Analogue quantum chemistry simulation

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Computing the electronic structure of molecules with high precision is a central challenge in the field of quantum chemistry. Despite the success of approximate methods, tackling this problem exactly with conventional computers remains a formidable task. Several theoretical1–2 and experimental3–5 attempts have been made to use quantum computers to solve chemistry problems, with early proof-of-principle realizations done digitally. An appealing alternative to the digital approach is analogue quantum simulation, which does not require a scalable quantum computer and has already been successfully applied to solve condensed matter physics problems6–8. However, not all available or planned setups can be used for quantum chemistry problems, because it is not known how to engineer the required Coulomb interactions between them. Here we present an analogue approach to the simulation of quantum chemistry problems that relies on the careful combination of two technologies: ultracold atoms in optical lattices and cavity quantum electrodynamics. In the proposed simulator, fermionic atoms hopping in an optical potential play the role of electrons, additional optical potentials provide the nuclear attraction, and a single-spin excitation in a Mott insulator mediates the electronic Coulomb repulsion with the help of a cavity mode. We determine the operational conditions of the simulator and test it using a simple molecule. Our work opens up the possibility of efficiently computing the electronic structures of molecules with analogue quantum simulation.

Quantum computers are expected to have a considerable impact in several areas of science because they will be able to tackle problems that are intractable with classical devices. Particularly relevant are quantum many-body problems involving several systems that interact with each other according to the rules of quantum physics9. Given the current theoretical and experimental progress, the most timely and important ones are quantum chemistry problems, which generally involve obtaining the ground-state energy of many electrons that interact with nuclei and with each other through Coulomb interactions. Current approaches to the simulation of chemistry problems with quantum computers follow the digital approach10–12, in which one breaks the complete Hamiltonian into gates that are applied in a time-dependent manner. An alternative way to address quantum many-body problems is analogue quantum simulation13. The idea is to use a well controlled quantum system (the simulator) and engineer its interactions according to the Hamiltonian under investigation. This approach has already been used to address questions that the most advanced classical computers cannot resolve6–8. The key feature is that their interactions are either local or short-range, being ideally suited for existing simulators. By contrast, analogue simulation of quantum chemistry requires engineering long-range (Coulomb) interactions between fermionic particles, and no system has been identified so far to fulfil such a requirement. This is why current efforts concentrate in digital simulation.

Here we show how to build an analogue simulator for quantum chemistry problems by bridging two paradigmatic systems, namely, ultracold atoms in optical lattices14–16 and cavity quantum electrodynamics (QED)17–21. Fermionic atoms trapped in a periodic three-dimensional (3D) optical potential play the role of electrons and are subject to additional optical potentials emulating their interaction with the nuclei. The key feature of the scheme is the trapping of another atomic species in a Mott insulator regime with several internal states such that its spin excitations mediate effective forces between the simulated electrons. We show that even though the interaction is local, one can induce Coulomb-like forces among the fermionic atoms in a scalable manner. Although the setup is discrete and finite, we show that precise results can be obtained for simple molecules with moderate lattice sizes. Apart from the standard advantages of analogue simulation over quantum computing regarding the required control13, the present scheme does not rely on a judicious choice of molecular orbitals12, but operates directly in real space, improving convergence to the exact result as the system size increases.

One of the main goals of quantum chemistry is to obtain the low-energy behaviour of $N_e$ electrons and several nuclei when the positions, $r_n$, of the nuclei are fixed. Using a cubic discretization, in real space of $N \times N \times N$ sites, the electronic Hamiltonian contains three terms, $H_{\text{qc}} = H_{\text{kin}} + H_{\text{nuc}} + H_{\text{e-e}}$ (using $\hbar = 1$ for the reduced Planck constant, and dropping the spin index)

\[
H_{\text{kin}} = - t \sum_{\langle i,j \rangle} f_i^\dagger f_j
\]

\[
H_{\text{nuc}} = - \sum_{n,i,j} Z_n V (\lvert j - r_i \rvert) f_i^\dagger f_j
\]

