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Study of parylene-coated NaI(Tl) at low temperatures for bolometric applications

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ABSTRACT

NaI(Tl) is a widely-used scintillator at room temperature, and it is particularly interesting as a target for dark matter searches. Its hygroscopic character however makes it unsuitable for many applications, in particular for bolometric particle detection at very low temperature. Despite that, a NaI scintillating bolometer would provide unique features for dark matter detection, like β/γ background rejection through particle discrimination and thermal quenching factors for nuclear with respect to electron recoils close to one. With the long-term goal of developing a scintillating NaI bolometer, we have tested NaI(Tl) crystals coated by vapor-deposited poly-p-xylylene (parylene) and studied their optical and mechanical behavior in the mK range. We present X-ray excited scintillation spectra of a parylene-coated NaI(Tl) sample at 1.5, 4 and 77 K, and measurements of the light output as function of the temperature over the 1.5–300 K range. At 1.5 K the wavelength of maximum emission is observed at 325 nm. Thermoluminescence peaks are found at around 60, 95 and 150 K. Tests of mechanical and optical resistance to thermal cycles of 45 g parylene-coated NaI(Tl) cylinders are also presented, and the adequacy and effectiveness of this coating technique is discussed.

1. Introduction

NaI(Tl) scintillators at room temperature have for several decades been extensively used as detectors for ionizing radiation, and especially for γ spectroscopy. Among other nuclear and high energy physics applications, NaI(Tl) has been used as target for direct detection of dark matter (DM) in a number of past and present experiments [1–5] and, in particular, by the DAMA/LIBRA collaboration, that for more than a decade has reported an annual modulation in the detector rate at very low energy, compatible with that expected for a galactic halo DM signal [6,7]. NaI(Tl) offers many advantages that make it appealing for this kind of search, like its high light yield (that translates into low energy threshold), the possibility of growing large radiopure crystals, the combination of light and heavy nuclei and the fact that 100% of its isotope content is sensitive to spin dependent interactions. The main disadvantages are its hygroscopic character and the unfavorable quenching factor for nuclear recoils (NR) with respect to electron ones (~0.3 for Na and ~0.1 for I).

In recent years scintillating bolometers have been extensively developed as particle detectors for rare event physics and, in particular, for DM searches [8–10]. This technique, based on the simultaneous reading of the light and heat produced by a particle interaction in a target at mK temperatures, offers unique advantages for rare events physics: wide spectrum of target materials, low energy threshold, thermal quenching factor close to 1, good energy resolution, and particle discrimination capability down to a few keV or a few tens of keV (depending on the light signal threshold), thanks to the dependence of the light yield on the particle ionizing power.

Using the same target and different technique than DAMA/LIBRA experiment could help to confirm (and even to clarify) one of the most important results of the last decade in dark mater direct detection. The special features offered by scintillating bolometers could discriminate if nuclear or electron recoils are responsible

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of the event excess and obtain more information about its spectral shape, combining differently than in DAMA/LIBRA I and Na signals, that in the best experimental scenario, could even be discriminated (by the very different corresponding light yield). In such a case, a positive signal could really quote mass and cross-section of the WIMP candidate in a very robust way. Considering the aforementioned quenching factors for NaI(Tl), the region of interest of DAMA/LIBRA (from 2 to 6 keVee in the light signal) corresponds to energies between approximately 7 and 20 keV for Na recoils and between 20 and 60 keV for I recoils. As far as the thermal signal in a NaI bolometer is expected to have a quenching factor close to one, it could allow to explore regions below the DAMA/LIBRA energy threshold, specially for Iodine recoils, provided a response similar to other tested scintillating bolometers with thresholds below 10 keV is achieved.

The scintillation at low temperature of pure and thallium-doped NaI has been studied along the years by several authors (see for example [11–17] and more recently [18–22]). Concerning light yield dependence with temperature, Van Sciver and Bogart [12] reported an increase in light by a factor of two for NaI at 77 K with respect to the NaI(Tl) scintillation at room temperature, when exciting with γ rays from a ^{137}Cs source. Nevertheless, in [22] the same light yield was observed for pure material at 50 K and NaI(Tl) at room temperature when exciting with α particles. Although for both kind of materials a higher light yield has been reported with decreasing temperature, the augmentation is more pronounced for the pure material and the maximum of emission occurs at lower temperatures: for pure NaI several authors found the maximum around 60 K for γ rays [12] and α particles [15,22], while for activated material and alpha excitation Sailer et al. [22] reported a double maximum at 250 K and 150 K followed by a strong decrease in light and a further increase at temperatures below 30 K. As concerns the emission spectrum, unactivated NaI emits mainly at 300 nm and, with less intensity, at 375 nm (this last line has been associated with a stoichiometric excess of iodine [13,17]), and the wavelength of the main emission band does not change when lowering the temperature to 5 K [16]. However, the relative intensity of the fluorescence emission bands identified in NaI(Tl) has been observed to be dependent on temperature. The emission of activated crystals is dominated by Tl^+ centers, that at room temperature produce a broad emission at 425 nm. At liquid nitrogen temperature this band narrows and moves to longer wavelengths; we have not found in literature any spectrum for lower temperatures. A line at 325 nm has also been identified for low levels of dopant at liquid nitrogen temperatures, which has been associated with Tl dimers (two Tl ions as near neighbors) [11,13,23]. The intensity of this line depends strongly on Tl concentration and excitation particle.

