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ABSTRACT

Room temperature quantum emitters have been reported in aluminum nitride grown on sapphire, but until now they have not been observed in epilayers grown on silicon. We report that epitaxial aluminum nitride grown on silicon by either plasma vapor deposition or metal-organic vapor phase epitaxy contains point-like emitters in the red to near-infrared part of the spectrum. We study the photon statistics and polarization of emission at a wavelength of 700–750 nm, showing signatures of quantized electronic states under pulsed and CW optical excitation. The discovery of quantum emitters in a material deposited directly on silicon can drive integration using industry standard 300 mm wafers, established complementary metal-oxide-semiconductor control electronics, and low marginal-cost mass-manufacturing.

Semiconductors host point-like defects which confine charges in three dimensions, some of which emit quantum light at room temperature. Quantum emitters (QEs) resulting from combinations of impurity atoms, vacancies, and structural defects have been discovered in wide-bandgap materials, such as diamond,[1] silicon carbide (SiC),[2] and hexagonal boron nitride (hBN).[3,4] QEs have recently been observed in silicon[5,6] at cryogenic temperatures, but even when a resonator enhances the photon extraction, they are currently limited to a few thousand detection events per second.[7] In cubic and hexagonal polymorphs of SiC, QEs have been identified in the near-infrared[8] and visible[9] regions with the former displaying an optically addressable spin.[2] Quantum dot based heteroepitaxial quantum light sources on silicon have been reported based on InGaAs quantum dots,[10] which only emit at cryogenic temperatures, and in GaN[11] emitting in the ultraviolet spectral range.

In contrast, QEs in the III-nitrides have been shown to emit from the visible out to the infrared at room temperature[12–14] with GaN demonstrating room temperature optically addressable spin states.[15] Some theoretical studies even suggest QEs in aluminum nitride have optically addressable spin states,[16–18] opening up new possibilities for spin–photon quantum interfaces. Given the apparent variety of QEs in these nitrides on sapphire, covering the range from the visible to the near-infrared, it is not agreed which QE arises from which complex. Theoretical studies have made several suggestions for complexes involving vacancies, oxygen and nitrogen contamination,[19,20] stacking faults,[21] and cubic inclusions.[14]

Nevertheless, AlN is a promising material for integrated photonics which could incorporate these QEs. Refined processing techniques have led to low loss optical devices in epitaxial AlN[22] in the infrared. Additionally, AlN's transparency in the visible spectrum can be used for heterogeneous integration of diamond color centers in nanophotonic chiplets.[23] Already, AlN QEs have been directly integrated into waveguide structures while retaining their single-photon emission properties,[24] which is a promising route to integration and multiplexing.

To date, most reports of AlN and GaN QEs are from metal-organic vapor phase epitaxy (MOVPE) of wurtzite crystal on sapphire substrates[12–14] aside from one report of QEs in GaN on SiC substrates.[25] However, MOVPE of III-nitrides on silicon (111) is also an established technology,[26,27] although no QEs have been reported.

From a practical point of view, epitaxy on silicon offers several advantages. Importantly, the ubiquitous adoption of silicon wafer processing presents many avenues for device fabrication and scalability as well as improved integration into preexisting silicon-based technologies. AlN grown on cheap, 300 mm wafers would be compatible with industry standard complementary metal-oxide-semiconductor (CMOS)[28] technology, enabling rapid adoption. Additionally, silicon has a higher thermal conductivity than sapphire, 145 W/mK (Ref. 29) compared to 36 W/mK (Ref. 30) at 300 K, aiding in device heat extraction.

We report the presence of QEs in epilayers of AlN grown on silicon that emit in the visible to near-infrared at room temperature. Studying two samples prepared by different epitaxial techniques, with differing crystalline structure, we find both samples contain similar point-like emitters. They display signatures of quantized states including saturating intensity at high power and autocorrelation functions with $g^{(2)}(0) < 0.5$, the accepted limit for an emission dominated by a single QE. We present data on their spectra, optical polarization, and photon statistics under pulsed and CW excitation, and compare them to QEs observed in AlN layers grown on sapphire.

