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Figs. S1 to S19
Tables S1 to S3

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# REPORTS

## Toward Heisenberg-Limited Spectroscopy with Multiparticle Entangled States

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**The precision in spectroscopy of any quantum system is fundamentally limited by the Heisenberg uncertainty relation for energy and time. For *N* systems, this limit requires that they be in a quantum-mechanically entangled state. We describe a scalable method of spectroscopy that can potentially take full advantage of entanglement to reach the Heisenberg limit and has the practical advantage that the spectroscopic information is transferred to states with optimal protection against readout noise. We demonstrate our method experimentally with three beryllium ions. The spectroscopic sensitivity attained is 1.45(2) times as high as that of a perfect experiment with three nonentangled particles.**

Quantum-mechanical entanglement in an ensemble of particles can enhance the signal-to-noise ratio in spectroscopy to a level that is fundamentally unattainable with non-entangled particles. The enhancement is ultimately limited by the Heisenberg uncertainty relation between energy and time. Experiments that approach the Heisenberg limit have been rare because of the difficulty of producing suitable entangled states and because of the lack of robust observables that realize the limit for a large number of particles when the intermediate operations and the readout of the final state are imperfect. The method described here relies solely on collective interactions without the need to individually address the particles, and the final observable is the global state population of the ensemble. Therefore, neither exact knowledge of the number of particles in the ensemble nor individual particle access in preparation or readout is required to reach the Heisenberg limit.

A transition between any two quantum states is formally equivalent to a transition between the two states of a spin-$\frac{1}{2}$ system. We will label the eigenstates of the $z$-component of spin $\tilde{S}_z$ as $|\downarrow\rangle$ and $|\uparrow\rangle$ with $\tilde{S}_z|\downarrow\rangle = -\frac{1}{2}|\downarrow\rangle$ and $\tilde{S}_z|\uparrow\rangle = \frac{1}{2}|\uparrow\rangle$, where we choose units such that $\hbar = 1$ and assume that spectroscopy is performed on an ensemble of $N$ such systems with the use of Ramsey interferometry (*1*). For each experiment, the spins are initialized to the state $|\downarrow, N\rangle \equiv |\downarrow\rangle_1|\downarrow\rangle_2 \cdots |\downarrow\rangle_N = |J = N/2, J_z = -N/2\rangle$, where the last expression is written in terms of the $N$-spin Bloch vector representation, where $\vec{J} = \sum_{i=1}^{N}\vec{S}_i$. The first Ramsey pulse and free precession for duration $T$ produces a state (in a rotating reference frame) that contains phase factors $\phi = (\omega - \omega_0)T$, where $\omega$ characterizes the frequency of the applied field and $\omega_0$ is the resonant transition frequency between states $|\downarrow\rangle$ and $|\uparrow\rangle$. The second Ramsey pulse renders the phase information accessible in a global-state observable.

In traditional Ramsey spectroscopy with unentangled atoms (*1*), both pulses implement a rotation $\tilde{R}_x \equiv \exp[i\frac{\pi}{2}\tilde{J}_x]$ (a "$\pi/2$ pulse"). The readout determines the number of atoms $\tilde{N}_\downarrow = N/2 - \tilde{J}_z$ in the $|\downarrow\rangle$ state, where $\langle\tilde{J}_z\rangle = \frac{N}{2}\cos[(\omega - \omega_0)T]$. The final uncertainty $\Delta(J_z)_{\text{final}}$ in $\langle\tilde{J}_z\rangle$ yields a corresponding phase uncertainty

$$\Delta\phi = \Delta\{(\omega - \omega_0)T\} = 1/\sqrt{N} \quad (1)$$

which can be termed the standard quantum limit (*2*, *3*). Any method is ultimately limited to a phase uncertainty of $\Delta\phi = 1/N$, usually called the Heisenberg limit (*4*, *5*).

