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# In-band pumped Kerr-lens mode-locked Tm,Ho-codoped calcium aluminate laser

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**Abstract:** We report on a Kerr-lens mode-locked Tm,Ho-codoped calcium aluminate laser with in-band pumping of the Tm ions by a spatially single-mode 1678 nm Raman fiber laser. The structurally disordered CaGdAlO<sub>4</sub> host crystal is also codoped also with the passive Lu ion for additional inhomogeneous line broadening. The Tm,Ho,Lu:CaGdAlO<sub>4</sub> laser generates soliton pulses as short as 79 fs at a central wavelength of 2073.6 nm via soft-aperture Kerr-lens mode-locking. The corresponding average output power amounts to 91 mW at a pulse repetition rate of ~86 MHz. The average output power can be scaled to 842 mW at the expense of slightly longer pulses of 155 fs at 2045.9 nm, which corresponds to a peak power of ~58 kW. To the best of our knowledge, this represents the first demonstration of an in-band pumped Kerr-lens mode-locked Tm,Ho solid-state laser at ~2 μm.

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## 1. Introduction

Mode-locked (ML) solid-state lasers emitting femtosecond pulses in the 2-μm spectral range at high (~100 MHz) repetition rates present unique opportunities for various applications. Such laser sources are interesting for time-resolved molecular spectroscopy, pump probe studies of semiconductor structures, and characterization of ultrafast detectors. At higher output powers, they can serve as pump sources for synchronously pumped optical parametric oscillators in the mid-infrared (mid-IR) spectral range beyond 5 μm, including frequency comb generation. They can also be used as seed sources for broadband, near-degenerate chirped-pulse optical parametric amplifiers (CPOAs) pumped near 1 μm, facilitating the generation of high-order harmonics and soft X-rays, or for regenerative laser amplifiers, boosting the single pulse energy at lower repetition rates. Finally, femtosecond ML solid-state lasers at ~2 μm are feasible for generating mid-IR supercontinuum and terahertz (THz) radiation [1].

Laser emission around 2 μm is commonly achieved using thulium (Tm<sup>3+</sup>) and holmium (Ho<sup>3+</sup>) ion based solid-state lasers according to the <sup>3</sup>F<sub>4</sub> → <sup>3</sup>H<sub>6</sub> (Tm<sup>3+</sup>) and <sup>5</sup>I<sub>7</sub> → <sup>5</sup>I<sub>8</sub> (Ho<sup>3+</sup>)

48 electronic transitions. Lasers employing singly  $\text{Tm}^{3+}$  (or  $\text{Ho}^{3+}$ ) doped as well as  $\text{Tm}^{3+}, \text{Ho}^{3+}$   
49 codoped materials exist. The latter approach is beneficial for several reasons: (i) the  $\text{Tm}^{3+}$  ion  
50 offers more convenient pump wavelengths available from AlGaAs diodes combined with  
51 efficient non-radiative energy transfer of the electronic excitation to the  $\text{Ho}^{3+}$  ion,  
52  ${}^3\text{F}_4(\text{Tm}^{3+}) \rightarrow {}^5\text{I}_7(\text{Ho}^{3+})$ ; (ii) the emission of the  $\text{Ho}^{3+}$  ion extends beyond  $2 \mu\text{m}$  thus avoiding the  
53 structured absorption of water vapors in atmospheric air which may prevent femtosecond pulse  
54 generation; (iii) the combined gain bandwidths of both ions may contribute to additional pulse  
55 shortening [1]. On the negative side, the substantial heat load caused by energy-transfer  
56 upconversion (ETU) from the metastable states of both active ions and energy losses due to the  
57 back energy transfer present some challenges for  $\text{Tm}^{3+}, \text{Ho}^{3+}$  codoped materials. It is therefore  
58 important to investigate new host materials that combine excellent spectroscopic and thermo-  
59 mechanical properties.

