I-1. PROJECT RESEARCHES

Project 3

PR3 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 second year of the project.

The research subjects reported here are as follows:

30P3-1 Technique of transferring radioactive atomic nuclei implanted in dry ice film

30P3-2 Search for isomer of fission product ^{144}La through the β^-–decay of ^{144}Ba by means of internal conversion electron measurements

30P3-3 Compton scattering asymmetry observed for γ rays from ^{146}La

30P3-5 Direct measurement of the internal pressure in ultrafine bubbles by angular correlation technique

30P3-6 Dynamic behavior of impurity indium ions in magnetite

30P3-7 Observation of local fields at the ^{111}Cd(←^{111m}Cd) sites in $CdTi_{0.95}Co_{0.05}O_3$

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*. (30P3-1) attacked this problem using a $^{146}LaO^+$ radioactive ion beam (two ^{146}La states with a half-life of 10 s and 6 s) available at KUR-ISOL and an apparatus designed for the present work. The removal efficiency was at most 30% and how to improve is now under consideration.

At KUR-ISOL, M. Shibata *et al*. (30P3-2) detected γ-rays and internal conversion electrons emitted in the β^- decay of ^{144}Ba (half-life of 11.5 s) using a 31% HPGe detector and a cooled Si(Li) detector, respectively, in order to search for an unknown isomer state in an odd-odd nucleus ^{144}La (a daughter of ^{144}Ba), which is in turn expected to be able to solve the so-called Q_β puzzle. They proceeded with analysis of data previously obtained and newly obtained. At present they could not observe their expected isomer, while they could determine several internal conversion coefficients and detected new γ-rays.

Y. Kojima *et al*. (30P3-3) have been trying to measure the linear polarization of γ-rays from short-lived β decaying nuclei using a clover detector as a Compton polarimeter and additional coaxial Ge detectors in order to obtain the multipolarities of those γ-rays. In AY 2017, they could not obtain clear Compton scattering asymmetry for the 258- and 410-keV γ rays from ^{146}La (half-life of 6.3 s) produced at KUR-ISOL. This was due to large chance coincidences with intense background radiations, in particular, γ rays from the beam collection port. In AY2018, they reduced background radiations by shielding the clover detector and could successfully obtain values of the polarization sensitivity for both the 258- and 410-keV γ rays from ^{146}La, consistent with those measured off-line for other long-lived nuclei, although the obtained values have large uncertainties.

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 live more than a month. Employing time-integral perturbed angular correlation (TIPAC) technique for ^{125}I(←^{125}Xe), M. Tanigaki *et al*. (30P3-5) 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.

More than three decades ago, magnetic hyperfine field at ^{111}Cd(←^{111}In) chemically doped in Fe_3O_4 was measured with time-differential perturbed angular correlation (TDPAC) technique to study the supertransferred magnetic field from the magnetic (Fe) ions via the oxygen ions to the nonmagnetic (Cd) ions. The location of In was assigned to the tetrahedral (*A*) site. However, recently, a paper was published stating that the location of In was the octahedral (*B*) site. W. Sato *et al.* succeeded in obtaining TDPAC spectra with damping free for the first time, concluded that the location of In is the *A* site, and moreover obtained a signal indicating that at high temperatures some In ions in the *A* sites move to the *B* sites, which is consistent with a density functional theory calculation. In order to observe this In motion at high temperatures, W. Sato *et al.* (30P3-6) synthesized 0.5 at.% In-doped Fe_3O_4 and took a ^{111}Cd(←^{111m}Cd) TDPAC spectrum for this oxide at 900 K (above the Curie temperature of 858 K). As anticipated, they could observe ^{111}Cd nuclear spin relaxation that was considered to be caused by thermally activated In motion.

In some cases, perovskites oxides ABO_3 induce ferroelectricity or assume better ferroelectricity when they are doped with a small amount of metal ions different from the constituent metals. Distortion of the crystal structure by doping can cause these changes. S. Komatsuda *et al.* (30P3-7) synthesized $CdTiO_3$ and $CdTi_{0.95}Co_{0.05}O_3$ and searched any structural difference between the two oxides by applying the room-temperature TDPAC method with ^{111}Cd(←^{111m}Cd) as local probes. They found no noticeable difference in the TDPAC spectra.

