A large-scale quantum simulator on a diamond surface at room temperature

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Strongly correlated quantum many-body systems may exhibit exotic phases, such as spin liquids and supersolids. Although their numerical simulation becomes intractable for as few as 50 particles, quantum simulators offer a route to overcome this computational barrier. However, proposed realizations either require stringent conditions such as low temperature/ultra-high vacuum, or are extremely hard to scale. Here, we propose a new solid-state architecture for a scalable quantum simulator that consists of strongly interacting nuclear spins attached to the diamond surface. Initialization, control and read-out of this quantum simulator can be accomplished with nitrogen-vacancy centers implanted in diamond. The system can be engineered to simulate a wide variety of strongly correlated spin models. Owing to the superior coherence time of nuclear spins and nitrogen-vacancy centers in diamonds, our proposal offers new opportunities towards large-scale quantum simulation at ambient conditions of temperature and pressure.

Many intriguing phenomena in condensed-matter systems originate from the interplay of strong interactions and frustrations. A representative example is frustrated quantum magnetism, where the spins cannot order to minimize all local interactions, and the ground state is highly degenerate\textsuperscript{2,3}. Together with long-range interactions between non-nearest neighbours, the frustrated quantum models give rise to intriguing quantum phases, for example supersolids\textsuperscript{3,4}. Moreover, they can also stabilize the long-sought quantum spin liquid\textsuperscript{5,6}, which has connections with high-temperature superconductivity\textsuperscript{7}. However, the properties of these systems have proved to be very hard to understand from numerical calculations, partly owing to the combination of long-range quantum correlations and the superposition principle of quantum mechanics. This principle implies that the required computational resources grow exponentially with the number of particles, making numerical approaches inefficient. Richard Feynman’s idea\textsuperscript{8} of quantum simulation provides a powerful solution to this problem: one could gain a deep insight into complex states of matter by experimentally simulating them with another well-controlled quantum system\textsuperscript{9}. Large-scale quantum simulations are expected to be a powerful tool\textsuperscript{10} for the investigation of fundamental problems in condensed-matter physics.

Quantum simulation has attracted extensive research interest in the past decade. Various architectures for quantum simulation have been constructed based on different systems, ranging from ultracold neutral atoms\textsuperscript{11-13}, trapped ions\textsuperscript{14,15}, and photonic systems\textsuperscript{16} to superconducting circuits in solid-state devices\textsuperscript{17}. The still challenging goal is to realize a large-scale quantum simulator (with less demanding technical requirements) which cannot be efficiently solved numerically with classical computers. In this work, we propose a scalable architecture that is of practical interest for large-scale quantum simulation. Our quantum simulator is based on lattices of interacting nuclear spins, which can be fabricated chemically on the diamond surface\textsuperscript{18,19} or by depositing functionalized graphene films\textsuperscript{20}. We propose to exploit the nitrogen-vacancy (NV) centres\textsuperscript{21} beneath the diamond surface as an efficient control element for cooling, spin–spin interaction engineering, and read-out of the nuclear-spin quantum simulator.

We explain how this simulator is constructed, and establish schemes for its initialization, control and read-out. We analyse its validity by detailed numerical studies, thus demonstrating the feasibility of our proposal within the current experimental capabilities.

Construction of hardware

We discuss two main paths for the fabrication of the hardware for our quantum simulator. First, large-scale lattices of nuclear spins can be constructed by chemically controlled termination of diamond surfaces. The fluorine (\textsuperscript{19}F with spin 1/2), oxygen (\textsuperscript{17}O with spin 5/2) and hydrogen/hydroxyl group (\textsuperscript{2}H with spin 1/2 and \textsuperscript{3}H with spin 1) termination of the diamond surface can be obtained from the process of chemical vapour deposition, or by functionalizing the diamond surface. As a representative example, we will mainly concentrate on the fluorine-terminated diamond surface, which has a positive electron affinity. The two most important diamond surfaces are the (111) and (100) surfaces, which constitute the crystal faces of polycrystalline chemical vapour deposition diamond films and can be selectively grown with appropriately controlled process parameters\textsuperscript{18}. The (111) surface of diamond is the natural cleavage plane of diamond, and has one dangling bond per surface carbon atom which is terminated by carbon–fluorine bonds on the fluorine-terminated diamond surface\textsuperscript{22}. The fluorine atoms naturally arrange in a triangular lattice with nearest-neighbour distances of about 2.5 Å (refs 18,19; Fig. 1a). The (100) surface of diamond shows two dangling bonds per surface carbon atom, which will undergo a reconstruction into a $2 \times 1$ geometry with neighbouring surface carbon atoms forming a π-bonded dimer. The remaining dangling bonds are terminated by carbon–fluorine bonds, which leads to a rectangular lattice of fluorine atoms\textsuperscript{18,19}, Fig. 1b. Functionalized graphene (fluorographene) provides a double-layer triangular lattice of fluorine atoms, Fig. 1c. This can be obtained through the mechanical cleavage of graphite fluoride, or by exposing graphene to atomic fluorine formed by decomposition of xenon difluoride (XeF\textsubscript{2}; ref. 20).

