Near-future optical quantum information processing relies on sources of pure and indistinguishable single-photons. Promising candidates include quantum dots, trapped ions, color centers in solids, and single-photon sources (SPSs) based on heralded spontaneous parametric down-conversion. The recent discovery of fluorescent defects in two-dimensional (2D) materials has added yet another class of quantum emitters to the solid-state color centers. Stable quantum emitters have been reported in the transition metal dichalcogenides WSe₂, MoSe₂, MoS₂, and WS₂. The optical transition energies for these emitters, however, are located in close vicinity to the electronic band gap. Thus, cryogenic cooling below 15 K is required to resolve the zero phonon lines (ZPLs). For room temperature quantum emission, defects hosted by large band gap materials are ideal, as has been demonstrated in 2D hexagonal boron nitride (hBN). In this case, the energy levels introduced by the defects into the band structure are well isolated. The large band gap of 6 eV also prevents nonradiative decay, which in turn allows for high quantum efficiencies. Unlike solid-state quantum emitters in 3D systems, the 2D crystal lattice of hBN allows for an intrinsically high extraction efficiency. More precisely, the single-photons emitters have an in-plane dipole resulting in out-of-plane emission, where the emitters are not surrounded by high refractive index materials. Hence, total internal or Fresnel reflection does not affect the collection of the single-photons. Furthermore, 2D crystals can be easily attached by van der Waals forces to components such as fibers or waveguides, making them suitable for integration with photonic networks. The exceptionally high thermal and chemical robustness of hBN benefits the durability of the quantum emitters, achieving long-term stable operation over a huge temperature range, as well as high resistance to ionizing radiation.

In spite of large experimental research efforts and theoretical calculations, the exact nature of the defects yet has to be determined. Furthermore, the identification is hampered by the large variations of the lifetime and ZPL wavelength from defect to defect. Lifetimes ranging from 0.3 up to 20 ns and ZPLs in the UV and full visible spectrum have been reported. In addition to naturally occurring defects, the emitters can also be created artificially using diverse methods, including chemical etching, plasma etching, irradiation with γ-rays, ions, and electrons as well as near-deterministic stress-induced activation. Although most researchers agree that quantum emitters in hBN provide a number of unique opportunities, the performance still lags behind that of 3D systems. This is mainly due to the complex stoichiometry of hBN and its sensitivity to reagent concentrations, which are challenging to control. Therefore, the interest is currently shifting to the design and fabrication of deterministic stress-induced activation.
Figure 1. Design and fabrication. (a) The microcavity consists of a hemispherical and flat mirror (only two stacks shown on either sides). The quantum emitter hosted by hBN emits confocally with the excitation laser. A PDMS spacer sets the cavity length. To prevent influence of the polymer on the emitter, the PDMS is etched in the middle. (b) Microscope image of the array of hemispheres (not all 64 shown). The surface profile of the hemisphere actually used for the cavity is shown in the right inset. The bottom inset shows the height profile through an arbitrarily chosen axis. The solid blue line shows an ideal cross section of a hemisphere with radius 2.7 μm. (c) Reflectivity of the coating measured by spectrophotometry, with R = 99.2% at the target wavelength λ = 565 nm. The inset shows the calculated cavity reflectivity based on the coating. (d) SEM image (immersion mode) of the mirror stacks, coated with a layer of gold. The sample is tilted by 52°, so the image is skewed in the vertical direction. The lighter areas in the cross section are regions which have been imaged with a magnification of 125 000× (see inset). The intense electron beam makes the surface reactive, and carbon-contaminations by residual organic materials in the SEM chamber are bonded at these areas. (e) Thickness change of a PDMS film with driving voltage reveals linear tuning with 102 nm·V⁻¹. (f) Design of the optics platform (all components to scale). A polarization maintaining fiber (blue) guides the excitation laser from the diode below the platform. The laser is focused to the diffraction limit into the cavity onto the defect. The single-photons transmit through the dichroic mirror (DM) and are additionally band-pass filtered (BPF). Next, they are split by a 50:50 beam splitter (BS) and fiber-coupled into multimode fibers.

