Dear Editor Noriaki Horiuchi,

Thank you very much for handling our manuscript. We appreciate reviewer #1's recommendation for publication. We are sincerely grateful for all the reviewers' comments, especially for a very special case of the alexandrite ($Cr^{3+}:BeAl_2O_4$) laser with high-temperature enhanced performances raised by reviewer #2. Obviously, reviewer #2 accepted our work in general, whose only concern was the novelty of our work, see "*This laser mechanism, ..., is in fact not a new mechanism but has actually been known to occur in other laser materials such as in alexandrite lasers since the 1980s (see references [1-3] below).*" As you will see, we respectfully but strongly disagree with this point.

Exactly, we studied and carried out experiments on alexandrite lasers (see Fig. 1&2). We clearly know the mechanism of our photon-phonon collaboratively pumped (PPCP) laser is **fundamentally different** from that of alexandrite one.

1. Different lasing mechanism

Alexandrite laser is only a particular case in Cr^{3+}-doped laser materials, possessing very distinctive electronic levels where two close $^4T_2$ and ^2E upper levels are generated by crystal-field Stark splitting. Its laser temperature enhancement originates from the temperature-dependent Boltzmann distribution in these two levels, whose population can be well described by traditional thermodynamic and statistical physics. Benefitting from a long lifetime of ^2E level, thermal excitation rates for the $^2E \rightarrow ^4T_2$ transition increase at high temperature, thus providing enhanced gain on the $^4T_2 \rightarrow ^4A_2$ laser transitions.

In our $Nd:YVO_4$ laser, there is no ^2E-like storage level with long lifetime. The newly created laser wavelengths associated with the upper levels owe entirely to the enhancement of electron-phonon coupling on the cost of phonons by simultaneously suppressing strong fluorescence emission. The higher temperature will provide more phonons, who acts as a pump source.

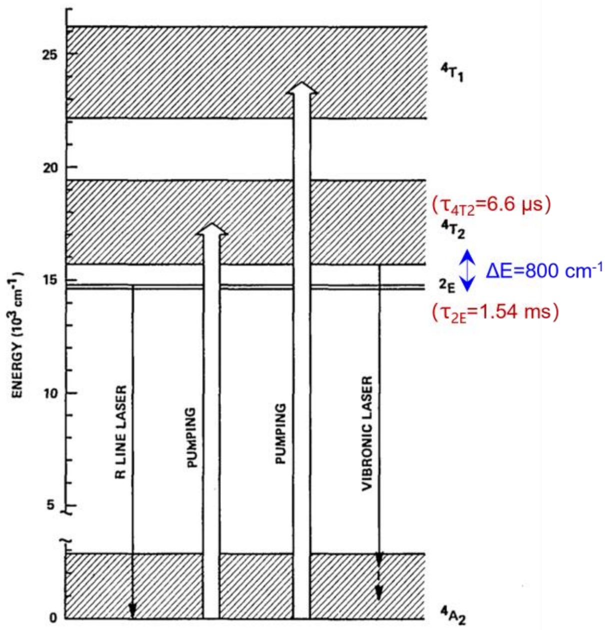

Figure 1. Energy-level diagram for alexandrite lasers. Thermal excitation from the ^2E storage level populates the $^4T_2$ state, which serves as the upper level for vibronic-laser operation [adapted from Opt. Lett. 7, 608-610 (1982)].

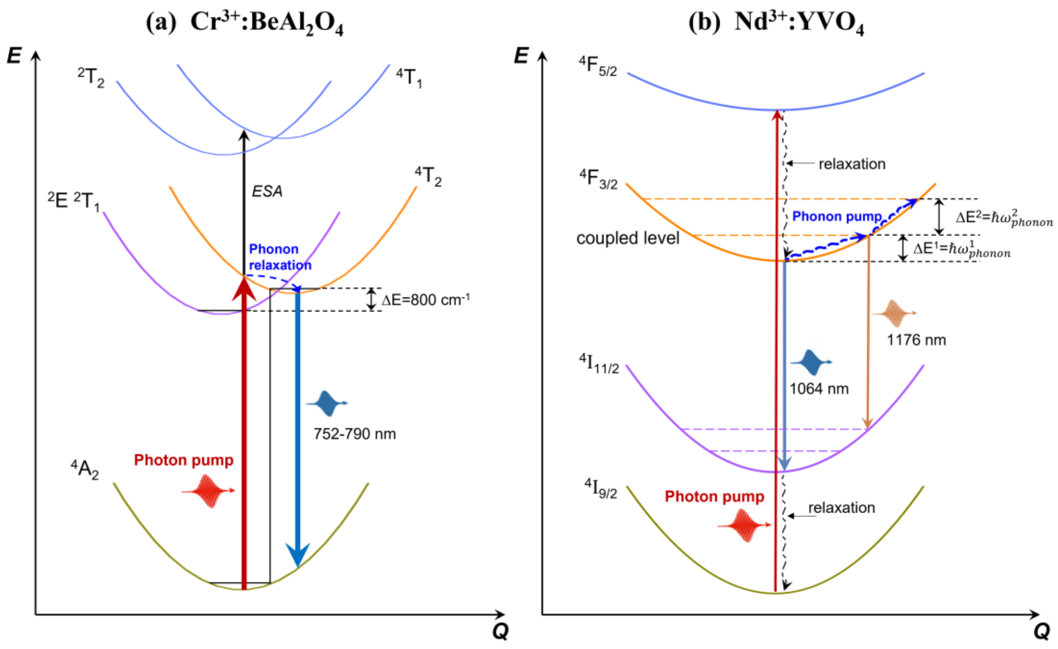

Figure 2. Different lasing mechanisms. (a) Alexandrite laser Cr^{3+}:$BeAl_2O_4$, (b) Our PPCP Nd:YVO_4 laser.

2. Different scope of application

The alexandrite crystal is the exceptional one with temperature enhanced lasers performance, due to its two close upper levels, to the best of our knowledge. Thus, it cannot be easily extended to other laser materials. Till now, only a few papers focus on this issue since it was first found in 1982 (see Table R1).

