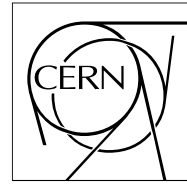


The Compact Muon Solenoid Experiment

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SYSTEMATIC STUDY OF THE SHORT-TERM INSTABILITY OF PbWO_4 SCINTILLATOR PARAMETERS UNDER IRRADIATION

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Abstract

The effect of irradiation on lead tungstate PbWO_4 (PWO) scintillator properties has been studied at different irradiation facilities. Lead tungstate crystals, grown with the oxide content in the melt tuned to the stoichiometry of pure sheelite or sheelite-like crystal types and doped with heterovalent, trivalent, and pentavalent impurities, have been studied in order to optimize their resistance to irradiation. A combination of a selective cleaning of raw materials, a tuning of the melt from crystallization to crystallization, and a destruction or compensation of the point-structure defects has to be used to minimize the short-term instability of PWO parameters under irradiation.

Submitted to Radiation Measurements

INTRODUCTION

The CMS Collaboration [1] R&D efforts related to lead tungstate PbWO_4 (PWO) scintillator technology development have already led to the reproducible production of a quite transparent, fast scintillator with acceptable light yield. In spite of significant progress in the improvement of PWO scintillation properties, the prototype technology for the production of PWO crystals with an acceptable level of transmission damage under irradiation is still under development. To solve this problem a systematic study of the variation under irradiation of PWO crystal properties, mainly produced in Russia, has been organized at different facilities. It allowed a fast or detailed investigation mode of the parameter changes for full-sized PWO elements (23 cm in length) at different dose rates. Such an organization made possible two research levels: a fast check of the produced crystals and a detailed investigation of the most important samples.

For a high-precision calorimeter the scintillator's light output must remain unchanged under irradiation in order not to produce non-uniformities of the crystal response due to a non-uniform irradiation. Moreover, the variation of the crystal response cannot exceed a few per cent and must be slow in order for the monitoring system to correct for these variations with a precision of about 0.2% [2]. Thus a short-term instability of the scintillator parameters dependent on dose rate or accumulated dose is not acceptable for the design of a precise calorimeter.

Although damage to the transmission of 23 cm long PWO scintillation elements is detectable and can achieve some tens of a per cent in the worst case, the value of the absorption induced by irradiation is rather small and does not exceed a few inverse meters corresponding to an attenuation length of more than one meter for the majority of crystals. Even for the worst crystals the total concentration of the damaged defects is of an order of magnitude of 10^{17} cm^{-3} to be compared to the lattice atom density of 10^{22} cm^{-3} . At such a level of defect content in crystals obtained from standard raw materials, the separate contribution of the impurities and of the point-structure defects to be recharged under irradiation cannot be distinguished. Although the separated contribution of the impurities could easily be studied by increasing their concentration, this is not possible for the point-structure defects.

Another aspect is related to the damage kinetics which describes the equilibrium between the damage/recovery processes and establishes the acceptable amount of the different kinds of defects. The transmission damage kinetics has already been described elsewhere [3].

In this paper the stability of the scintillating centers under irradiation is established and the conclusion is that the damage results from the attenuation of the scintillation light by color centers. The origin of the damaged centers is discussed. Also the results of a systematic investigation of the tuning of crystal growth conditions (stoichiometry scan) based on 56 full-sized crystals produced by the Bogoroditsk Techno-Chemical Plant in Russia are presented. Ways to suppress or compensate the remaining defects by specific doping of PWO crystals are proposed. For the last point the results based on a large statistical sample of full-sized crystals are presented in Ref. [4].

MEASUREMENTS AND IRRADIATION FACILITIES

The changes of absorption in the sample due to the irradiation are evaluated using the radiation-induced absorption coefficient k_{ind} defined as:

$$k_{\text{ind}} = (1/L) \ln (T_0(\lambda)/T(\lambda)) \quad (1)$$

where $T(\lambda)$ stands for the transmission after the irradiation, $T_0(\lambda)$ stands for the initial transmission, L is the length of the PWO sample. A clear correlation was observed for each

crystal between the collected light loss measured at the back of the crystal on a photomultiplier tube (PMT) and the longitudinal transmission loss characterized by the induced absorption coefficient k defined in equation (1). The number of photons detected on the PMT before (N_0) and after irradiation (N) is given at the first order by:

$$N_0 = I_0 \times \exp (-k \times L_{\text{mean}}) , \quad (2)$$

$$N = I \times \exp (-(k + k_{\text{ind}}) \times L_{\text{mean}}) , \quad (3)$$

where I_0 and I are the scintillation yields, not necessarily equal before and after irradiation, L_{mean} is the mean free path for photons between the point of emission and the PMT, k is the initial absorption coefficient at the wavelength of peak emission of PWO (about 500 nm) and k_{ind} is the induced absorption coefficient at the same wavelength after irradiation. Strictly speaking, equations 2 and 3 are first order approximations of more complex equations [5]. In particular, with this formalism, L_{mean} is a function of the transparency of the crystal, given by k and k_{ind} , of its other optical characteristics (surface quality, coating or wrapping) and of the photodetector. L_{mean} decreases when k (or $k + k_{\text{ind}}$) increases. However, for a small induced absorption, it can be considered as constant for a given crystal. Assuming (2) and (3), simple mathematics leads to:

$$k_{\text{ind}} \times L_{\text{mean}} = \text{Ln}(N_0 / N) - \text{Ln}(I_0 / I) . \quad (4)$$

The evolution of k_{ind} versus $\text{Ln}(N_0 / N)$ during recovery after irradiation is shown in Fig. 1 for a large number of crystals of different initial transmission. Each crystal has a different slope which depends on the value of L_{mean} . However, all the lines point to the origin within the measurement and fitting errors, which proves that the scintillation yield did not change within the errors (about 1%) under irradiation for all these crystals. It indicates that at least the fundamental properties of the radiating centers in PWO are not influenced by irradiation.

To clarify the features of the damage origin of the scintillating crystal under irradiation, as many parameters as possible have to be measured in an irradiation environment. But practically, only just the optical transmission of PWO crystals shows a detectable change under irradiation and is sufficient to explain the loss of the collected light. Such a change of the transmission in a spectral region is measured directly by a single- or double-beam spectrophotometer after the irradiation. Also, its relative change can be determined by recording the intensity of a monitoring light injected through the wrapped crystal equipped with a properly protected photodetector during the irradiation. The relative change of the monitor signal from the initial value is then proportional to the transmission change. Depending on the absorption spectrum induced by the irradiation, the monitoring signal variation at some wavelengths is correlated with the decrease of the particle signal detected in the scintillator. The best combination for this study is the measurement of the transmission, the monitoring, and the particle signals in the same experimental set-up implemented in beam facilities but accessible only for a limited time each year. So independent investigation of the crystal transmission or monitor signal changes is implemented at irradiation by the widely used ^{60}Co source.

Keeping in mind that under LHC irradiation PWO will suffer a dose rate of 15–20 rad/h, the following irradiation facilities have been used both for very fast and detailed study of the crystals under irradiation:

COCASE Facility, at Saclay, Dapnia [6]. It consists of the ^{60}Co (1.25 MeV, 14 Ci) source with geometrical tuning of the dose rate in the region below 2 R/min. Relative change of the monitoring signals in the spectral region 380–800 nm is measured. During the irradiation the monitoring light pulses are generated by a spectrometer made of a computer-driven fast-tuning

monochromator, a Xe pulse lamp, fiber optics feeding light to the crystals under test, and electronics for recording the short- and long-term instability of the light pulses. Such a system provides registration of the monitor signal changes at specific wavelengths in the range mentioned above. The minimal available wavelength step is 5 nm. Also, longitudinal transmission is measured by a Perkin-Elmer spectrophotometer in the region 300–850 nm for the crystal before and after its irradiation.

⁶⁰Co therapy unit of the Geneva Hospital provides the irradiation of the crystals with an irradiation rate of 420 R/min. Change of the crystal transversal and longitudinal transmission, light yield, and scintillation kinetics are measured 40 minutes after the irradiation by a set of spectrometers located at CERN.

INP ⁶⁰Co source facility. It consists of the ⁶⁰Co well-shaped source with fixed irradiation rate of 14000 R/min. Change of the crystal transmission is measured 60 minutes after irradiation by a Varian ‘Cary1E’ spectrophotometer in the range 300–900 nm. Control of other scintillation parameters is also carried out.

INR irradiation facility. A microtron MK-25 and linear electron accelerator (both of 25 MeV electron energy) are used as γ sources. Irradiation rates are tuned from 0.1 to 3000 R/min. Longitudinal transmission is measured in the range 390–650 nm during the irradiation by a specially developed spectrophotometer based on a transparent grating.

CERN H4 beam [7] is also sometimes used as an irradiation facility. It includes specially constructed thermostabilized equipment for the simultaneous measurement of the time dependence of the particle signal and monitor signal in PWO scintillators. Light monitoring signals are generated by the same type of spectrometer as the one which is used in the COCASE facility.

CERN X5 beam facilities are under development. With a ¹³⁷Cs source and the possibility to probe the crystal in parallel with a muon or an electron beam, they will be equipped as far as possible with the best measuring systems tested in the above-mentioned facilities. They should become the main facilities for crystal tests at the CMS mass production stage.