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In addition to the nuclear spins, we shall introduce NV centres by shallow implantation a few nanometres below the surface of diamond\textsuperscript{23,24}. This constitutes a fundamental ingredient of our quantum simulator, as it allows the initialization and read-out of the nuclear spins. Let us remark that in contrast to graphene, fluorographene exhibits a large band gap of more than 3.5 eV, which is larger than the optical gap of NV centres in diamond (\textasciitilde 2.9 eV). This avoids the unwanted fluorescence quenching of NV centres and is thus crucial for using the NV centres for the control of the nuclear spin arrays.

**Engineering of the interacting Hamiltonian**

The nuclear spins interact with each other via magnetic dipole–dipole interactions as $V_{ij} = g(r_{ij})[s_i \cdot s_j - 3(s_i \cdot r_{ij})(s_j \cdot r_{ij})]$, where $s_i$ are the nuclear spin operators, $g(r_{ij}) = (\hbar^2 \mu_0 / 2m_i \cdot r_{ij}^3)$, and $g_i$ and $g_j$ are gyromagnetic ratios of the $i$th and $j$th nuclear spin, which are connected by the vector $r_{ij} = r_i - r_j$. Because nearest-neighbour fluorine nuclear spins are at a distance of 2.5 Å, a coupling strength of $g(2.5) \approx 6.8$ KHz is achieved. A strong magnetic field, which leads to energy level shifts exceeding the nuclear spin coupling strength, simplifies the effective Hamiltonian due to the rotating wave approximation to an XXZ model

$$V_0 = g(r_{ij}) \left[ s_i^z s_j^z - \Delta (s_i^+ s_j^- + s_i^- s_j^+) \right]$$

with $\Delta = 1/2$. Our calculations verify that such a rotating wave approximation can already be well satisfied for a magnetic field as low as 750 G (3 MHz), see Supplementary Information. We denote the diamond surface on which the nuclear spin lattice is constructed as the $X$–$Y$ plane, while the vector perpendicular to it defines the $Z$ axis (spatial axes), and the magnetic field direction as the $\hat{m}$ axis, which gives the quantization of nuclear spins. The spin–spin interaction strength can be tuned by changing the direction of the magnetic field as

$$g(r_{ij}) = g(r_{ij})(1 - 3 \cos^2 \theta_{ij})$$

where $\theta_{ij}$ is the relative angle between $r_{ij}$ and $\hat{m}$. Thus, by changing the direction of the external magnetic fields, we can control the spatial anisotropy and the sign of the interaction strength, which determines whether the interaction is ferromagnetic or anti-ferromagnetic and thereby induces spin frustration. The value of the spin anisotropy $\Delta$ can be tuned with gradient magnetic fields or radio-frequency fields.