PR3-1 Technique of Transferring Radioactive Atomic Nuclei Implanted in Dry Ice Film

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INTRODUCTION: The nuclear charge radius is one of the fundamental parameters that gives information about the effective interactions on nuclear structure, which has been investigated by electron scattering, measurements of muonic atom X-ray energies and optical isotope shifts. Recently the study for unstable nuclei has been advanced by optical isotope shifts, while the study using other methods is little progress mainly because a large amount of radioactivity is needed [1]. The results obtained from these three methods provide complementary information each other. Therefore, R&D for more efficient systems of electron scattering and muonic X-ray measurements is being promoted.

Measurement of the emitted X-ray energy from muonic atoms is a powerful method for the investigation. The muon and the electron have similar properties except for their mass difference, thus a negative muon (μ^-) can be captured in a Bohr orbit of a nucleus just as an electron and then a muonic atom is formed. Because the mass of a muon is 207 times the electron mass, its lower orbits are close to the nucleus and the energies of the states depend sensitively on the nuclear charge distribution. By means of the muonic X-ray spectroscopy, therefore, information on the nuclear charge distribution can be obtained. 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 (D) film in which nuclei of interest (A) are implanted beforehand, thereby muonic atoms being formed through the highly-efficient muon transfer reaction: $\mu^-D + A \rightarrow D + \mu^-A$. The feasibility of this method was demonstrated and promising results were obtained [2]. However, several problems have to be solved before this method is applied to experiments for unstable nuclei. In this study, one such problem, concerned with handling of residual radioactivities in deuterium film after experiments, in particular their highly efficient recovery, was approached experimentally 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 copper blocks and one CO_2 gas diffuser in the vacuum chamber. Each copper block can be cooled with LN_2 flow internally and movable independently in a horizontal direction between the center of the chamber which is on the axis of the RI beam and the γ-ray measurement port while keeping LN_2 flow. Dry ice film was formed on the surface of one cooled block (catcher) by sprayed CO_2 gas through the diffuser. In the present work, about 10^6 ions of $^{146}LaO^+$ ($T_{1/2}$ = 10 s and 6 s) were implanted into dry ice film on the catcher in every run. After the implantation, the other pre-cooled block (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 ($T_{1/2}$ = 14 m, daughter of ^{146}La) and ^{146}Pr ($T_{1/2}$ = 24 m, daughter of ^{146}Ce) ware released from the catcher and were re-trapped on the trap together with CO_2. The trapped efficiency was measured by detecting the γ-rays emitted from each of the blocks.

RESULTS AND DISCUSSION: In this work, the trapped efficiencies were measured by changing the formation condition of dry ice film on the catcher. Specifically, a single layer film or a multilayer film was formed while changing the spraying speed and total amount of carbon dioxide gas. However, since it takes a long time for one run, the experiments under so many conditions were not performed. Incidentally, a multilayer film was made by alternately repeating formation of dry ice film and ion implantation.

While the detail data analysis also is in progress, the trapped efficiencies remained at most 30% in any run. (For now, the efficiency is defined as the ratio of the radioactivities on the trap to those remaining on the catcher, and the contribution of the radioactivities escaped somewhere other than the trap is not taken into account.) From a practical point of view, these efficiencies are not so high, but the most notable point is that radioactivities remained on the catcher in all runs.

The range in dry ice of $^{146}La^+$ whose acceleration energy is 27 keV is about 27 nm according to SRIM code, on the other hand, a visually recognizable dry ice film was formed on the catcher in any run. That is, the thickness of the dry ice film is considered to be sufficient to stop ^{146}La ions within the film. The cause for the remaining radioactivities on the catcher is not clear. So far, the possible reasons are (1) formation of a non-ideal dry ice film, or (2) chemical interaction between the implanted atoms and the catcher surface material in the process of sublimation of dry ice, etc. If there is a cause in (2), it means that an important issue has been found for applying the solid hydrogen method to experiments on unstable nuclei.

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PR3-2 Search for Isomer of Fission Product ^{144}La through the β⁻-decay of ^{144}Ba by means of Internal Conversion Electron Measurements

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INTRODUCTION: Determination of precise decay scheme of fission products of ^{235}U are very important for both nuclear engineering and nuclear physics. Information of nuclear structure are also useful to study nucleosynthesis. Especially, relatively long-lived isomeric states are important to evaluate the r- or s-process of neutron capture in nucleosynthesis. In the last year, we focused on to search for unobserved isomeric states of some elements which are expected from the systematics for doubly-odd nuclei around mass number 150 with an energy-sum γ-ray spectrometry using a clover detector. [1] In this year, to solve the "Q_β-puzzle" in doubly-odd ^{144}La [2], internal conversion electron (ICE) measurements with a cooled Si(Li) detector were carried out to search for isomeric transitions and to determine of their multipolarities, and also data analyses of γ singles, γ–γ and x-γ coincidences were proceeded.

