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# Extensive studies of $CaMoO_4$ scintillation crystals for search of dark matter experiments

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We present results of extensive studies of CaMoO4 crystals for dark matter experiments. Light emission kinetics and absolute light yield of the crystals were measured thoroughly at room temperature. The light emission kinetics of the crystal has a complicated multi-exponential behavior with the main component with 15-16 μs decay time. The observed fast components with 12-46 ns decay time have small (≤1%) contribution to the total light yield. The latter was measured to be ~3000 photons per MeV at room temperature (22°C). The temperature dependence of the crystals parameters were measured in the wide range of 1-300K. It is shown that CaMoO4 crystals are very interesting for dark matter experiments and for neutrinoless double-beta experiments too.

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## 1. Introduction

The last decade of the 20th century has been marked by a tremendous scientific achievement – the discovery of neutrino oscillations which the finiteness of the neutrino rest mass is inferred [1, 2]. The next step of experimental endeavors is in the direction of shedding light onto the nature of neutrino by establishing them as either Dirac or Majorana particle and in the mass determination. This task is being undertaken by experiments searching neutrinoless double beta decays of some specific atomic nuclei [3]. There are a number of experiments presently running or being planned or installed [4, 5, 6, 7]. The half life times in the order of $10^{26}$ years require high purity setups with rigid background suppression. In this respect it seems the $^{100}$Mo isotope is very promising due to its rather high reaction Q-value $Q_{\beta\beta}$ ~ 3 MeV which is above the main background lines from natural radioactive decay [8]. Keeping in mind this fact we studied scintillation properties of CaMoO$_4$ crystals. The scintillation properties of crystals will play a crucial role in designing new experiments. Therefore, it is of utmost importance to know well the crystal scintillation parameters like its light yield and light emission kinetics.

### 1.1 Light emission kinetics

The crystal was grown by the Czochlarski method and it has cubic dimensions of $20\times20\times20$ mm$^3$. So far it has been claimed in a number of publications that CaMoO$_4$ crystals have just a simple single exponential scintillation emission with only one decay time constant of 15-17 μs [9, 10, 11, 12]. Only in reference [13] the existence of a fast scintillation component with a decay time constant of 10 ns was reported. However, it was observed at low, cryogenic temperature of ~10 K.

In our measurements we observed for the first time the fast scintillation emission components of CaMoO$_4$ crystal at room temperature seen after irradiation by α-particles with $E_\alpha$=5.5 MeV from a $^{238}$Pu and by γ-quanta with $E_\gamma$=662 keV from $^{137}$Cs. The scintillation emission kinetics has been measured by a time correlated single photon counting technique (TCSPC) [14, 15]. Two opposite sides of the crystal were viewed by fast 1” photoelectron multipliers (PMTs) (R1398 by HAMAMATSU Photonics). One of them was directly optically coupled to the crystal and it was used to form trigger signals of the set-up. Another PMT was fixed at a certain distance from the crystal to provide single photoelectron level of illumination from the crystal, which is necessary for the TCSPC technique. The time spread of the trigger signals is less than 1 ns (FWHM). The lateral sides of the crystal were wrapped by TYVEK diffuse reflective paper from 3M [16]. The anode signals of both PMTs were amplified by fast amplifiers with ~1 ns rise time and fed to constant fraction discriminators with thresholds set to ~0.25 pe, where 1 pe is a mean charge of the PMTs single photoelectron pulses. The output signals of the discriminators were used as SRART and STOP signals for a time-to-digital-converter (TDC). Two different TDCs were used to measure the crystal scintillation light emission kinetics. The first, “slow”, TDC was used for measurements of full emission kinetics of the crystal and has 200 μs range and 67 ns step. Another, “fast”, TDC with 5 μs range and 75 ps step was used for thorough studies of fast components of the crystal scintillation light.

It turned out that the scintillation emission kinetics of the crystal at room temperature with irradiation by α-particles and γ-quanta has a rather complicated multi-exponential character. It can be seen in Fig. 1, where the whole light emission kinetics of $CaMoO_4$ crystal, registered in the wide time range of 120 μs, is presented.