RECHARGEABLE DEFECTS IN PWO CRYSTALS

Radiation damage in the crystal appears to be due to the charge-state change of the existing point defects. They are divided into two groups: the substitution defects and the point-structure defects. The radiation damage process is therefore driven by the change of the defects’ electronic states which provides recharged ions and color centers and the creation or suppression of their associated absorption bands.

The data obtained to date for the radiation-induced effects in PWO crystals are summarized as follows:

Irradiation of PWO crystals grown without specific tuning generates two absorption bands with maxima around 360 nm and 620 nm. Usually, induced absorption in such crystals reaches a value of about 10 m^{-1} in the region 400–600 nm.

There is a correlation between the initial absorption of crystals at the wavelength near 360 nm and the absorption induced by irradiation in the green–red region as shown in Fig. 2.

The value of the absorption induced by radiation in the green–red region is correlated with the presence of red luminescence emitting centers, however, the blue and green luminescence radiating centers themselves are not damaged by irradiation.

The concentration of the defects able to create metastable color centers under irradiation does not depend on the atmosphere of the crystal growth but is influenced by the purity of the raw material.

In conclusion only some impurities and point-structure defects contribute to the induced absorption in PWO crystals.

The point-structure defects in PWO crystals appear to be due to stoichiometric violation in the melt. This change may be casual in some fragments of the crystal or systematic if, for instance, the deficiency of one sort of atom becomes dominant. The dominating defects in the grown PWO crystal are due to lead leakage from the melt, however, two phases — pure sheelite and sheelite-like type crystals [8] or their solid solution appear at the lead tungstate crystallization. Contrary to pure sheelite, sheelite-like crystals have already some deficiency of lead, but organized in a superstructure with a large range charge compensation. One can expect that lead leakage creates cation vacancies $V_{c(\text{Pb})}$ whose concentration has three extremes in the system "pure sheelite–sheelite-like type" and depends on the portion of the pure sheelite in the solid solution (Fig. 3). In the crystals grown from purified raw materials such vacancies are compensated by anion vacancies V_o or different F-centers [9]. Cation vacancy locally generates O hole centers which may be separated or associated to form $(2O)^{2-}$ centers with anti-parallel spins of electrons rotating around two oxygen atoms and the cation vacancy. These hole centers being recharged under irradiation create metastable color centers with associated absorption bands. As we observed that radiation-induced effects in PWO crystals do not depend on the atmosphere of the crystal growth, we concluded that anion vacancies V_o do not contribute significantly to the discussed phenomena.

Among the impurities, even if they are present in small amounts, those which have charge transfer or interconfiguration transitions cause a detectable change of the crystal transmission. Let us consider some ions with charge transfer (CT) metal-ligand transition. Although the majority of the **3d** elements are involved in such transitions, only different Fe ions produce detrimental effects in PbWO_4 crystals. In wide-gap oxide crystals Fe^{3+} ions in tetrahedral coordination have $O^{2-}\text{-Fe}^{3+}$ transitions in the region above $35\,000\text{ cm}^{-1}$ (280 nm) [10]. In PWO they contribute to edge absorption or generate O centers with an absorption band around 350 nm [10]. The Fe^{2+} ions being reduced from trivalent state under the irradiation also have CT bands in the region above $28\,000\text{ cm}^{-1}$ (350 nm). Both ions have relatively low intensity of the intraconfiguration **d–d** transitions in the visible and infrared region but decrease the total PWO light yield due to overlapping of the CT levels and the radiating levels of the regular and irregular tungsten groups. An acceptable concentration of Fe in PWO crystals was already specified in raw materials during the PWO scintillation kinetics optimization [11].

Among the ions having interconfiguration transitions, the most important class is made of lanthanoids or rare-earth elements. Replacing divalent Pb ions in the host. Trivalent ions Ce^{3+} , Pr^{3+} , and Nd^{3+} have 4f–5d interconfiguration transitions. However, such ions are not converted in the reduced valent state under irradiation. Their absorption bands related to the 4f–5d transitions are in the UV region and do not influence the radiating properties of PWO. The Eu and Yb ions are easily reduced from a trivalent to a divalent stable state under irradiation by the capture of electrons created under irradiation. In the trivalent state they have just f–f transitions exhibiting weak narrow absorption bands in the visible and infrared regions. However, Eu^{2+} have $4f^75d^0\text{--}4f^65d^1$ transitions which generate a wide absorption band in the region below 400 nm in oxide crystals [12]. The Yb^{2+} ion has three interconfiguration transitions $4f^{14}5d^0\text{--}4f^{13}5d^1$ and its associated absorption bands overlap all visible regions [13] in oxide crystals. Both divalent ions have no interconfiguration luminescence at room temperature in PWO. The absorption spectra in Eu and Yb doped crystals induced by irradiation are shown in Fig. 4. They indicate that the presence of such heterovalent ions has to be carefully specified in the raw materials at a level below several ppm.

SUPPRESSION OF THE RECHARGEABLE DEFECTS IN PWO CRYSTALS

As previously noted [3], the radiation induced absorption k in the crystal is described by the following equation:

$$k = \sum_i \frac{k_i^{\text{sat}} S}{S + \omega_i d_i} \left\{ 1 - \exp \left[- \left(\omega_i + \frac{S}{d_i} \right) t \right] \right\}. \quad (5)$$

At fixed dose rate S , it depends with time t on the amount N_i of the defects of kind i to be recharged, $k_i^{\text{sat}} = N_i \sigma_i$ with σ_i the absorbing cross section of the center i , the recovery probability ω_i and damage constant d_i .

Thus, the minimization of the observed transmission damage for a given dose rate S is achieved by the decrease of the total amount of slowly decaying color centers N_i or by the increase of their recovery probability ω_i . As it was determined at a dose rate of 5 rad/min and with a reasonable damage constant value, the defects with recovery times of less than a few minutes do not provide a significant contribution to the transmission damage of full-sized PWO crystals.

Specified impurity ions are extracted from the raw materials by chemical purification, so their contribution to the creation of rechargeable defects in PWO crystals can be easily minimized before crystal growth. To prevent the appearance of rechargeable point-structure defects in the crystal, it is important to control the perfection of the crystal host structure. In the PWO case, it is realized by the choice of the crystal structure to a pure sheelite or a sheelite-like crystal type by a precise tuning of the oxide content in the melt. Figure 5 shows the results of the influence of the stoichiometric variation on the value of the absorption induced by irradiation in full-sized PWO scintillators. Such variation has been achieved by the addition of an appropriate oxide excess in the melt. Qualitatively the stoichiometric scan results agree quite well with the expectations as shown in Fig. 3. The stoichiometric scan has given the possibility to reach an optimal combination of the oxides in the raw material and to minimize the induced damage in full-size elements. However, it has not led to a complete suppression of the damaged defects. This is due to the difficulty to control the stoichiometry when a considerable part of the melt is pulled into the crystal causing a significant gradient of defects from the seed to the tail part. It cannot be fully compensated by the tuning of the melt at the beginning of the crystallization. Thus, in addition to the structure tuning, an additional compensation or destruction of the defects is required. These phenomena have been investigated in the pure sheelite crystals and will be studied soon in the sheelite-like structure as well.

The destruction of the defects is achieved by the doping of PWO crystals by pentavalent Nb metal ion as described in Refs. [9, 14]. The pentavalent Nb doping ion creates its own stable hole center when substituting to the W ion. The creation in the crystal of an additional amount of such stable hole centers decreases the amount of the unstable O hole centers needed for cation vacancy compensation. Hole centers generated by Nb can also take part in the recharging processes under irradiation. However, owing to the stability of the valence state of the Nb^{5+} ions, such a reduced hole center has a short decay time. Therefore it does not contribute to the generation of a stable color center but rather favors the fast recombination of free carriers created by irradiation. As the Nb concentration in the PWO crystal becomes similar or higher than the V_c one, such a fast channel of electron-hole recombination does not leave any chance for a slow recharge process of O hole centers near cation vacancies. With a further increase of Nb doping concentration, the probability of a local V_c compensation is increased by the destruction of hole centers near cation vacancy. Most probably they create well-compensated and therefore stable associated defects like $\text{Nb}^{5+}V_0V_cO^-$. However, a practical concentration of the Nb doping is limited to some hundreds of ppm as described in Ref. [11]. Hole centers near

cation vacancies may be compensated by the injection of stable trivalent ions replacing divalent lead ions in the crystal. The most convenient ions for such an application are Y, La and Lu which have no intracenter optical transitions and ionic radii compatible with the PWO structure.

EXPERIMENTAL RESULTS AND DISCUSSION

A large number of full-size doped and undoped crystals obtained after the optimization of the melt by stoichiometric scan have been investigated. Reference samples and full-sized elements obtained from crystals doped with La, Lu, Nb with concentration of the activator close to the optimum have been irradiated.

