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Application of low energy electron beam excitation of dense gases and liquids for astro-particle physics

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A table-top setup for characterizing gaseous and liquid scintillating material is described. The key part of the instrumentation is a 300 nm thin ceramic membrane made from silicon nitride and silicon oxide. Low energy electrons with 12 keV particle energy are sent through this membrane into the material to be tested. Both fully continuous and pulsed beams can be used. Time resolved optical spectroscopy is used as the diagnostic method. The fluorescence of nitrogen and solutions of liquid scintillation material are described as examples for applying the technique.

Key words: particle detectors, electron beam, optical spectroscopy.

Introduction

Detection and characterization of rare events in which highly energetic or weakly interacting particles arrive on earth and interact with a detector material is an important subject of experimental particle-astrophysics. Because of the intrinsically low count rate it is important to study the properties of the detector material in detail before large and expensive detectors containing large quantities of the substance are installed and put into operation. Essentially all online particle detection schemes finally rely on charged particles. Both primary and secondary reaction products interacting with the detector material may lead to ionization in the detector. The charge is collected and registered using electric fields. The second important processes are inelastic collisions of charged particles in which they excite atoms or molecules which subsequently emit photons which can then be registered as fluorescent light.

Radioactive sources are routinely used for studying the performance of particle detectors. The response time of the detector can be precisely measured with this technique. The activity of sources necessary for recording the spectral emission characteristic of scintillation detectors, however, is normally above values which are allowed to be handled in regular laboratories. A way around this problem is, of course, to use particle beams for exciting the detector material. In that case the light intensity can be controlled by the beam current. This, however, requires the use of large facilities with limited access. Other ways for obtaining higher excitation densities are the production of charged particles via photo effect (X-ray excitation) or direct excitation of the scintillating material using ultraviolet photons. In these cases, however, it is questionable whether the emission spectrum is the same as for actual particle beam excitation.

Here we describe a technique in which a low energy electron beam is used to induce fluorescent light in gases or liquids. The main advantage is that it is available as a table-top setup. Due to a particle energy of typically 12 keV it produces no hard X-rays. The energy is however high enough to simulate high energy particle beam excitation in a realistic way since the cross sections for producing fluorescent light are maximum for electrons of typically only tens of electron volts.

1. Experimental technique

The experimental setup is schematically shown in Fig. 1.

It consists of one part with a high vacuum and as a second part a cell filled with the material to be

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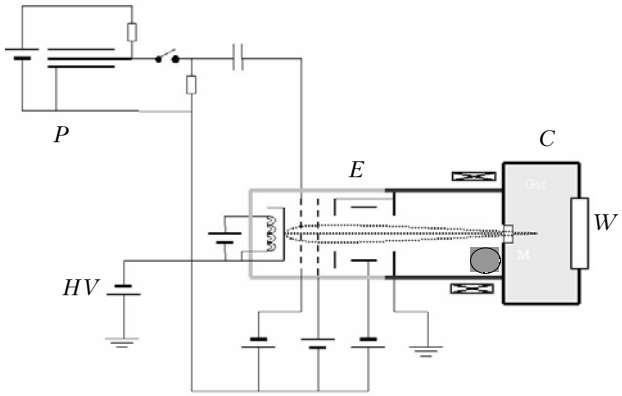

Fig. 1. Material (gas or liquid) to be tested is placed into the target cell *C*. There it is irradiated with an electron beam produced and accelerated in the electron tube *E*. A high voltage (*HV*) on the order of 12 kV is used for particle acceleration. Various grids and a pair of dipole magnets are used to control the electron beam with respect to current, focus, and direction. As an option, a pulsing unit (*P*), e.g. in the form of the cable pulser schematically shown here, can be added

tested, either a gas or a liquid. An electron beam is formed and accelerated in the vacuum part and sent into the target cell through a very thin membrane. This membrane is typically 300 nm thick and made from ceramic material. So far silicon nitride membranes are used which are available from solid state industry. The internal stress of Si_3N_4 has to be reduced for obtaining stable membranes. This is either achieved by changing the stoichiometry of silicon nitride or by forming a double layer of silicon-nitride and oxide. The latter technique is routinely used in our experiments. The free standing membranes are typically 0.7×0.7 mm^2 in size, manufactured on a 5×5 mm^2 section of $a \approx 0.5$ mm thick silicon wafer. For laboratory applications this wafer section is glued onto a flange separating the vacuum and medium- filled parts of the setup.