Our proposed "phonon-pumping" is a universal physical mechanism for many solid-state lasers without such limitation. Here, we would like to share our very recent experimental results of the phonon-pumped laser in Nd:YAG crystal (λ=1151 and 1166 nm), showing its great expansibility and importance from the application point of view (Figs. R4-R7).

3. Different temperature-dependent relation

In alexandrite lasers, high temperature also tends to populate the terminal $^4A_2$ levels. Therefore, the laser performance is the best one with a maximally populated initial level and a minimally terminal one at an optimal temperature (T_m) to balance the above two conflicting effects. The typical temperature-dependent performance is V-shape [see IEEE J. Quantum Electron. 16, 1302 (1980), T_m ~ 42°C and Opt. Lett. 7, 608 (1982), T_m ~ 225°C].

In our PPCP lasers, higher temperature brings more phonons, which will continuously improve laser performances in principle. Most importantly, besides qualitative explanation, we quantitatively obtain a lasing threshold-temperature relation $P_{th} = C/T_{th}$, based on both theoretical analysis and experimental results (C is a constant depending on active material). We believe such an in-depth and solid study has provided a clear picture of our phonon-pumped lasers.

4. The role of phonons

Alexandrite crystal has a large S factor, and the thermal phonons participated in the lasing process play an average effect for homogeneously spectral broadening [S~2.13 & $\hbar\omega_{ph}$~524 cm^{-1} in our recent paper Opt. Express 31, 9790 (2023)]. Therefore, with temperature increasing from 10 to 50 °C, the laser wavelength has a significant redshift from 761 to 772 nm (see Table R1 & Fig. R1). The laser wavelength is challenging to keep unchanged, not to mention controllable phonon modes.

Our $Nd:YVO_4$ crystal has a tiny S factor (0.002-0.013). Only specific-symmetry phonons that follow selection rules, such as A_{1g} or E_g phonon mode coupled to $\Gamma_6 \rightarrow \Gamma_7$ transition, are amplified to create new laser upper levels. Thus, we can preciously design and specify laser wavelength maintaining at 1176 nm (A_{1g}) or 1168 nm (E_g). From this perspective, our study may offer an ideal platform to quantitatively verify and study the quantized electron-phonon coupling effect, which may only appear in the textbook of solid-state physics.

Regarding reviewer #3's criticism about our physical picture and paper title, we can understand that it would take a long time to recognize a new laser mechanism. However, you can see that reviewers #1 and #2 have accepted our proposed collaborative pumping laser mechanism in the second round of review. Furthermore, reviewer #2 considers the current description is better, see"*..., and the revised figure 1 is helpful in better explaining their scheme...*" Although we would like to answer, explain, and discuss any technical issues, there is no tangible target in reviewer #3's comments. In the revised version, we try our best to improve the presentation to make our case more acceptable, especially for broad readership with different background.

Please find attached our detailed response to the second round of the reviewing process. All these new results are added to the revised manuscript and supplementary materials. We believe the current work does meet the strict standards on novelty, innovation, and impact for publication in ***Nature Photonics***.

Yours sincerely,
Haohai Yu, Huaijin Zhang and Yan-Feng Chen (On behalf of all authors)

Response to reviewer's comments

Reviewer #1

Comment:

The authors properly answered my questions and the manuscript is fine for publication.

Response:

We sincerely appreciate the reviewer's positive comments and recommendation.

Reviewer #2

Comment:

The authors claim a new type of laser that is collaboratively pumped with both photons and phonons. They present data that shows a conventional power threshold and input-output characteristic for a Nd:YVO_4, laser operating at 1176 nm. The laser also shows an unusual temperature threshold for laser action- i.e. above a critical temperature at constant pump power the laser increases its output with increasing temperature. The authors have made a detailed study of the temperature dependence, the underpinning laser physics and its use to generate laser light on several different transitions of Nd:YVO_4.

Following detailed and careful revisions, the authors have further clarified what they mean by "collaborative pumping with both photons and phonons", and the revised figure 1 is helpful in better explaining their scheme. (their previous version of figure 1 was presenting a quite different thermal interaction only in the ground state of the laser energy states). The "phonon pumping" as now described is a thermal population of higher lying vibronic states in the upper laser level, which gives rise to the anomalous temperature dependence of laser threshold, as shown in figure S1.

Response:

We thank the reviewer for his/her constructive comments. These comments greatly help us have a deep and comprehensive insight into our Nd:YVO_4 lasers.

Comment:

This laser mechanism, and the resulting change in performance with temperature, is in fact not a new mechanism but has actually been known to occur in other laser materials such as in alexandrite lasers since the 1980s (see references [1]-[3] below). I am not however aware of previous reports of this phenomenon in Nd-based lasers, so there is novelty in this careful scientific study. However, although this is an interesting result in Nd:YVO_4 that will be of interest to solid-state laser physicists, I do not believe that the phenomenon is in itself new, and therefore do not feel that the work is of sufficient novelty and wide interest for publication in Nature Photonics.

I therefore recommend resubmission to another journal (perhaps: Light, Science and Applications, Scientific Reports or Optics Letters).

[1] S Guch and CE Jones "Alexandrite-laser performance at high temperature", Optics Letters 7, 608 (1982)

[2] U Demirbas, A Sennaroglu and FX Kärtner, "Temperature dependence of Alexandrite effective emission cross section and small signal gain over the 25-450 °C range", Optics Materials Express 9, 3352 (2019)

[3] W Koechner "Solid-State Laser Engineering" Chapter 2.5.1 (Springer)

Response:

We thank the reviewer for raising this critical issue. However, we respectfully disagree with this point. Since we studied and carried out experiments on alexandrite lasers, we clearly know the mechanism of our photon-phonon collaboratively pumped laser is **fundamentally different** from that of alexandrite one.

