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Nanostructured materials for particle detectors

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This paper discusses the opportunities and the challenges related to the use of nanostructured material for particle detectors.
These materials are subject of intense research and application works since the nineties. Nevertheless, a direct application in technology related to particle detection in accelerator or underground experiments can not yet be found. Despite of this, several R&Ds based on these advanced materials are pursued by few INFN research groups and in the world.
Nanotechnology is advancing rapidly and it is worthwhile to be aware of its potentiality with the ambition to improve the performance of existing detectors or conceive new ones as required in future generation of experiments.
Here the discussion is limited to nanostructured materials where detector applications are envisaged and some INFN groups are involved: colloidal quantum dots, metal nanowires, carbon nanotubes and graphene sheets, which are examples of materials with dimensionality 0, 1 and 2.

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1. Introduction

Over the past decades, in academic and industrial sectors a lot of interest raised on nanostructured materials thanks to their impressive variation in electrical, magnetic, and optical properties with respect to "infinitely extended materials". Nanostructured materials are made of building blocks having one or more dimensions with a size ranging from 1 to 100 nm. This leads to a countable number of atoms and electrons confined in nanoscale structures and with a physical behaviour ranging from semiconductor to metal.

The lower dimensionality of the nanostructured material exhibits new phenomena, such as plasmonic enhancement of light absorption and multiexciton generation ([1]), and it has the remarkable feature of size-based spectral tuning. In addition to that, new kind of devices, such as graphene based FETs or THz bolometers, could amplify a charge signal or sense a temperature pulse of a traditional bulk materials, in order to boost significantly the signal-to-noise ratio. Finally, several attempts were done to employ these materials as electrical conductors for realising interconnection techniques and electrodes transparent to light or to electrons.

Particle detector technology, in general, and physics driven particle detection, in particular, could benefit from technological advances of these "new materials". Anyway, it is important to be aware that particle detectors have peculiar properties difficult to meet in emerging materials, sensors or devices, such as very high efficiency, fill factor near to one, radiation hardness, ageing, large area and volume and affordable cost. Nevertheless, this is not always the case, for example nano-technology researches in photovoltaic cells, flat screens and organic electronics promise large area and low cost devices. In fact, the synthesis of nanostructured material, based on a "bottom-up" approach, could be often cost effective, radically different from the "top-down" approach where lithographic methods are used, like in the fabrication of conventional submicron solid state devices and MEMs.

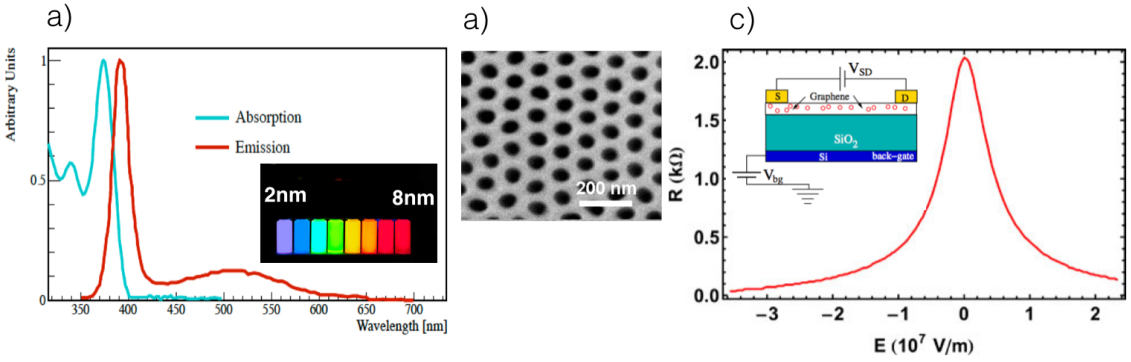

Figure 1: a) Absorption and emission spectra of CdS QDs. b) Alumina nanotemplate for CNT and NW growth. c) Drain-source resistance of a graphene sheet as a function of transverse electric field in G-FET.

[1]The remarkable property to convert one absorbed photon in dozens of free carriers is subject to extensive work in photovoltaic cells but could inspire new concepts in photodetectors like no-drift based detectors [1].

2. Colloidal quantum dots

Quantum dots (QDs), like to nanoparticles, are spherical assemblies of nanometer size, but exhibit semiconductor behaviour, with a band gap E_g inversely proportional to the radius R:

$$E_g \propto \frac{1}{R}, \tag{2.1}$$

which is a typical feature of all semiconductor nanostructured materials [2]. The quantum dots act like perfectly tunable wavelength shifters, and therefore have obvious application for scintillator-based detectors.

