**Answers (in blue) to the reviewers’ comments:**

**Reviewer: 1**

This paper reports two new results but it is not clearly explained why these results are important, rather than merely being incremental advances in an ongoing study of Pr:YLF emission by this research group. If the authors can revise their paper by presenting possible specific applications of these two results, I think the manuscript would be of broader interest.

Yes, we agree with this comment and we believe through emphasizing the application of the achieved laser that it would be much more interesting to readers. C-cut Pr:YLF makes laser emission at 696 and 719 nm realized much readily. We added the following sentences into the revised manuscript.

“In addition, actually, the lasers have great potential applications in data storage, micro-sized projectors, color display, biomedical and lithographic techniques, etc. For example, a monochromatic irradiation at 696 nm laser can be used for determining photoresolution of the Cr(III)tris-oxalato complex [11]. In addition, a 696 nm laser was also reported to play an important role in enzyme catalytic steps of protochlorophyllide (Pchlide) photoreduction [12] and analysis of Spectrophotometric Chlorophyll Equations for methanol [13]. On the other hand, the 719 nm laser is one of the absorption bands in Synechocystis [14], which is meaningful for the study of synechococcus elongates.”

References:

[11] G. L. J. A. Rikken and E. Raupach, “Enantioselective magnetochiral photochemistry” Nature 405, 932-935 (2000)

[12] Derren J. Heyes and C. Neil Hunter, “Making light work of enzyme catalysis: protochlorophyllide oxidoreductase” Trends in Biochemical Sciences 30 (11), 642-649 (2005)

[13] Raymond J. Ritchie, “Consistent Sets of Spectrophotometric Chlorophyll Equations for Acetone, Methanol and Ethanol Solvents” Photosynthesis Research 89 (1), 27-41 (2006)

[14] Lars-Olof Pålsson, Jan P. Dekker, Eberhard Schlodder, René Monshouwer and Rienk van Grondelle, “Polarized site-selective fluorescence spectroscopy of the long-wavelength emitting chlorophylls in isolated Photosystem I particles of Synechococcus elongatus” Photosynthesis Research 48(**1**), 239-246 (1996)

The first result is laser emission at 719 nm rather than 721 nm by cutting the crystal in a different orientation. But is that enough of a wavelength shift to be significant? In particular, it is not explained whether one could get shifts on the order of 2 nm by simply changing the temperature of the crystal? Since their sample is mounted in a copper block, why didn’t they use fluid flow cooling/heating of the block to investigate the temperature dependence of the emission?

In this work, we present simultaneous two-wavelength laser at 696 and 719 nm by using a common method, i.e. employing a c-cut laser gain medium instead of a-cut one which is often used to achieve π-polarized 698 and 721 nm with higher gain. In fact, in our lab we have nice water chiller and at the beginning of this experiment we did try to change the temperature of the Pr:YLF by using the chiller with temperature varying from 16 to 30oC, however, we observed that the output laser wavelength almost not shifted. Using the pump source with maximum output power of about 2 W, no obvious thermal lensing effect inside the laser crystal has been observed since a good stability and no saturation of output power.

The second result, which is the one announced in the abstract, is dual laser emission at two far red wavelengths. But no application has been suggested that requires such dual lasing. Can the authors suggest any?

On the one hand, the two wavelengths are respectively important in specific applications, which have already been depicted above. On the other hand, the dual wavelength laser could also be interesting in some applications. We added the following sentence into the revised manuscript.

“The dual-wavelength deep red laser could be interesting for THz wave generation (~13.7 THz) by difference frequency or UV generation (~353.5 nm) by sum-frequency mixing with the aid of nonlinear crystals.”

The title should not start with an acronym. Instead “LD” should be spelled out. For that matter, is it important that the laser is diode pumped? (If so, why?) Isn’t it more important to specify in the title that the laser is cw?