\[
H_{\text{e-e}} = \sum_{i,j} V (\lvert i - j \rvert) f_i^\dagger f_j f_i^\dagger f_j
\]

where $f_i$ are annihilation operators of electrons at site $i$ fulfilling $\langle f_i^\dagger f_j \rangle = \delta_{ij}$ and $(i,j)$ denote nearest-neighbour sites. $H_{\text{kin}}$ describes electron hopping at rate $t$, $H_{\text{nuc}}$ represents the electron–nucleus attraction when the nuclei are at positions $r_n$ and $H_{\text{e-e}}$ accounts for the electron–electron repulsion. In both $H_{\text{nuc}}$ and $H_{\text{e-e}}$, the attractive repellent potential has the standard Coulomb form, $V(r) = V_0/r$. The connection between the length/energy scales of the discrete Hamiltonian $H_{\text{qc}}$ and the continuum one is given by:

\[
a_0 = \frac{2t}{V_0} \quad \text{and} \quad R_y = \frac{V_0^2}{4t^2}
\]

where $a_0$, $a$ and $R_y$ are the Bohr radius, lattice spacing and Rydberg energy, respectively. Thus, we work in a regime:

\[
1 \ll \frac{2t}{V_0} \ll \frac{N}{N_0^{1/3}}
\]

such that the first inequality prevents discretization effects and the second one guarantees that the molecule fits in the volume of the simulator. Our simulator then requires three components (see Fig. 1a): (i) cold spin-polarized fermionic atoms hopping in a 3D optical potential with a tunable tunnelling rate, $J_0$, which play the role of electrons14. We consider spinless fermions, but the spin degree of freedom can be included using an extra internal level22 (see Methods). (ii) Additional potentials
to emulate the attraction between fermions and nuclei. Given that this is a single-particle Hamiltonian, it can be created through optical Stark shifts with an adequate spatial modulation. For example, one can use holographic techniques\(^{24,25}\) with judiciously optimized phase masks to engineer a Coulomb-like spatial potential at the fermionic positions (see Methods). (iii) The most difficult part to simulate is \(H_\text{fermions}\), because it involves repulsive interactions between the fermions with a \(1/r\) dependence. Inspired by how virtual photons mediate Coulomb interactions in QED, we use a spin excitation of another atomic species forming a Mott insulator to mediate the Coulomb forces between fermions (see Fig. 1b). This species is composed of \(N_0^b\) atoms trapped in an optical potential with the same spacing as that of the fermions and with two additional internal atomic states, \(|a⟩\) and \(|b⟩\), which describe spin excitations. Spin excitations in the \(|a⟩\) state interact repulsively and locally with the fermionic atoms with strength \(U\), and propagate through the long-range couplings induced by a cavity mode with rate \(J\) (refs.17-21). The \(|b⟩\) internal state is subject to a different optical potential, such that its itinerant spin excitation propagates through standard nearest-neighbour exchange\(^{26}\) at rate \(J\). Furthermore, an external field (Raman laser or a radiofrequency field) drives the \(|a⟩\)–\(|b⟩\) transition with coupling strength \(g\) and detuning \(\Delta\). The complete simulator Hamiltonian after the elimination of the cavity mode is \(H_{\text{sim}} = H_{\text{kin}} + H_{\text{nat}} + H_{\text{M}}\), with

\[
H_{\text{M}} = \Delta \sum_j b_j^\dagger b_j + \sum_{(ij)} b_i^\dagger b_j + \frac{J}{N^2} \sum_{i,j} a_i^\dagger a_j + U \sum_{i,j} a_i^\dagger a_i^\dagger f_i f_j + g \sum_{j} (a_j^\dagger b_j + b_j^\dagger a_j)
\]

where \(a_i\) and \(b_j\) are annihilation operators for \(|a⟩\) and \(|b⟩\) spin excitations, respectively, at site \(j\). Intuitively, the on-site interaction \(U\) localizes the \(|a⟩\) and \(|b⟩\) excitations around the fermions, renormalizing their tunnelling rates and creating an effective interaction. Mathematically, one can eliminate the Mott insulator excitations adiabatically and derive the effective dynamics for the fermions (see Supplementary Information

