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Solid-State ^{229}Th Nuclear Laser with Two-Photon Pumping

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Abstract

The radiative excitation of the 8.3 eV isomeric state of thorium-229 is an outstanding challenge due to the lack of tunable far-ultraviolet (F-UV) sources. In this work, we propose an efficient two-photon pumping scheme for thorium-229 using the optonuclear quadrupolar effect, which only requires a 300 nm UV-B pumping laser. We further demonstrate that population inversion between the nuclear isomeric and ground states can be achieved at room temperature using a two-step pumping process. The nuclear laser, which has been pursued for decades, may be realized using a Watt-level UV-B pumping laser and ultrawide bandgap thorium compounds (e.g., ThF_4 , Na_2ThF_6 , or K_2ThF_6) as the gain medium.

Introduction. Thorium-229 (^{229}Th) nucleus exhibits a long-lived isomeric state ($^{229*}\text{Th}$) with an ultra-low energy of $\omega_{\text{is}} \approx 8.3$ eV above the ground state [1–3], in stark contrast to the typical nuclear excitation energies (keV to MeV) [4]. Such a low-energy isomeric state elicits considerable interest in understanding the underlying nuclear structure [5,6], the coupling between nuclear and electronic excitations [7–9], as well as in developing various applications such as creating a nuclear clock frequency standard [1,10–12] and determining fundamental physical constants [13,14].

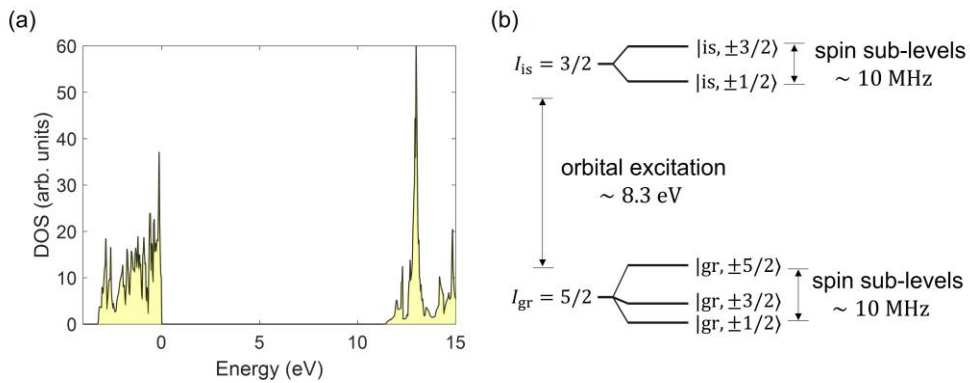
^{229}Th also provides unique opportunities for building nuclear lasers, which were envisioned more than half a century ago, but have not been realized yet [15–17]. Besides the conceptual importance of nuclear lasing, nuclear lasers also feature short wavelengths and narrow linewidth that could facilitate various applications. ^{229}Th nuclear lasers, if realized, could also offer a direct and convenient approach to optically pump ^{229}Th nuclei, which is crucial for

35 many applications involving ^{229}Th , including nuclear clock [1,10–12]. However, the
36 construction of nuclear lasers is a demanding task that requires interdisciplinary research in
37 nuclear physics, materials science, and photonics. A major challenge is that typical lasers
38 require population inversion and hence efficient pumping to the excited states. However, it is
39 notoriously difficult to pump to nuclear excited states. Historically, it has been proposed to use
40 X-ray radiations, slow neutron capture, or other nuclear reactions to pump the nuclear excited
41 states, all of which are not so efficient [15]. For ^{229}Th , it is possible to populate the isomeric
42 states optically, thanks to the small transition energy within the far-ultraviolet (F-UV)
43 regime [17]. Unfortunately, F-UV sources resonant with the ^{229}Th isomeric transition are still
44 under development, and the direct laser excitation of ^{229}Th has not been experimentally
45 demonstrated yet [1]. Moreover, even if direct F-UV pumping becomes available, population
46 inversion still cannot be achieved if only two states are involved, and at least one auxiliary state
47 is necessary. For nuclear lasers, it is not straightforward to find the auxiliary state [17], because
48 the energy gaps between nuclear spin sub-levels ($10^{-9} \sim 10^{-6}$ eV) are too small compared
49 with typical laser frequency (~ 1 eV), thus requiring prohibitively low temperatures [17].
50 Meanwhile, the energy gaps between nuclear orbital excited states are too large – the second
51 excited state of ^{229}Th is around 29 keV above the ground state – thus requiring demanding X-
52 ray sources.