The most popular quantum dots are chemically synthesised in a colloidal solution and they are composed by elements from periodic groups III-V or II-VI (ZnS, ZnSe, CdS, CdSe, CdTe, ...). Large batches of colloidal QDs are commercially available in kg scale. A coating of organic ligands is used to suspend the quantum dots in the solvent, like toluene, but polar solvents, like water, are also possible. The most common quantum dots are a core of CdSe crystals, which has a size from 2 nm to 8 nm leading to emission spectra between 450 nm and 650 nm (Fig.:1a). A shell of another crystal, for example ZnS, is sometimes used to passivate the secondary surface states, which affect the QDs optical properties. These are referred to as core-shell quantum dots.

Quantum dots manifest stronger light absorption than traditional scintillation materials for the same concentration in the primary scintillator. They absorb the light shorter than a characteristic wavelength and reemit in a narrow resonance range of about 25 nm around this wavelength (Fig.:1a). This is different from traditional scintillators, where the absorption and emission spectra have two distinct broad peaks (Stoke shift). Apparently, the partial overlap between absorption and emission spectra does not compromise the envisaged high quantum yield. Anyway, another phenomenon could cause the loss of the reemitted light, this is the energy transfer to the solvent by means of non-radiative dipole-dipole interaction, termed non-radiative Fluorescence Resonant Energy Transfer (FRET).

2.1 QDs in liquid scintillator

A very advanced study on colloidal quantum dots performance, as liquid scintillator in toluene, has been performed in ref [2] and compared to PPO. The spectrum from a ^{90}Sr beta source was measured with 20 mL of toluene, showing a reasonable signal yields for 1.5 g/L of quantum dots from different suppliers. Anyway, the signal yield is lower than PPO with a concentration of 5 g/L. It is worth to underline that higher quantum dots concentrations could reduce the attenuation length, due to the self-absorption, and increase the non-radiative FRET, with respect to the radiative one. The possibility to tune the emission wavelength to the photosensor response spectrum should further improve the signal yield of quantum dots. In the same reference a physics driven approach to the application of quantum dots is clearly stated. In fact, quantum dots are particularly interesting for neutrino physics due to the nuclear properties of their elements: Cd and Gd have high thermal neutron cross sections, good features for antineutrino detection, while Cd, Se, and Te are candidate isotopes for neutrinoless double-beta decay searches.

[2] It is worth to notice that quantum confinement predicts $1/R^2$ and classic theory based on dielectric properties of sphere predicts 1/R.

2.2 QDs in plastic scintillator

Further chemistry can be done to embed the colloidal quantum dots into a solid matrix (PMMA, GLASS, SU8, MEH-PPV) in order to realise a new class of free standing scintillators or wavelength shifters (Qdots-WLS experiment in CSN5).

The energy resolution of this solid scintillator is in principle between the one of cheap plastic scintillators and the one of expensive scintillation crystals, thanks to the band-gap value of several eV, but with a fabrication cost similar to plastic scintillators. The response of quantum-dot-doped solid scintillator has been studied for gamma rays [3], electrons [4] and thermal neutrons [5].

QD/organic composites with large enough QDs concentration should produce excitations predominantly in the inorganic semiconductor QDs. These excitations are FRET-transferred to the organic transparent material, which can emit Stokes-shifted photons, as in organic scintillator, or can dissociate electron-hole pairs, as in organic photosensors[3]. Theoretically, the expected yield of the QD/organic composite should be a factor of 10 higher than not-doped organic plastic. Unfortunately, only a maximum gain of 2 is experimentally observed for a quantum dots volume fraction of 0.15 ([4]), likely due to non-radiative FRET.

2.3 Some issues for QDs

Intermittency phenomena and on/off emission are observed in all nanostructures, such as colloidal quantum dots, nanorods, nanowires and some organic dyes. The phenomena are named blinking and could have a negative impact on efficiency and dark count rate. This is also true for another phenomenon affecting nanostructures, named quenching: this is the off emission due to environmental boundaries, for example when Au nanoparticles are present.

The decay time of colloidal quantum dots is quite fast, few hundred of ns, but not fast enough for the demanding high energy physics applications. From this respect, more promising are the quantum dots, quantum wires or nanowires deposited by Metal-Organic Chemical Vapour Deposition (MOCVD) on a substrate, such as GaAs/AlGaAs, where a decay time of the order of 10-100 ps is measured [6], [7].