We now revise the title into “Diode-pumped continuous-wave dual-wavelength c-cut Pr3+:LiYF4 laser at 696 nm and 719 nm”, which covers the two points including the full spelling of “LD” and “cw” mode of the laser as the reviewer suggested. The reason for using laser diode (LD) to pump the Pr:YLF crystal is that at first LDs are cheap, compact and robust laser sources for various applications including using as pump source. Second, at present, we admit that there existing some other laser source for pumping Praseodymium laser materials, such as optically pumped semiconductor laser (OPSL) [1] or particularly made solid state laser by second-harmonic generation of 0.94 μm Nd3+ lasers [2]. However, LDs are still the most popular ones for pumping laser materials, such laser system is just we often call diode-pumped all-solid-state lasers (DPASSLs).

References:

[1] A. Richter, E. Heumann, and G. Huber, “Power scaling of semiconductor laser pumped Praseodymium-lasers,” Opt. Express 15 (8), 5172 (2007)

[2] B. Xu, P. Camy, J. L. Doualan, Z. P. Cai, and R. Moncorgé, “Visible laser operation of Pr3+-doped fluoride crystals pumped by a 469 nm blue laser,” Opt. Express 19 (2), 1191 (2011)

The abstract refers to “beam propagation factors” whereas the end of the introduction calls them“beam quality factors.” Be consistent.

We unified them as “beam propagation factors” in the revised manuscript.

I don’t see the advantage of adding mirror M1 to the setup and going through the analysis associated with Eqs. (2) and (3). Isn’t it easier and conceptually simpler to measure the spectra (as in Fig. 4) at different pump powers and directly compare the area under one peak to the area under the other peak to determine the fraction of the total laser power output at each wavelength?

Here, by using mirror M1 to have an idea about the individual output power of the dual-wavelength laser is absolutely correct and effective method. There must be some other methods to do it and we are also not interested in comparing these method since it’s not our point in this work. We feel that it could be interesting to measure the output power by using the suggested method by the reviewer; however, we are also wondering that whether the method can provide accurate values about the two powers of the lasers first because the areas under the peaks are not so easy to calculate and second because the areas are much dependent on the measurement of the spectra during the experiments.

The transmissivity of the output coupler is given at the two emission wavelengths. But what is its reflectivity at the pump wavelengths?

The output coupler has a high transmission (>92%) at the pump wavelengths. We added the information into the revised manuscript.

The Pr:YLF crystal is reported to have a concentration of 0.5 at.% and length of 1 cm. Why were these two particular values chosen and was any attempt made to vary them to optimize the laser output power or slope efficiency?

First, a relatively low concentration as 0.5 at.% was chosen to decrease possible non-radiative cross-relaxation processes [3]. Second, since the concentration was already determined, the length of the laser crystal can then be decided to be about 10mm in our work to assure enough absorption of the pump power. However, in fact, we also wondered whether the long crystal could introduce some additional losses, which, on the contrary, degrades the laser performance. Theoretical optimization will be done in the near future in our group, but not presented in this work.

References:

[3] T. Gun, P. Metz and G. Huber, “Power scaling of laser diode pumped Pr3+:LiYF4 cw lasers: efficient laser operation at 522.6 nm, 545.9 nm, 607.2 nm, and 639.5 nm”, Opt. Lett. 36 (6), 1002–1004 (2011)

A reference should be given to explain why τ ≈ 49 µs is a reasonable value.

The following reference gives an fluorescence lifetime to be 38µs for 0.5% doped Pr:YLF. We used 0.15% as the round-trip loss during the calculation because it shows good agreement with our experimental results of the laser threshold.

In the revised manuscript, we wrote the following sentences to make it clear.

“To fit our laser results, L1≈L2≈0.15% and τ≈38 µs [16] were taken into Eq. (1), and thus we can simply obtain Pth,1= 589.0 mW and Pth,2= 570.2 mW.”

