PRELIMINARY MEASUREMENT ON POTENTIAL LUMINESCENT COATING MATERIAL FOR THE ESS TARGET IMAGING SYSTEMS

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Abstract

We present in this paper the preliminary measurements performed on luminescent materials to be investigated and eventually coated on the ESS target wheel, the Proton Beam Window separating the end of the ESS Linac and the entrance of the ESS target area, and the ESS Dump. Among all the properties of the luminescent material required for the target imaging systems, luminescence yield and luminescent lifetime are essential for two reasons. The first one is trivial, since this material is the source for the imaging system and sets its potential performance. The lifetime is not generally of importance, unless the object is moving, or time dependence measurements are to be done. In our case, the target wheel is moving, and measurement of the beam density current may have to be performed at the 10μs scale. Thus luminescence lifetime of the coating material should be known and measured. In this paper, we present the luminescence measurements of the photo-luminescent lifetime of several materials currently under studies to be used eventually for the first beam on target.

INTRODUCTION

One of the challenges presented by high power proton beams for neutron spallation is the level of control required to prevent any damage on material of the target that must not be illuminated by the beam, or on the target itself not to be damaged. This level of control can be partly achieved by means of imaging the beam power density distribution deposited on the surface of the target. For such a system to be deployed at ESS, a luminescent material has to be identified and qualified. The qualification of such a material goes through reviews and tests of the candidate material properties, such as its luminescence yield, decay time, temperature sensitiveness, but also the thermo-mechanical resistance of the coated material to the beam impact and to high radiation dose, and to its luminescence properties under extremely high fluence, and finally, the qualified control of the industrial coated process. In this paper we address one of these aspect which is the measurement of the luminescence decay time. There are several ways to measure this decay time. Two possible ways are the time domain response, provided by a direct measurement of the luminescent pulse excited by a short proton bunch, and frequency domain response, from the measurement of the amplitude and phase luminescence excited by a frequency modulated source. The first method uses short bunches, and for instance short proton bunches delivered by facility like HiRadMat can be used for such a measurement. The second method is developed on a test bench for luminescent materials which can be photo-excited. The main objective of this test bench is to get time decay without the need of an accelerator of short bunched proton beams, and without time access restrictions. The method developed by Lakowicz *et al.* [1], consists in measuring the amplitude and phase response of the luminescent material under intensity modulated source and as function of the modulation frequency. The method is complementary to the time-domain lifetime measurement. It can provide a fine measurement of multi-exponential intensity decays but also non-exponential decays resulting from resonance energy transfer, time-dependent relaxation or collisional quenching. In the following, we firstly describe the experimental setup for the test bench for the frequency-domain luminescent lifetime measurement. Then we will present results from frequency-domain lifetime measurements on the chromium alumina coated for the SNS target imaging system. We will then discuss the method and the results obtained with it and within the perspective of using a similar luminescent coating material for the ESS target imaging system. Finally, we conclude on the performance and usage of the frequency-domain lifetime bench towards the selection process of the luminescent coated material for the ESS target imaging system.

EXPERIMENTAL SETUP

Frequency-Domain Lifetime Bench Setup

Our frequency domain lifetime measurement setup is shown in the Fig. 1. It consists of a modulated UV source (Thorlabs UV source DC3100-365), and two optical path, one towards the reference detector (Thorlabs PDA10), and the other towards the photo-luminescent sample. The luminescent light from the sample is detected by means of another fibre coupling assembly, which incorporates a filter for the source wavelength, and a second detector ((Thorlabs PDA10 or PDA36, depending on the bandwidth and sensitivity required). The signals from the two detectors together with the source modulation reference signal are sent to an oscilloscope, where the data is acquired. A detail description of the setup, the method and the analysis is explain in [1] and in [2].

Model for the Frequency-Domain Lifetime Measurement

As described in [2], the lifetime decay can be measured by fitting the amplitude and phase response of the lumines-

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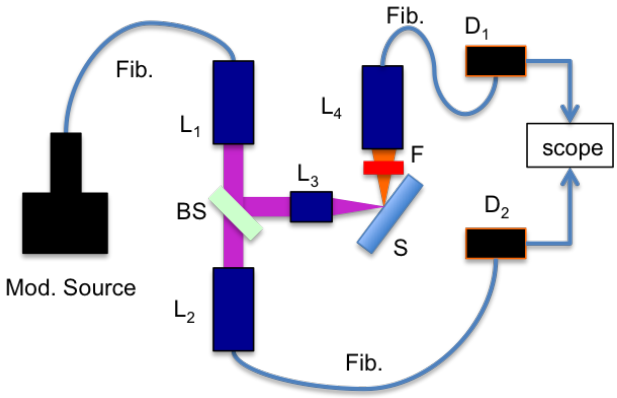

Figure 1: Frequency Domain lifetime measurement setup. The Modulated source is coupled to a fiber bundle (Fib.), and then collimated. The UV light is separated into two branch by a 90:10 beam spliter (BS), one coupling the 10% of the UV light intensity to another fiber connected to a selected detector for its sensitivity and bandwidth, and the other to focus the 90% UV onto the sample (S). The luminescent light is collected by a fiber coupling lens assembly that includes a filter F, which blocks the UV light. The fiber is connected to the second detector also selected according to bandwidth and sensitivity at the emitted luminescent wavelength. All signals are acquired in an oscilloscope using the electrical signal from the source as a reference trigger.