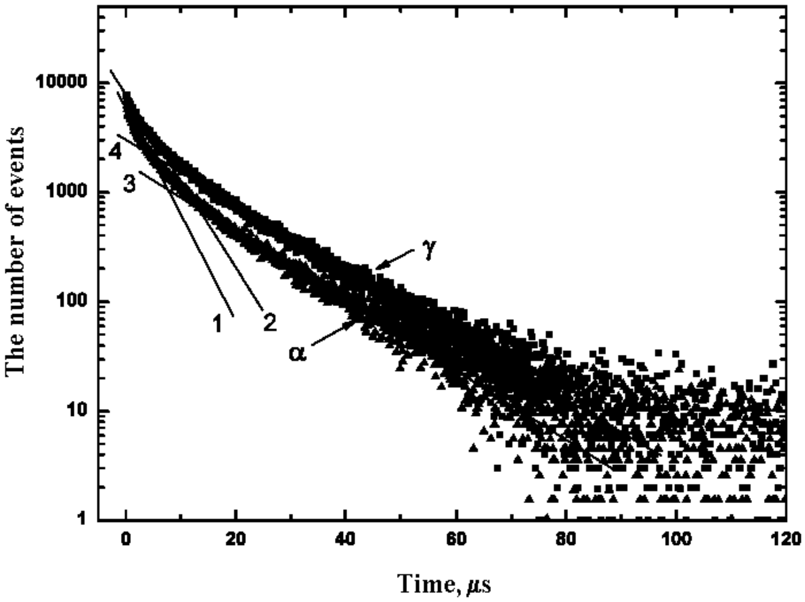

Fig. 1. The scintillation emission kinetics of $CaMoO_4$ crystal after α-particles and γ-quanta irradiation in the time range of 0-120 μs. The insert shows another measurement in the time range up to 450 ns.

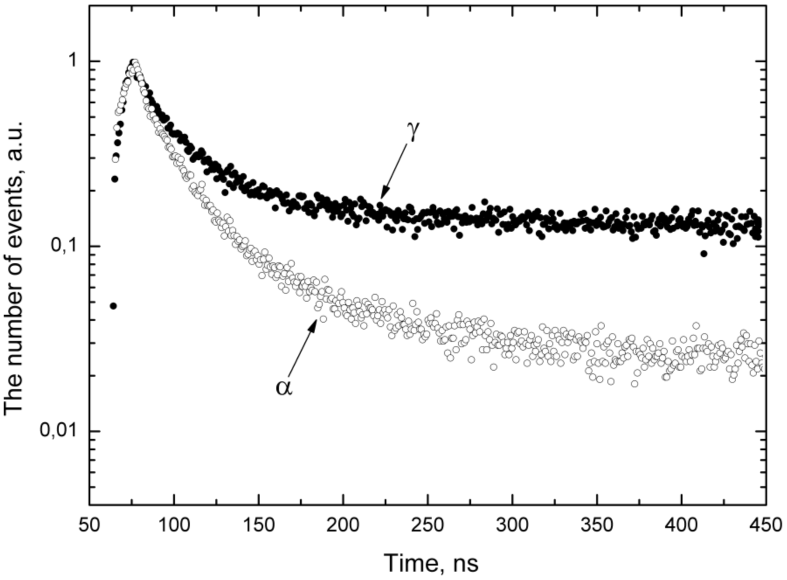

Fig. 2. The scintillation emission kinetics of $CaMoO_4$ crystal after α-particles and γ-quanta irradiation in the time range of 0-450 ns. The insert shows another measurement in the time range up to 450 ns.

All components of the crystal scintillation emission can be subdivided into three groups: slow, intermediate and fast. The first, slow, group consists of two components with decay time constants of 5 μs (29%) and 15.8 μs (65%) for γ-quanta as well as 4.6 μs (35%) and 15.2 μs (58%) for α-particles, the components contributions to the crystal total light yield are shown in brackets. The intermediate components have decay time constants of 1.4 μs (5.5%) and 1 μs (6%) for γ-quanta and α-particles respectively. The slow and the intermediate components can be clearly seen in Fig. 1.