Regular electron guns which are normally used in cathode ray tubes ("CRT tubes", for monochrome TV or monitor screens) can be used for producing the electron beam. We normally limit the acceleration voltage to 12 keV to avoid unwanted production of harder X-rays. For these electrons only about 15% of the electron beam power is deposited in the membranes described above. The rest is used for excitation of the target material. Maximum electron beam currents of about 10 μA (continuous or time averaged) can be used. This corresponds to a beam power of 0.1 W deposited in the material under study. The time structure of the beams which can be produced with CRT tubes ranges from 5 ns to fully continuous. The technique and details concerning the energy loss in the membranes and the power deposition in the target material have been carefully studied and described in previous publications [1–8].

Preparation of the scintillating material depends on the type of material under study. Gases are filled into a closed cell which is part of a vacuum and gas filling system. Purification methods similar to the ones which are used for actual particle detector systems must be included in the setup. Liquids can either be simply put on top of the membrane as a droplet as described below or put into an appropriate container similar to the one shown in Fig. 1. Since the range of the low energy electrons is only on the order of 1 mm in gases at atmospheric pressure and 1 μm in liquids, respectively, the optical absorption in the material has to be carefully considered. Also the optical window of the cell through which the scintillation light is coupled into the diagnostic part of the setup and other optical elements in the setup such as mirrors, lenses, optical fibers, and diffraction gratings etc. have to be selected with respect to the spectral range which has to be studied.

Two different spectrometers were used as the diagnostic tools for the experiments described below. A compact spectrometer (OCEAN OPTICS HR2000) was used for the characterization of liquid scintillator material excited by continuous electron beams. The light was guided to the monochromator with a 600 μm thick fiber optic cable. Light was coupled into the fiber with a small quartz lens. The resolution of the spectrometer was approximately 0.5 nm.

The emission from nitrogen and air used as the fluorescent material in some of the cosmic ray detectors, such as the Pierre Auger detector, was studied using a $f = 30$ cm criss cross Cerny–Turner vacuum monochromator (McPherson 218). This monochromator has a resolution of 0.03 nm for a 10 μm setting of the entrance and exit slits. A phototube with a MgF_2 entrance window and a multialkali cathode (S20) operated in the counting mode was used as the detector. This detection system covers a wavelength range from 110 nm, the cut off of the MgF_2 optics, up to approximately 700 nm. It can be used in a scanning mode for recording wavelength spectra. Time spectra are recorded by selecting a fixed wavelength with the monochromator, operating the electron gun in a pulsed mode, and recording the photomultiplier counts with a time- to amplitude-converter (TAC). So far, a minimum width of 5 ns for the electron beam pulses has been reached [7].

2. Emission from nitrogen and air

There is presently significant interest in the details of light emission from nitrogen and atmospheric air since the atmosphere is used as the scintillating material for recording so called extensive air showers. These are showers of secondary particles which are initiated by primary cosmic ray particles, some of which have energies of more than 10^{20} eV [9]. The majority of the secondary particles are electrons and positrons with energies covering a wide range. The fluorescence of the atmosphere in dark, moonless nights is used for detecting air showers in addition to ground based Cherenkov detectors. The fluorescence of air is predominately light emitted from a molecular transition (B to C electronic states) in nitrogen. Light emitted from air showers is collected with large mirrors behind UV filters since the C–B light is emitted

mainly in the 300 to 400 nm spectral range. Various experiments have been performed for studying the electron beam induced nitrogen emission [10]. With the table top setup described above the fluorescence induced by very low energy electrons can be studied down to excitation energy threshold. This is a rather relevant energy range since the excitation cross sections are large just above threshold [11].