cence to the modulated excitation. The signal intensity decay modulation and phase can be predicted for any decay law by means of the following sine and cosine transforms of the intensity decay $I(t)$:

$$N_\omega = \frac{\int_0^\infty I(t)\sin(\omega t)}{I(t)} \quad (1)$$

$$D_\omega = \frac{\int_0^\infty I(t)\cos(\omega t)}{I(t)} \quad (2)$$

with ω the radial modulation frequency. For a signal composed of exponential decays, Eq. 1 and 2 can be written as:

$$N_\omega = \frac{\sum_i \frac{\alpha_i \omega \tau_i^2}{1+\omega^2\tau_i^2}}{\sum_i \alpha_i \tau_i} \quad (3)$$

$$D_\omega = \frac{\sum_i \frac{\alpha_i \tau_i}{1+\omega^2\tau_i^2}}{\sum_i \alpha_i \tau_i} \quad (4)$$

then the calculated phase and modulation amplitude of the signal response is given by:

$$\phi_{c\omega} = \arctan\left(\frac{N_\omega}{D_\omega}\right) \quad (5)$$

$$m_{c\omega} = \left(N_\omega^2 + D_\omega^2\right)^{1/2} \quad (6)$$

We have been implementing this model in Matlab, and built a fit function based on Marquadt Levenberg non-linear fitting algorithm. The validity of the model, i.e. the probability that the number of time decay values and their associated weights chosen for the fit is appreciated by means of a χ^2 calculation:

$$\chi^2 = \frac{1}{\nu}\sum \frac{|m - m_{c\omega}|^2}{\delta m^2} + \frac{|\phi - \phi_{c\omega}|^2}{\delta\phi^2} \quad (7)$$

with ν the number of free parameters, typically the number of measurement points less the number of fit parameters; δm and $\delta\phi$ are the experiment error bars on the measurement of the amplitude and phase response; m and ϕ the amplitude and phase response.

The validity of the model is given by a tabulation depending on ν. The result of the χ^2 has to be less than the values predicted using the Normal probability density distribution. In the other case, the result implies that random noise could also be responsible for the obtained measurement result[1].

RESULTS WITH THE FREQUENCY DOMAIN LIFETIME METHOD

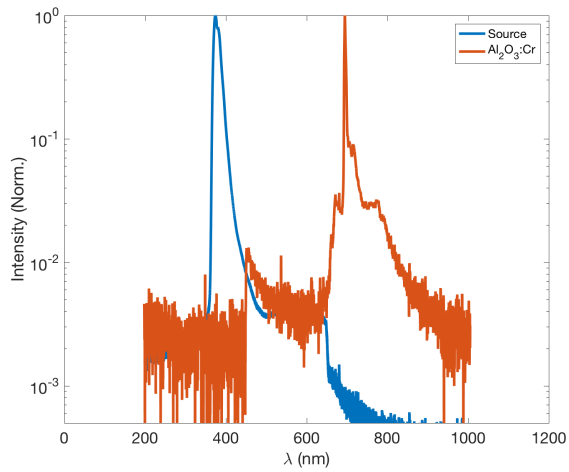

Figure 2: Spectra of the source at 365nm and the chromium alumina photo-excitation response, exhibiting the so-called R lines at around 693 nm and 695 nm

The first measurements we performed on the test bench is to acquire the spectrum of the source, and the photo-excitation spectrum, with and without the bandpass filter applied to suppress most of the light from the source. Figure 2 present these spectra. The spectrometre resolution is 0.7 nm r.m.s. So it doesn't resolve the R lines and the other lines that can be seen in the sides of the main peak. These lines will contribute all to the fluorescence lifetime. The frequency domain method is able in principle to retrieve the lifetime and weight of all these lines. However, in this case, the model should be extended to the spectral dimension.

[1] More detail and explanation can be found in the Encyclopedia of Mathematics online https://www.encyclopediaofmath.org/

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Then Eq. 3 and 4 contain also a wavelength dependency. We will not derive this, since our model integrates over all wavelengths from 450 nm to 1100 nm. Based on values from the literature [3] and also values extracted from measurements in the time domain performed at SNS [4, 5], the apparent lifetime of the chromium alumina is of the order of 1 ms. Taking this into account, we expect the decay of the modulation to happen in the range from DC to 10 kHz. Therefore we selected the range of frequencies for the measurement of our sample to be from 50 Hz to 10 kHz. The results of the measurement is presented in the Fig. 3. The top graph shows the amplitude modulation for a sine modulation of the source as function of the modulation frequency. The bottom graph is the phase of the signal less the phase of the source signal, with respect to the reference signal as picked up from the second branch of the setup (Fig. 1). In the graph, the results from the most probable fit, associated with the acceptable χ^2 test result and small residual from the fit is also shown. The trial of model started with a single time decay, and then we increased the number of parameters of the fit, and calculated for each of the results the corresponding χ^2 parameter. The results are shown in the table 1. The values of the decays found are mostly in the sub-ms and ms range. The χ^2 values are shown in the table, together with the residual of the fit. The minimum for these two quantities are for 5 and 3 decay times for the χ^2 and for the residual respectively. The smallest residual indicates the best fit, and the smallest χ^2 the largest deviation from the null-hypothesis, i.e. the most probable fit. In practice, increasing the number of fit parameters should result in a significant decrease of the χ^2 test, until it converges and start growing again, until it disqualifies the hypothesis of the initial free independent number of time decays. So from the last two lines of the Tab. 1, the most probable result is then between 3 and 4 independent time decays.