60 The tetragonal (sp. gr.  $I4/mmm$ ) calcium rare-earth (*RE*) aluminate  $\text{CaREAlO}_4$  crystals  
61 where *RE* denotes Gd or Y, doped with  $\text{Tm}^{3+}$  and  $\text{Ho}^{3+}$  ions represent one of the most promising  
62 laser materials for generating ultrashort pulses slightly above  $2 \mu\text{m}$  [2]. The Gd- compound is  
63 abbreviated in the literature as CALGO and its Y counterpart – as CALYO. The doping of these  
64 crystals with laser-active lanthanide (*Ln*) ions is relatively easy due to the presence of the *RE*  
65 sites. Both crystals melt congruently which facilitates their growth by the conventional  
66 Czochralski (Cz) method [3, 4]. The  $\text{CaREAlO}_4$  crystals are optically uniaxial, and their  
67 intrinsic birefringence and notable polarization anisotropy of the spectroscopic properties of  
68 the dopant ions result in linearly polarized laser emission with weak depolarization losses in  
69 high power laser operation (for *a*-cut crystals). Calcium *RE* aluminates crystallize in a  $\text{K}_2\text{NiF}_4$ -  
70 type structure with the  $\text{Ca}^{2+}$  and  $\text{RE}^{3+}$  cations statistically distributed over the same Wyckoff  
71 site ( $4e, C_{4v}$ -symmetry) leading to structural disorder [5]. Despite the disordered structure, the  
72  $\text{CaREAlO}_4$  crystals still possess relatively high thermal conductivity with a weak dependence  
73 on the active  $\text{RE}^{3+}$  doping level [6, 7].

74 When co-doped with  $\text{Tm}^{3+}$  and  $\text{Ho}^{3+}$  ions, these crystals exhibit inhomogeneous broadening  
75 of absorption and emission spectral bands [8]. Due to their broad, flat and smooth gain profiles,  
76 they are extremely suitable laser materials for sub-100 fs pulse generation at  $\sim 2 \mu\text{m}$ . Pumped  
77 with a continuous-wave (CW) Ti:sapphire laser at 795.3 nm, a Tm,Ho:CALYO laser ML by a  
78 Semiconductor Saturable Absorber Mirror (SESAM) generated 87 fs pulses at 2042.6 nm with  
79 an average output power of 27 mW [9]. Shorter pulses of 52 fs at 2015 nm with an average  
80 output power of 376 mW were obtained from a SESAM ML Tm,Ho:CALGO laser using the  
81 same pump source [8].

82 The structural disorder characteristic of CALGO and CALYO crystals can be augmented  
83 by a compositional disorder, resulting in additional inhomogeneous spectral line broadening.  
84 This contributes to smoothening and flattening of the gain profiles of the  $\text{Tm}^{3+}$  and  $\text{Ho}^{3+}$  ions.  
85 Compositional disorder can be attained e.g. by introducing optically passive  $\text{Lu}^{3+}$  ions into the  
86 CALGO host [10]. Note that the doping level of Lu is at present limited since the stoichiometric  
87 compound  $\text{CaLuAlO}_4$  does not exist. Still the larger ionic radius difference to the Gd host  
88 forming ion (compared to Y) means that larger effect can be expected by Lu doping of CALGO  
89 compared to CALYO. It was shown that such Tm,Ho,Lu:CALGO crystals feature an additional  
90 spectral broadening mechanism due to the compositional disorder leading to red-shifted gain  
91 profiles [11]. Using a GaSb-based SESAM as a saturable absorber (SA) and a Ti:sapphire laser  
92 tuned to 801.3 nm as a pump source, a Tm,Ho,Lu:CALGO laser delivered soliton-like pulses  
93 as short as 46 fs at 2033 nm with an average output power of 121 mW at 78 MHz [12].