**Cooling of the quantum simulator**

The reliable preparation of the quantum simulator in a low-entropy state is a prerequisite for the observation of quantum phases. Here we explain in detail how the nuclear spin lattice can be initialized to a well defined spin direction using dynamical nuclear polarization via optical pumping NV centres in diamond, Fig. 2, which works at room temperature (similar to the laser cooling of trapped ions). The entire process consists of repetitive cycles. In each cycle, the NV centre is first prepared in the $| + \rangle = \sqrt{1/2}|m_z = 0 \rangle + |m_z = +1 \rangle$ state by optical pumping to the $| m_z = 0 \rangle$ state using a green laser (532 nm) and a subsequent $\pi/2$ microwave pulse. A microwave field (with Rabi frequency $\Omega_{NV}$) on resonance with the electronic transition $| m_z = 0 \rangle \leftrightarrow | m_z = +1 \rangle$ will lock the NV electron spin state. If the Hartmann–Hahn condition with nuclear spins\textsuperscript{25} is satisfied, the NV electron spin polarization will be transferred to the nuclear spins\textsuperscript{26}. Although an individual NV centre suffices, this process can be made swifter by using a few NVs (Supplementary Information). Owing to their proximity, the interactions between nuclear spins exceed those between the electron spin of the NV centre and the nuclear spins. Therefore, it will be advantageous to effectively decouple nuclear spin interactions, which also facilitates estimates of the cooling efficiency. To this end, we apply a radio frequency field with the amplitude $\Omega_{rf}$ whose frequency is detuned from the Larmor frequency of the nuclear spins $\omega_0$ by $\Delta p$. The energy conserving terms of nuclear spin interactions cancel each other when $\Omega_{rf} = \sqrt{2} \Delta p$ (ref. 27), and the anti-rotating terms are suppressed as long as the energy mismatch $\omega_0 = (\Omega_{rf}^2 + \Delta p^2)^{1/2} \gg \gamma_0$ is fulfilled. Thus, the nuclear spins behave as isolated particles, and the Hartmann–Hahn condition becomes $\Delta_{NV} = (\omega_0 - \Delta p) + (\Omega_{rf}^2 + \Delta p^2)^{1/2}$, see Supplementary Information. Such a mechanism for the isolation of the nuclear spins (as we will discuss in the following section) can also reduce the perturbation on the nuclear spin state during the read-out process, which will be beneficial for the accuracy of the measurement.

To verify the validity of this idea, we have used a Chebyshev expansion\textsuperscript{28} to calculate polarization dynamics with the exact Hamiltonian of a $3 \times 3$ nuclear spin lattice, assuming a distance of 5 nm from the NV centre. It can be seen from Fig. 3a that the coupling between nuclear spins is effectively eliminated. We compare the exact numerical calculations with the results for isolated nuclear spins under the spin temperature approximation in which
coherences between nuclear spins are neglected, Fig. 3b; these show a good agreement. To achieve an ultra-high polarization given by the spin temperature approximation, one can introduce magnetic noise to remove coherence among nuclear spins in between polarization cycles. From the spin temperature approximation, one can estimate the polarization rate, and the required polarization time scales linearly with the total number of nuclear spins $N$ and the inverse effective temperature. The ultimate polarization efficiency will be limited by the relaxation time $T_1$ of nuclei, which can be as long as a few hours even at room temperature. The polarization cycle time sets the required coherence time of NV centres and nuclear spins to a few hundreds of microseconds, which is readily achievable with the current experimental techniques in diamond samples. The polarization efficiency can also be improved by optimizing the polarization cycle time and exploiting several NV centres. Once the nuclear spins have been initialized, by performing an adiabatic passage, one can prepare the system in the ground state of the engineered interacting Hamiltonian (similar to other types of quantum simulator); see Supplementary Information for an explicit example which demonstrates that adiabatic state preparation permits the observation of different quantum phases. During the operation of quantum simulation, a green laser can be used to decouple the nuclear spins from the NV electron spin.

Measurement of the quantum simulator

Before discussing the static and dynamical properties of the proposed quantum simulator, let us describe the measurement schemes and demonstrate their viability by means of numerical simulations. The main goal is to measure observables that can provide information about the nuclear spin state. It is challenging to measure nuclear spins directly because of their small magnetic moments. However, NV centres implanted beneath the diamond surface will provide the solution as a measurement interface for nuclear spin states. Before the measurement, we apply a radio-frequency pulse to map the nuclear spin basis from $|\uparrow\rangle$ and $|\downarrow\rangle$ to $|\uparrow\rangle$ and $|\downarrow\rangle$, in which the nuclear spins can be effectively decoupled from each other by a continuous driving field, as described in the process of initialization. The NV centre is prepared in the state $|\uparrow\rangle$ ($\mu = \pm$), and is driven to match the Hartmann–Hahn condition between the electron spin of the NV centre and the nuclear spins. After the electron spin interacts with the nuclear spins for time $\tau$, we measure the population of state $|\psi\rangle$ of the NV centre, which is given by $P_{\psi} = \tau^2 \sum_i (g^+_{\psi} g^-_{\psi}) (\hat{S}_i^+ \hat{S}_i^-)$ in second order in $\tau$, where $\mu, \nu = \pm$ and $g^+_{\psi}, g^-_{\psi}$ represent the rate of polarization exchange between the NV centre and the nuclear spins (Supplementary Information). The above observables include both local contributions of individual nuclear spins (for $i = j$) and two-point correlations of nuclear pairs (for $i \neq j$).

