Robust Quantum Enhanced Sensing via Antisymmetric Rabi Spectroscopy

Jiahao Huang\(^1\), Sijie Chen\(^2,3\), Min Zhuang\(^1\), and Chaohong Lee\(^1,2,3\)

\(^1\)College of Physics and Optoelectronic Engineering, Shenzhen University, Shenzhen 518060, China
\(^2\)Guangdong Provincial Key Laboratory of Quantum Engineering and Quantum Metrology,
School of Physics and Astronomy, Sun Yat-Sen University (Zhuhai Campus), Zhuhai 519082, China and
\(^3\)State Key Laboratory of Optoelectronic Materials and Technologies,
Sun Yat-Sen University (Guangzhou Campus), Guangzhou 510275, China

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Atomic spectroscopy, an essential tool for frequency estimation, is widely used in quantum sensing. A key for achieving quantum enhanced sensing is employing atom-atom interaction to generate entanglement. However, atom-atom interaction always induces collision shift, which brings systematic error in determining the resonance frequency. Contradiction between utilizing entanglement and suppressing collision shift generally exists in atomic spectroscopy. Here, we propose an antisymmetric Rabi spectroscopy protocol without collision shift for arbitrary atom-atom interactions. We analytically find that, the antisymmetric point, which can be used for determining the resonance frequency, has no collision shift. By employing atom-atom interaction, the spectrum resolution can be dramatically improved and the measurement precision may beat the standard quantum limit. Moreover, unlike the quantum-enhanced Ramsey interferometry via spin squeezing, our scheme is robust against detection noises. Our antisymmetric Rabi spectroscopy protocol has promising applications in various practical quantum sensors such as atomic clocks and atomic magnetometers.

Introduction. – Atomic spectroscopy, the basis for precise measurement of transition frequencies, has been extensively applied in fundamental sciences as well as high-precision sensors [1]. Most quantum sensors employ a pair of discrete energy levels relevant to the physical quantity to be measured. By using atomic spectroscopy to determine the transition frequency, one can estimate the physical quantity to be measured [2] and build various practical quantum sensors such as atomic clocks [3–5], magnetometers [6–8], gyroscopes [9] and gravimeters [10].

On the one hand, atom-atom interaction can be used to generate the desired entanglement for improving the measurement precision from the standard quantum limit (SQL) to the Heisenberg limit (HL) [11–15]. On the other hand, in atomic spectroscopy [16–19], atom-atom interaction always results in collision shift [20] which brings systematic error in determining the resonance frequency [21]. Many schemes have been developed to suppress the influences of atom-atom collisions [22–24].

Generally, in high-precision quantum sensing via atomic spectroscopy, there is a contradiction between utilizing entanglement and suppressing collision shift. To achieve high-precision Ramsey spectroscopy, one can use atom-atom interaction to prepare the desired entanglement and then turn off atom-atom interaction for interrogation. In this way, the collision shift can be small, but it requires precise time-dependent manipulation of atom-atom interactions. In Rabi spectroscopy, the coupling field and the atom-atom interaction coexist. It has been demonstrated that one may achieve a Rabi spectroscopy towards the HL with two correlated ions [25]. However, in such a protocol, the system should be split into two orthogonal subspaces with different parities as different probes, which adds additional complexity to the existing Rabi protocols. Therefore, it starves for an efficient spectroscopy protocol without collision shift and meanwhile one can utilize atom-atom interactions to improve the measurement precision beyond the SQL.

In this Letter, we propose a robust quantum enhanced frequency estimation protocol via antisymmetric Rabi spectroscopy, which can be used in various quantum sensors. Instead of preparing all atoms in the lower level for the conventional Rabi spectroscopy, our protocol inputs an equal superposition state of the two sensor levels before applying the coupling field for implementing Rabi oscillation. We analytically find that the Rabi spectroscopy becomes exactly antisymmetric with respect to the detection of arbitrary atom-atom interactions. This means the absence of collision shift in our protocol. More importantly, the measurement precision can be improved beyond the SQL by tuning the atom-atom interaction and the Rabi frequency. Compared to the spin-squeezing-enhanced Ramsey spectroscopy protocol, our protocol is much more robust against detection noises.

Conventional Rabi oscillation in an atomic ensemble. – We consider an ensemble of N two-level atoms, which can be regraded as identical pseudospin-\(\frac{1}{2}\) particles obeying a collective spin \(\hat{J}\) with \(\hat{J}_x = \sum_{n=1}^{N} \sigma_x^{(n)}\), \(\hat{J}_y = \sum_{n=1}^{N} \sigma_y^{(n)}\), and \(\hat{J}_z = \sum_{n=1}^{N} \sigma_z^{(n)}\). Here \(\sigma_x^{(n)}, \sigma_y^{(n)}, \sigma_z^{(n)}\) are the Pauli matrices for the \(n\)-th atom. In the Schwinger representation, \(\hat{J}_x = \frac{1}{2}(\hat{a}^{\dagger}\hat{b} + \hat{b}^{\dagger}\hat{a})\), \(\hat{J}_y = \frac{1}{2}(\hat{a}^{\dagger}\hat{b} - \hat{b}^{\dagger}\hat{a})\), \(\hat{J}_z = \frac{1}{2}(\hat{b}^{\dagger}\hat{b} - \hat{a}^{\dagger}\hat{a})\), where \(\hat{a}\) and \(\hat{b}\) are the annihilation operators for atoms in \(|\uparrow\rangle\) and \(|\downarrow\rangle\), respectively. These collective spin operators obey the general angular momentum commutation relations \([\hat{J}_i, \hat{J}_j] = i\hbar \epsilon_{ijk} \hat{J}_k\) with \(i, j, k = x, y, z\) and \(\epsilon_{ijk}\) the Levi-Civita symbol. Thus, an arbitrary state can be expressed by \(|\Psi\rangle = \sum_{m=-N/2}^{N/2} C_m |J, m\rangle\), where \(|J, m\rangle\) is the Dicke basis denoting \((N/2 - m)\) atoms in \(|\uparrow\rangle\) and \((N/2 + m)\) atoms in \(|\downarrow\rangle\).
A well-known spectroscopic method for frequency estimation is implementing Rabi oscillations. In a non-interacting atomic ensemble driven by an external coupling field, its Rabi oscillations obey the Hamiltonian $\hat{H} = \omega_0 \hat{J}_z + \Omega (e^{i\omega t} + e^{-i\omega t}) \hat{J}_z$, where $\omega_0$ is the atomic transition frequency, $\omega$ is the frequency of the coupling field, and $\Omega$ is the Rabi frequency. For simplicity, we set $h = 1$ hereafter. In the rotating-frame with rotating-wave approximation (RWA), the Hamiltonian becomes

$$\hat{H}_R = \Omega \hat{J}_x + \delta \hat{J}_z,$$

where the detuning $\delta = \omega_0 - \omega$. Without atom-atom interactions, the system state can be described by a spin coherent state (SCS) $|\theta, \phi\rangle_{SCS} = (\cos \theta/2 |\uparrow\rangle + e^{i\phi}\sin \theta/2 |\downarrow\rangle)^\otimes N$, in which all atoms are in the same quantum state. Conventionally, one prepare an initial SCS $|\psi\rangle_0 = |\pi, 0\rangle_{SCS} = (|\downarrow\rangle)^\otimes N$ with all atoms in $|\downarrow\rangle$. Then, the initial state evolves according to $|\psi(t)\rangle = e^{-i\hat{H}_R t} |\psi\rangle_0$. At time $T$, we have the half population difference

$$\langle \hat{J}_z(T) \rangle = \langle \psi(T)|\hat{J}_z|\psi(T)\rangle = -\frac{N}{2} \frac{\delta^2 + \Omega^2 \cos(\sqrt{\Omega^2 + \delta^2} T)}{\Omega^2 + \delta^2},$$

which is a symmetric function with respect to $\delta = 0$. In practice, one would choose $\Omega T = \pi$ and measure the half population difference $\langle \hat{J}_z(T) \rangle$. It attains the maximum at $\delta = 0$ so that can be used as the frequency locking signal, see the blue solid line in Fig. 1 (b).

