**PR12**   **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 first year of the project. Unfortunately, no experiments in all six research subjects of the project (26P12) were executed owing to the suspension of the reactor operation. Here, we report some results which were obtained in previous periods and have already been published in journals.

The research subjects (PRS) reported here are as follows:

PRS-1 $\beta^-$ Decay of $^{150}$Ce to $^{150}$Pr

PRS-2 Interaction between He and Elements with $A = 140$ in Fe

PRS-3 Atmosphere Dependence of Stability of Local Fields in Al-Doped ZnO

PRS-4 Extranuclear Dynamic Motion of $^{111}$Cd($\leftarrow$ $^{111}$Ag) Doped in AgI Nanoparticles

## Main Results and Contents of This Report

At KUR-ISOL (the on-line isotope separator installed at the Kyoto University Research Reactor), Y. Kojima *et al.* (PRS-1) studied the $\beta^-$ decay of $^{150}$Ce having a half-life of 6.05(7) s produced by the thermal-neutron-induced fission of $^{235}$U. From $\gamma$-ray singles and $\gamma$–$\gamma$ coincidence measurements, they newly found 9 excited states and 37 $\gamma$-rays and constructed a decay scheme containing 18 excited states and 55 $\gamma$-rays which include the newly found excited states and $\gamma$-rays. They concluded that the dominant $\beta$-feeding at the 110-keV excited state of $^{150}$Pr arising from the $\beta^-$ decay of $^{150}$Ce to be an allowed unhindered spin-flip transition. They furthermore proposed the Nilsson configurations of the ground state and the 110-keV excited state of $^{150}$Pr. A part of the results are described in the next page.

In order to examine whether Ce (or rather, La and Ba) and He form complexes having a definite geometrical structure in Fe as suggested in first-principles density functional theory calculations, Y. Ohkubo *et al.* (PRS-2) projected 100-keV $^{140}$Cs$^+$ at KUR-ISOL and then 4-keV He$^+$ using an ion beam gun into an Fe foil and took time-differential perturbed-angular-correlation (TDPAC) spectra at room-temperature of $^{140}$Ce arising through $^{140}$Ba-$^{140}$La from $^{140}$Cs in He-doped Fe, unannealed and annealed in vacuum at various temperatures. They observed no clear signal of such complexes in the TDPAC spectra. However, the TDPAC spectra indicate that Ce and He form complexes having a variety of geometrical structures. Comparison with TDPAC results reported by another research group on $^{111}$Cd arising from $^{111}$In in He-doped stainless steel shows that the parent atoms (La and Ba) of $^{140}$Ce trap He atoms more efficiently than In atoms do, indicating stronger bonding of He to the former atoms, while different from the present case, $^{111}$Cd (In)–He complexes form a unique geometrical structure. A part of the results are described in the following second page.

S. Komatsuda *et al.* (PRS-3) investigated thermal behavior and interacting nature of 100-ppm Al and ~100-ppt In impurities doped in a semiconductor ZnO by means of the TDPAC technique with the $^{111}$In($\rightarrow$ $^{111}$Cd) probe. They observed the following contrasting interactions between Al and In impurities depending on different atmospheric conditions: (1) in air, Al and In impurities irreversibly associate with each other in the process of thermal diffusion of these two species, but (2) in vacuum, these bound states formed in air dissociate by heat treatment at temperature higher than 873 K, and the dissociation reaction is enhanced with increasing temperature. Considering that the dissociation is triggered by the oxygen vacancy formation near the locally associated In–Al structure and using the TDPAC data obtained, they estimated the activation energy of the oxygen–vacancy formation to be 0.72(6) eV. A part of the results are described in the following third page.