With a field gradient, we can estimate quantities such as structure factors, which are very important for the study of quantum phase transitions and for inferring entanglement properties, from the observables $A_{\alpha\beta} = \tau^2 \sum_i (g^+_{\alpha} g^-_{\beta}) \cos(q \cdot (r_i - r_j))/(s_i^+ s_j^- + s_i^- s_j^+)$ with $\alpha, \beta = x, y, z$. By one single magnetic tip with the state-of-art technology, it is possible to generate a gradient field as large as 15 G (about ten times larger than the coupling between fluorine nuclear spins) over the lattice constant (2.5 Å). We remark that the scheme also works by exploiting an ensemble of NV centres (Supplementary Information). The validity of our measurement scheme is numerically tested in the context of witnessing quantum phase transitions as discussed in the following section.

Frustrated quantum magnetism and supersolids

Our system can simulate quantum spin models with the tunable spin–spin interaction $g(r_{ij})$: positive $g(r_{ij})$ corresponds to anti-ferromagnetic (AF) interactions, and negative $g(r_{ij})$ corresponds to ferromagnetic (F) interactions. In the triangular lattice of the fluorine simulator, the nearest-neighbour nuclear spin interactions are denoted as $J_{ij} = J_{ij}, J_{ij}, J_{ij}$, with $\alpha = \{1, 0, 0\} \delta_1 = (1/2, \sqrt{3}/2, 0)$ and $\delta_2 = (-1/2, \sqrt{3}/2, 0)$. The long-range interactions and spin frustration make it hard to perform numerical simulations using the Quantum Monte Carlo method owing to the subtle sign problem. Here, we exactly diagonalize the system on a $4 \times 4$ lattice using the Lanczos algorithm under periodic boundary conditions to provide evidence for various phases of quantum magnetism.

In the situation where $J_{ij} = J_{ij} = g_{ij}$ is positive (anti-ferromagnetic), and $J_{ij} = J_{ij} = g_{ij}(1 - (9/4) \cos^2 \theta)$ is negative for $\cos \theta \geq 2/3$ (the magnetic field direction is $m = (\cos \theta, \sin \theta, \sin \theta)$), the triangle which consists of $J_1 = J_2 = J_3$ is spin frustrated (Fig. 4a). For small values of $J_2/J_1$, the system consists of 1D (anti-ferromagnetic) chains with weak intra-chain interactions that induce ferromagnetic order in the sublattice of every two 1D chains and are characterized by the (normalized) spin structure factor $S_q = (1/N^2) \sum_\mathbf{r} \mathbf{e}^{i \mathbf{q} \cdot \mathbf{r} + \mathbf{q} \cdot \mathbf{r}}$ with $\mathbf{q} = (\pi, 0)$. As $|J_2/J_1|$ increases, the competition between anti-ferromagnetic ($J_1$) and ferromagnetic ($J_3$) interactions leads to the ferromagnetic phase above the point $|J_2/J_1| = 1$, corresponding to the spin structure factor $S_0(0, 0)$ (Fig. 4a, lower left). Note that the largest non-nearest-neighbour interaction $J_3$ is ferromagnetic and is essential for the emergence of...
the ferromagnetic phase which is absent for the short-range model (Supplementary Information).