Low energy electron beam induced fluorescence of nitrogen has been studied with respect to various aspects. Lifetime measurements and water vapor quenching of the C—B transitions is described in ref. [7]. The influence of pressure on the population of the lowest vibrational levels of the C state in nitrogen is described in ref. [4]. To demonstrate the quality of time resolved data which can be obtained with the table-top setup described here, where excellent counting statistics can be achieved due to an essentially unlimited counting time, the time dependent light emission of the 337 nm C ($v' = 0$) to B ($v'' = 0$) transition is shown in Fig. 2. The temperature dependence of the decay rate of the nitrogen C—B transition has been studied from about 200 to 300 K to simulate the temperature variation which is experienced by the extensive air showers in the atmosphere [12].

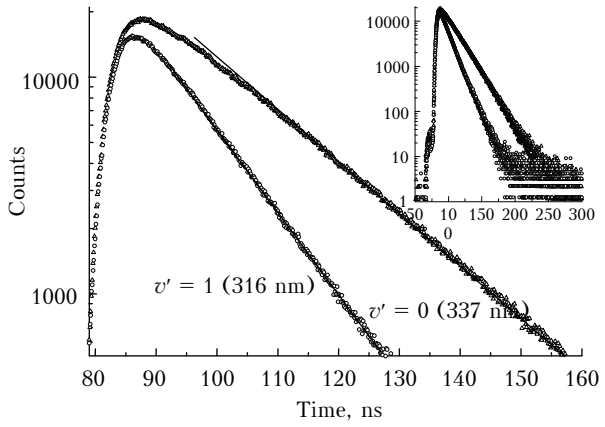

Fig. 2. The time dependence of the onset of light emission following excitation of nitrogen with a 5 ns electron beam pulse is shown for the two molecular transitions at 316 and 337 nm, respectively. These emissions originate from the $v' = 1$ and $v' = 0$ upper vibrational levels of the electronic C and B states of the nitrogen molecule. The lines represent exponential fits to the data starting at times when the time structures of the emissions can be represented by single exponential decays (see ref. [4] for details). The inset shows the full dynamic range of the data. Note that the time spectrum covers almost 4 decades in intensity

As a recent result an emission spectrum from ambient laboratory air is shown in Fig. 3. It was recorded with 12 keV, 5 μA fully continuous electron beam excitation. A gas flow was produced with a fan in front of the membrane where the electrons entered the gas to avoid modification of the target gas due to electron beam induced chemical reactions. The light emitted from the excited volume was imaged onto the entrance slit of the monochromator by an elliptical mirror coated with an Al—MgF_2 reflective layer. The point-like source

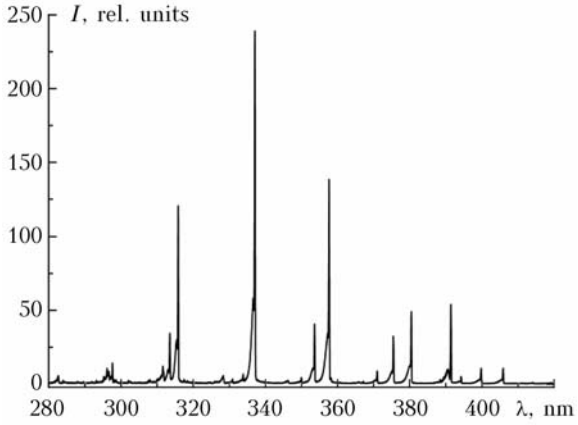

Fig. 3. Overview spectrum of the emission from electron beam excited ambient air. The spectral region shown is relevant for the detection of so called "extensive air showers" induced by high energy cosmic ray particles

in combination with a high quality mirror optics allowed a good signal to noise ratio for the spectrum although the efficiency for light production is low for ambient air mainly because of quenching by oxygen. We have measured the number of photons emitted per energy deposited by the electron beam (fluorescence yield) in a separate experiment. A value of (90.4 ± 6) photon/MeV has been measured for the 337 nm ($v' = 0$ to $v'' = 0$) transition of the C—B band in pure nitrogen at 1 bar. Details of this measurement will be published elsewhere. An expanded view of the spectrum shown in Fig. 3 is shown in Fig. 4. It covers the region of the B to X transition of N_2^+. The rotational structure becomes visible for this transition with the resolution of 0.15 nm determined by the slit width of 50 μm used for recording these data.