Superionic conductivity observed for AgI emerges only at the high-temperature ($\geq$ 420 K) $\alpha$ phase because of temperature-dependent crystal structures, which is a barrier to the practical applications of this compound. Recently, this problem was attacked by a novel technique: powder AgI coated with poly-N-vinyl-2-pyrrolidone (PVP) can drastically enhance the ionic transport property at room temperature. W. Sato *et al.* (PRS-4) studied dynamic behavior of the extranuclear field relative to the $^{111}$Cd($\leftarrow$ $^{111}$Ag) probe nucleus incorporated in AgI, using the TDPAC method. For PVP-coated AgI nanoparticles, they observed nuclear spin relaxation of the probe at room temperature, which indicates that Ag$^+$ ions in the PVP-coated sample make hopping motion from site to site at this low temperature. From the TDPAC data obtained, they estimated the activation energy for the dynamic motion to be 46(10) meV. A part of the results are described in the following fourth page.

# β⁻ decay of $^{150}$Ce to $^{150}$Pr

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**INTRODUCTION:** Decay data provide valuable information about unstable nuclides. In particular, the β-branching ratio $I_\beta$ is one the most important properties from the nuclear structure viewpoint. For even-even $^{150}$Ce, Fogelberg *et al.* briefly reported an intense β-transition to the 110-keV level in $^{150}$Pr [1]. If this β-feeding is an allowed unhindered transition, it will lead to the assignment of the Nilsson orbital for $^{150}$Pr. However, it is uncertain whether this transition is firmly an allowed unhindered transition because of the following reasons. First, ref. [1] did not provide detailed experimental data on $^{150}$Ce. Second, the other published data for $^{150}$Ce are very scarce and partly inconsistent with each other [2,3].

In the present work, decay studies on $^{150}$Ce have been performed using the on-line isotope separator at KUR (KUR-ISOL) [4] to investigate low-energy level structures in $^{150}$Pr. Note that this work was recently published [5]. This report is an outline of the paper.

**EXPERIMENTS:** The $^{150}$Ce nuclides were produced by the thermal neutron induced fission of $^{235}$U. The mass-separated source was periodically moved to a detector port with time intervals of 12.2 s.

Time-dependent γ-ray singles and γ–γ coincidence measurements were performed with two Ge detectors in a measurement period of 46 h. The energy and efficiency calibrations were made using standard γ-ray sources.

**RESULTS:** The half-life and coincidence relationships with the Pr KX-ray were analyzed to assign the parent nuclide of each γ-ray observed in this work. From these analyses, 57 γ-rays including 39 new γ-transitions were found to be due to the decay of $^{150}$Ce.

Energy spectra gated on the γ-rays from $^{150}$Ce were analyzed to establish γ-ray cascade relations. For example, Fig. 1 is a spectrum gated on the 110-keV γ-ray. This spectrum shows that the 110-keV γ-ray is coincident with 13 γ-rays. From these cascade relations and also from energy sum rules, the excited levels in $^{150}$Pr and the γ-ray placements were unambiguously proposed. The decay scheme includes 18 excited levels. Nine levels of them were newly proposed. The 717-keV level seen in ref. [3] was not observed in this work.

The γ-ray intensities relative to that of the 110-keV tran-

sition were deduced from peak counts observed in the singles spectrum or in coincident spectrum. Multipolarities of three γ-rays were also estimated from the intensity ratio between KX and γ-ray peaks observed in the coincident spectrum: $M1/E2$ for the 103- and 154-keV γ-rays, and $E1$ for the 110-keV γ-ray.

Finally, the $I_\beta$ and log-*ft* values were calculated from the transition intensity (γ and conversion electron) imbalance for each excited level. A small log-*ft* value of 4.9 was observed for the 110-keV level. It leads to spin-parity $J^\pi = 1^+$ for this level.