EXPERIMENTS: The experiments were carried out using the on-line mass separator KUR-ISOL at the Kyoto University Reactor. 93%-enriched $^{235}UF_4$ target of 72 mg were inserted in a through-hole facility and the nuclei of interests were produced with the thermal neutron-induced fission of ^{235}U. The fission products were transported by a He-N_2 gas jet system and were ionized by a thermal-ionization type ion source. The mass-separated radioactive beam was implanted in an aluminized Mylar tape set in the computer controlled tape transport system. The tape was moved with a predetermined period to reduce the background from their daughter nuclei. At the measuring position, the γ rays and ICEs were measured, simultaneously, with a 31% HPGe detector (ORTEC GMX, 57 mm⌀ × 68 mm*L*, energy resolution 2.4 keV at 1408 keV) and a cooled Si(Li) detector (Eurisys, 500 mm^2 × 6 mm*L*), respectively. The detectors were set at 180° direction and the source to detector distances were set at 15 mm and 13 mm for the Ge and the Si(Li) detector, respectively. The detailed setup of the detector were described elsewhere [3]. The data were taken with the list mode including the time information using a VME based data acquisition system (A3100 by IWATSU). The collection-measurement cycle was set at 22 s - 22 s for ^{144}Ba ($T_{1/2}$ = 11.5 s), and was repeated for 10 hours. The detectors were shielded with 10 cm-thick lead blocks and borated polyethylene ones outside them in order to reduce the background neutrons and γ radiations. For the efficiency calibration in the geometrical condition as well as the energy calibration, the daughter nucleus of ^{144}La and ^{146}La were measured for 8 hours, respectively, since these nuclei contain well-evaluated pure E2 transitions.

RESULTS: The measured singles ICE spectrum in the decay of ^{144}Ba was shown in Fig. 1. The ICE peaks were observed up to the 260 keV transition. The internal conversion coefficients (ICCs) were determined with the ratio of the measured counts of ICEs to the corresponding γ-rays by normalized by the pure E2 transitions as described above. The ICC of the 260 keV transitions were determined and only the upper limit values of the 418, 422, 431, 516, 571 and 584 keV transitions were determined owing to low statistics. On the other hand, the ICCs of the 43, 69, 82, 104, 115, 153 keV transitions were determined from the x-γ coincidence method. Preliminarily, adding the result of γ-γ coincidence relations, 115 γ-rays including the newly observed 28 ones and the multipolarities of 13 transitions and 28 excited levels were incorporated in the decay scheme. These results were mostly in agreement with the previous results [4]. Nevertheless, isomeric transitions to give the answer of "Q_β-puzzle" were not observed in this experiment.

CONCLUSIONS: The measurements of internal conversion electrons and the analyses of γ-rays of doubly-odd fission product ^{144}La were carried out through the β-decay of ^{144}Ba. The expected long-lived isomer and γ transition having higher multipolarity could not be observed, consequently, the Q_β-puzzle in ^{144}La could not be solved. The construction of detailed decay scheme is in progress.

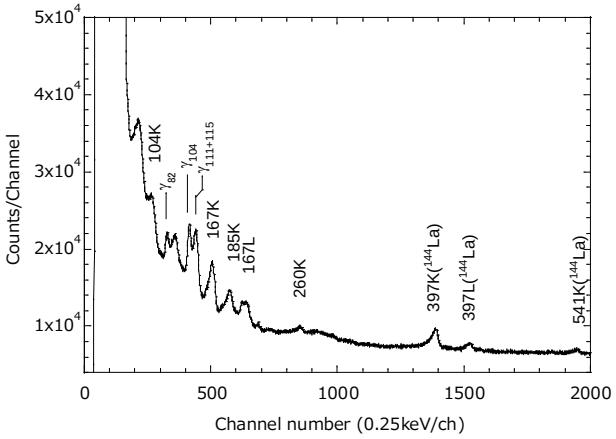

Fig. 1. Singles spectrum of internal conversion electrons in the decay of ^{144}Ba measured with a cooled Si(Li) detector. The electron peaks for daughter nucleus ^{144}La, which were used for the calibration of the detector, are also observed.