Figure 6 shows the initial transmission of crystals of the two sheelite types. A pure sheelite crystal grown without melt correction or any kind of compensation (Fig. 6a) has a clearly detectable absorption band with a maximum near 360 nm. As mentioned above, such crystals have shown a rather large radiation transmission damage in the spectral region 400–600 nm. A good correlation between the presence of this band in the crystal absorption spectrum and the value of the induced absorption has been observed [15] and confirmed by our research. The Nb doping at the level of some tens of ppm suppresses this absorption band, but not completely. La or Lu doping with a concentration of about a hundred ppm completely suppresses the absorption band and the transmission spectrum is not too far from ideal, as expected from Fresnel reflection losses. Both types of activators decrease the amount of $O^{\cdot-}$ centers near cation vacancies and suppress the 360 nm absorption band associated with them.

In sheelite-like crystals, the concentration of the V_c is considerably reduced and both Nb and La provide complete suppression of the $O^{\cdot-}$ centers with a slightly better transmission cut-off for Nb. The transmission spectra of such crystals are shown in Fig. 6b.

In Figure 7 the influence of suppression and compensation of the point-structure defects in the lead tungstate crystals is presented for pure sheelite crystals. Crystals grown from corrected melt, but without doping, show the appearance of two color centers with wide absorption bands maxima around 360 and 620 nm as indicated on Fig. 7a. The first band is due to the additional creation of $O^{\cdot-}$ centers in the irradiated crystal. Such metastable centers are probably stabilized around other macro defects or impurity traces in the host. The second absorption band is connected to the creation of the metastable color center $O^{\cdot-}V_cO^{\cdot-} + e^- \rightarrow O^{2-}V_cO^{\cdot-}$ due to the capture of one electron e^- . In the crystals obtained without tuning of the melt, the intensity of these absorption bands induced by the same irradiation conditions are 4–5 times larger than for crystals grown from optimized melt.

Figure 7b, c show induced absorption of the crystals doped by trivalent Lu and La ions. The absorption band with maximum at 620 nm is suppressed in these samples but a new band with maxima at 520 nm appears under irradiation. If the amount of the activator is too small, a partly compensated center $O^{2-}V_cO^{\cdot-} + La(Lu)^{3+}$ appears, jointly with the completely compensated centers $O^{2-}V_cO^{2-} + La(Lu)^{3+}$. Such a center capturing an electron gives a metastable color center $O^{2-}V_cO^{2-} + La(Lu)^{3+}$ which causes the 520 nm absorption band. When the activator concentration is increased, the concentration of partially compensated centers is reduced producing a decrease of the 520 nm induced absorption band. La doping has a positive influence on the PWO radiation hardness of small PWO crystals grown without additional tuning of the crystal to stoichiometry as described in Ref. [16]. Thus, the activation by trivalent ions may be used as an additive to stoichiometric tuning to compensate rechargeable hole centers near the cation vacancy.

Figure 7d shows an absorption spectrum of PWO:Nb crystal induced by irradiation. Owing to the destruction of the complex defects $O^{\cdot-}V_cO^{\cdot-}$ for an activator concentration close to the optimal value, the 620 nm band in the induced absorption spectrum disappears. Also the

360 nm band is significantly suppressed due to the reasons described above. Thus, the Nb activation can be used as an additive to stoichiometric tuning in order to destroy rechargeable hole centers near the cation vacancy.

The data of the transmission loss at 420 and 500 nm for the full-sized elements irradiated at the COCASE facility are shown on Fig. 8a, b. Crystal 1769 was obtained at the beginning of the series of crystallizations with stoichiometric tuning of the melt and defects compensation by La doping. Crystal 1772 is the last crystal from this series. Crystal 1755 was obtained at the end of a series of crystallizations with stoichiometric tuning of the melt and defect compensation by Lu doping. Crystal 1773 was obtained at the beginning of a series of crystallizations with stoichiometric tuning of the melt and defects destruction by Nb doping. The results show that for crystals grown from the tuned melt with additional compensation of the defects or their destruction by appropriate doping, the relative fall of the monitor signal in the relevant spectral region is below 5% for a dose rate similar to the one expected at LHC for the CMS electromagnetic calorimeter.

The results of a full-size scintillation element study in the COCASE and H4 facilities with the same type of light readout (PMT XP1921) are shown on Fig. 9a, b. The element was produced from an ingot grown with tuned stoichiometry and doped with La with a concentration at half of the optimal value. The crystal was irradiated laterally uniformly along 18 cm by a ^{60}Co source for the COCASE test and longitudinally by a 120 GeV/c electron in the CERN H4 beam line with a similar dose rate of about 17 rad/h. In our study some crystals were also tested in the COCASE facilities with PMT and in the H4 beam with APD readout. These data are presented in Table 1. They indicate that at least to a first approximation the source irradiation data correlate with the particle signal change under the beam irradiation. The model of the transmission damage kinetics quoted above [3] allows to estimate the level of the induced absorption with a source which should correspond to a relative stability of the electron signal in a PWO calorimeter. A 5% electron peak position fall should correspond to an induced absorption less than 0.5 m^{-1} at 500 nm with an irradiation rate above 100 R/min.

Table 1

Crystal irradiation at the COCASE and H4 facilities for an accumulated dose of 200 rad

Scintillation element number*	1552	1557	1554	1474	1283	1564	1696	1694
Relative fall of the monitor signal at 500 nm in COCASE facilities, (%)	25	18	21	9	8	4	5	30
Relative electron peak position shift, (%) [17]	22	17	21	3	5	4	7	22

* Crystal numbers refer to the CMS database.

CONCLUSIONS

Data from the irradiation of doped crystals allowed us to specify the raw material needed to grow crystals exhibiting low damage under irradiation. The specification is complemented by the requirements for impurities having charge transfer and interconfiguration transitions.

The transmission damage of PWO scintillating crystals under irradiation is suppressed by the simultaneous use of a combination of crystal host structure control due to stoichiometric tuning and compensation or destruction of the remaining defects.

The damaged defects in PWO crystals are destroyed by Nb doping at a level of several tens of ppm.

The compensation of the damaged defects in crystals is achieved by trivalent ion compensation like La, Lu at the level of some tens of ppm.

Crystals which show an induced absorption of less than 0.5 m^{-1} at 500 nm at a dose rate of more than 100 R/min in source irradiation conditions will give an acceptable level of the loss of light signal induced by electrons in the irradiation conditions of the CMS electromagnetic calorimeter.

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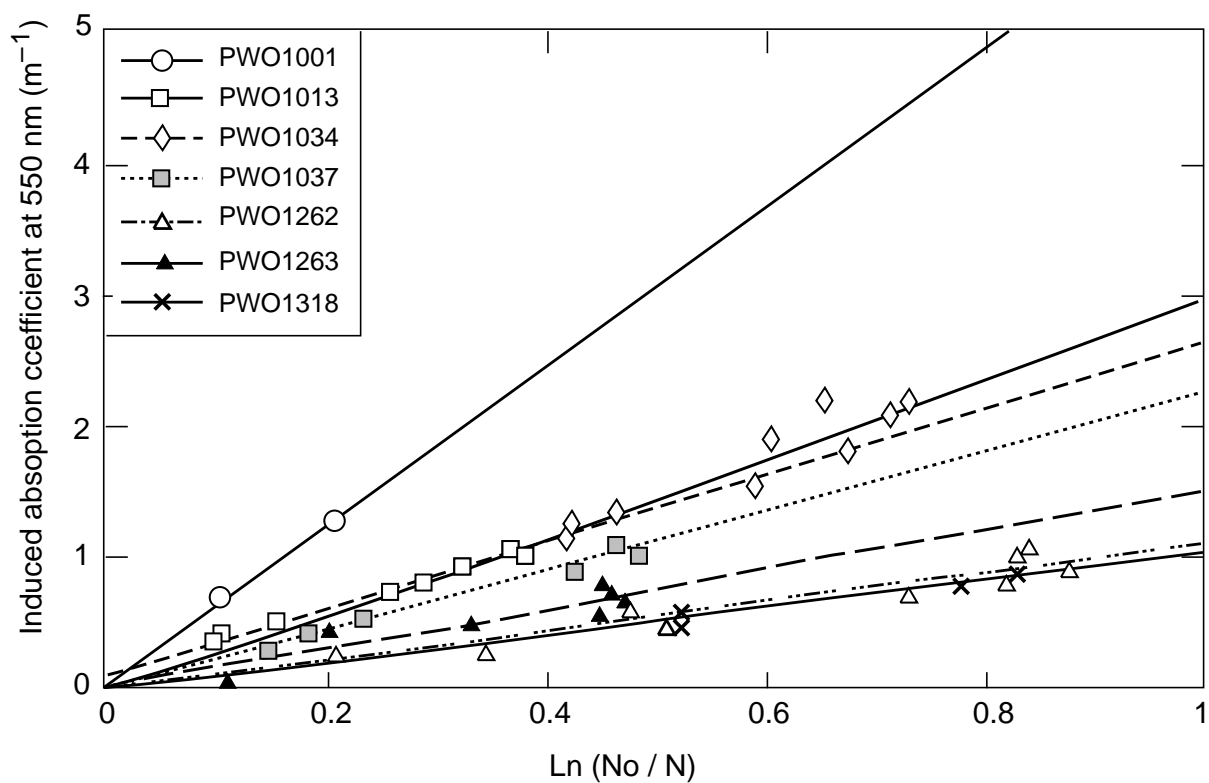


Fig. 1: Induced absorption coefficient at 550 nm versus Log (N_0/N). Crystals numbers refer to the CMS database.