The third group includes fast components of the $CaMoO_4$ crystal scintillation. In Fig. 2 the fast components are clearly seen for both, α-particles and γ-quanta irradiation. In fact there are two fast components with decay time constants of 12.0 ns and 36.0 ns for α-particles and 15 ns and 46 ns for γ-quanta. The fast components summed contribution to the crystal's scintillation total light yield does not exceed 1%. The fast components emitted under α-particles irradiation yield two times more light than in case of γ-quanta, constituting 1% and 0.5% of the total light yield respectively.

Thus the slow components with ~15 μs decay time make the main contribution to the $CaMoO_4$ crystal scintillation light yield. Observed multi-component character of scintillation emission of the $CaMoO_4$ crystal with fast components with decay time of 10-50 ns should be taken into account by future experiments

## 2. Scintillation Light Yield of $CaMoO_4$ scintillation crystals.

Light yield measurements with these crystals are not so easy issue. The long decay time of the crystal imposes notable experimental problems. For evaluation of the crystal scintillation light yield we proposed a simple and rather robust method. The method is based on the detailed knowledge of thoroughly measured scintillation light yield of the crystal, as described in the previous section of the paper and in [17]. Moreover we measured other important optical parameters of the crystal including its refractive index, transparency and emission spectrum.

Light emission spectrum of the crystal was measured using UV LED emitting at 255 nm and a monochromator. The emission spectrum reaches a maximum at 540-560 nm with a long tail going far into the red region of the spectrum, Fig.3. It makes the crystal to be "yellow" and expose additional experimental problems for absolute light yield measurements. The shape of the measured spectrum (filled rectangles) coincides well with spectrum (line) reported in [11-13]. To register enough photons one should use a proper photodetector with sensitivity matching the crystal's light emission spectrum. Another important point is the crystal has a large refractive index n – its direct measurement shows that n=2.0±0.1 in the wavelength range of 470-600 nm. The fact forces us to be careful in choosing a proper optical contact between the crystal and photodetector and measure directly photons collection on the photocathode of photodetector. The optical grease BC-630 was used for providing optical coupling of the crystal and PMT. The grease refrefractive index is 1.59 minimizing photon lost en route from the crystal to PMT photocathode. We chose photomultiplier XP5302B for its high sensitivity in green region of spectrum. The PMT has a mean quantum efficiency of 16.3% convoluted with the crystal emission spectrum within region of 400-650 nm. It should be noted here that the maximum quantum efiiciency of the PMT photocathode exceeds 40% at 350-560 nm, Fig. 4. Effective collection efficiency of photoelectrons to the first dynode of XP5302B is close to 100%. Single photoelectron charge spectrum was measured carefully for the PMT and the excess-noise factor

(F-factor) has the value of 1.3 for the PMT. The crystal was wrapped by VM2000 foil with high reflactivity at wavelenths higher than 380 nm. The photon collection efficiency was measured to be ~0.85. The crystal was illuminated by 5 MeV $\alpha$-particles and 662 keV $\gamma$-quanta from $^{241}$Am and $^{137}$Cs radioactive sources respectively. Anode pulses of the PMT were fed directly to the input of spectrometric amplifier CANBERRA 2020 with 12 $\mu$s integration time. Output pulses of the amplifier were measured by multichannel analyzer CANBERRA 3100. All measurements were done at room temperature (22°C).