**Fig. 1 | Schematic representation of the analogue simulator.** a, Fermionic atoms, playing the role of electrons, are trapped in a periodic 3D cubic potential. Their hopping simulates the kinetic energy term of the electrons, and they are subject to additional optical potentials that emulate the nuclear interaction. b, Coulomb repulsion among the fermions is mediated by a spin excitation of a Mott insulator with three internal levels. Excitations in level \(|b⟩\) are allowed to propagate through spin-exchange interactions with rate \(J\) (inset, top). Excitations in level \(|a⟩\) experience a strong repulsive interaction with the fermions and interact with a cavity mode. These two levels are coupled through either a microwave or a two-photon Raman transition (inset, bottom). c, The complete simulator for the \(\text{H}_2\) molecule. Although a 2D lattice is pictured, the experimental proposal presented here refers to a 3D optical lattice.

**Fig. 2 | Atomic hydrogen spectrum dependence on the effective Bohr radius.** a, Lower part of the spectrum of the atomic Hamiltonian \(H_\text{nat}\), for a cubic lattice of \(N = 100\). Different symbols represent different energy orbitals, and the first three atomic levels \((n = 1, 2, 3)\) of the continuum Hamiltonian are represented by the dashed lines. In the blue-shaded region, the hopping parameter is \(t_F/V_0 < 0.5\), the Bohr radius is smaller than the lattice spacing, and energies are highly affected by the cut-off of the nuclear potential. As the hopping parameter \(t_F/V_0\) increases, the simulator effectively zooms in on the system, as we show in the bottom insets, which present the fermionic density of the second-lowest energy orbitals shown in the main graph. By increasing \(t_F/V_0\) we include more lattice sites in the simulation, reducing systematic deviations as \((t_F/V_0)^{-2}\) (red dashed line in top inset), as we show in the top inset for the lowest energy state (see Supplementary Information section 1). At higher values of \(t_F/V_0\), solutions suffer from finite-size effects. b, Axial cut in the central positions of the lattice for the first nine eigenstates of \(H_{\text{nat}}\), for \(t_F/V_0 = 2\) and \(N = 150\). c, By choosing the appropriate Bohr radius, the same orbital can be obtained with \(N = 1,000\) (top; \(t_F/V_0 = 150\)) or \(N = 20\) (bottom; \(t_F/V_0 = 3\)), where the discretization of the system is more noticeable.
in the projective basis until convergence is observed. For a Coulomb potential (blue dots), the result agrees with an accurate solution in the nonrelativistic regime\(^\text{28,29}\) (dashed line). As \(L\) decreases, the exponential decay in the Yukawa potential prevails, underestimating Coulomb repulsion and lowering the molecular potential. c, This underestimation of the repulsive potential is more evident when the condition \(N \ll L/a\) is violated (inset). d, By changing the ratio \(F\) between the electronic and nuclear potentials, one can explore artificial repulsive interactions that form pseudomolecules in more relaxed experimental conditions. The dotted line represents the limit of zero repulsion in the absence of a mediating excitation.

We now benchmark our simulator for moderate system sizes using numerical simulations. In Fig. 2 we solve the hydrogen problem in a lattice to explore discretization and finite-size effects by comparing the energies of the low-lying excited states with that of the continuum. We show that an error of 0.3% with respect to the exact energy can be obtained for systems of \(N = 100\). In Fig. 3 we analyse the accuracy of the simulator for the simplest molecule, \(H_2\). First, we compute exactly the energy of the spin excitation that mediates the fermionic repulsion, and \(\rho_M = N_e/N_M\). This mapping between \(H_\text{sym}\) and \(H_\text{qc}\) holds as long as

\[I_c \ll U\]  

\[I_c \rho_M\]  

\[V_{\text{e}} N_e^{7/3} \ll f(aN/L)^2\]

