Novel Iron Doped Materials for Mid-Infrared Laser Applications

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Introduction

Recent progress in technology of Cr:ZnSe and Fe:ZnSe gain media enabled efficient lasers operating over 1.8-3.3 and 3.6-5.2 μm. Nevertheless, there is strong demand for effective gain materials for the 3.0-3.9 μm spectral range not nicely covered by Cr:ZnSe and Fe:ZnSe amplification bands. Therefore, a search of new perspective materials for laser applications in this spectral range is in a great demand. The ^5D is ground states of the Fe^{2+} ions which is split into the doublet ^5E and triplet $^5T_2$ in the crystal field. The order and energy difference between the ^5E state and the $^5T_2$ states depends on ionic valence state, bond lengths and crystal file symmetry.

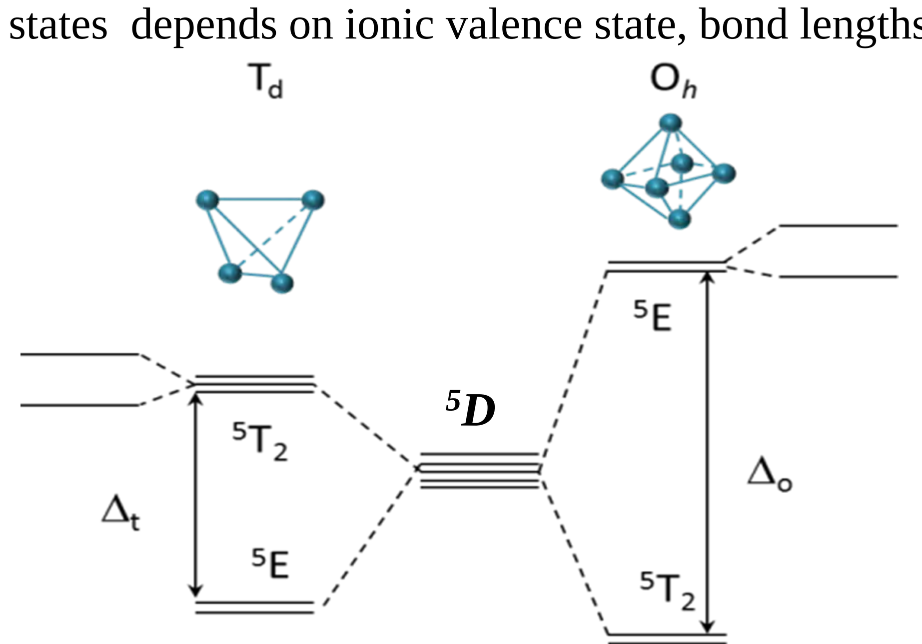


Figure 1. Energy Levels of Fe^{2+} ions in tetrahedral and a octahedral sites

The doublet ^5E is the ground state of the Fe^{2+} ions in the tetrahedral sides, while the triplet $^5T_2$ is the ground state in the octahedral sites. In addition, the crystal filed theory predicts approximately twice smaller energy splitting in tetrahedral site in comparison with octahedral site [1,2]. Therefore, crystal with tetrahedral coordination of iron ions are more promising for mid-IR applications.

In this research, we characterized $Fe:MgAl_2O_4$ (spinel), $Fe:ZnAl_2O_4$ (gahnite) and Fe:InP crystals as promising laser materials for this mid-IR spectral range.

Samples Fabrications

$MgAl_2O_4$, and $ZnAl_2O_4$ crystals are the most representative crystals from oxide spinel group with chemical formula AB_2O_4 where A and B metal ions have valence +2 and +3 correspondingly. Oxygen ions form face-centered cubic structure with 32 octahedral (O_h) and 64 tetrahedral (T_d) sites inside unit cell. In the normal spinel, 1/8 of tetrahedral sites are occupied by A^{2+} ions and half of the O_h occupied by B^{3+} ions.

