

Hyperbolic phonon polaritons with positive and negative phase velocities in suspended $\alpha\text{-MoO}_3$

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ABSTRACT

Sample suspension is a valuable method to improve the mechanical, thermal, electronic, and optical properties of low-dimensional materials. In terms of confined light-matter waves—the polaritons, sample suspension can elongate the wavelength of polaritons with a positive phase velocity. Previous work demonstrates a wavelength elongation of $\sim 10\%$ for hyperbolic phonon polaritons (HPPs) in uniaxial crystals of hexagonal boron nitride (hBN). In this work, we report the alteration of HPPs in biaxial α -phase molybdenum trioxide (α -MoO₃) by sample suspension. Our combined infrared nano-imaging experiments and electromagnetic theory reveal a wavelength elongation $> 60\%$ and a propagation length increase $> 140\%$, due to the simultaneous wavelength elongation and dissipation elimination in the suspended specimen. We have also examined HPPs in α -MoO₃ with a negative phase velocity. The sample suspension shortens the HPP wavelength and simultaneously reduces the dissipation due to the unique permittivity tensor. The HPPs with improved figures of merits in the suspended specimen may be developed for nano-polaritonic circuits, biochemical sensing, emission engineering, and energy transfer.

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Phonon polaritons^{1–4} are hybrid electromagnetic waves mixed by lattice vibrations and free-space photons in polar materials. These light-matter waves possess large optical confinement—a much smaller wavelength λ than free-space light λ_0 —and, therefore, facilitate the control of wave properties, optical energy, and light-matter interactions at small scales. Importantly, phonon polaritons are immune to Ohmic loss to provide complementary merits to their electronic counterpart—plasmon polaritons. In addition to the virtues of phonon polaritons established in conventional polar systems,^{5–7} recent investigations of phonon polaritons in van der Waals (vdW) materials revealed new advances.^{8–20} In particular, anisotropic lattice vibrations in vdW materials lead to natural hyperbolicity^{9,10,15} where principal components of the permittivity tensor have opposite signs: $\epsilon_i \epsilon_j < 0$, $i, j = x, y$, or z . The natural hyperbolicity emerges with multiple branches of phonon polaritons that provide an exceptionally high photonic density of states and optical access at the interatomic length scale. These

advances have been utilized for nanophotonic applications, including super-resolution imaging,^{11,21} efficient energy control,²² and extreme light manipulation.^{23,24}

Further developments of hyperbolic phonon polaritons (HPPs) require the effective altering and tuning of these nanoscale light-matter modes. Due to their lattice vibration nature, HPPs can be altered by either changing the intrinsic properties of the HPPs materials, such as element intercalation^{25–27} and carrier injection,²⁸ or engineering the surrounding permittivity by implementing graphene,²⁹ phase change materials,^{30–33} or sample suspension.^{34–36} Among all the approaches, sample suspension offers the simultaneous dispersion altering and dissipation reduction of HPPs by eliminating dielectric losses from the substrates. Prior works^{34–36} on suspended hexagonal boron nitride (hBN) reported a $\sim 10\%$ wavelength elongation $\Delta\lambda$ of HPPs with positive phase velocity ($v_{\text{phase}} > 0$) in uniaxial materials ($\epsilon_x = \epsilon_y \neq \epsilon_z$). In this work, we investigated the sample suspension on

altering HPPs with both positive and negative phase velocity, in a representative biaxial material ($\epsilon_x \neq \epsilon_y \neq \epsilon_z$)— α -phase molybdenum trioxide (α -MoO₃).^{16,18,27} The response of biaxial materials differs in every axis in the Cartesian coordinate and can offer rich degrees of freedom to engineer physical properties in vdW heterostructures. Our work on suspended α -MoO₃ demonstrates a wavelength elongation of HPPs $\Delta\lambda$ more than 60% and a propagation length elongation ΔL up to 140%. The wavelength of HPPs also reveals the opposite altering effect: it is elongated ($\Delta\lambda > 0$) with positive phase velocity $v_{\text{phase}} > 0$ while shortened ($\Delta\lambda < 0$) with negative phase velocity $v_{\text{phase}} < 0$.

