Quantum entanglement is one of the key ingredients that fundamentally distinguishes quantum mechanics from classical mechanics (1). It is considered an essential resource for quantum information processing but remains generally challenging to create experimentally. One mechanism of realizing quantum entanglement between particles is using the electric dipole-dipole interaction, as demonstrated in systems such as Rydberg atoms (2) and silicon quantum dots (3). Polar molecules possess permanent, long-range, and spatially anisotropic electric dipoles whose interaction could be harnessed for high-fidelity entanglement generation. Therefore, they have been proposed as a powerful platform for realizing quantum simulations of strongly interacting many-body dynamics (4–7) and for scalable quantum computing (8–12). Ultracold molecules can be produced through either direct laser cooling (13–17), assembly of individual ultracold atoms (18–26), or optoelectrical Sisyphus cooling (25, 26). Current experimental progress has established the capability of preparing and manipulating ultracold molecules with high fidelity (27–33). In particular, reconfigurable tweezer arrays offer an attractive quantum platform owing to their scalability and potential for single-site addressability (24, 34–39). Toward this aim, molecules have been demonstrated to have long qubit (rotational and hyperfine state) coherence times, which are substantially longer than the predicted molecule-molecule dipolar gate times (28, 31, 39, 40). In this work, we report the entanglement of pairs of molecules—the critical ingredient in quantum computing and simulation based on molecules—by leveraging long molecular coherence and intrinsic dipolar interactions with individual particle addressability.

A key step toward using ultracold polar molecules for quantum simulations and multiparticle quantum gates is the generation of coherent dipole-dipole couplings between molecules (12, 41), which has been shown in sparsely filled three-dimensional (3D) lattices (27) and 2D layers (42) and in a molecular quantum gas microscope (39). Here, we used resonant electric dipole-dipole coupling for coherent exchange of rotational angular momentum between pairs of laser-cooled CaF molecules individually trapped in optical tweezers. With the ability to tune the angle of the molecular quantization axis in the lab frame, our system realizes both ferromagnetic and antiferromagnetic couplings in a quantum XY spin-exchange model by effectively encoding a spin-1/2 system into molecular rotational states. Using this spin-exchange Hamiltonian, we performed iSWAP two-qubit gate operations. An iSWAP gate, when sequentially combined with single-qubit operations, can generate bipartite entanglement deterministically and form a universal set of quantum gates, which is the essential resource for all quantum information applications (12, 43). We found that the fidelity of generated Bell states is presently limited by the thermal motion of molecules within the tweezer traps. Finally, we demonstrate the use of an interleaved dual tweezer-array system, which allows for robust single-site addressability and fast gate operations between molecules. This, combined with expected improved molecular cooling (44), has the potential to greatly increase the fidelity of two-qubit operations in this system. This work is parallel and concurrent with that of another study (45).

**Initial-state preparation of CaF molecules in a 1D optical tweezer array**

We began the experiment by loading CaF molecules from a cryogenic buffer gas beam source (46) into a radio-frequency magneto-optical trap (46). Next, we loaded the molecules into a 1D optical lattice using A-enhanced gray molasses
cooling on the electronic $X^2Σ^+→A^2Π_{1/2}$ transition (47, 48) and optically transported them into a glass cell using a focus-tunable moving lattice, as characterized previously (49).

We used a microscope objective with 0.6 numerical aperture to project a 1D optical tweezer array in the glass cell, as well as collect fluorescence from the molecules during A-imaging (47). The array is formed by passing single-frequency 776-nm laser light through a shear-mode acousto-optic deflector (AOD) driven with a multitone waveform (37), which is generated by a high-speed arbitrary waveform generator (AWG). This allows for control of the position of individual optical tweezer sites as well as trap depth. We started with a 20-site array formed by a single AOD (Fig. 1A).

