I. Project Research

Project 8

PR8 Project Research on Nuclear Spectroscopy and Condensed Matter Physics Using Short-Lived Nuclei

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Objective and Participating Research Subjects

Because of the suspension of the reactor (KUR) operation for the last three periods, we could not execute any irradiation experiments in a three-year project research named the same title of the present project research. In the late August of this period, the operation of KUR restarted and we also restarted the project research, the main objectives of which are the investigation of the nuclear structure of unstable neutron-rich nuclei and also the local properties of matters using short-lived nuclei.

Unfortunately, several troubles interrupted the operation of KUR and moreover a different trouble made the on-line isotope separator (KUR-ISOL) unavailable for several weeks so that two research subjects (29P8-1 and 29P8-4) were not executed at all and the other research subjects of the project (29P8) were carried out incompletely.

The research subjects reported here are as follows:

29P8-2 Isomer search for fission products around $A = 150$ with energy-sum γ-ray spectrometry
29P8-3 Compton polarimeter for β-delayed γ rays using clover detector
29P8-5 Nuclear spin relaxation of ^{111}Cd at the A site in a spinel oxide $CdIn_2O_4$
29P8-6 Observation of local fields at the ^{111}Cd(← ^{111m}Cd) sites in cadmium titanate

Main Points Described in the Reports in the Following Four Pages

M. Shibata *et al*. (29P8-2) took γ-ray spectra associated with the β^- decay of ^{144}Ba, ^{150}Ce, and ^{152}Ce in the singles and add-back modes of their γ-ray detection system with a clover detector in order to search for an unknown isomer state in an odd-odd nucleus ^{144}La in particular, which is in turn expected to be able to solve the so-called Q_β puzzle in ^{144}La. The trick of searching for an unknown isomer state with the clover detector is as follows: For an isomeric γ-ray transition, the intensity is higher in the add-back mode than in the singles mode, whereas the intensity of a γ-ray in a cascade is lower in the add-back mode than in the singles mode. They obtained ^{144}Ba, ^{150}Ce, and ^{152}Ce as thermal neutron-induced fission products of ^{235}U at the KUR-ISOL. The detailed data analyses are in progress.

Y. Kojima *et al*. (29P8-3) are trying to measure the linear polarization of γ-rays from short-lived β decaying nuclei using the above mentioned clover detector and additional coaxial Ge detectors in order to obtain the multipolarities of those γ-rays. The clover detector is used as a Compton polarimeter. They first measured the γ-ray energy dependence of the polarization sensitivity of the polarimeter using standard γ-ray sources ^{60}Co, ^{134}Cs, and ^{152}Eu, and then measured the Compton scattering asymmetry for ^{140}Cs and ^{146}La which were produced and separated at KUR-ISOL. They obtained preliminary asymmetry values for the 258-keV and 410-keV γ-rays from ^{146}La.

W. Sato *et al*. (29P8-5) studied the local fields at the two metal sites of $CdIn_2O_4$, which is expected to be optoelectronic, using the time-differential perturbed-angular-correlation (TDPAC) technique. They used two TDPAC nuclear systems with a common probe nuclear state, the 245-keV ^{111}Cd state having a nuclear spin of 5/2: one arising from ^{111}In (commercially available) and the other from ^{111m}Cd (produced using a pneumatic system at KUR). It is natural to consider that ^{111}In and ^{111m}Cd occupy the In site and the Cd site in $CdIn_2O_4$, respectively and that the 245-keV ^{111}Cd state arising from ^{111}In and that from ^{111m}Cd feel the corresponding static electric field gradients. This is the case for the former. However, unexpectedly, the 245-keV ^{111}Cd state arising from ^{111m}Cd feels a dynamic perturbation. The cause of the dynamic perturbation is under consideration.

S. Komatsuda *et al*. aim at obtaining with the ^{111}Cd(← ^{111m}Cd)-TDPAC technique microscopic information on the ferroelectric to paraelectric phase transition in perovskite oxides $CdTiO_3$ and Ca-doped $CdTiO_3$. They first of all examined the preparation condition for $CdTiO_3$ by taking a room-temperature TDPAC spectrum of ^{111}Cd(← ^{111m}Cd) in $CdTiO_3$ (29P8-6). Because ^{111m}Cd is short lived (its half-life is 49 min), it is necessary to shorten the preparation process as much as possible. They confirmed together with the powder X-ray diffraction method that for the preparation condition they used, ^{111m}Cd occupies the Cd site of $CdTiO_3$ prepared with no secondary phase.