53 In this work, we propose a nuclear laser based on ^{229}Th with a two-step two-photon pumping,
54 which could potentially overcome the aforementioned challenges. As the gain medium, we
55 suggest using Thorium compounds such as Na_2ThF_6 , which can provide a high number density
56 of ^{229}Th and an ultrawide bandgap, $E_g > \omega_{\text{is}}$ [18]. The ultrawide bandgap also forbids internal
57 conversion (IC) of the isomeric state [1,4], because the large ionization energy prevents this
58 non-radiative nuclear decay process that excites and ejects a valence electron [19]. These
59 properties are advantageous for the nuclear laser. Then, we introduce the two-photon pumping
60 of the ^{229}Th isomeric states based on the optonuclear quadrupolar (ONQ) effect [20,21], which
61 is an efficient interface between two photons and the nuclei. The ONQ pumping requires only
62 a near-ultraviolet laser operating at $\omega_{\text{in}} = \frac{\omega_{\text{is}}}{2} \approx 4.1$ eV, which is in the UV-B regime and
63 could be much easier to build than a F-UV laser [22]. In contrast to the pumping scheme based
64 on the electronic bridge (EB) effect [7,9,23–26], the ONQ pumping avoids the usage of lasers
65 resonant with electronic transitions and can thus significantly suppress the heating in the solid-
66 state gain medium. This is important when the ^{229}Th density is high and the pumping laser is
67 strong, whereby the heating power density will be high. Under a sub-Watt-level pumping laser,
68 the ONQ pumping could be fast enough for the experimental observation of the radiative
69 excitations of $^{229*}\text{Th}$, which has not been realized yet. We further propose a two-step pumping

70 process to achieve population inversion at room temperature, taking advantage of the long
 71 relaxation time of ^{229}Th nuclear states. We show that the peak power of the nuclear laser can
 72 reach Watt-level when the gain medium size is about $1\mu\text{m} \times 1\mu\text{m} \times 1\text{mm}$. By selecting
 73 different nuclear spin sub-levels, the nuclear laser can have tunable chirality as well [27]. The
 74 nuclear laser can have narrow linewidth, and can naturally match the resonance condition for
 75 optically pumping the ^{229}Th nuclear clock. While ^{229}Th can be pumped with other schemes
 76 under development or operation [2], the nuclear laser pumping may have its own advantage.
 77 For example, the central frequency of the nuclear laser can potentially be tuned with ultra-fine
 78 resolution using e.g., the Mössbauer effect. Further investigation is required to explore the
 79 potential applications of the nuclear laser.

80 **^{229}Th in ultrawide bandgap Th-compounds.** The radiative transitions between the isomeric
 81 (angular momentum $I_{\text{is}} = 3/2$) and ground states ($I_{\text{gr}} = 5/2$) of ^{229}Th have both $M1$
 82 (magnetic dipole) and $E2$ (electric quadrupole) channels. Some detailed information on the
 83 isomeric transition, including the selection rules, is summarized in Section 2 in Ref. [28]
 84 (Supplementary Materials, which also cites Refs. [5,6,29–38]). The spontaneous gamma-decay
 85 of $^{229*}\text{Th}$ is dominated by the $M1$ process with a decay rate of $\gamma_{\text{is}}^{\gamma} \sim 10^{-4}$ Hz [4]. The IC,
 86 while fast [39], can be forbidden if the isomeric transition energy ω_{is} is below the electron
 87 ionization energy, so the nuclear transition does not have enough energy to kick out an electron.
 88 In this case, the total decay rate of $^{229*}\text{Th}$ is $\gamma_{\text{is}}^{\text{decay}} = \gamma_{\text{is}}^{\gamma}$ [40]. It has been shown that using
 89 trapped ionized ^{229}Th [41] or ^{229}Th dopants in ultrawide bandgap compounds (e.g. CaF_2 [42])
 90 can forbid IC. For nuclear lasers, it is desirable to have a large number density of ^{229}Th . Hence,
 91 we instead suggest using natural Th-compounds, which can have a number density of up to
 92 10^{26} m^{-3} if ^{229}Th is enriched to 1% isotopic abundance. Some candidate compounds are ThF_4 ,
 93 Na_2ThF_6 , and K_2ThF_6 , all of which have electronic bandgaps $E_g \gtrsim 10$ eV according to
 94 experiments [18] as well as our many-body G_0W_0 calculations (Section 1.1 in Ref. [28]).