One way to increase phase sensitivity over Eq. 1 is through "spin squeezing" (*6*–*14*). Such squeezing can, for example, be realized by transferring squeezing from a mechanical (*3*, *7*) or field (*9*, *11*) mode to the spins. It has been shown that the initial spin state $|\downarrow, N\rangle$ could be squeezed by an interaction of the form $U_{\text{sqz}} = \exp[i\chi\tilde{J}_x^2]$ with a suitable coupling parameter $\chi$ (*8*). This operator can be implemented in atomic systems (*10*, *12*), and it was demonstrated experimentally on two spins (*13*). It can also be implemented with rotations and phase gates (*12*, *15*, *16*). Spin squeezing has also been realized recently in the context of quantum nondemolition measurements (*14*).

The above methods rely on measuring the orientation of the Bloch vector $\vec{J}$; however, other input states and measurements can also be used, such as initial states of the form $|J = N/2, J_z = 0\rangle$ and a variance measurement operator $\tilde{J}_z^2 - \langle\tilde{J}_z\rangle^2$ (*17*, *18*). Another method (*5*) uses an $N$-particle Greenberger Horne Zeilinger (GHZ) state (*19*, *20*)

$$\Psi_{\text{GHZ}} \equiv \frac{1}{\sqrt{2}}(|\downarrow, N\rangle + e^{i\zeta}|\uparrow, N\rangle)$$

produced by a "generalized Ramsey pulse," where $|\uparrow, N\rangle \equiv |J = N/2, J_z = +N/2\rangle$ and $\zeta$ is a phase factor. After free precession, the final pulse is a rotation $\tilde{R}_x$, applied to all particles, and the measured observable is the parity $\tilde{\Pi} \equiv \Pi_{i=1}^{N} 2(\tilde{S}_z)_i$. This protocol can reach the Heisenberg limit for any value of $(\omega - \omega_0)T$, but has the disadvantage that measurement of the parity is very difficult for large values of $N$ because it requires the ability to distinguish between odd and even numbers of particles in $|\downarrow\rangle$ in the presence of noise. Both protocols have been demonstrated experimentally on two spins (*13*).

The method described here combines ideas from precision spectroscopy and quantum information processing. The goal is to first encode a fiducial initial state into a state

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of maximal spectroscopic sensitivity that acquires the phase information in an optimal way. The state is then decoded to make the phase information accessible through a collective observable that is robust and simple to read out. The encoding and decoding can be accomplished with the same ensemble unitary transformations; their form is independent of the exact value of $N$ and depends instead on whether this number is odd or even (which can be straightforwardly determined). Upon readout, the final state will collapse to either $|\downarrow, N\rangle$ or $|\uparrow, N\rangle$, a two-state outcome that is maximally immune to detection noise.

In atomic systems, the initial state $|\downarrow, N\rangle$ can be prepared by optical pumping. The operator $U_N \equiv \exp[i[\frac{\pi}{2}]\,\tilde{J}_x^2]$ can produce $\Psi_{\mathrm{GHZ}}$, an entangled state with maximal phase sensitivity, if $N$ is even; if $N$ is odd, a further rotation completes the encoding $U_N \equiv \exp[i[\frac{\pi}{2}]\,\tilde{J}_x]\exp[i[\frac{\pi}{2}]\,\tilde{J}_x^2]$ (*10*). We can also show that

$$U_N = \frac{\exp\left(-i\,\frac{\pi}{4E}\right)}{\sqrt{2}}$$
$$(1+i^{N+E}\sigma_1\sigma_2\cdots\sigma_N) \qquad (2)$$

with $E = 1$ when $N$ is even and $E = 2$ when $N$ is odd. This transformation applied to the initial state yields (up to a global phase)

$$|\Psi_{\mathrm{GHZ}}\rangle = U_N|\downarrow, N\rangle = \frac{1}{\sqrt{2}}$$
$$(|\downarrow, N\rangle + i^{N+E}|\uparrow, N\rangle) \qquad (3)$$

After free precession for duration $T$,

$$|\Psi_{\mathrm{GHZ}}\rangle \rightarrow |\Psi_{prec}\rangle = \frac{1}{\sqrt{2}}\,(e^{-i\frac{N}{2}\phi}|\downarrow, N\rangle +$$
$$i^{N+E}e^{i\frac{N}{2}\phi}|\uparrow, N\rangle)$$
$$= \frac{1}{\sqrt{2}}\,\{\cos(\frac{N}{2}\phi)(|\downarrow, N\rangle +$$
$$i^{N+E}|\uparrow, N\rangle) - i\,\sin(\frac{N}{2}\phi)$$
$$(|\downarrow, N\rangle - i^{N+E}|\uparrow, N\rangle)\} \qquad (4)$$