Finally, the CdSe / ZnS colloidal quantum dots photoluminescence intensity shows a factor of two decrease just after 10 kRad of ^{137}Cs gamma irradiation [8] . This poor radiation hardness is likely to preclude application in accelerator experiments where radiation hardness is needed, but could open opportunities in dosimetry detector for medical applications (NADIR experiment in CSN5).

3. Carbon nanotubes

Carbon nanotubes (CNTs) have high conductivity, high aspect ratio and outstanding mechanical strength. Several studies were carried out on CNTs since about a decade from different INFN groups (experiments GIN, SINPHONIA, PARIDE, NANOCHANT, CANTES, SERENA and ESOPO in Commissione Scientifica Nazionale 5 [CSN5]). These groups use different deposition techniques in collaboration with italian research institutes mastering the technology: Chemical

[3]Trap-free, conjugated organic materials are now available for electronics and opto-devices. Anyway, the slow response and the ageing still limit their applications in nuclear detectors.

Vapour Deposition (CVD) INFN-NA and UNI-AQ [9], spray deposition INFN-BA and CNR-BA [10] and CVD growth in a template of porous anodic alumina (Al_2O_3) INFN-BO and CNR-IMM-BO (see [11] and Fig.:1b). In all cases multi-wall nanotubes (MWCNTs) are produced, which are easier to produced than single-wall nanotubes (SWNTs) [4]. The drawback of MWCNTs is related to the observed spread in size, consequently, a spread in band-gap.

To date there is not yet experience in INFN groups of nanostructures growth starting from a matrix of regular and dense nanoparticles (for example nanodroplets of Fe or Ni) based on the physical principles of Vapor Phase or Vapor-Liquid-Solid Phase growth inside traditional reactors (CVD, MOCVD, ...) [12]. These methodologies allow to grow very regular arrays of single nanotubes or nanowires, with a behaviour ranging from metallic to semiconductor. The 1D nanostructures grow between the substrate (common reference plane) and the catalytic metallic droplet seeds, which remains on top of the nanostructure (single nanocontacts). This opens the theoretical possibility of detectors with a spatial resolution of few tens of nanometers.

Several photodetector devices, made by CNT layers, were realised and studied by INFN groups, showing that the light is absorbed with high efficiency and converted in free charge carriers on the semiconductor substrate by ohmic conduction or tunneling.

In reference [10], semi-insulating GaAs substrates were covered on the front-side by a 0.5 μm layer of MWCNTs by dispersion spray using an ultrasonic atomizer. The MWCNT side was electrically connected by a ITO layer and the substrate back-side by a TiAu layer. Current-voltage (IV) curves were measured in dark conditions and with light ranging from IR to UV. Good ohmic contacts between ITO and GaAs are observed. A 15% quantum yield at 170-180 nm is measured only if the MWCNT film is present, showing the beneficial effect of the MWCNT layer in UV photoresponse. The quantum yield increases up to 34% if the GaAs substrate is covered by MWCNT film on both sides.

In reference [9], MWCNTs were grown by CVD on a 60 nm thick insulating silicon nitride layer, deposited on the front-plane of n-type Si substrate. Two small spherical Au/Pt electrodes were also deposited on the insulating silicon nitride and grounded. On the back-plane the silicon nitride was absent, instead a thin n^+ layer was implanted to ensure a good ohmic contact with a large Au/Pt pad electrode. Surprisingly, the IV curve shows a negative differential resistance when the applied voltage has a value of 2.4 V and the light impinges on the CNT carpet. This behaviour is typical of a tunneling diode where electrons can tunnel through some resonant states at a certain voltage level. Also in this CNT device the conversion efficiency of light to photocurrent is very good: about 92% at 730 nm and about 43% at 378 nm.

A very large array of well aligned SWNTs are instead required for the innovative proposal illustrated in reference [13] for a directional dark matter detector. The proposal relies on the theoretical prediction of channeling for a carbon ion ejected from the nanotube wall due to a scattering with a massive particle. The ion channeling provides the required anisotropic response of the active material. A TPC+GEM gas chamber is proposed to detect the scattered carbon ions with a low energy threshold. In fact, the channeling probability is expected to be strongly dependent on the relative orientation of the nanotube axis and the massive projectile particle. Anyway, the experi-

[4] Single-walled carbon nanotubes are rolled-up graphene sheets with unique electronic properties, which can change from semiconductor to metallic, according to graphene sheet tilting angle.

mental observation of the channeling phenomena in nanostructures is already a very ambitious and relevant scientific goal.

