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Accelerated Discovery of Elpasolite Scintillators

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Motivation

- Cubic elpasolite halides (A_2BLnX_6) have demonstrated desirable scintillation properties including high luminosity, good energy resolution, excellent proportionality and the ability to discriminate gammas from capture neutrons by a pulse shape discrimination technique.
- The isotropic nature of cubic elpasolites alleviate the manufacturability challenges and facilitate the growth of large single crystals and the production of transparent ceramics with high production yield and reduced costs.

Objectives

- Develop and evaluate high Z cerium doped elpasolite halides for scintillator applications.
- Establish a structure and property correlation between host, activator and scintillation performance such as band gap, fluorescence spectrum, stokes shift and decay times.
- Identify key technical challenges for producing large size transparent ceramic scintillators.

Technical Approach

- Streamline synthesis and crystal growth of elpasolite halide compounds.
- Perform photoluminescence measurements to pre-screen material candidates.
- Exploit cation/anion substitution to adjust the bandgap, excitation/emission characteristics and enhance scintillation performance.
- Explore advanced concepts for performance enhancement.

Accomplishments

- Successfully grew 23 different elpasolite halide compounds and solid solutions. 19 elpasolite halides have shown scintillation responses.
- Performed basic material characterization for all the new compounds such as structural refinement, thermal analysis, photoluminescence and radioluminescence.
- Developed and obtained a better understanding between the host and the activator for designing new scintillators.
- Discovered a new promising sorohalide scintillator $Cs_3Gd_2Br_9:Ce^{3+}$ with optical quantum efficiency > 91% and decay time less than 35 ns.
- Demonstrated a sensitizing effect in elpasolite and sorohalide compounds.

Material Development, Synthesis, and Crystal Growth

- Cubic elpasolite (Fm-3m)