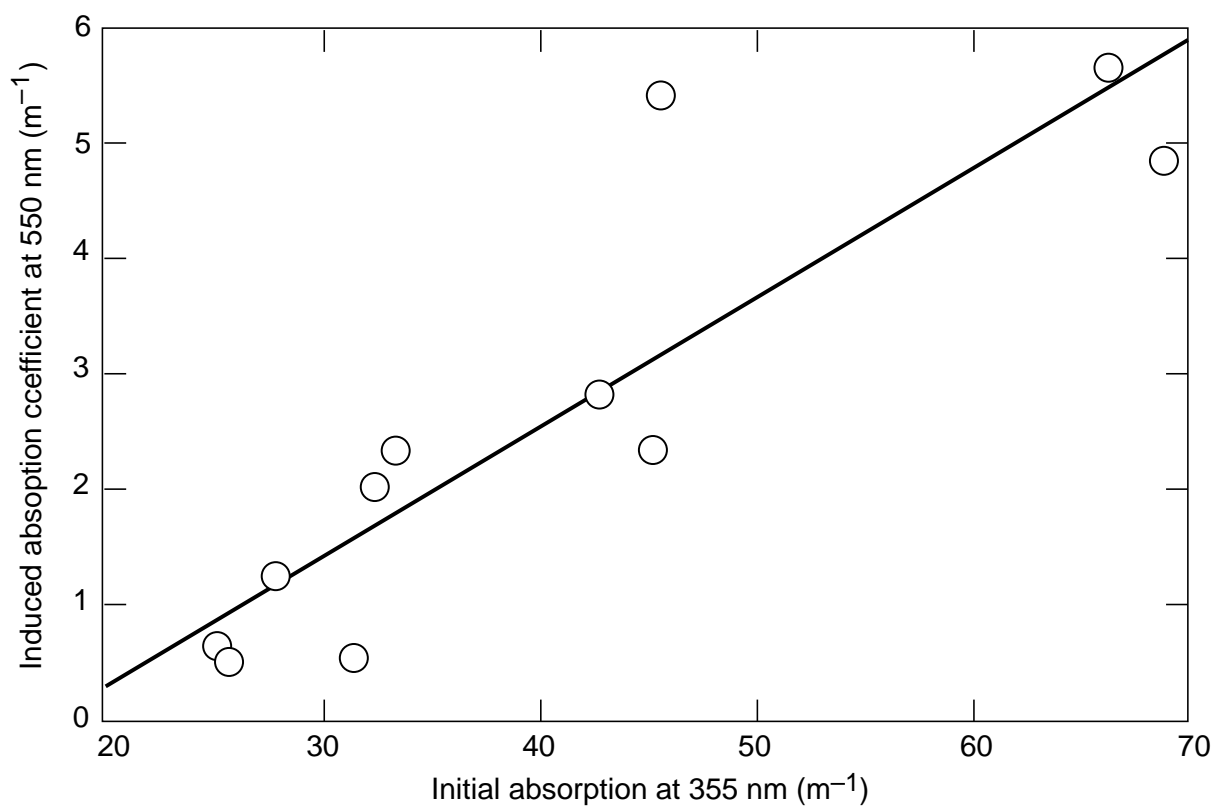


Fig. 2: Induced absorption coefficient of 550 nm versus initial absorption at 355 nm.

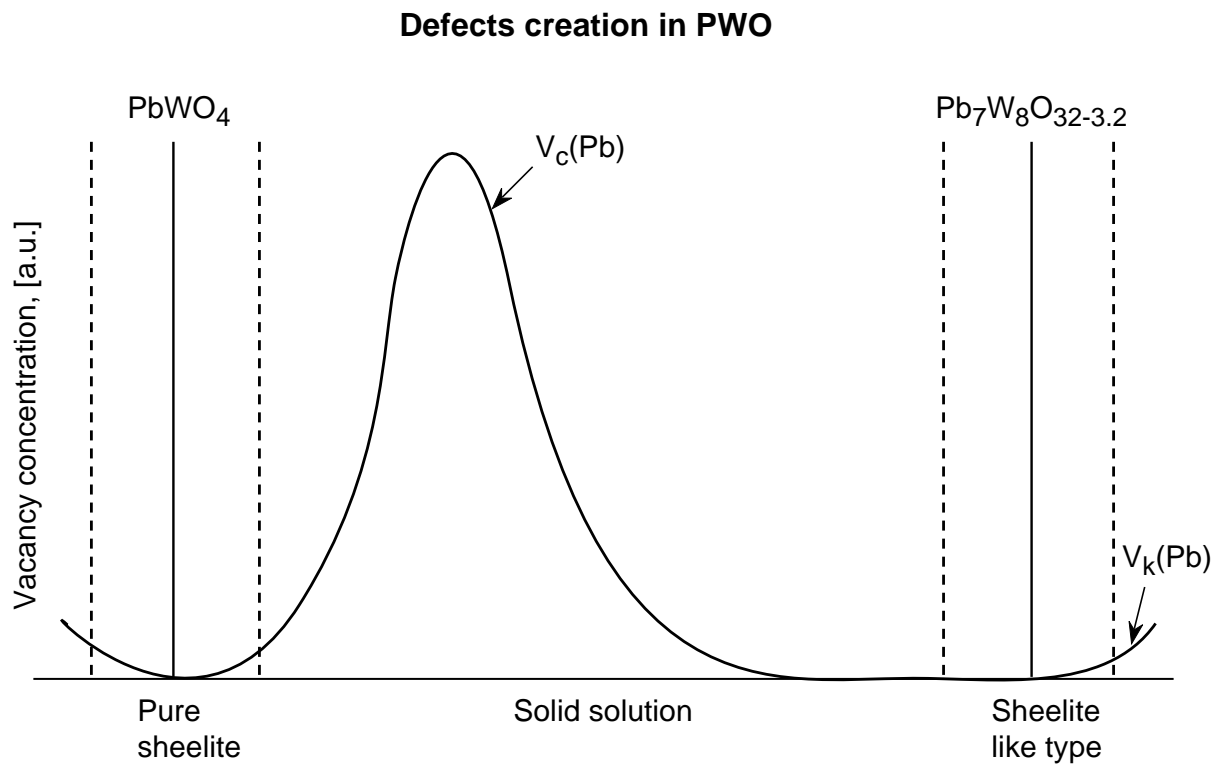


Fig. 3: Qualitative distribution of the cation vacancy V_c defects in PWO crystals with its structure.

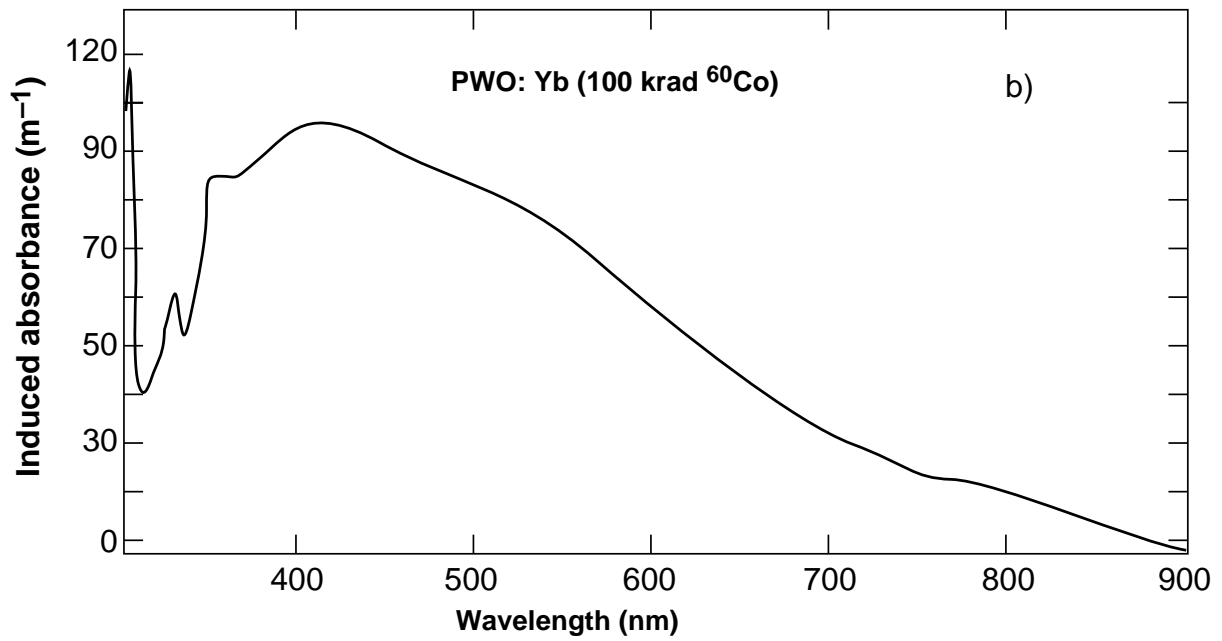
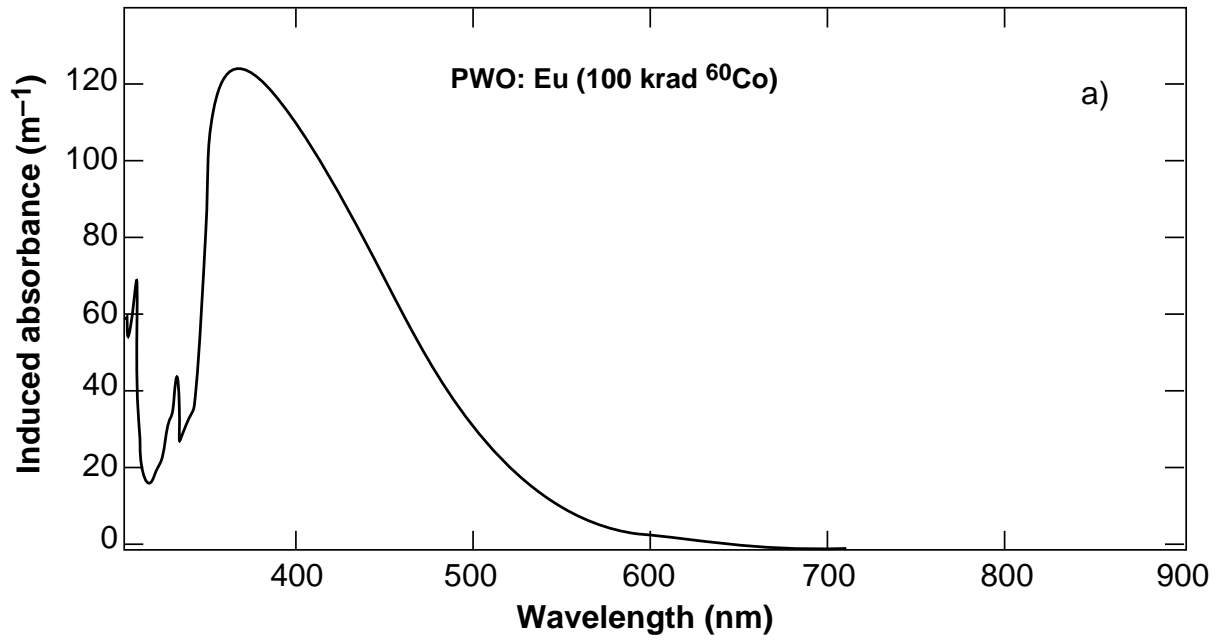