PR8-1 Isomer Search for Fission Products around *A*=150 with Energy-sum γ-ray Spectrometry

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INTRODUCTION: Decay scheme information of fission products of ^{235}U are important for both nuclear engineering and nuclear physics. Concerning fission products around mass number 150, doubly-odd nuclei have not been studied well and the decay information are scarce compared to odd mass nuclei. Especially, isomeric states are expected in the doubly-odd nuclei. For example, a long-lived isomeric state at 150.3 keV whose half-life is longer than 5s in ^{156}Pm has been proposed [1] and ^{152,154}Pm and ^{152}Pr also have isomeric states. These isomeric states are interpreted as the spin flip-flop transition. Therefore the other isotopes are expected to have isomeric states.

As described in ref. [2], in ^{144}La, three different Q_β-values were proposed by the β-ray spectra gated by γ-rays. One of the reason of the differences are considered to be discrepancies of decay schemes because the gated γ-rays are placed at different position between the decay schemes. Another Q_β-value was proposed by the total absorption detector [2], nevertheless, the problem still remains. There is a possibility that an unidentified isomer exists. To solve the "Q_β-puzzle", reconstruction of the decay scheme and search for the isomer are desired.

As described in ref. [3, 4], our clover detector has four identical Ge crystals and each Ge works independently. The detector can measure in not only singles mode but also add-back mode by using a VME-based data acquisition system. In the energy-sum γ-ray spectrometry, sum peaks of cascade γ-rays are observed more intensively in the add-back mode, and each cascade γ-ray is observed weakly compared to that of singles mode. Therefore the long-lived isomeric transitions are expected to be observed intensively in the add-back mode.

In this experiments, the β-decay study of ^{144}Ba, ^{150}Ce and ^{152}Ce were carried out by means of the energy-sum γ-ray spectrometry using the clover detector.

EXPERIMENTS: The experiments were performed at the on-line mass separator KUR-ISOL at the Kyoto University Reactor, a 72 mg of 93%-enriched $^{235}UF_4$ target being inserted in a through-hole. The isotopes of interests were produced with the thermal neutron-induced fission of ^{235}U. The produced radioactive isotopes were transported by a gas jet of He-N_2 and were ionized by a thermal-ionization type ion source. The mass-separated radioactive beam was incident on an aluminized Mylar tape set in the computer controlled tape transport system. The tape was moved a predetermined period to reduce the background from their daughter nuclei. The γ-rays associated with the decay of each isotope were measured by means of energy-sum γ-ray spectrometry. For ^{144}Ba ($T_{1/2}$=11.5 s), the collection–measurement cycle was set at 22 s–22 s and was repeated for 10 hours. The detector was shielded with 10 cm-thick lead blocks and 10 cm-thick borated polyethylene blocks outside them in order to reduce the background neutrons and γ-radiation.

RESULTS: The measured γ-ray spectra associated with the β decay of ^{144}Ba in the singles and add-back modes are shown in Fig. 1. The closed circles indicate the coincidence γ-rays with the La x-rays. At present, the excited states and γ-rays of daughter nucleus ^{144}La are proposed up to 1240 keV and 922.4 keV, respectively. The evaluated Q_β-values of ^{144}Ba is 3083 keV [5], then, higher-lying levels and γ-rays are expected be observed by the experiment using the clover detector. The detailed analysis is in progress also for ^{150}Ce and ^{152}Ce.

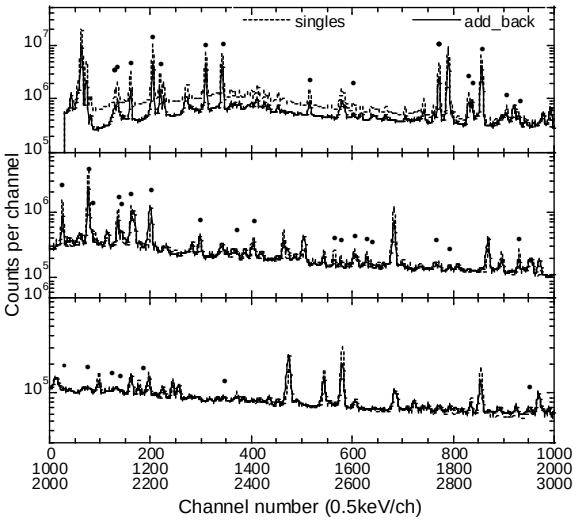

Fig. 1 Singles and add-back spectra for γ-rays associated with the β decay of ^{144}Ba. The closed circle indicates the coincidence γ-ray with the La x-rays in the preliminary analysis.