The confocal microcavity consists of a hemispherical and a flat mirror, with the hBN flake hosting the quantum emitter transferred to the focal point of the cavity (see Figure 1(a)). The hemisphere spatially confines the cavity mode to the location of the emitter and is fabricated using I₂-enhanced focused ion beam milling. We fabricated arrays of 64 hemispheres per substrate with varying geometrical parameters. The surface roughness could be minimized by adding I₂-gas during the milling process. With the FIB we can achieve radii of curvature down to <3 μm (see Figure 1(b)). We initially characterized the hemispheres using an atomic force microscope (AFM) and phase-shift interferometry (PSI). The characteristic parameters extracted with both methods agree well, which allows us to use the much faster PSI for the characterizations. The hemisphere profile shown in Figure 1(b) has a radius of 2.7 μm and root-mean-square deviations <1 nm from an ideal hemisphere (see Supporting Information S1). Note that we did not fabricate full hemispheres and the shapes deviate at the edges (which is due to a conductive coating to prevent charging effects during the milling). Both the flat and concave substrate are coated with 9 pairs of alternating dielectric quarter wave stacks (SiO₂/TiO₂), deposited using plasma sputtering. We measured a reflectivity behind state-of-the-art SPs. Moreover, the reported quality of single-photons from hBN is not sufficient for practical quantum information processing like quantum key distribution (QKD) or photonic quantum computing.

A straightforward path for improving the performance of a spontaneous emission process is to use the Purcell effect by coupling the emitter to an optical resonator. The optical resonator reduces the number of modes the emitter can couple to, thereby enhancing emission into the resonant modes. This even works in the “bad-emitter” regime, when the emitter line width is larger than the cavity line width. Work on cavity-integration of emitters in 2D materials has been reported, with quantum emitters hosted by WSe₂ coupled to plasmonic nanocavities and microcavities. Quantum emitters hosted by hBN have been coupled to plasmonic nanocavities.

Hexagonal boron nitride can also be used to fabricate photonic crystal cavities, however, this makes the required spectral matching between optical cavity mode and emitter difficult. Yet, the performance is still not sufficient for use in quantum information experiments.

In this article, we report room temperature single-photon emission from multilayer hBN flakes coupled with a microcavity. The plano-concave cavity fully suppresses the phonon sideband (PSB) and other off-resonant noise, while at the same time greatly enhances directionality and the spontaneous emission rate. The hemisphere is fabricated using focused ion beam (FIB) milling, allowing for a small radius of the accurate and precise curvature. This leads to an ultrasmall mode volume on the order of λ³. We fully characterize the SPS and assess its feasibility for quantum key distribution and quantum computing. Moreover, the single-photon source in its current configuration is compact and fully self-contained within 10 × 10 × 10 cm³, including all optics, driving electronics and control units.