First, we summarized the temperature-dependent alexandrite laser performances in **Table R1**. We agree that some alexandrite lasers have an anomalous temperature dependence on laser threshold. In previous reports, the output power increases while lasing threshold decreases at high temperatures [Opt. Lett. 7, 608 (1982)].

We also performed a similar alexandrite laser experiment in our lab. As shown in **Fig. R1**, the lasing threshold does indeed decrease with the increasing water-cooling temperatures, as well as higher output power. **However, the laser wavelength inevitably has a significant red shift from 761 to 772 nm, which is vital evidence to show the difference.** We will give a detailed explanation as follows.

Table R1. A summary of temperature-dependent alexandrite laser performances

Crystal	Pump source	Temperature range	Laser wavelength	Lasing threshold	Output power	Reference
0.4at.% Cr^{3+}:$BeAl_2O_4$ diameter:2.3mm length: 40mm	flash-lamp pulse: 104 μs	34-310 ℃	752-790 nm λ red shift at high temperature	first decrease, then increase; P_{th} is minimum at 225 ℃	P_{out} first increase, then decrease, P_{out} is maximum at 225℃	*Opt. Lett.* 7, 608 (1982)
c-cut 0.12at.% Cr^{3+}:$BeAl_2O_4$ diameter:6.35mm length: 96mm	flash-lamp pulse: 200 μs	15-64 ℃	λ ~ 750 nm	first decrease, then increase; P_{th} is minimum at 42 ℃	P_{out} increase at high temperature	*IEEE J. Quantum Electron.* 16, 1302 (1980)
c-cut 0.13at.% Cr^{3+}:$BeAl_2O_4$ diameter: 5mm length: 7mm	blue laser diode (λ=444 nm)	78-400 K	T<220K, λ=680 nm; T=260-400K, λ~745-751 nm λ red shift at high temperature	---	T<220K, P_{out} decrease at high temperature. T=260-400K, P_{out} first increase, then decrease. P_{out} is maximum at 350 K	*Laser Phys.* 27, 115801 (2017)
Brewster-cut 0.24at.% Cr^{3+}:$BeAl_2O_4$ diameter:4×4mm^2 length: 8mm	bule laser diode (λ=444 nm)	10-60 ℃	T=10-30 ℃ λ=762-760 nm; T=25-60 ℃ λ=774-772 nm. λ blue shift at high temperature	---	P_{out} decrease at high temperature	*Opt. Express* 31, 5832 (2023)
c-cut 0.2at.% Cr^{3+}:$BeAl_2O_4$ diameter: 4×4mm^2 length: 10mm	red laser diode (λ=638 nm)	10-50 ℃	λ=761-772 nm λ red shift at high temperature	P_{th} decrease at high temperature	P_{out} increase at high temperature	*Our experimental results*

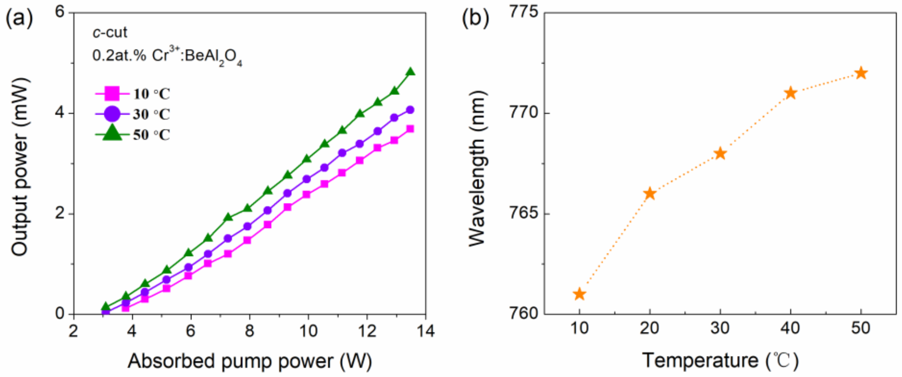

Figure R1. Temperature-dependent alexandrite lasers. (a) output power, (b) laser wavelength.

The physical model of alexandrite laser

This anomalous temperature-dependent threshold of the alexandrite laser can be attributed to the Boltzmann distribution of ^2E and $^4T_2$ levels generated by the Stark splitting. Due to the long lifetime of 1.54 ms, a metastable electronic ^2E level provide the capacity for significant energy storage. The energy difference between ^2E and $^4T_2$ is 800 cm^{-1}. The resulting $^4T_2 \rightarrow {}^4A_2$ transitions are natural at a finite temperature. At high temperatures, a successive thermal population of the $^4T_2$ level occurs in accordance with the Boltzmann distribution, where the thermal excitation rates for the $^2E \rightarrow {}^4T_2$ transition increase, thus providing enhanced gain on the $^4T_2 \rightarrow {}^4A_2$ vibronic laser transitions. This laser physical mechanism is plotted in **Fig. R2**.

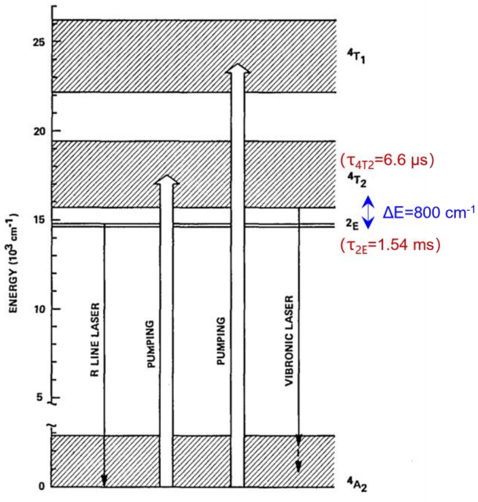

Figure R2. Energy-level diagram for alexandrite lasers. Thermal excitation from the ^2E storage level populates the $^4T_2$ state, which serves as the upper level for vibronic-laser operation [adapted from Opt. Lett. 7, 608 (1982)].