CONCLUSIONS: The β-decay study for the doubly-add nuclei of fission products were carried out by means of the energy-sum γ-ray spectrometry. The measurement by clover detector coupled with ISOL is effective to the decay study. Especially, there are some newly observed γ-ray in the decay of ^{144}Ba. To solve the Q_β-puzzle in ^{144}La, further analyses are desired.

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PR8-2 Compton Polarimeter for β-delayed γ Rays Using Clover Detector

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INTRODUCTION: Gamma-ray multipolarities are one of the most important properties for nuclei. For example, experimental data on them and also those on spins and parities of nuclear levels are necessary for discussion on nuclear deformation.

The linear polarization of γ rays is useful in determining the multipolarities. The linear polarization is usually measured using a Compton polarimeter consisting of two or more γ-ray detectors because the Compton scattering is a polarization-sensitive process. 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. This method is often used for in-beam γ-ray spectroscopies. However, it rarely applies for γ rays following decays of short-lived nuclei. This is because the latter case needs an additional detector (directional detector) in order to define the *reference plane* and the triple coincidence measurement makes the overall detection efficiency low.

Our group is trying to apply a clover Ge detector [1] to the linear polarization measurements for short-lived β-decaying nuclei. The clover detector contains four large Ge crystals (80mmϕ, 90mm in length) packed closely. Thus, we expect that the clover detector enables the polarization measurement for β-delayed γ rays to be made. In this report, we present the status of this research and preliminary results of the first on-line experiment at KUR-ISOL [2].

EXPERIMENTS: The detection system consisted of the clover detector as a polarimeter, a coaxial Ge detector as a directional detector, and another coaxial detector to deduce correction factors of detection efficiencies. The pre-amplifier signals from the detectors were processed by a VME-based data acquisition system and recorded in a list mode.

First, ^{60}Co (40 kBq), ^{134}Cs(9.5 kBq) and ^{152}Eu(27 kBq) standard sources were measured to reveal properties of the system such as energy dependence of the polarization sensitivity. In order to find the optimum measuring condition, we repeated measurements, placing the clover and coaxial Ge detectors (relative efficiencies of 38% and 60%) in different geometry arrangements.

Then, we measured short-lived ^{140}Cs and ^{146}La nuclei. They were prepared at KUR-ISOL, following the thermal neutron induced fission of ^{235}U. The fission products were thermalized in the target chamber, and transported to an ion source by gas jet stream. After ionization, the nuclei were mass-separated and implanted into an aluminized Mylar tape. Each detector was placed at 10 cm from the source (see below). The measuring time was 3 h for ^{140}Cs and 17 h for ^{146}La.

RESULTS: The list data were analyzed using an off-line sorting program. Signals recorded within 490 ns were taken as coincident events.

Asymmetry of the Compton scattering was observed for E2-E2 γ cascades in the decay of ^{60}Co, ^{134}Cs and ^{152}Eu. The polarimeter sensitivity Q was deduced using the experimental and theoretical asymmetry value [3]. The best performance was obtained when the source-to-detector distance is 10 cm and the directional detector is the 60% coaxial detector. The energy dependence of the sensitivity Q in this condition was shown in Fig. 1. The 344- and 411-keV γ rays from ^{152}Eu show large uncertainties because of poor statistics. We need a further measurement for ^{152}Eu.

From the measurements at KUR-ISOL, preliminary asymmetry values were obtained for ^{146}La to be about −0.05 for 258-keV and 0.03 for 410-keV E2 γ ray. Further measurements are needed to improve the precision.

Another problem found in the experiments at KUR-ISOL is intense background radiations. They cause random coincidence and reduce counts associated with the Compton scattering. To solve this problem, a heavier shield should be placed around the clover detector. Also, data analysis to remove (or reduce) random summing effects is planned.

SUMMARY: It has been found that a clover Ge detector works as a Compton polarimeter for β-delayed γ rays. However, further improvement is required to use the polarimeter at KUR-ISOL.