### DESIGN AND FABRICATION

The confocal microcavity consists of a hemispherical and a flat mirror, with the hBN flake hosting the quantum emitter transferred to the focal point of the cavity (see Figure 1(a)). The hemisphere spatially confines the cavity mode to the location of the emitter and is fabricated using I₂-enhanced focused ion beam milling. We fabricated arrays of 64 hemispheres per substrate with varying geometrical parameters. The surface roughness could be minimized by adding I₂-gas during the milling process. With the FIB we can achieve radii of curvature down to <3 μm (see Figure 1(b)). We initially characterized the hemispheres using an atomic force microscope (AFM) and phase-shift interferometry (PSI). The characteristic parameters extracted with both methods agree well, which allows us to use the much faster PSI for the characterizations. The hemisphere profile shown in Figure 1(b) has a radius of 2.7 μm and root-mean-square deviations <1 nm from an ideal hemisphere (see Supporting Information S1). Note that we did not fabricate full hemispheres and the shapes deviate at the edges (which is due to a conductive coating to prevent charging effects during the milling). Both the flat and concave substrate are coated with 9 pairs of alternating dielectric quarter wave stacks (SiO₂/TiO₂), deposited using plasma sputtering. We measured a reflectivity...
Photoluminescence reveals an excited state lifetime of 897(8) ps. The exponential fit function is convoluted with the system response (SR). (c) The cavity narrows the spectrum down to 0.224 nm (fwhm). The spectrum has been recorded using a high-resolution Fourier-transform spectrometer. From a Lorentzian fit we extract the ZPL at 565.85(5) nm and a line width (fwhm) of 5.76(34) nm. (b) Time-resolved photoluminescence reveals an excited state lifetime of 897(8) ps. The exponential fit function is convoluted with the system response (SR). (c) The cavity narrows the spectrum down to 0.224 nm (fwhm). The spectrum has been recorded using a high-resolution Fourier-transform spectrometer. From a Lorentzian fit we extract the ZPL at 565.85(5) nm and a line width (fwhm) of 5.76(34) nm. (b) Time-resolved photoluminescence reveals an excited state lifetime of 897(8) ps. The exponential fit function is convoluted with the system response (SR). (c) The cavity narrows the spectrum down to 0.224 nm (fwhm). The spectrum has been recorded using a high-resolution Fourier-transform spectrometer. From a Lorentzian fit we extract the ZPL at 565.85(5) nm and a line width (fwhm) of 5.76(34) nm.
defects were located using confocal PL mapping under off-resonant excitation at 522 nm. As hBN itself is optically inactive in the visible spectrum, all emission originates from the defects or surface contaminants. Each crystal is scanned with a resolution of 0.5 μm. For the cavity, we selected a suitable defect with a ZPL at 565.85(5) nm and a Lorentzian line width (fwhm) of 5.76(34) nm (see Figure 2(a)). The PL spectrum shows the typical asymmetric line shape. Note that this is not a result of partial suppression of the long pass filter used to block the pump laser (see Methods), but rather the PSB being adjacent to the ZPL. The defect emits 63.2% into its ZPL. We note that the emission >580 nm originates from surface contaminants activated during the annealing and is usually filtered out (see Methods). Alternatively, annealing in a reactive environment can burn off these contaminants. Time-resolved PL (TRPL) reveals a single-exponential decay with a lifetime of 366(19) ps (see Methods). For a fair comparison of the free-space and cavity-enhanced lifetimes we compare the lifetimes of 366(19) ps and 897(8) ps from the TRPL measurement is likely more accurate. While g_2(0)(r = 0) = 0.018(36) (see Figure 2(d)) and from the fit we extract a lifetime of 366(19) ps (see Methods). For a fair comparison of the cavity-enhanced lifetimes we compare the correlation function measurements in free-space and with the cavity. The g_2(r) for the uncoupled emitter dips only to 0.051(23) and has a lifetime of 837(30) ps. The lifetimes measured with time-resolved PL and extracted from the g_2(r) measurements agree reasonably well, even though we note that the 897(8) ps from the TRPL measurement is likely more accurate. While g_2 = 0 is within the error margin for the cavity-enhanced emitter, more accurate measurements are required to reduce the error margin to extract the true value of g_2. A small error margin on correlation function measurements can typically be achieved with ultrashort pulsed excitation. We also calculated the background correction term \( f \) and found that it is smaller than the significant digits of our measurement result (<5 × 10^{-5}), so we conclude that any deviation from 0 is not due to detector dark counts, but rather other noise sources excited through the laser. If we directly compare g_2 of the uncoupled and cavity-enhanced emitter, however, we see a reduction of a factor of 2.83. Such reduction can typically be achieved in the “bad-emitter” regime and means that off-resonant noise sources are successfully suppressed. A narrower cavity line width could thus further reduce g_2. It is worth noting that many applications require the generation of single-photons on-demand, which can be achieved by pulsed excitation of the emitter. As already mentioned, our pulsed laser is within the stopband of the cavity, making it impossible to match excitation and emission wavelength for this laser due to the large free spectral range. The values for the second-order correlation function, however, can be equal for both continuous and pulsed excitation schemes, allowing for simply replacing the continuous excitation laser in our experiment with a suitable pulsed laser for potential quantum information experiments (see below).