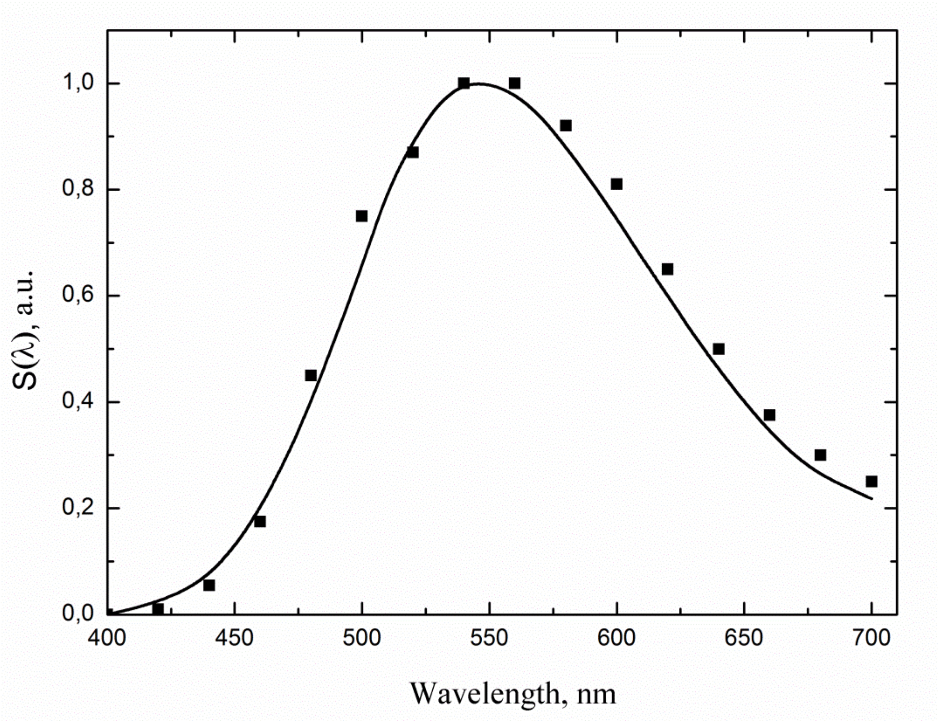

Fig. 3. Photoluminescence emission spectrum of CaMoO$_4$ crystal measured under irradiation by UV LED with $\lambda_{max}$ =255 nm.

The charge spectrum measured by such a simple set-up under irradiation by 662 keV $\gamma$-quanta from $^{137}$Cs source is shown in Fig. 5. The full absorption peak is clearly seen. The careful analysis of the peak shows that the peak corresponds to ~165 photoelectrons and light yield of the crystal with 400-650 nm spectrum region is evaluated to be ~3000 photons/MeV. All above mentioned data on the PMT sensitivity, photoelectron collection efficiency, excess noise factor, as well as, materials reflectivity, the crystal emission spectrum etc, were taken into account in this analysis. It should be noted here that the measured scintillation light yield is not absolute light yield, it is just a part of the total light yield which lays within 400-650 nm.

Another important parameter of scintillation crystals for experiments searching rare events like dark matter particles and neutrinoless double beta decay is crystals $\alpha/\beta$ ratio. This parameter reflects distinct response of crystals to $\alpha$-particles and $\gamma$-quanta irradiations due to different ionization density [18, 19]. We measured the $\alpha/\beta$ ratio for CaMoO$_4$ crystal by simultaneous irradiation with $^{241}$Am and $^{137}$Cs radioactive sources. Knowing energies of $\alpha$-particles and $\gamma$-quanta it is easy to define this ratio to be $\alpha/\beta$ = 0.25.

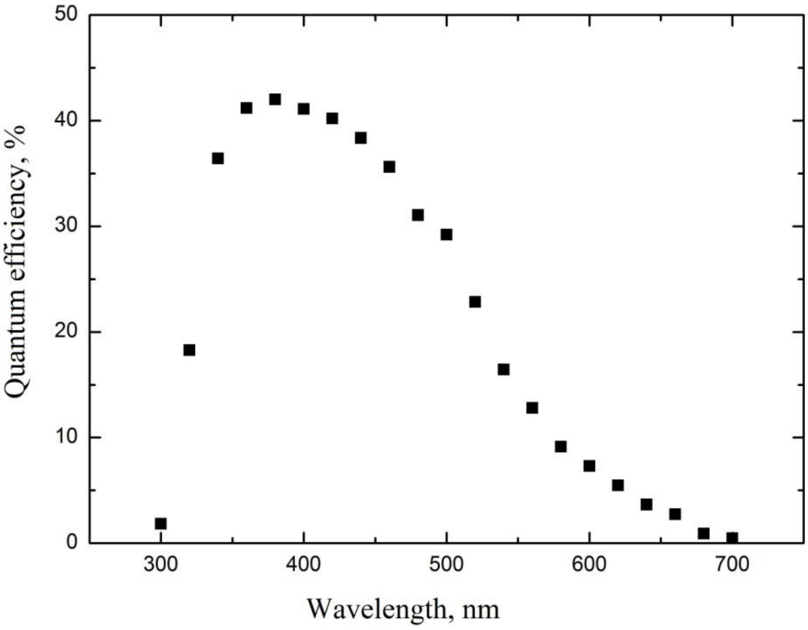

Fig. 4. Dependence of quantum effiency of XP5302B PMT on wavelength.