We loaded the molecules into the tweezers by overlapping the array with a transported molecular cloud that is held in an optical dipole trap in the presence of the $A$-cooling light. We observed an average probability of 35% for loading a single site with a single molecule. After loading the array, we applied a $A$-imaging pulse and collected fluorescence onto a camera to identify the tweezer sites that are loaded with single molecules in the ground electronic and vibrational state (or empty) with a detection fidelity of 98(1)% and nondestructive detection fidelity of 94(1)% (37, 47). Because the $A$-cooling technique relies on closed photon cycling between the $X, N = 1$ and $A, J = 1/2$ manifolds ($N$ is the rotational angular momentum, and $J$ is the total angular momentum, excluding nuclear spin), only molecules in the $X, N = 1$ rotational manifold can be loaded into the tweezers and detected in this phase of the experiment.

Imaging distributes the population of molecules over all 12 hyperfine states. To prepare the molecules in a single quantum state, we used a combination of optical pumping and microwave transfer. Using short $X→A$ laser pulses resonant with all hyperfine levels in the $X$, $v = 0, N = 1$ manifold except the $|X, N = 1, F = 0, m_F = 0⟩$ state ($\nu$ is the vibrational quantum number; $F$ is the total angular momentum, including nuclear spin; $m_F$ is the magnetic quantum number of $F$), we pumped most of the molecular population into the latter state. We then linearly ramped down the trap depth of the tweezers in 5 ms and applied a bias magnetic field of $3.2$ G. A subsequent microwave $\pi$-pulse transfers the population from $|X, N = 1, F = 0, m_F = 0⟩$ to the $|X, N = 0, F = 1, m_F = 0⟩$ state (Fig. 1B). Finally, we applied an $X→A$ laser pulse that contains frequencies to drive all the hyperfine components in order to remove any molecular population left over in the $X, N = 1$ manifold. In the end, all the remaining molecules in the array were in the $|X, N = 0, F = 1, m_F = 0⟩$ state, initializing the qubits and effectively encoding a spin-$1/2$ model in the subspace spanned by the $|X, N = 0, F = 1, m_F = 0⟩ = |↑⟩$ and $|X, N = 0, F = 1, m_F = 0⟩ = |↓⟩$ states (Fig. 1B). The total state preparation and detection efficiency is 60(2)%, which is limited by the residual population in other hyperfine states within $X, N = 1$ and imperfect imaging fidelity. This state preparation efficiency can be improved by an optimized optical pumping scheme (45).

**Single-molecule rotational coherence time**

To observe high-fidelity dipolar spin-exchange interactions, a long rotational coherence time comparable to the timescale of the dipolar interaction is required. With a sample of molecules at finite temperature in optical tweezers, it is important to control the differential ac Stark shift broadening caused by the molecule’s thermal motion in the tweezer trap (28, 50–52). To suppress this broadening, the tweezer laser is linearly polarized at a “magic” angle relative to the quantization axis that is defined by the applied bias magnetic field, as detailed previously (31) (Fig. 1A). To maximize the single-qubit decoupling sequences. The coherence time is fitted as a Gaussian $1/e$ decay time constant.

**Fig. 2. Rotational coherence and dependence of dipolar interactions on tweezer spacing. (A) The Ramsey sequence with XY8 dynamical decoupling used in this work. $X_{\pi/2}$ represents a $\pi$-pulse, which rotates the quantum state around the $x$ axis of the Bloch sphere by 90°. Similarly, $X_Y$ ($Y_x$) represents a 180° rotation around the $x$ axis ($y$ axis) on the Bloch sphere. At the top, “$X^N$” means that the block of XY8 pulses is repeated multiple times during the evolution time. (B) Measured single-molecule rotational coherence time between $|↑⟩$ ($N = 1$) and $|↓⟩$ ($N = 0$) using Ramsey, single-$\pi$-pulse spin echo, and the XY8 dynamical detuning method to improve the coherence time. (C) Dipolar spin-exchange oscillation at various tweezer spacings, with fitted decay-time constant and oscillation period shown in the legends. (D) Dipolar spin-exchange interaction strength $J$ versus the tweezer spacing $|R|$. The dashed orange line is the theoretical prediction of $J$ at zero temperature. The solid blue line is the simulated result of $J$ with the thermal motion of the molecules taken into account (59). In (B) and (C), error bars represent one standard deviation of uncertainty.
coherence time, it is beneficial to adiabatically lower the trap depth as much as possible without unduly spilling molecules from the trap. However, this is not optimal for maximizing the number of observed dipolar spin-exchange oscillation cycles. At finite temperature, the instantaneous dipolar interaction strength fluctuates owing to thermal motion, which becomes more prominent in a shallow trap. This is the main mechanism of dephasing. To mitigate this issue, we confined the molecules more tightly by operating the tweezer at a higher trap light intensity for which the magic angle is close to 90°. The work described in this manuscript was performed under these conditions.