Condition (8) enforces that the \(|a\) excitation is localized symmetrically only around the position of the fermions; condition (9) guarantees that neither the tunnelling of the fermions nor the interaction with the \(|b\) excitations dominate over the cavity interaction; and condition (10) ensures that the Yukawa potential does not depend on the fermionic positions. Furthermore, to obtain a truly Coulomb repulsion, the length \(L\) must be larger than the fermionic lattice with \(N\) sites but smaller than the Mott insulator size, that is:

\[N \ll L/a < N_M\]

When inequalities (5), (8)–(11) are satisfied, the exact solution in the continuum limit is recovered in the limit \(N_M > N \to \infty\). Thus, the finite size of the simulator is what ultimately limits the precision of the simulation.
Online content

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**METHODS**

Here we provide further details and candidates for the experimental implementation of some of the key components of the proposed method.

**Holographic engineering of the nuclear potential.** Holographic techniques are a possible approach to engineering the 3D Coulomb potential ‘seen’ by the fermionic atoms, \( H_{\text{lat}} \). Following this method, the potential is created by imprinting a phase pattern on an incoming laser, as in ref. 23; where it was used to experimentally induce microcrystals for single atoms. The same technique can be applied to engineer various intensity patterns at the lattice sites where the fermions are trapped by choosing an appropriate phase mask. To do this, we particularized the algorithm used in that implementation (Gerchberg–Saxton algorithm\(^{36, 37}\), G-S) to identify the phase mask that provides the required geometry (a 3D Coulomb potential, \( V_i = V(\|l_i - n\|) \)).

We then use the ‘ping-pong’ strategy described by the discretized G-S algorithm to retrieve the phases of the holographic field \( u \) associated with our intensity distribution \( \{u_{ij}\} = V_j \). This algorithm iterates over the following steps 2-3: (i) the field is transformed to reciprocal space via a fast Fourier transform (FFT), obtaining \( \hat{u} = \text{FFT}(u) \); (ii) We restrict the calculation to those \( k \) vectors that satisfy the physical constraints of the system; that is, they must sit on an Ewald sphere of radius \( k_0 = 2\pi/\lambda \), where \( \lambda \) is the wavelength of the monochromatic input light. The remaining contributions are neglected, defining a constrained field in Fourier space, \( u^* \); (iii) We use an inverse FFT (IFFT) to transform the field back to real space, \( u = \text{IFFT}(u^*) \). At this point, \( u^* \) satisfies diffraction laws but may differ from the goal potential. (iv) The objective potential is combined with the phases of the constrained solution, \( \varphi = \arg(u^*) \), giving \( u = |u|^2 \exp(i\varphi) \). To improve the accuracy, we use a refined phase frame of size \( (n_\text{ref}N) \times (n_\text{ref}N) \), with \( N \) being the number of lattice sites per side and \( n_\text{ref} \) a refining factor (see insets in Extended Data Fig. 1). Because the trapped atoms are only affected by the value of the holographic potential at the lattice sites, these are the only coordinates where the field is updated in step (iv).

In Extended Data Fig. 1 we show the result of applying this optimization process to the Coulomb potential. By quantifying the normalized relative error\(^{22}\)

\[
\epsilon = \frac{1}{N^2} \sum_j \left( \frac{|V_j - \tilde{V}_j|^2}{\tilde{V}_j^2} \right) \tag{12}
\]

between the desired Coulomb potential \( V_j \) and the intensity pattern obtained numerically (\( V_j = |u_j|^2 \)), we observe that the accuracy obtained for \( n_\text{ref} = 3 \) already provides normalized relative errors smaller than 0.3% for \( N = 30 \). More precise results can be obtained by increasing the number of iterations of the algorithm or the refining factor.

**Candidate atomic species.** The proposed method is based on the interplay between two atomic species: (i) fermionic atoms, which have two internal levels and play the role of the electronic spin; and (ii) mediating atoms, which must have three levels available: \( |0\rangle \) for the ground state in the Mott insulator, \( |a\rangle \) for the spin excitation and \( |b\rangle \) for the state that tunnels and induces the effective repulsion.