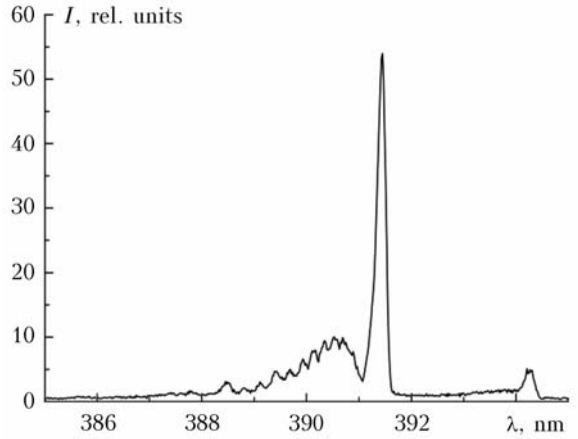

Fig. 4. An expanded region from 385 to 395 nm of the spectrum of ambient air (Fig. 3) is shown. The molecular band is in this case the B-X ($v' = 0$ to $v'' = 0$) transition in N_2^+. The nominal resolution of the detection system used is 0.15 nm given by the slit width of 50 μm

An analysis of the spectrum which can be compared with intensity ratios for the various vibrational transitions reported in the literature [13] is given in Table 1.

Table 1

Relative intensities of the nitrogen emission bands are listed as taken from ref. [13] (I_1, air at 800 mbar) and derived from integrating the intensities shown in Fig. 3 I_2. A tungsten lamp was used for calibrating the setup in ref. [13] and a deuterium lamp for the data in Fig. 3. The strong and systematic deviations listed in the last column show that a more careful study on this issue is needed

Transition	λ, nm	I_1, %	I_2, %	Deviation, %
2*P*(1,0)	315.9	39.3	49.1	20.8
2*P*(0,0)	337.1	100	100	0
2*P*(1,2)	353.7	21.35	16.0	–33.8
2*P*(0,1)	357.7	67.4	54.0	–24.8
2*P*(1,3)	375.6	17.87	12.8	–39.6
2*P*(0,2)	380.5	27.2	20.5	–32.5
1*N*(0,0)	391.4	28.0	21.3	–31.3
2*P*(2,5)	394.3	3.36	2.1	–57.3
2*P*(1,4)	399.8	8.38	4.9	–71.4
2*P*(0,3)	405.0	8.07	5.0	–60.2

It lists the intensities of the vibrational transitions normalized to the strong $v' = 0$ to $v'' = 0$ transition at about 337 nm. Note, that the relative spectral response of the monochromator-detector system had been calibrated using a deuterium lamp and the spectrum in Fig. 3 represents the intensity distribution corrected with respect to this sensitivity calibration. Reflection of the UV light on the silicon nitride membrane has been measured and included in the calibration procedure. The rather large discrepancies found between the data published in ref. [13] and in this paper suggest that it will be necessary to perform a careful study of the relative intensities of the particle beam induced light emission from air. Note, that the sensitivity calibration of optical detection systems in the wavelength range between about 300 and 400 nm is particularly difficult since both the intensity of deuterium and tungsten lamps with known emission spectra are weak in this region. The variation of intensity with wavelength is opposite for these two types of calibration lamps: decreasing with increasing wavelength for the deuterium lamp and increasing for the tungsten lamp. This allows a consistency check which still needs to be performed for the setup described here.