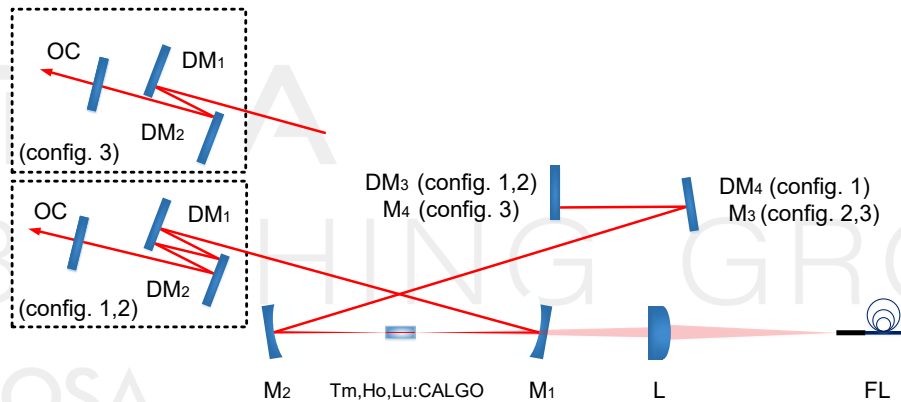
94 In contrast to the traditional pumping of  $\text{Tm}^{3+}$ -doped laser materials near 800 nm which  
95 corresponds to the  ${}^3\text{H}_6 \rightarrow {}^3\text{H}_4$  transition in absorption, we explore the direct excitation of the  
96  $\text{Tm}^{3+}$  ion to the  ${}^3\text{F}_4$  state ( ${}^3\text{H}_6 \rightarrow {}^3\text{F}_4$ , the so-called Tm in-band pumping). This approach offers  
97 a reduction in the heat load by avoiding the non-radiative relaxation path from the  ${}^3\text{H}_4$  and  ${}^3\text{H}_5$   
98 excited-states of  $\text{Tm}^{3+}$ . In-band pumping has been employed before for Tm,Ho codoped  
99 materials in  $\sim 2 \mu\text{m}$  solid-state lasers in the bulk and waveguide geometries for achieving

100 efficient and power scalable operation in the CW regime [13, 14]. More recently also the ML  
 101 regime was studied using a SESAM [15-17]. In the present work we demonstrate that in-band  
 102 pumping is feasible also for Kerr-lens mode-locked (KLM) Tm,Ho lasers.

103 We demonstrate, to the best of our knowledge, the first in-band pumped solid-state KLM  
 104 laser emitting femtosecond pulses beyond 2  $\mu\text{m}$ , employing a Tm,Ho codoped crystal. The  
 105 Tm,Ho,Lu:CaGdAlO<sub>4</sub> laser generates soliton pulses as short as 79 fs at 2073.6 nm, with an  
 106 average output power scalable to almost the Watt-level at longer pulse durations of 155 fs  
 107 centered at 2045.9 nm.

## 108 2. Experimental setup

109 KLM operation was investigated in the X-folded standing-wave cavity depicted in Fig. 1. A 6-  
 110 mm long rectangular element was cut from a high-quality Tm,Ho,Lu:CaGdAlO<sub>4</sub> crystal with  
 111 measured doping levels of 4.48 at.% Tm<sup>3+</sup>, 0.54 at.% Ho<sup>3+</sup>, and 5.51 at.% Lu<sup>3+</sup> corresponding  
 112 to a chemical formula of CaGd<sub>0.8947</sub>Lu<sub>0.0551</sub>Tm<sub>0.0448</sub>Ho<sub>0.0054</sub>AlO<sub>4</sub> to enable light propagation  
 113 along the *a*-axis (*a*-cut). Its 3 × 3 mm<sup>2</sup> input/output faces were polished to laser-grade quality  
 114 and anti-reflection coated for both the pump and laser wavelengths. The sample was mounted  
 115 in a water-cooled copper holder kept at a temperature of 12°C and placed between two dichroic  
 116 folding mirrors M<sub>1</sub> and M<sub>2</sub> (radius of curvature, RoC = -100 mm).



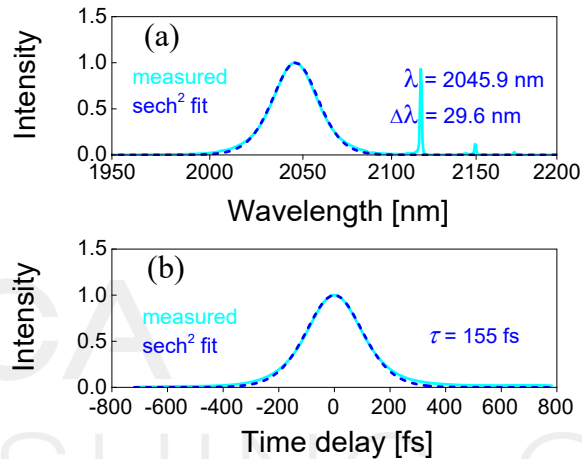
117  
 118 **Fig. 1.** Schematic of the KLM Tm,Ho,Lu:CALGO laser. FL: Raman-shifted Er-fiber laser; L:  
 119 spherical focusing lens; M<sub>1</sub> and M<sub>2</sub>: concave mirrors; M<sub>3</sub> and M<sub>4</sub>: plane mirrors; DM<sub>1</sub> - DM<sub>4</sub>:  
 120 flat dispersive mirrors; OC: plane-wedged output coupler.

