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^{83m}Kr, a potentially powerful PAC probe

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Abstract In the decay of ^{83}Rb to ^{83m}Kr and the subsequent decay to the ^{83}Kr ground state a 553–9.4 keV γ-γ and a 17.85–9.4 keV e^--γ cascade are populated. The intermediate 9.4 keV $7/2^+$ state with a half-life of 154 ns is a perfect candidate for the application of the perturbed angular correlation (PAC) technique. Thus, it is possible to investigate the lattice environment of the implanted probes via the electric quadrupole interaction of the 9.4 keV $7/2^+$ state with the electric field gradient produced by the host lattices. Details of the production of this new PAC probe and planned measurements will be discussed.

Keywords PAC · Perturbed angular correlation · ^{83}Rb · ^{83m}Kr · KATRIN

1 Introduction

The 17.85 keV conversion electron line emitted in the decay of ^{83m}Kr has been shown to be a perfect calibration source for The **Ka**rlsruhe **Tri**tium **N**eutrino (KATRIN) experiment [1]. Therefore, the production and implantation of the parent activity ^{83}Rb has been studied in Bonn [2]. Recently, however, precision measurements of the 17.85 keV conversion electron line emitted from ^{83}Rb implanted Pt and Au foils showed an unexpected double line structure [3]. A possible cause could be different lattice environments of the electron emitting nuclei.

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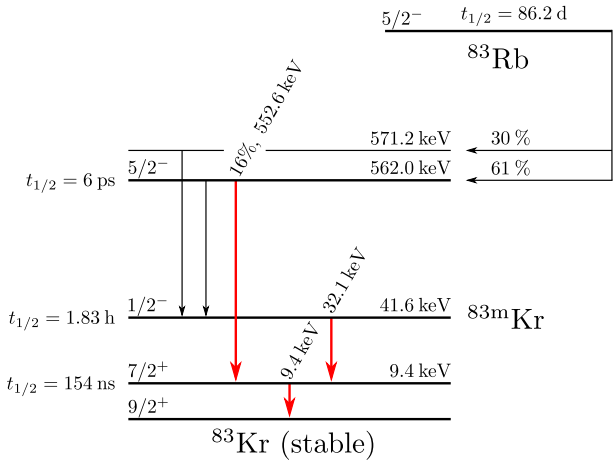

Fig. 1 Simplified decay scheme of ^{83}Rb/^{83m}Kr with possible PAC cascades

2 Properties of the probe

^{83}Rb mainly decays through two channels in which two cascades, a 553–9.4 keV γ-γ and a 17.85–9.4 keV e$^-$-γ cascade are populated (all data on ^{83}Rb, ^{83}Kr and ^{83m}Kr taken from [4] unless stated otherwise). These have the same intermediate state, the 9.4 keV 7/2$^+$ ^{83}Kr state with a half-life of 154 ns. This is a perfect situation for the application of the perturbed angular correlation technique (PAC) observing the electric quadrupole interaction of this intermediate state with the electric field gradient (EFG) produced by host lattices. The specific properties of these two cascades provide the possibility to examine two different lattice environments using the same probe nucleus.

The half-life of ^{83m}Kr ($t_{1/2}$ = 1.83 h), the parent activity of the 17.85–9.4 keV e$^-$-γ cascade in ^{83}Kr, is quite short as compared to typical times it takes to prepare samples, for instance, by implantation and subsequent annealing steps. However, in the decay of ^{83}Rb the isomeric state ^{83m}Kr is populated with 75 % (Fig. 1) and can thus serve as a generator for ^{83m}Kr. During the rather long half-life of 1.83 h of the cascades initial level there is considerable time for the ^{83m}Kr probe nucleus to interact with its environment e.g. undergo diffusion processes. The 17.85 keV K-conversion electron yield is sufficiently high due to its conversion coefficient of $\alpha_K = 462$. The γ-yield of the second transition is reduced to 5.5 % by the emission of conversion elecrons. However, this poses no problem for the measurements.

In 16 % of the ^{83}Rb decays a 553 keV–9.4 keV γ-γ cascade is populated, which circumvents the ^{83m}Kr isomeric state. Due to this fact and the rather short half-life of 6 ps of its initial level one can assume that the hyperfine interaction observation takes place before any change in lattice site can occur. In contrast to the first cascade where the lattice site of a ^{83m}Kr probe is studied, here the position of the ^{83}Rb is decisive.

The ratio $Q_{ex}/Q_{gr} = 1.958(2)$ of the quadrupole moment of the 9.4 keV intermediate state and the ^{83}Kr ground state have been measured by Mößbauer measurements very accurately in various Kr compounds by Holloway et al. [5]. Thus, a value of $Q_{ex} = +0.491(5)$ b can be derived using the ground state moment of

$Q_{gr} = +0.251(5)$ b given by Kuiper [6]. Further, the shielding factor for Kr is relatively large $(1 - \gamma_\infty) = 85$ [7]. So, assuming a typical lattice EFG of 1×10^{16} V/cm^2 one can calculate that the quadrupole interaction frequency has a value of ≈ 59 MHz corresponding to a period of 140 ns which should be well observable.