Fig. 4: Absorption of PWO:Eu a) and PWO:Yb b) crystals induced by irradiation. ^{60}Co source, 14000 R/min irradiation rate, accumulated dose 100 krad.

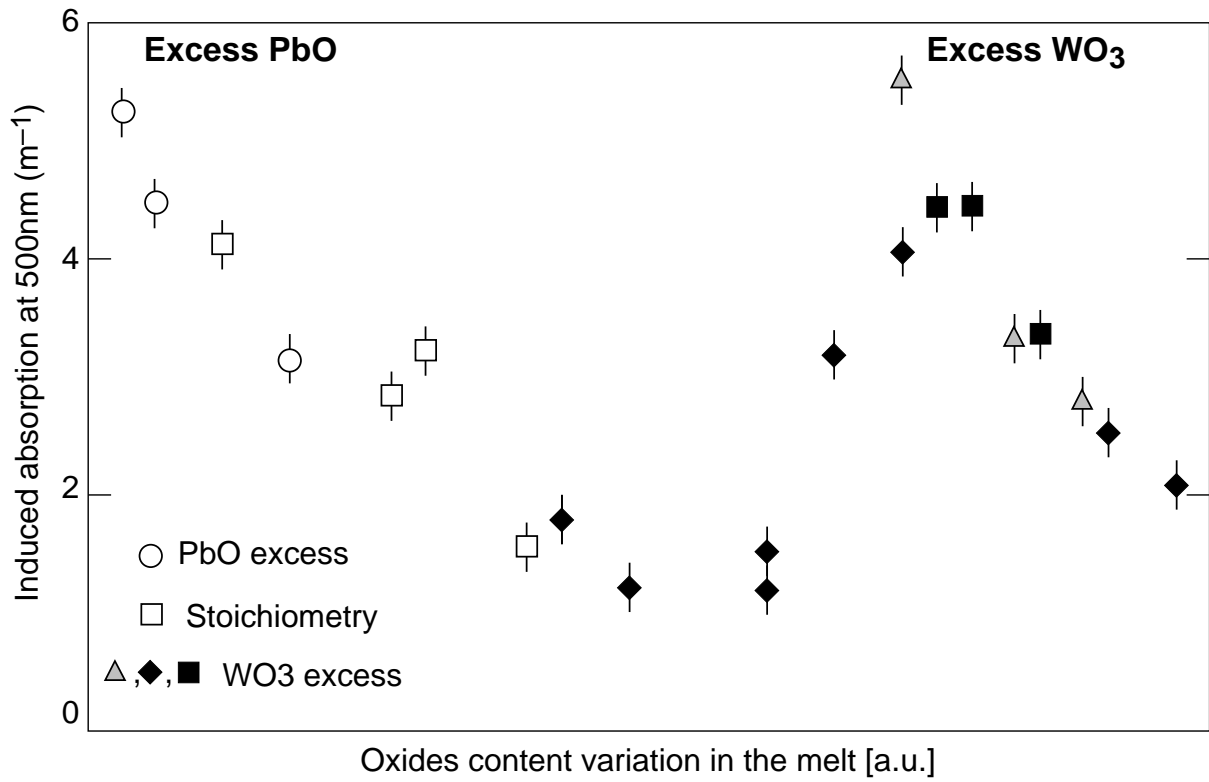


Fig. 5: Influence of the melt stoichiometry tuning on the induced absorption value at 500 nm. ⁶⁰Co source, 300 R/min irradiation rate, accumulated dose 50 krad.

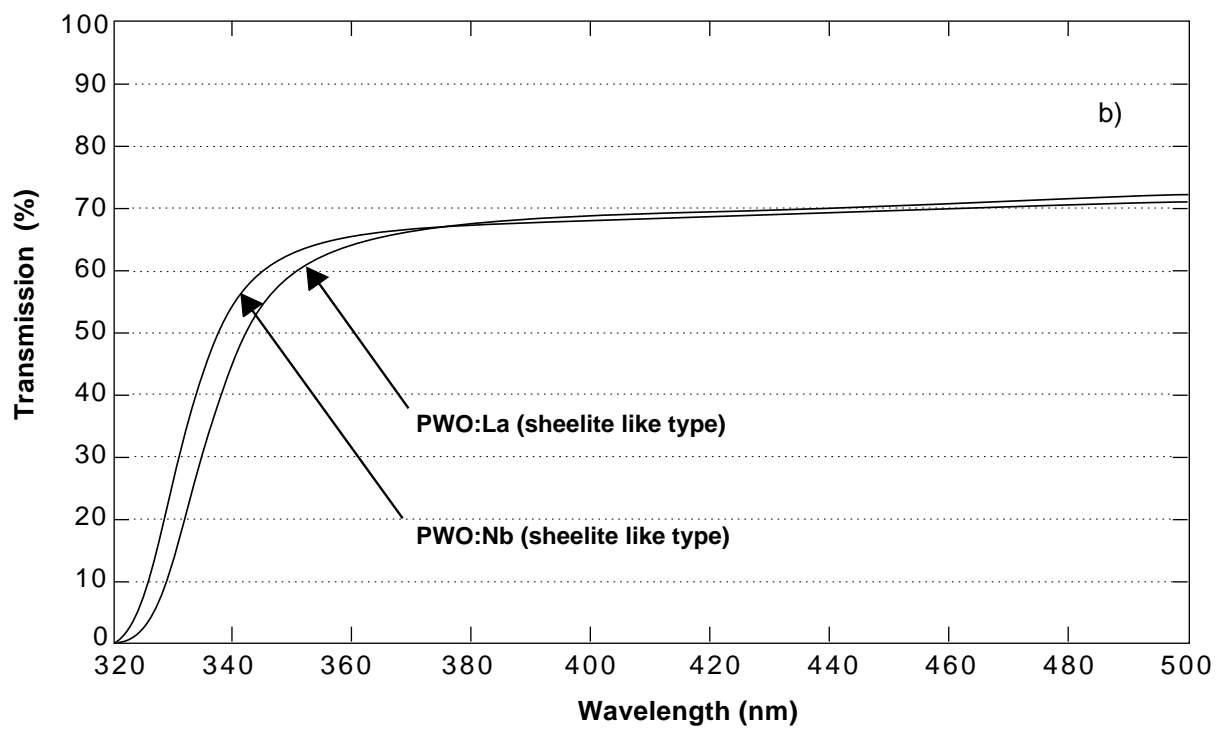
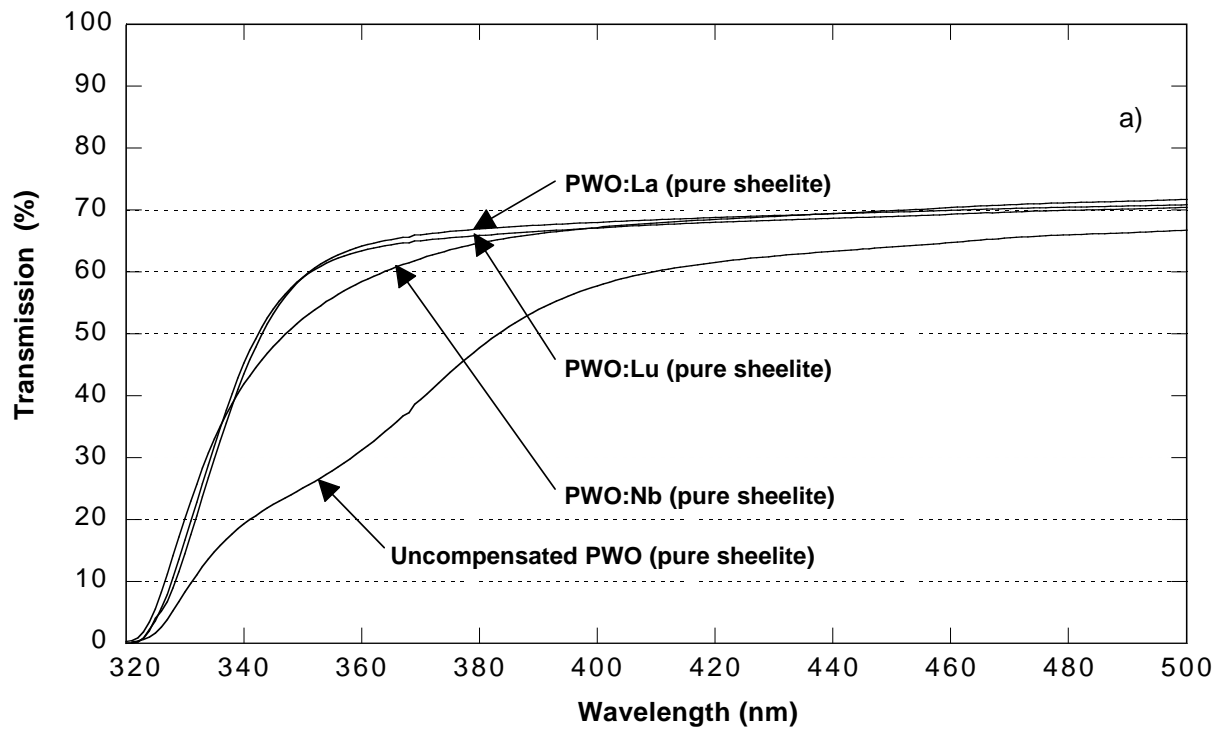