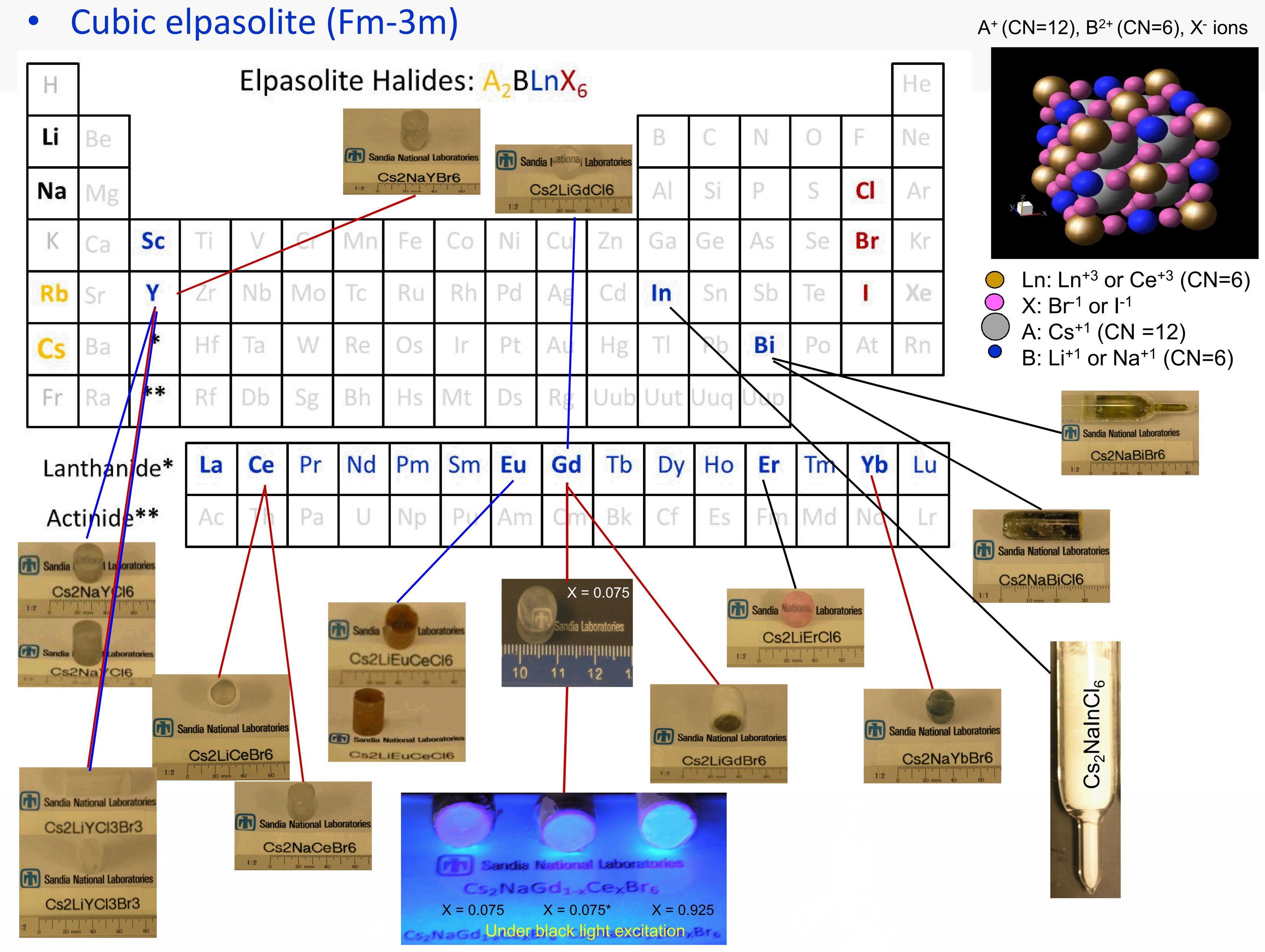

Fig. 1 Selected elpasolite halides and their single crystals grown in Sandia.