The difference between alexandrite laser and our Nd:YVO$_4$ laser

Here, we must emphasize that our "phonon-pumped" Nd:YVO$_4$ laser is fundamentally different from those alexandrite lasers. The alexandrite laser is a *traditional vibronic laser*. Its laser performance is determined by the electron-phonon coupling induced spectral broadening. In comparison, our Nd:YVO$_4$ laser is a *phonon-pumped* lasing process accompanied by lattice phonon annihilation at high temperature. This is a prerequisite for lasing at 1176 nm and 1168 nm in Nd:YVO$_4$ crystal. This would be a new and universal physical mechanism for many solid-state lasers.

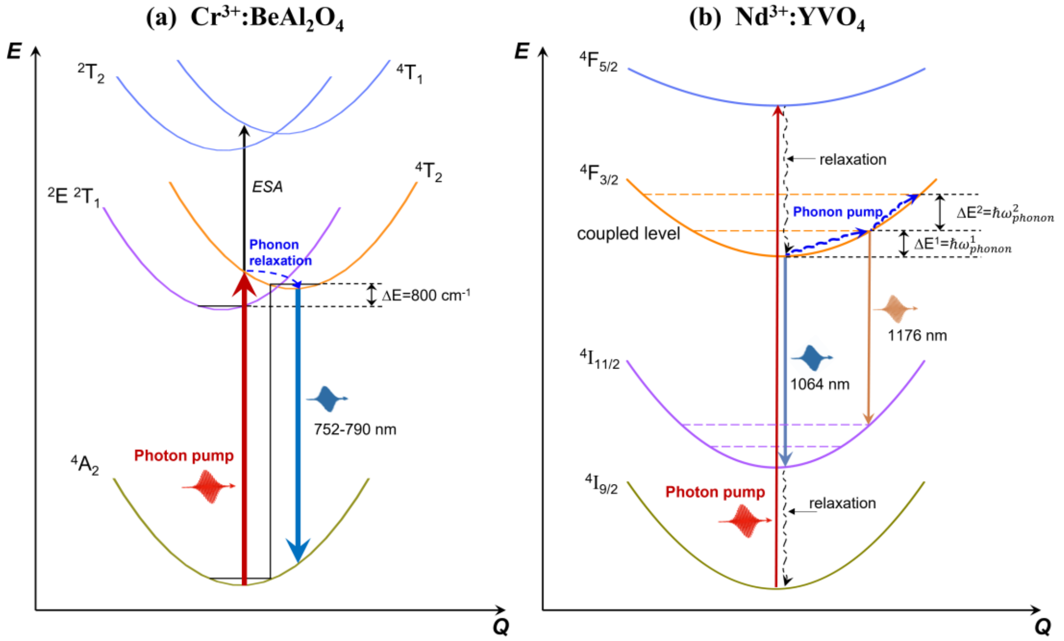

Figure R3. The configurational coordinate diagram of energy levels of Cr^{3+} ion and Nd^{3+} ion. (a) alexandrite laser ($Cr^{3+}:BeAl_2O_4$), (b) $Nd:YVO_4$.

We used a configurational coordination model to show the difference. As shown in **Fig. R3**, the main differences include two aspects: (1) energy level difference ΔE, (2) laser low-level.

1. Energy level difference ΔE

Alexandrite is an intermediate crystal field material, in which ^2E and $^4T_2$ states are coupled. Their energy difference is 800 cm^{-1}, determined by the crystal field effect and independent of phonon energy. The lifetimes are 1.54 ms and 6.6 μs, respectively. The two *R* lines emitted from ^2E occur in the vicinity of 680 nm, as for ruby. Because of the vibronic nature of the alexandrite laser, the fluorescence spectrum and corresponding gain cross-section become homogeneously broadening at high temperatures [Opt. Mater. Express 9, 3352-3370 (2019)]. Therefore, as the temperature increases, the gain of alexandrite increases, the gain peak shifts to a longer wavelength, and the fluorescence lifetime decreases. **This temperature-dependent wavelength shift is a typical behavior of vibronic lasers, indicating the spectral broadening is a classical electron-phonon coupling effect with average participated phonon energy** [$h\omega_{ph}$ ~524 cm^{-1} in our recent paper, Opt. Express 31, 9790-9799 (2023)]. The laser wavelength is unable to control owing to the strong electron-phonon coupling effect and laser mode competition.

In $Nd:YVO_4$, the "phonon pumping" effect creates new vibronic levels in laser transitions. **The quantized phonon energy determines the position of both initial and terminal levels for the laser emission**. The population of phonon states follows the Bose-Einstein distribution. Therefore, its laser wavelength is determined by the phonon energy. Under A_{1g} mode phonon pumping ($h\omega_{ph}$ =891 cm^{-1}), its lasing wavelength maintain at 1176 nm without a large wavelength shift (Δλ < 0.5 nm at 296-338 K, Supplementary Fig. S30). Moreover, we can move it to 1168 nm by choosing E_g mode phonon-pumping ($h\omega_{ph}$ =839 cm^{-1}) via a birefringent filter (main text Fig. 4). That is exactly why we call it a phonon-pumped laser, not a traditional vibronic laser.

2. Laser terminal level

In the alexandrite laser, the terminal laser level is a set of vibrational states well above the ground state. The laser initial level is a level 800 cm^{-1} above a long-lived storage level and in thermal equilibrium with it. The laser terminal level is very close to the ground-state level. Therefore, raising the temperature also tends to populate the terminal levels, especially those which lie closest to the ground level and which therefore correspond to the highest-energy (shortest-wavelength) photons. Since laser performance is the highest with a maximally populated initial level and a minimally populated terminal level, it can be seen that increasing the temperature has two conflicting effects. As a result, there would be an optimal temperature T_m for alexandrite lasers, showing a typical V-shape temperature-dependent performance as demonstrated by Walling [IEEE J. Quantum Electron. 16, 1302 (1980), T_m ~ 42℃] and Guch [Opt. Lett. 7, 608 (1982), T_m ~ 225℃].