Fig. 6: Initial transmission of lead tungstate samples of 1 cm lengths of pure sheelite type a) and sheelite type b) doped by different activators. $T = 300$ K.

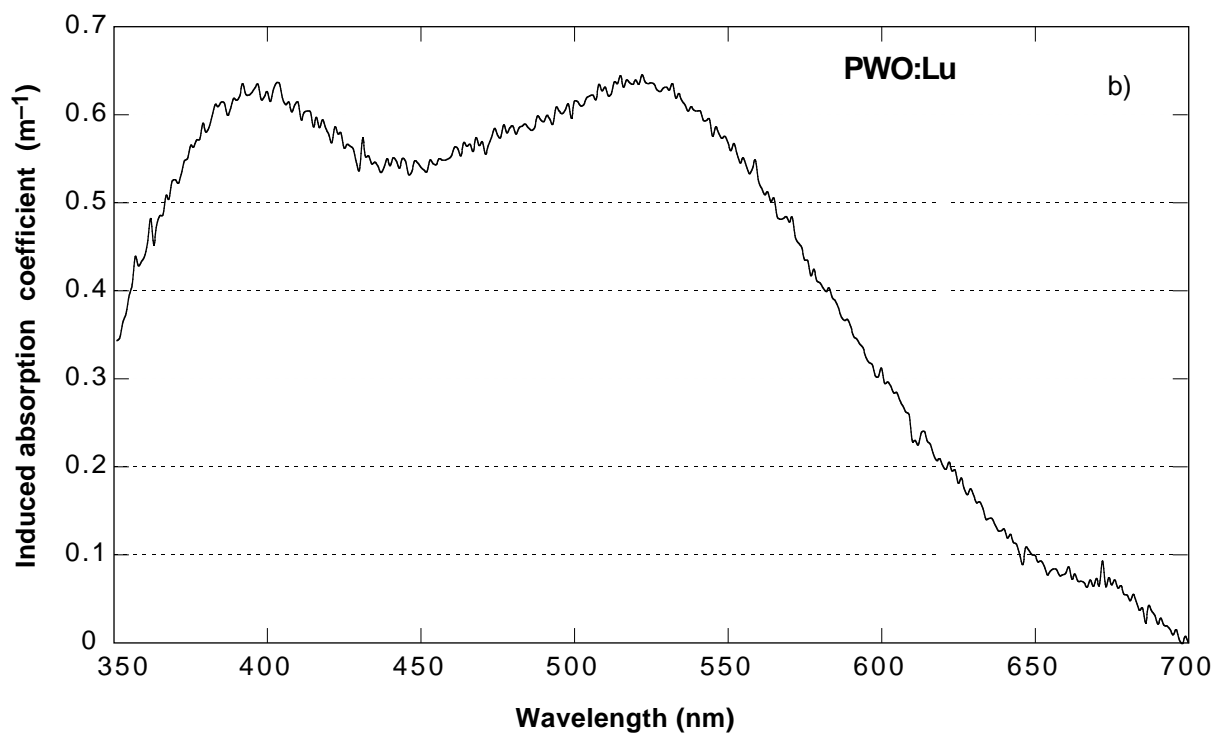
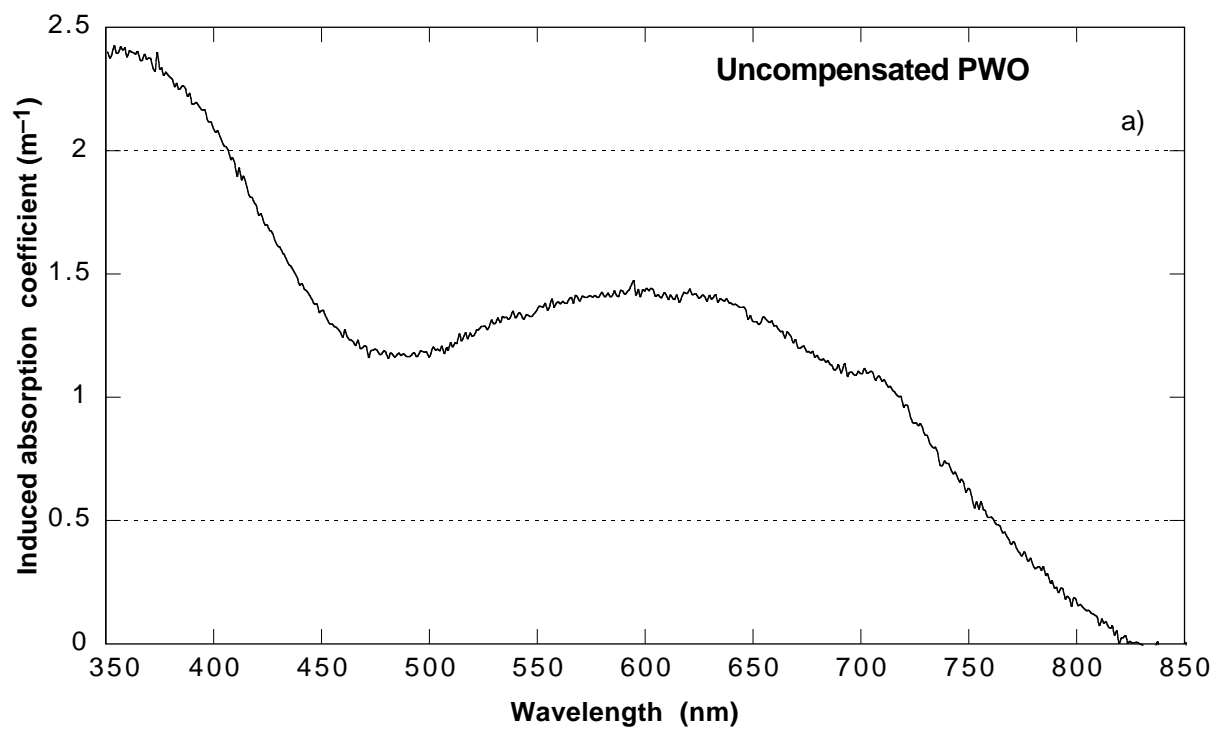


Fig. 7: Induced by irradiation absorption spectra in different PWO crystals:
 a) - grown in the conditions optimized by stoichiometry scan;
 b) - grown in the conditions optimized by stoichiometry scan and doped by Lu.

