method

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INTRODUCTION: Strontium titanate (SrTiO$_3$) is a cubic perovskite oxide. SrTiO$_3$ doped with metal ions as impurities exhibits a wide variety of electronic properties. Especially, SrTiO$_3$ doped with trivalent metal ions at Ti^{4+} site has been attracted much attention because of their excellent photocatalytic activity [1]. For a practical use of SrTiO$_3$, it is necessary to obtain more microscopic information on the impurity site. Therefore, we investigated the local structures at the In^{3+} site in SrTiO$_3$ by means of the time-differential perturbed angular correlation (TDPAC) method using the ^{111}Cd($\leftarrow^{111}$In) probe.

EXPERIMENTS: Polycrystalline SrTiO$_3$ sample was prepared from stoichiometric mixture of SrCO$_3$ and TiO$_2$. The powders were mixed in a mortar and pressed into a disk. For TDPAC measurements, commercially available ^{111}In solution was added in droplets onto the disk. The disk was sintered in air at 1473 K for 24 h for preparation of doped SrTiO$_3$ sample. It was confirmed from the powder XRD pattern for the nonradioactive sample that calcination at 1473 K for 24 h is sufficient to synthesize a single phase SrTiO$_3$. The TDPAC measurement was carried out for the 171-245 keV cascade γ rays of ^{111}Cd($\leftarrow^{111}$In) probe with the intermediate state of $I = 5/2$ having a half-life of 85.0 ns.

RESULTS: Figure 1 shows the TDPAC spectrum of ^{111}Cd($\leftarrow^{111}$In) probe in SrTiO$_3$. The measurement was performed at room temperature. The directional anisotropy on the ordinate, $A_{22}G_{22}(t)$, was deduced with the following simple operation for delayed coincidence events of the cascade:

$$A_{22}G_{22}(t) = \frac{2\left[N(\pi,t) - N(\pi/2,t)\right]}{N(\pi,t) + 2N(\pi/2,t)}. \quad (1)$$

Here, A_{22} denotes the angular correlation coefficient, $G_{22}(t)$ the time-differential perturbation factor as a function of the time interval, t, between the relevant cascade γ-ray emissions, and $N(\theta,t)$ the number of the coincidence events observed at angle, θ. The oscillatory structure observed in Fig.1 reflects electrostatic interactions between the probe nucleus and the extranuclear field because the sample consists of no magnetic materials. We thus performed a least-squares fit to the spectrum in Fig. 1 with $G_{22}(t)$ expressed as

$$G_{22}(t) = \sigma_{2,0} + \sum_{n=1}^{3} \sigma_{2,n} \cos(\omega_n t). \quad (2)$$

For all symbols in eq.(2), refer to our previous paper[2]. The spectrum in Fig. 1 can be reproduced by a fit with three unique quadrupole frequencies. As for one of these components, the quadrupole frequency shows a zero. On the basis of the cubic structure of SrTiO$_3$, this signifies the ^{111}Cd($\leftarrow^{111}$In) probes occupy defect-free substitutional Sr or Ti sites in SrTiO$_3$. The remaining two components shows two well-defined EFG values which is characterized by the quadrupole frequencies and its distributions $\omega_{Q1} = 48.9(1)$ Mrad/s, $\delta_1 = 0\%$ and $\omega_{Q2} = 52.1(2)$ Mrad/s, $\delta_2 = 0\%$, respectively. These non-zero frequencies imply that defects were associated with ^{111}In probes. It has already been reported that the oxygen vacancy is formed by substitution of trivalent metal ions such as Fe^{3+} ion into Ti^{4+} for charge compensation [3]. Therefore, we assume that part of the ^{111}In probes is associated with the oxygen vacancy, substituting at Ti or Sr site. For more information on the microstructure of SrTiO$_3$, TDPAC measurements with ^{111}Cd($\leftarrow^{111m}$Cd) probe are now in progress.
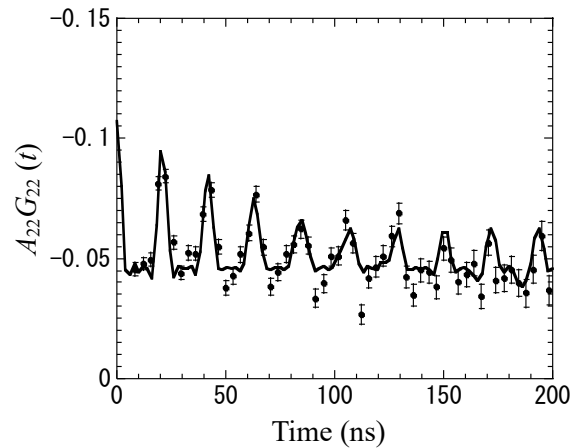
Fig. 1. TDPAC spectrum of ^{111}Cd($\leftarrow^{111}$In) probe in SrTiO$_3$ at room temperature. The line is the result of a least-squares fit with eq.(2).

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