We measured the rotational qubit coherence time under conditions of three different microwave pulse sequences used to drive transitions between |↑⟩ and |↓⟩ and observed the population in |↑⟩. With a Ramsey sequence, we observed a single-qubit coherence time of \( \tau_c = 3.6(6) \) ms. By adding a single π spin-echo pulse, \( \tau_c \) is extended to 33(5) ms. We used active magnetic field cancellation to remove long-term drift on the order of 10 mG, but the system we used does not remove magnetic field fluctuation within a power line cycle time of less than 1/60 Hz. This limits the effective interval between spin-echo pulses to an integer multiple of 60 Hz, making it difficult to measure fast dipolar oscillations with only a single π-pulse. Instead, we used dynamical decoupling schemes to preserve the qubit coherence. This technique is used in a variety of quantum information systems (53, 54), including molecular systems (27, 30, 45). We chose the XY8 dynamical decoupling sequence (Fig. 2A) with a cycle length of 1.6 ms (55), which is much shorter than 1/60 Hz, and achieved a coherence time of \( \tau_c = 630(90) \) ms. This dynamical decoupling was used for all of our measurements except where noted. Figure 2B shows the measured contrast versus time for single-particle oscillations between |↑⟩ and |↓⟩, from which \( \tau_c \) is determined.

**Coherent dipolar spin-exchange interaction**

The dipolar spin-exchange interaction Hamiltonian (40) is

\[
H_{dp} = \frac{J}{2} \left( S_1^+ S_2^- + S_1^- S_2^+ \right) = J \left( S_1^x S_2^x + S_1^y S_2^y \right)
\]

where, respectively, \( S_i^x, S_i^y, S_i^z \) is the spin-\( i \) raising (lowering, Pauli-X, Pauli-Y) operator for molecule number \( i \) in a tweezer pair. \( J \) is the dipolar interaction strength that can be further expressed as

\[
J = \frac{d^2}{4\pi\epsilon_0 r^3} (1 - 3\cos^2 \theta)
\]

where \( d \) is the transition dipole moment between the |↑⟩ and |↓⟩ state (\( d = 1 \) Debye), \( \epsilon_0 \) is the vacuum permittivity, \( r \) is the intermolecular spacing, and \( \theta \) is the angle between the quantization axis and the intermolecular axis direction. Apart from molecular systems (27, 30, 33), this XY spin Hamiltonian was previously studied with Rydberg atoms in optical tweezers (56) and atoms in optical lattices (57, 58).

We set the bias magnetic field perpendicular to both the \( k \)-vector of the tweezer light and the direction of the 1D tweezer array. The polarization of the linearly polarized tweezer light is rotated to the tweezer array direction (i.e., a magic angle close to 90°). This configuration provides the largest “magic trap depth” (the tightest confinement) at a given magnetic field.