Hyperbolic phonon polaritons (HPPs) were investigated by nano-imaging suspended α -MoO₃ using scattering-type scanning near-field optical microscopy [s-SNOM, Fig. 1(a)]. The suspended α -MoO₃ devices were fabricated by mechanical exfoliation and the polymer-based dry transfer method.³⁷ Thin slabs of α -MoO₃ were first mechanically exfoliated from bulk crystals synthesized by thermal physical deposition.³⁸ Due to the anisotropic crystal structure, the crystallographic axes of α -MoO₃ can be identified^{16,18,27} according to the long straight edge of the exfoliated slabs using optical microscopy and atomic force microscopy (AFM). The exfoliated α -MoO₃ slab with a thickness of 247 nm was transferred onto the Si/SiO₂ substrate with 3 μm wide air trenches to suspend part of the α -MoO₃ slab [Fig. 1(a)]. s-SNOM was then performed to image the standing wave fringes of HPPs in the α -MoO₃ device. In the s-SNOM experiment [Fig. 1(a)],^{10,16–18,29,39–43} an AFM tip was illuminated by infrared (IR) lasers and acted as an antenna⁴⁴ to generate strong optical near-fields underneath the tip apex. These strong near-fields bridge the momentum mismatch and transfer energy between free space photons and HPPs. HPPs were, therefore, launched from the AFM tip and propagated inside the α -MoO₃ slab. The propagating HPPs got reflected at crystal edges, interfered with newly launched HPPs from the AFM tip, and formed standing wave interferences between the crystal edges and the AFM tip. Once the α -MoO₃ device is scanned underneath the tip, these standing wave interferences were recorded as HPP fringes parallel to the crystal edges in the s-SNOM amplitude $S(\omega)$ images [Figs. 1(b) and 1(c)]. The period of the imaged fringes Δ equals to half of the HPP wavelength $\Delta = \lambda/2$.^{10,16–18,29,39–43}

We performed the s-SNOM imaging on suspended α -MoO₃ in two Reststrahlen bands: the lower band with the frequency ω from

820 to 972 cm^{-1} and the upper band with ω from 958 to 1004 cm^{-1} . In the lower band, principal components of the permittivity tensor have opposite signs: $\epsilon_x < 0$, $\epsilon_y > 0$, and $\epsilon_z > 0$. HPPs in α -MoO₃ possess a positive phase velocity $v_{\text{phase}} > 0$ ¹² and an open hyperbolic “X” shape wavefront. HPP fringes, therefore, can only be observed parallel to the y [001] direction [Fig. 1(b)], with the strongest fringe closest to the edge followed by damped ones away from the edge (white dotted line). Although sharing a similar oscillation form, HPPs in suspended α -MoO₃ exhibit a longer fringe period (therefore a longer wavelength) than those in supported α -MoO₃. A direct signature from the s-SNOM image [Fig. 1(b)] is HPP fringes in suspended α -MoO₃ can still be observed far away from the crystal edge, whereas those in the supported α -MoO₃ were already damped. Quantitative details of the s-SNOM results can be obtained from the line profiles [Fig. 2(a)] cut along dashed lines in the s-SNOM image. At a representative frequency $\omega = 920 \text{ cm}^{-1}$, HPP wavelength in suspended α -MoO₃ is $\lambda_{\text{sus}} = 1280 \text{ nm}$, while in supported α -MoO₃, $\lambda_{\text{sup}} = 800 \text{ nm}$. Both s-SNOM line profiles and their Fourier Transform [FT, Fig. 2(b)] reveal a wavelength elongation $\Delta\lambda = (\lambda_{\text{sus}} - \lambda_{\text{sup}})/\lambda_{\text{sup}} = 60\%$ by sample suspension, much larger than $\Delta\lambda \sim 12\%$ reported in hBN.^{34–36} The electromagnetic field of HPPs spans from the inside to the outside of the polaritonic medium (see the [supplementary material](#) Sec. II for details). Sample suspension changes the outside permittivity and, thus, can alter the properties of HPPs. The more significant wavelength elongation by suspending α -MoO₃ is attributed to the smaller confinement of HPPs in α -MoO₃ than hBN. The elongation of the HPP wavelength was also reported recently in gradually suspended α -MoO₃.⁴⁵ In the following, we provide combined experimental and theoretical results revealing the alteration of HPP wavelength and propagation length over the broad Reststrahlen bands. This evident wavelength elongation has been observed at other frequencies inside the lower Reststrahlen band, see the frequency ω —confinement λ_0/λ and elongation $\Delta\lambda$ dispersion in Figs. 2(d)–2(e). We also modeled the HPP dispersion using Fresnel and Fabry–Pérot theory (see the [supplementary material](#) Sec. I). These modeling results [curves in Figs. 2(d) and 2(e)] agree well with our s-SNOM data [dots in Figs. 2(d) and 2(e)]. Note that in addition to the tip-launched HPP fringe period $\Delta = \lambda/2$, fringe period $\Delta = \lambda$ originating from the edge-launched HPPs⁴⁶ also appeared; see the corresponding α and β peaks in the FT of the