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PR3-3 Compton Scattering Asymmetry Observed for γ Rays from ^{146}La

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INTRODUCTION: Gamma-ray multipolarities are one of the most important data for nuclei because they relates to γ-ray emission process. The linear polarization is useful in determining the γ-ray multipolarities, and is usually measured using a Compton polarimeter consisting of two or more γ-ray detectors. Our group is trying to apply a clover Ge detector [1] to the linear polarization measurements for short-lived β-decaying nuclei. 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 *P* which has been calculated for various multipolarities.

In AY2017, we tried linear polarization measurements for nuclides produced by on-line isotope separator KUR-ISOL [2]. However, we could not clearly observe the Compton scattering asymmetry [3]. As a result of detailed analysis, we found that the main reason was due to chance coincidences with intense background radiations.

In this report, we present the preliminary results of improved experiments in AY2018.

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 clover detector contains four large Ge crystals packed closely. The size of each crystal is 80 mm in diameter and 90 mm in length. 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 data on the pulse height and the detection time were recorded in event-by-event mode.

Gamma rays from ^{146}La (half-life of 6.3 s) were measured at KUR-ISOL. The ^{146}La nuclei were produced by the thermal-neutron-induced fission of ^{235}U. The fission products were thermalized in the target chamber, and transported to a surface ionization-type ion source using gas jet stream. After ionization, 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 13 s. The measuring time was 13 h.

To reduce background radiations, in particular, γ rays from the beam collection port, heavy shield was placed around the clover detector. The shield was extremely effective. Counts due to the background γ rays were reduced to about 1/100 compared with those in experiments in AY2017.

RESULTS: The list data were analyzed using an off-line sorting program. In this analysis, we focused on E2-E2 γ cascade (258-410 keV γ cascade) in the β decay of ^{146}La. Signals recorded within 570 ns were taken as coincident events.

From coincidence spectra gating on the 258-keV γ ray, we found that the Compton scattering perpendicular to the reference plane occurs 6% more often than that parallel to the reference plane for the 410-keV γ ray. After correcting detection efficiencies, the asymmetry value *A* was found to be 0.055(27). Asymmetry was also observed for 258-keV γ ray. The sensitivity $Q = A/P$ was deduced for each γ ray, and compared with values obtained using standard radioactive sources of ^{60}Co, ^{134}Cs and ^{152}Eu. As shown in Fig. 1, the sensitivities obtained in the on-line measurement agree with the values obtained in the off-line measurement, while the statistical uncertainties for ^{146}La are somewhat large.

CONCLUSION: We succeeded in observation of asymmetry of Compton scattering events in the on-line measurements. We plan to measure γ rays from nuclei produced by KUR-ISOL to deduce unknown multipolarities.

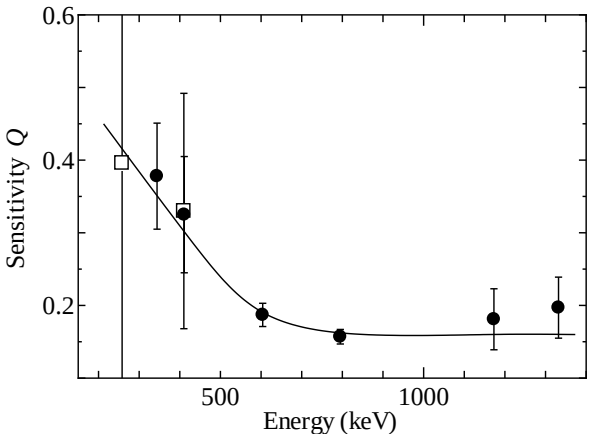

Fig. 1. The polarization sensitivity as a function of incident γ-ray energy. Open and filled marks show data obtained at KUR-ISOL and off-line measurements, respectively. The curve is only a guide to the eye.

