Direct Search for Low Energy Nuclear Isomeric Transition of Th-229m With TES Detector

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***Abstract*—Precise knowledge of the energy and lifetime of ^{229m}Th isomeric state has notable importance as a basis for a nuclear clock. Such a clock would be capable to extend precision on the oscillator frequency by up to four orders of magnitude compared to the presently best atomic clocks. However, the technique proposed for the clock requires that the isomeric state energy is accessible with existing laser systems. Previous measurement placed this state at ∼8 eV (150 nm), in the Vacuum Ultra Violet (VUV) range of the electromagnetic spectrum. A precise direct measurement of the energy of this state is necessary to determine whether the nuclear clock can be made using existing laser technology. We are developing a cryogenic microcalorimeter to measure the energy and lifetime of the ^{229m}Th isomeric state directly. The experiment will use a ^{233}U source whose alpha-decay will populate the ^{229m}Th isomeric state with 2% probability. The subsequent decay of ^{229m}Th will be measured by a Transition Edge Sensor (TES) with <1 eV resolution. Such a technique will allow to observe all possible types of decays of ^{229m}Th in the range of energy from 3 to 50 eV and lifetimes >5 microseconds. The single-photon TES has sufficient resolving power combined with high efficiency in the whole energy band for this experiment. Here we present a prototype of TES based on a 200 nm thick iridium-gold (Ir/Au) film which was tested with a pulsed laser source and demonstrated ∼0.8 eV energy resolution and 5.8 ± 2.1 μs signal recovery time.**

***Index Terms*—Iridium, microcalorimeter, nuclear decay, thorium, transition edge sensor.**

I. Introduction

THE recent direct observation of the isomeric state ^{229m}Th makes ^{229}Th nucleus to be a realistic and the only candidate known to the date for nuclear clock developments [1], [2]. Its energy above the ground level is ∼8 eV (corresponding to ∼150 nm, VUV) which can be reached with modern laser systems [3], [4]. Precise direct measurement of ^{229m}Th transition to the ground state can be achieved with the use of cryogenic microcalorimeters which, unlike conventional detectors,[1] would be equally sensitive not only to radiative, but also to the dominant internal electron conversion (IC) channel.

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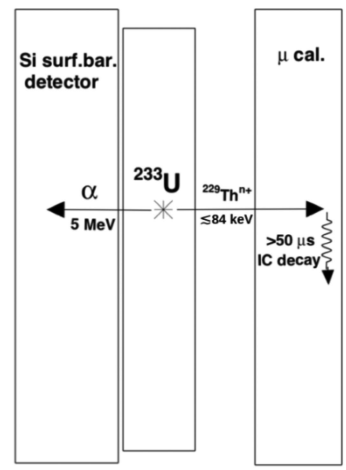

Fig. 1. Schematic drawing of the experimental setup to measure ^{229m}Th isomeric transition energy using a TES microcalorimeter. A ^{233}U source is deposited as a thin film on top of a silicon surface-barrier detector which registers an alpha-particle from a ^{233}U decay. A recoiling ^{229}Th ion being in the isomeric state with 2% probability implants into the TES which detects its consequent transition.

The aim of the experiment is to determine directly the energy of the ^{229m}Th isomeric transition. ^{229m}Th will be enclosed inside the microcalorimeter to ensure the maximal energy absorption from the dominant internal conversion (IC) decay channel (∼ 10 µs lifetime) as well as from rare radiative decays (∼ 10^4 s lifetime). ^{229m}Th will be implanted into TES bulk by its up to 84 keV kinetic energy released in alpha-decay process of the ^{233}U mother isotope: $^{233}_{92}\mathrm{U} \rightarrow {}^{229(m)}_{90}\mathrm{Th} + {}^{4}_{2}\alpha$.

Fig. 1 demonstrates the measurement principle. We will use a ^{233}U source deposited as a thin film on the active surface of a silicon detector for tagging the alpha-decay that produces ^{229m}Th ions recoiling outwards the U film itself. These ions, occupying the isomeric state with 2% probability, will implant in the TES microcalorimeter [6]. There, the ions will be neutralized via

[1] Such as high-purity germanium (HPGe) detectors [5] or Fano-Noise-Limited CCDs [1].