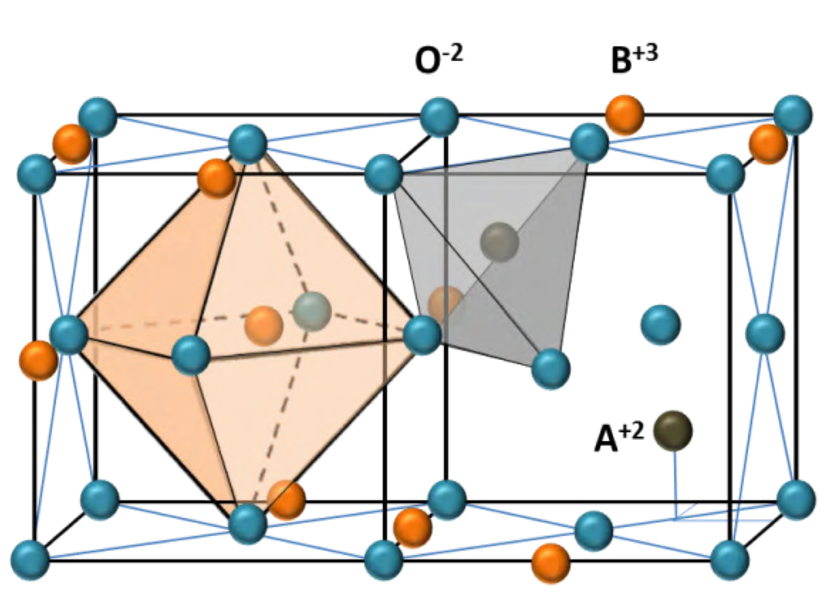
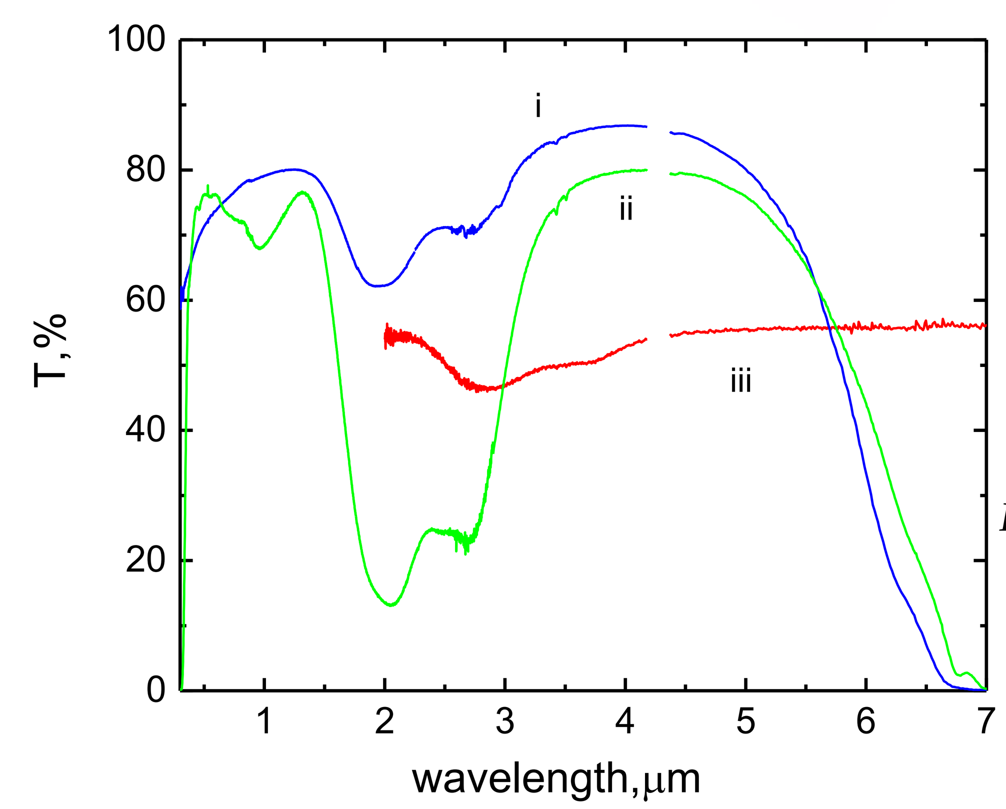


Figure 2. Crystal structure of spinel crystals

In our study we used $Fe:MgAl_2O_4$ single crystal doped by thermal diffusion methods and $FeZnAl_2O_4$ ceramic samples doped during fabrication process.
Indium phosphide (InP) crystals have zincblend crystal structure where each ion is located in a tetrahedral site consisting of four ions of the opposite type [1]. In our study we used Fe:InP single crystal doped during crystal growth process with nominal concentration of iron 10^{18} cm^{-3}.