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PR3-4 **Direct 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 micro meter [1], recently attract a lot of attentions because of their multifunctionalities. While applications of ultrafine bubble are extended in wide variety of fields, fundamental studies on ultrafine bubble itself are not well extended because of its small size, smaller than the wave length of radiant ray. Some are even suspicious of the existence of ultrafine bubble itself.
In nuclear physics, the angular correlations measurements of iodine isotopes in Xe gas were reportedly difficult [2][3] because of their large dependences on the pressure of Xe gas [4][5][6]. This means that the gas pressure in a ultrafine bubbles of Xe can be directly measured by a technique of nuclear physics, i.e., the angular correlation measurement of radioactive iodines inside ultrafine bubbles, .
In this paper, the measurement of the internal pres- sure in Xe ultrafine bubble through the angular correla- tion measurement of 55 - 188 keV γ-γ cascade in ^{125}I is described.

EXPERIMENTS: The water containing ultrafine bubbles of natural Xe gas with the average diameter of 200 nm in pure water was generated via the pressurized dissolution method. 4 cm^3 of ultrafine bubble water and saturated solution of natural Xe were separately packed to 4.5 cm^3 polypropylene (PP) cylinders for the irradiation at the reactor and were irradiated by thermal neutrons with the total dose of 6.9×10^{16} n/cm^2 at the slant exposure tube of Kyoto University Research Reactor (KUR) to activate ^{125}Xe ($T_{1/2}$ = 16.9 h) through the thermal neutron capture reaction of ^{124}Xe(n,γ)^{125}Xe. The γ-γ cascades were detected by a conventional fast-slow coincidence method with a 1 cm^3 CdZnTe detector (KROMEK GR1-A) and one of two 1.5-inch ϕ × 1-inch thick BaF_2 detectors. Two BaF_2 detectors are placed at the fixed angle of 90°, and a CdZnTe detector on a turntable moves around the PP cylinder containing the irradiated sample. With this configuration, the angular correlations of both 90° - θ and 180° + θ are measured at a time. The correlation function of γ-γ cascade under the existence of hyperfine interactions is given as,

$$W(\theta) = 1 + A_{22}\overline{G_{22}(\infty)}P_2(\cos\theta),$$

where A_{22}, $\overline{G_{22}(\infty)}$ are asymmetry parameter and the attenuation factor caused by the hyperfine interaction, respectively. $A_{22}\overline{G_{22}(\infty)}$ for 55-188 keV cascade in ^{125}I was determined to be +0.097 ± 0.037 from the obtained angular correlation (Fig. 1), by taking into account the corrections of detector solid angles, size of source and the contribution of ^{125}I existing as the solute in the water.

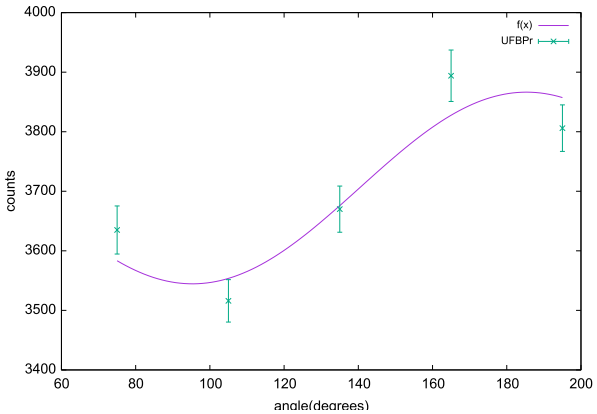

Fig. 1 Angular correlation of 188 - 55 keV γ-ray cascade of 188 keV state in ^{125}I in ultrafine bubble water.

RESULTS: The pressure dependence of $A_{22}\overline{G_{22}(\infty)}$ for the 203 keV state in ^{127}I in Xe gas was well studied by Berek [5], and the result of ^{127}I can be converted to that of ^{125}I by simply applying the ratio of A_{22} for these states [6].
From the obtained pressure dependence of $A_{22}\overline{G_{22}(\infty)}$, the internal pressure of ultrafine bubble is tentatively determined to be $3.4^{+5.4}_{-1.1}\times10^5$ Pa.
The present result is smaller than that expected from Young-Laplace equation, 1.6×10^6 Pa. Some kind of compensation schemes for the present discrepancy are required to maintain an ultrafine bubble. One possible explanation is the repulsive force by the electrostatic charge on the surfaces of ultrafine bubbles. We are planning to measure ζ potential for ultrafine bubble to evaluate the surface charge.