**DISCUSSION:** The very intense β-feeding to the 110-keV level is an allowed transition, and should be explained by selection rules for β-decays of deformed nuclei [6]. Near the Nilsson orbitals observed in the neighboring nuclides, only the spin-flip transition between ν3/2⁻[532] and π5/2⁻[532] satisfies the selection rules. Thus, this β-transition is interpreted as one of the coupled neutron in 3/2⁻[532] orbital in $^{150}$Ce changing to a proton in the vacant 5/2⁻[532] orbital in $^{150}$Pr. This configuration yields $J^\pi = 1^+$, and agrees with our spin-parity assignment. The configuration of π5/2⁺[413]-ν3/2⁻[521] was also proposed for the ground state of $^{150}$Pr from the systematics of the neighboring nuclides and from coupling rules.

**CONCLUSIONS:** A detailed decay scheme of $^{150}$Ce, which includes 37 new γ-rays and 9 new excited levels, was proposed from γ-ray measurements. On the basis of the systematics and observation of the allowed spin-flip transition, the Nilsson configurations were newly proposed for the ground and 110-keV level in $^{150}$Pr.
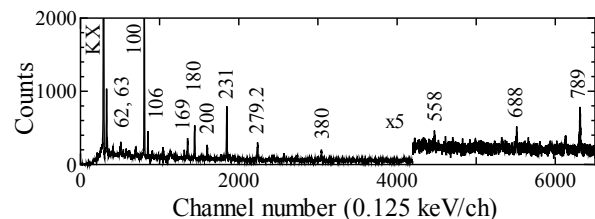


Fig. 1. A γ-ray spectrum gated on the 110-keV γ-transition. Coincident γ-rays are indicated with their energies in units of keV.

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**Interaction between He and Elements with $A$ = 140 in Fe**

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**INTRODUCTION**: As far as we know, the state of impurities He and Ce in Fe has not been studied experimentally until our recent work [1]. This knowledge is important in materials science related to nuclear fusion reactor materials. With a nuclear spectroscopic technique, the time-differential perturbed angular correlation (TDPAC) technique, we obtained the result that the parent atoms (La and Ba) of $^{140}$Ce very efficiently trap He and they form complexes in Fe, which do not have a definite geometrical structure. The present report describes the latter result. See [1] for the full description.

**EXPERIMENTS**: 100-keV $^{140}$Cs$^+$ (~1.4 × 10$^{12}$/0.2 cm$^2$) were projected into an Fe foil at KUR-ISOL. $^{140}$Cs decays through $^{140}$Ba and then $^{140}$La to $^{140}$Ce. We name this sample $^{140}$CeFe. Three additional samples were prepared using the $^{140}$CeFe. The first, second and third were prepared by projecting 4-keV He$^+$ of ~4 × 10$^{13}$, ~5 × 10$^{14}$ and ~2 × 10$^{16}$/cm$^2$ in this sequence into the $^{140}$CeFe. We name them He-doped(1)$^{140}$CeFe, He-doped(2)$^{140}$CeFe and He-doped(3)$^{140}$CeFe, respectively. The profile of the He concentration in Fe overlaps that for $^{140}$Cs. Room-temperature TDPAC spectra of $^{140}$Ce were taken for these four samples.

**RESULTS**: Fig. 1 (from top to bottom) shows the TDPAC spectra obtained at room temperature for $^{140}$CeFe, He-doped(1)$^{140}$CeFe, He-doped(2)$^{140}$CeFe and He-doped (3)$^{140}$CeFe, all four with no annealing. The top spectrum is the one appearing in [2]. As explained there, the oscillation pattern seen in the spectrum is due to a unique static magnetic hyperfine interaction at $^{140}$Ce occupying the Fe substitutional site with no lattice defects nearby (the Larmor frequency equals 1.927(7) Grad/s). Hereafter, we call these $^{140}$Ce "good" $^{140}$Ce. From the value of the amplitude of the oscillation, "good" $^{140}$Ce are only about 30% of the $^{140}$Ce implanted in Fe. In Fig. 1 are also shown the characteristic values for each oscillation component seen in the TDPAC spectra. As the dose of He increases to ~5 × 10$^{14}$/cm$^2$, the oscillation amplitude gets reduced more and more (note that the amplitude is not damped with time), and at the dose of ~2 × 10$^{16}$/cm$^2$ (He-doped(3)$^{140}$CeFe) no clear oscillation is observed. The dose of 2 × 10$^{16}$/cm$^2$ corresponds to 0.15 dpa (displacement per atom) at the maximum. This large number may imply that He ions directly displace the parent atoms of "good" $^{140}$Ce from the substitutional sites. However, based on the fact that the oscillation amplitude is reduced by as much as ~50% even for the