4. Surface Plasmon Resonance in metallic nanowires

Surface plasmons (SPs) are collective oscillations of conduction electrons excited by an external electromagnetic wave and bounded at the metal-dielectric or metal-semiconductor interface. For a metallic nanostructure the SPs create opposite charge on opposite surfaces. The dipole-like restoring forces strongly excite Surface Plasmon Resonances (SPRs) [14], which can increase the electromagnetic near-field of an order of magnitude more than the external wave.

The key features of the SPRs depend on the radius R of the nanostructure. The resonance spectrum is of the order of R meaning visible or near infrared, and the resonance bandwidth goes like 1/R meaning a large bandwidth for small particles. Absorption-to-scattering ratio for electromagnetic far field is proportional to R meaning that small particles strongly absorb light, while large particles are dominated by light scattering. Another very peculiar feature of light absorption by SPR is the antenna-like behaviour, which allows, at the same time, very high effective fill factor and light concentration in a small area.

A nanostructured light-sensing layer can be optimized for high fill factor and for a specific absorption spectral region. In addition to these, it can be coupled to several semiconductor substrates, which could be independently optimized to convert light into ionization charge with high collection efficiency.

The INFN-BO group which developed the alumina template for oriented-CNT growth by CVD (Fig.:1b), used the same template to electrochemically growth oriented-metal nanowires (Ag, Ni, Co, Fe, Cu) in collaboration with CNR-IMM-BO. The nanowires (NW), grown in the template, have a cylindrical monocrystalline structure and they are vertically aligned within the matrix pores. The nanowire geometry is forced to be the same of the alumina template pores, which is controlled by the anodising process parameters and duration (diameter from 10 to 100 nm, pitch from 10 to 200 nm and the length up to several μm).

Recently, they observed that the presence of Ag nanowires, in anodic alumina pores, introduces a bump in the transmission spectrum, around the Ag nanowire SPR wavelength, of about 325 μm. For a 5 μm Ag nanowire length the transmission spectrum is about 10% higher with respect to the porous alumina without nanowires. This clearly shows a plasmonic resonance coupling of the impinging light with the nanowire entrance ends (light absorption), together with a plasmonic wave propagation along the nanowires (waveguide) and, finally, a reemission of the light on the nanowire exit ends (light source). These effects are similar to what is described in reference [15] .

In the experiment REDSOX2 of CSN5, the plasmonic resonance and its propagation in metallic nanowires are going to be exploited to improve the quantum efficiency in the UV-VI-IR spectral regions of Silicon Drift Detectors (SSDs). The challenging idea is to replace the existing Si_3N_4 Anti-Reflecting-Coating of the SSD, which is responsible for the cut-off below 500 nm, with a matrix of nanowire in porous alumina of 0.5-1 μm thickness. The nanowires, having a plasmonic resonance below the cut-off, should extend the SSD quantum yield up to 300 nm.

5. Interconnection with nanomaterials

Some promising research on small-pitch interconnection technology, using nanostructured material, could brought to lower cost and higher current density, having a benefit impact in solid state pixel detectors. In addition to that, 3D stacking of circuits is one of the key technologies for future silicon scaling.

The CERN laboratory, in collaboration with a Finland farm, is studying the possibility to use carbon nanofibers (CNF) to realise low-cost, low-mass and small-pitch bump bonding. The carbon nanofibers are grown by CVD in the passivation openings of the sensor and coated by a polymer. The chip is interconnect to the sensor by bumping the Ni/Au Under Bump Metalization (UBM) deposited on its opening with the CNF islands. Before bumping, the CNF free ends are exposed to air by etching the polymer and, after bumping, chip and sensor are glued by melting the not-etched polymer fraction.

Through-Silicon-Vias (TSVs) play a critical role for 3D chip stacking and for four-side buttable pixel sensors to avoid dead area boundaries. TSVs are generally made by metals, but a possible alternative it is to use aligned CVD carbon nanotubes to fill the pre-etched silicon holes. This could reduce the cost, speed-up the process and achieve high aspect ratio (10:1) in TSV technology.

There are two primary limitations hindering the potential of CNTs: relatively high synthesis temperature (600-800 ^{0}C) and low volume fraction ($< 10\%$). To overcome the limitations, CNTs are grown on a sacrificial silicon wafer and post-growth densification is applied. Consequently, the CNTs are aligned and inserted into the TSV wafer with pre-etched holes. After the silicon template is etched, the CNT-hole gaps are filled with epoxy resin. Very promising results were obtaining with the above illustrated methodology [16].