Taking into account the atom-atom interaction, the original Hamiltonian becomes $\hat{H}' = \omega_0 \hat{J}_z + \chi \hat{J}_+^2 + \Omega (e^{i\omega t} + e^{-i\omega t}) \hat{J}_z$, where $\chi$ characterizes the strength of effective atom-atom interaction [see Fig. 1 (a)]. In the rotating-frame with RWA, the Hamiltonian reads

$$\hat{H}'_R = \Omega \hat{J}_x + \chi \hat{J}_+^2 + \delta \hat{J}_z.$$

When $\chi$ is non-negligible, the atomic collision causes a frequency shift and the locking signal is no longer symmetric with $\delta = 0$. For example, when $\chi = 0.01\Omega$, an obvious frequency shift can be observed, see the green dashed line in Fig. 1 (b). While for $\chi = 0.02\Omega$, the frequency shift is larger and the lineshape also distorted, see the orange dash-dotted line in Fig. 1 (b).

Thus in the conventional Rabi spectroscopy, the atom-atom interaction always induces a collision shift that de-creasing the accuracy for estimating the transition frequency. Besides, since the maximum is around $\delta = 0$, its signal slope equals zero and the standard deviation $\Delta \delta$ at this point is diverged. In general, it is necessary to find out two symmetric points (with respect to $\delta = 0$) to determine the on-resonance point. These limit the performances of the Rabi spectroscopy. In the following, we analytically analyze the quantum sensing via anti-symmetric Rabi spectroscopy and demonstrate how to achieve better frequency estimation at the on-resonance point $\delta = 0$.

**Antisymmetric Rabi spectroscopy.** – Let’s turn back to the non-interacting case ($\chi = 0$). For the Rabi oscillation from an arbitrary initial state $|\psi\rangle_0$, we analytically obtain its half population difference $\langle J_z(T) \rangle$ at time $T$,

$$\langle \hat{J}_z(T) \rangle = \langle \psi(T)|\hat{J}_z|\psi(T)\rangle = 0$$

and

$$\langle \hat{J}_+ \rangle_0 = \left( \sum_{k=1}^{\infty} \frac{1}{(2k)!} x^{2k} \right) \langle \psi(T)|\hat{J}_+|\psi(T)\rangle_0 + \left( \sum_{k=1}^{\infty} \frac{1}{(2k-1)!} y^{2k-1} \right) \langle \psi(T)|\hat{J}_z|\psi(T)\rangle_0$$

$$= \langle \hat{J}_+ \rangle_0 \left( 1 + \sum_{k=1}^{\infty} \frac{1}{(2k)!} z^{2k} \right) + \langle \hat{J}_\Delta \rangle_0 \left( \sum_{k=1}^{\infty} \frac{1}{(2k-1)!} y^{2k-1} \right) \langle \psi(T)|\hat{J}_z|\psi(T)\rangle_0$$

$$= \langle \hat{J}_\Delta \rangle_0 X(\Omega, \delta, T) + \langle \hat{J}_\Delta \rangle_0 Y(\Omega, \delta, T) + \langle \hat{J}_\Delta \rangle_0 Z(\Omega, \delta, T),$$

where $x_k = \Omega T, y_k = y_{2k-1}(\delta T), z_k = -y_{2k-1}(\Omega T)$ and $y_{2k-1} = -(\delta T) x_{2k} + (\Omega T) z_{2k}$ for $k > 1$. The sums $X(\Omega, \delta, T) = \sqrt{\Omega^2 + \delta^2} / \Omega$, $Y(\Omega, \delta, T) = \Omega \sin(\sqrt{\Omega^2 + \delta^2} T)$, and $Z(\Omega, \delta, T) = \Omega^2 \cos(\sqrt{\Omega^2 + \delta^2} T)$ can be analytically calculated, see more details [26]. If $|\psi\rangle_0 = |\pi, 0\rangle_{SCS}$, we have $\langle \hat{J}_\Delta \rangle_0 = -\frac{\sqrt{\Omega^2 + \delta^2}}{\Omega^2 + \delta^2}$ and $\langle \hat{J}_\Delta \rangle_0 = 0$ and Eq. (4) gives Eq. (2).

According to Eq. (4), one can analytically give the relation between the half population difference $\langle \hat{J}_z(T) \rangle$
and the detuning $\delta$ for any initial state $|\psi\rangle_0$. Since $X(\Omega, \delta, t) = -X(\Omega, \delta, t)$, $Y(\Omega, \delta, t) = Y(\Omega, \delta, t)$ and $Z(\Omega, \delta, t) = Z(\Omega, \delta, t)$, we find that, when $\langle \hat{J}_z \rangle_0 = \langle J_y \rangle_0 = 0$, the half population difference $\langle \hat{J}_z (T) \rangle = \frac{\Omega \delta}{\Omega^2 + \delta^2} \left[ 1 \cos \sqrt{\Omega^2 + \delta^2 T} \right] \langle \hat{J}_z \rangle_0$ is exactly antisymmetric with respect to $\delta = 0$. For an example, inputting the initial state $|\psi\rangle_0 = |\pi/2, 0\rangle_{SCS}$ whose $\langle \hat{J}_z \rangle_0 = \frac{\pi}{2}$ and $\langle \hat{J}_y \rangle_0 = 0$, we have

$$\langle \hat{J}_z (T) \rangle = \frac{N}{2} \frac{\Omega \delta}{\Omega^2 + \delta^2} \left[ 1 \cos \sqrt{\Omega^2 + \delta^2 T} \right].$$

Obviously, $\langle \hat{J}_z (T) \rangle$ is an antisymmetric function of $\delta$ for arbitrary $\Omega$ and $T$, see the blue solid line in Fig. 1 (c). Compared to the conventional Rabi spectroscopy with $|\psi\rangle_0 = |\pi, 0\rangle_{SCS}$, the slope of the signal at on-resonance point $\delta = 0$ becomes sharp and the corresponding measurement precision becomes high [19, 27].

To implement the antisymmetric Rabi spectroscopy, different from the conventional Rabi spectroscopy, one needs to change the input state to $|\psi\rangle_0 = |\pi/2, 0\rangle_{SCS} = e^{i \frac{\pi}{2} J_y} |\pi, 0\rangle_{SCS}$. The desired input state can be easily prepared by a short $\pi/2$ pulse (with large Rabi frequency) along $y$ axis onto the state of all atoms in $|\downarrow\rangle$. The antisymmetric Rabi spectroscopy keeps work under imperfect $\pi/2$ pulse [26].

Amazingly, in the presence of atom-atom interaction, there is no collision shift in the antisymmetric Rabi spectroscopy. We find that the frequency locking signal is always antisymmetric with respect to $\delta = 0$ for arbitrary interaction strength $\chi$, see Fig. 1 (c). Since the antisymmetry of the signal preserves for arbitrary $\chi$, the antisymmetric point has no collision shift and can be used as the on-resonance point. The key for antisymmetric Rabi spectroscopy originates the symmetry of Hamiltonian (3) [28, 29]. Under the transformation $a \rightarrow b(a)$, $\hat{J}_x \rightarrow \hat{J}_x$ and $\hat{J}_z \rightarrow -\hat{J}_z$. The first two terms $\Omega \hat{J}_x$ and $\chi \hat{J}_Z^2$, which have even parity symmetry, are invariant under the transformation of exchanging $|\uparrow\rangle$ and $|\downarrow\rangle$. The last term $\delta \hat{J}_z$, which has odd parity symmetry, changes according to $\delta \hat{J}_z \rightarrow -\delta \hat{J}_z$ when $|\uparrow\rangle \rightarrow |\downarrow\rangle$.