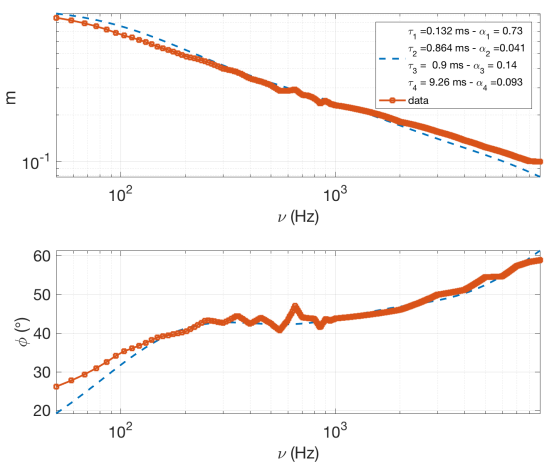

Figure 3: Luminescence frequency response for the Chromium Alumina coated on the SNS target.

Table 1: Results of the non-linear fit using models with 1,2, ...etc. , n_τ time decays. The decay time values τ_i are given in ms. The associated weights α_i are rounded and $\sum_i \alpha_i = 1$. The last two lines show the values of the χ^2 test (Eq. 7) and the residual values of the fit returned from the fitting function; Measurement on the data lead to $\delta m = 0.055$ and $\delta\phi = 0.2$. The total number of sample points is 49, thus $\nu = 49 - n_\tau$,.

#	1	2	3	4	5	6
τ_1	0.7	0.18	0.10	0.13	0.14	0.18
α_1	1	0.84	0.66	0.73	0.75	0.84
τ_2	-	5.97	0.65	0.86	1.11	3.26
α_2	-	0.16	0.23	0.04	0.10	0.035
τ_3	-	-	8.94	0.90	1.03	3.49
α_3	-	-	0.1	0.14	0.05	0.039
τ_4	-	-	-	9.26	6.73	3.98
α_4	-	-	-	0.09	0.04	0.036
τ_5	-	-	-	-	12.47	11.59
α_5	-	-	-	-	0.04	0.01
τ_6	-	-	-	-	-	14.1
α_6	-	-	-	-	-	0.037
χ^2	306.2	4.3	1.14	1.17	1.13	1.81
Res.	99.2	2.44	0.27	0.36	0.485	2.00

Time Decay and ESS Target Imaging

One of the Imaging system under design for the ESS will be delivering an image of each of the pulses sent to the Target Wheel (TW), with the requirements to determine the footprint of the proton power density distribution to be within the nominal area, the average power density to be uniform within 20% of the nominal average power density. The calculation to be performed on the image are relatively simple on a still image. However, the TW is moving at ≈ 7.8 mm/ms. With the luminescent lifetime decays measured, the image of a beamlet footprint will continue to emit over many ms, extended the beam footprint in the TW motion by many mm. This might render impossible the task to evaluate 99% of the power density footprint to be within ±4 accuracy from the nominal position of the beam on target. For still targets, like for instance the Tuning Dump and the Proton Beam Window (PBW), beamlet images, measurement of the power density footprint position will be possible. More advanced diagnostics though, at a fraction of the 2.86 ms pulse, might not be possible with this luminescent material.

CONCLUDING REMARKS

A bench test for the measurement of the lifetime decay of the luminescent material to be coated on the ESS target and Dump have been deployed. The setup is operational and luminescent material in-use at SNS as been measured as a potential candidate material for ESS Target Imaging Systems. Further improvement of the setup might follow.

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For instance the selection of the detectors to allow a larger bandwidth extending in the MHz range will be necessary for the measurement of faster decays of new materials yet to be tested. Also the detection with an oscilloscope might be improved by using dedicated acquisition electronics. This should improve signal to noise and phase noise, although in this measurement, the noise figure mostly come from the detectors itself.

The bench is part of a series of qualification tests derived in order to down-select pre-selected luminescent materials for their proton luminescent yield, their yield at 200 degrees, their lifetime under high radiation dose, and high displacement per atom, their narrow-band spectrum and for their luminescence lifetime decay. The sample material measured has been passing most of these selection criteria, but doesn't satisfy entirely the last criteria. However, this material constitute so far the best known luminescent material for high power target beams. The search for a similar material, and the studies to understand its qualifying properties for the usage in high power target beams continues in order to provide the required luminescent material for the ESS.

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