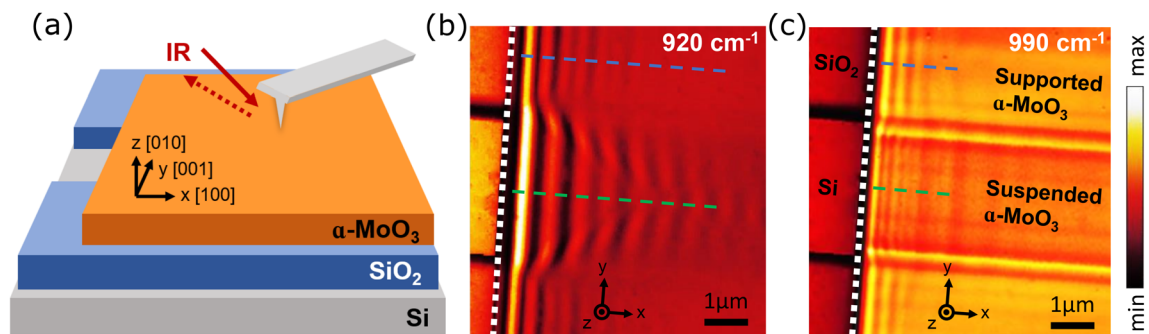


FIG. 1. Scattering-type scanning near-field optical microscopy (s-SNOM) imaging of hyperbolic phonon polaritons (HPPs) in suspended and supported α -MoO₃. (a) Schematics of the s-SNOM imaging experiment on the partially suspended α -MoO₃ slab. (b) s-SNOM amplitude image of HPPs in the lower Reststrahlen band at the frequency $\omega = 920 \text{ cm}^{-1}$. (c) s-SNOM amplitude image of HPPs in the upper Reststrahlen band at the frequency $\omega = 990 \text{ cm}^{-1}$. Line profiles of the s-SNOM data in Figs. 2(a) and 3(a) were extracted along dashed lines and averaged with a width of 2 μm in Fig. 1(c), 2 μm along the blue dashed line, and 200 nm along the green dashed line in Fig. 1(b). Scale bar: 1 μm .

s-SNOM line profiles in Fig. 2(b). Nevertheless, since tip-launched HPPs with $\Delta = \lambda/2$ dominate in the s-SNOM data (the β peak is stronger than the α peak), our analysis will rely on them.

Alongside the elongation of HPP wavelength λ , sample suspension also elongates the propagation length L of HPPs in α -MoO₃. In Fig. 2(c), s-SNOM line profiles from suspended and supported α -MoO₃ were replotted by inverse FT¹¹ of the β peaks [Fig. 2(b)] and then fitted with the envelope of the damped sinusoidal function:^{46,47}

$$S(\omega) = Ax^{-1/2}e^{-x/L}, \quad (1)$$

where x is the distance from the edge and A is a constant. The propagation length L relates to the wavelength λ and dissipation γ : $L = \lambda/4\pi\gamma$. With the wavelength λ input from the experiment [Figs. 2(a) and 2(b)], L can be extracted at the best fit of Eq. (1) [dashed curves, Fig. 2(c)] to the s-SNOM data [solid curves, Fig. 2(c)]. At $\omega = 920 \text{ cm}^{-1}$, the propagation length of HPPs from suspended α -MoO₃ is $L_{\text{sus}} = 2037 \text{ nm}$, whereas in supported α -MoO₃, $L_{\text{sup}} = 848 \text{ nm}$. Remarkably, the elongation of HPP propagation length $\Delta L = (L_{\text{sus}} - L_{\text{sup}})/L_{\text{sup}}$ reaches 140% in our experiments, with the corresponding dissipation reduction $\Delta\gamma = (\gamma_{\text{sus}} - \gamma_{\text{sup}})/\gamma_{\text{sup}} \sim -33\%$. This evidently elongated propagation length L is attributed to the simultaneous wavelength elongation and dissipation reduction via the sample suspension. In Fig. 2(f), we summarize the propagation length elongation ΔL for HPPs in the lower Restrahlen band, where the s-SNOM data (black squares) agree well with the simulation (black curve), and both reveal an ΔL above 100% in the entire lower Restrahlen band.