For an initial state $|\psi\rangle_0 = \sum_{m=-J}^J C_m(0)|J, m\rangle$ with $C_m(0) = C_{-m}(0)$ (e.g. $|\psi\rangle_0 = \frac{\pi}{2}, 0\rangle_{SCS}$), when $\delta = 0$, the evolved state will always possess even parity symmetry (that is $C_m(t) = C_{-m}(t)$) [26]. Thus, the half population difference $\langle \hat{J}_z (t) \rangle = \sum_{m=-J}^J m|C_m(t)|^2 = 0$ at the on-resonance point $\delta = 0$. However, if $\delta \neq 0$, $\delta \hat{J}_z$ and $-\delta \hat{J}_z$ rotate along opposite directions, which results in the antisymmetry property $\langle \hat{J}_z (t) \rangle = -\langle \hat{J}_z (-\delta, t) \rangle$ (see the detailed proof [26]). The above results are also valid for individual atoms with $\chi = 0$.

Quantum enhanced sensors via antisymmetric Rabi spectroscopy. Since atom-atom interaction may generate the desired entanglement for achieving quantum enhanced sensing, can we employ atom-atom interaction to improve antisymmetric Rabi spectroscopy? Here, we show how to utilize atom-atom interaction to improve measurement precision. Our scheme can be realized with state-of-the-art techniques in an atomic Bose-Einstein condensate (BEC) [30–32], an optical cavity system with light-mediated interactions [33–35], and etc.

In the following, we take the system of Bose condensed atoms for an example [31]. Another example in collective cavity-QED system [36] is shown in [26]. We compare the locking signals using our scheme with and without atom-atom interaction. We choose $\chi = 2\pi \times 0.063$ Hz, which is a typical atom-atom interaction strength for one-axis twisting with the total atom number $N \approx 400$ in experiment [31, 37]. The Rabi frequency is chosen as $\Omega = N \chi/2 = 2\pi \times 12.6$ Hz, which is the optimal condition for twist-and-turn dynamics [38–40] whose Hamiltonian is similar to Eq. (3). For $N = 400$, we numerically find the optimal evolution time $T = 0.06/\chi \approx 0.1516$ s. In comparison with the conventional Rabi spectroscopy via non-interacting atoms, we choose $T = 0.1516$ s and $\Omega = 2\pi \times 3.3$ Hz which satisfy the optimal condition $\Omega T = \pi$. As shown in Fig. 2 (a) and (b), the locking signal with $\chi > 0$ oscillates dramatically (especially near $\delta = 0$) compared with the ones of $\chi = 0$. Unlike the conventional Rabi spectroscopy, the resolution of the antisymmetric Rabi spectroscopy can be greatly enhanced in the presence of atom-atom interaction. The slope at the on-resonance point is much sharper which indicates

![FIG. 2. (Color online) (a) The scaled population difference $\langle \hat{J}_z (T) \rangle / N$ versus the detuning $\delta$ and (c) the measurement precision $\Delta \omega_\delta$ versus $\delta$ for individual atoms ($\chi = 0$) with ($\Omega = 2\pi \times 3.3$ Hz, $T = 0.1516$ s) satisfying the optimal condition $\Omega T = \pi$, the half population difference $\langle \hat{J}_z \rangle_0 = 0$ becomes sharp and the corresponding measurement precision becomes high [19, 27].](image-url)
Comparison to Ramsey interferometry with spin squeezed states. - Ramsey interferometry is another widely used method for atomic spectroscopy with narrower spectrum compared with Rabi spectroscopy [17–19]. By using entangled states such as spin squeezed states, the measurement precision of the Ramsey spectroscopy can be improved beyond the SQL [41–43]. Here, we find that our antisymmetric Rabi spectrum can exhibit higher resolution and better robustness against detection noises than the Ramsey scheme.

To compare with our scheme, we consider the input state of the Ramsey spectroscopy is an optimal spin squeezed state generated by one-axis twisting (OAT) during \( T_p = 31/6 N^{-2/3}/\chi \) [44]. To make the squeezed direction perpendicular to \( z \) axis with \( \alpha \) the optimal rotation angle, a rotation \( e^{i\alpha J_z} \) has to be applied [45, 46]. Then the state undergoes phase accumulation within a duration of \( T_R \). In comparison with our scheme under the same temporal resource, we set \( T_R = T - T_p \). Finally, a \( \pi/2 \) pulse along \( x \) axis is applied and the population difference measurement is performed.

The comparison of the scaled population difference \( \langle \hat{J}_z(T) \rangle/N \) between our scheme and the conventional OAT Ramsey scheme is shown in Fig. 3. At the on-resonance point, the signal slope of our scheme is much sharper and the measurement precision attains the same level, see Fig. 3 (a) and (b). Besides, we find that the phase measurement precision scaling of our scheme is similar to the conventional OAT Ramsey protocol. The standard deviation of the phase \( \Delta(\omega_0T) \) scales as \( N^{-5/6} \), which is consistent with the standard OAT scaling, see Fig. 3 (c). The process in our scheme is much simpler. Unlike the Ramsey protocol with separated entangled state preparation and interrogation stages, which needs time-dependent control of the atom-atom interaction and the coupling field, our scheme does not need those time-dependent control.

In practice, the detection may become imperfect due to the presence of unavoidable detection noises. Considering the Gaussian detection noise \( \sigma^2 \), the variance of the population difference becomes \( \Delta^2 \hat{J}_z = \Delta^2 \hat{J}_z + \sigma^2 \) and the measurement precision is \( \Delta \omega_0 = \Delta \hat{J}_z / [\langle \partial^2(\hat{J}_z(T))/\partial \omega_0^2 \rangle] \). As shown in Fig. 3 (d), under the shot noise \( \sigma = \sqrt{N} \), our scheme is much more robust against the detection noise. This is because that our scheme simultaneously magnifies the quantum fluctuation and the slope of the signal. When the quantum fluctuation submerges the classical noise, our scheme may achieve excellent robustness against noises [47–49]. In addition, our scheme does not need to prepare specific entangled state and to perform nonlinear detection [33, 47, 48, 50], which is much simpler compared to the protocols of Ramsey interferometry with other entangled states [51, 52].

Summary and discussions. - We have presented a novel protocol for achieving robust quantum enhanced sensing via antisymmetric Rabi spectroscopy. The antisymmetric Rabi spectroscopy can be easily implemented by inputting an equal superposition state for Rabi oscillations. The antisymmetric Rabi spectroscopy have many advantages compared with the conventional ones: (i) It has no collision shift for arbitrary atom-atom interaction; (ii) Its resolution can be dramatically improved beyond the SQL by using atom-atom interaction; and (iii) It is robust against detection noise. Our scheme does not require large modification to the existing experimental setups. It can be easily realized with state-of-the-art techniques in Bose-condensed atomic systems [31, 32, 53], cavity systems with light-mediated interactions [35, 36, 54], and trapped ions [55]. Our work revolutionizes the Rabi spectroscopy with higher resolution, measurement precision and robustness, which offers a new way for quantum enhanced frequency measurement.

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Email: hjiahao@mail2.sysu.edu.cn, eqjiahao@gmail.com
Email: chleecn@szu.edu.cn, chleecn@gmail.com

[1] C. L. Degen, F. Reinhardt, and P. Cappellaro, “Quantum sensing,” Rev. Mod. Phys. 89, 035002 (2017).

[2] John Kitching, Svenja Knappe, and Elizabeth A. Donley, “Atomic sensors – a review,” IEEE Sensors Journal 11, 1749–1758 (2011).

[3] S. A. Diddams, J. C. Bergquist, S. R. Jefferts, and C. W. Oates, “Standards of Time and Frequency at the Outset of the 21st Century.” Science 306, 1318–1324 (2004).

[4] E. Oelker, R. B. Hutson, C. J. Kennedy, L. Sonderhouse, T. Bothwell, A. Goban, D. Kedar, C. Sanner, J. M. Robinson, G. E. Marti, D. G. Matei, T. Legero, M. Giunta, R. Holzwarth, F. Richle, U. Sterr, and J. Ye, “Demonstration of 4.8x10^-17 stability at 1 s for two independent optical clocks,” Nature Photonics 13, 714–719 (2019).