A very distinct property of nanostructures is the lower melting point with respect to bulk material of the same composition. This suggests to investigate the use of nanostructures for low temperature soldering/bonding applications. Low temperature wafer bonding has been demonstrated at 200 ^{0}C by depositing Oblique Angle Cu nanorod arrays. For higher bonding temperatures, the bonding structure further evolved by pore reduction and grain growth, until a full densification is achieved upon 400 ^{0}C. The bonding with Cu nanorods exhibits a more homogeneous structure than with a Cu blanket film. The study proves that Cu nanorod arrays are feasible adhesive layers for low-temperature wafer bonding [17] .

6. Graphene

Graphene is the only freestanding two-dimensional material: an atomic monolayer of carbon atoms with a thickness of 0.335 nm [18]. Thanks to its hexagonal honeycomb structure of sp_2 hybridized carbon atoms, graphene is stronger than steel and has better electronic conducibility than copper and silver. The non-hybridized p_z atomic orbitals result in a cone-shape valence and conduction bands (Dirac cones), which meet to a point (Dirac point) with a zero band gap (semimetal structure). The cone-like energy dispersion relation results in free massless Dirac electrons with no inertia, because of the uniform velocity (v_e = dE/dk). Therefore, a very high free carrier mobility and conductivity can be reached along the graphene sheet.

Since its discovery, the development of a graphene-based FET was very fast (G-FET in Fig.:1c), reaching quite soon a cut-off frequency of 100 GHz. However, the absence of an energy gap makes graphene sheets unsuitable for digital switching devices. In addition to that, a peculiar saturation behaviour limits the G-FET radio-frequency performance [19] .

It is significant to mention that carbon atom displacement energy is quite large for graphene, this is of 15-22 eV as graphite [5]. This means that graphene is potentially a radiation hard material [20].

There are two INFN experiments in CSN5 devoted to develop graphene-based detector. The first experiment is named GARFIELD and it has the primary goal to implement a full-capability graphene production within the INFN-LNF laboratory. The experiment grows graphene by Chemical Vapour Deposition, which is suitable for producing large-area multi-domain monolayers on copper foils.

Recently, an international collaboration, using this method, realised monolayers of graphene on Cu substrate. Subsequently up to three graphene monolayers have been transferred to a copper mesh. The copper mesh-graphene stacks were inserted into a GEM detector to measure their charge transparency [21]. A strong asymmetry in transmission between low energy electrons and ions is observed in this graphene triple layer, opening the possibility to reduce the ion feedback and space charge distorsion in high rate Time Projection Chambers.

The second experiment using graphene is named GBTD. It aims to demonstrate single photon detection capability from UV to THz radiation by developing ultra-cryogenic bolometer based on graphene as thermometer, for dark matter and double-beta decay neutrino experiment. In this work commercial high quality graphene is purchased on a substrate, complete of contacts and isolation cover.

6.1 Sensing a MIP with G-FET

A small vertical electric field can shift the graphene Fermi level upward (or downward) with respect to the Dirac point, which means that the holes (or electrons) became the majority carriers (ambipolar conductivity, see Fig.:1c). At the Dirac point, the free carrier modulation is extremely sensitive to external electric and magnetic fields. This makes graphene suitable for a huge number of sensing applications (photons, gas, chemical, biomolecular detectors).

Until now, the real challenge to detect a Minimum Ionization Particle (MIP), using the graphene built-in FET structure, it has been addressed only by TCAD simulations [22]. In this work an ionizing particle, crossing a traditional bulk material, such as Silicon or GaAs, is sensed by measuring the in-plane electric resistance of a deposited graphene sheet. A strong sensitivity is expected if the graphene sheet is biased exactly at the Dirac point, where the variation of the resistivity, with respect to the transverse electric field, is very high (Fig.:1c). In fact, the drifting ionized charges produce an electric field with a transverse component to the graphene sheet, modulating the graphene free carrier density. A clear advantage of such a structure is the possibility to use a variety of substrates, not necessarily of detector grade quality.

[5] The graphene displacement energy is less than diamond-like material, which is 30-48 eV but higher than Silicon, which is 13.5 eV.