Despite the high scintillation efficiency that has been observed at low temperatures and the intrinsic interest of this material as a DM target, NaI (pure or activated) has, to our knowledge, never been tested as a bolometer at very low temperatures. In order to do this, several problems have to be faced: its relatively high specific heat (although no measurements at very low temperature have been reported in the literature, the low Debye temperature (θ_D=164 K [24]) anticipates a volumetric heat capacity around 7 times that of Ge); the large coefficient of thermal expansion (1% between 300 K and 4 K [25]) which compromises the mechanical resistance of the crystal-sensor coupling under thermal cycles; and its high hygroscopicity which complicates enormously the bolometer design, manufacturing and operation.

Coating the NaI crystal with an appropriate material acting as humidity barrier could minimize the manipulation and cooling complications related to the hygroscopicity. The coating material should fulfill a number of requirements: transparency in the NaI/NaI(Tl) emission band, resistance to thermal cycles from room temperature to below 100 mK, radiopurity, low heat capacity in the mK range, good thermal conductivity, compatibility with some selected epoxies for the thermal coupling to the sensor, etc. In the case of non scintillating materials, the coating can cause a leakage of $\alpha/\beta/\gamma$ surface events into the (less scintillating) NR band, increasing the background and limiting the DM sensitivity of the bolometer. Given the relative high rate of events attributed to DM in the DAMA/LIBRA experiment (S_m=0.0116 ± 0.0013 c/keVee/kg/day [7], that supposing a modulation amplitude amounting a 7% of the total results in an expected DM interaction rate of around 0.7 c/kg/day between 2 and 6 keVee), the presence of a modulated component in the NR band should be detected even with backgrounds of the order of 0.5 c/kg/d in the ROI for exposures of the order of several kg $\times$ year. For non scintillating coating materials surface-events rejection methods based on pulse shape analysis could also be explored to reduce background level. Anyhow, having as main goal the development of a NaI or NaI(Tl) scintillating bolometer to be applied in the search for DM, coated bolometers should be taken as an “easy to mount” approach; they could allow to carry out thermal and scintillation quenching studies. Nevertheless, to avoid the possible drawbacks coatings could imply in thermal and background issues for the building of a competitive bolometer, we do not discard as final detector design to build naked NaI bolometers inside a sealed enclosure.

This paper studies the low temperature light output and the mechanical resistance to the cooling down process of parylene-coated NaI(Tl) crystals. Activated materials were selected for the study because *a priori* their luminescence spectrum matches better the parylene transmission band [26].

The structure of this article is as follows: we start with a brief description of the parylene conformal coating process, its properties and the experimental details of the deposition done for this work (Section 2). Section 3 is a study of the scintillation at low temperature of a parylene-coated NaI(Tl) sample. The experimental procedures, equipment and sample preparation are described in SubSection 3.1, while in SubSections 3.2 and 3.3 X-ray scintillation spectra and light yield vs temperature curves are presented. Section 4 is a study of the mechanical and optical resistance of the samples to thermal cycles. Finally, conclusions are drawn in Section 5.

2. Parylene conformal coating

Parylene is the generic name for a commonly used polymer family based on poly-p-xylylene. It can be deposited in thin films by vapor-phase condensation polymerization: the solid dimer (dichloro-p-cylophane) is sublimed at more than 100 °C and then pyrolized at more than 600 °C to produce the monomers. These molecules are conducted to a vacuum chamber and spread throughout it, being adsorbed and simultaneously polymerizing on all the exposed surfaces. In this way a conformal and very homogeneous layer is formed, whose thickness depends on the exposition time and can be made as thin as several hundredths of a micron or as thick as 100 microns. Deposition is pinhole-free, it can be accomplished at room temperature (avoiding thermal stresses on the sample) and solvents are not required. The resulting parylene layer has been tested under thermal cycles, showing mechanical resistance to thermal stresses and is widely used in the space industry. On the other hand, the deposited film is expected to be as radiopure as the starting materials, due to the absence of additional solvents and the vacuum chamber deposition at a dedicated facility.

Several types of parylene are available. Among them, parylene C, in which one of the aromatic hydrogens is substituted by a chlorine atom, has superior barrier properties against moisture trans-