95
 96 **Figure 1.** (a) Electronic density of states of Na_2ThF_6 from G_0W_0 calculations. (b) Nuclear energy level
 97 diagram of ^{229}Th in Na_2ThF_6 .

99 We will use Na_2ThF_6 as an example. According to our calculations, the electronic bandgap of
 100 Na_2ThF_6 is $E_g \approx 11$ eV (Figure 1a), yielding $\omega_{\text{is}} < E_g < \frac{3}{2}\omega_{\text{is}}$. The splitting between
 101 nuclear spin sub-levels due to the nuclear quadrupolar interaction is on the order of 10 MHz
 102 (40 neV, Figure 1b), equivalent to a temperature of mK. Hence, under ambient conditions, the
 103 five ground-state sub-levels are almost equally populated. In contrast, ω_{is} is much greater
 104 than the thermal energy, and the isomeric sub-levels should have zero population at thermal
 105 equilibrium. Another important parameter for the nuclear laser is the drift (inhomogeneous
 106 broadening) $\gamma_{\text{is}}^{\text{drift}}$ of the isomeric transition energy in solid-state compounds, which could
 107 result from magnetic interactions, temperature, and strain effects [31]. The magnetic dipole
 108 interaction between nearby nuclei is on the order of kHz [43], while our calculations (Section
 109 1.1 in Ref. [28]) indicate that $\gamma_{\text{is}}^{\text{drift}}$ can be kept below 10 kHz if the variance of strain
 110 (temperature) is below 10^{-2} % (1 K). Hence, we will assume $\gamma_{\text{is}}^{\text{drift}} \sim 10$ kHz in the
 111 following. The small drift in the isomeric transition energy also indicates that the nuclear laser
 112 can have a narrow linewidth.

113 **Two-photon pumping via the optonuclear quadrupolar effect.** As discussed before, the
 114 pumping of the nuclear isomeric excited state is a key challenge for nuclear lasers. In this
 115 section, we demonstrate the two-photon pumping of ^{229}Th based on the ONQ effect [20,21].
 116 Specifically, the nuclear state can be influenced by the nuclear quadrupolar ($E2$) interaction
 117 $\mathcal{H}_{E2} = \mathcal{M}_{E2}\mathcal{V}$, which is an electromagnetic interaction between the nuclear electric
 118 quadrupolar moment \mathcal{M}_{E2} and the electric field gradient (EFG) \mathcal{V} at the site of the nucleus.
 119 External fields can modulate \mathcal{V} , which can in turn control the nuclear states. In fact, the
 120 gamma-decay through the $E2$ channel is the consequence of the oscillating EFG of a resonant
 121 photon [44]. However, the EFG of a VUV photon is too weak, and thus the $E2$ channel for
 122 gamma-decay is inefficient compared with the $M1$ channel for bare $^{229*}\text{Th}$ in vacuum [6].

123 The situation is different when electrons come into play. The electric field generated by
 124 electrons can vary by $\Delta\mathcal{E}_e \gtrsim 1$ V/Å over the atomic scale a_0 , with a_0 the Bohr radius.
 125 Hence, the EFG \mathcal{V}_e generated by electrons can reach $\mathcal{V}_e \sim \frac{\Delta\mathcal{E}_e}{a_0} \sim 1$ V/Å², leading to a strong
 126 nuclear quadrupolar interaction (MHz to GHz). When the electronic states are perturbed, the
 127 change in the nuclear quadrupolar interaction is proportionally strong. This fact helps explain
 128 the fast IC of $^{229*}\text{Th}$ through the $E2$ channel [19].

129 Particularly, the electronic states can be perturbed by two-photon transitions. This is the origin
 130 of various well-known second-order nonlinear optical effects – two photons drive the electronic