The ratio of free-space (or rather half-sided cavity) to cavity-coupled lifetime is \( f = 2.29 \). The effective Purcell enhancement is given by

\[
F_p^{\text{eff}} = \frac{3}{4\pi^2} \lambda^3 \frac{Q^{\text{eff}}}{V}
\]

with \( Q^{\text{eff}} \) being the effective quality factor and \( V \) being the cavity mode volume. This can be calculated by integrating over the Gaussian fundamental mode in a resonator:

\[
V = \pi \int_0^{2\pi} \int_0^\infty L' \, d\phi \, dz \, \sin^2 \left( \frac{2\pi n}{\lambda} \right) \exp \left( -2r^2 / w_0^2 \right)
\]

\[
= \frac{\pi}{4} L' w_0^2
\]

where \( L' \) is the effective cavity length (see below for the calculation) and \( w_0 = \lambda / (2\pi L' r - r^2) \) is the cavity waist, determined by the effective cavity length and \( r \), the radius of the hemisphere. We calculate the mode volume to be \( 1.76 \lambda^3 \). In the “bad-emitter” regime the effective quality factor \( Q^{\text{eff}} = \delta / (\delta_{\text{res}} + \delta_{\text{in}}) \) has to be used, which is dominated by the emitter dynamics. It should be mentioned that this is only an approximation and it is more accurate to calculate the overlap integral of the photonic density of states of the cavity and electronic density of states of the emitter. In addition, this effective Purcell factor is different from the ratio \( f \), because the dielectric environment of the mirror is modifying the available density of states, whereas the Purcell factor is the ratio of vacuum (or true free-space) to cavity lifetime. We calculate the effective Purcell factor to be 4.07. This also allows for the direct calculation of the quantum efficiency, given by

\[
\eta = \frac{f - 1}{f + F_p^{\text{eff}} - \epsilon f}
\]

where \( \epsilon \) is the Purcell factor caused by the mirror and is determined by finite-difference time-domain (FDTD) simulations. For our mirror we find \( \epsilon = 1.68 \) (see Supporting Information S5) and thus the quantum efficiency is 51.3%.

The cavity also modifies the power saturation behavior (see Figure 2(e)), with an increased single-photon count rate even at lower excitation power. This is a result of the Purcell enhancement, which makes the emitter brighter, but also from the increased collection efficiency of the cavity, as the emitter predominantly emits into the cavity mode. The low excitation power also assists the single-photon count rate stability, because at low excitation power the emitters show no blinking or photobleaching. This is particularly important as the
photobleaching increases with decreasing wavelength and due to the stopband of the cavity our excitation laser is at 450 nm. Note that the count rates at the single-photon avalanche diodes (SPADs) shown in Figure 2(e) are the raw count rates, not corrected for transmission loss or detector efficiency. The quantum emitter also emits linearly polarized light (see Figure 2(f)) with a degree of polarization (DOP) of 90.4%. The fit is obtained using a \( \cos^2(\theta) \) function. A high polarization contrast is crucial for QKD applications which use polarization encoding. Increasing the DOP of not fully polarized light is always accompanied by loss, and so it sets an upper bound on the efficiency of the SPS.

Since the cavity length is tunable, the single-photon wavelength can also be tuned. Effectively, the tuning range is the line width of the free-space emission. The cavity is only sampling the free-space emission spectrum, however, so the actual single-photon count rate is the spectral overlap integral of optical cavity mode and emitter. This results in the emission rate decreasing with increasing cavity detuning.

### Simulations

**Numerical Modeling.** We can use FDTD simulations to calculate the electric field distribution of the dipole emitter in the cavity. The electric field intensity \( |E|^2 \) is shown in Figure 3(a). The simulations also show that resonance does not occur at a physical mirror separation \( L' \) which is a multiple of \( \lambda/2 \). This is due to the finite penetration depth of the electric field into the dielectric mirror stacks, leading to an effective cavity length. The penetration depth \( \xi \) thereby is given by

\[
\xi = \frac{q\lambda/2 - L'}{2}
\]

The physical mirror separation \( L' \) is determined by maximizing the intracavity electric field (see Supporting Information S5). Our simulations yield \( \xi = 122 \text{ nm} \). When designing the thickness of the PDMS spacer, this has to be taken into account. To reduce the computational time we simulated the longitudinal mode \( q = 5 \) instead of the experimentally realized \( q = 8 \). Nevertheless, the parameter \( \xi \) is not affected by this beyond the numerical precision of the simulation.

**Applications in Quantum Technologies.** We now turn to an evaluation of the SPS for two of the most common quantum information applications: quantum key distribution and quantum computing. In principle, the photon quality is sufficient for quantum key distribution, outperforming state-of-the-art protocols based on weak coherent pulses with decay states on short and medium distances (see Supporting Information S6). On long distances decay state protocols become more efficient. However, when evaluating the performance, it is important to keep in mind that QKD requires single-photon generation on-demand (e.g., through pulsed excitation), which has not been demonstrated yet with the current device. Moreover, since the photophysics of different emitters varies, it is not clear how well quantum emitters in hBN can be scaled. More statistics of cavity-coupled emitters are required to explore the potential of employment in quantum networks.

