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Bright and ultra-fast scintillation from a semiconductor?

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ABSTRACT

Semiconductor scintillators are worth studying because they include both the highest luminosities and shortest decay times of all known scintillators. Moreover, many semiconductors have the heaviest stable elements (Tl, Hg, Pb, and Bi) as a major constituent and a high ion pair yield that is proportional to the energy deposited. We review the scintillation properties of semiconductors activated by native defects, isoelectronic impurities, donors and acceptors with special emphasis on those that have exceptionally high luminosities (e.g. ZnO:Zn; ZnS:Ag, Cl; CdS:Ag, Cl) and those that have ultra-fast decay times (e.g. ZnO:Ga; CdS:In). We discuss underlying mechanisms that are consistent with these properties and the possibilities for achieving (1) 200,000 photons/MeV and 1% fwhm energy resolution for 662 keV gamma rays, (2) ultra-fast (ns) decay times and coincident resolving times of 30 ps fwhm for time-of-flight positron emission tomography, and (3) both a high luminosity and an ultra-fast decay time from the same scintillator at cryogenic temperatures.

1. Introduction

A surprising variety of scintillation emissions have been observed from semiconductors, and most of them are quite different than those that occur in ionic crystals. Among the brightest is ZnS:Ag, Cl which produces so much scintillation light that alpha particles can be detected by the naked eye [1]. This also demonstrates that in semiconductors slow charged particles are almost as efficient in producing scintillation light as fast charged particles. Other semiconductor scintillators (e.g. ZnO:Ga, CdS:In [2]) have decay times in the 1 ns range, as short as core-valence scintillators (e.g. CsF [3,4], BaF$_2$ [5]) and the fastest organic scintillators (e.g. EJ-232, BC-422).

In Section 2 we summarize and compare the ionization processes that occur in semiconductors and ionic crystals. In Sections 3–7 we describe scintillation from semiconductors with native defects (Section 3), isoelectronic doping (Section 4), donor–acceptor complexes (Section 5), shallow (n-type) doping (Section 6), and cryogenic excitons (Section 7). Section 8 concludes with a review of the fundamental limits to the scintillation properties of semiconductors and possible approaches for designing compositions that approach those limits.

2. Ionization and excitation processes

During an ionization event in a crystal, electrons are ejected from their energy levels in the core and valence bands to higher levels in the conduction band. The missing electrons (holes) in the valence bands rapidly rise to the top of the uppermost valence band and the excited electrons rapidly drop to the bottom of the conduction band. Typically half of the deposited energy is converted to heat during this thermalization process. In a few crystals (e.g. CsF, BaF$_2$) the energy difference between an upper core band and the valence band is less than the band gap, resulting in an ultra-fast core-valence scintillation component.

In semiconductor crystals these charge carriers will be spatially diffuse until they recombine or are trapped on defects. Room temperature scintillation is based on doping with a combination of donor, acceptor, or isoelectronic impurities. At cryogenic temperatures (typically below 20 K) the charge carriers can remain spatially diffuse and combine to form free excitons or defect-trapped excitons. In direct-gap semiconductors this cryogenic scintillation can be bright and ultra-fast.

In pure ionic crystals it is energetically favorable for the hole to localize because the energy to be gained by lattice relaxation is greater than the energy required to localize the hole charge. The result is called a self-trapped hole that forms rapidly during thermalization. The self-trapped holes can attract ionization electrons to become self-trapped excitons that are usually quenched at room temperature [6,7]. The unpaired spin associated with the hole and the diffuse electron spin form a triplet state which has a radiative lifetime of typically 10^{-6} s because the transition is spin

forbidden [8]. It is interesting that BaF_2 has both a core-valence emission and a useful self-trapped exciton emission at room temperature. In ionic crystals that contain a luminescent ion as a major constituent (e.g. $Bi_4Ge_3O_{12}$) the charge carriers combine to excite that ion. Alternatively, a native defect (vacancy, interstitial, antisite) can trap the carriers and become a exciton (e.g. ZnO:Zn). Most inorganic scintillators in common use are ionic crystals doped with a luminescent ion (e.g. Tl^+, Eu^{2+}, Ce^{3+}, and Pr^{3+}) that is excited by efficient trapping of the charge carriers. The luminescent ions Eu^{2+}, Ce^{3+}, Pr^{3+} have a single 4f electron in the ground state that is excited to the 5d level. The 5d–4f radiative transition is an allowed dipole transition but the decay times are limited by the poor spatial overlap between the d and f wavefunctions.