For the other situation, \( J_0 = J_1 = J_2 = g(a)(1 - (3/4)\cos^2 \theta) \) is always positive (anti-ferromagnetic), and \( J_0 = J_1 = g(a)(1 - 3\cos^2 \theta) \) is negative (ferromagnetic) for \( \cos \theta > 1/3 \), where \( \theta \) is defined by the magnetic field direction as \( \hat{m} = (\cos \theta, 0, \sin \theta) \). Considering only the nearest-neighbour interactions, the system is non-frustrated, and is expected to be ferromagnetic in the \( \hat{a}_1 \) direction and anti-ferromagnetic in the \( \hat{a}_2 \) direction (F–AF phase), which is identified by the spin structure factor \( S_{q} \) with \( q = (0, \pm 2\pi/\sqrt{3}) \). As the value of \( \cos \theta \) approaches 1, the non-nearest-neighbour interactions \( J_0, J_1, J_2 = (1 - (9/4)\cos^2 \theta)/3\sqrt{3} \) become comparable to \( J_2 \). The competition between them leads to a magnetic phase identified with the spin structure factor \( S_{q} \) with \( q = (0, \pm \pi/\sqrt{3}) \). The spins are ferromagnetic in the \( \hat{a}_1 \) direction, whereas they are anti-ferromagnetic between next-nearest-neighbour chains in the \( \hat{a}_2 \) direction (Fig. 4b, lower right).

We also check the feasibility of using NV centres to identify different magnetic phases. We numerically calculate the dynamics during measurement and obtain the estimation of spin structure factors. Owing to the limit of computational overhead, we consider an example of a 4 × 4 nuclear spin lattice with periodic boundary conditions. By applying a gradient field we can generate the relative phases between different nuclear spins corresponding to the spin structure factor, which can then be estimated by the observable \( S_{q}(\hat{a}_1, \pm \Delta \hat{a}_1) \) via NV centres. We find that the estimation is in good agreement with the results from exact diagonalization, and that it can witness quantum phase transitions between different magnetic orders, Fig. 4a,b (lower right) and Supplementary Information for more details.

The nuclear spin Hamiltonian can be mapped to the hard-core boson model by the Holstein–Primakoff transformation. Note that quantum simulation of similar models with polar molecules in optical lattices has been proposed\(^{36,37}\) and numerically studied\(^{38,39}\) for long-range repulsive dipole interactions (Vij). Our system possesses long-range interactions (both Vij and Vij), thus offering a platform to investigate rich phases of hard-core bosons with potentially novel features. Indeed, these models with frustration and long-range interactions pose considerable challenges to the classical numerical simulations because of the sign problem for 2D systems. We simulate such a model with the directed loop algorithm in the stochastic series expansion representation of the

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**Discussion and outlook**

In addition to the static properties of quantum phases such as spin liquids and topological phases\(^4\), the present quantum simulator is capable of studying quantum many-body dynamics, such as quantum quenches and the generation of multi-particle entanglement (Supplementary Information). It would also be interesting to exploit the ideas in room temperature quantum information processing with NV centres\(^5\). The prerequisite technologies for the implementation of such a quantum simulator, such as the techniques for charge state manipulation of NV centres in (surface-functionalized) diamond\(^6\), shallow NV implantation\(^7\),\(^8\), coherent control and readout of NVs\(^9\),\(^4\),\(^6\), and Hartmann–Hahn spin locking with NVs, are currently being developed. The proposal will simulate the interest of material scientists in fabricating other candidate systems, for example, a few layers of \(^{13}\)C grown inside diamond\(^10\) and graphene heterostructures assembled layer by layer in a desired sequence\(^11\). The relevant tools in this work may also be beneficial for further research in coherent control on surfaces/mono-layer films.