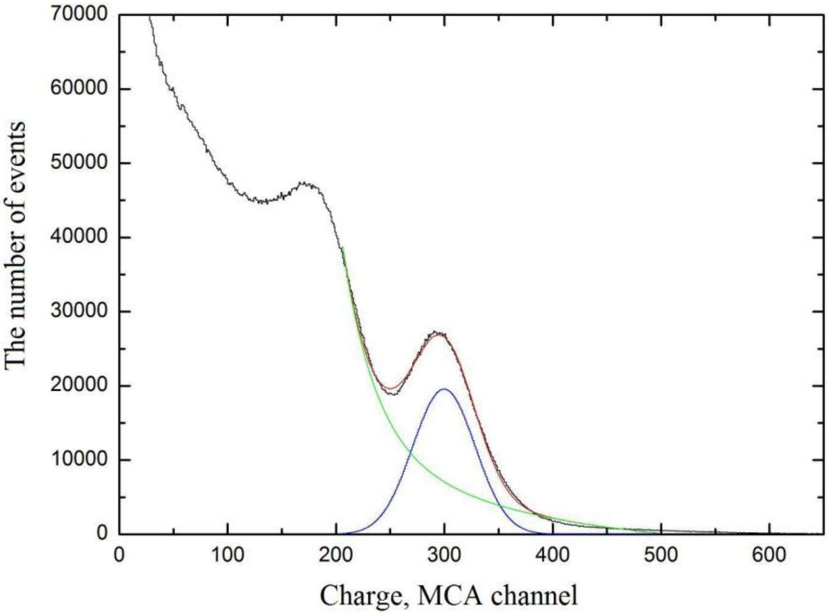

Fig. 5. Charge spectrum measured with CaMoO$_4$ crystal under irradiation by 662 keV $\gamma$-quanta from $^{241}$Am radioactive source.

Temperature dependence of the crystal scintillation parameters in the wide range of 1-300K are presently under studies with liquid helium optical cryostat. The cryostat equipped with two quartz widows. Ther crystal inserted into cryostat cooled down and viewed by two PMTs from outside. The crystal irradiated by $\alpha$-particles from $^{241}$Am radioactive source which is immersed into the cryostat and fixed close to the crystal. Thus the PMTs of the set-up are at constant room temperature and one can not worry about on temperature dependence of the PMTs parameters – sensitivity, gain etc. Waveforms of output signals of both PMTs are digitized and processed to measure the total charge and arrival times of scintillation photons. Preliminary results demonstrates that the crystal scintillation paraneters temperature dependences differ substantially from previously reported in [11, 12, 13], for scintillation light yield in particular. In Fig. 6 the temperature dependence of the crystal light yield is shown. The light yield increases by a factor of more than 2 as temperature decreases from 300 K to 250 K reaching a plateau till 200 K. With temperature decreasing further the light yield decreases and starts increase slightly after 50 K and finally drops down after 10K getting less than at room temperature. At the same time the crystal's light emission kinetics is getting steadily larger and larger as temperature decreases. The decay time of the main scintillation component of the crystal encreases from 16 μs at 300 K up to ~600 μs at 10 K.