In comparison, the laser low-level of our $Nd:YVO_4$ is $^4I_{11/2}$ level, not ground-state level $^4I_{9/2}$. There is a large energy difference of 1966 cm^{-1}. In general, the population of $^4I_{11/2}$ level can be neglected in the calculation of steady-state population inversion. As a result, our lasing threshold would continuously decrease with increasing temperatures in principle ($P_{th} = C/T_{th}$), until the intrinsic damage at high temperature. The minimum lasing threshold is determined by the thermal load capacity of the laser crystal and electron-phonon coupling intensity.

This "phonon-pumping" mechanism is universal for solid-state laser materials.

The alexandrite laser is only a special case in Cr^{3+}-doped laser materials. There is no anomalous temperature dependence of laser threshold in other Cr^{3+}-doped crystals, *e.g.*, ruby. However, our proposed "phonon-pumping" is applicable in other rare-earth and transition-metal laser crystals.

To prove this, we present some similar laser results in Nd:YAG crystal. As shown in **Fig. R4**, under high temperatures, Nd:YAG crystal exhibits a strengthened vibronic emission at 1151 and 1166 nm. Via a rational cavity design to suppress the conventional lasing at 946, 1064, 1122, and 1319 nm, we successfully realized a dual-wavelength lasing at 1151 and 1166 nm. These two wavelengths can be well explained by the phonon-pumped laser theory. According to our calculations, the participated phonon of Nd:YAG crystal is one E_g mode at 340 cm^{-1} for 1122 nm$\xrightarrow{+340cm^{-1}}$1151 nm, and one T_{2g} mode at 719 cm^{-1} for 1064 nm$\xrightarrow{+719cm^{-1}}$1166 nm. As shown in **Fig. R5**, the lasing thresholds of 1151 and 1166 nm gradually reduce and the output power gradually increases at high temperature. This case is similar to that of $Nd:YVO_4$ laser at 1176 nm.

In addition, we also observe a phase transformation from 1064 nm to 1151(&1166) nm lasing with increasing temperatures (**Fig. R6**). This is the biggest highlight of this work. The phonon pumping is related to the temperature T_{th} in the critical region. With an incident pump power at 12 W, the critical temperature T_{th} locates around 315 K for laser transition from phonon-free 1064 nm to phonon-pumped 1151(&1166) nm. When the pump power increases to 12.5 W, the critical temperature T_{th} becomes 311 K. This tendency of temperature shift is consistent with our "phonon-pumping" model. The laser wavelengths at different temperatures are plotted in **Fig. R7**. We can see that the laser wavelengths don't have a large shift at high temperatures, indicating that 1151 and 1166 nm emissions are novel "phonon-pumping" lasers, not traditional vibronic lasers.

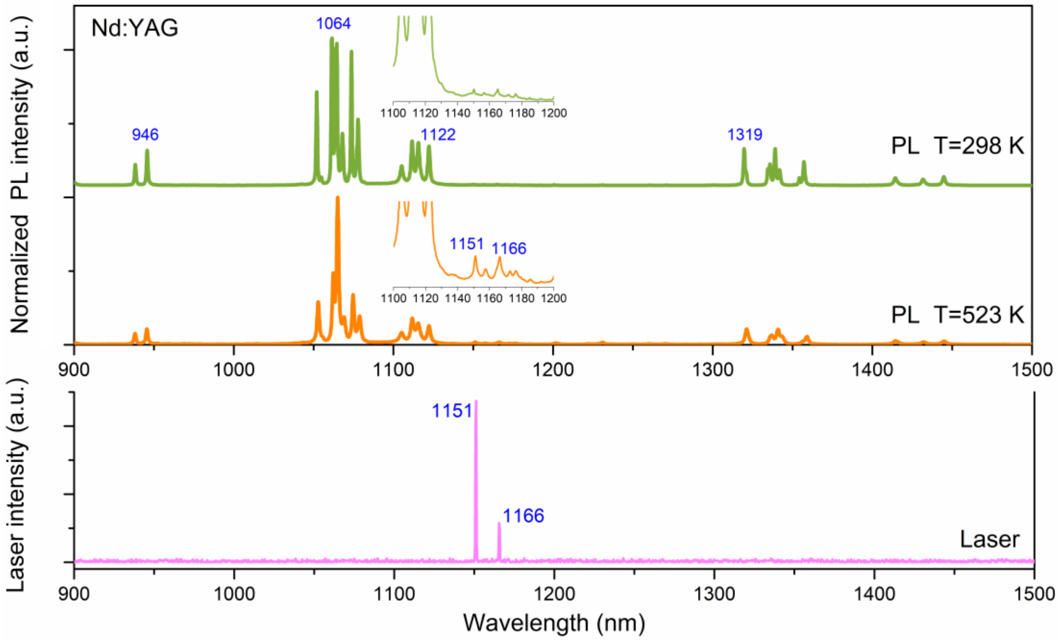

Figure R4. Fluorescence spectra (T=298 K/523 K) and phonon-pumped laser spectra of Nd:YAG crystal.

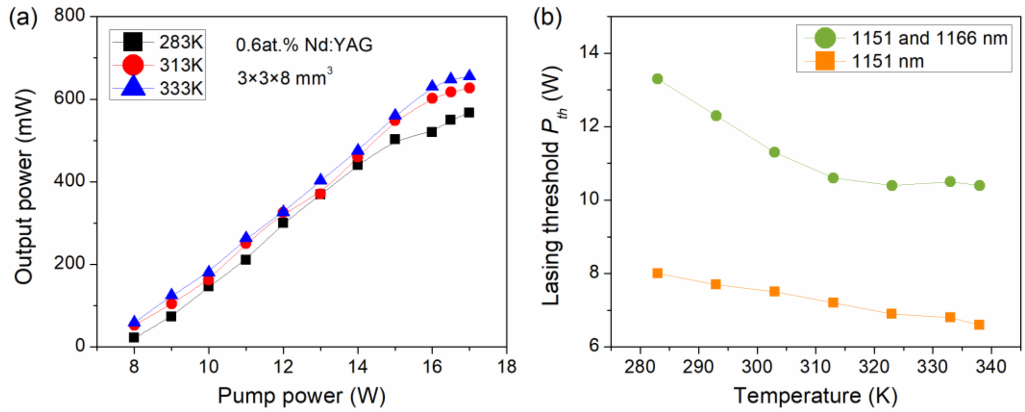

Figure R5. Temperature-dependent lasing performances at 1151 and 1166 nm laser in Nd:YAG crystal. (a) Output power versus pump power, (b) lasing threshold. Here, 1151 nm laser first starts. Then, 1151 and 1166 nm lasers start oscillation simultaneously.