121 We employed a home-made spatially single-mode 6 W Raman-shifted Er-fiber laser  
 122 emitting unpolarized radiation at 1678 nm [17] as a pump source to address the <sup>3</sup>H<sub>6</sub> → <sup>3</sup>F<sub>4</sub> Tm<sup>3+</sup>  
 123 transition. Its linewidth (full width at the half maximum, FWHM) was 0.8 nm and the  
 124 maximum incident pump power reached 5.5 W. The pump radiation was focused into the  
 125 crystal with a spherical focusing lens (*f* = 75 mm), resulting in a beam waist (radius) of 18  $\mu\text{m}$ .  
 126 Four flat dispersive mirrors (DMs) with negative group delay dispersion (GDD) of -1000 fs<sup>2</sup>  
 127 per bounce at ~2  $\mu\text{m}$  were used to balance the self-phase modulation (SPM) induced by the  
 128 Kerr nonlinearity of the laser crystal for soliton-like pulse shaping.

## 129 3. Mode-locked laser operation

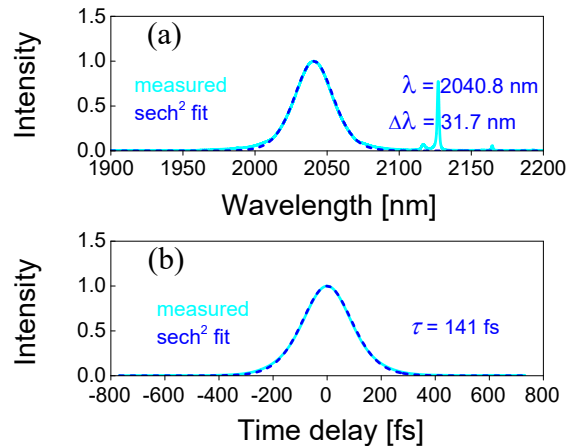
130 KLM operation was initially investigated by implementing four DMs providing an overall  
 131 negative GDD of -11000 fs<sup>2</sup> and an output coupling *T*<sub>OC</sub> of 3%, see Fig. 1 (configuration 1).  
 132 The cavity geometry was adjusted with enlargement of the separation between the two folding  
 133 mirrors surrounding the laser crystal by a few hundred microns, positioning it close to the edge  
 134 of the stability region. Stable KLM was then initiated by either knocking on the output coupler  
 135 (OC) or translating the flat rear mirror (DM<sub>3</sub> or M<sub>3</sub>). Soliton pulses were directly generated  
 136 from the ML laser, producing an average output power of 842 mW. The absorbed pump power

137 in a single pass was estimated to be 3.72 W. The optical spectrum of the ML laser had a  
 138 bandwidth (FWHM) of 29.6 nm at a central wavelength of 2045.9 nm, assuming a sech<sup>2</sup>-shaped  
 139 spectral profile, see Fig. 2(a). The autocorrelation trace (AC) based on second harmonic  
 140 generation (SHG) gave a deconvolved pulse duration of 155 fs (FWHM) assuming a sech<sup>2</sup>-  
 141 shaped temporal intensity profile, as shown in Fig. 2(b). The corresponding time-bandwidth  
 142 product (TBP) was 0.329, which only slightly exceeded the value for ideal unchirped soliton  
 143 pulses (0.315). The physical cavity length was ~1.75 m, resulting in a pulse repetition rate of  
 144 82.57 MHz. The corresponding peak power amounted to 57.9 kW. The presence of a narrow  
 145 satellite peak at ~2117 nm is attributed to the limited reflectivity bandwidth of the cavity  
 146 mirrors and the associated abrupt change in the GDD. This could potentially be improved by  
 147 using new cavity mirrors with optimized spectral coverage.