In addition to the significantly improved figures of merits, the wavelength λ of HPPs in α -MoO₃ was alternated oppositely, in the

lower and upper Restrahlen bands, via the sample suspension. In the upper band, principal components of the permittivity tensor are $\epsilon_x > 0$, $\epsilon_y > 0$, and $\epsilon_z < 0$. HPPs in α -MoO₃ possess a negative phase velocity $v_{\text{phase}} < 0$ ¹² and a closed elliptic wavefront. At a representative IR frequency $\omega = 990 \text{ cm}^{-1}$ [Fig. 1(c)], HPP fringes parallel to the edge were observed in both suspended α -MoO₃ and supported α -MoO₃. In contrast to wavelength elongation $\Delta\lambda > 0$ in the lower band, in the upper band, the HPP wavelength gets shortened $\Delta\lambda < 0$ by sample suspension [Fig. 3(a)]: $\lambda_{\text{sus}} = 539 \text{ nm}$, $\lambda_{\text{sup}} = 875 \text{ nm}$, and $\Delta\lambda = (\lambda_{\text{sus}} - \lambda_{\text{sup}})/\lambda_{\text{sup}} = -38\%$ at $\omega = 990 \text{ cm}^{-1}$. This shortening of the HPP wavelength ($\Delta\lambda < 0$) was observed at other frequencies inside the upper Restrahlen band, see the combined s-SNOM data (dots) and modeling results (curve) in Figs. 3(d) and 3(e). In our experiments, the wavelength shortening reaches—40% in the upper Restrahlen band [Fig. 3(e)]. Note that while the sample suspension leads to opposite wavelength alteration in the lower and upper Restrahlen bands, it reduces the HPP dissipation in both bands due to the elimination of the substrate dissipation. In the upper band, the HPP dissipation was reduced from 0.06 to 0.045 via the sample suspension [Fig. 3(c)].

The opposite alteration of HPP wavelength was attributed to the difference in the permittivity tensor of α -MoO₃ in the lower and upper Restrahlen bands. HPPs in the α -MoO₃ slab correspond to the Fabry-Pérot quantization condition:⁴⁸

$$k = k_1 + ik_2 = \frac{\rho}{d} \left[\arctan\left(\frac{\rho}{\epsilon_z}\right) + \arctan\left(\frac{\epsilon_{\text{sub}}\rho}{\epsilon_z}\right) + \pi l \right], \quad (2)$$

$$\rho = i\sqrt{\epsilon_z/(\epsilon_x \cos^2 \alpha + \epsilon_y \sin^2 \alpha)}, \quad (3)$$