3. Emission from liquid scintillators

Testing liquid scintillation material with particle beams faces the same problem as testing gases. A window has to be used for separating the liquid from the vacuum in which the particles are accelerated. Here we present the results from first test experiments with liquid scintillators using the setup with the thin silicon nitride/oxide windows described above. In the context of a study of the liquid scintillator material for the proposed LENA (Low Energy Neutrino Astronomy) detector [14, 15] a set of spectroscopic test experiments have been performed. The goal was to verify that extensive studies of the emission spectra using ultraviolet light from a deuterium lamp for excitation are consistent with spectra obtained with particle excitation.

A schematic drawing of the experimental setup is shown in Fig. 5. The surface tension of the material under study allowed us to just put the liquid on top of the membrane through which the electrons exit from the vacuum. A thin layer of the scintillating liquid is excited by the electron beam producing a visible glow. Part of this light was collected with a quartz lens into a 600 μm diameter quartz fiber and guided to the spectrograph as described above. Fully dc beams were used in these experiments.

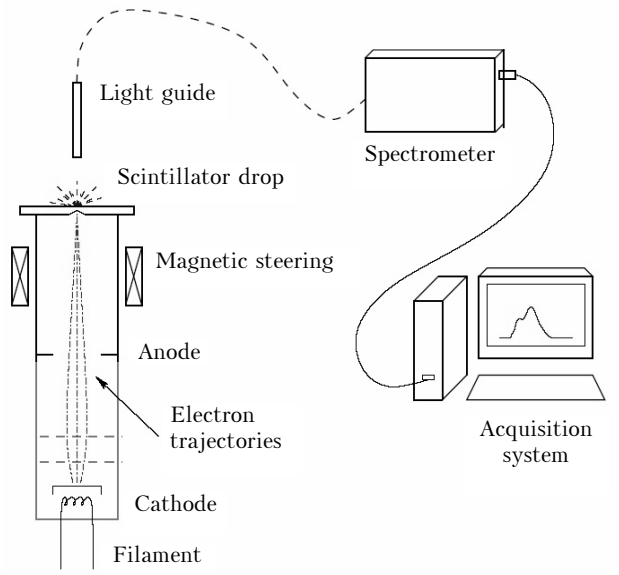

Fig. 5. Schematic drawing of the setup for recording spectra emitted from an electron beam excited liquid scintillator material

The scintillation liquids studied here were mixtures of the solvents PXE (phenyl-o-xylylethan, $C_{16}H_{18}$) and LAB (linear-alkyl-benzene, $C_9H_{12} + (CH_2)_m$) and the fluorescent materials PPO (2,5-diphenyl-oxazole, $C_{15}H_{11}NO$), bisMSB (1,4-bis-(o-methylstyryl, $C_{24}H_{22}$), and PMP (1-phenyl-3-mesityl, $C_{18}H_{20}N_2$). These are mixtures which are relevant for many future neutrino detectors. Typical admixtures of the fluorescent material to the solvents range from 20 mg/l to 2 g/l. Examples of the emission spectra are shown in Fig. 6 in comparison with spectra which were recorded with UV excitation.

For recording these comparative spectra the UV light was applied to the samples from one side and the fluorescence light was recorded from the same side. Note, that the light is produced in a very thin layer next to the entrance foil and has to traverse the liquid drop before it enters the fiber optics of the spectrometer in the case of electron beam excitation. The differences which appear in the spectra at short wavelengths below 400 and 350 nm, respectively can be attributed to this geometrical difference in the setup. The fact that the light produced by the electron beam has to traverse the droplet before it is registered