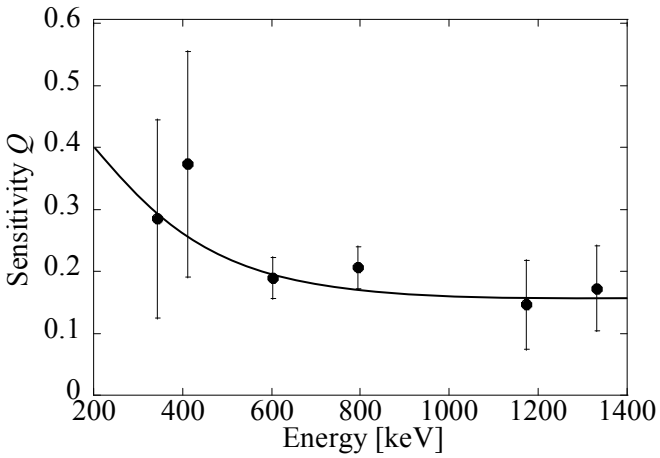

Fig. 1. The polarization sensitivity as a function of incident γ-ray energy. The curve is only a guide to the eye.

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PR8-3 Nuclear Spin Relaxation of ^{111}Cd at the *A* Site in a Spinel Oxide $CdIn_2O_4$

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INTRODUCTION: Spinel oxides exhibit various physical properties depending on the constituent metal elements and on their residence positions. Optoelectronic response is one of the most interesting phenomena observed for the oxide group [1]. From the view point of fundamental physics as well as technology applications, therefore, an atomic level investigation of the physical properties of these oxides is very important. In the present work, we measured local fields at the metal sites in $CdIn_2O_4$, an expected optoelectronic material, to obtain microscopic information by means of time-differential perturbed angular correlation (TDPAC) spectroscopy with two different probes ^{111}In($\rightarrow^{111}$Cd) and ^{111m}Cd($\rightarrow^{111}$Cd). In this report, unexpected nuclear spin relaxation of the ^{111m}Cd($\rightarrow^{111}$Cd) probe is presented [2].

EXPERIMENTS: Well-mixed stoichiometric amounts of CdO and In_2O_3 were pressed into a disk, and sintered in air at 1373 K for 45 min. After the confirmation of successful synthesis of $CdIn_2O_4$ by a powder X-ray diffraction pattern, the sample was doped with ^{111}In($\rightarrow^{111}$Cd) in HCl solution and underwent heat treatment in air at 1373 K for 3 h for the thermal diffusion of the radioisotope. A TDPAC measurement was then performed for the ^{111}In($\rightarrow^{111}$Cd) probe on the 171-245 keV cascade γ-rays with the intermediate state of $I = 5/2$ having a half-life of 85.0 ns.

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 well mixed in a mortar with $CdIn_2O_4$ prepared in advance, and sintered in air at 1373 K for 45 min. A TDPAC measurement was carried out for the ^{111m}Cd($\rightarrow^{111}$Cd) probe on the 151-245 keV cascade γ-rays with the same intermediate state as the ^{111}In($\rightarrow^{111}$Cd) probe. 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 coincidence events observed at an angle θ.

RESULTS: The room-temperature TDPAC spectrum of ^{111}In($\rightarrow^{111}$Cd) embedded in $CdIn_2O_4$ is shown in Fig. 1(a). The spectrum shows oscillatory structure characteristic of the electric quadrupole interaction of the probe nucleus on the intermediate state (I = 5/2) with the extranuclear charge distribution. 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 frequencies ω_n:

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

Because In ions are known to reside in the distorted octahedral *B* site in $CdIn_2O_4$, it is plausible that the present ^{111}In($\rightarrow^{111}$Cd) probe also occupies the *B* site; the quite large quadrupole frequency ω_Q = 14.04 Mrad s^{-1} is consistent with this interpretation.

Figure 1(b) shows the room-temperature TDPAC spectrum of the ^{111m}Cd($\rightarrow^{111}$Cd) probe incorporated in $CdIn_2O_4$. An unexpected pattern was observed in the spectrum; the directional anisotropy of the γ-γ cascade shows a fast attenuation, implying nuclear spin relaxation caused by the extranuclear perturbation. A possible cause of the spin relaxation is dynamic fluctuation of the probe nucleus relative to the extranuclear charge distribution. Taking this assumption into account, the spectra were tentatively fitted with the following time-differential perturbation factor:

$$G_{22}(t) = \exp(-t/\tau_c) G_{22}^{\text{static}}(t). \qquad (3)$$

Here, we assumed slow fluctuation in the relative motion of the extranuclear field. In Eq. (3), τ_c (= 39(4) ns at room temperature) stands for the reorientational correlation time of the probe nucleus and $G_{22}^{\text{static}}(t)$ is the static perturbation factor. Because it is considered that ^{111m}Cd probes mainly occupy the tetrahedral *A* site in $CdIn_2O_4$, however, it is difficult to ascribe the relaxation to the thermal motion of the probe ions confined in the small tetrahedral space. Further detailed investigation in a wide temperature range would be needed for the determination of the origin of the spectral damping.