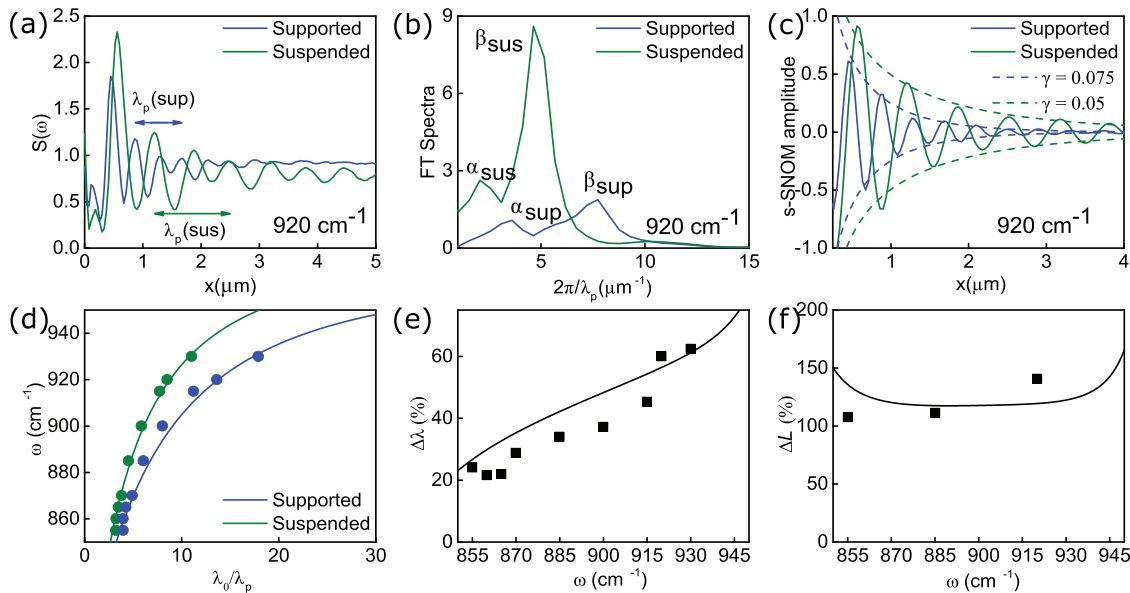


FIG. 2. Altering HPPs in α -MoO₃ by sample suspension in the lower Restrahlen band. a) s-SNOM line profiles taken along dashed lines in Fig. 1(b) from suspended α -MoO₃ and supported α -MoO₃. b) Fourier Transform (FT) of the s-SNOM line profiles in Fig. 2(a). α and β peaks correspond to the edge and tip launched HPPs, respectively.⁴⁶ c) Extracted s-SNOM line profiles containing only tip launched HPPs by inverse FT of the β peaks in Fig. 2(b). d) Frequency ω —confinement λ_0/λ_p dispersion of HPPs in suspended (green) and supported (blue) α -MoO₃. Experimental data are plotted with dots, and the modeling results are plotted with curves. e) The dependence of wavelength elongation $\Delta\lambda$ on the frequency ω . Experimental data are plotted with squares, and the modeling results are plotted with curves. f) The dependence of propagation length elongation ΔL on the frequency ω . Experimental data are plotted with squares, and the modeling results are plotted with curves.