Notably, QKD only requires minimal entropy on all degrees of freedom which are not used for qubit encoding. Other quantum information protocols, however, do require truly indistinguishable single-photons. An example are entangling gates for single-photons for use in one-way quantum computing.51 A measure of how indistinguishable consecutively emitted single-photons are is the interference contrast \( I \) in a Hong-Ou-Mandel experiment.52 Unfortunately, as our cavity is pumped continuously, we cannot directly measure \( I \). Nevertheless, we can at least theoretically calculate the expected indistinguishability. The indistinguishability of a quantum emitter with pure dephasing is given by

\[
I = \frac{\gamma}{\gamma + \gamma^*}
\]

where \( \gamma \) is the emission rate and \( \gamma^* \) is the pure dephasing rate. At room temperature we find \( I = 2 \times 10^{-3} \), meaning only 1 in 5000 photons would interfere in a Hong-Ou-Mandel experiment. Even such a strongly dephasing emitter, however, can reach a regime of high indistinguishability, when coupled with a high-Q cavity. In the limit of weak coupling \( I \) modifies to

\[
I = \frac{\gamma + \kappa R/(\kappa + R)}{\gamma + \kappa + 2R}
\]

where the parameter \( R = \frac{4g^2}{\kappa + \gamma + \gamma^*} \) is the effective transfer rate between the emitter and the cavity, \( \kappa \) is the cavity line width and \( g \) is the cavity coupling strength.53 For our cavity parameters we find \( I = 5.3 \times 10^{-3} \). While this is an improvement by a factor of 26, it is still far beyond being useful for fault-tolerant quantum computing. The indistinguishability for generalized cavity line width and coupling strength is shown in Figure 3(b). Note that in the limit of
strong coupling it is also possible to achieve a high indistinguishability. With the coupling strength typically <1 GHz, a narrow cavity line width is required to maximize $I$. Figure 3(c) shows that $I > 90\%$ requires a cavity line width less than 124 MHz. At a reflectivity of 99.95%, this line width limits the free spectral range (FSR) to 779 GHz. Compared with the free-space emission line width (5.41 THz) this means that the spectral profile of the cavity would be comb-shaped, with the cavity sampling the emitter spectrum at multiples of the FSR. Single-photons originating from different comb peaks are of course distinguishable, so a high indistinguishability requires filtering out only one peak (for example with another cavity). This, however, is balanced by a loss in efficiency. To overcome this, the natural line width of the emitter into free-space must be narrowed. Cryogenic cooling is one option to narrow the line width sufficiently.

## CONCLUSION

We have demonstrated coupling of a quantum emitter hosted by multilayer hBN to a confocal microcavity. The hemispherical geometries have been fabricated using FIB milling with sub-nm precision. The cavity mode volume is of the order of $\lambda^3$. The cavity improves the spectral purity of the emitter substantially, with the fwhm decreasing from 5.76 to 0.224 nm. Moreover, the cavity suppresses off-resonant noise, which allows us to improve its single-photon purity. The excited state lifetime of the emitter is also shortened by the Purcell effect by a factor of 2.3. The emission of the cavity is linearly polarized and stable over long timeframes, with no signs of photobleaching or blinking. The cavity also features a linearly tunable PDMS spacer between both mirrors, which allows in situ tuning of the single-photon line over the full free-space ZPL of the quantum emitter. This would allow us to fabricate multiple identical single-photon sources, by locking all to the same emission wavelength, making this approach fully scalable. Furthermore, the complete SPS is portable and fully self-contained within $10 \times 10 \times 10$ cm$^3$. The microcavity platform can also be easily adapted to other quantum emitters in 2D materials and offers a promising path toward scalable quantum information processing.