Two samples of AlN on silicon are examined in this work. The first is a planar crystalline AlN epilayer 150 nm thick grown by MOVPE. The epilayer was grown in a vertical-flow high-speed (1000 rpm.) rotating disk reactor (Veeco Propel^{TM}) with trimethylaluminum (TMAl) and ammonia (NH_3) as the precursors and hydrogen (H2) as the carrier gas. For this growth, emissivity corrected pyrometry was used to control the substrate temperature and reflectometry was used to continuously monitor the surface morphology in real-time. Before growth, the 6-in. p-type Si (111) substrate was heated inside the reactor at 1100 °C to remove the native surface oxide *in situ* without any chemical treatment. This was followed with a change of temperature to grow a thin (10 nm) nucleation layer at a low temperature of 750 °C with a high V/III molar ratio (750). Subsequently, the rest of the epilayer (140 nm) was grown at 1070 °C with a low V/III molar flow of 75 at a growth rate of 370 nm/h. The XRD linewidth is 0.35° for the (002) reflection. The second sample is a commercially available 200 nm epilayer of AlN on a p-doped silicon (111) substrate from Kyma Technologies. It is grown via plasma vapor deposition of nanocolumns (PVDNCs), which results in many close-packed pillar crystals extending vertically to the sample surface with an aspect ratio of 5:1. The room temperature confocal scan maps in Figs. 1(a) and 1(d) show point-like emitters. Each map is composed of three scans at different excitation laser polarization angles to reveal the true density of QEs[31] with minimal selection bias. Figure 1(b) shows an atomic force microscope (AFM) image of the MOVPE epilayer surface, revealing it to have a low surface roughness of 3.2 nm RMS over the 2×2 μm area. In contrast, the AFM image of the PVDNC sample in Fig. 1(e) shows the presence of many close-packed nanocolumns with a density of ~600 μm^{-2}. The surface roughness is 9.1 nm RMS over the same area. Each individual nanocolumn has a width of approximately 40 nm extending from the substrate to the sample surface. The PVDNC AlN surface is c-plane (0001) with an XRD linewidth of 1° for the (002) reflection.

It is possible to isolate single QEs as highlighted in Figs. 1(a) and 1(d). Figures 1(c) and 1(f) show room temperature photoluminescence (PL) spectra excited from the circled QEs in Figs. 1(a) and 1(d). Each spectrum shows a broad emission peak with full width at half

