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Mid infrared lasers for remote sensing applications

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ABSTRACT

To accurately measure the concentrations of atmospheric gasses, especially the gasses with low concentrations, strong absorption features must be accessed. Each molecular species or constituent has characteristic mid-infrared absorption features by which either column content or range resolved concentrations can be measured. Because of these characteristic absorption features the mid infrared spectral region is known as the fingerprint region. However, as noted by the Decadal Survey, mid-infrared solid-state lasers needed for DIAL systems are not available. The primary reason is associated with short upper laser level lifetimes of mid infrared transitions. Energy gaps between the energy levels that produce mid-infrared laser transitions are small, promoting rapid nonradiative quenching. Non-radiative quenching is a multiphonon process, the more phonons needed, the smaller the effect. More low energy phonons are required to span an energy gap than high energy phonons. Thus, low energy phonon materials have less nonradiative quenching compared to high energy phonon materials. Common laser materials, such as oxides like YAG, are high phonon energy materials, while fluorides, chlorides and bromides are low phonon materials. Work at NASA Langley is focused on a systematic search for novel lanthanide-doped mid-infrared solid-state lasers using both quantum mechanical models (theoretical) and spectroscopy (experimental) techniques. Only the best candidates are chosen for laser studies. The capabilities of modeling materials, experimental challenges, material properties, spectroscopy, and prospects for lanthanide-doped mid-infrared solid-state laser devices will be presented.

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1. Introduction

Mid infrared (MIR) differential absorption lidar (DIAL) systems can provide vital data needed by atmospheric scientists to understand atmospheric chemistry. The Decadal Survey [1] recommended missions to measure atmospheric constituents including CO₂, CH₄, CO, O₃, NO₂, SO₂, and CH₂O. All of the atmospheric constituents noted above have unique and useful characteristic absorption features in the mid infrared, as shown in Table 1 where significant absorption of a constituent exists. In addition, the characteristic absorption can be found in spectral windows where the atmospheric transmission is relatively good, as shown in Fig. 1. Thus, when developed, mid infrared lasers will be able to measure all of the atmospheric constituents requested by the Decadal Survey.

While there are a variety of choices for MIR laser, each has various tradeoffs and the application to specific measurements must be considered as well [2]. Semiconductor Lasers (Lead-Salt, Quantum Cascade and Antimonide III–V) are possibilities, but may require

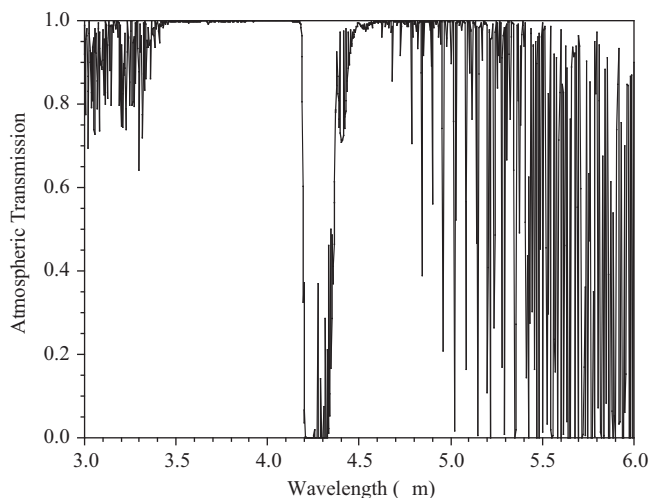
cryogenic cooling and have highly divergent, astigmatic beams. Solid State lasers, such as Cr²⁺ II–VI lasers, are generally low gain materials, resulting in low power/energy. Parametric frequency conversion, such as optical parametric oscillator (OPO) and difference frequency generation (DFG) suffer from a variety of drawbacks including phase matching, design complexity, alignment sensitivity, and laser induced damage (LID). Laser pumped Ln³⁺ (lanthanide) lasers offer a main advantage of pump energy storage, as well as high power or energy output in a narrow spectral bandwidth and diffraction limited beam, which are some specific reasons to select them over other technologies for some remote sensing applications. A wide variety of pulse widths and pulse repetition frequencies (from cw to 1 GHz) can also be achieved. Ln³⁺ lasers, in particular, offer broad tunability, efficient operation via direct diode pumping, a variety of pulse lengths and repetition frequencies in pulsed mode, and good beam quality. Operating in the MIR the Ln³⁺ lasers can access atmospheric transmission windows in the ~3–5 μm and 7–10 μm regions. The ~5–7 μm region is not so useful due to the presence of high water vapor absorption. Some explanation should be made regarding the meaning of mid infrared, which can be rather ambiguous. Generally it is used to encompass the 3–8 μm region, which comprises the mid wavelength infrared (MWIR) and long

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Table 1
Atmospheric constituents identified by characteristic absorption features.

