Snowmass2021 - Letter of Interest

[Novel Low Workfunction Semiconductors for Dark Matter, Neutrino Phenomena and x-ray Astronomy]

Instrumentation Frontier Topical Groups: (check all that apply \Box/\blacksquare)

- □ (IF1) Quantum Sensors
- (IF2) Photon Detectors
- (IF3) Solid State Detectors and Tracking
- □ (IF4) Trigger and DAQ
- □ (IF5) Micro Pattern Gas Detectors (MPGDs)
- (IF6) Calorimetry
- □ (IF7) Electronics/ASICs
- □ (IF8) Noble Elements
- (IF9) Cross Cutting and Systems Integration

Other Topical Group(s):

- ✓ (CF1) Dark Matter: Particle Like
- ✓ (CF3) Dark Matter: Cosmic Probes
- (CF7) Cosmic Probes of Fundamental Physics
- ✓ (NF10) Neutrino Detectors

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ABSTRACT: In frontier physics, precision calorimetry from photons and charged and neutral massive particles have been crucial to major discoveries. Better resolution and low detection thresholds are of great interest for dark matter searches, solar and reactor neutrino detection and oscillations, neutrino mass measurements, x-ray astronomy, and double-beta decay. A Challenge for semiconductor detectors: $\sigma_{E}\sim10-100$ eV. We discuss semiconductor photocathode materials *Cs3Sb(S-11) and Ag-O-Cs(S-1)* as possible bulk detectors.

Novel Low Workfunction Semiconductors for Dark Matter and Neutrino Phenomena

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ABSTRACT: In frontier physics, precision calorimetry from photons and charged and neutral massive particles have been crucial major discoveries. Better resolution and low detection thresholds are of great interest for dark matter searches, solar and reactor neutrino detection and oscillations, neutrino mass measurements, x-ray astronomy, and double-beta decay. A Challenge for semiconductor detectors: $\sigma = -10-100 \text{ eV}$. We discuss the semiconductor photocathode materials $Cs_3Sb(S-11)$ and Ag-O-Cs(S-1) as possible bulk detectors.

Semiconductor detectors benefit from high atomic number (Z), density (ρ), and low electron-hole pair production energy E_{pair} or E_p (Ethreshold pair production). For the lowest bandgaps E_g , operation must be cooled to LHe or lower to avoid thermal noise generation. The probability per unit time that an electron-hole pair is thermally generated is $P(T) = CT^{y_2} e^{E_g/2kT}$ where T temperature, E_g the band gap energy, k the Boltzmann constant and C is a constant characteristic of the material. For operation at room temperatures, $Eg \ge 1.3 \text{ eV}$ is desired so thermally generated carriers do not dominate low energy events. For high resolution, the pair energy Ep = Eg + Ea (Ea=electron affinity), also called the threshold energy E_{th} . $E_{th}(=Ep)$ is similar to/related to the work function ϕ but can be more or less than the vacuum level VL. Typically in alkali photocathodes the ϕ is slightly more than Eg+Ea(Fig 1). Poor photoemitters are like the right diagram, like Si or Ge, where most of the electron-hole pairs do not have enough energy to be above the vacuum level (work function) to escape. However, the electron-hole pairs are above the conduction band and can be drifted to electrodes or a junction.



Figure 1: Left: Energetics for alkali photocathode vacuum emission – the pair energy Ea+Eg is near the vacuum level. ML: Cs_3Sb semi-conductor energy level band diagram – the energy for pair production is ~Ea +Eg= 1.9-2.0 eV, near the vacuum level and like the 2nd class shown. M.R and Right: Cs_3Sb and Cs_3Bi have similar pair energies Ep proportional to Ea+Eg. Cs_3Sb bandgap inhibits thermal energy promoting carriers to the Fermi level, operating with no cooling for low energy events, such as coherent neutrino scatterings.

For a given energy deposition in a detector material (the full energy photoelectric peak), to 0th order the energy resolution limit $\sqrt[4]{-\sigma(F/n)}$, where n~E/Ep is the number of generated carriers; F the Fano factor as small as ~0.1 for semiconductor detectors (Ge~0.06). A basic figure of merit M for a detector material at energies <500 KeV is M ~ $\rho Z^{35}/\sigma$. For higher energy, the reciprocal of the radiation length Lrad scales as a lower power of Z , ie ~Z². There are many examples of semiconductors as diodes or drift cells for calorimetry. Ge reaches resolutions of ~0.2% at 662 KeV (±1400 eV) and peak/Compton ratios of 30 or even higher, but Eg = 0.7 eV and Ep = 2.96 eV, too large for extending present Dark matter and double-beta decay searches. Silicon pair energy Ep=3.6 eV is similarly too large, as is GaAs. By contrast, some semiconductor materials normally used for vacuum photocathodes have much lower Ep. Cs₃Sb (S-11 photocathode-like) has a ~2eV pair energy. Cs-Ag-O (S-1 like) has a surface averaged pair energy/work function Ep=0.7 eV in commercial vacuum phototube applications. Several research studies demonstrate that small patches (1-2 mm₂)on thermal evaporated S-1 Cathodes have pair energies as low as Ep=0.3-0.4 eV).