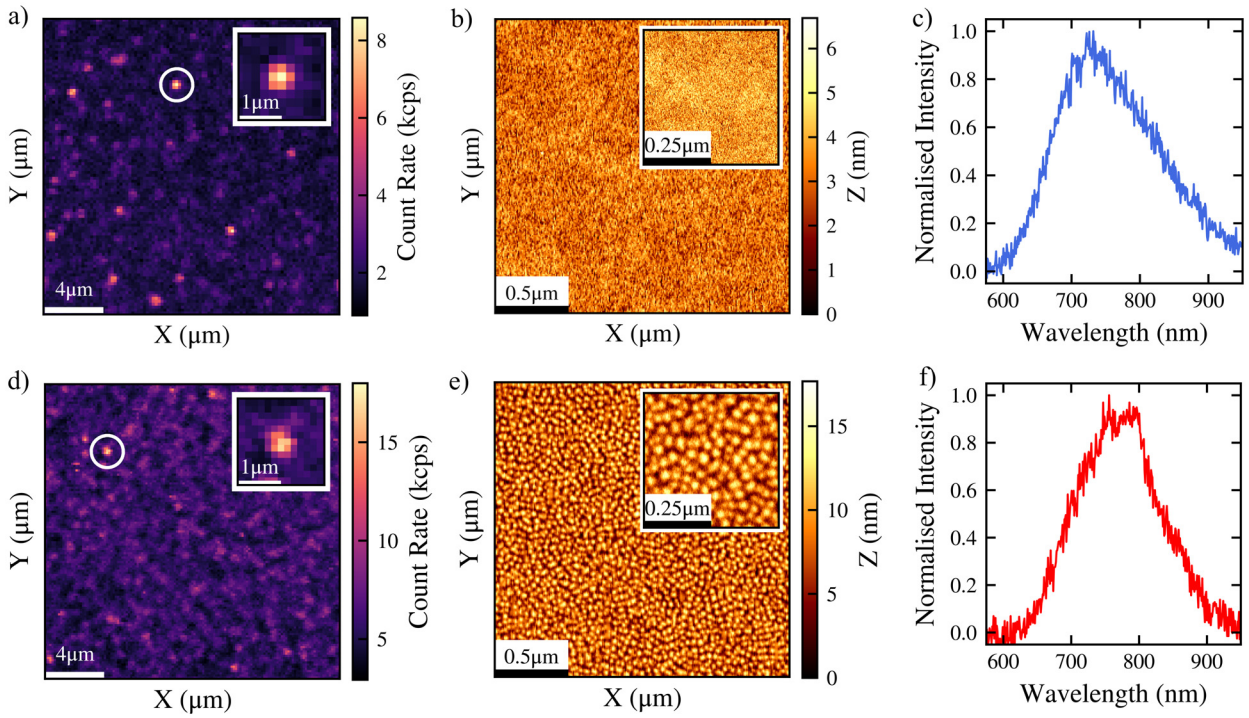

FIG. 1. Confocal scan map, atomic force microscopy map, and room temperature photoluminescence spectrum for the MOVPE sample, (a)–(c), and the PVDNC sample, (d)–(f), respectively.

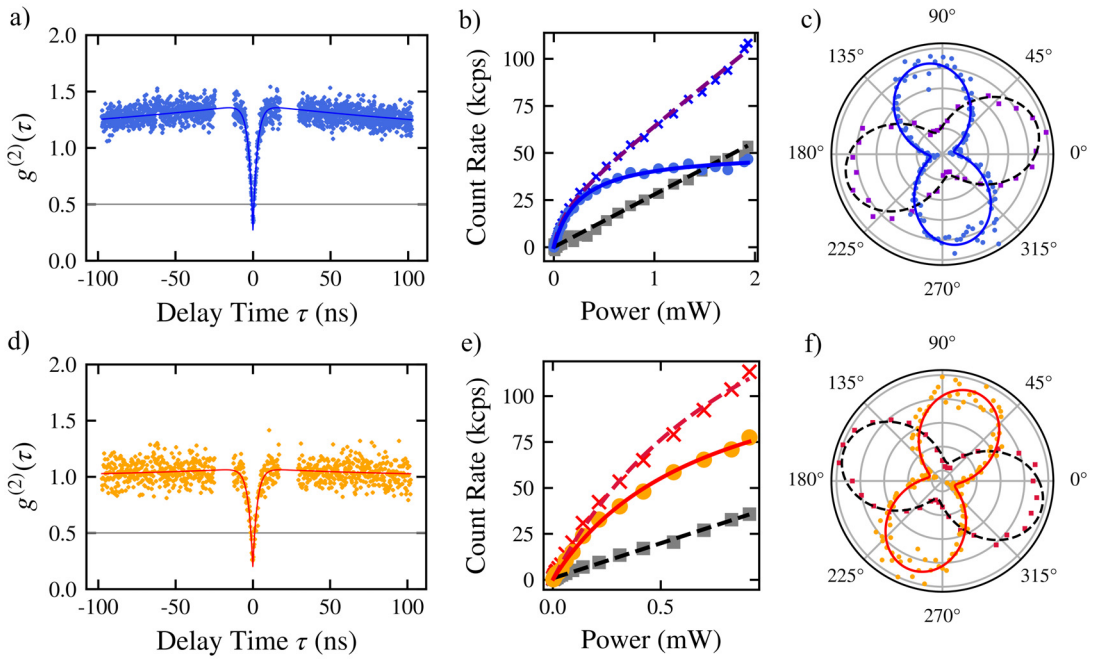

FIG. 2. Room temperature physical properties of MOVPE (a)–(c) and PVDNC (d)–(f) samples. (a) and (d) Room temperature autocorrelation measurements. (b) and (e) Power-dependent photon counting measurement. The signal, background, and corrected data are marked by crosses, squares, and circles, respectively. (c) and (f) Background-corrected polarization-resolved measurement. Excitation marked by square markers and dashed lines. Collection marked by circles and solid lines.

maximum ∼200 nm, which does not change with laser power. No zero phonon line is resolved, which suggests a strong coupling to phonons in the crystal. An absence of an obvious zero phonon line is also common in many QEs in AlN on sapphire samples[12] which tend to emit at ∼600 nm. In comparison to AlN QEs fabricated by aluminum ion implantation,[32] the QEs presented here occur at wavelengths approximately 50 nm longer but have comparable radiative lifetimes.