131 orbital motions, which in turn generate e.g., electromagnetic waves (sum or difference
 132 frequency generation) or phonons (Raman scattering). Similarly, electronic orbital motions can
 133 also generate oscillating EFG and hence oscillating nuclear quadrupolar interaction, which can
 134 influence the nuclear states. This is the ONQ effect [20,21], which can be described by the
 135 Hamiltonian $\mathcal{H}_{E2}^{\text{ONQ}}(t) = \sum_{ij} \mathcal{D}_{ij,\pm}^{pq} \mathcal{E}_p \mathcal{E}_q e^{i(\omega_p \pm \omega_q)t} + h.c.$, where $\mathcal{E}_{p,q}$ and $\omega_{p,q}$ are the
 136 electric field strength and the frequency of the two photons p and q , respectively. The \pm
 137 sign indicates the sum (+) or difference (−) frequency process. The response function $\mathcal{D}_{ij,\pm}^{pq} =$
 138 $\mathcal{M}_{E2} \frac{\partial^2 \mathcal{V}}{\partial \mathcal{E}_p \mathcal{E}_q}$ can be expressed as [20,21]

$$\mathcal{D}_{ij,\pm}^{pq} = \mathcal{M}_{E2} \sum_{mnl} \frac{[\mathcal{V}_{ij}]_{mn}}{\omega_{mn} - (\omega_p \pm \omega_q)} \times \left\{ \frac{f_{lm}[r_p]_{nl}[r_q]_{lm}}{\omega_{ml} - \omega_p} - \frac{f_{nl}[r_q]_{nl}[r_p]_{lm}}{\omega_{ln} - \omega_p} \right\} + (p \leftrightarrow q) \quad (1)$$

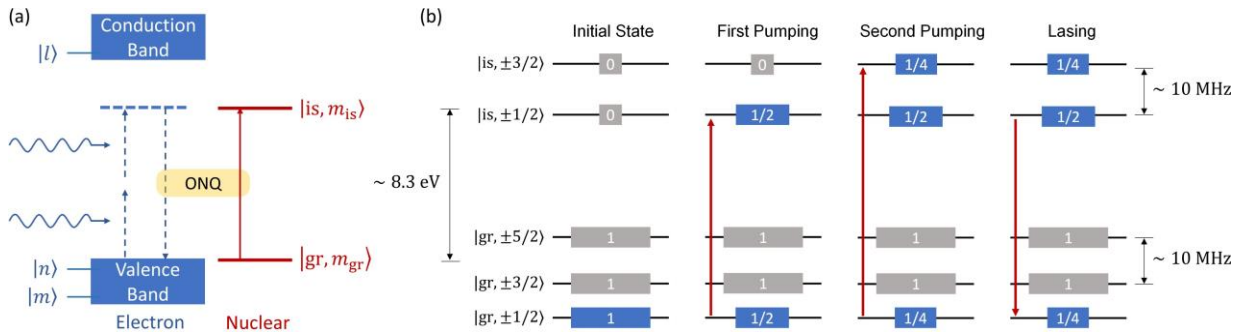
139 where $(p \leftrightarrow q)$ indicates the exchange of the p and q subscripts. $[r_i]_{nl} \equiv \langle n | r_i | l \rangle$ and
 140 $[\mathcal{V}_{ij}]_{mn} = \frac{e}{4\pi\epsilon_0} \left\langle m \left| \frac{3r_i r_j - \delta_{ij} r^2}{r^5} \right| n \right\rangle$ are the electron position and EFG operators, respectively,
 141 where m, n, l label the electronic states, and ϵ_0 is the vacuum permittivity. ω_{mn} (f_{mn}) is
 142 the energy (occupation) differences between two electronic states $|m\rangle$ and $|n\rangle$ (Planck
 143 constant $\hbar = 1$). Here, we focus on the sum-frequency term $e^{i(\omega_p + \omega_q)t}$, which can pump the
 144 electrons to a (virtual) excited state with an energy of $\omega_p + \omega_q$. Then, the nuclear excitation
 145 can be realized by a swap process between the electronic and nuclear excitations – electrons
 146 (virtually) jump back to the ground state, while the nucleus jumps to the isomeric state (Figure
 147 2a). This process is enabled by the electron-nuclear interactions, which can have $M1$, $E2$, and
 148 other higher-order channels. In the case of Na_2ThF_4 , there are no net electron spins, so the
 149 $M1$ channel is absent. To leading order, we only need to consider the $E2$ channel.