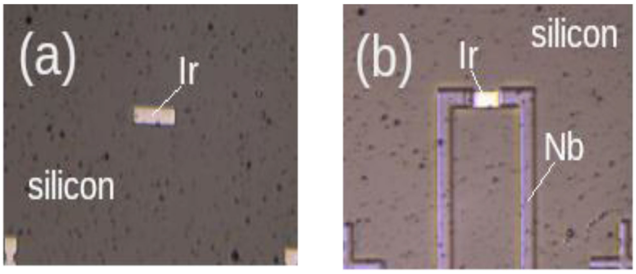

Fig. 2. (a) Ir detector film on the silicon substrate after patterning; (b) Ir detector film with Nb wiring fabrication.

charge exchange with metallic bulk, which will allow the IC decay, since the first ionization potential of thorium (6.3 eV) is lower than the isomeric excitation energy. Finally, the consequent transition to the ground state will be fully detected. Since the half-life of the ^{229m}Th state is very short, development of a fast TES working in 10 eV energy domain with a sub-eV resolution is required. Moreover, a large half-life of ^{233}U mother isotope ($\sim$1.6 $\times$ 10^5 y) and small penetration depth of thorium ions make it necessary to design a detector with a large active area. A detector array of 2 $\times$ 10^4 μm^2 active area with signal rise-time of few μs and fall-time smaller than 7 μs placed 1 mm away from a Ø10mm ^{233}U source with an activity of about 5 kBq would yield an acceptable detected event rate of about 1 event per day.

In this paper, we report on development of a first prototype of TES detector based on an iridium-gold bi-layer.

II. Prototype Fabrication

In the following, we explain the fabrication process of the Ir/Au TES. First, a 150 nm thick layer of iridium was deposited on a silicon substrate chip by means of pulsed laser deposition [7] in a dedicated set-up. A 400 μm^2 detector, shown in Fig. 2 (a), was then patterned in the following steps: a) photo-lithography with a positive photoresist ma-P1275 [8] to cover the detector area, b) RIE-etching of the uncovered area to remove excess iridium from the substrate, c) lift-off of the photoresist mask with acetone.

Afterwards, the detector was wired to its readout SQUID system (see Fig. 2 (b)). For that, a) photo-lithography with a negative photoresist ma-N440 [8] was done to pattern the wires, b) a thin ($\sim$200 nm) niobium film was sputter-deposited inside a dedicated vacuum chamber, c) the excess material was lifted off with acetone. The niobium wiring covered about 100 μm^2 of the detector area.

To shield inactive parts of the detector system from visible photons,[2] which may produce indirect thermal pulses generating background in the detector, a gold-plated PMMA mask was made. The bonding pads of the Nb wiring were covered with the Kapton film strips and a layer of PMMA photoresist has been deposited on top of the chip. Then, 50 nm of Au was thermally evaporated covering the PMMA mask and the Ir film. The Kapton strips were removed freeing the Nb bonding pads. A scheme representing the final layer configuration of the TES can be seen in Fig. 3.

[2]Only a tiny fraction of light is illuminating the detector due to a large distance between the end of the optical fiber delivering the light and the chip surface, see next section "Calibration measurements".

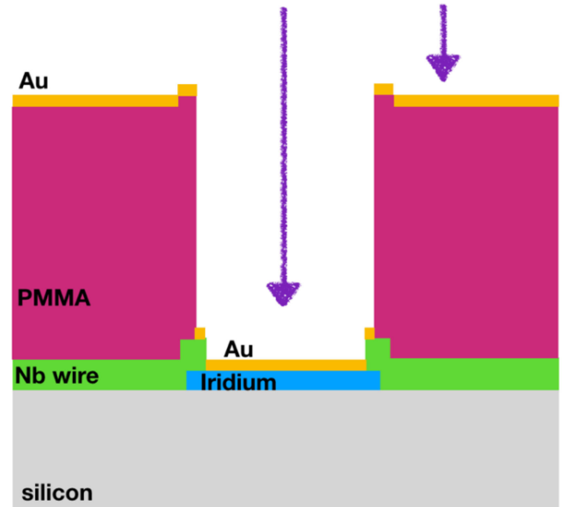

Fig. 3. Scheme of the section of the Ir/Au TES prototype after fabrication. Arrows indicate irradiation direction.