Figures 2(a) and 2(d) shows autocorrelation measurements performed with a Hanbury-Brown and Twiss (HBT) interferometer on exemplar QEs in each sample. Both autocorrelations dip below the conventional single-emitter limit of $g^{(2)}(0) = 0.5$ at time zero. The curves are fit with the empirical function given in the following equation:[1]

$$g^{(2)}(\tau) = 1 + \alpha_1 e^{-|\tau|/t_1} + \alpha_2 e^{-|\tau|/t_2}, \quad (1)$$

where $t_{1,2}$ are the antibunching and bunching timescales, respectively, and $\alpha_{1,2}$ are the antibunching and bunching amplitudes. A reflection of the avalanche photodiode back-flash (photoemission during photodetection)[33] from within our fiber HBT interferometer is masked between delay times of $\pm(15 \rightarrow 27)$ ns. The autocorrelation parameters for these data, recorded at excitation powers 100 and 343 μW, respectively (250 and 860 W/mm^2), are shown in Table I:

Figures 2(b) and 2(e) show power-dependent photon counting measurements, demonstrating saturation of the QEs intensity. The data are fit with the following equation:

$$I(P) = \frac{I_\infty P}{P + P_{sat}}, \quad (2)$$

where I_∞ is the intensity as the pump power tends toward infinity and P_{sat} parameterizes the power when the intensity reaches half its saturated value. The intensity dependent measurement was performed with the excitation laser aligned to the absorption dipole of the emitter. I_∞ is found to be 50 ± 1 kcps with $P_{sat} = 0.30 \pm 0.01$ mW for the MOVPE QE. For the PVDNC sample, I_∞ is found to be 134 ± 5 kcps with $P_{sat} = 0.71 \pm 0.04$ mW. These intensities are comparable to what has been previously reported in other GaN[14,15,34] and AlN samples,[12,32,35] which is not surprising given the similar radiative lifetimes, sample design, and experimental configuration.

Polarization-resolved absorption and emission measurements in Figs. 2(c) and 2(f) are fit according to the following equation:

$$I(\theta) = a + b\cos^2(\theta - \theta_D), \quad (3)$$

where a and b are offset and amplitude values, respectively. The visibility of the dipole, η, is defined as $(I_{max} - I_{min})/(I_{max} + I_{min})$ and θ_D is the dipole orientation. Visibilities exceeding 0.90 are achieved for the absorption and emission dipole in both examples. Non-parallel linear absorption and emission dipoles are also reported in some AlN on

TABLE I. Autocorrelation parameters for both samples. $g^{(2)}(0)$ and its error are estimated from the raw data, and the other parameters are from a least squares fit.

	MOVPE	PVDNC
$g^{(2)}(0)$	0.36 ± 0.03	0.24 ± 0.04
τ_1 (ns)	2.8 ± 0.1	2.9 ± 0.2
τ_2 (ns)	237.4 ± 14.1	97.8 ± 31.2
α_1	-1.12 ± 0.02	-0.88 ± 0.04
α_2	0.39 ± 0.01	0.08 ± 0.01

sapphire QEs,[12,35] which has been attributed to indirect excitation of the transition via and excited state.[35] This feature could be beneficial for crossed-polarization filtering of the excitation. We have not found any preferred orientation of the emitters along any crystal axes, as is also the case for QEs in AlN on sapphire samples.[31]

The power-dependent autocorrelation properties are presented in Fig. 3 for the PVDNC QE. At the highest excitation power of 1.10 mW, $g^{(2)}(0) = 0.46 \pm 0.03$ is observed. In the limit of vanishing excitation power, the antibunching timescale is extrapolated to (3.0 ± 0.3) ns.

Deviation from perfect antibunching can be attributed to a weak broadband background fluorescence from the sample. The origin of this background emission so far from the AlN band edge is unknown, but we hypothesize that it arises from scattered emission of multiple nearby QEs. The signal-to-background ratio (SBR) can be defined as the ratio of fluorescence from the QE to the background fluorescence, which is assumed to have Poissonian statistics. The background-limited value of $g^{(2)}(0)$ is calculated using the following equation:[36]