Absorption measurements

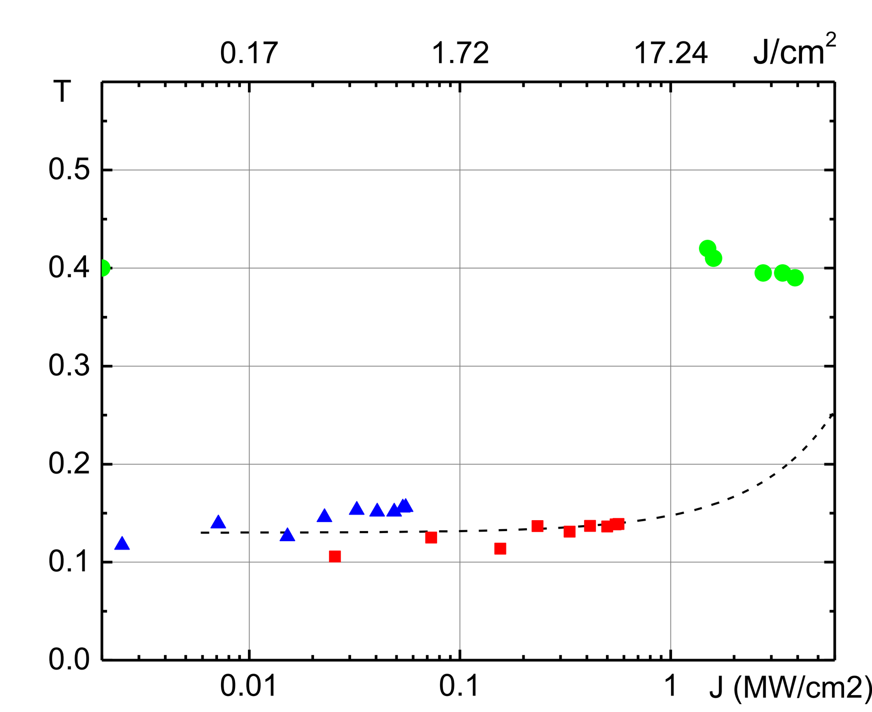


Spectroscopy Analysis:

- Visible and IR absorption spectra were measured at room temperature using Shimadzu UV-VIS-NIR-3101PC and Prestige-21 (Shimadzu) FTIR spectrometer .

Figure 3. Transmission spectra of $Fe:MgAl_2O_4$ (curve i); $Fe:ZnAl_2O_4$ (curve ii); and Fe:InP (curve iiii) samples.

Saturation of Absorption



Experimental setup:

- Saturation of the absorption in $Fe:ZnAl_2O_4$ samples was studied under excitation by radiations of Q-switched Ho:YAG @ 2.09μm and Er:YAG @ 2.9μm lasers

Figure 4. Transmission spectra of $Fe:ZnAl_2O_4$ sample vs pump energy density at 2.09 μm wavelength (circles) and 2.94μm wavelength(triangles and squares); dashed line shows theoretical dependence with upper level life time 50 ps.

Photoluminescence measurements

Experimental setup:

- The non-selective mid-IR photoluminescence of Fe^{2+} ions was measured using HgCdTe detector with temporal resolution ~80 ns.
- The long-wavelength pass optical filter with cut-off wavelength at 3.5 μm was installed before HgCdTe detector.
- The Q-switched Er:YAG @ 2.94 μm and Ho:YAG @ 2.09μm laser were used for direct excitation of the $^5E \rightarrow {}^5T_2$ transition of Fe^{2+} ions in the tetrahedral sites.
- A Q-switched Nd:YAG @ 1.064μm laser was used to study excitation of Fe^{2+} ions in the InP crystal via photoionization transition

Discussion

- In addition to broad absorption band of Fe^{2+} ions in tetrahedral sites between 2-3 μm, the absorption spectra of $Fe:ZnAl_2O_4$ ceramic samples reveal Fe^{2+} ions in octahedral sites with maximum of the absorption band around 1μm.
- The thermal diffusion of iron in $MgAl_2O_4$ single crystal could be used for fabrication of $Fe:MgAl_2O_4$ samples with ion concentration ~5x10^{19} cm^{-3}.
- The radiative life time for $^5E \leftrightarrow {}^5T_2$ transition of Fe^{2+} ions in tetrahedral sites in the $MgAl_2O_4$ crystal was estimated using following equation:

$$\tau_{rad} = \frac{1}{8\pi c n^2}\left[\frac{g_1}{g_2}\int \tilde{\nu}^2 \sigma_{abs}\, d\tilde{\nu}\right]^{-1}$$

, and was calculated to be 35 μs.
- Absence of the absorption saturation (up to pump power density ~1MW/cm^2) and absence of the mid-IR photoluminescence in $Fe:ZnAl_2O_4$ sample at room temperature, allow estimation of the nonradioactive decay time faster than 50 ps
- The photoluminescence signals in Fe:InP crystal were detected under direct excitation of Fe^{2+} ions by radiation at 2.94 μm as well as under excitation via photo-ionization transition by 1.064 μm radiation.

Conclusions and Future Work

- Fe:InP has the potential to be an effective mid-IR laser material, but would require optimization of material doping technology to increase concentration of Fe^{2+} ions to ~10^{19} cm^{-3}.
- Due to strong quenching of the mid-IR photoluminescence, $Fe:ZnAl_2O_4$ and $Fe:MgAl_2O_4$ crystals are not suitable for laser application at room temperature. The possibility of utilization these crystal should be tested at low temperature.

References

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2. V.V. Fedorov, S.B. Mirov, A. Gallian, D.V. Badikov, M.P. Frolov, Yu.V. Korostelin, V.I. Kozlovsky, A.I. Landman, Yu.P. Podmar'kov, V.A. Akimov, A.A. Voronov "3.77–5.05-mm Tunable Solid-State Lasers Based on Fe^{2+}-Doped ZnSe Crystals Operating at Low and Room Temperatures", IEEE Journal of Quantum Electronics, 42(9), 907-917 (2006).

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