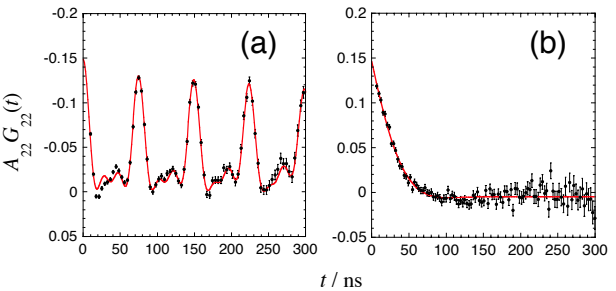

Fig. 1. TDPAC spectra (a) of ^{111}In($\rightarrow^{111}$Cd) and (b) of ^{111m}Cd($\rightarrow^{111}$Cd) in $CdIn_2O_4$ measured at room temperature.

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PR8-4 Observation of Local Fields at the ^{111}Cd(←^{111m}Cd) Sites in Cadmium Titanate

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INTRODUCTION: The perovskite oxides ABO_3 exhibit various electronic properties, depending on their combination of A and B ions. The cadmium titanate ($CdTiO_3$) has been attracting much attention as a ferroelectric oxide, and its technological applications are strongly expected. It is reported that $CdTiO_3$ exhibits a phase transition from the ferroelectric to paraelectric at about 85 K, and this transition temperature is affected by calcium (Ca) doping on the Cd site in $CdTiO_3$[1]. Because it is necessary for a practical use of $CdTiO_3$ to obtain more microscopic information on this transition mechanism, we investigated the local structures in $CdTiO_3$ and Ca-doped $CdTiO_3$ by means of the time-differential perturbed angular correlation (TDPAC) method using the ^{111}Cd(←^{111m}Cd) probe. In the present work, in order to ascertain the doping condition of the ^{111}Cd(←^{111m}Cd) probe, we initially examined the site occupied by the ^{111}Cd(←^{111m}Cd) in undoped $CdTiO_3$.

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 powder and nonradioactive CdO powder to synthesize the polycrystalline $CdTiO_3$ powder. The powders were mixed in the mortar. The powders were pressed into disks and sintered in air at 1373 K for 45 min. It was confirmed from the powder XRD pattern for the nonradioactive sample 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 the TDPAC spectrum of ^{111}Cd(←^{111m}Cd) probe in $CdTiO_3$. 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:

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

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 spectrum in Fig. 1 with $G_{22}(t)$ expressed as

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

For all symbols in eq.(2), refer to our previous paper[2]. The electric field gradient (EFG) and asymmetry parameter value were estimated to be V_{zz} = 5.5(9)×10^{21} Vm^{-2} (η =0.60). This EFG value shows a good agreement with that obtained for ^{111m}Cd probe in $CdTiO_3$ sample[3]. Since the present spectrum can be reproduced by the fit assuming a single component, ^{111m}Cd probes occupy the Cd site in $CdTiO_3$. Relating to the η value, the large η value is probably due to the distortion of their orthorhombic structure. For more information on the microstructure of $CdTiO_3$, TDPAC measurements over a wide range of temperature and sample preparation of Ca-doped $CdTiO_3$ are now in progress.

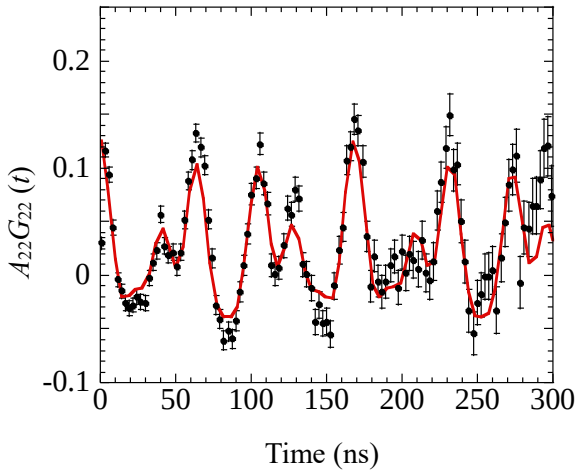

Fig. 1. TDPAC spectrum of ^{111}Cd(←^{111m}Cd) probe in $CdTiO_3$ at room temperature. The line is the result of a least-squares fit with eq.(2).

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