$$g^{(2)}(0) = \frac{C_N - (1-\rho^2)}{\rho^2}, \tag{4}$$

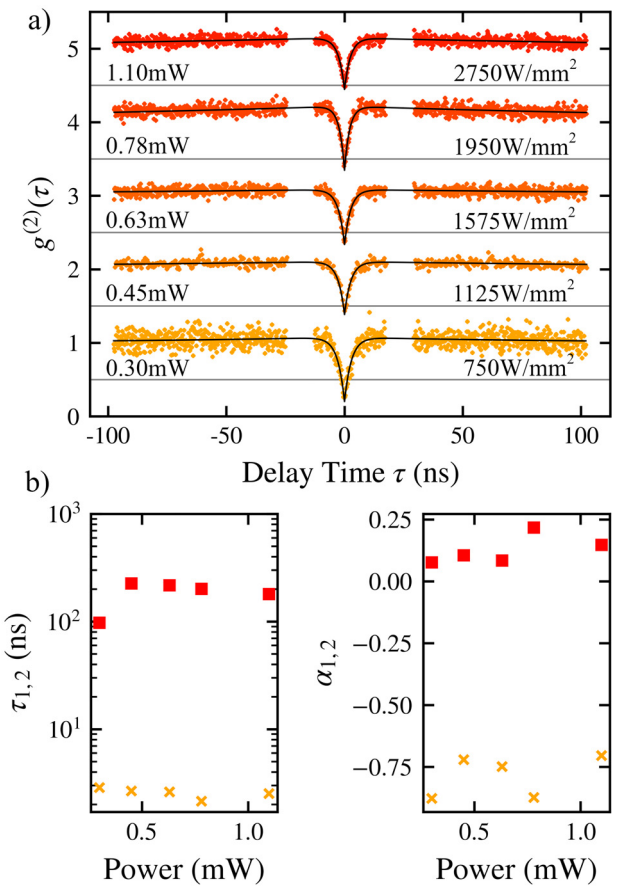

FIG. 3. (a) Power-dependent autocorrelation measurements from an emitter in the PVDNC sample under continuous wave optical excitation. (b) Fit values from Eq. (1). τ_1 and α_1 are marked by crosses, while τ_2 and α_2 are marked by squares.

where $\rho = \mathrm{SBR}/(\mathrm{SBR}+1)$. At low excitation power, SBR = 5.6 corresponding to a background-limited value of $g^{(2)}(0) = 0.27$, which is consistent with data in Fig. 3.

The lifetime of the PL emission was fitted with a bi-exponential decay with lifetimes of (2.2 ± 0.1) ns and (6.9 ± 0.9) ns, and amplitudes 0.88 and 0.11, respectively. This biexponential decay indicates the presence of two decay paths. Other III-nitride QEs report monoexponential lifetimes in the scale of few nanoseconds for GaN[13,14] and hBN.[37] It appears the proximity of the surfaces in the nanocolumnar sample has not substantially reduced the lifetime, probably because radiative recombination in the III-nitrides is known to be slow.[38] Evidence of single-photon emission is shown in Fig. 4(b) where $g^{(2)}(0) = 0.41 \pm 0.03$ under pulsed optical excitation. The data are overlaid with a series of biexponential peaks determined from a least squares fitting routine. $g^{(2)}(0)$ is calculated by the ratio of the central peak area, shaded red, to the average of many satellite peak areas, example shaded blue. We note the correlation does not go to zero between peaks due to the long decay time. Moreover, there is a finite peak at time zero we attribute to the finite proportion of Poissonian background emission, as also seen in CW correlations.

The unknown origin of these QEs means we are not able to say with certainty how they differ from those seen in AlN grown on sapphire, which tend to emit at ≈ 600 nm. The absence of a zero phonon line in the emitters seen in the AlN on Si samples is consistent with many QEs seen in AlN on sapphire samples.[12,39] The ability to create