All atoms need to see the same lattice, although with tunable tunneling amplitudes. The fermions must additionally see the external potential and have to interact with the internal state of the mediating atoms. The scattering lengths for the interactions corresponding to levels \( |0\rangle \), \( |a\rangle \) and \( |b\rangle \) do not need to be the same, and we require the scattering length between the fermions and the mediating atom in state \( |b\rangle \) to be negligible, so that the scaling of the moving excitation is unchanged. Additionally, the mediating atoms in \( |a\rangle \) have to be exposed to the cavity mode. We note that the atoms that mediate the fermionic interactions can correspond to both bosons and fermions, as there is only a single excitation.

Over the past years, many atomic species have been trapped, condensed and used in experiments with optical lattices. For illustration, let us give a particular example using fermionic and hyperfine alkaline-earth atoms. These atoms offer a rich internal structure, with long-lived excited metastable states \( ^{3}\text{P}_2 \) and \( ^{3}\text{P}_0 \).

We can use \(^{88}\text{Sr}\) as a simulator for electrons, which has a nuclear spin of \( I = 9/2 \). Similarly to ref. 23, we can encode the spin of the simulated electron into the system by using a Raman transition of \( 1S_0 \) to \( 1S_2 \), where the spin is encoded in the magnetic state \( |\uparrow\rangle \) or \( |\downarrow\rangle \), corresponding to the occupied \( |0\rangle \) or empty \( |3\rangle \) levels. For this, we use \(^{87}\text{Sr}\) as a simulator for electrons, which has a nuclear spin of \( I = 9/2 \).

The fermionic states can be written as \( |n, \psi\rangle = \frac{1}{\sqrt{N}} \sum_{k} \psi_k |k\rangle |n\rangle \), where \( |n\rangle \) is the fermionic Fock state and \( \psi_k \) are the normalized single-particle orbitals. Finally, we adiabatically introduce the electronic repulsion, completing the Hamiltonian.

As an initial state, we begin with both fermions tightly trapped in a lattice site \( j_0 \). As discussed in the main text, this corresponds to growing the Bohr radius and therefore decreasing the effective distance \( d/a_0 \) (even though \( d/a \) remains fixed). As only the kinetic and attractive terms of the Hamiltonian are present, there is no interaction between the two fermions, and the resulting eigenstate corresponds to two independent ground-state electrons associated with \( ^{3}\text{P}_0 \).

**Measurement.** From the chemistry perspective, all relevant quantities can be expressed in terms of the fermionic density. This is the approach used, for example, in density functional theory methods\(^{37}\). The fermionic density is obtained by using the Green’s functions and reconstruct the fermionic density. Although this is very complex in practice, it could be achieved with gas microscopy techniques (see ref. 38, for instance).

An alternative approach would be to measure the energy of the system. In addition to constructing molecular potentials, scanning the energy at different nuclear configurations can provide additional information, such as the magnitude of the molecular forces (Hellmann–Feynman theorem\(^{39}\)). For this, three quantities need to be close to the \( ^{3}\text{P}_0 \rightarrow 1S_0 \) resonance without affecting the other states. Two-photon Raman transitions can then couple levels \( ^{3}\text{P}_2 \) and \( ^{3}\text{P}_0 \) appropriately, or one can also use a two-photon transition with level \( 1S_0 \). We also note that having nuclear spin 0 is not a problem in this case, because the mediated electronic repulsion is spin-independent. By choosing an isotope of Sr, an optical lattice with the same spacing can be engineered at the bosonic state \( ^{3}\text{S}_0 \), similarly to the fermionic atoms. Furthermore, the lattice depth and spacing in \( ^{3}\text{P}_2 \) can be independently controlled (for example by using magic wavelengths\(^{31}\)) and its scattering length can be tuned (for example, using non-resonant light\(^{34}\)).
to be measured simultaneously: the expected values of the kinetic energy, \( (K) \), the nuclear attraction, \( \langle V_n \rangle \), and the electronic repulsion, \( \langle V_e \rangle \), such that the total energy is \( E = (K) + \langle V_n \rangle + \langle V_e \rangle \). By using sudden quenches of the Hamiltonian, such contributions can be independently converted into kinetic energy. One can then perform a time-of-flight measurement of the fermionic atoms expelled from the lattice using, for example, ionization or fluorescence techniques. As one can observe, the measured quantities will not correspond to eigenstates of the original Hamiltonian, which will introduce a variance proportional to the number of fermions. One could then repeat this procedure to gain statistical significance. Once the equilibrium point of the molecular potential is identified, the procedure can be greatly simplified, because only the \( (K) \) measurement is needed to read the total energy at that point, according to the virial theorem for molecules\(^{39}\).