The phase information is encoded in the phase difference of the two parts of the superposition and cannot be inferred directly by measuring probability amplitudes. The decoding step transfers this phase information into amplitude information,

$$|\Psi_{final}\rangle = U_N|\Psi_{prec}\rangle = -i\,\sin(\frac{N}{2}\phi)|\downarrow, N\rangle$$
$$+i^{N+E}\cos(\frac{N}{2}\phi)|\uparrow, N\rangle \qquad (5)$$

In the readout, this state will collapse to either $|\downarrow, N\rangle$, with probability $P_{(\downarrow, N)} = \frac{1}{2}(1 - \cos[N\,\phi])$ or $|\uparrow, N\rangle$, with probability $1 - P_{(\downarrow, N)}$. In the parlance of quantum information, this two-state outcome has the largest possible Hamming distance in the $N$-qubit measurement basis (*21*) and is therefore optimally protected against readout errors. For example, when detecting atoms by fluorescence scattering, one would typically use the electron-shelving technique (*22*), where the state $|\downarrow, N\rangle$ yields $N$ times the fluorescence rate of a single atom in $|\downarrow\rangle$, whereas $|\uparrow, N\rangle$ does not fluoresce. To accurately determine the probability $P_{(\downarrow, N)}(\phi)$, we can repeat the experimental sequence $M$ times and count the number of experiments $N_f(\phi)$ where fluorescence is detected. In the limit of many experiments $S_\downarrow(\phi) = N_f(\phi)/M$ converges toward $P_{(\downarrow, N)}(\phi)$.

Without added readout noise, the phase sensitivity is the same for any value of $\phi$ (*2*, *3*). However in a practical spectrometer where technical noise is present, we would sample $S_\downarrow(\phi)$ at two values $\phi = (\omega - \omega_0)T = \pm\pi/(2N)$, where the slope is maximum. By using electronic feedback to make $\langle S_\downarrow(+\pi/(2N))\rangle = \langle S_\downarrow(-\pi/(2N))\rangle$, we would force the mean applied frequency to be equal to $\omega_0$. The phase uncertainty for our method can be inferred by noting that the signal appears as that from a measurement on a single quantum system (we derived one collective two-state measurement from the ensemble) with a phase sensitivity $N(\omega - \omega_0)T$. The phase sensitivity is therefore increased $N$-fold compared with unentangled particles, but this gain is offset by the fact that we have a measurement of one quantum system versus $N$ systems in the case of uncorrelated atoms. The net effect is a phase uncertainty reduced by $\sqrt{N}$, yielding the Heisenberg limit $\Delta\{(\omega - \omega_0)T\} = 1/N$.

We demonstrated our method experimentally on three $^9Be^+$ ions confined in a linear Paul trap with an axial center-of-mass (COM) frequency of $\omega_{\mathrm{COM}}/(2\pi) = 3.86$ MHz and radial COM frequencies of about 11 MHz (*23*). The ions were cooled to the ground state of all three axial motional modes (*24*) and then initialized in the $|F = 2, m_F = -2\rangle \equiv |\downarrow\rangle$ hyperfine ground state by optical pumping ($|F = 1, m_F = -1\rangle \equiv |\uparrow\rangle$). The encoding step $U_3$ was realized with the use of two-photon stimulated Raman transitions for the rotations (*25*) and a phase gate $G$ (*16*). The phase gate was embedded in the first leg of a spin-echo sequence $\tilde{R}_y\tilde{I}_D\tilde{R}_y^2 G\,\tilde{R}_y$ where $\tilde{R}_y = \exp[i\frac{\pi}{2}\tilde{J}_y]$, $\tilde{I}_D$ is a delay to match the duration of $G$, and $\tilde{R}_y^2$ is the refocusing $\pi$ pulse. For the phase gate $G$, two laser beams with a relative detuning $\omega_{\mathrm{COM}} + \delta$ ($\delta << \omega_{\mathrm{COM}}$) exert an axial state-dependent optical dipole force on the three ions (*16*, *26*). The spacing of the ions was chosen such that the force was in phase on all three ions when they are in the same state. If this dipole force is applied for a duration $t_g = 2\pi/\delta$, the motion of the center of mass mode is excited and de-excited in such a way that each state combination on which a nonzero net force acts traverses a circle in phase space (*16*) and acquires a phase given by the area circumscribed in phase space, proportional to the square of the net force. In the experiment, the strength and detuning were adjusted to yield a phase of $2\pi$ on $|\downarrow, 3\rangle$ relative to $|\uparrow\downarrow\uparrow\rangle$ (*27*). The operation of the pulse sequence produced the GHZ state