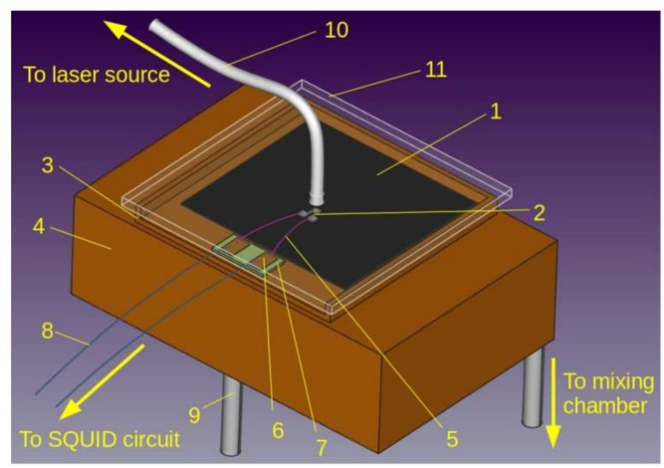

Fig. 4. Schematic representation of the prototype calibration setup. The silicon chip (1) with the detector (2) was attached to a copper holder (3) placed on a heavy copper block (4). The detector wiring pads were connected via bonding (5) to copper pads (6) sitting on an insulating layer (7) at the copper holder. From the copper pads two wires (8) lead to the SQUID circuit. The block was mounted on the mixing chamber of the cryostat with steel tubes (9). The optical fiber (10) delivering the light from the pulsed laser source outside the cryostat was fixed into a hole of a Plexiglas lid (11) above the detector.

III. Calibration Measurements

The chip with Ir/Au TES was mounted onto a heavy copper block at the mixing chamber of the Oxford Kelvinox K25 cryostat, as shown in Fig. 4. A VTT K3B DC SQUID, thermally anchored inside the cryostat at the mixing chamber and shielded from external electromagnetic sources with a Nb box, was used read out TES response. The TES was connected in series to

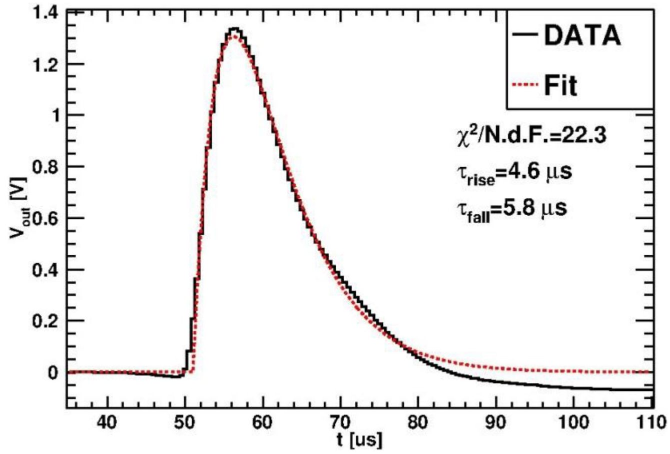

Fig. 5. Average measured signal from the detector and its amplification chain in response to the single photon absorption and its fit with (1). The decay tail of the signal is distorted by the electronic filtering and is not described by (1), therefore the signal was fit up to t = 70 us.

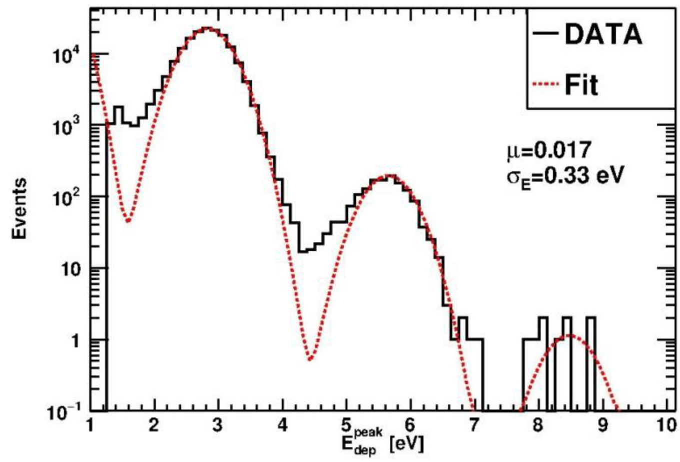

Fig. 6. Deposited energy spectrum with single-, double- and triple- photon peaks calibrated using the positions of baseline and the single photon peak.

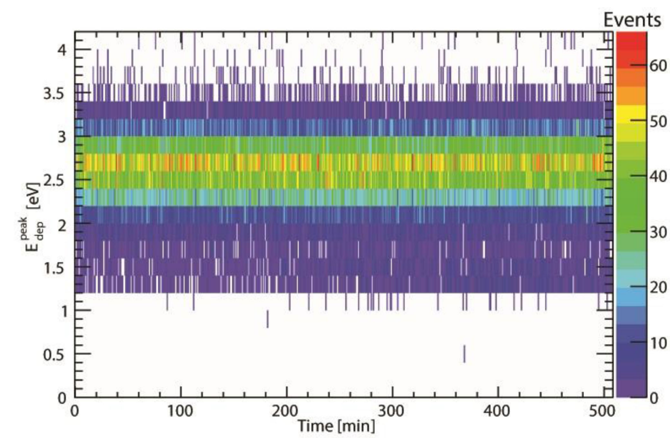

Fig. 7. Stability of the measured single-photon peak position with time during a continuous run.