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PR3-5

Dynamic Behavior of Impurity Indium Ions in Magnetite

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INTRODUCTION: Magnetite (Fe_3O_4) is a promising material applicable to functional devices in such fields as spintronics, drug delivery systems, and chemical catalysts. There are many vacant spaces inherent in the compound comprised of the sublattices called tetrahedral *A* site and octahedral *B* site, and these vacancies could make it possible for impurity ions to move around from site to site. In our previous work, we observed thermal migration of impurity indium ions from the *A* site to the *B* site at temperatures higher than the Curie temperature (T_C = 858 K) by means of time-differential perturbed angular correlation (TDPAC) spectroscopy with the ^{111}In($\rightarrow^{111}$Cd) probe [1]. It is expected that this phenomenon possibly leads to impurity-induced ionic conductivity. However, the observation of the movement of In ions was indirect; we merely measured the field at the probe site after the migration of the parent nucleus (^{111}In), namely, after the disintegration to the daughter nucleus (^{111}Cd). It is thus essential to observe the movement of In ions through "the eyes" of witnessing nuclei. For that purpose, in this work, we performed TDPAC spectroscopy for the observation of dynamic motion of impurity In ions adopting the ^{111m}Cd($\rightarrow^{111}$Cd) probe nuclei as the witnessing eyes. In the report, direct evidence of dynamic motion of In ions is presented.

EXPERIMENTS: A stoichiometric amount of In_2O_3 was mixed well with Fe_3O_4 so as to synthesize 0.5 at.% In-doped Fe_3O_4 (IFO). The mixture was pressed into a disk, and sintered in vacuum at 1073 K for 3 h. It was confirmed that any second phase of the In impurities did not appear in its powder X-ray diffraction pattern.

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 in a mortar with IFO prepared in advance, and sintered in vacuum at 1173 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:

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

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.

RESULTS: The TDPAC spectra of ^{111m}Cd($\rightarrow^{111}$Cd) embedded in IFO are shown in Fig. 1. The room-temperature spectrum shows an oscillatory structure characteristic of static magnetic interaction with the internal field. Thus, we analyzed the spectrum with the following time-differential perturbation factor $G_{22}(t)$ assuming a relative width δ to the centroid of the Larmor frequency ω_L:

$$G_{22}(t) = \frac{1}{5}\left\{1 + 2\exp\left(-\frac{1}{2}\delta^2\omega_L^2t^2\right)\cos(\omega_L t) + 2\left[\exp\left(-\frac{1}{2}\delta^2\omega_L^2t^2\right)\right]^4\cos(2\omega_L t)\right\} \quad . \qquad (2)$$

The analytical results are as follows: ω_L = 171.9(5) Mrad/s, δ = 3.6(4)%. The ω_L value shows good agreement with that obtained in the previous work [1], verifying that Cd as well as In ions occupy the *A* site. At 900 K ($>T_C$), we observed exponential relaxation in the spectrum, which signifies nuclear spin relaxation caused by dynamic perturbation arising from the fluctuation of the extranuclear field. Thus we analyzed the spectrum with

$$G_{22}(t) = \exp(-\lambda t) \quad , \qquad (3)$$

where λ is the relaxation constant. Taking into account the unperturbed nature of the spectrum for Fe_3O_4 doped only with the ^{111}In($\rightarrow^{111}$Cd) probe measured at 873 K ($>T_C$), this relaxation should be attributed to the doping effect of macroscopic amount of In. Thus it is evident that the observed nuclear spin relaxation was caused by thermally activated dynamic motion of In ions. The present result shows direct observation of In motion through the eyes of ^{111m}Cd.

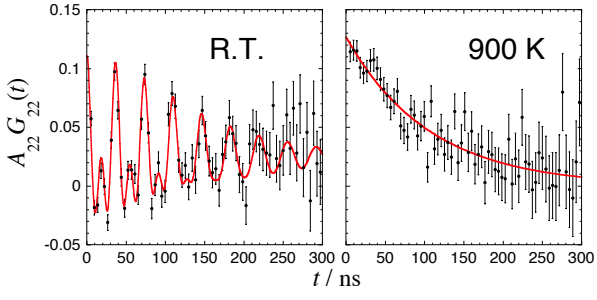

Fig. 1. TDPAC spectra of ^{111m}Cd($\rightarrow^{111}$Cd) in 0.5 at.% In-doped Fe_3O_4 measured at temperatures indicated.