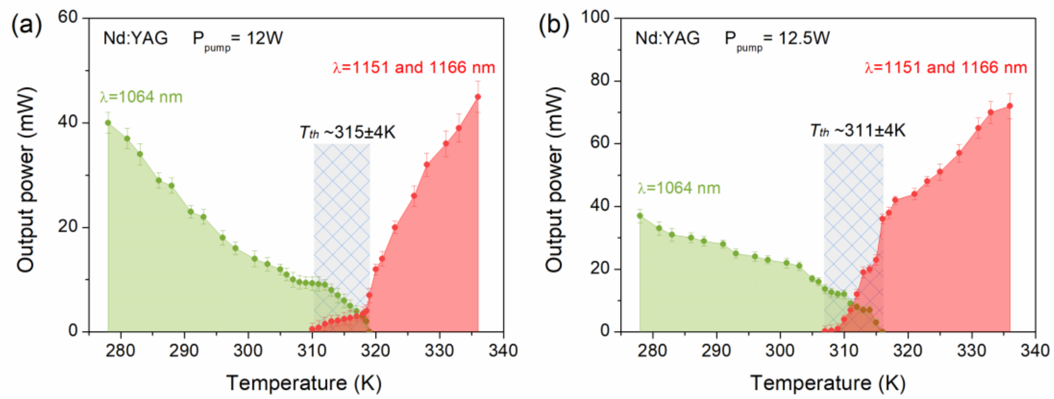

Figure R6. Power-temperature diagram of 1064 and 1151(&1166) nm laser in Nd:YAG crystal. (a) The incident pump power is 12 W. (b) The incident pump power is 12.5 W. T_{th} is a temperature threshold for phonon-pumped lasing.

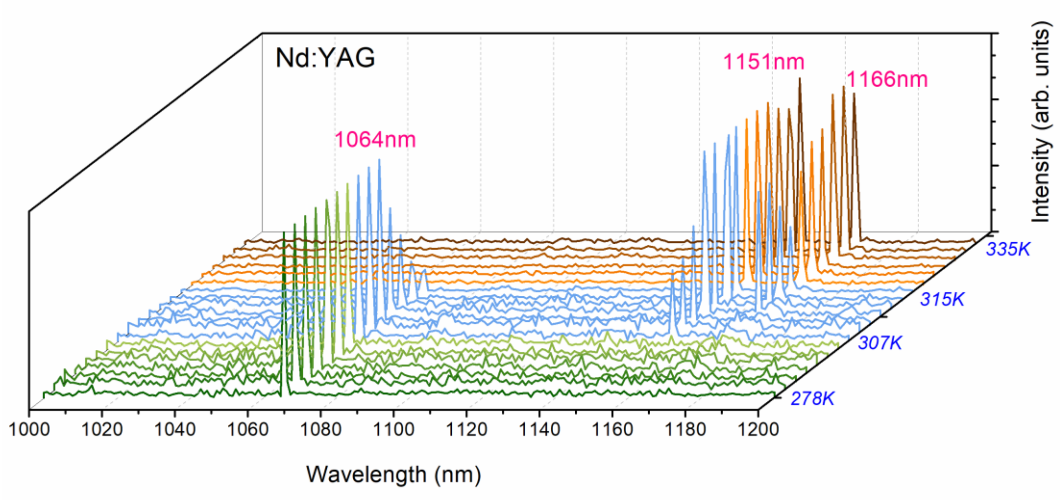

Figure R7. The phonon-pumped laser wavelengths at different temperatures (P_{pump}=12.5 W).

Finally, we make a comparison between alexandrite ($Cr^{3+}:BeAl_2O_4$), $Nd:YVO_4$, and Nd:YAG lasers. The alexandrite lasers belong to traditional phonon-assisted vibronic lasers around 760-770 nm, but $Nd:YVO_4$ and Nd:YAG lasers are phonon-pumped lasers at the wavelength of 1176 and 1151(&1166) nm, respectively. **These new laser wavelengths are first reported in Nd^{3+}-doped crystals since their first invention in 1960s.** These lasers have a distinct photon-pumping and phonon-pumping threshold relation of $P_{th}\times T_{th}=C$.

Table 2. A comparison between alexandrite ($Cr^{3+}:BeAl_2O_4$), $Nd:YVO_4$, and Nd:YAG lasers.

Crystal	pump	ΔE (cm^{-1})	phonon energy (cm^{-1})	laser wavelength λ (nm)	P_{th} (W)	T_{th} (K)	$P_{th}\times T_{th}$=C	C (W•K)
$Cr^{3+}:BeAl_2O_4$	638 nm LD	$\Delta E = {}^4T_2 - {}^2E$ = 800 cm^{-1}	average $h\omega_{ph}$=524 cm^{-1}	761-772	watt-level	No	No	---
$Nd^{3+}:YVO_4$	808nm LD	determined by the phonon energy and selection rules	A_{1g}=891 cm^{-1} E_g=839 cm^{-1}	1176, 1168	watt-level	293-308 K	Yes	3388±158 (λ=1176 nm)
Nd^{3+}:YAG	808nm LD	determined by the phonon energy and selection rules	E_g=340 cm^{-1} T_{2g}=719 cm^{-1}	1151, 1166	watt-level	311-315 K	Yes	3834±53 (λ=1151 nm)

Of course, we expect this "phonon-pump" mechanism should be also applied in alexandrite lasers. If we design a special cavity to suppress lasing below 850 nm, some phonon-pumped lasing can be expected at longer wavelengths. It can be modulated by the temperature and selection rules of electron-phonon coupling effect. However, at present this idea is a great challenge for alexandrite laser, because there is a strong excited-state absorption at long wavelengths beyond 850 nm [Opt. Express 26, 7771-7785 (2018); Opt. Mater. Express 9, 3352-3370 (2019)].