3. Bright scintillation from a semiconductor with a native defect

ZnO: Zn is an example of a bright scintillator where the emission center is a native defect. It is sold as a commercial phosphor (P15 or P24) and can be produced by adding approximately 100 ppm Zn metal to ZnO or by annealing ZnO in hydrogen to produce oxygen vacancies. The bright green emission is centered at 510 nm (2.43 eV), 0.94 eV less than the band gap of 3.37 eV. The decay time is several microseconds. Vanheusden et al. used EPR and PL measurements to determine that the emission mechanism was due to the radiative transition of an electron bound to the vacancy (i.e. a singly ionized oxygen vacancy) to a hole in the valence band produced by the excitation [9,10].

4. Scintillation from semiconductors with isoelectronic impurities

By replacing an element with another that has the same electronic structure states can be introduced into the gap. If the replacement is more electropositive (or electronegative) it can trap holes (or electrons) and be a center for radiative recombination with the other carrier [11].

4.1. Semiconductors doped with isoelectronic hole traps

CdS:Te and ZnSe:Te are examples of commercially available scintillators that use tellurium as an isoelectronic hole trap [12,13]. Sulfur and selenium have very similar electronegativities (2.58 and 2.55 eV, respectively). Tellurium has an electronegativity of 2.1 eV and acts as a hole trap when it replaces one of the former.

CdS:Te has a broad emission spectrum with a maximum at 640 nm (1.94 eV), 0.55 eV from the 2.49 eV band edge and the main decay components are 270 ns and 3.0 µs [14]. At room temperature configurational thermal quenching reduces the luminosity [15].

4.2. Semiconductors doped with isoelectronic electron traps

ZnSe:O and ZnTe:O are examples of scintillators with an isoelectronic impurity that traps electrons.

ZnSe:O is a scintillator with a peak emission wavelength of 595 nm (2.08 eV), 0.62 eV less than the band gap of 2.70 eV [16]. Its scintillation luminosity is 1.28 times that of CsI:Tl and there are two decay times of 4 µs and 12 µs [17]. A small crystal exhibited a pulse height resolution of 7.4% fwhm for 662 keV gamma rays [17].

ZnTe:O is a scintillator with a peak emission wavelength at 680 nm (1.82 eV), 0.44 eV below the band gap of 2.26 eV [18–20].

Our calculations show that oxygen on a tellurium site introduces an empty donor state about 0.4 eV below the conduction band minimum. Ionization electrons trap on those donor sites and then attract ionization holes to form excitons that have long decay times.

5. Bright scintillation from semiconductors with donor–acceptor complexes

ZnS:Ag, Cl is a commercial phosphor (P11) with an emission peak at 450 nm (2.76 eV), 0.78 eV from the band edge at 3.54 eV. Many combinations of dopants and codopants have been reported, but the highest luminosity is achieved by using Ag (an acceptor) as the primary dopant and Cl (a donor) as the codopant. It is a well-studied phosphor and the high luminosity is attributed to a donor–acceptor complex that is very efficient in trapping ionization electrons and holes [21–25]. Our measurements show that 1) the rise time is sub-ns, 2) the decay is complex with components as long as 100 µs, and 3) the luminosity is about six times higher than $YAlO_3$:Ce phosphor powder. In spite of the rapid and efficient production of the excited states, the donor electron/acceptor hole spatial overlap is poor and the decay time is long.

CdS:Ag, Cl is a similar phosphor with a high luminosity and an emission peak at 620 nm (2.0 eV), 0.5 eV from the band edge at 2.5 eV [26]. CdS:Ag, Cl can be blended with ZnS:Ag, Cl in any ratio to adjust the emission wavelength from 450 to 620 nm. We have found that the rise and decay times are similar to those of ZnS (Ag, Cl).

6. Ultra-fast scintillation from semiconductors with shallow (n-type) donor impurities

6.1. n-type ZnO

ZnO:Ga has been long known as an ultra-fast near-band-edge scintillator [2,27]. The gallium donor ion on a zinc site is charge compensated with electrons whose energy is close to the bottom of the conduction band. Above the Mott critical density of about 7×10^{19} cm^{-3} (0.17%) the donor electrons from a conducting band as their wavefunctions overlap [28,29]. The highest luminosity of ZnO:Ga occurs at gallium concentrations above 0.05% and when hydrogen is used in the synthesis, which is consistent with the notion that H^+ acts as a hole-trapping acceptor and radiative center (e.g. ZnO:Ga, H) [30,31]. ZnO:Sc has also been found to be an ultra-fast scintillator [32].

6.2. n-type CdS

CdS:In is an ultra-fast near-band-edge scintillator where indium is the donor ion [2,33]. When indium is used to provide donor electrons and tellurium is used as an isoelectronic hole trap (CdS:In, Te), fast emission occurs near the emission wavelength of CdS:Te with a decay time of 3.3 ns [34,35]. This shows that delocalized electrons that are not in a definite spin state relative to the tellurium-bound hole can increase the radiative recombination rate by more than 100-fold.