Severe drawbacks of this detector schema are the slow collection speed of the ionized charge, due to the not optimal electric field configuration in the bulk material, and a fast build-up of localized charge, which must be cleared otherwise the device will stop working immediately. These issues are also faced by proposing a DEPFET-like structure. In this device the radiation-induced charge carriers are very quickly drifted below the G-FET by an underneath n-well. When the readout is completed, the charges are removed by a lateral clear-gate.

6.2 Graphene-based hot-electron bolometer for THz radiations

A bolometer operates at cryogenic temperatures, typically no more than few degrees from absolute zero. It measures the energy E released in the active material by an incident radiation, sensing the pulse temperature ΔT. The high sensitivity is reached by a small thermal capacitance C, because $\Delta T=\frac{E}{C}$, and by a small thermal conductance G, because the time response $\tau=\frac{C}{G}$ is long.

Graphene is an optimal hot-electron bolometer [23], the most sensitive kind of bolometer for THz radiations. At low temperature it has a small electron thermal capacitance ($C_e \propto T$), due to the low density of states near the Dirac point, and a small electron thermal conductivity, due to the low electron-phonon interaction ($G_e \propto T^3$). The graphene response turn out to be fast, being of the order of few μs. Finally, the coupling to an antenna of few 100 μm linear size, required for a THz detector, can be designed with high efficiency, because graphene has a low impedence above this frequency range.

The figure of merit of a bolometer is anyway the energy resolving power $R=\frac{E}{\delta E}$ for the complete device. A bolometer, in addition to the intrinsic energy resolution $\delta E_{int} \approx \sqrt{k_B T^2 C_e}$, due to the thermodynamical fluctuations of the internal energy (phonon noise), has also a noise contribution introduced by the readout system. Unfortunately, the resistance of the graphene with metallic (non-superconducting) contacts is essentially temperature independent. Due to this the traditional readout schema of negative electrothermal feedback for reducing the intrinsic noise, can not be used.

A lot of interesting ideas were proposed to readout the graphene electron temperature, but here we report only the two most simple: measuring the Johnson noise related to the resistance, which is temperature dependent, and using the superconductive contacts, which introduce temperature dependence in the resistance. The Johnson noise readout schema, in non-linear mode, predicts an energy resolving power of about R=2.2 for an area of 4.5 μm^2 at T = 0.1 ^{0}K and allows to perform single photon detection. Superconductive contact readout schema can work with much larger sensor area but can not resolve single photons. For example, for an area of 1000 μm^2 at T = 0.1 ^{0}K, the energy resolving power is predicted to be about R=0.5, and the device can work only as power detector and not as calorimeter.

Based on the measured electron-phonon coupling a graphene-based bolometer should have a Noise Equivalent Power (NEP) of about 33 fW$\frac{fW}{\sqrt{Hz}}$ at 5 ^{0}K. This is about 10 times better than silicon bolometers and similar to superconducting Transition Edge Sensors (TES) working at 4.2 ^{0}K. Anyway, the response time of graphene bolometer is three order of magnitude smaller than TES. If the extrapolation from 5 ^{0}K to 0.1 ^{0}K of the graphene electron thermal conductivity and capacitance will be confirmed experimentally, the expected ultimate NEP could be less than 10^{-4} $\frac{fW}{\sqrt{Hz}}$ with a response time of 3 μs (similar NEP is reached by TES for IR photons but with much smaller bandwidth) [24] .

7. Conclusions

Nanostructured materials are new to high-energy physics but a lot of research and applications are pursued in other scientific fields. Several nice ideas are proposed also for detectors in this last decade [25], but they turned out to be technologically too difficult or too naive to be competitive with traditional concepts. This could reveal an intrinsic difficulty or a too early technological development phase.

The very striking peculiarity of a particle detectors is to detect the passage of a single penetrating particle with high efficiency over a large area. A goal that is not easy to reach by unconventional sensors. This is traditionally accomplished by sensing free charge carriers or photons or temperature pulse generated directly, or indirectly, in the active material by the primary particle. The active material is typically a bulk material having very low dark counts (low leakage current or very low background of photons or phonons). In this way a measurable electric signal can be obtained by low noise electronic amplifiers occupying small volumes.

In this scenario, INFN could play a relevant role in new ideas, device simulation, advanced front-end, interconnects, daq and data analysis, but it should create a synergic alliance with the other research institutes, which must have the expertise in the relevant nanotechnology fields. Although, new detector concepts, based on these enabling technologies, could be a high-risk investment, they could open up exciting possibilities for the next generation of experiments. The INFN scientific tradition and excellence could be the crucial ingredients of success and help the institute to maintain the current national-wide scientific and technological leadership.

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