Comment:

There are also some areas where the authors should correct before resubmission:

1. The authors should add the previous work on Alexandrite lasers to figure S1 and table S1 and repitch their introduction and conclusions in the context of this previous work.

2. Line 64 "… By selecting and amplifying specific-symmetry phonons, …" I do not believe that the authors are amplifying phonons, but are amplifying photons – this needs to be corrected..

Response:

We thank the reviewer's comments.

1. We add the previous works on alexandrite lasers to Figure S1 and Table S1 (see **Fig. R8**).
2. In Line 64, Page 3, we corrected this sentence to "… By selecting and amplifying specific-symmetry photons,…".

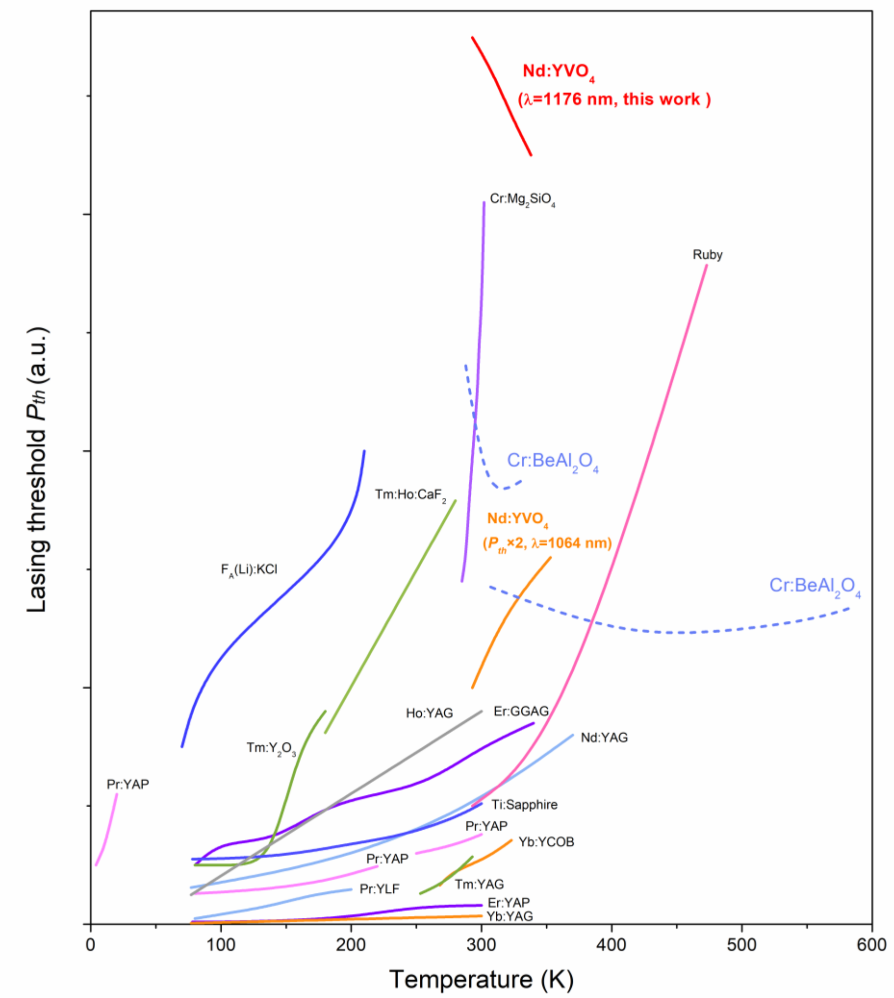

Figure R8. A summary of temperature dependent lasing threshold in solid-state lasers.

Action:

(1) Introduction section (Page 2), we added one sentence in the revision, "*Only a few alexandrite lasers exhibited the reduced P_{th} at high temperatures owing to its special crystal filed splitting and thermally-enhanced $^4T_2$ population.*[21-23]"

Refs 21-23: Opt. Lett. 7, 608 (1982); Opt. Mater. Express 9, 3352 (2019); Laser Phys. 27, 115801 (2017).

(2) Discussion and Conclusion section (Page 14), we added some sentences to emphasize the difference between our $Nd:YVO_4$ laser and alexandrite laser. "*Alexandrite laser is only a particular case in Cr^{3+}-doped laser materials, possessing very distinctive electronic upper levels where two close $^4T_2$ and ^2E upper levels are generated by crystal-*

field Stark splitting. Its laser temperature enhancement originates from the temperature-dependent Boltzmann distribution in these two levels, whose population can be well described by traditional thermodynamic and statistical physics. However, in our Nd:YVO_4 laser, these newly created laser wavelengths were associated with the upper levels owe entirely to the enhancement of electron-phonon coupling on the cost of phonons. The higher temperature will provide more phonons, who acts as a pump source."

(3) Additional *Fig. S36-38, Table S5, and explanatory text* have been added in the Supplementary materials to show the difference between Nd:YVO_4 laser and alexandrite laser.

As the reviewer can see, all the issues are well addressed based on the new experiments and analysis. Thanks to his/her comments, our manuscript's quality has significantly improved. We believe the current version does meet the strict standards on novelty, innovation, and impact for publication in ***Nature Photonics***. We sincerely hope the reviewer can change his/her recommendation.