148 **Fig. 2.** KLM Tm,Ho,Lu:CALGO laser: (a) optical spectrum and (b) autocorrelation trace for  
 149  $T_{OC} = 3\%$ . Dashed curves - sech<sup>2</sup> fits.  
 150

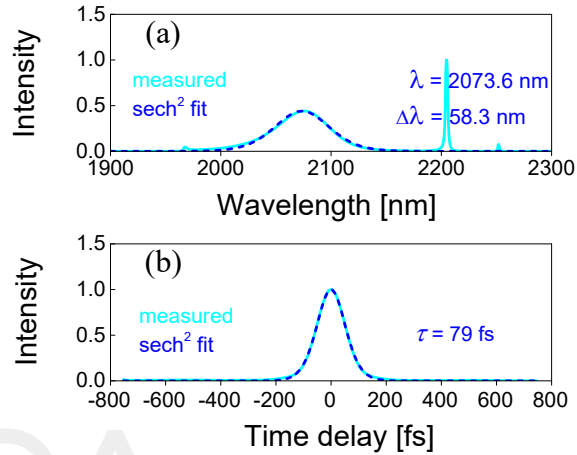
151 By reducing  $T_{OC}$  to 1.5% and optimizing the total negative GDD to -9000 fs<sup>2</sup> by a different  
 152 set of DMs, cf. Fig. 1 (configuration 2), slightly shorter pulses were achieved. Their  
 153 characteristics are displayed in Fig. 3.



154 **Fig. 3.** KLM Tm,Ho:Ca(Gd,Lu)AlO<sub>4</sub> laser: (a) optical spectrum and (b) autocorrelation trace  
 155 for  $T_{OC} = 1.5\%$ .  
 156

157 Assuming a sech<sup>2</sup>-shaped spectral intensity profile, the emission bandwidth (FWHM)  
 158 increased to 31.7 nm at a central wavelength of 2040.8 nm, see Fig. 3(a). The recorded SHG-

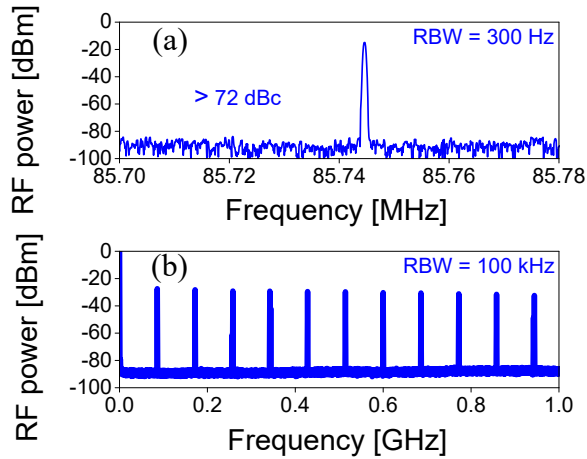
159 based intensity AC closely matched a sech<sup>2</sup>-shaped temporal intensity profile, resulting in an  
 160 estimated pulse duration of 141 fs (FWHM), see Fig. 3(b). The corresponding TBP was  
 161 calculated to be 0.322, very close to the Fourier-transform-limit. Under these conditions, the  
 162 average output power reached 398 mW, for an absorbed pump power of 3.86 W, corresponding  
 163 to a peak power of 29 kW. A similar satellite peak was observed in this case, too, related again  
 164 to unmanageable GDD.



165  
 166 **Fig. 4.** KLM Tm,Ho:Ca(Gd,Lu)AlO<sub>4</sub> laser: (a) optical spectrum and (b) autocorrelation trace  
 167 for  $T_{oc} = 0.5\%$ .

168 The shortest pulse duration was achieved by further reducing the OC transmittance to 0.5%  
 169 and decreasing the overall negative GDD to -4000 fs<sup>2</sup>, see Fig. 1 (configuration 3). The  
 170 characteristics of the shortest pulses are shown in Fig. 4. The laser spectrum exhibited a FWHM  
 171 as large as 58.3 nm, centered at 2073.6 nm, see Fig. 4(a). The pulse duration was assessed from  
 172 the recorded SHG-based intensity AC trace, which was almost ideally fitted with a sech<sup>2</sup>-shaped  
 173 temporal intensity profile, yielding a deconvolved FWHM of 79 fs. The corresponding TBP  
 174 was 0.321. Under these conditions, the maximum average output power dropped to 91 mW, for  
 175 an absorbed pump power of 3.1 W, corresponding to a peak power of 11.8 kW. The narrow  
 176 satellite peak was more pronounced in this case, and it could not be suppressed by the cavity  
 177 alignment. The limitation for further pulse shortening in this KLM laser is mainly related to the  
 178 reflectivity bandwidth of the pump mirror (M<sub>1</sub>) which exhibits a significant reflectivity  
 179 reduction well below 2000 nm. We believe that this could be improved in the future with  
 180 broadband pump mirror as well as optimized GDD management.