$$|\Psi_{\mathrm{GHZ}}\rangle_3 = \tilde{R}_y\tilde{I}_D\tilde{R}_y^2 G\tilde{R}_y|\downarrow, 3\rangle$$
$$= U_3|\downarrow, 3\rangle = \frac{1}{\sqrt{2}}(|\downarrow, 3\rangle + i|\uparrow, 3\rangle) \qquad (6)$$

The total duration of the pulse sequence was 43 μs, with phase gate $G$ duration of 14 μs. The GHZ-state fidelity is determined in two steps (*20*): (i) The populations $p_{\downarrow 3}$ and $p_{\uparrow 3}$ are measured, and (ii) the coherence between the two components is determined by applying a $\pi/2$ pulse to all ions in state $|\Psi_{\mathrm{GHZ}}\rangle_3$ with a phase difference $\phi_1$ relative to the pulses used in the gate and determining the total parity $\langle\tilde{\Pi}\rangle = 2(p_{\downarrow\downarrow\uparrow} + p_{\downarrow\uparrow\downarrow} + p_{\uparrow\downarrow\downarrow} + p_{\uparrow 3}) - 1$ (*20*). The measured parity trace (Fig. 1) sinusoidally oscillates with period $2\pi/3$ as expected for a maximally correlated state. From the contrast (0.84(1)) and the measured populations ($p_{\downarrow 3} = 0.53(1)$, $p_{\uparrow 3} = 0.40(2)$) we determine the fidelity of the GHZ state to be $F_{\mathrm{GHZ}} = 0.89(3)$.

To complete the spectroscopy algorithm, a free precession period and a decoding pulse sequence are added after creation of $|\Psi_{\max}\rangle$. For our demonstration we replaced the precession phase by an offset phase $\phi_2$ in the $\pi/2$ pulses of the decoding sequence. The decoding operator $\tilde{R}_{\phi_2}G\tilde{R}_{\phi_2}^2\tilde{I}_D\tilde{R}_{\phi_2}$ was applied 2 μs after the encoding sequence and the total fluorescence of the three ions in a 400 μs detection period was recorded versus the offset phase $\phi_2$.

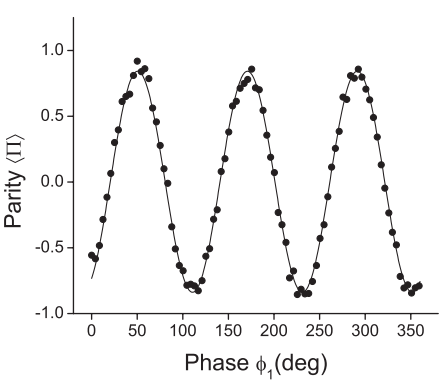


**Fig. 1.** Experimentally determined parity Π used to determine the fidelity of the three-particle GHZ state, plotted as a function of the phase difference $\phi_1$ of the analysis $\pi/2$ pulse. The solid line is a fitted sinusoid oscillating with $3\phi_1$ yielding a contrast of 0.84(1). deg, degrees.

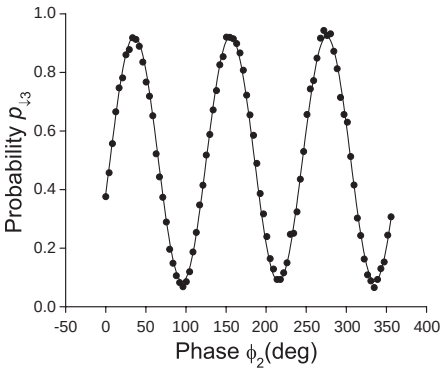


**Fig. 2.** Average probability of the three ions to be measured in $|\downarrow, 3\rangle$ as a function of the offset phase $\phi_2$. The fringes show a sinusoidal oscillation with period $2\pi/3$ and fringe contrast 0.84(1). deg, degrees.