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PR3-6 Observation of Local Fields at the ^{111}Cd(←^{111m}Cd) Sites in $CdTi_{0.95}Co_{0.05}O_3$

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INTRODUCTION: The perovskite oxides ABO_3 such as cadmium titanate ($CdTiO_3$) has been attracting much attention as a ferroelectric oxide, and its technological applications are strongly expected. It is reported that substitution of impurity ions at the *A* or *B* sites induces good ferroelectricity because of a distortion of crystal symmetry[1]. Therefore, it is necessary for a practical use of $CdTiO_3$ to obtain microscopic information on this doping effect. In this study, in order to investigate the effect of Co doping on crystal symmetry, we observed the local structures in $CdTiO_3$ and $CdTi_{0.95}Co_{0.05}O_3$ by means of the time-differential perturbed angular correlation (TDPAC) method using the ^{111}Cd(←^{111m}Cd) probe.

EXPERIMENTS: About 3 mg of CdO enriched with ^{110}Cd was irradiated with thermal neutrons in a pneumatic tube at Kyoto University Reactor, and radioactive ^{111m}Cd was generated by ^{110}Cd(n, γ)^{111m}Cd reaction. The neutron-irradiated CdO powder was then added into stoichiometric amount of TiO_2, Co_3O_4 and nonradioactive CdO powder to synthesize the polycrystalline $CdTiO_3$ and $CdTi_{0.95}Co_{0.05}O_3$ powder. The powders were mixed in the mortar. Each powder was pressed into disks and sintered in air at 1373 K for 45 min, respectively. It was confirmed from the powder XRD pattern for the nonradioactive $CdTiO_3$ and $CdTi_{0.95}Co_{0.05}O_3$ samples that calcining at 1373 K for 45 min is sufficient to synthesize a single phase $CdTiO_3$. The TDPAC measurement was carried out for the 151-245 keV cascade γ rays of the ^{111}Cd(←^{111m}Cd) probe with the intermediate state of I = 5/2 having a half-life of 85.0 ns.

RESULTS: Figure 1 shows TDPAC spectra of ^{111}Cd(←^{111m}Cd) probe in $CdTiO_3$ and $CdTi_{0.95}Co_{0.05}O_3$. The measurements were 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:

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

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 least squares fits to the spectra in Fig. 1 with $G_{22}(t)$ expressed as

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

For all symbols in eq.(2), refer to our previous paper[2]. The electric field gradient (EFG) and asymmetry parameter values obtained for $CdTiO_3$ were estimated to be V_{zz} = 5.5(9)×10^{21} Vm^{-2} (η=0.40). This EFG value shows a good agreement with that obtained for ^{111m}Cd probe in $CdTiO_3$ sample[3]. Since the present spectra can be reproduced by the fit assuming a single component, ^{111m}Cd probes occupy the Cd site in $CdTiO_3$ and $CdTi_{0.95}Co_{0.05}O_3$. For $CdTi_{0.95}Co_{0.05}O_3$ sample, V_{zz} and η values show good agreement with those for $CdTiO_3$ sample: V_{zz} = 5.6(9)× 10^{21} Vm^{-2} (η=0.40). These TDPAC parameters show that substitution of Co ions at Ti site has no effect for crystal symmetry at ^{111}Cd probe. The present experimental result suggests that doping into *B* site has negligible effects on crystal symmetry and microstructure for $CdTiO_3$.

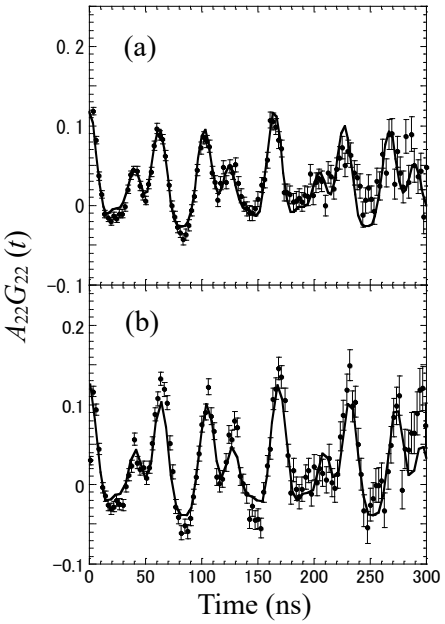

Fig. 1. TDPAC spectra of ^{111}Cd(←^{111m}Cd) probe in (a) $CdTi_{0.95}Co_{0.05}O_3$ and (b) $CdTiO_3$ at room temperature. The line is the result of a least-squares fit with eq.(2).

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