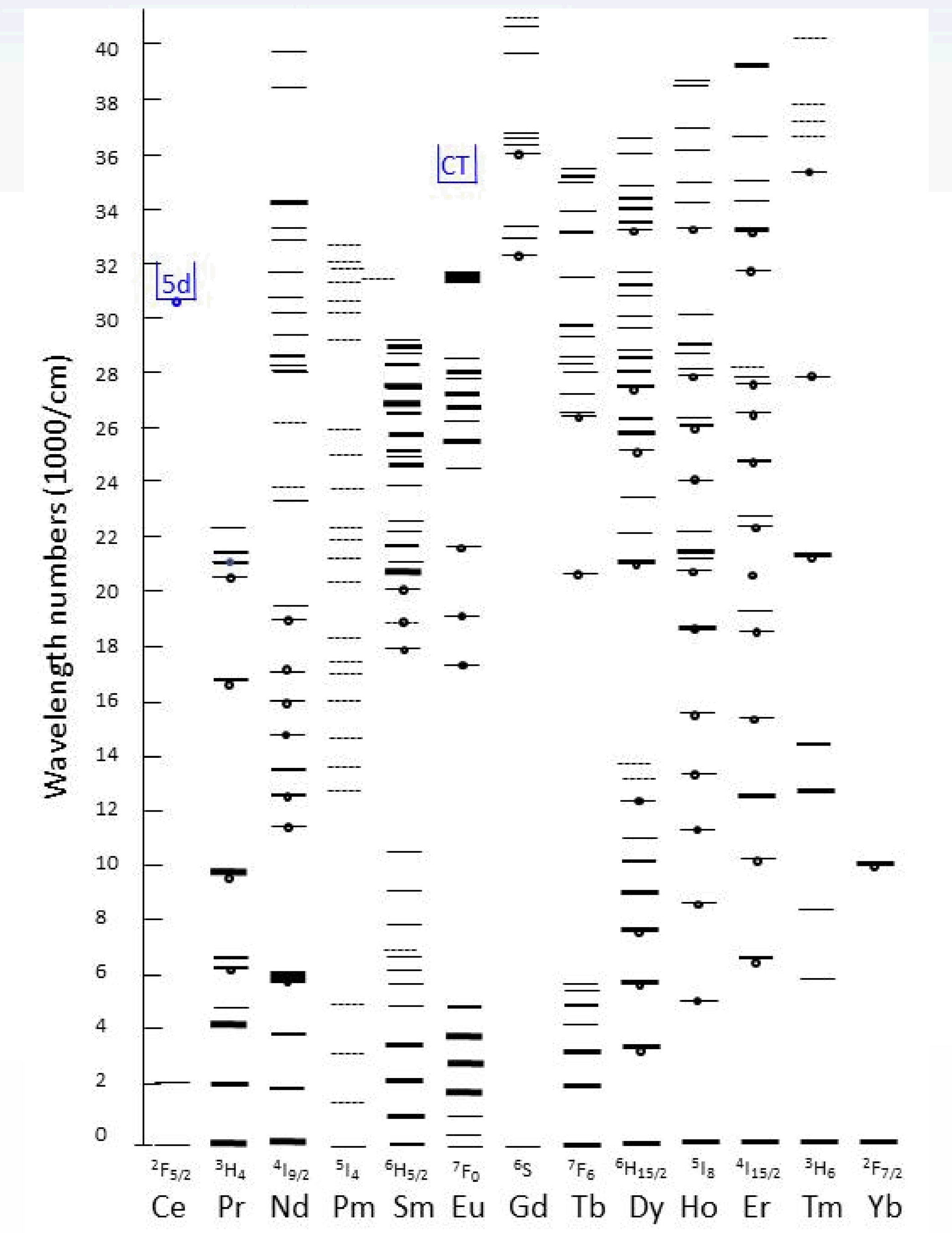

Fig. 2 Spectra and energy levels of rare earth ions in crystals

- Optical transparent single crystals of different elpasolite halides were successfully grown for structural and scintillation evaluation.
- Scintillator hosts with only a few 4f energy levels that have large energy gaps between sublevels tend to have less nonradiative interactions and are more suitable for Ce^{+3} activators (Dieke diagram Figure 2).

Photoluminescence and Radioluminescence

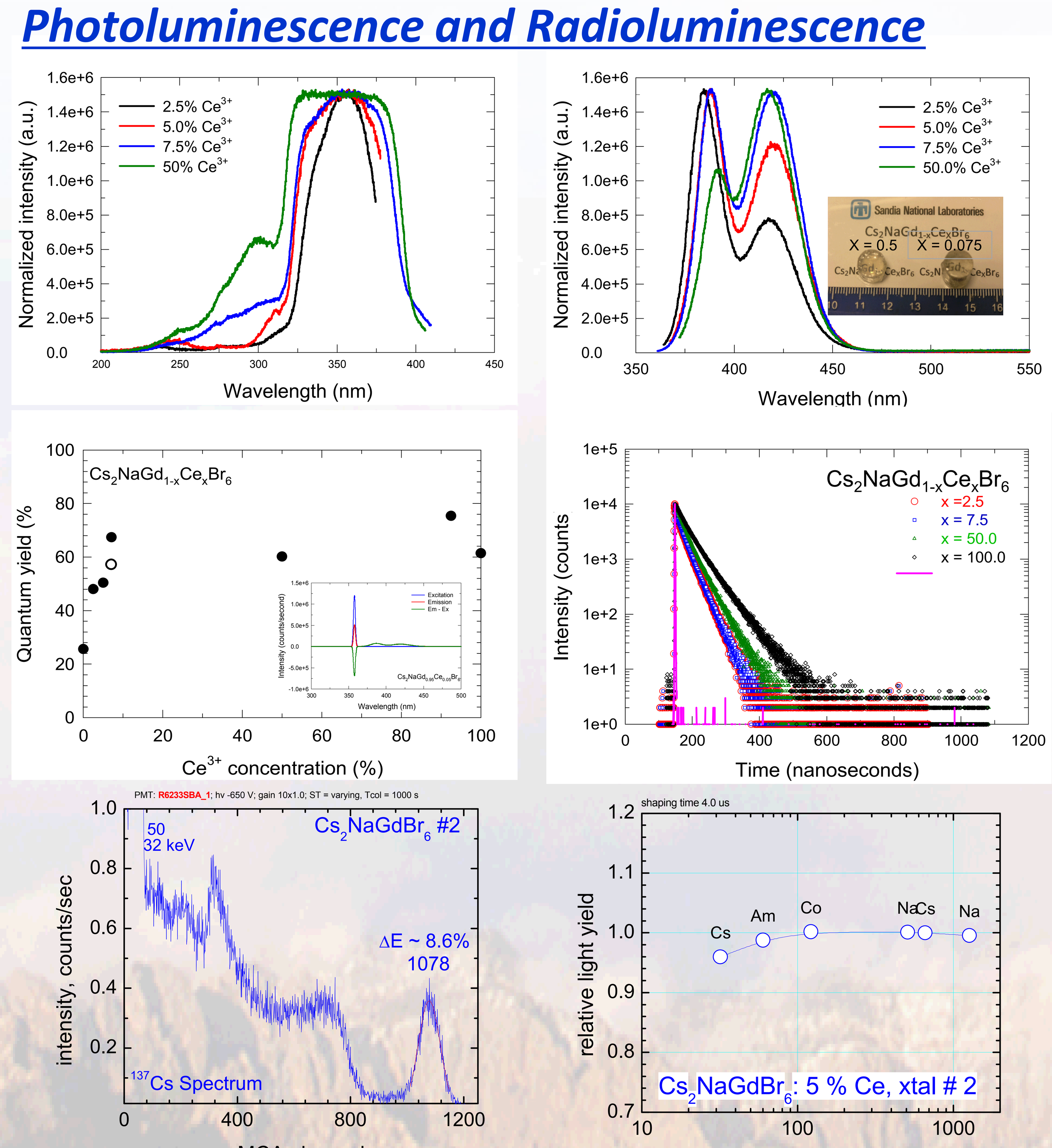

Fig.3 Photoluminescence and radioluminescence responses for $Cs_2NaGd_{1-x}CeBr_6$ solid solutions.