**Experimental considerations.** A reliable simulation of the quantum chemistry Hamiltonian requires that our simulator, described by equation (6), satisfies inequalities (5), (8)–(11). We are, however, aware that there will be other experimental imperfections that may impose extra conditions and that will have to be analysed in detail to optimize the performance of the simulation. The most relevant ones are:

1. Finite temperature leads to thermal fluctuations, which may spoil the simulation by populating undesirable higher-energy bands. Thus, these fluctuations will lead to defects in the Mott insulator (see below) and may also influence the internal states of the atoms. The latter influence, however, can be very well controlled in atomic systems, given that we only need the atoms to be initially in a polarized state, which is reasonably easy to prepare.

2. Dephasing can be initiated by fluctuations in the transitions or by magnetic fields (as internal levels are being used). This would limit the potential of the system as a quantum simulator. However, the first effect is small in the case of microwave or Raman transitions, and the second one can be controlled under the conditions used for condensed matter simulations\(^{9,10}\).

3. Inexact fermionic filling. Because fermions play the role of electrons, an inexact number of fermionic atoms hopping in the lattice translates into an erroneous effective charge in the simulated molecule. These errors can be post-selected by measuring the number of fermionic atoms after the simulation is performed.

4. Defects in the Mott insulator. The absence of Mott particles in a given lattice site will locally modify the effective fermion potential. Fermions hopping to this site cannot mediate its repulsive interaction through spin excitations, perturbing the simulated molecular orbital around this position. Importantly, such defects will not affect the potential far from the fermion so that the final performance of the simulation will scale with the density of defects instead of with their number.

5. Spatial inhomogeneities in cavity coupling. In the simulator Hamiltonian of equation (6), we assumed that the \( j \) excitations couple homogeneously to the cavity mode. In general, there might be some inhomogeneities that translate into a Hamiltonian:

\[
\frac{J}{N_W} \sum_{ij} f_\text{ij} a'_i a'_j
\]

The fluctuations of \( f_\text{ij} \) around 1 will induce an extra decoherence time of \( \tau_{\text{inh}} \), which must also be smaller than our simulator parameters. In state-of-the-art experiments, optical cavities at wavelengths of 780 nm and with a beam waist of around 60 µm are already available\(^{40}\), which would roughly allow 50–100 local minima of the standing wave to sit in a homogeneous region.

6. Cavity and atom losses. Even though the cavity-mediated interactions are mediated by a virtual population of photons, the cavity decay introduces extra decoherence into the system due to the emission of these virtual photons. Moreover, the atomic excited states, which are also virtually populated, may decay to free space, introducing losses. Thus, the cooperativity of the cavity QED system must be large to avoid both type of losses.

(7) Three-body losses. Because we have fermions and there can be at most one atom per lattice site, this type of loss should be small.

Therefore, most of the possible errors of the simulation are either already under control in current experiments\(^{6,10}\) or depend only on the number of defects.

**Code and data availability**

The computer code developed for this study is available from the corresponding authors upon reasonable request. All data supporting the findings of this study can be generated using the numerical methods described within Methods and Supplementary Information and are available upon reasonable request.