[5] Daniel Benedetto Orenes, Robert J. Sewell, Jérôme Lodewyck, and Morgan W. Mitchell, “Improving Short-Term Stability in Optical Lattice Clocks by Quantum Nondemolition Measurements,” Phys. Rev. Lett. 128, 153201 (2021).

[6] I. Baumgart, J.-M. Cai, A. Retzker, M. B. Plenio, and Ch Wunderlich, “Ultrasensitive Magnetometer using a Single Atom,” Phys. Rev. Lett. 116, 240801 (2016).

[7] S. J. Smullin, I. M. Savukov, G. Vasilakis, R. K. Ghosh, and M. V. Romalis, “Low-noise high-density alkali-metal scalar magnetometer,” Phys. Rev. A 80, 033420 (2009).

[8] Hao Shi, Ma Jie, Xiaofeng Li, Jie Liu, Chao Li, and S. J. Smullin, I. M. Savukov, G. Vasilakis, R. K. Ghosh, and M. V. Romalis, “Low-noise high-density alkali-metal scalar magnetometer,” Phys. Rev. A 80, 033420 (2009).

[9] J. H. Simpson, J. T. Fraser, and I. A. Greenwood, “An optically pumped nuclear magnetic resonance gyroscope,” IEEE Transactions on Aerospace 1, 1107–1110 (1963).

[10] Stuart S Zsigeti, Onur Hosten, and Simon A. Haine, “Improving cold-atom sensors with quantum entanglement: Prospects and challenges,” Applied Physics Letters 118, 140501 (2021).

[11] Vittorio Giovannetti, Seth Lloyd, and Lorenzo Maccone, “Quantum metrology,” Phys. Rev. Lett. 96, 010401 (2006).

[12] Chaohong Lee, “Adiabatic mach-zehnder interferometry on a quantized josephson junction,” Phys. Rev. Lett. 97, 150402 (2006).

[13] Jiahao Huang, Shuyuan Wu, Honghua Zhong, and Chaohong Lee, “Quantum Metrology with Cold Atoms, Vol. 2” (2014) pp. 365–415.

[14] Luca Pezzè, Augusto Smerzi, Markus K. Oberthaler, Roman Schmied, and Philipp Treutlein, “Quantum metrology with nonclassical states of atomic ensembles,” Rev. Mod. Phys. 90, 035005 (2018).

[15] Bo Lu, Cheng-Yin Han, Min Zhuang, Yong-Guan Ke, Jia-Hao Huang, and Chao-Hong Lee, “Non-gaussian entangled states and quantum metrology with ultracold atomic ensemble,” Acta Physica Sinica 68, 040306 (2019).

[16] I I Rabi, S Millman, P Kusch, and J R Zacharias, “The molecular beam resonance method for measuring nuclear magnetic moments,” Phys. Rev 53, 318 (1938).

[17] Norman F. Ramsey, “A new molecular beam resonance method,” Physical Review 76, 996–996 (1949).

[18] Norman F. Ramsey, “A molecular beam resonance method with separated oscillating fields,” Physical Review 87, 695–690 (1950).

[19] Norman F. Ramsey, “The method of successive oscillation fields,” Physics Today 33, 25–30 (1980).

[20] Andrew D Ludlow, Martin M Boyd, Jun Ye, Ekkehard Peik, and P. O. Schmidt, “Optical atomic clocks,” Rev. Mod. Phys. 87, 637–701 (2015).

[21] S. J. J. M. F. Kokkelmans, B. J. Verhaar, K. Gibble, and D. J. Heinzen, “Predictions for laser-cooled rb clocks,” Phys. Rev. A 56, R4389–R4392 (1997).

[22] K. Szymaniec, W. Chalupczak, E. Tiesinga, C. J. Williams, S. Weyers, and R. Wynands, “Cancellation of the Collisional Frequency Shift in Caesium Fountain Clocks,” Phys. Rev. Lett. 98, 153002 (2007).

[23] Zhenhua Yu and C. J. Pethick, “Clock shifts of optical transitions in ultracold atomic gases,” Phys. Rev. Lett. 104, 010801 (2010).

[24] Sangkyung Lee, Chang Yong Park, Won Kyu Lee, and Dai Huyuk Yu, “Cancellation of collisional frequency shifts in optical lattice clocks with Rabi spectroscopy,” New Journal of Physics 18 (2016).

[25] Ravid Shani, Tom Manovitz, Yotam Shapira, Nitzan Akerman, and Roee Ozeri, “Toward heisenberg-limited rabi spectroscopy,” Phys. Rev. Lett. 120, 243603 (2018).

[26] See Supplemental Material for details on: (i) Derivation of the half population difference Eq. (4), (ii) Measurement precisions for noninteracting systems, (iii) Analytical analysis on antisymmetric locking signal for noninteracting systems, (iv) Analytical analysis on antisymmetric locking signal for interacting systems, (v) Influences of imperfect π/2 pulse for preparing the initial state, (vi) Experimental feasibility via Bose condensed atoms, and (vii) Experimental feasibility via collective Cavity-QED system.

[27] Christian Sanner, Nils Huntemann, Richard Lange, Christian Tamm, and Ekkehard Peik, “Autobalanced rabi spectroscopy,” Phys. Rev. Lett. 120, 53602 (2018).

[28] A. Trenkwalder, G. Spagnoli, G. Semechni, S. Cooper, M. Landini, P. Castillo, L. Pezzé, G. Modugno, M. Inguscio, A. Smerzi, and M. Fattori, “Quantum phase transitions with parity-symmetry breaking and hysteresis,” Nature Physics 12, 826–829 (2016).

[29] Min Zhuang, Jiahao Huang, Yongguan Ke, and Chao-Hong Lee, “Symmetry-protected quantum adiabatic evolution in spontaneous symmetry-breaking transitions,” Annales der Physik 532, 1900471 (2020).

[30] Max F. Riedel, Pascal Böhi, Yun Li, Theodor W. Hänsch, Alice Sinatra, and Philipp Treutlein, “Atomchip-based generation of entanglement for quantum metrology,” Nature 464, 1170–1173 (2010).

[31] C. Gross, T. Zibold, E. Nicklas, J. Estève, and M. K. Oberthaler, “Nonlinear atom interferometer surpasses classical precision limit,” Nature 464, 1165–1169 (2010).

[32] Caspar F. Ockeloen, Roman Schmied, Max F. Riedel, and Philipp Treutlein, “Quantum metro-
ogy with a scanning probe atom interferometer,” Phys. Rev. Lett. 111, 143001 (2013).

[33] Emily Davis, Gregory Bentsen, and Monika Schleier-Smith, “Approaching the heisenberg limit without single-particle detection,” Phys. Rev. Lett. 116, 053601 (2016).

[34] Simone Colombo, Edwin Pedrozo-Peafiel, Albert F. Adiyatullin, Zeyang Li, Enrique Mendez, Chi Shu, and Vladan Vuletić, “Time-reversal-based quantum metrology with many-body entangled states,” (2021), arXiv:2106.03754 [quant-ph].

[35] Zeyang Li, Boris Braverman, Simone Colombo, Chi Shu, Akio Kawasaki, Albert F. Adiyatullin, Edwin Pedrozo-Peafiel, Enrique Mendez, and Vladan Vuletić, “Collective spin-light and light-mediated spin-spin interactions in an optical cavity,” PRX Quantum 3, 020308 (2022).

[36] Graham P. Greve, Chengyi Luo, Baochen Wu, and James K. Thompson, “Entanglement-Enhanced Matter-Wave Interferometry in a High-Finesse Cavity,” (2021), arXiv:2110.14027.

[37] B. Juliá-Díaz, E. Torrontegui, J. Martorell, J. G. Muga, and A. Polls, “Fast generation of spin-squeezed states in bosonic josephson junctions,” Phys. Rev. A 86, 063623 (2012).

[38] W. Mussel, H. Strobel, D. Linnemann, T. Zibold, B. Juliá-Díaz, and M. K. Oberthaler, “Twist-and-turn spin squeezing in bose-einstein condensates,” Phys. Rev. A 92, 023603 (2015).