He-doped(1)$^{140}$CeFe (~4 × 10$^{13}$/cm$^2$ He corresponding to only 0.0003 dpa), it is unlikely for He ions to directly displace them. We think that He atoms diffused and bonded to the parent atoms of "good" $^{140}$Ce (interstitial He atoms diffuse much more easily than vacancies: the corresponding calculated activation energies are 0.06 and 0.68 eV, respectively). As the dose of He increases, the number of the parent atoms of "good" $^{140}$Ce decreases. If $^{140}$Ce–He complexes take one or a few definite geometrical structures, these would be reflected in the TDPAC spectra and it may be possible to determine whether He atoms are at substitutional or interstitial sites. The observation that the oscillation amplitude is only reduced but not damped with time under the He implantation indicates that $^{140}$Ce–He complexes take a variety of geometrical structures, accordingly these $^{140}$Ce bonded by He feeling various electric field gradients due to He. It is natural to think that many He atoms surround the parent atom and that there is a distribution of the number of He atoms attached to it.
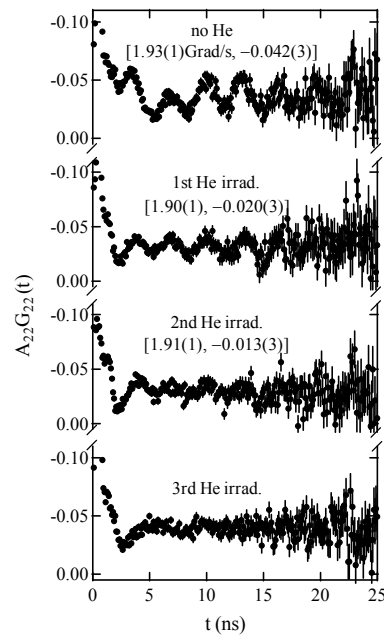


Fig. 1. Room-temperature TDPAC spectra for $^{140}$CeFe before He irradiation, He-doped(1)$^{140}$CeFe, He-doped(2)$^{140}$CeFe, and He-doped(3)$^{140}$CeFe, all with no annealing. The two values in the square brackets in each panel are the Larmor frequency (Grad/s) and the amplitude of the oscillation component.

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## PR12-3      Atmosphere Dependence of Stability of Local Fields in Al-Doped ZnO

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**INTRODUCTION:** Impurity-induced properties emerging in ZnO has been attracting increasing attention toward its application to functional materials in a wide field of industry. For a practical use of ZnO as a conduction-controlling device, it is of great importance to study the physical and chemical states of doped impurities and/or oxygen vacancies. From this point of view, we have investigated the factors determining the local structures and their stability of Al-doped ZnO samples synthesized on different conditions by means of the time-differential perturbed angular correlation (TDPAC) method. In a series of our TDPAC studies, drastic change of the local structure was observed for Al-doped ZnO sample heat-treated in vacuum [1,2]. In order to provide further insight into the above atmosphere dependence, in the present work, we performed an additional TDPAC measurement for 100 ppm Al-doped ZnO annealed under argon gas flow.