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Extended Data Fig. 1 | Results of the G-S algorithm.  

a. We apply the G-S algorithm to identify the phase mask associated with a holographic 3D Coulomb potential on a lattice of $N^3$ sites. Fixing the origin at the central site, we choose the nucleus position as $\mathbf{n} = [(2n_{\text{div}})^{-1}, 0, 0]$ (the first coordinate is shifted so that the lattice induces a natural cutoff). 

b. An axial central cut of the potential (yellow markers) in direction $z$ (see aligned set of sites in b; red), created by a phase mask composed of $(n_{\text{div}}N) \times (n_{\text{div}}N)$ cells for $n_{\text{div}} = 3$ (see inset), compared to the objective Coulomb potential (blue solid line). In step (ii) of the algorithm, the Ewald sphere is discretized using a parallel projection, as in ref. 31. The field is initiated with random phases. Parameters: $N = 30$ and 7,000 iterations of the G-S algorithm. 

b. Location of the axial cut shown in a.
Extended Data Fig. 2 | Numerical simulation of the adiabatic preparation of the ground state of $H_2$ with the simulator (particularized for a 2D lattice). Red dashed lines follow the adiabatic evolution of the initial state $|\psi_0\rangle$ and arrows point to the direction of evolution.

a, Preparation of the bosonic state through steps I(a)–I(c) (see Methods and Extended Data Table 1). Continuous lines indicate the exact energy of the two lowest energy states. For the adiabatic evolution we use Trotterized $\Delta t = 0.5$ and evolution with $|\Delta U|/(U^2 \Delta t) = 3 \times 10^{-4}$. b, Steps II–III of the preparation of the fermionic state. In the simulation, we use the Trotterized time evolution in intervals of $\Delta tV_0 = 0.05$. In step II, the kinetic term is adiabatically introduced in steps of $\Delta tV/(V_0^2 \Delta t) = 0.005$. In step III, the electronic repulsion is tuned up as $\Delta tV/(V_0^2 \Delta t) = 0.02$. Here yellow (blue) continuous lines follow the exact energy levels of $H_{qc}$, as calculated by imaginary-time evolution with (without) the effect of electronic repulsion. The top insets show the Mott excitation (a) fermionic population (b) in the lattice at the times indicated in the figure. The final point of the evolution shown in b corresponds to $d/a_0 = 1$. Parameters: $N = 60$, $U/J_c = 1.5$, $d/a_0 = 10$. 
Extended Data Table 1 | Evolution of the main parameters of the system during adiabatic preparation with steps I–III presented in Methods

| Stage | On-site interaction | Attractive potential | Fermionic hopping | Raman coupling |
|-------|---------------------|----------------------|-------------------|---------------|
| I(a)  | $U \rightarrow 0$   | 0                    | 0                 | 0             |
| I(b)  | 0                   | $V_0$                | 0                 | 0             |
| I(c)  | 0 $\rightarrow U$   | $V_0$                | 0                 | 0             |
| II    | $U$                 | $V_0$                | 0 $\rightarrow t_F$ | 0             |
| III   | $U$                 | $V_0$                | $t_F$             | 0 $\rightarrow g$ |

To simplify the preparation of $|\psi\rangle_1$, step I (illustrated in Extended Data Fig. 2) has been divided into three consecutive substeps: (a): starting from state $|\psi_0\rangle$, the on-site interaction $U \rightarrow 0$ is adiabatically cancelled; this brings the Mott excitation into a delocalized single excitation shared by all the atoms in the insulator, $\sum_j |n_j^0\rangle N^{3/2}$.  (b): as discussed in Methods, one can use a dynamic double-well potential to move the fermionic wavepackets in opposite directions. We note that because fermionic and bosonic species are decoupled at this point ($U \rightarrow 0$), the Mott excitation will remain in the symmetric state reached in (a). (c): we adiabatically restore the on-site repulsion $U \rightarrow 0$; the bosonic state then evolves to a superposition localized at the new position of the fermionic atoms, $(a_0 + a_n)|\psi_0\rangle/\sqrt{2}$. 