[39] Safoura S. Mirkhalaf, Samuel P. Nolan, and Simon A. Haine, “Robustifying twist-and-turn entanglement with interaction-based readout,” Phys. Rev. A 97, 053618 (2018).

[40] Giacomo Sorelli, Manuel Gesser, Augusto Smerzi, and Luca Pezzè, “Fast and optimal generation of entanglement in bosonic josephson junctions,” Phys. Rev. A 99, 022329 (2019).

[41] D. J. Wineland, J. J. Bollinger, W. M. Itano, F. L. Moore, and D. J. Heinzen, “Spin squeezing and reduced quantum noise in spectroscopy,” Phys. Rev. A 46, R6797–R6800 (1992).

[42] D. J. Wineland, J. J. Bollinger, W. M. Itano, and D. J. Heinzen, “Squeezed atomic states and projection noise in spectroscopy,” Phys. Rev. A 50, 67–88 (1994).

[43] Anne Louchet-Chauvet, Jürgen Appel, Jelmer J. Renema, Daniel Oblak, Niels Kjaergaard, and Eugene S. Polzik, “Entanglement-assisted atomic clock beyond the projection noise limit,” New Journal of Physics 12, 065032 (2010).

[44] Masahiro Kitagawa and Masahito Ueda, “Squeezed spin states,” Phys. Rev. A 47, 5138–5143 (1993).

[45] Jian Ma, Xiaoguang Wang, C.P. Sun, and Franco Nori, “Quantum spin squeezing,” Physics Reports 509, 89–165 (2011).

[46] Christian Gross, “Spin squeezing, entanglement and quantum metrology with bose-einstein condensates,” Journal of Physics B 45, 103001 (2012).

[47] Samuel P. Nolan, Stuart S. Szegi, and Simon A. Haine, “Optimal and robust quantum metrology using interaction-based readouts,” Phys. Rev. Lett. 119, 193601 (2017).

[48] Simon A. Haine, “Using interaction-based readouts to approach the ultimate limit of detection-noise robustness for quantum-enhanced metrology in collective spin systems,” Phys. Rev. A 98, 030303 (2018).

[49] Jiahao Huang, Min Zhuang, and Chaohong Lee, “Non-gaussian precision metrology via driving through quantum phase transitions,” Phys. Rev. A 97, 032116 (2018).

[50] Jiahao Huang, Min Zhuang, Bo Lu, Yongguan Ke, and Chaohong Lee, “Achieving heisenberg-limited metrology with spin cat states via interaction-based readout,” Phys. Rev. A 98, 012129 (2018).

[51] Onur Hosten, Nils J. Engelsen, Rajiv Krishnakumar, and Mark A. Kasevich, “Measurement noise 100 times lower than the quantum-projection limit using entangled atoms,” Nature 529, 505–508 (2016).

[52] Jinyang Li, Gregorio R. M. da Silva, Schuyler Kain, Gour Pari, Renu Tripathi, and Selim M. Shahriar, “Spin Squeezing Induced Enhancement of Sensitivity of an Atomic Clock using Coherent Population Trapping,” (2021), arXiv:2112.08013.

[53] Helmut Strobel, Wolfgang Muessel, Daniel Linnemann, Tilman Zibold, David B. Hume, L. Pezze, Augusto Smerzi, and Markus K. Oberthaler, “Fisher information and entanglement of non-gaussian spin states,” Science 345, 424–427 (2014).

[54] Edwin Pedrozo-Peafiel, Simone Colombo, Chi Shu, Albert F. Adiyatullin, Zeyang Li, Enrique Mendez, Boris Braverman, Akio Kawasaki, Daisuke Akamatsu, Yanhong Xiao, and Vladan Vuletić, “Entanglement on an optical atomic-clock transition,” Nature 588, 414–418 (2020).

[55] Kevin A. Gilmore, Matthew Affolter, Robert J. Lewis-Swan, Diego Barberena, Elena Jordan, Ana Maria Rey, and John J. Bollinger, “Quantum-enhanced sensing of displacements and electric fields with two-dimensional trapped-ion crystals,” Science 373, 673–678 (2021).

[56] Christian Gross, “Spin squeezing and non-linear atom interferometry with bose-einstein condensates,” PhD dissertation (2010).
Derivation of the half population difference Eq. (4)

In order to analytically calculate \( \langle \hat{J}_z(T) \rangle = \langle \psi(T) | \hat{J}_z | \psi(T) \rangle \), we can make use of the formula \( e^{A}Be^{-A} = \sum_{n=0}^{\infty} \frac{1}{n!} [A^{(n)}, B] \). For Hamiltonian \( \hat{H}_x \), we have

\[
[\hat{H}_x^{(0)}, \hat{J}_z] = \hat{J}_z, \tag{S1}
\]

\[
[\hat{H}_x^{(1)}, \hat{J}_z] = [\hat{H}_x, \hat{J}_z] = \Omega T \hat{J}_z, \tag{S2}
\]

\[
[\hat{H}_x^{(2)}, \hat{J}_z] = [\hat{H}_x, \Omega T \hat{J}_z] = -\Omega T \hat{J}_z + (\Omega T)(\delta T) \hat{J}_z. \tag{S3}
\]

When proceeding to higher order, we find that \( [\hat{H}_x^{(n)}, \hat{J}_z] \) will only contain the linear operators of \( \hat{J}_z \), \( \hat{J}_y \), and \( \hat{J}_x \). Then, we have

\[
[\hat{H}_x^{(n)}, \hat{J}_z] = x_n \hat{J}_x + y_n \hat{J}_y + z_n \hat{J}_z. \tag{S4}
\]

The coefficients satisfy

\[
x_{n+1} = (\delta T)y_n, \tag{S5}
\]

\[
z_{n+1} = -\Omega T z_n - \delta T x_n, \tag{S6}
\]

\[
y_{n+1} = -\delta T y_n + (\Omega T) z_n. \tag{S7}
\]

From Eqs. (S1)-(S3), we have \( x_0 = 0, x_1 = 0, x_2 = (\Omega T)(\delta T); y_0 = 0, y_1 = \Omega T, y_2 = 0; z_0 = 1, z_1 = 0, z_2 = -\delta T \). Using the recursive relations between these coefficients, we obtain

\[
x_{2n+2} = -\Omega^2 + \delta^2 \Omega^2 x_{2n} + x_{2n+1} = 0, \tag{S8}
\]

\[
z_{2n+2} = -\Omega^2 + \delta^2 \Omega^2 z_{2n} + z_{2n+1} = 0, \tag{S9}
\]

\[
y_{2n+3} = -\Omega^2 + \delta^2 \Omega^2 y_{2n+1} + y_{2n+2} = 0 \tag{S10}
\]

with \( n = 0, 1, 2, \ldots \). Thus, we can analytically give the expressions for the coefficients as

\[
x_{2n+2} = (-1)^n \frac{\Omega \delta}{\Omega^2 + \delta^2} \left( \sqrt{\Omega^2 + \delta^2} T \right)^{2n+2}, \tag{S11}
\]

\[
z_{2n+2} = (-1)^n \frac{\Omega^2}{\Omega^2 + \delta^2} \left( \sqrt{\Omega^2 + \delta^2} T \right)^{2n+2}, \tag{S12}
\]

\[
y_{2n+3} = (-1)^n \frac{\Omega}{\sqrt{\Omega^2 + \delta^2}} \left( \sqrt{\Omega^2 + \delta^2} T \right)^{2n+1}. \tag{S13}
\]

Finally, we can analytically obtain the sums for the coefficients,

\[
X(\Omega, \delta, T) = \sum_{k=1}^{\infty} \frac{x_{2k}}{(2k)!}, \tag{S14}
\]

\[
= \frac{\Omega \delta}{\Omega^2 + \delta^2} \left[ 1 - \cos(\sqrt{\Omega^2 + \delta^2} T) \right], \tag{S14}
\]

\[
Y(\Omega, \delta, T) = \sum_{k=1}^{\infty} \frac{y_{2k-1}}{(2k-1)!} = \frac{\Omega \sin(\sqrt{\Omega^2 + \delta^2} T)}{\sqrt{\Omega^2 + \delta^2}}. \tag{S15}
\]

\[
Z(\Omega, \delta, T) = \sum_{k=0}^{\infty} \frac{z_{2k}}{(2k)!} = \frac{\delta^2 + \Omega^2 \cos(\sqrt{\Omega^2 + \delta^2} T)}{\Omega^2 + \delta^2}. \tag{S16}
\]

Thus, we get the final result of Eq. (3) for arbitrary initial state,

\[
\langle \hat{J}_z(T) \rangle = X(\hat{J}_x)|_0 + Y(\hat{J}_y)|_0 + Z(\hat{J}_z)|_0. \tag{S17}
\]

with \( X = X(\Omega, \delta, T), Y = Y(\Omega, \delta, T), Z = Z(\Omega, \delta, T) \).