During the detection period, we obtained on average 0.2 counts after preparing the state $|\uparrow, 3\rangle$ and 30 counts after preparing the state $|\downarrow, 3\rangle$. If the number of counts after a spectroscopy experiment was below 15, we assumed the outcome to be $|\uparrow, 3\rangle$; otherwise, we assumed the outcome to be $|\downarrow, 3\rangle$. The fringes resulting from averaging 1000 such experiments show a sinusoidal oscillation with period length $2\pi/3$ (Fig. 2). The fringe contrast (0.84(1)) yields a phase sensitivity that is $0.84 \times \sqrt{3} = 1.45(2)$ as high as that of a perfect Ramsey experiment with three unentangled particles. With perfect operations and readout, the method would yield a fringe contrast equal to 1 and thus reach the Heisenberg limit, giving an increase of phase sensitivity of $\sqrt{3}$ ($=\sqrt{N}$).

The gain obtained by using GHZ states can be offset by the corresponding faster decoherence if the free precession time is comparable to the dephasing time (*28*). However, the Ramsey time is often limited by other practical considerations, such as the desire to servo an oscillator to an atomic transition in a time that is much shorter than the decoherence time or by limitations imposed by the spectrum of the oscillator that drives the transitions (*29*, *30*). In those cases, the entanglement provided by generalized GHZ states gives the expected gain.

We prepared a GHZ state of three atomic ions with a fidelity of 0.89(3). We used this state to demonstrate a new method for precision spectroscopy on entangled particles that can reach the Heisenberg limit and is of practical use because of its immunity to errors in final state detection. Because only collective ensemble preparation and detection pulses are used, the extension of the method to larger numbers of particles ($N$) is straightforward. For large values of $N$, the required interaction proportional to $\hat{J}_x^2$ can be implemented with either the approach described in (*10*) or by further generalization of the phase gate because the operators $e^{i\chi\hat{J}_x^2}$ and $e^{i\chi\hat{J}_z^2}$ are equivalent up to a rotation applied uniformly on all ions. We want the phase-space-displacement dipole forces on all ions (in the same state) to be the same, which is the case in a one-dimensional (or two-dimensional) array of trapped ions, if the forces act on a center-of-mass mode whose motion is perpendicular to the ion string (or plane) (*31*). The readout is robust against noise because the observable is a two-state "all-on" versus "all-off" signal. Despite experimental imperfections, our demonstration of the method on three ions shows a gain of 45% over that of a perfect Ramsey experiment with non-entangled states.


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# Control and Measurement of Three-Qubit Entangled States


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**We report the deterministic creation of maximally entangled three-qubit states—specifically the Greenberger-Horne-Zeilinger (GHZ) state and the W state—with a trapped-ion quantum computer. We read out one of the qubits selectively and show how GHZ and W states are affected by this local measurement. Additionally, we demonstrate conditional operations controlled by the results from reading out one qubit. Tripartite entanglement is deterministically transformed into bipartite entanglement by local operations only. These operations are the measurement of one qubit of a GHZ state in a rotated basis and, conditioned on this measurement result, the application of single-qubit rotations.**


Quantum information processing rests on the ability to deliberately initialize, control, and manipulate a set of quantum bits (qubits) forming a quantum register (*1*). Carrying out an algorithm then consists of sequences of quantum gate operations that generate multipartite entangled states of this quantum register. Eventually, the outcome of the computation is obtained by measuring the state of the individual qubits. For the realization of some important algorithms such as quantum error correction (*1*–*5*) and teleportation (*6*), a subset of the quantum register must be read out selectively, and subsequent operations on other qubits must be conditioned on the measurement result. The capability of entangling a scalable quantum register is the key ingredient for quantum information processing as well as for many-party quantum communication. Whereas entanglement with two or more