References:

[16] K. Hashimoto and F. Kannari, “High-power GaN diode-pumped continuous wave Pr 3+-doped LiYF4 laser” Optics Letters 32 (17), 2493-2495 (2007)

I would prefer the horizontal axis of Fig. 6 to be in minutes rather than seconds, to make it clearer that the stability was measured over about a half-hour period.

Yes, we absolutely agree with this comment. We now substitute the *x*-axis with minutes rather than seconds in Fig. 6 (now revised as Fig. 7).

I am not sure it is necessary to superscript the authors’ names with “1” given that all of them have the same address.

We thank the reviewer for his/her so careful checking of our submitted manuscript. Yes, it is not necessary to put “1” as superscript for the authors. We therefore deleted these “1” in the revised manuscript.

**Reviewer: 2**

The scheme of the Pr levels has to be presented in the paper.

We thank the reviewer for his/her advice, the energy level scheme of Pr:YLF including the 696 and 719 nm lines is now presented in the revised manuscript as Fig. 1.

Following to the paper the laser threshold is 552 mW. From the black line (Ptotal) in Fig. 3 it looks like the laser threshold is ~800mW. Need to clarify.

The ~800mW (absorbed pump power, Pabs) is not the laser threshold because at 800 mW the laser output power was already up to 16.5 mW. We insist the laser threshold to be about 552 mW, at which the laser output power was just a little bit above zero. We therefore clarified the threshold by adding the following sentences in the revised manuscript.

“According to the measurement of the total output power, laser behavior was observed at absorbed pump power of about 552 mW corresponding to laser threshold. In addition, from 552 mW to 800 mW, the output powers increase almost linearly to about 16.5 mW. When the absorbed pump power was above 800 mW, it can be seen from Fig. 4 that the total output powers increased sharply than before. Therefore, the output powers curve took on an obvious turning point in Fig. 4.”

The radius of the pump beam was measured, but the radius of the laser emission beam was calculated. Why? Need to clarify too.

The laser beam size was calculated according to a standard ABCD law, which assumes a TEM00 mode inside the laser cavity, namely diffraction-limited beam. In our experiment, the generated laser had good beam quality (near diffraction-limited) and therefore we directly use the calculated laser beam size to estimate the laser threshold. In fact, according to the following formula



the laser beam waist diameter 2*w*0 inside the laser crystal can be easily deduced. Here, *M*2 is the output beam propagation factor, *wz* the measured output beam size, *z* the distance between the measurement location and the laser medium. Using this formula, the laser waist size was about 56.6 μm (radius), which is approximately equal to the calculated value of 55 μm.

In the revised manuscript, we clarify this issue by the following sentences.

“The output laser should have better beam quality than the measured values because additional optics were used during the measurement of the M2 factors, which more or less degraded the output beam quality. Anyway, the M2 factors show that the output laser had a near diffraction-limited beam quality. Therefore, during the calculation of the laser threshold, the calculated laser beam size (55 μm) based on ABCD law seems reasonable. In fact, using the following formula



where *wz* is the measured output beam size, *z* the distance between the measurement location and the laser crystal, *w*0 the laser beam waist radius inside the laser crystal. The measured *w*0 can be yielded to be about 62 μm showing good agreement with the calculated value.”

The authors use transitions 3P0 -> 3F3 and 3P0 –> 3F4 for their dual-wavelength laser. Can other transitions in the Pr ions be used for the dual-wavelength generation?

As we know, to operate dual- or multi-wavelength laser, it is important to produce a balanced gain for these potential lasing wavelengths. In fact, at present, in Pr3+ ions, multiple wavelength lasers have already been achieved in (696, 698nm) lines corresponding to transition 3P0 -> 3F3 [4].

References:

[4]B. Xu, Y. J. Cheng, B. Qu, S. Y. Luo, H. Y. Xu, Z. P. Cai, P. Camy, J. L. Doualan and R. Moncorge, “InGaN-LD-pumped Pr3+:LiYF4 continuous-wave deep red lasers at 697.6 and 695.8 nm”, Opt. & Laser Tech. 67, 146–149 (2015).