	Ζ	$\rho(g/cc)$	Egap(eV)	Epair	μ e, μ hole cm ₂ /V/s	Lrad(cm)
Ge	32	5.3	0.7	2.98	≤3900, ≤1900	2.3
Si	14	2.3	1.1	3.6	≤1400, ≤450	9.4
GaAs	31,33	5.3	1.4	4.4	≤8500, ≤400	2.3
$Cs_{3}Sb$	55,51	4.6	1.6	2.0-2.1	10,000-106	1.9
Ag-O-Cs	55,47,8	7.1	< 0.3	0.4-0.7	Predicted ~5,000	1.8-2.0

 Table 1: Semiconductor properties

1. Cesium antimonide (Cs₃Sb) is a weakly bound cubic semiconductor, lattice constant of 9.15 Angstroms, normally p-type from Sb excess displacement defects, with relatively well-known properties: a very low electron affinity of 0.4-0.5 eV, and room-temperature intrinsic resistivity ~1,000 Ω -m, about 10^{7.8} times higher than Sb, comparable to the 500 Ω -m of Ge at 77K, and a mobility μ - even in the highly defect filled films of photocathodes which exceeds - 10^4 cm²/Vs. Indeed, on theoretical grounds μ could exceed 10^6 , like InSb [2]. For calorimetry, as compared with Ge at 2.98 eV, the pair energy for Cs₃Sb is a remarkably low 2 eV [3], [4], [5] and therefore, in principle, Cs₃Sb has an intrinsic resolution better than Ge by $\sim \sqrt{(2.98/2)}$. On the other hand, the bandgap is 1.6 eV, exceeding Si (1.1 eV) and even CdTe (1.47 eV) so that thermal noise is minimal at room temperature. The thermal noise current at room temperature is between 10,000-20,000 times lower in Cs₃Sb than Silicon, which alone makes it interesting as a detector. That the pair energy Ep is only 0.4 eV above Eg is the secret to this potentially breakthrough material, quite unlike Si, Ge and the like. The density of Cs₃Sb is 4.6, compared to 5.3 for Ge, however Z of Cs₃Sb is 55 and 51 (average ~54), compared with 32 for Ge. The result is that photoelectric absorption – as an example of a radiation detector function - is about 4-5 times higher for Cs₃Sb compared with Ge for any given thickness at ~662KeV gamma-rays, and the Peak/Compton ratio should exceed Ge, provided a low-defect crystal can be obtained. Moreover, the radiation length is short. As a detector for low energy deposition photons or dE/dx, Cs₃Sb may be superior Ge in many respects. Because of its larger atomic number its photoelectric absorption is larger, its peak to Compton ratio is larger, and its radiation length is shorter. It has better resolution because its pair energy is lower. It does not need to be cooled for applications at room temperature, because its bandgap is large, larger than Si. Compared to Ge, the cost of the highly purified raw material is negligible. A shortcoming is that, like NaI, it must be protected from the atmosphere. It can be grown from a melt similarly to that of NaI or CsI. The Phase diagram for Cs+Sb melts form Cs3Sb at 725 °C. Recently it has been shown that one molecular thickness of boron carbide (BN) or one layer of graphene can protect it from air, important for practical detectors. An atomic BN layer (< 100°C[6]) lowers the pair energy from 2 eV to 1.6 eV[7],[8].Crystals of Cs₃Sb have been grown ad bulk properties measured [9]; we know of no studies of Cs₃Sb used as a dE/dx detector. Because benefits in many applications (and cost-savings) we assert that the cost and effort to grow crystals, and test them as low energy deposition detectors is an interesting addition to very low energy calorimeters.

2. Ag-O-Cs is the basis for the S-1 photocathode. It is no longer used for vacuum photoelectron detectors since

it has a noise/dark current at room temperature 4-6 orders of magnitude greater than the best bi-alkali photocathodes and 3-4 orders of magnitude larger than the Cs₃Sb photocathode (S-11). We propose turning that bug into a feature! Ag-O-Cs has the lowest work-function or pair energy of any other material, as low as 0.4 eV [10],[11]. A thick tile cooled to <1K° could be a superior Dark Matter Detector, capable of generating ~100 electrons for ~50 eV deposited. Deposition on Si may lower it further by band bending or by the electrostatic potential induced by protecting it with an atomic layer of BN and perhaps by graphene. The structure of Ag₂O amenable to incorporation of Cs. The basic reaction forming an Ag-O-Cs cathode is represented by Ag₂O+2Cs >- 2Ag+Cs₂O.The silver atoms remain in the lattice and are essential for the energetics in the energy bands The standard formation processes of Ag-O-Cs photocathodes are described for photocathode formation[12]; in this application, there is no need to preserve semi-transparency.



Fig 2: (L) Dark current measurements or pnotocathodes[PMT handbook] – Note the large dark current from Ag-O-Cs from low workfunction. (Right) Relative changes in photoemission (curve I) and thermionic emission(curve II) (arbitrary units) during the reaction of silver oxide with Cs vs the Cs:O ratio[13] – the material is optimized when the Cs:O ratio is 2.0.

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