Reviewer #3

Comment:

Fortunately, the revision did improve the manuscript, certainly linguistically but also concerning some technical aspects.

Still, I'm left with the feeling that the authors do not have a clear picture about what is going on. In the first version, it was lasing to a virtual level, now the levels are real, namely vibronic states. But for the authors, this does not matter; they left the title unchanged and so neglect the objections of all 3 reviewers. Also the authors were not able to underpin their model, despite the questions from the reviewers.

Because of the (still) poor presentation, and the lack of theoretical substantiation of the experimental results, my advice for publication in NP remains negative.

Response:

Thank you for your comments. According to your suggestions, we revised and improved our manuscript consequently.

1. First, we believe we have a clear picture of our laser model.

In the first submission, it was lasing to a virtual level, and in the second version, it is a phonon-pumped state coupled to laser up-level. However, these two concepts are not contradictory. Both them can be applied to describe the phonon-involved lasing process. For example, the first phonon-involved lasing in Ni^{2+}:MgF_2 was explained by a transition from $^3T_2$ level to a virtual phonon level (**Fig. R9a**). Later, a configurational model with vibronic states was applied to explain *F*-center laser and transition-metal lasers (**Fig. R9c**). Till now, both two models are still utilized in solid-state laser engineering to describe phonon-involved lasers, such as Tm^{3+}:KLuW (**Fig. R9b**) and Ti^{3+}:Sapphire (**Fig. R9d**). Therefore, both virtual level and phonon-pumped states are correct for vibronic lasers.

In the current revision, we choose the phonon-pumped states because it is convenient to describe our phonon-pumping model.

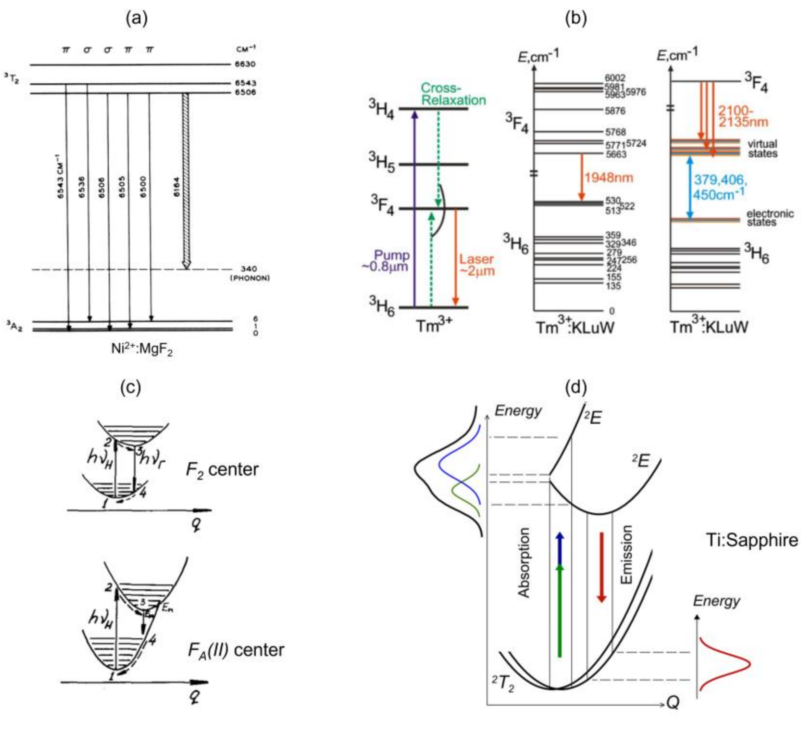

Figure R9. Laser physical model. (a) Ni^{2+}:MgF_2, adapted from *Phys. Rev. Lett.* **11**, 318 (1963); (b) Tm^{3+}:KLuW, adapted from *J. Opt. Soc. Am. B* **33**, D19 (2016); (c) *F*-center laser, adapted from *IEEE J. Quantum Electron.* 24, 1052 (1988); (d) Ti:Sapphire laser, adapted from *Topics in Applied Physics*, 95, 3 (2004).

2. We consider our title of "Photon-phonon collaboratively pumped laser" is sufficiently accurate to underpin the laser physic of Nd:YVO_4 laser. As for new Nd:YAG laser results, we firmly believe that it is indeed a cooperative pumping process with photon absorption and phonon absorption simultaneously. Two threshold parameters, P_{th} and T_{th}, can be observed in experiments, and satisfy a simple relation of $P\times T=Constant$. Meanwhile, a laser phase transition from phonon-free 1064 nm to phonon-pumped 1151(&1166) nm was demonstrated. Therefore, we are confident in such a phonon-pumping process. It would be a universal laser model for solid-state lasers.

3. Based on the phonon-pump mechanism and rational cavity, we created new laser wavelengths in Nd:YVO_4 (λ=1176 and 1168 nm) and Nd:YAG crystal (λ=1151 and 1166 nm). Since these crystals were invented in 1960s, this work represents a new forward step for these well-known laser crystals. In addition, an anomalous temperature dependence of laser threshold was discovered in them. These experimental results give us, also a broad community of laser physicists, some new degree of freedom to recognize electronic transitions in laser. More impressively, phonon acts as a carrier of energy, momentum, and angular momentum, which should be reconsidered in the area of laser generation, manipulation and detection. Therefore, this work is extremely matchable with the aim and scope of *Nature Photonics*.

We hope reviewer #3 can change his/her recommendation because such a new (at least potential) laser mechanism is a significant step forward in laser physics and laser technology, thus, deserve to be published in *Nature Photonics* to get a broader audience.

A list of changes:

Main text

We revised the introduction and discussion parts.

Several sentences have been added to address the reviewers' comments, especially for lasing mechanism, and discussion of temperature-dependent alexandrite laser performances. We emphasize the difference between alexandrite laser and our phonon-pumped laser.

References

Refs 21-23 have been added in the main text.

Supplementary information

Additional Fig. S36-38, Table S5, and explanatory text have been added in the supplementary materials.