We first prepared pairs of tweezers spaced at \( R \approx 5 \mu m \), with each site loaded with a single molecule prepared in the |↓⟩ state (or empty), yielding the |↓↓⟩ state. At this separation, the dipolar interaction strength \( J \) is negligible (\( J/\hbar < 3 \) Hz, where \( \hbar \) is Planck’s constant). In a time period of -1 ms, we then moved the even numbered sites toward the odd numbered sites by sweeping the AOD frequency tones of the even sites, which reduces the separation and has the effect of increasing the dipolar interaction strength. We then applied a \( \frac{\pi}{2} \)-pulse to prepare both molecules in the superposition state \((|\uparrow\rangle + |\downarrow\rangle)/\sqrt{2} \). Then, we applied XY8 dynamical decoupling sequence of microwave pulses. Under the time evolution of dipolar spin-exchange Hamiltonian (\( H_{dp} \)), a relative phase accumulated between \((|\uparrow\rangle + |\downarrow\rangle)/\sqrt{2} \) and \((|\uparrow\rangle + |\downarrow\rangle)/\sqrt{2} \). (Because the XY8 sequence only contains \( \pi \)-pulses, it will not affect the phase accumulation during the evolution under \( H_{dp} \)). After a wait time, we applied another \( \frac{\pi}{2} \)-pulse and then moved the molecules apart. To read out the final qubit state, we used a second \( \Lambda \)-imaging pulse to project the system to |↑⟩. We selected the data where both sites in a tweezer pair are initially loaded with single molecules. Our measurement yielded the probability \( P_1 \), of detecting both molecules in the |↑⟩ state. To summarize, starting from the initial state |↓⟩, a microwave pulse sequence creates a final state that evolves in time as \( \psi(t) = \frac{1}{2} \left( 1 - e^{-\frac{\tau}{T}} \right) |\downarrow\rangle - \frac{1}{2} \left( 1 + e^{-\frac{\tau}{T}} \right) |\uparrow\rangle \), resulting in the probability \( P_1 = \cos^2 \frac{\Delta}{T} = \frac{1}{2} (1 + \cos \frac{\Delta}{T}) \), which oscillates at an angular frequency of \( \omega_f = \frac{\Delta}{T} \), where \( \hbar \) is reduced Planck’s constant.

At smaller tweezer spacings, we observed increased \( \omega_f \) because of the stronger dipolar interaction between the two molecules (Fig. 2C). By fitting the data to an exponentially decaying sinusoidal model, we extracted the dipolar oscillation cycle period \( T \) and contrast decay time constant \( \tau_T \). The dipolar spin-exchange strength \( J \) at different tweezer spacings can then be calculated from \( T \) (Fig. 2D). We found that the measured \( J \) is slightly smaller than the theoretical prediction and deviates more as the spacing decreases. This can be explained by the finite temperature of the molecules causing the effective intermolecular spacing \( |R| \) to be larger than the tweezer spacing \( |\hat{R}| \). We used Monte Carlo simulations to describe the behavior of the molecules in the tweezer, including the thermal motion of the molecules, and show in Fig. 2D that the simulated results agree with the experimental data. Additionally, the simulation captures that the thermal motion reduces the observed number of coherent dipolar oscillations as tweezer spacing is decreased (59).

**Anisotropy of the dipolar interaction**

The general dipole-dipole Hamiltonian described in Eqs. 1 and 2 is inherently anisotropic owing to the \( \theta \)-dependent term. We studied the effect of the anisotropy of spin exchange experimentally by varying the angle \( \theta \) and measuring the corresponding dipolar interaction strength. The angle \( \theta \) is varied by rotating the quantization axis relative to the line between the centers of the tweezers. The quantization axis is set by...
the applied bias magnetic field and can be changed by tuning the current through two pairs of magnetic field coils while simultaneously rotating the tweezer light polarization to maintain the same magic angle.

In Fig. 3, we show $P_{\uparrow\downarrow}$ at three characteristic angles $\theta = 0^\circ$, $54.7^\circ$, and $90^\circ$, all taken with a $|R| = 2 \mu$m tweezer spacing. We observed dipolar spin-exchange oscillations in both the $\theta = 0^\circ$ and $\theta = 90^\circ$ configurations, with the oscillation at $\theta = 0^\circ$ being at twice the frequency as that at $\theta = 90^\circ$. No clear oscillation is observed for the $\theta = 54.7^\circ$ configuration, which was as expected because the dipolar interaction averages to zero at this angle. These results agree with the absolute value of the magnitude of the anisotropic term in each configuration. We also observed a larger number of oscillation cycles at $\theta = 90^\circ$ than at $\theta = 0^\circ$. Our Monte Carlo simulation indicated that thermal motion dephasing is the dominant cause. At $\theta = 0^\circ$, the larger motional wave function spread in the more weakly trapped axial direction of the optical tweezers results in a large fluctuation of the instantaneous value of $\theta$. The $\theta = 90^\circ$ configuration is less affected because of the tight confinement of the optical tweezers in the radial direction.