**EXPERIMENTS:** For the synthesis of 100 ppm Al-doped ZnO, stoichiometric amounts of $Al(NO_3)_3 \cdot 9H_2O$ and ZnO powder were mixed in ethanol. The suspension was heated to evaporate the ethanol until dryness. The powder was pressed into two disks and sintered in air at 1273 K for 3 h. For TDPAC measurements, commercially available ^{111}In solution was added in droplets onto the sintered disks at the concentration of 100 ppt. The disks again underwent heat treatment in air at 1373 K for 2 h. After that, a TDPAC measurement was performed for one of these samples prepared in air. The other sample was ground into powder and annealed under argon gas flow at 1373 K for 24 h. A TDPAC measurement was carried out for the [111]In($\rightarrow$[111]Cd) probe on the 171-245 keV cascade $\gamma$ rays with the intermediate state of $I = 5/2$ having a half-life of 85.0 ns.

**RESULTS:** Fig. 1 shows the TDPAC spectra of [111]In($\rightarrow$[111]Cd) probe in 100 ppm Al-doped ZnO annealed (a) in air and (b) in argon gas. The directional anisotropy on the ordinate, $A_{22}G_{22}(t)$, was deduced by the following relation for delayed coincidence events of the cascade:

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

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 $\gamma$-ray emissions, and $N(\theta,t)$ the number of the coincidence events observed at angle, $\theta$. The spectrum in Fig.1(a) shows that 100 ppm Al and 100 ppt ^{111}In are locally-associated in ZnO matrix, suggesting that there is a strong attractive force between Al and In in ZnO [1,2]. For the spectrum in Fig. 1(b), two different components were observed: one is that appearing in the spectrum in Fig. 1(a) and the other is that observed for undoped ZnO[3]. This observation implies that the probe gradually resides solely at the substitutional Zn site after annealing under argon gas in the same way as the case for the sample heat-treated in vacuum. These observations suggest that the drastic change of local structure formed by Al and In ions is closely related to the presence of $O_2$ gas. In oxide compounds, in general, oxygen vacancies are likely to be formed during annealing process at a lower partial pressure of atmospheric oxygen. Therefore, it is suggested that the local association of Al and ^{111}In becomes unstable in anaerobic atmosphere due to the formation of oxygen vacancies in the sample. For further information on the stability of the local structure, dependence of partial pressure of oxygen for the dissociation process needs to be investigated.
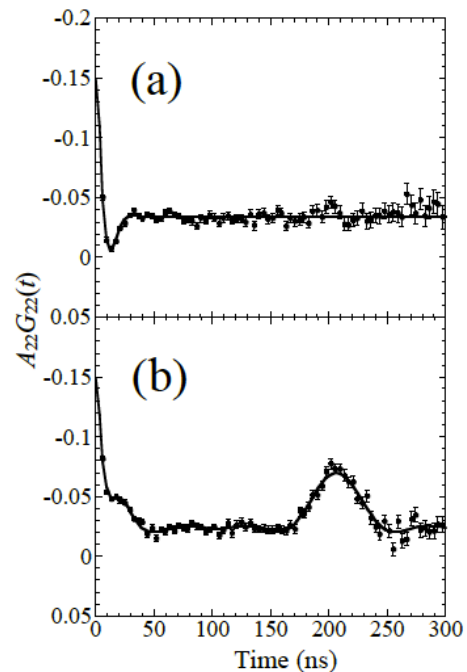


Fig. 1.   TDPAC spectra of [111]In($\rightarrow$[111]Cd) in 100 ppm Al-doped ZnO annealed (a) in air and (b) in argon gas. The TDPAC measurements were performed at room temperature.

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# PR12-4 Extranuclear Dynamic Motion of $^{111}$Cd($\leftarrow ^{111}$Ag) Doped in AgI Nanoparticles

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**INTRODUCTION**: Silver iodide, AgI, is a promising metal halide showing superionic conductivity as in its $\alpha$ phase, and its application to functional materials such as solid electrolytes is thus highly expected. The conducting phenomenon, however, only emerges at elevated temperature (> 420 K) because the crystal structure depends on temperature; this has indeed been a barrier to the practical applications of this compound.