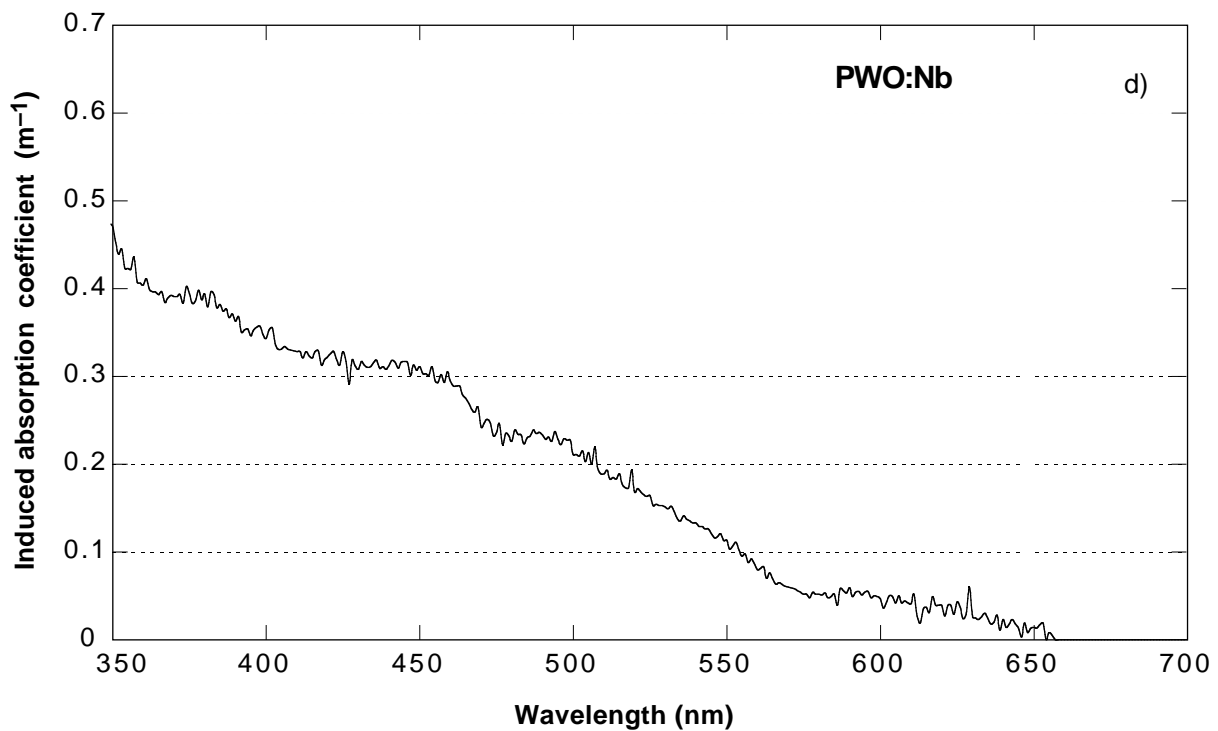
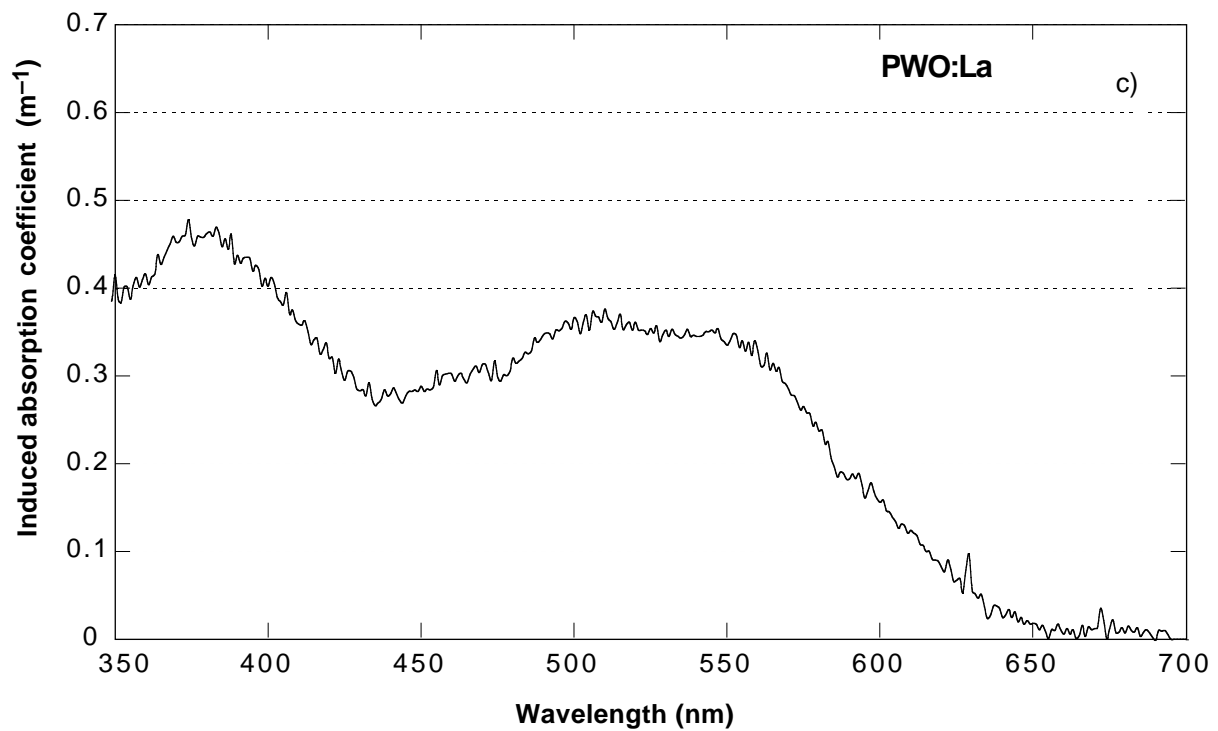


Fig. 7: Induced by irradiation absorption spectra in different PWO crystals:
 c) - grown in the conditions optimized by stoichiometry scan and doped by La;
 d) - grown in the conditions optimized by stoichiometry scan and doped by Nb. ^{60}Co source,
 14 000 R/min irradiation rate, 1000 Gy accumulated dose.

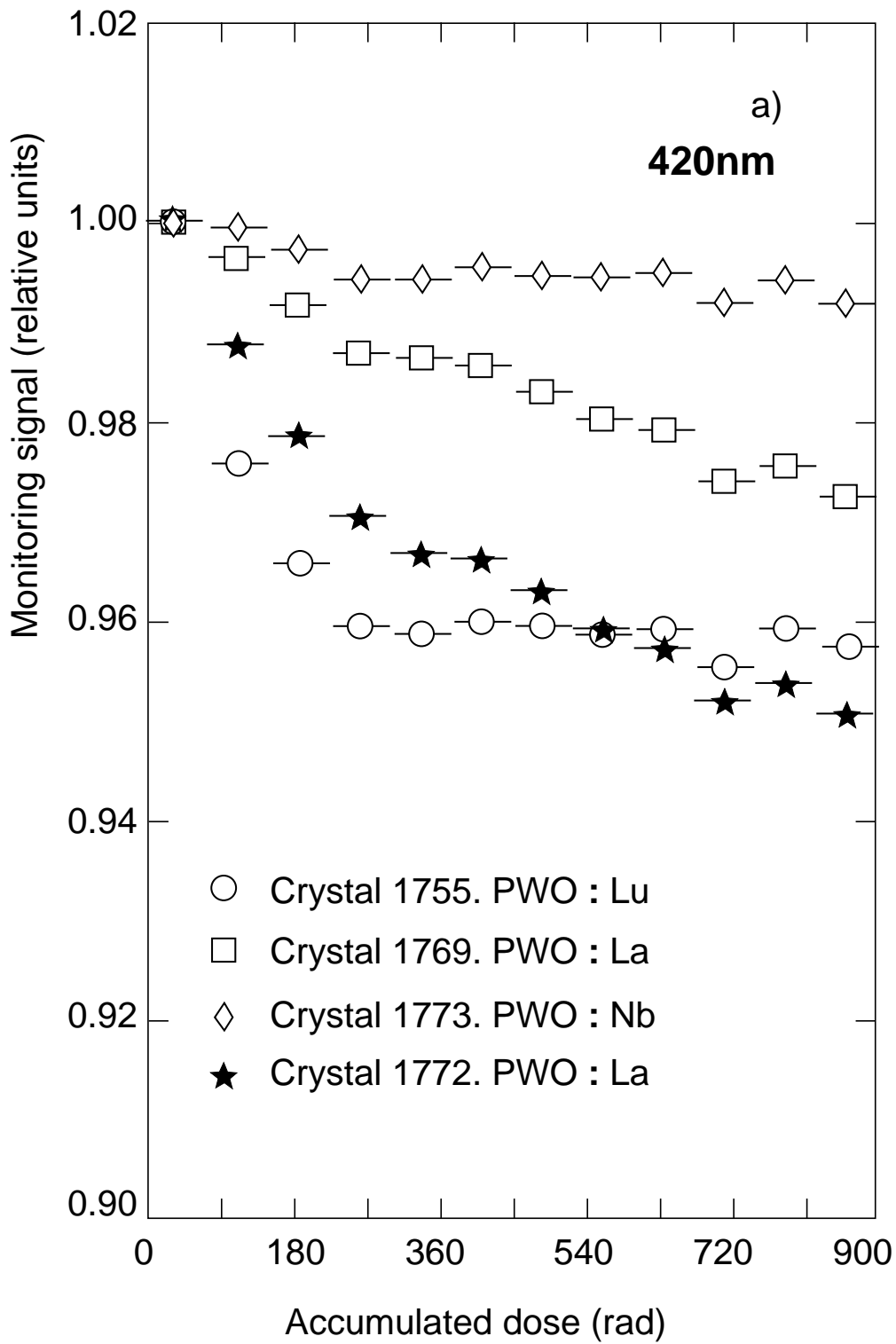


Fig. 8: Relative change of the monitoring signal at 420 nm a) obtained with crystals grown with stoichiometry tuning and additional doping by La, Lu and Nb. COCASE facilities, rate of dose accumulation 17 rad/h.