an input coil, inductively coupled to the SQUID. The SQUID inductively coupled to a feedback coil was connected to an amplification feedback circuit from Magnicon XXF-1 set [9]. The output of the circuit was filtered with a SR560 low-noise voltage amplifier. The ultimate passband of the readout system was from 0.3 kHz to 80 kHz. The calibration of the TES was performed using a 439 nm (2.824 eV) pulsed laser source (PLP-10-044, Hamamatsu Photonics K.K) with 100 ps pulse-length. The laser was connected to an optical fiber embedded inside the cryostat.[3] There, on the detector side, the optical fiber (Ø50 μm, numerical aperture 0.22) was fixed into a hole in a Plexiglas lid which was microscopically aligned to the detector, ∼2 mm away from the TES surface. This gave ∼1 mm wide light spot on the chip. In this way, the response of the TES and its amplification chain to single-photon absorption could be measured. The measurements were done with the base temperature of about 90 mK. The detector had a superconducting transition around 110 mK.

The response of the TES and its amplification chain to a single-photon absorption shown in Fig. 5 was measured by averaging the signal over all single-photon events. The signal was fitted by empirical signal model described by the amplitude A, the rise-time $\tau_{\rm rise}$ and the fall-time $\tau_{\rm fall}$:

$$V\ (t) = \ A \times \left[e^{\frac{-t}{\tau_{fall}}} - e^{\frac{-t}{\tau_{rise}}} \right] \qquad (1)$$

resulting in $\tau_{\rm rise} = 4.6 \pm 1.7\ \mu$s, $\tau_{\rm fall} = 5.8 \pm 2.1\ \mu$s.

The distribution of the energy deposited onto the active TES area was measured and is shown in Fig. 6. In the spectrum, one can identify three Gaussian-like single-, double- and triple-photon absorption peaks which appear due to the detector response being relatively slow compared to the laser pulses. An exponential tail at low energy is due to accidental triggering on detector noise and is cut by the electronics threshold. The three well-separated photon absorption peaks were fitted with a convolution of Poisson distribution and a Gaussian describing finite detector resolution ($E_{\rm dep}$ is energy deposited in the detector, E_γ is energy of the incident photon):

$$N\ (E_{dep}) = \sum_n e^{\frac{-\left(\frac{E_{dep}}{E_\gamma} - n\right)^2}{2\sigma_E^2}} \frac{\mu^n}{n!} e^{-\mu} \qquad (2)$$

giving $\sigma_{\rm E} = 0.335 \pm 0.001$ eV corresponding to energy resolution $\Delta E_{\rm FWHM} = 2\sqrt{2\ln(2)}\sigma_E$ ∼0.789 eV and the mean number of absorbed photons per pulse of $\mu = 0.017 \pm 0.0005$ eV.

The result parameters, which are in agreement with a previous measurement of thermal conductance of Ir TES [10], are within the experiment requirements.

Due to small geometrical acceptance of the TES the detected event rate is expected to be low, of the order of 1 event per day, which requires detector response to be stable on the correspondingly large time-scale. A continuous measurement of the signal from the prototype detector demonstrated that the

[3]The cryostat has an optical fiber connector on its outer flange which allows to connect a laser source outside and an optical fiber inside the cryostat. The optical fiber inside the cryostat is passed along the ^{3}He/^{4}He circulation system towards the lowest part where the detector is mounted. It is wound and fixed with heat-conducting glue at a couple of stages for a more efficient thermalisation.

single-photon peak position variation was less than 0.02 eV (0.7%) over ∼8 hours, see Fig. 7. A fit to the single photon peak position distribution in time gave the linear coefficient of $3.6 \times 10^{-5} \pm 2.4 \times 10^{-5}$ eV/subset compatible with zero within 1.5 RMS.

IV. Conclusion

In this work we reported about the development and test of a single Ir/Au TES prototype for the experiment for direct detection of ^{229m}Th isomeric transition. The TES was demonstrated to satisfy the experimental requirements on the response speed and resolution and to have a stable response. Such a technique will allow to observe all possible types of decays of ^{229m}Th in the range of energy from 3 to 50 eV and lifetimes of >5 μs.

The last step in the development of the detector will be fabrication of an array of such TESs, for which in the first order reproducibility of the presented TES with the same characteristics has to be checked. Finally, we plan to fabricate and test an array of TESs connected in parallel together and organized in groups each with a single-channel DC SQUID readout. The size and number of single detectors *n* will be optimized, as appropriate, with regard to the trade-off between the active area, which scales with *n,* and the energy resolution, which scales as $\sim n^{1/2}$. The relative response uniformity of *n* TESs should be set to 3% in order to give a negligible impact on the energy resolution, about 10% at 8 eV.

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