Recently, an epoch-making technique has broken through this situation: powder AgI coated with poly-N-vinyl-2-pyrrolidone (PVP) can drastically enhance the ionic transport property at room temperature, recording the conductivity of $1.5 \times 10^{-2}$ $\Omega^{-1}$ cm$^{-1}$[1]. According to the report, this achievement is due to successful control of the particle size as small as nanoscale. In addition to the property in the bulk, it is of great importance to obtain complementary information on the site-to-site hopping motion of Ag$^+$ ions on an atomic scale for a detailed understanding of ionic conductivity of this binary solid. For that purpose, in the present work, we have applied the time-differential perturbed angular correlation (TDPAC) technique with the $^{111}$Cd($\leftarrow ^{111}$Ag) probe nuclei to observe dynamic behavior of Ag$^+$ ions. We here report successful evaluation of the activation energy of the dynamic motion of Ag$^+$ ions [2,3].

**EXPERIMENTS**: Pd foil was irradiated with thermal neutrons in Kyoto University Reactor to produce the TDPAC probe. After radioequilibrium was achieved between $^{111}$Pd and $^{111}$Ag, the Pd foil was dissolved in HNO$_3$ aq. solution, and carrier-free $^{111}$Ag was isolated by an anion-exchange chromatography. The separated $^{111}$Ag was incorporated together in PVP-coated AgI sample when the powder sample was synthesized by precipitation of their raw materials. We confirmed by transmission electron microscopy that microscopic particles with sizes of 10-100 nm were expectedly synthesized for $^{111}$Ag-free PVP-coated AgI.

TDPAC measurements of the $^{111}$Cd($\leftarrow ^{111}$Ag) probe were performed for the synthesized sample at various temperatures. The directional anisotropy, $A_{22}G_{22}(t)$, was deduced with the following relation:

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

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 $\gamma$-ray emissions, and $N(\theta, t)$ the number of the delayed coincidence events observed at an angle, $\theta$.

**RESULTS**: It was found that TDPAC spectra of the $^{111}$Cd($\leftarrow ^{111}$Ag) probe introduced in PVP-coated AgI measured at different temperatures show exponential relaxation of the directional anisotropy, which signifies that the probe is dynamically perturbed by the extranuclear field. This phenomenon is understandable for the spectra of the sample as in the $\alpha$ phase (> 420 K). What is to be noted here is that the spectral relaxation was also observed even at room temperature. It was found from this observation that Ag$^+$ ions surrounding the probe nuclei exhibit site-to-site hopping motion in the PVP-coated AgI nanoparticle even at this low temperature. Fig. 1 shows the temperature dependence of the relaxation constants. From a least-squares fit to the data with an Arrhenius-type equation, the activation energy, $E_a$, of the hopping motion was successfully estimated to be $E_a = 46$ (10) meV. This value shows fair agreement with that obtained for the superionic conductivity: $E_a = 60$ meV [1]. The present result is significant in that superionic conduction can be probed with local dynamics observed through nuclear spin relaxation.
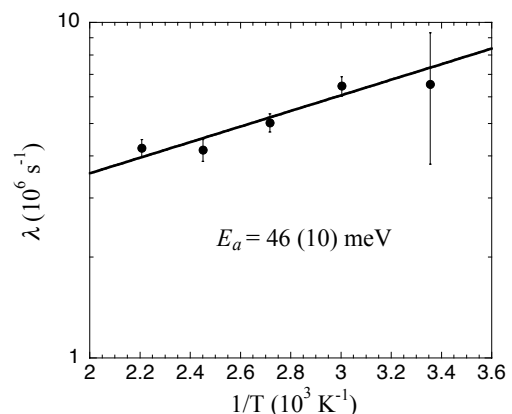


Fig. 1. Temperature dependence of the relaxation constant. The line is the result of a least-squares fit with an Arrhenius-type equation.

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