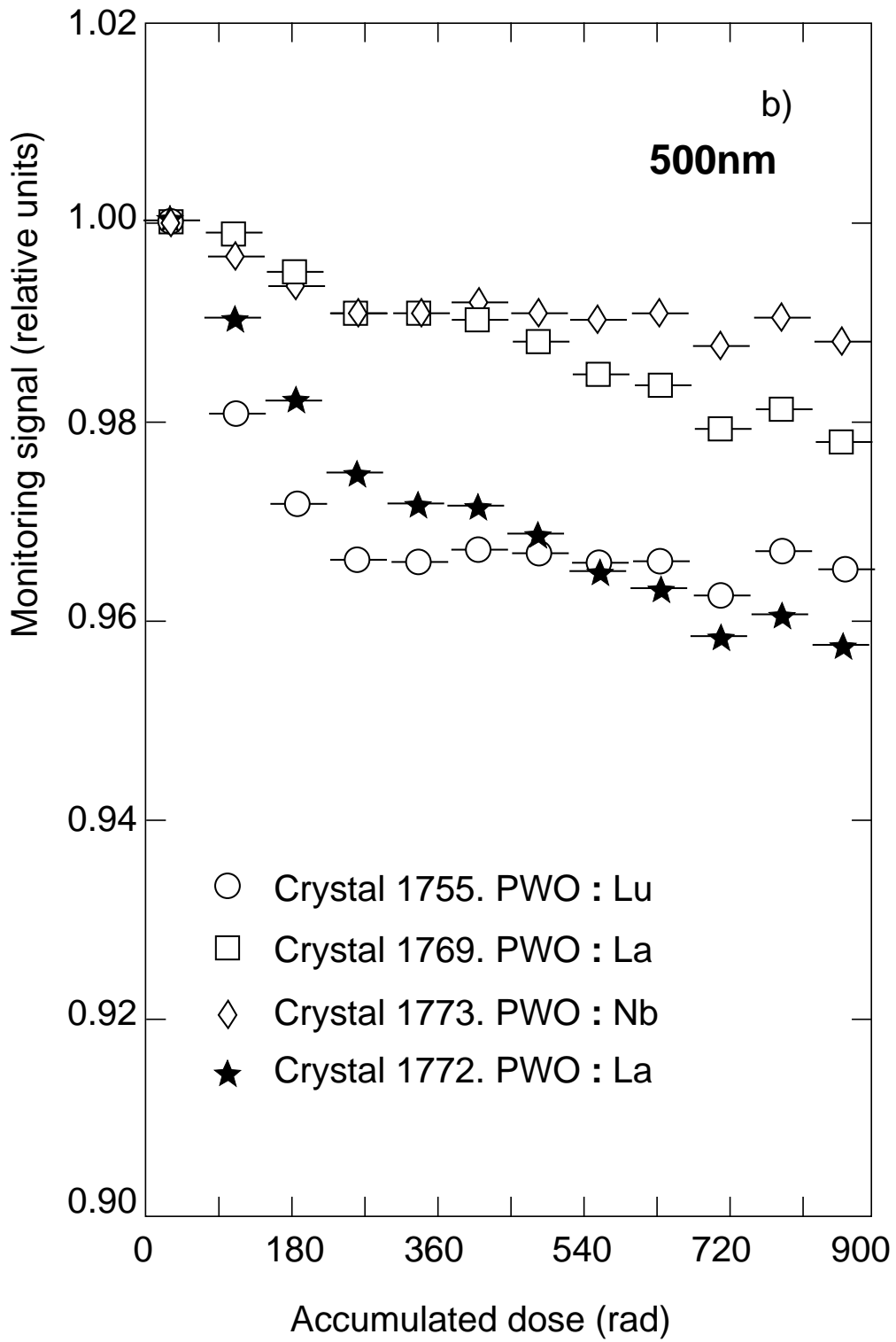


Fig. 8: Relative change of the monitoring signal at 500 nm b) obtained with crystals grown with stoichiometry tuning and additional doping by La, Lu and Nb. COCASE facilities, rate of dose accumulation 17 rad/h.

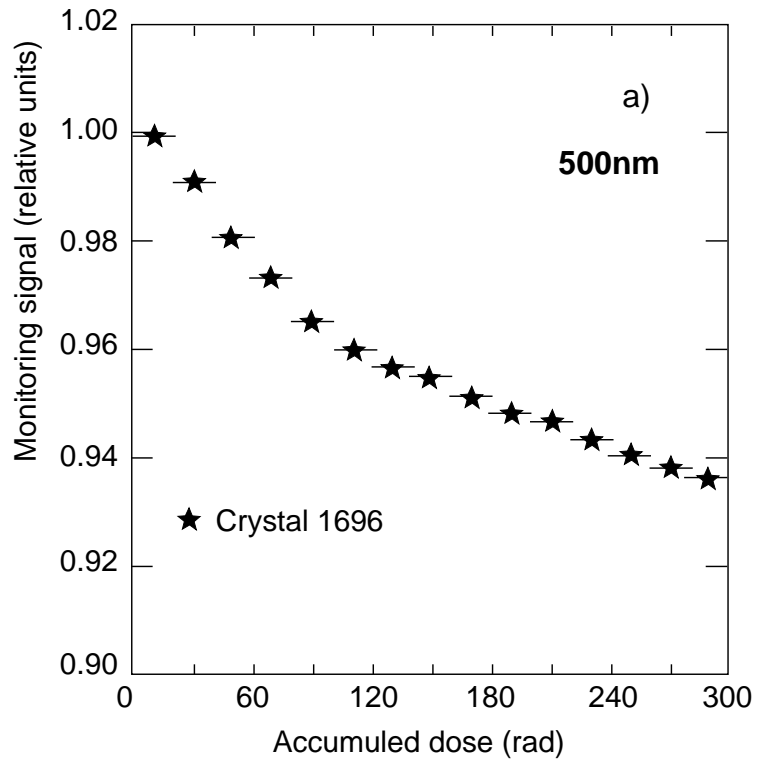


Fig. 9: The relative changes of monitor signal versus accumulated dose for the same crystal irradiated in the COCASE facilities.

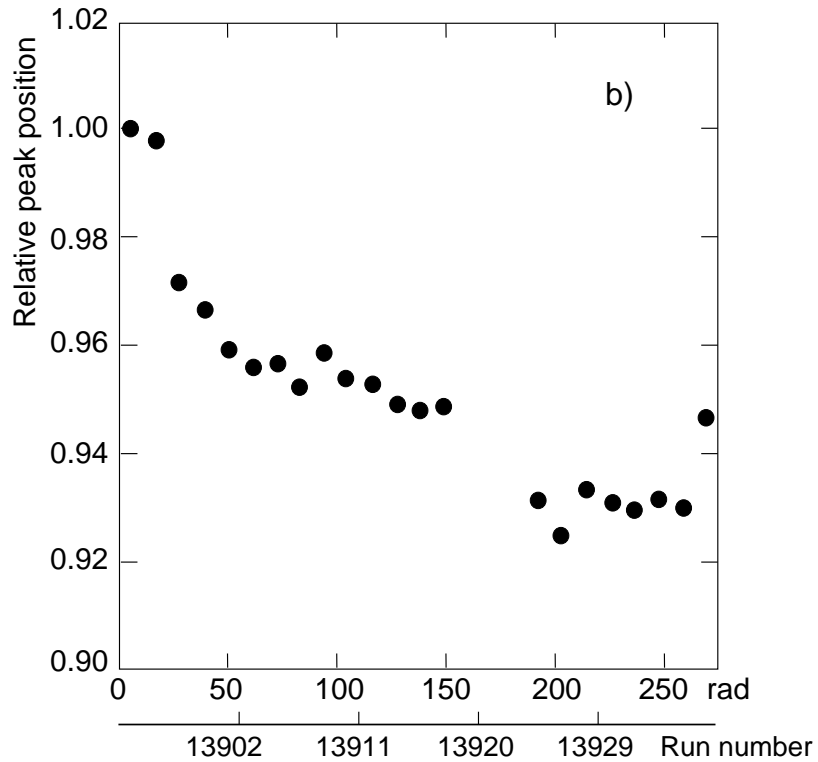


Fig. 9: The relative peak position of the electron signal of energy 120 GeV/c irradiated at H4. Corresponding run numbers are plotted at the bottom of the horizontal scale.