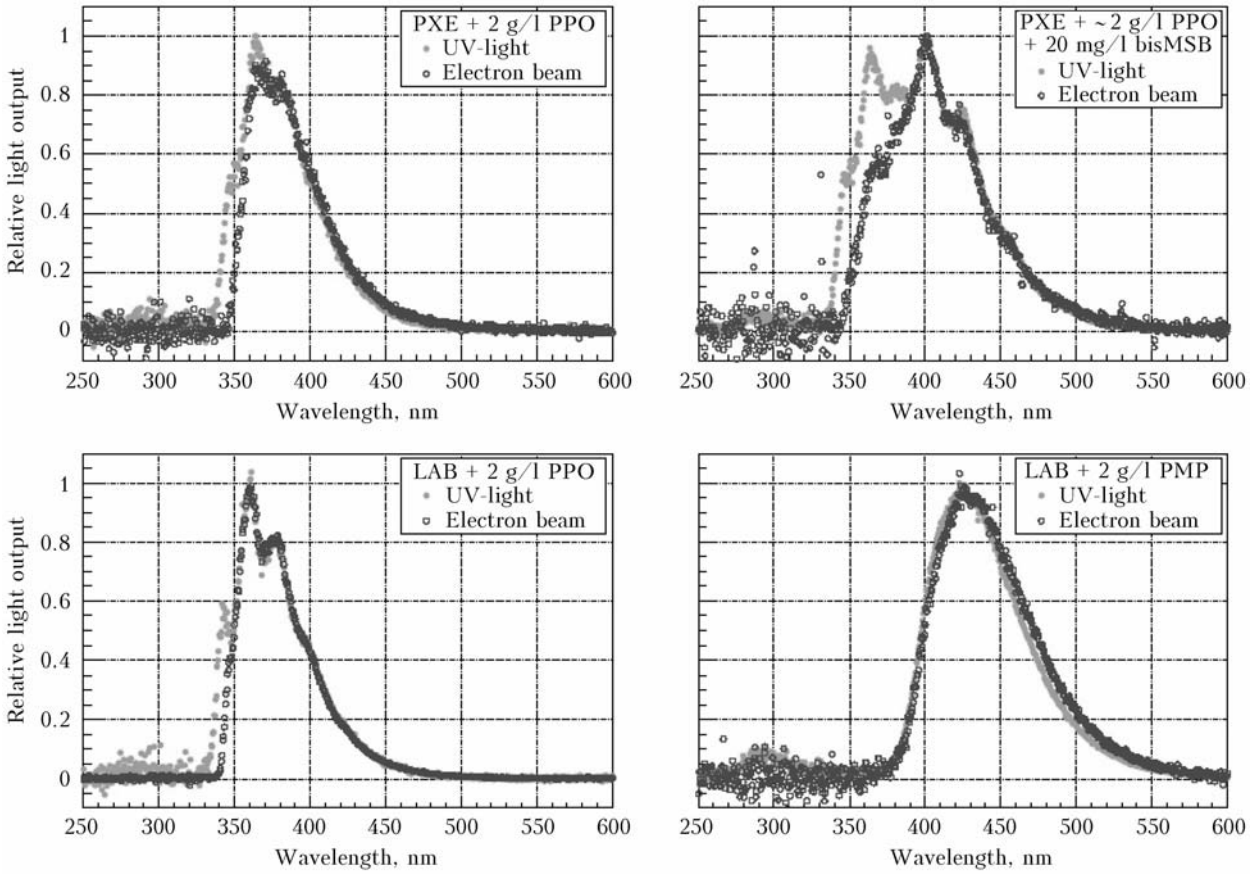

Fig. 6. Emission spectra obtained for the liquid scintillator material indicated for both excitation by an electron beam and UV photons from a deuterium lamp, respectively

leads to absorption features at the short wavelength side of the spectra. Only the mixture of LAB with 2 g/l PMP where the emission starts at rather long wavelengths around 400 nm is unaffected by any absorption feature. The interesting and positive result of this test, however, is the observation that the shape of the spectra beyond these absorption features towards longer wavelengths is practically identical for UV and electron beam excitation.

4. Summary and outlook

It has been demonstrated that gaseous and liquid scintillator materials can be studied using electron beam excitation with a compact, table-top setup. The key technology is a thin ceramic entrance foil through which the electrons enter the gas or liquid. Wavelength – as well as time spectra can be recorded. Whereas wavelength integrated time spectra can often also be measured using radioactive sources for excitation, the technology described here has the advantage that the beam power is high enough to record high quality wavelength spectra despite unavoidable losses of light in monochromators or spectrometers. The technology described here is also used to develop ultraviolet light sources [16] and will be used to study further gaseous and liquid scintillator materials in the future.

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