Measurement precisions for noninteracting systems

Below we show how to calculate measurement precisions for noninteracting systems (i.e. \( \chi = 0 \)). Given \( \delta \) and \( \Omega \), we first need to analytically obtain the expressions of \( \langle \hat{J}_z(T) \rangle \). It is easy to find that

\[
\langle \hat{J}_z^2(T) \rangle = \langle \psi(T) | \hat{J}_z^2 | \psi(T) \rangle = \langle \psi \frac{e^{|H_T} t e^{-iH_T t} \rangle}{\psi} = 0 \tag{S18}
\]

\[
Y \langle \hat{J}_y(t) \rangle = Y \langle \hat{J}_y(t) \rangle + Z \langle \hat{J}_z(t) \rangle = 0 \tag{S18}
\]

\[
X = X^2(\Omega, \delta, T), \quad Y = Y^2(\Omega, \delta, T), \quad ZZ = Z^2(\Omega, \delta, T), \quad XY = X(\Omega, \delta, T) Y(\Omega, \delta, T), \quad YZ = Y(\Omega, \delta, T) Z(\Omega, \delta, T), \quad XZ = X(\Omega, \delta, T) Z(\Omega, \delta, T), \quad \{\bullet, \bullet\} = \bullet \cdot + \bullet \cdot \] denotes the anti-commutator. According to Eqs. (S17) and (S18), we can explicitly write the expression,

\[
\Delta \hat{J}_z = \sqrt{\langle \hat{J}_z^2(T) \rangle - \langle \hat{J}_z(T) \rangle^2}. \tag{S19}
\]

Besides, the deviation \( \partial \langle \hat{J}_z(T) \rangle / \partial \delta \) can also be analytically obtained, which reads

\[
\frac{\partial \langle \hat{J}_z(T) \rangle}{\partial \delta} = X' \langle \hat{J}_x \rangle + Y' \langle \hat{J}_y \rangle + Z' \langle \hat{J}_z \rangle, \tag{S20}
\]

with
FIG. S1. The first, second and third column represent the results with initial state $|\psi_0\rangle = |\pi/2, 0\rangle_{SCS}$, $|\pi/2, \pi/2\rangle_{SCS}$, and $|\pi, 0\rangle_{SCS}$, respectively. The first, second, third and fourth row respectively correspond to the population difference, absolute slope of the population difference, variance of population difference and the measurement precision versus detuning. Here, atom number $N = 100$, $\Omega T = \pi$.

\[
X' = \frac{\Omega}{\Omega^2 + \delta^2} \left[ 1 - \cos(\sqrt{\Omega^2 + \delta^2} T) \right] + \frac{\delta^2 \Omega T}{(\Omega^2 + \delta^2)^{3/2}} \left[ \sin(\sqrt{\Omega^2 + \delta^2} T) \right] \\
- \frac{2\delta^2 \Omega}{(\Omega^2 + \delta^2)^2} \left[ 1 - \cos(\sqrt{\Omega^2 + \delta^2} T) \right]. \tag{S21}
\]

\[
Y' = \frac{\delta \Omega T}{\Omega^2 + \delta^2} \cos(\sqrt{\Omega^2 + \delta^2} T) - \frac{\delta \Omega}{(\Omega^2 + \delta^2)^{3/2}} \sin(\sqrt{\Omega^2 + \delta^2} T), \tag{S22}
\]

\[
Z' = \frac{2\delta}{\Omega^2 + \delta^2} + \frac{2\delta^3}{(\Omega^2 + \delta^2)^2} - \frac{\delta \Omega^2 T}{(\Omega^2 + \delta^2)^{3/2}} \sin(\sqrt{\Omega^2 + \delta^2} T) \\
- \frac{2\delta \Omega^2}{(\Omega^2 + \delta^2)^2} \cos(\sqrt{\Omega^2 + \delta^2} T). \tag{S23}
\]

Finally, according to Eqs. (S17) - (S25), can also be analytically expressed. Since $\delta = \omega_0 - \omega$, the

\[
\Delta \delta = \frac{\Delta \dot{J}_z}{|\dot{\partial}(J_z(T))/\partial \delta|} \tag{S24}
\]
standard deviation $\Delta \omega_0 = \Delta \delta$ mathematically. For convenience, we calculate the measurement precision $\Delta \omega_0$ by $\Delta \delta$ instead.

**Analytical analysis on antisymmetric locking signal for noninteracting systems**

Below we analyze the antisymmetric locking signal for noninteracting systems. From Eqs. (S14) - (S17), one can easily find that $X(\Omega, \delta, T) = -X(\Omega, -\delta, T)$, $Y(\Omega, \delta, T) = Y(\Omega, -\delta, T)$, and $Z(\Omega, \delta, T) = Z(\Omega, -\delta, T)$ so that $\langle J_z(\delta, T) \rangle = -\langle J_z(-\delta, T) \rangle$ if $\langle J_y \rangle_0 = \langle J_z \rangle_0 = 0$. For example, if the initial state is $|\psi\rangle_0 = |\pi/2, 0\rangle_{SCS}$, $\langle J_z \rangle_0 = \frac{\pi}{2}$ and $\langle J_z \rangle_0 = 0$, the final population difference $\langle J_z(T) \rangle$ is antisymmetric with $\delta = 0$. While for $|\psi\rangle_0 = |\pi/2, \pi/2\rangle_{SCS}$ and $|\psi\rangle_0 = |\pi, 0\rangle_{SCS}$ (both with $\langle J_z \rangle_0 = 0$), the final population difference $\langle J_z(T) \rangle$ is symmetric with $\delta = 0$. The comparisons with these three different initial states are shown in the first row of Fig. S1.

From Eq. (S25), we can plot the absolute slope of population difference $|\partial \langle J_z(T) \rangle / \partial \delta|$ versus detuning, see the second row of Fig. S1. There is a peak at $\delta = 0$ for $|\psi\rangle_0 = |\pi/2, 0\rangle_{SCS}$, while the slope equals 0 for both $|\psi\rangle_0 = |\pi/2, \pi/2\rangle_{SCS}$ and $|\psi\rangle_0 = |\pi, 0\rangle_{SCS}$.

From Eq. (S19), we can also plot the variance of population difference $\Delta^2 J_z$ versus detuning, see the third row of Fig. S1. Finally, according to Eq. (S24), we can analytically obtain the measurement precision $\Delta \delta$ versus detuning, see the last row of Fig. S1. It is obviously shown that, only $|\psi\rangle_0 = |\pi/2, 0\rangle_{SCS}$ can achieve high-precision measurement at the locking point $\delta = 0$, while for $|\psi\rangle_0 = |\pi/2, \pi/2\rangle_{SCS}$ and $|\psi\rangle_0 = |\pi, 0\rangle_{SCS}$, the measurement precisions are diverged.

The slope of the signal at the locking point $\delta = 0$ can be analytically written as

$$ \frac{\partial \langle J_z(T) \rangle}{\partial \delta} \bigg|_{\delta=0} = \frac{N}{2} \left[ 1 - \cos \Omega T \right]. $$

(S25)

When $\Omega T = \pi$, $\frac{\partial \langle J_z(T) \rangle}{\partial \delta} \bigg|_{\delta=0} = N/\Omega$ attains its maximum, which indicates the high sensitivity for frequency locking. At the locking point $\delta = 0$, $\Delta \delta \propto 1/\sqrt{N}$, which exhibits the SQL scaling.