7. Scintillation from semiconductors at cryogenic temperatures

In this section we describe semiconductors at cryogenic temperatures. Semiconductors have an advantage over ionic crystals in that ionization holes do not spontaneously self-trap by lattice deformation [6,7] but can bind with ionization electrons to form

free excitons and those in turn can bind to defects such as donors, acceptors, or isoelectronic ions [36]. Their radiative recombination can produce efficient near band edge scintillation that can also be ultra-fast in the case of direct gap semiconductors. Cryogenic temperatures are required for high luminosity to reduce thermal broadening and overlap between the absorption and emission lines and because the binding energies are tens of meV and higher temperatures result in dissociation and trapping on native defects, which are almost always non-radiative.

7.1. Undoped ZnO

ZnO has a density of 5.606 g/cm^3, a band gap of 3.3 eV, and a free exciton dissociation energy of 30 meV. Wilkinson et al. measured two ultra-fast emissions from undoped ZnO at 16 K, the free exciton and a neutral donor-bound exciton. Their decay times were 400 ps and 50 ps, respectively [37].

7.2. Undoped PbI_2

PbI_2 has a density of 6.16 g/cm^3, a band gap of 2.55 eV, and a free exciton dissociation energy of 73 meV [38]. At cryogenic temperatures emission arises both from the free and bound excitons. [38–40] and from native donors and acceptors [41–43]. At 14 K the scintillation luminosity is 40,000 photons/MeV, the emission peak wavelength is 520 nm (0.18 eV below the band edge), and the decay spectrum is complex, with 22% of the light in the first 3 ns [33,44,45]. Although some purification occurred during the crystal growth process, sample-to-sample variations in luminosity indicated that the luminosity could be improved with further purification.

7.3. Undoped HgI_2

HgI_2 has a density of 6.28 g/cm^3, a band gap of 2.37 eV, and a free exciton dissociation energy of 35 meV [46]. At 14 K an unpurified sample showed a luminosity of 13,000 photons/MeV and a 2 ns decay time [33,44].

7.4. Undoped TlBr

TlBr has a density of 7.557 g/cm^3, a direct band gap of 3.016 eV, an indirect band gap of 2.68 eV, and a free exciton dissociation energy of 6.5 meV [Grabner1971]. Both indirect and weaker direct electron-hole recombination was measured at 8.5 K [47]. Purified crystals show good luminosity at 10 K but the decay time is long ($\sim$1 ms) because the radiative transition is indirect. Undoped, Pb-doped and Se-doped crystals showed emission peaks at 2.75, 2.22, and 1.87 eV, respectively [48].

8. Future directions for improved semiconductor scintillators

In this section we describe several strategies for designing semiconductor scintillators with combinations of stopping power, luminosity, and decay time that offer substantial improvements over currently available scintillators.

8.1. Gamma-ray spectroscopy

Improved scintillators for gamma ray spectroscopy requires a combination of high atomic number, high density, and high light output at room temperature. A semiconductor with a band gap of 2.5 eV and one electron-hole pair per 5 eV of ionization energy will produce 124,000 electron-hole pairs at 662 keV. If only 55,000 of these are converted into photoelectrons in the photodetector

and the response is proportional, the statistical energy resolution would be 1% fwhm.

CdS:Ag, Cl has a density of 4.8 g/cm^3, an effective atomic number similar to that of NaI and a density 31% higher. Its proportionality and luminosity combined with the high quantum efficiency of silicon photodetectors make possible an energy resolution that could approach 1% fwhm for 662 keV gamma rays. The primary limitation is the long decay time, which would limit the maximum detection rate to several kHz. Commercial crystal growth would be similar to that of CdS:Te [14].

TlBr has a density of 7.56 g/cm^3 and a gamma-ray stopping power close to that of $Bi_4Ge_3O_{12}$. Since Pb and Se have been shown to be donors and acceptors in TlBr [48], doping with a PbSe donor–acceptor complex might result in a high luminosity analogous to ZnS(Ag,Cl). The decay time is also expected to be as long, which will limit the maximum event rate. Crystals of KRS-5 (thallium bromoiodide) are grown commercially in large sizes for far infra-red windows.

One promising possibility for fast, high luminosity, room-temperature scintillation is a heavy-atom semiconductor that can be 1) doped with a donor (or acceptor) to provide a built-in population of delocalized electrons (or holes), and 2) doped with a second impurity to efficiently trap and localize carriers of the other sign [49]. It is important that the concentration of deloca-lized carriers be high enough for efficient radiative recombination but low enough to limit non-radiative recombination by Auger excitation [50–52]. It is also important that the localized trapping by the second impurity results in a lattice relaxation and Stokes shift that is large enough to avoid self-absorption but low enough to avoid configurational thermal quenching. Note that CdS:In, Te meets all these requirements except one: its luminosity is reduced by thermal quenching at room temperature [15].