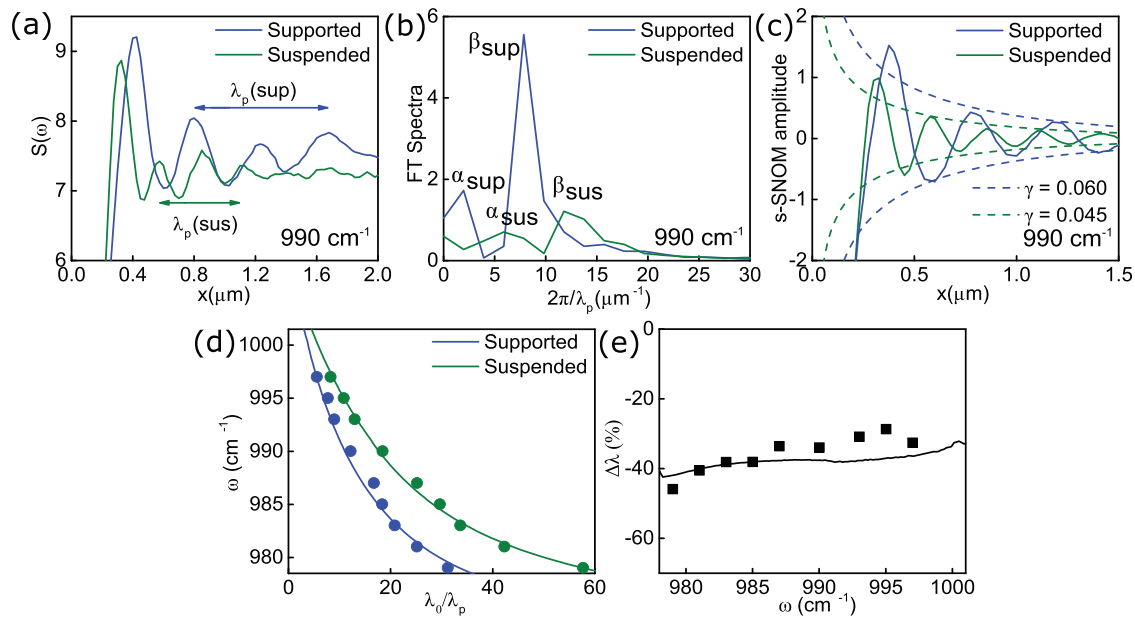


FIG. 3. Altering HPPs in α -MoO₃ by sample suspension in the upper Restrahlen band. (a) s-SNOM line profiles taken along dashed lines in Fig. 1(c) from suspended α -MoO₃ and supported α -MoO₃. (b) Fourier Transform (FT) of the s-SNOM line profiles in Fig. 3(a). α and β peaks correspond to the edge and tip launched HPPs, respectively.⁴⁶ (c) Extracted s-SNOM line profiles containing only tip launched HPPs by inverse FT of the β peaks in Fig. 3(b). (d) Frequency ω —confinement λ_0/λ_p dispersion of HPPs in suspended (green) and supported (blue) α -MoO₃. Experimental data are plotted with dots, and the modeling results are plotted with curves. (e) The dependence of wavelength elongation $\Delta\lambda$ on the frequency ω . Experimental data are plotted with squares, and the modeling results are plotted with curves.

where $k = k_1 + ik_2$ is the momentum of HPPs with an angle α to the $x[100]$ direction, and it relates to the HPP wavelength λ and dissipation γ with $k_1 = 2\pi/\lambda$ and $k_2 = \gamma k_1$. d is the thickness of α -MoO₃, $l = 0, 1, 2, \dots$, is the mode index, and in this work, we focus on the fundamental mode $l = 0$. ϵ_{sub} is the permittivity of the substrate. For HPPs with fringes parallel to the $y[001]$ direction, their momentum k is along the $x[100]$ direction, therefore $\alpha = 0$. The HPP wavelength λ depends on the substrate's permittivity ϵ_{sub} , see the derivative of Eq. (2),

$$\frac{\partial k_1}{\partial \epsilon_{\text{sub}}} = \frac{1}{d} \text{Re} \left(\frac{\epsilon_z}{\epsilon_{\text{sub}}^2 - \epsilon_z \epsilon_x} \right). \quad (4)$$

In the lower Restrahlen band, $\epsilon_x < 0$, $\epsilon_y > 0$, and $\epsilon_z > 0$, $\frac{\partial k_1}{\partial \epsilon_{\text{sub}}} > 0$, the sample suspension (substrate changed from SiO₂ $\epsilon_{\text{SiO}_2} > 1$ to air $\epsilon_{\text{air}} = 1$) corresponds to the wavelength elongation $\Delta\lambda > 0$. Whereas in the upper Restrahlen band, $\epsilon_x > 0$, $\epsilon_y > 0$, and $\epsilon_z < 0$, $\frac{\partial k_1}{\partial \epsilon_{\text{sub}}} < 0$, the sample suspension (substrate changed from SiO₂ $\epsilon_{\text{SiO}_2} > 1$ to air $\epsilon_{\text{air}} = 1$) corresponds to the wavelength shortening $\Delta\lambda < 0$.

Combined experimental data and theoretical results in Figs. 1–3 reveal the evident alteration of HPPs in α -MoO₃ by the sample suspension. The alteration of HPP wavelength $\Delta\lambda$ (%) and propagation length ΔL (%) in suspended α -MoO₃ is demonstrated to be ~ 5 times larger than those previously reported in hBN.³⁴ Due to the difference in the permittivity tensor, HPPs in the lower and upper Restrahlen band exhibit positive ($\Delta\lambda > 0$) and negative ($\Delta\lambda < 0$) wavelength alterations, whereas the dissipation is reduced ($\Delta\gamma < 0$) in both bands. Importantly, in the upper Restrahlen band, optical confinement (λ_0/λ) and propagation dissipation (γ) of HPPs were simultaneously optimized by the sample suspension. These enhanced figures of merits

may facilitate improved nanophotonic functionalities using HPPs, including biochemical sensing, emission engineering, nanoscale energy transfer, and nano-polaritonic circuits.

See the [supplementary material](#) for the modeling of dispersion and field distribution of HPPs.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

DATA AVAILABILITY

The data that support the findings of this study are available within the article and its [supplementary material](#).

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