**Analytical analysis on antisymmetric locking signal for interacting systems**

In this section, we illustrate the mathematical analysis on the antisymmetric locking signal $\langle J_z(\delta, T) \rangle$ (versus $\delta$) when atom-atom interaction is taken into account.

In the Schrodinger picture, the final state evolved from the initial state $|\psi\rangle_0$ at time $T$ can be calculated as

$$ |\psi(T)\rangle_f = e^{i\hat{H}_0 T} e^{-i(\hat{J}_y^2 + \Omega J_z + \delta J_z) T} |\psi\rangle_0. $$

(S26)

In order to analytically analyze the principle of Rabi spectroscopy in the presence of atom-atom interaction, we work in the interaction picture. In the reference of $H_0 = \chi \hat{J}_z^2 + \delta \hat{J}_z$, we have the Hamiltonian

$$ \hat{H}_I = e^{i\hat{H}_0 T} \Omega \hat{J}_z e^{-i\hat{H}_0 T} $$

$$ = \frac{\Omega}{2} e^{i(\hat{J}_+^2 + \delta \hat{J}_z)T} \left( \hat{J}_+ + \hat{J}_- \right) e^{-i(\hat{J}_+^2 + \delta \hat{J}_z)T} $$

$$ = \frac{\Omega}{2} \left( \hat{J}_+^2 + \hat{J}_-^2 \right), $$

(S27)

where $J^I = e^{i(\hat{J}_+^2 + \delta \hat{J}_z)T} \hat{J}_z \hat{J}_- = 0$. Since $\frac{d}{dt} \hat{J}_z^I = i \chi \hat{J}_+^2 + \delta \hat{J}_z \hat{J}_-^I$, $\hat{J}_z^I = 0$, and $\{ \hat{J}_z, \hat{J}_\pm \} = \hat{J}_z \left( 2 \hat{J}_z \pm 1 \right)$, we have

$$ \frac{d}{dt} \hat{J}_z^I = i \left[ \chi \hat{J}_z^2 + \delta \hat{J}_z \hat{J}_- \right] $$

$$ = i \chi \hat{J}_z^2 \hat{J}_+^I + i \delta \left[ \hat{J}_z, \hat{J}_-^I \right] $$

$$ = \pm i \chi \left( \hat{J}_z, \hat{J}_-^I \right) + i \delta \left[ \hat{J}_z, \hat{J}_-^I \right] $$

$$ = \pm i \hat{J}_z^I \chi \left( 2 \hat{J}_z \pm 1 \right) + \delta. $$

(S28)

Therefore, the raising and lowering operators in the interaction picture can be given as

$$ \hat{J}_z^I = \hat{J}_z^I e^{i\chi t + \delta (2\chi t + \hat{J}_z^I)}. $$

(S29)

The state in interaction picture can be expanded in terms of Dicke basis, i.e.,

$$ |\psi(t)\rangle^I = \sum_{m=-N/2}^{N/2} C(t) |J, m\rangle, $$

(S30)

where $|\psi(t)\rangle^I = e^{i\hat{H}_0 t} |\psi(t)\rangle$ with $|\psi(t)\rangle = \sum_{m=-N/2}^{N/2} C(t) |J, m\rangle$ the evolved state in Schrodinger picture. Hence, $C(t) = e^{i(\chi m^2 - \delta m) t} C_0(m)$ and $C_0(m) = C_{\lambda}(0)$.

The time-evolution in interaction picture obeys

$$ i \frac{d}{dt} |\psi(t)\rangle^I = \hat{H}_I |\psi(t)\rangle^I = \frac{\Omega}{2} \left( \hat{J}_+^I + \hat{J}_-^I \right) |\psi(t)\rangle^I. $$

(S31)

Owing to the relations $\hat{J}_+ |J, m\rangle = \sqrt{(J-m)(J+m+1)} |J, m+1\rangle = \lambda^+_m |J, m+1\rangle$ and $\hat{J}_- |J, m\rangle = \sqrt{(J+m)(J-m-1)} |J, m-1\rangle = \lambda^-_m |J, m-1\rangle$, we can get the equations for the coefficients $C_m(t)$. For a given $C_m(t)$, the equation reads
\begin{equation}
|\psi_0 = |\pi/2, 0\rangle_{SCS}, |\pi/2, \pi/2\rangle_{SCS}, \text{ and } |\pi, 0\rangle_{SCS}\rangle,
\end{equation}

FIG. S2. The first, second and third column represents the results with initial state $|\psi_0 = |\pi/2, 0\rangle_{SCS}, |\pi/2, \pi/2\rangle_{SCS}, \text{ and } |\pi, 0\rangle_{SCS}\rangle$, respectively. The first, second, third and fourth row respectively correspond to the population difference, absolute slope of the population difference, variance of population difference and the measurement precision versus detuning. Here, atom number $N = 100$, $\chi = 2\pi \times 0.063$ Hz, $\Omega = 2\pi \times 3.15$ Hz, and $T = 0.198/\chi = 0.5$ s.

\begin{equation}
\dot{C}_m^I(t) = \frac{\Omega}{2} e^{i\chi t} \left[ \lambda^+_m C_{m-1}^I(t) e^{i\delta t + 2i\chi t(m-1)} + \lambda^-_{m+1} C_{m+1}^I(t) e^{-i\delta t - 2i\chi t(m+1)} \right]
\end{equation}

Since $\lambda^+_m = \lambda^-_{-m}$, we also have

\begin{equation}
\dot{C}_{-m}^I(t) = \frac{\Omega}{2} e^{-i\chi t} \left[ \lambda^+_m C_{m-1}^I(t) e^{i\delta t + 2i\chi t m} + \lambda^-_{m+1} C_{m+1}^I(t) e^{-i\delta t - 2i\chi t m} \right].
\end{equation}

In the following, we verify the antisymmetry property of $\langle \hat{J}_z(T) \rangle$ versus $\delta = 0$. For fixed $\Omega$ and $\chi$ at time $t$, $C_m^I(\delta, t)$
FIG. S3. The locking signal versus detuning with imperfect $\pi/2$ pulse. The deviation to a perfect $\pi/2$ is characterized by $\epsilon$. The first row are the results of non-interacting atoms ($\chi=0$) with $\Omega = 2\pi \times 1$ Hz and $T = 0.5$ s. The second row are the results of interacting atoms ($\chi = 2\pi \times 0.063$ Hz) with $\Omega = 2\pi \times 3.15$ Hz and $T = 0.5$ s. Here, we choose $N = 100$.

is dependent on $\delta$. According to Eqs. (S32) and (S33), we have

$$i\dot{C}_m^I(\delta, t) = \frac{\Omega}{2} e^{-i\chi t} \left[ \lambda_{m-1}^+ C_{m-1}^I(t) e^{i\delta t + 2i\chi t} + \lambda_{m+1}^- C_{m+1}^I(t) e^{-i\delta t - 2i\chi t} \right],$$

(S34)

$$i\dot{C}_{-m}^I(-\delta, t) = \frac{\Omega}{2} e^{-i\chi t} \left[ \lambda_{m-1}^- C_{-(m-1)}^I(t) e^{i\delta t + 2i\chi t} + \lambda_{m+1}^+ C_{-(m+1)}^I(t) e^{-i\delta t - 2i\chi t} \right].$$

(S35)

If the coefficients of the initial state $|\psi(0)\rangle = |\psi_0\rangle = \sum_{m=-N/2}^{N/2} C_m(0)|J, m\rangle$ satisfy the condition

$$C_m(0) = C_{-m}(0)$$

(S36)

for arbitrary $m$, we can immediately find that $\dot{C}_m^I(\delta, t) = \dot{C}_{-m}^I(-\delta, t)$ and therefore

$$C_m^I(\delta, t) = C_{-m}^I(-\delta, t).$$

(S37)