8.2. Time-of-flight positron emission tomography

In positron tomography the spatial distribution of positron annihilation points is imaged in 3D. It is used for the non-invasive imaging of radiotracers in nuclear medicine to study disease in the brain and the heart, and whole-body imaging is used to detect metastatic tumors. Good image resolution requires that the annihilation photons are stopped in a small detector volume so high density and atomic number are important. If the two back-to-back 511 keV annihilation photons can be detected with good timing resolution, the annihilation point can be determined in 3D. A timing uncertainty of 30 ps corresponds to a spatial uncertainty of 4.5 mm along the line of flight.

One excellent candidate would be a heavy-atom direct-gap semiconductor at cryogenic temperatures. With suitable purifica-tion and doping essentially all the electron-hole pairs will form weakly bound excitons with fast ($\sim$1 ns), efficient radiative recombination. An annihilation photon will produce about 100,000 electron-hole pairs in a semiconductor with a band gap of 2.5 eV. Depending on the overlap between the emission and absorption spectra some photons will be delayed, but an initial intensity of only 1000 photoelectrons/ns is needed in the photo-detector to provide a timing precision of 30 ps fwhm [53].

A good room-temperature candidate is one previously described in Section 8.1: a heavy-atom semiconductor with donor (or acceptor) doping to provide a built-in population of delocalized electrons (or holes) and a second impurity to efficiently trap carriers of the other sign (e.g. heavy-atom analogs of ZnO:Ga, H or CdS:In, Te). In this case some non-radiative processes such as Auger and thermal quenching can be tolerated since an ultra-fast scintillator with a modest light yield can still provide excellent timing accuracy. It is important that the impurity bound exciton have sufficient Stokes shift to avoid unacceptable self absorption

and that other carrier trapping impurities are at a sufficiently low concentration.

Note that CdS:In, Te meets all these requirements except one: the photoelectric fraction is too low for efficient full-energy absorption in small crystals [54].

8.3. Fast timing at room temperature

While the decay time of many scintillators can be decreased by quenching (e.g. thermal quenching, impurity quenching) this also decreases the efficiency and does not improve the intensity in terms of photons/ns. The three classes of room-temperature scintillators whose ultra-fast emissions are fundamental and not the result of quenching are: organic, core-valence, and n-type semiconductor. These all have one thing in common- a population of delocalized electrons in the ground state. In the case of organic scintillators the pi orbital electrons are delocalized. In the case of core-valence scintillators the valence band electrons are delocalized. In the case of n-type (or p-type) scintillators the donor (or acceptor) carriers are delocalized. This suggests that because delocalized electrons are not in well-defined atomic quantum states their transitions are not quantum forbidden.

8.4. Ultra-bright, ultra-fast scintillation at cryogenic temperatures

As presented in Section 7, direct-gap semiconductors can be both bright and ultra-fast at cryogenic temperatures. Apparently the combination of purification and low temperatures reduces many of the carrier trapping processes that occur at room temperature. This is a promising direction for applications that are compatible with cryogenic temperatures.

9. Conclusions

Semiconductor scintillators with proportional luminosities near 200,000 photons/MeV have been demonstrated using donor–acceptor complexes. Challenges for the future are finding combinations of (1) host semiconductors that can be grown as large, clear crystals and (2) donor–acceptor complexes that are not only efficient in trapping ionization carriers but also that have good spatial overlap and acceptable decay times. Since semiconductor scintillators are highly proportional, a photoelectron yield of 55,000 should result in an energy resolution of 1% fwhm.

Semiconductor scintillators with ns decay times and reasonable luminosities have been demonstrated using n-type doping. Challenges for the future are in finding combinations of (1) host semiconductors that can be grown as large, clear crystals, (2) shallow donors (or acceptors) to provide a built-in supply of delocalized carriers, and (3) acceptors (or donors) that efficiently trap ionization carriers of the other type and serve as efficient radiative centers. A scintillator able to provide an initial intensity of only 1000 photoelectrons/ns should result in a timing resolution of 30 ps fwhm.

Donor–acceptor complexes can be bright but none have been reported to be fast. n-type semiconductors can be ultra-fast but their luminosity can be limited by Auger quenching. It is possible that only bound excitons at cryogenic temperatures can provide both ultra-high luminosity and ultra-fast decay time in the same scintillator. Challenges for the future are finding combinations of (1) direct-gap host semiconductors that can be grown as large, clear, ultra-pure crystals and (2) native defects or impurities that can bind free excitons so that the radiative recombination is efficient and has sufficient Stokes shift to avoid reabsorption.

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