## METHODS

**FIB Milling.** Borosilicate glass substrates with a size of $18 \times 18$ mm$^2 \times 160$ $\mu$m have been coated with 100 nm gold using electron-beam thermal evaporation to prevent substrate charging effects. The ion accelerating voltage in the FIB (FEI Helios 600 NanoLab) is 30 kV with currents $\leq 0.28$ nA. The dose rate is encoded in the RGB color of a hemispherical field at a gas flow of 300 ccm$^3$/min) removes the PMMA around the hBN flakes. The PMMA below the flake is decomposed during the annealing, which also stabilizes the optical emission properties (more details have been published previously).

**Quantum Emitter Fabrication.** The flat mirrors have been coated with 300 nm 950 PMMA A4. Multilayer hBN flakes have been exfoliated from bulk (HQGraphene) and transferred onto the PMMA layer by dry contact. Oxygen plasma etching (500 W for 2 min generated from a microwave field at a gas flow of 300 ccm$^3$/min) removes the PMMA around the flake as well as creates the quantum emitters. The PMMA below the flake is decomposed during the annealing, which also stabilizes the optical emission properties (more details have been published previously).

**Optical Characterization.** Each flake has been scanned using a custom-built confocal micro-photoluminescence setup with a resolution of 0.5 $\mu$m and a spectrum has been recorded at each scanning position. The excitation laser with a wavelength of $\lambda = 222$ nm is nonresonant with the optical transition energy of the defect. The laser light is blocked with a Semrock RazorEdge ultrasteep long-pass edge filter. With a laser pulse length of 300 fs at a repetition rate of 20.8 MHz, the setup also allows us to measure the excited state lifetime. The pulses are split into trigger and excitation pulses, and the photoluminescence is detected by a SPAD (Micro Photon Devices). The time correlation between trigger pulse and arrival time of the photoluminescence is given by a time-to-digital converter (PicoQuant PicoHarp 300). The photoluminescence is coupled via a grating to the SPAD, which makes the TRPL wavelength-sensitive. This allows us to measure the lifetime of the ZPL only. The second-order correlation function measurements have been performed using two SPADs in the exit ports of a beam splitter and under continuous excitation.

**Spectral Ellipsometry.** The emitter is centered onto the mirror, making it easy to locate on the mirror. Nearby crystal flakes serve as markers for localizing the flake with the precharacterized defect. For aligning the crystal long working distance objectives in a custom-built microscope have been used, illuminated with a near-infrared LED where all

\[ g^{(2)}(t) = 1 - A e^{-\tau_1/\rho} + B e^{-\tau_2/\rho} \]  

with the anti- and bunching amplitudes $A$ and $B$, and the decay times $\tau_1$ and $\tau_2$. The experimental data is normalized such that $g^{(2)}(\tau \to \infty) = 1$. The background corrected $g^{(2)}$ is given by

\[ g^{(2)}_b = g^{(2)} - \frac{(1 - \rho^2)}{\rho^2} \]  

with $\rho = \text{SNR}/(\text{SNR} + 1)$, where SNR is the signal-to-noise ratio. In addition to the long-pass filter, the photoluminescence for correlation function measurements is band-pass filtered around the ZPL. We utilize linear variable filters (Delta Optical Thin Film 3G LVLPW and 3G LVSWP) to tune center and bandwidth of the band-pass filtering system.

**Cavity Alignment.** The emitter on the mirror has been located and characterized in free-space. The hBN flake capable of hosting a defect (determined by flake thickness) was centered onto the mirror, making it easy to locate on the mirror. Nearby crystal flakes serve as markers for localizing the flake with the precharacterized defect. For aligning the crystal long working distance objectives in a custom-built microscope have been used, illuminated with a near-infrared LED where all

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coatings are transparent. Each component, held and aligned with vacuum tweezers on a motorized 6-axis nanopositioning stage, is glued one after another. The adhesive used was UHU Plus Endfest 300, a two component epoxy glue, and was cured for 24 h at room temperature. Where required a second layer of glue was added and cured for another 24 h. Diagnostics were used to provide feedback for a good alignment, e.g., single-photon detection rate when aligning the cavity, combined with the spectrometer to check the cavity mode profile.

**ASSOCIATED CONTENT**

### Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsphoto-nics.9b00314.

(1) Hemisphere fabrication; (2) spin speed curves PDMS solution; (3) photographs of the device; (4) transverse mode spacing; (5) FDTD simulations; (6) QKD simulations (PDF)

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**Notes**

The authors declare no competing financial interest.

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