New Discovery – Sorohalides $Cs_3Gd_2Br_9:Ce^{3+}$

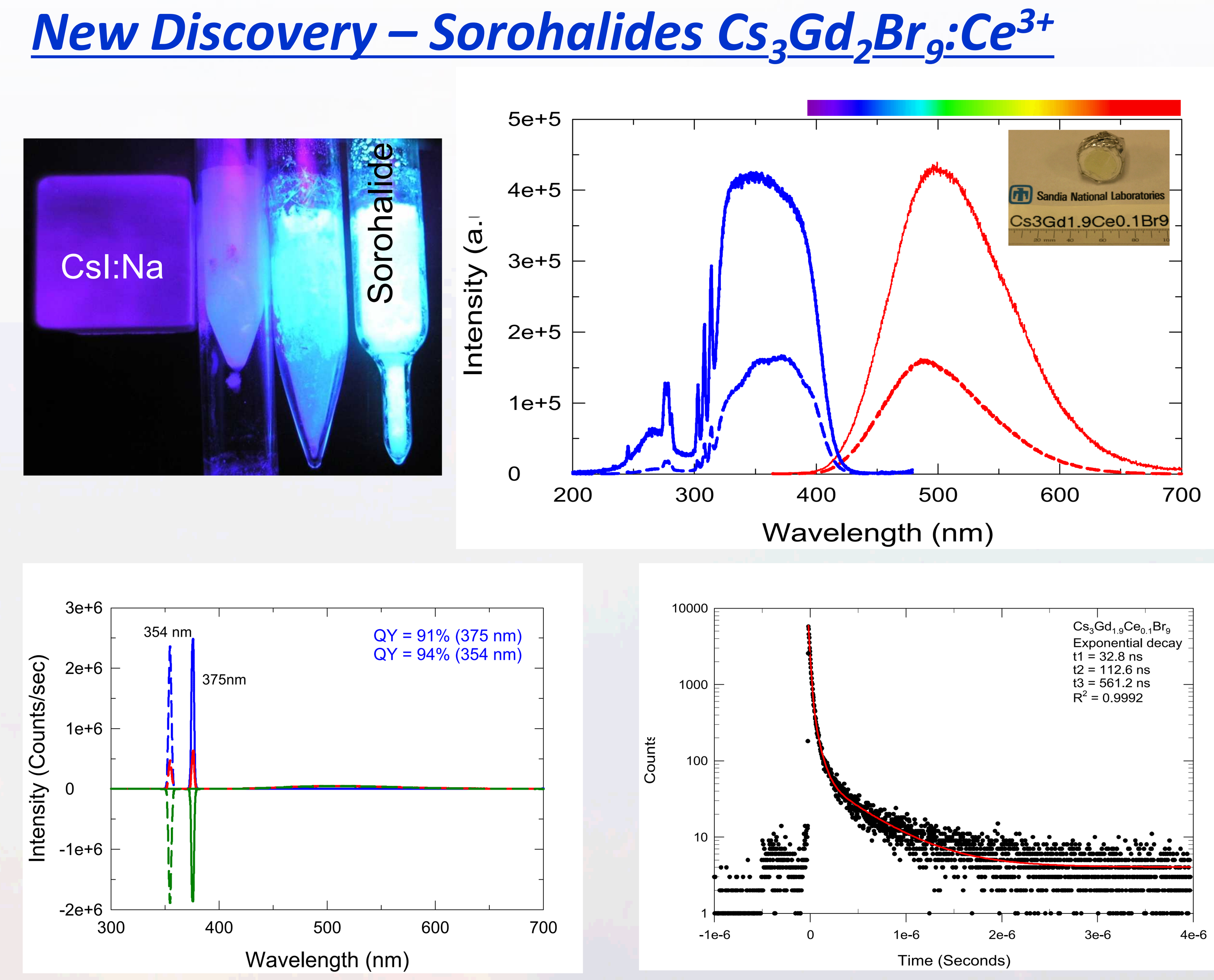

Fig.4 Photoluminescence, optical quantum yield, and decay time measurements for $Cs_3Gd_2Br_9:Ce^{3+}$.

- The new discovered sorohalide possesses excellent optical quantum yield (>91%) and a short decay time.
- The sorohalide has a large stokes shift (~100nm).
- Photon energy absorbed by the host can be effectively transferred to Ce^{3+} and enhance the light yield.

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