150 When $\omega_p + \omega_q < E_g$, the electronic transition is virtual, but the nuclear transition can be a real
 151 resonant transition when $\omega_p + \omega_q = \omega_{is}$. When the laser frequencies (ω_p , ω_q or $\omega_p + \omega_q$)
 152 are resonant with an electronic transition, the electrons can be resonantly pumped to electronic
 153 excited states, and the \mathcal{D} tensor will be substantially enhanced. In this case, the ONQ effect is
 154 in principle equivalent to the EB process [7,9,23–26]. We would like to emphasize that a
 155 unique advantage of the ONQ effect is that the laser frequencies can be off-resonant with
 156 electronic transitions (below bandgap), which can significantly suppress the one-photon
 157 absorption of laser energy and the resultant heating. This difference with the EB process is
 158 particularly important when the number density of Th is high and the pumping laser is strong,
 159 both of which are desirable for the solid-state nuclear laser. Additionally, the EB process is not

160 favorable in an ultra-wide bandgap thorium compound, as the one-photon resonant transition
 161 requires a laser with frequency >10 eV, which is hard to construct. Therefore, we believe the
 162 ONQ effect can be more advantageous than the EB process regarding building the nuclear laser.
 163 For an order-of-magnitude estimation, we only consider the (m, n, l) pair that has ω_{mn}, ω_{ml}
 164 close to E_g , which makes a major contribution to \mathcal{D} . We also use $\langle m | \frac{3r_i r_j - \delta_{ij} r^2}{r^5} | n \rangle \approx \frac{1}{a_0^3}$ and
 165 $[r_i]_{mn} \approx a_0$, as the spatial distribution of the electronic states is characterized by a_0 . We also
 166 set $\omega_p = \omega_q = \omega_{in} = \frac{\omega_{is}}{2}$. Finally, one has $\mathcal{D}_+ \sim \mathcal{M}_{E2} \frac{g_S e^3}{4\pi\epsilon_0 a_0} \frac{1}{(E_g - \omega_{is})(E_g - \omega_{is}/2)}$, where $g_S =$
 167 2 is the electron spin degeneracy.

168 The pumping rate to the isomeric state is $R = \frac{4|\langle gr, m_{gr} | \mathcal{D}_+ | is, m_{is} \rangle|^2 \mathcal{E}^4}{\Gamma_{\text{pump}}}$ with $\Gamma_{\text{pump}} \sim \gamma_{is}^{\text{decay}} +$
 169 $\gamma_{is}^{\text{drift}} + \kappa_{in}$, where κ_{in} is the linewidth of the pumping laser. One has $R[\text{Hz}] \sim 10^{-5} \times$
 170 $\mathcal{E}^4[\text{MV}^4 \cdot \text{m}^{-4}]$ when Γ_{pump} is on 10 kHz scale. When $\mathcal{E} = 1 \text{ MV} \cdot \text{m}^{-1}$, one has $R \sim$
 171 10^{-5} Hz . If a $[10\mu\text{m}]^3 \text{ Na}_2\text{ThF}_6$ sample with a ^{229}Th number density of 10^{26} m^{-3} is used,
 172 then there will be $\sim 10^6$ excitations to, and $\sim 10^2$ radiative decays from the isomeric state
 173 per second. This could be fast enough for the experimental observation of the nuclear radiative
 174 emission.

175



176

177 **Figure 2** (a) Two-photon pumping scheme based on the ONQ effect. Two-photons pumps a (virtual)
 178 electronic excitation, which is then swapped to a real nuclear excitation through the nuclear quadrupolar
 179 interaction. (b) The two-step pumping scheme to achieve population inversion. Numbers in the boxes
 180 indicate normalized populations. States with grey boxes do not participate in the nuclear pumping/lasing
 181 process.

182

183 **Population inversion under two-step pumping.** While the two-photon process discussed
 184 above can pump the isomeric state, it cannot lead to a population inversion, because it only

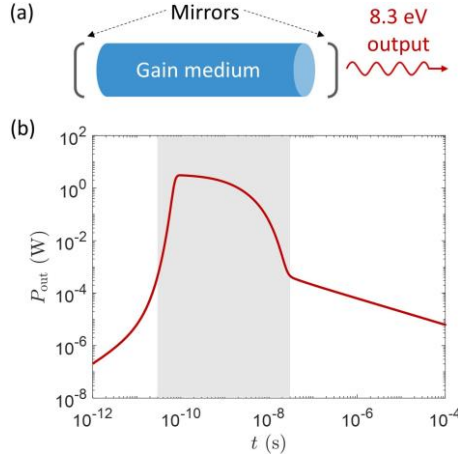
185 involves two nuclear states. For population inversion, at least one other auxiliary state is
 186 necessary [33]. The second nuclear orbital excited state is not an ideal choice because its high
 187 energy (29 keV) necessitates demanding X-ray sources. A more practical option is to use the
 188 nuclear spin sub-levels. However, the energy splitting between these spin sub-levels is too
 189 small. Hence, if the common one-step pumping scheme is used, then an effective population
 190 can be achieved only under a cryogenic temperature of millikelvin (Section 3 in Ref. [28]).