**Methods**

**Tuning spin anisotropy with radio-frequency fields.** The value of the spin anisotropy \(\Delta\) can be tuned with gradient magnetic fields to modulate the hopping coupling \((s_i^+s_j^- + s_j^+s_i^-)\) while keeping the repulsive interaction \(s_i^s_j^\) unchanged\(^3\). In this way the interplay between geometrical frustration and the effects of quantum fluctuations on the realization of a spin liquid phase could be tested\(^12\). As an alternative scheme with higher nuclear spin species, for example \(^{13}\)H with spin \(I = 1/2\), one can tune the spin anisotropy \(\Delta\) by applying continuous fields corresponding to the nuclear spin transitions \(|m = 1\rangle \leftrightarrow |m = 0\rangle\). We can extract information from the average nuclear spin magnetization by \(\Delta_\alpha = P_1^\alpha - P_2^\alpha - 2\tau\sum_i (g_i^\alpha)^2\langle s_i^\alpha\rangle\), and the transverse correlation as \(\Delta_\alpha = P_2^\alpha - P_1^\alpha - 2\tau\sum_i (g_i^\alpha)^2\langle s_i^\alpha s_i^\alpha\rangle\). The correlation function along the other directions, \(\Delta_{\alpha\beta}\), can be obtained by applying the Hadamard operation \(O_H = |1\rangle|+\rangle |+\rangle |1\rangle\) with \(|\beta\rangle = \sqrt{2}|\uparrow\rangle |\uparrow\rangle\), and the phase transformation \(O_t = |1\rangle|+\rangle |+\rangle |1\rangle\) on the nuclear spin state before the measurement. To measure observables such as the structure factor, we can use a gradient field with which the nuclear spin at the position \(r_i\) experiences a magnetic field \(b_i = r_i \times b\), and gains a position-dependent phase \(\phi_i = \tau_i \cdot q\) after time \(t_p\), where \(q = (g_i^\alpha, g_i^\beta) = \gamma_i \mu_i B_i(r_i)\) after a certain time \(t_p\), and \(\gamma_i\) is the gyromagnetic ratio of nuclear spins. By performing the same measurement as before, we can obtain \(\Delta_{\alpha\beta} = t^2\sum_i (g_i^\alpha)^2\cos(q \cdot r_i)\langle s_i^\alpha s_i^\alpha\rangle\) and \(\Delta_{\alpha\beta}\) in a similar way.

**Figure 6 | Tuning spin anisotropy with dressed states.** Simulation of an effective spin 1/2 by the dressed states from a higher spin by applying radio frequency driving fields (with Rabi frequency \(\Omega_0\) and detuning \(\Delta\)).

**Map to hard-core boson model.** The nuclear spin Hamiltonian can be mapped to the hard-core boson model by the Holstein–Primakoff transformation as

\[
H_0 = \sum_{n \geq 0} \left( | \Omega_0 n \theta | n - t (\delta a^\dagger a^\dagger + \delta a) \right) + \nu \sum_n n
\]

with \(n = m + 1/2\) (\(n = 0, 1\)). Here, the chemical potential is \(\mu = (\gamma_0 B) - \sum g(r_i)\), the repulsive interaction is \(V_r = g(r_i)\), and the hopping is \(t = \Delta/2g(r_i)\).

The system can therefore simulate the hard-core boson model, which demonstrates interesting phases such as long-range off-diagonal order superfluids, and moreover a supersolid phase characterized by both long-range off-diagonal and diagonal order. With the magnetic field along the direction \(m = \cos \theta_0 + \sin \theta_0 \hat{z} = (-1/2) \cos \theta_0, \sqrt{3}/2 \cos \theta_0, \sin \theta_0\), the nearest-neighbour interactions are \(V_r = V_r = g(1 - 3/4 \cos \theta_0)\) and \(V_r = V_r = g(1 - 3/4 \cos \theta_0)\).

By changing the value of the magnetic direction angle \(\theta_0\), we can gradually tune the geometric frustration as quantified by the ratio \(V_r/V_r\). For \(\cos \theta_0 \in [0, \sqrt{3}/7]\), the values of \(V_r\) for all interactions have the same sign (including long-range interactions), and one can simulate such a model with Quantum Monte Carlo methods. By comparison with the short-range model, we find that the long-range interaction significantly enhances the superfluidity (Supplementary Information). One can also use a gradient field to selectively tune hopping interactions. For example, with a gradient field that decreases along the direction \(\Delta \theta = (\sqrt{3}/2, 1/2)\), the hopping interaction will be suppressed except in the direction \(\Delta \theta = (1/2, 3/2)\).

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Author contributions

M.B.P. proposed the idea and developed it further together with F.J. and J.C. J.C. carried out the numerical and analytical work with advice from F.J., M.B.P. and A.R. All authors discussed the results. J.C. drafted the manuscript with input from F.J., M.B.P. and A.R.

Additional information

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Competing financial interests

The authors declare no competing financial interests.