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Inorganic liquid hosts for mid-infrared lanthanide lasers

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Inorganic liquids such as phosphorous oxytrichloride and selenium oxychloride were very heavily researched as laser hosts many years ago. The only laser ion used was Nd³⁺, and as more convenient solid hosts such as glasses and YAG became available work on the liquids ceased. In this early work the lasers were large flashlamp pumped systems, and in this application there are further disadvantages arising from thermal expansion and high stimulated Raman cross sections, aside from the corrosion and toxicity problems in circulating systems sometimes containing litres of material.

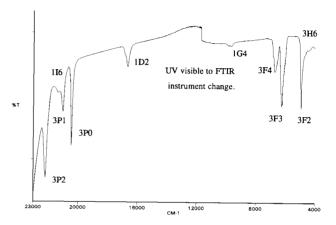
There are currently no routinely available crystalline or glass hosts with wide ranging potential for laser action beyond three microns. Aside from the obvious requirement of transparency at the pump and laser wavelengths low nonradiative relaxation rates are essential for the laser upper level. In most cases this requirement is more restrictive than that of transparency, in a solid requiring the highest phonon energy to be typically less than one-fifth of the laser transition energy. Some solvent systems, in particular phosphorous tribromide acidified with appropriate Lewis acids, provide very low maximum vibrational energies (381 cm-1 in this case) and hence should provide infrared transparency and low nonradiative rates. There is just one report in the literature on this system,1 for the 1.06 micron neodymium transition.

In modern diode pumped lasers many of the disadvantages of the liquids are mitigated. The volume of material needed is minute, the geometry inhibits convection, and unnecessary thermal load is minimised. The liquids have the great advantage that dopant levels can be varied, codopant systems explored without costly crystal growth. They can be contained in sealed cells with standard nonhydroscopic infrared transparent windows without difficulty.

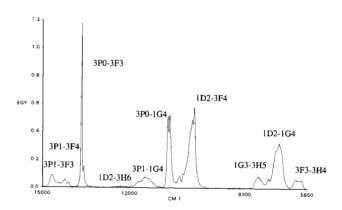
In order to realise the potential of these systems it is essential that the solvent is of very high purity. We have developed purification methods for the phosphorous tribromide, aluminum tribromide, and antimony tribromide, which yield infrared transparent, unquenched, scatter free solutions of anhydrous lanthanides.

The energy levels below 20,500 cm⁻¹, lifetimes and oscillator strengths of Pr³⁺, Nd³⁺, Sm³⁺, Dy³⁺, Ho³⁺, Er³⁺, Tm³⁺, and Yb³⁺ in this solvent system have been measured and are reported in detail. As an example in Figs. 1 and 2 we show absorption and fluorescence spectra of PrBr₃ in PBr₃, and Table 1 gives the resulting energy levels. The potential of these materials for long wavelength diode pumped lasers, based on these results, is discussed.

 S. Bondarev, et al., Soviet Journal of Quantum Electronics 6(2), 202–204 (1976).



CWC2 Fig. 1. NIR/MIR absorption spectrum of Pr3⁺ in PBr3/Al2Br6/SbBr3.



CWC2 Fig. 2. Fluorescence spectrum of Pr3⁺ in PBr3/Al2Br6/SbBr3.

CWC2 Table 1. Pr^{3+} doped $PBR_3/AlBr_3/SbBr_3$

Pr ^{III} 4f level	Energy cm ⁻¹	I_{rel}	f_{rel}	ΔE cm ⁻¹	Energy ZBLAN
³ P ₂	22174	93	1	271	22734
¹ I ₆	21500	2	_		_
³ P ₁	21010	31	0.283	246	21459
³ P ₀	20450	100	0.617	103	20877
¹ D ₂	16682	22	0.286	250	17036
¹ G ₄	9682	3	0.075	489	9709
$^{3}F_{4}$	6714	32	0.420	333	6920
³ F ₃	6272	73	0.849	230	6494
³ F ₂	4954	43	0.394	113	5128
³ H ₆	4450	1	_		4310
³ H ₅	2137	_	_		2167