191 For room-temperature nuclear lasing, we propose a two-step pumping process (Figure 2b).
 192 Initially, the system is at thermal equilibrium, so the ground state sub-levels have
 193 (approximately) the same population (normalized to $f = 1$), while the isomeric states are
 194 empty ($f = 0$). For the first step of the pumping process, we use a two-photon pumping laser
 195 resonant with the $|gr, \pm 1/2\rangle \leftrightarrow |is, \pm 1/2\rangle$ transition. Provided the pumping rate is large
 196 enough ($R \gg \gamma_{is}^{\text{decay}}$), the $|is, \pm 1/2\rangle$ state will have almost the same population ($f = \frac{1}{2}$) as
 197 the $|gr, \pm 1/2\rangle$ state when the pumping saturates. Then, we switch to a second pumping laser
 198 resonant with the $|gr, \pm 1/2\rangle \leftrightarrow |is, \pm 3/2\rangle$ transition, which again equalizes the final
 199 population ($f = \frac{1}{4}$) on these two states. This results in the population inversion between
 200 $|is, \pm 1/2\rangle$ ($f = \frac{1}{2}$) and $|gr, \pm 1/2\rangle$ ($f = \frac{1}{4}$), and nuclear lasing between these two states would
 201 start when the laser resonator is tuned on resonance. A greater population inversion can be
 202 achieved if a multi-step pumping sequence is used (Section 3.2 in Ref. [28]). A caveat is that
 203 the two-photon pumping rate R should be faster than the nuclear spin relaxation, which tends
 204 to equalize the population among nuclear spin sub-levels at room temperature and destroy the
 205 population inversion. Considering that the nuclear spin relaxation rate is usually on the order
 206 of Hz at room temperature [45,46], a pumping rate of $R = 100$ Hz should suffice. The
 207 corresponding pumping electric field is $\mathcal{E}_{\text{in}} \approx 0.56$ MV/cm, and the laser power is $P_{\text{in}} \approx$
 208 4.2 W when the spot size is $[1\mu\text{m}]^2$. With a pumping rate of $R = 100$ Hz, it takes $0.01 \sim$
 209 0.1 s to reach the two-level saturation. In addition, we remark that the linewidth of the
 210 pumping laser should be smaller than the splitting between nuclear spin sub-levels (around 10
 211 MHz in Na_2ThF_6), so that it can be resonant with just one nuclear transition at a time.

212 **Experimental setup and performance of the nuclear laser.** Next, we discuss the basic
 213 experimental setup of the nuclear laser. We assume the gain medium (e.g., Na_2ThF_6) has a
 214 volume $V = Sl$, with S the cross-sectional area and l the length. The total number of *active*
 215 ^{229}Th nuclei is $N_{\text{Th}} = f_n \rho_n V$, where ρ_n is the number density of ^{229}Th , while $f_n \approx 1$ is the
 216 Lamb–Mössbauer factor (also known as the Debye-Waller factor) [47]. For clarity, we fix
 217 $f_n \rho_n \sim 10^{26} \text{ m}^{-3}$, which can be achieved when ^{229}Th is enriched to $\sim 1\%$ abundance. Note
 218 that an abundance of ^{229}Th exceeding 75% has been realized before [48]. Given a gain medium

219 with a $1\mu\text{m} \times 1\mu\text{m} \times 1\text{mm}$ dimension (see below), the total weight of ^{229}Th nuclei is around
 220 4×10^{-11} gram, much smaller than the current global stock of ^{229}Th (tens of grams [1]). The
 221 total pumping rate is $\mathcal{R} \equiv N_{\text{Th}}R \propto S l \mathcal{E}^4$, while the total power of the pumping laser is $P_{\text{in}} \propto$
 222 $S \mathcal{E}^2$, yielding $\mathcal{R} \propto \frac{P_{\text{in}}^2 l}{S}$. Hence, a smaller S improves \mathcal{R} , as it is typical for nonlinear two-
 223 photon processes [34]. Considering that the wavelength of the pumping laser is around 300 nm,
 224 we suggest using $S = [1\mu\text{m}]^2$. On the other hand, the length l should not be too small,
 225 because the performance of the pulsed nuclear laser, including peak power and number of
 226 photons per pulse, increases with l . For demonstrative purposes, we will use $l = 1$ mm
 227 hereafter, but we have not optimized these parameters. The gain medium is thus a nanowire of
 228 size $1\mu\text{m} \times 1\mu\text{m} \times 1\text{mm}$. Actually, nanowire lasers have been demonstrated before based on
 229 electronic transitions [49].