Molecular species	Wavelength windows (μm)		
	3.00–4.00	4.50–5.00	7.0–10.0
CH ₄	3.15–3.57		< 7.5–8.3
CO		4.50–4.87	
O ₃		4.70–4.79	9.37–9.90
NO ₂	3.40–3.85	4.45–4.70	7.50–8.10
SO ₂			7.13–7.75
CH ₂ O	3.20–3.40		

**Fig. 1.** Atmospheric transmission from 3 to 6 μm .**Table 2**
Infrared spectral ranges.

Division name	Abbreviation	Wavelength (μm)
Near infrared	NIR	0.75–1.4
Short wavelength infrared	SWIR	1.4–3
Mid wavelength infrared	MWIR	3–8
Long-wavelength infrared	LWIR	8–15
Far infrared	FIR	15–1000

wavelength infrared (LWIR) divisions, and is sometimes used at shorter wavelengths such as 2 μm . In Table 2, the various divisions of the infrared spectral ranges are given for reference.

Most lanthanide or transition metal doped solid-state materials can produce laser wavelengths in the ultraviolet (UV), visible (VIS), or short wavelength infrared (SWIR). However, it becomes increasingly difficult to generate wavelengths in the mid-wavelength infrared (MWIR) and beyond. We will use the term MIR to refer to the MWIR wavelength ranges as defined in Table 2. The great challenge in solid-state lasers for MIR operation is finding materials that offer low phonon energies. Solid-state lasers that have wavelengths longer than about 2.5 μm are usually quenched by nonradiative processes. These nonradiative processes are caused by high energy crystal lattice vibrations called phonons. When the energy gap between 2 energetically adjacent manifolds is less than 5 times the maximum phonon energy, the nonradiative transitions tend to quench the upper manifold. Hence, to develop an efficient MIR laser with a wavelength longer than ~ 2.5 μm requires a low phonon energy host material.

In general oxides have the largest phonon energies, followed by fluorides, chlorides, bromides and iodides. However, having a low phonon energy is not the only criterion in choosing a material. The

Table 3
Crystal phonon energies and optical transparency range.

Formula	Material name	Phonon energies (cm^{-1})	Transparency (μm)
YVO ₄	Yttrium vanadate	880–980	0.35–4.8
Y ₃ Al ₅ O ₁₂	Yttrium aluminum garnet	700–850	0.21–5.2
YAlO ₃	Yttrium aluminum oxide	550–600	0.20–7.0
Y ₂ O ₃	Yttrium oxide	400–600	0.29–7.1
YLiF ₄	Yttrium lithium fluoride	400–560	0.12–8.0
CaGa ₂ S ₄	Calcium gallium sulfide	350–400	0.34–12
BaY ₂ F ₈	Barium yttrium fluoride	350–415	0.20–9.5
KYF ₄	Potassium yttrium fluoride	350–400	0.15–9.0
LaF ₃	Lanthanum fluoride	300–350	0.20–10
CaF ₂	Calcium fluoride	280–330	0.20–10
LaCl ₃	Lanthanum chloride	240–260	0.30–25
KPb ₂ Cl ₅	Potassium lead chloride	200	0.30–16
LaBr ₃	Lanthanum bromide	175	0.30–20
CsCdBr ₃	Cesium cadmium bromide	150–160	0.40–25
KPb ₂ Br ₅	Potassium lead bromide	140	0.40–25

optical transparency is important as well. Table 3 lists the phonon energy and transparency range for a variety of crystal laser materials.

Clearly the oxides do not make good choices either in phonon energy or optical transparency. This leaves choices of fluorides, chlorides and bromides for materials with appealing phonon energies. Another issue to consider is the hygroscopic nature of the material, or the ability to readily absorb moisture. Fluorides tend to be weakly hygroscopic, while chlorides and bromides show varying degrees depending on the composition. For example, LaCl₃ and LaBr₃ are highly hygroscopic, while KPb₂Cl₅ and KPb₂Br₅ are only mildly hygroscopic. Prudent material choices, based on Table 3 as well as hygroscopic issues are BaY₂F₈, KYF₄, LaF₃, KPb₂Cl₅, CsCdBr₃ and KPb₂Br₅ materials.

Keeping the importance of the host material in mind we will proceed in a systematic way, discussing the lanthanide ions and manifold-to-manifold pairs which offer MIR transitions, followed by material selection and laser prospects. Some emission spectroscopy results for praseodymium (Pr) in the 3–6 μm region is also presented.

2. Materials and methods

Selection of the laser material and the specific lanthanide series atom can be made essentially independently. The choice of laser material exerts only a small influence on the 4f electrons because of the shielding of the 5s and 5p electrons. Consequently, the choice of laser material affects the energy levels and manifolds only slightly. Thus, a promising pair of manifolds in a particular laser material will have essentially the same pair of manifolds in another material. If the particular wavelength is not critical, the largest effect in the choice of laser material is the maximum phonon energy.

2.1. Laser ions and manifold pairs

Lanthanide series atoms were reviewed to identify potential manifold pairs suitable for mid infrared lasers. The qualities sought include

1. An energy gap between a pair of manifolds that generates the desired wavelength.
2. As large of an energy gap as practical between the upper and lower laser manifolds.

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