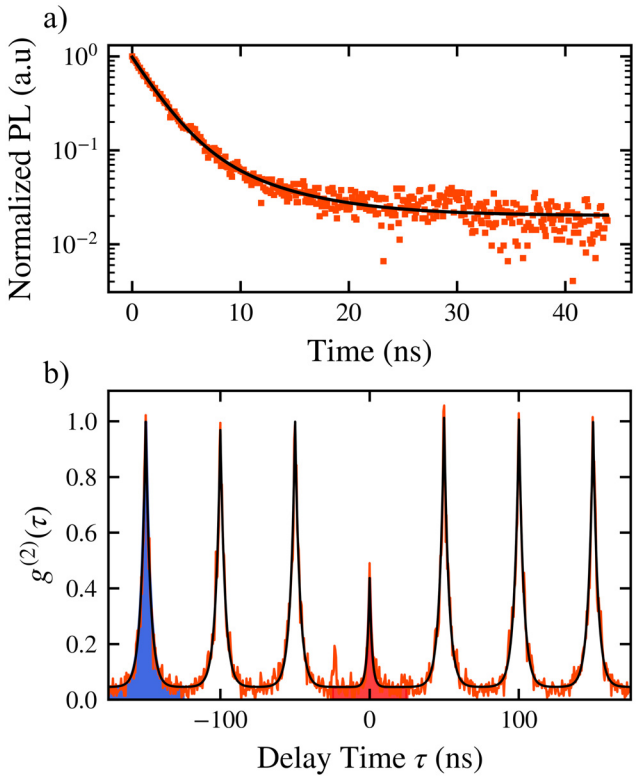

FIG. 4. (a) PL lifetime measurement of the same emitter in the PVDNC sample. (b) Pulsed autocorrelation measurement.

QEs in AlN on sapphire sample by Al-implantation[32] suggests the radiative center involves vacancies created by ion damage. In any case, spectral measurements are insufficient to uniquely determine the origin of each emitter. Future work could correlate spectroscopic maps with structural information on the sample, as has been reported in GaN samples,[40] where it was shown samples with higher densities of threading dislocations had higher densities of QEs, even though QEs were not located at the position of the threading dislocation. A determination of the origin of QEs in these III-nitrides would inform future work to engineer their position, energy, and photo-physical properties.

In this work, thin films of aluminum nitride grown on a silicon substrate were found to host QEs that emit quantum light at room temperature. The presence of quantized electronic states were verified through both continuous wave and pulsed autocorrelation measurements. Future work should focus on reducing the background emission, which pollutes the autocorrelations, enhancing the photon extraction efficiency and achieving deterministic control over the QE location. Thereafter, integration with silicon control microelectronics and silicon avalanche photodiodes for monolithic on-chip detection can be investigated, potentially leading to applications in quantum communications, imaging, and sensing.

A scanning confocal 4*f* optical microscope was used to map QEs in the AlN film, where they appeared as diffraction limited spots. A 0.9 numerical aperture refractive objective was used to focus the excitation laser onto the sample and collect fluorescence. Emission from a QE is isolated using a 532 nm dichroic mirror in the collection path, a 550 nm long pass filter, and a linear polarizer aligned to the emitter dipole suppressing the intensity of the unpolarized background. Spectral measurements were performed on a 328 mm spectrometer equipped with a 80 lines/mm grating and a silicon CCD chosen to span a range greater than the QE spectrum. For polarization-resolved measurements, a half-wave plate and a thin film linear polarizer were used in the excitation and collection paths, respectively. Photon counting measurements were performed by fiber coupled SPCM-AQRH silicon avalanche photodiodes (APDs) from Excelitas. All CW optical measurements are performed under 532 nm excitation and pulsed measurements with a Picoquant 520 nm laser. Fits to the CW autocorrelation data are convolved with the instrument response function of the APDs to account for detector jitter (~300 ps).

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Joseph Kevin Cannon: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Writing – original draft (equal); Writing – review & editing (equal). **Sam Bishop:** Methodology (equal); Software (equal); Writing – original draft (equal); Writing – review & editing (equal). **Katie Eggleton:** Investigation (equal); Writing – review & editing (equal). **Hüseyin Bilge Yağcı:** Writing – review & editing (equal). **Rachel N. Clark:** Software (equal); Writing – review & editing (equal). **Sherif Ibrahim:** Investigation (equal); Writing – review & editing (equal). **John Patrick Hadden:** Funding acquisition (equal); Software (equal); Writing – review & editing (equal). **Saptarsi Ghosh:** Resources (equal); Writing – review & editing (equal). **Menno J. Kappers:** Resources (equal); Writing – review & editing (equal). **Rachel A. Oliver:** Resources (equal); Writing – review & editing (equal). **Anthony J. Bennett:** Conceptualization (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (equal); Supervision (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

Data supporting the findings of this study are available in The Cardiff University Research Portal at http://doi.org/10.17035/d.2023.0248937510 (Ref. 41).

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