230



231

232 **Figure 3** (a) A simplified setup of the nuclear laser. (b) Time evolution of the output power of the
 233 nuclear laser. The parameters of the nuclear laser are described in the main text. The shaded area
 234 indicates a nuclear laser pulse.

235

236 We further assume that the nanowire gain medium is confined by two mirrors on two sides,
 237 which forms an optical cavity (Figure 3a). The left mirror is a total reflector with 100%
 238 reflectivity for the 8.3 eV cavity photon, while the right mirror is a partial reflector with a
 239 transmissivity of T , which serves as the output channel of the cavity photons. The stimulated
 240 emission (absorption) rate of the cavity photons can be expressed as $K =$

241 $\frac{4|\langle gr, m_{gr} | \mathcal{M}_{M1} | is, m_{is} \rangle|^2 \mathcal{B}_{zpf}^2}{\Gamma_0}$, where $\Gamma_0 \sim \gamma_{\text{is}}^{\text{decay}} + \gamma_{\text{is}}^{\text{drift}}$ is the total broadening of the isomeric

242 states, \mathcal{M}_{M1} is the nuclear magnetic dipole transition dipole, while $\mathcal{B}_{zpf} = \sqrt{\frac{\mu_0 \omega_{\text{is}}}{2V}}$ is the

243 zero-point magnetic field of the cavity photon with μ_0 the vacuum permittivity. Note that we
244 only consider the $M1$ channel, as it is much more efficient than the $E2$ channel for radiative
245 transitions that do not involve electrons [32].

246 The performance of the nuclear laser can be evaluated using the semi-classical rate equations
247 (details in Section 4.1 in Ref. [28]). A typical time evolution of the output power P_{out} is
248 plotted in Figure 3b, where one can clearly see a nuclear laser pulse with a peak power of above
249 1 Watt and a duration of about 10 ns (shaded area in Figure 3b). A total of 4.6×10^9
250 nuclear gamma photons (8.3 eV) will be emitted per pulse. Because these highly coherent and
251 collinear photons come from the stimulated emission of the nuclear excited states, our device
252 would qualify as a gamma-ray laser (“graser”). In Section 4.2 of Ref. [28], we also show that
253 the losses and temperature rise in the gain medium are minor and would not influence the
254 operation of the nuclear laser.

255 Here we would like to remark on some potential challenges in constructing the nuclear laser
256 proposed in this work. First, the nuclear lasing requires fast pumping of the nuclear isomeric
257 state, so a ~ 4.1 eV UV- B laser with narrow linewidth and high power, which are assumed
258 to be 10 kHz and 1 W in the discussions above, would be necessary. Such a laser would be
259 challenging to build. But we expect it could be easier than building an ~ 8.3 eV F- UV laser.
260 Additionally, our proposal implicitly assumes that the isomeric transition energy ω_{is} is known
261 with high precision, which has not been realized yet. Fortunately, ω_{is} has been measured with
262 increasing precision [50] recently. Potentially, the two-photon ONQ pumping proposed in the
263 work can be used to measure ω_{is} . To this purpose, one needs a pumping laser with tunable
264 frequency. On the other hand, the pumping rate does not necessarily need to be high, so a laser
265 with relatively wide linewidth and low output power may be sufficient.

266 In summary, we propose a two-photon pumping scheme to populate the long-lived nuclear
267 isomers $^{229\text{m}}\text{Th}$ based on the ONQ effect in solid crystals. This pumping scheme could be used
268 in nuclear clocks based on ^{229}Th as well. We further propose a nuclear gamma-ray laser (as this
269 emission originates from nuclear isomeric transition) that utilizes ultrawide bandgap ^{229}Th -
270 compounds as the gain medium and a two-step, two-photon scheme to achieve population
271 inversion at room temperature. Pulsed nuclear lasing should be realizable with a Watt-level
272 pumping laser. The nuclear laser with narrow linewidth might be useful for various applications
273 in e.g., nano-imaging, nuclear clock, and quantum information processing.

274

275

276

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281

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