181 The radio frequency (RF) spectra of the shortest pulses were recorded to verify the stability  
 182 of the ML operation in different frequency span ranges, as shown in Fig. 5. The fundamental  
 183 beat note located at 85.74 MHz exhibits a high extinction ratio of >72 dBc above carrier, see  
 184 Fig. 5(a). The uniform harmonics on a 1-GHz frequency span reveal high stability of the single-  
 185 pulse CW-ML operation without any Q-switching instabilities, see Fig. 5(b).



**Fig. 5.** RF spectra of the KLM Tm,Ho,Lu:CALGO laser: (a) fundamental beat note at 85.74 MHz recorded with a resolution bandwidth (RBW) of 300 Hz, and (b) harmonics on a 1-GHz frequency span recorded with a RBW of 100 kHz.

**Table 1. Output Characteristics of the KLM Tm,Ho,Lu:CALGO Laser**

$T_{OC}$ , %	$P_{out}$ , mW	$P_{abs}$ , W	$\Delta\tau$ , fs	$\lambda_{em}$ , nm	$\Delta\lambda_{em}$ , nm	TPB
3	842	3.72	155	2045.9	29.6	0.329
1.5	398	3.86	141	2040.8	31.7	0.322
0.5	91	3.1	79	2073.6	58.3	0.321

$T_{OC}$  – output coupler transmittance,  $P_{out}$  – average output power,  $P_{abs}$  – absorbed pump power,  $\Delta\tau$  – pulse duration (FWHM),  $\lambda_{em}$  – central emission wavelength,  $\Delta\lambda_{em}$  – emission bandwidth (FWHM), TBP – time bandwidth product.

In Table 1, we summarize the output characteristics of the KLM Tm,Ho,Lu:CALGO laser achieved in the present work. The laser polarization for all the studied OCs was  $\sigma$  and it was naturally selected due to its higher laser gain cross-section. In all aspects the present results were superior to the SESAM ML laser based on the same material and pump source [17].

#### 4. Conclusion

To conclude, Tm in-band pumping directly to the  $^3F_4$  state is a viable route for attaining power scaling to the Watt-level of Kerr-lens mode-locked Tm,Ho crystalline lasers. It allows to suppress the thermal effects in the gain medium. This pumping scheme can be implemented by using Raman-shifted Er-fiber or short wavelength Tm-fiber lasers. In the present work, a proof-of-principle of this concept of ultrafast laser development is demonstrated with a Tm $^{3+}$  and Ho $^{3+}$  codoped tetragonal calcium rare-earth aluminate crystal Ca(Gd,Lu)AlO $_4$  combining attractive thermo-optical and spectroscopic properties. The latter properties stem from its highly disordered nature: both structural disorder (a random distribution of Ca $^{2+}$  and Ln $^{3+}$  cations over the same Wyckoff site) and compositional one (a partial replacement of Gd $^{3+}$  by Lu $^{3+}$ ). Soliton pulses as short as 79 fs were generated at 2073.6 nm with an average output power of 91 mW and a pulse repetition rate at 85.74 MHz. The average output power could be further scaled up to 842 mW at the expense of somewhat longer pulse duration of 155 fs. It shall be emphasized that the crystal *a*-cut for the present work was selected to utilize the  $\pi$ -polarization for higher absorption of the unpolarized pump beam. Further improvements can be expected employing polarized pump sources. It shall be also outlined that for other crystalline hosts, in-band pumping can have an additional benefit of higher pump efficiency when the absorption of the  $^3F_4$  level of Tm is higher compared to the ( $^3H_4$ ) 800 nm range [18].

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222 **Disclosures.** The authors declare no conflicts of interest.

223 **Data availability.** Data underlying the results presented in this paper are not publicly available at this time but may  
224 be obtained from the authors upon reasonable request.

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