**Fidelity of created Bell states**

Dipolar spin-exchange can be used in a two-qubit iSWAP gate to generate entanglement between molecules in neighboring tweezers (22). At a dipolar spin-exchange interaction time of $t = T/4 = 19.2$ ms, where $T$ is the time the molecular pair stays in the entanglement phase, the system has evolved into a maximally entangled state known as a Bell state. To test the fidelity of the Bell state that was generated in our system, we applied a third $\pi$-pulse (see Fig. 4 legend) around a variable rotation axis on the Bloch sphere (angled $\phi$ relative to the $x$ axis on the equatorial plane) (Fig. 4A). By varying $\phi$ and measuring the survival probability of all four possible final-state outcomes, one can construct the parity quantity $\Pi$ (43, 60):

$$\Pi = \langle \Pi \rangle = 8^{\frac{1}{4}} \sum_{i=1}^{4} S_i \tilde{S}_i \sum_{j=1}^{4} S_j \tilde{S}_j$$

where $\Pi$ is determined to be $\Theta = 0^\circ$: $\Pi = 0$, $\Theta = 90^\circ$: $\Pi = 1$. Here, $\langle \rangle$ denotes averaging over all occurrences where both sites in a tweezer pair are initially loaded with single molecules, and $S_i \tilde{S}_j$ represents the Pauli-Z operator on the number $i$ molecule in a pair.

Starting from a Bell state, this sequence will result in a $4\pi$ oscillation in $\Pi$ as $\phi$ is varied from 0 to $2\pi$ (61). In Fig. 4B, $\Pi$ is displayed for both the $\theta = 0^\circ$ and $\theta = 90^\circ$ configurations. Extracting the contrast of the oscillation for the $\theta = 90^\circ$ case, we measured a Bell state fidelity of $F = 0.32(2)$ and a state preparation and measurement (SPAM)-corrected fidelity of $F_{SPAM} = 0.89(7)$. The phase of the parity oscillation reveals the sign of the anisotropic term ($1 - 3\cos^2 \theta$), also seen previously (65).

Our data show that the $\theta = 0^\circ$ configuration leads to a negative $J$, which corresponds to a ferromagnetic interaction, and the $\theta = 90^\circ$ configuration leads to a positive $J$, which corresponds to an antiferromagnetic interaction.

$F_{SPAM}$ is measured to be significantly higher than $F$, with imperfect state preparation and detection being the surmised cause. In detail, during initial-state preparation, molecules that fail to be prepared in the desired $|N = 0, F = 1, m_F = 0\rangle$ are intentionally removed by a resonant laser pulse, resulting in an empty trap or traps. During the final readout, if the molecule is in the $|\uparrow\downarrow\rangle$ state or the trap is simply empty, both would appear dark. We used a $\pi$-pulse and additional imaging pulse (Fig. 4C) to distinguish between these two cases. If the molecule is in the $|\uparrow\downarrow\rangle$ state, it will be transferred to the $|\uparrow\downarrow\rangle$ state and detected, whereas an empty trap will remain dark. This information is used to exclude the cases of empty traps during the final readout, which improves the contrast of the parity oscillation (Fig. 4D). The resulting Bell state fidelity corrected for measurement error is determined to be $F_{ex} = 0.89(6)$, which is higher than the threshold of $F_{TH} = 0.5$ (60), showing, under these conditions, the conditional preparation of entanglement of two molecules in a tweezer pair. The evolution of the system to create a Bell state is an iSWAP operation, and, although we have not fully characterized the system as an iSWAP gate, the Bell state fidelity indicates how it would perform.