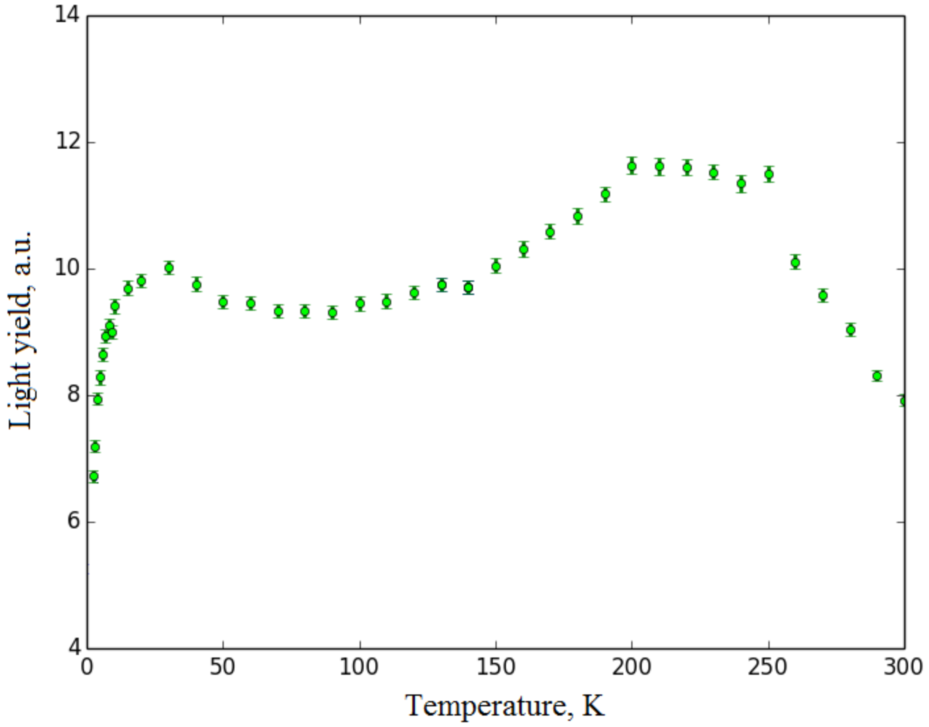

Fig. 6. Charge spectrum measured with $CaMoO_4$ crystal under irradiation by 662 keV γ-quanta from $^{241}$Am radioactive source.

## 3. Conclusion.

$CaMoO_4$ scintillation crystal is a very promising material for use in experiments searching for rare events such as dark matter and neutrinoless double beta decay experiments. The crystal light yield and emission kinetics are thoroughly studied. For optimal operation of such crystals in experiments it is desirable to find an optimal photodetector with sensitivity matching well the emission spectrum of crystals. Cooling the crystal down to liquid helium temperatures demonstrates rather controversial results. So further tempearature studies of the crystal basic scintillation parameters are urgently needed.

## 4. Aknowledgements.

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## References

[1] M. Maltoni, T. Schwetz, M. A. Tortola and J. W. F. Valle, *New J. Phys.* **6** (2004) 122.

[2] G. L. Fogli et al., Phys. Rev. D 78 (2008) 033010.

[3] F. T. Avignone, S. Elliott and J. Engel, Rev. Mod. Phys. **80** (2008) 481.

[4] GERDA, Proposal to the LNGS (2004), http://www. mpi-hd.mpg.de/gerda.

[5] MAJORANA, White paper (2003) nucl-ex/0311013

[6] S. Pirro et al. , Nucl. Instr. and Methods A 559 (2006) 352

[7] R. Arnold et al., Phys. Rev. Lett. 95 (2005) 182302

[8] Yu. Zdesenko, Rev. Mod. Phys., 2002, V.74, P.663.

[9] S. B. Mikhrin et al., Nucl. Instr. and Meth. A. 2002. V.486. P.295.

[10] S. Belogurov et al., IEEE Trans. Nucl. Sci. 2005, V.52, N.4, P.1131.

[11] V. B. Mikhailik et al., Nucl. Instr. and Meth. A. 2007. V.583, P.350.

[12] A. N. Annenkov et al., Nucl. Instr. and Meth. A. 2008. V.584, P.334

[13] V. B. Mikhailik et al., J. Phys.: Condens. Matter, 2005, V.17, P.7209.

[14] D. V. O’Connor, D.Philips, Time-Correlated Single Photon Counting, London, Academic Press, 1984.

[15] W. Becker, Advanced Time-Correlated Photon Counting Techniques, Dortmund, Springer, 2006.

[16] TYVEK registered by 3M Company, St. Paul, MN 55144.

[17] R.V. Vasiliev et al. Instruments and Experimental Techniques. 2010. Vol.53. No.6. P.795.

[18] J.B. Birks. The theory and practice of scintillation counting. New York. 1964.

[19] G.F. Knoll. Radiation detection and measurement. New York. 1989.