At time $T$, the final population difference

$$\langle \hat{J}_z(T) \rangle = \langle \psi(T)|\hat{J}_z|\psi(T)\rangle = \langle \psi(T)|\hat{J}_z|\psi(T)\rangle^T = \sum_{m=-J}^{J} m|C_m^I(T)|^2.$$  

(S38)

For $\delta$, $\langle \hat{J}_z(\delta, T) \rangle = \sum_{m=-J}^{J} m|C_m^I(\delta, T)|^2$. While for $-\delta$, $\langle \hat{J}_z(-\delta, T) \rangle = \sum_{m=-J}^{J} m|C_m^I(-\delta, T)|^2 = \sum_{m=-J}^{J} -m|C_m^I(-\delta, T)|^2$.
Thus, we finally prove that

$$\langle \hat{J}_z(\delta, T) \rangle = -\langle \hat{J}_z(-\delta, T) \rangle. \quad (S36)$$

We choose the initial state as $|\psi\rangle_0 = |\pi/2, 0\rangle_{SCS} = \sum_{m=-J}^{J} \frac{(2J)!}{(J+m)! (J-m)!} |J, m\rangle$ in the main text which satisfies the condition $\langle\hat{J}_z(0)\rangle = 0$, see the first row in Fig. S2. For comparison, we also show the population difference versus detuning with initial states $|\psi\rangle_0 = |\pi/2, \pi/2\rangle_{SCS}$, and $|\pi, 0\rangle_{SCS}$. For these two initial states, $C_m(0)$ does not always equal to $C_{-m}(0)$, thus the population difference is neither antisymmetric nor symmetric with respect to $\delta = 0$, see the first row in Fig. S2.

The absolute slope of the population difference, variance of population difference and the measurement precision versus detuning are also shown in the last three rows of Fig. S2.

**Influences of imperfect $\pi/2$ pulse for preparing the initial state**

To generate the input state $|\psi\rangle_0 = |\pi/2, 0\rangle_{SCS}$ for Rabi oscillation in our scheme, it is necessary to apply a $\pi/2$ pulse along $y$ axis onto the state of all atoms in $| \downarrow \rangle$. Imperfect $\pi/2$ pulse can be characterized by

$$e^{i(\frac{\omega}{2} + \epsilon) \hat{J}_y}, \quad (S40)$$

where $\epsilon \pi$ is the deviation from $\pi/2$.

When $\epsilon = 0$, it is a perfect $\pi/2$ pulse along $y$ axis. Thus, the results for $\epsilon = 0$ are consistent with the previous ones. When $\epsilon \neq 0$, the signal of $\langle \hat{J}_z \rangle$ will deviate from the perfect one, see Fig. S3 (a) and (c). However, the deviations for $\pm\epsilon$ are antisymmetric with respect to 0, i.e.,

$$\langle \hat{J}_z(\delta, \epsilon) \rangle - \langle \hat{J}_z(-\delta, -\epsilon) \rangle = -(\langle \hat{J}_z(\delta, \epsilon) \rangle - \langle \hat{J}_z(-\delta, \epsilon = 0) \rangle).$$

Therefore, the mean signal $\frac{1}{2} \langle \hat{J}_z(\delta, \epsilon) + \hat{J}_z(-\delta, -\epsilon) \rangle$ becomes antisymmetric with respect to $\delta = 0$, see Fig. S3 (b) and (d).

For $\chi = 0$, the mean signal is the same with the perfect one. While for nonzero $\chi$, the mean signal differs from the perfect one depending on $\epsilon$ and $\chi$. However, the antisymmetry will still possess. Thus in practise, one can scan the pulse duration to obtain the condition for perfect $\pi/2$ pulse ($\epsilon = 0$) by analyzing the antisymmetry of the signal. The influences of the imperfect pulse can be easily eliminated in experiments.

**Experimental feasibility via Bose condensed atoms**

A Bose-Einstein condensate of $^{87}$Rubidium has been considered as a promising candidate to create coherent spin squeezing based on two hyperfine states $|31, 46, 56\rangle$. The $|F, m_F\rangle = |1, 1\rangle$ and $|2, -1\rangle$ states in the lower and upper hyperfine manifold are suitable states for the high-precision measurement experiment. They fulfill the two major requirements: the tunability of interspecies interactions and their Zeeman energy shifts are to first order common mode with respect to magnetic fields.

The two states are coupled by a two-photon transition comprising of two frequencies in microwave regime and radio-frequency regime, respectively. The Rabi frequency strength is controlled by the intensity of the electromagnetic radiation. In the typical Bose condensed atomic system of $^{87}$Rb with atom number $N = 400$, for the
FIG. S5. (a) The final population difference at time $T$ versus $\delta$ and (b) the measurement precision versus $T$ at $\delta = 0$ with individual atoms ($\chi = 0$) and interacting atoms ($\chi = 2\pi \times 10$ Hz). For $\chi = 2\pi \times 10$ Hz, we choose $\Omega = 2\pi \times 3500$ Hz, $T = 0.636$ ms. For $\chi = 0$, we choose $\Omega = 2\pi \times 785$ Hz, $T = 0.636$ ms under the optimal condition $\Omega T = \pi$. (c) is the enlarged orange shaded region in (b). (d) The final population difference at time $T$ versus $\delta$ and (e) the measurement precision versus $T$ at $\delta = 0$ using our scheme and Ramsey scheme with spin squeezed state generated via one-axis twisting. For the latter scheme, we consider optimal squeezing with $T_p = 3^{1/6}N^{-2/3}/\chi$ and the Ramsey interrogation time $T_R = T - T_p$. For both schemes, $\chi = 2\pi \times 10$ Hz, $T = 0.636$ ms. (f) is the enlarged orange shaded region in (e). Here, atom number $N = 700$.

For the conventional Rabi spectroscopy, to achieve better measurement precision, the Rabi frequency should be small. Here, we choose $\Omega = 2\pi \times 1$ Hz with $T = 0.5$ s for simulation. Considering the background atom-atom interaction, the spectrum has a small shift and the line-shape is no longer symmetric with $\delta = 0$, see Fig. S4 (a). This collision shift is harmful for accurately determining the resonance frequency. In contrast, using the antisymmetric Rabi spectroscopy, the spectrum is always antisymmetric with $\delta = 0$, see Fig. S4 (b). The resonance point will not alter when atom-atom interaction is taken into account. Other results for tunable atom-atom interaction are shown in Fig. 2 and Fig. 3 in the main text, in which the measurement precision can reach beyond SQL with suitably chosen atom-atom interaction $\chi$, Rabi frequency $\Omega$ and evolution time $T$.

**Experimental feasibility via collective Cavity-QED system**

Our scheme can also be applied to other many-body quantum systems, such as the collective cavity-QED system. For the atomic ensemble in an optical cavity [35, 54], the one-axis twisting can be generated by quantum non-demolition measurements and cavity-mediated spin interaction. In Ref. [36], the spin squeezed states created by quantum non-demolition measurements and cavity-mediated spin interaction are demonstrated, which provides an ideal platform for achieving the quantum enhanced measurement via antisymmetric Rabi spectroscopy.
In the experiment [36], the one-axis twisting strength can be up to $\chi_{\text{OAT}} \approx 2\pi \times 10 \text{ Hz}$ with atom number $N \approx 700$. Thus, in our simulation we choose $\chi = 2\pi \times 10 \text{ Hz}$ and $N = 700$. With Rabi frequency $\Omega = N\chi/2 = 2\pi \times 3500 \text{ Hz}$, the optimal evolution time can be numerically obtained as $T = 0.636 \text{ ms}$. Under the same evolution time, we choose $\Omega = 2\pi \times 785 \text{ Hz}$ in the case of $\chi = 0$ for comparison. Besides, we also compare the results of using Ramsey spectroscopy with optimal one-axis twisting spin squeezed state. The results are shown in Fig. S5, which are similar with the ones in Bose condensed atomic system. According to these results, our protocol can also greatly improve the resolution of the spectrum and the frequency measurement precision in the collective cavity-QED systems.