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**Fig. 4. Parity measurements showing the creation of Bell state pairs.**

**A** The microwave pulse and detection sequence used in the parity oscillation measurement. The XY8 block is the same as that in Fig. 2A. $\phi_{n/2}$ denotes a $\frac{\pi}{2}$-pulse with a microwave phase shifted by $\phi$ relative to the first $X_{n/2}$ pulse in the sequence, effectively rotating the quantum state around an axis that is angled $\phi$ relative to the $x$ axis on the Bloch sphere. The pulse in yellow with a camera symbol represents imaging of the molecules. Note that the first imaging pulse for tweezer-loading identification, as well as the microwave pulses for initial-state preparation, are applied before this sequence and not shown in this figure. **B** Parity oscillation at $\theta = 0^\circ$ and $\theta = 90^\circ$. $A$ is the fitted parity oscillation amplitude. **C** With the addition of a $\pi$-pulse and a third imaging step to the sequence shown in (A), molecules are verified to be present throughout the entanglement and readout process. **D** Parity oscillation for $\theta = 90^\circ$, with empty traps excluded and corrected for measurement error, using the sequence depicted in (C). In (B) and (D), error bars represent one standard deviation of uncertainty.
Toward arbitrary initial-state preparation
Motivated by the desire to perform robust single-site addressing, instead of using a single AOD to generate all sites in the array, we switched to using one AOD to generate the odd numbered sites and another AOD to generate the even numbered sites. This allowed for convenient independent trap-depth control over each molecule in a pair and uniformity across the array. Additionally, by offsetting the frequency of the tweezer light of the even and odd sites, molecules can be moved in close proximity without experiencing the heating that can arise in a single AOD system (34, 38). For a given trap depth, the differential as Stark shift results in molecules in even numbered sites being away from the resonance of the |↑↓⟩−|↓↑⟩ microwave transition, therefore allowing separate microwave addressing of the odd sites. By applying a microwave π-pulse when odd sites are detuned away, we could prepare an antiferromagnetic initial state |↑⟩. Under $H_{\text{dip}}$, an initial state |↑⟩ evolves as $|ψ(t)⟩ = \cos \frac{\pi}{2} |↑⟩ - i \sin \frac{\pi}{2} |↓⟩$, and $P_\uparrow = \cos^2 \frac{\pi}{2} = \frac{1}{2} (1 + \cos \frac{\pi}{2})$ will thus oscillate at an angular frequency of $\frac{\pi}{2}$.

To demonstrate individual addressing, we first prepared the molecules (both in the even and odd sites) in initial state |↓⟩. We then adiabatically ramped the trap depth of the odd sites to seven times that of the even sites, so as to detune the transition of the molecules in the odd sites out of resonance. The microwave π-pulse then only transfers the molecules in the even sites from |↓⟩ to |↑⟩. This creates an antiferromagnetic state |↓↑⟩. By then applying a microwave pulse with variable length of time and detecting molecules in the |↑⟩ state, we observed Rabi oscillations in both even and odd sites with opposite phase (Fig. 5A). To observe dipolar spin exchange, we moved the tweezers to a smaller spacing ($|\vec{R}| = 2 \mu m$). As with the single AOD system, we applied the XY8 dynamical decoupling pulses and then separated the pairs for detection. The resultant outcome probabilities are shown in Fig. 5B, with a clear display of spin exchange.

Conclusions and outlook
We observed dipolar spin-exchange interactions and created Bell-state entangled pairs with single CaF molecules trapped in optical tweezers. We studied the dipolar interaction and entanglement by tuning the spacing of the optical tweezers and the angle of the electric dipole quantization axis. By applying detection at the end of the entanglement sequence to include only cases where two molecules are present, we determined a Bell state fidelity of $F_{\text{ex}} = 0.89(6)$ through a parity oscillation measurement. Parity measurements confirm that the interaction in this system can be tuned between ferromagnetic and antiferromagnetic.

The coherence time of dipolar interactions and the single-molecule rotational coherence time are both limited by the finite temperature of the molecules. Implementation of further cooling using other techniques, for example, Raman sideband cooling (44), would substantially reduce motional dephasing, extend the single-qubit rotational coherence time (37), and thus increase the two-qubit gate fidelity (2). The approach presented in this work can be extended to ultracold polyatomic molecules, which have robust parity doublet states that give rise to an advantageous Stark level structure (62, 63).

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**SUPPLEMENTARY MATERIALS**

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Supplementary Text

Figs. S1 to S3

Reference (66)

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