3 Experimental details

Efficient reactions for the production of Rb are the ^{nat}Kr (p,xn) Rb and the ^{81}Br (α,2n) Rb process. The former has a maximum reaction cross-section of $\sigma = 500$ mb and its suitability has been investigated by Vénos et al. [8]. The main challenges here are the necessary use of a high pressure Kr gas target and the technique to remove and store the Rb produced from the gas stream. In Bonn we exploited the second route, namely the ^{81}Br (α, 2n) ^{83}Rb reaction.

The element bromine, the only non-metallic element liquid at room temperature, has two stable isotopes with roughly equal isotopic abundances of 51 % for ^{79}Br and 49 % for ^{81}Br. The desired isotope ^{83}Rb can be produced via the ^{81}Br (α, 2n) reaction, with the large maximum cross section of $\sigma = 1300$ mb at an α-particle energy of 26 MeV [9, 10]. In order to achieve a production rate as large as possible we refrained from using a Br compound and aimed at a liquid Br_2 target. This ensures the highest ^{81}Br density and allows for an effective removal of the beam energy deposited in the target. However, since liquid Br is an extremely aggressive agent the only suitable material to contain the target is quartz. So, app. 3 cm^3 of liquid Br were filled into a quartz tube of 6 mm outer diameter with 200 µm wall thickness and 80 mm length. Thus, in the centre of the tube a layer of app. 5.6 mm of liquid Br was hit by the beam. A constant flow of demineralised water from the back provided the necessary cooling of the quartz tube. At the beam centre a 100 µm Al foil separated the target and the cooling water from the accelerator vacuum. A detailed simulation using the NIST energy loss tables [11] yielded the following results for in these layers considering α-particles with a primary energy of 54 MeV. After losing slightly less than 10 MeV by passing through 100 µm Al, 100 µm H_2O and 200 µm SiO_2 the particles enter the liquid Br with an energy of 45.4 MeV in the centre of the quartz tube, where they travel about 1 mm before they are stopped. Typical production efficiencies of 0.2 MBq/Ah were measured.

The 54 MeV α-beam was provided by the Bonn isochronous cyclotron. In the present experiments, a maximum beam current of 0.5 µA was used. Thus, a power of 27 W is deposited in the target, which is well within the limits of the cooling circuits capacity.

After the irradiation, and some cool off time the top of the quartz ampoule was opened and the excess Br was allowed to evaporate by passing the Br fumes through a sodium thiosulfate solution for 72 h. The spectra (HPGe) of the now empty quartz ampoule showed that the ^{83}Rb and ^{84}Rb activity ($t_{1/2} = 32.8$ d [12]) had remained completely inside the ampoule, presumably in the form of RbBr. The latter is an unavoidable side product that is produced by a ^{81}Br (α,n) ^{84}Rb reaction during the irradiation. The activity was washed out and transferred to the ion source of the **Bon**n **I**sotope **S**eparator (BONIS). A tungsten surface ionisation oven was used which had to be quite long in order to separate the evaporation zone with app. 300 °C and the

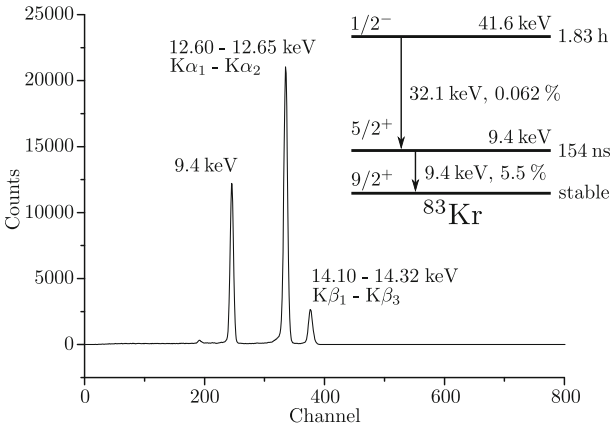

Fig. 2 Low energy spectrum of ^{83m}Kr recorded with an AMPTEK XR-100CR x-ray detector.and an excerpt of the decay scheme of ^{83m}Kr. For the 32.1 keV and 9.4 keV transitions the γ-yields are given. X-ray data taken from [14]

ionisation zone with 2500 °C. Implantation efficiencies of the order of 15 % were achieved.

Energy spectra of the ^{83}Rb/^{83m}Kr after implantation into Cu were taken with an AMPTEK XR-100CR x-ray detector and are shown in Fig. 2. For the actual measurements it is planned to use APD detectors coupled to LSO scintillator crystals for the 9.4 keV x-rays and windowless APDs for the detection of the 17.85 keV conversion electrons. These detectors are small, easy to handle and provide a 100 % detection probability in both cases. It has been shown by Baron et al. [13] that in this way a sufficient energy resolution and at the same time an excellent time resolution can be achieved.

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