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Spin-orbit quantum impurity in a topological magnet

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Quantum states induced by single-atomic impurities are at the frontier of physics and material science. While such states have been reported in high-temperature superconductors and dilute magnetic semiconductors, they are unexplored in topological magnets which can feature spin-orbit tunability. Here we use spin-polarized scanning tunneling microscopy/spectroscopy (STM/S) to study the engineered quantum impurity in a topological magnet $\text{Co}_3\text{Sn}_2\text{S}_2$. We find that each substituted In impurity introduces a striking localized bound state. Our systematic magnetization-polarized probe reveals that this bound state is spin-down polarized, in lock with a negative orbital magnetization. Moreover, the magnetic bound states of neighboring impurities interact to form quantized orbitals, exhibiting an intriguing spin-orbit splitting, analogous to the splitting of the topological fermion line. Our work collectively demonstrates the strong spin-orbit effect of the single-atomic impurity at the quantum level, suggesting that a nonmagnetic impurity can introduce spin-orbit coupled magnetic resonance in topological magnets.

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Understanding the single-atomic impurity state in a quantum material is a fundamental problem with widespread implications in physics and technology^{1–8}. For instance, the Zn impurity state in a high-temperature superconductor uncovers the Cooper pairing symmetry³, the Mn impurity state in a semiconductor elucidates the ferromagnetic coupling⁴, and the interstitial Fe impurity in a superconductor with topological surface states creates Majorana-like state⁵. Besides being a local probe of the quantum materials, the impurity state with discrete or (magnetic) bistable quantum levels is valuable for the quantum technology^{6–8}. Most known single-atomic impurity states are, however, either from the spin or orbital channel, limiting their tunability at the quantum level. Recently, spin-orbit coupled kagome magnets have emerged as a new class of quantum materials suitable for microscopic research^{9–19}. In particular, we notice that the In doped $\text{Co}_3\text{Sn}_2\text{S}_2$ exhibits strongly altered bulk magnetic and transport properties, including reductions of magnetism, suppressions of metallicity, and variations of anomalous Hall conductivity^{20–22}. These effects imply a striking, yet not understood quantum state associated with each nonmagnetic In impurity in this topological magnet. Therefore, a single crystal of $\text{Co}_3\text{Sn}_2\text{S}_2$ containing a dilute concentration of In impurities is considered a natural and promising quantum material for experiments on atomic impurity state with spin-orbit tunability. Here we report our spin-polarized STM/S study of 1% In doped $\text{Co}_3\text{Sn}_2\text{S}_2$, which uncovers a spin-orbit quantum impurity state.

Results

Engineered atomic impurity. $\text{Co}_3\text{Sn}_2\text{S}_2$ has a layered crystal structure and a ferromagnetic ground state (Curie temperature, $T_C = 170$ K) with the *c*-axis magnetization arising from the Co kagome lattice. Cleaving preferentially breaks its S-Sn bond, which leads to the S and Sn terminated surfaces. Previous STM studies have dominantly observed two surfaces, one with largely

vacancy defects and the other with adatom defects^{11,13,14}. Several factors challenge their assignment, as the two surfaces have identical lattice symmetries, their interlayer distance is sub-Å in scale, and STM topographic image convolutes the spatial variation of the integrated local density of states and the geometrical corrugations²³. Decisive experimental evidence for surface identification can be found by imaging the symmetry-dictated surface boundary and the layer-selective chemical dopants^{23–25}. In previous work¹¹, we have determined that the vacancy surface is the Sn layer and the adatom surface is the S layer by imaging their surface boundary determined by the crystalline symmetry. We further conclude this assignment by doping the bulk $\text{Co}_3\text{Sn}_2\text{S}_2$ single crystals with 1% In impurities and imaging the layer-selective In-dopants. The In impurities preferentially replace the Sn atoms according to previous experimental and theoretical studies^{20,21}, as well as our recent systematic single-crystal growth²². Indeed, on the vacancy surface identified¹¹ to be Sn, we observe dilute substitutional atoms with consistent concentration (Fig. 1a), suggesting these impurities to be In atoms.

Spatial feature of single impurity resonance. By performing an extensive study on the electronic properties of the In impurities, we find that each impurity repeatedly features a sharp state at the negative energy as shown in Fig. 1b. First-principles calculations show that each In impurity is almost nonmagnetic but introduces a strong resonance (Fig. 1c), similar to the experimental data. The calculations further reveal that the impurity resonance arises from a spin-down state (opposite to the bulk magnetization direction) and resides in the spin-down bandgap. Thus, the magnetic resonance state likely arises from the local impurity perturbation of the spin-polarized band structure. As the low-energy band structure is dominated by Co 3*d* orbitals, the resonant impurity state of the In atom also implies that there a strong local impact on the Co kagome lattice in real-space. To explore the detailed

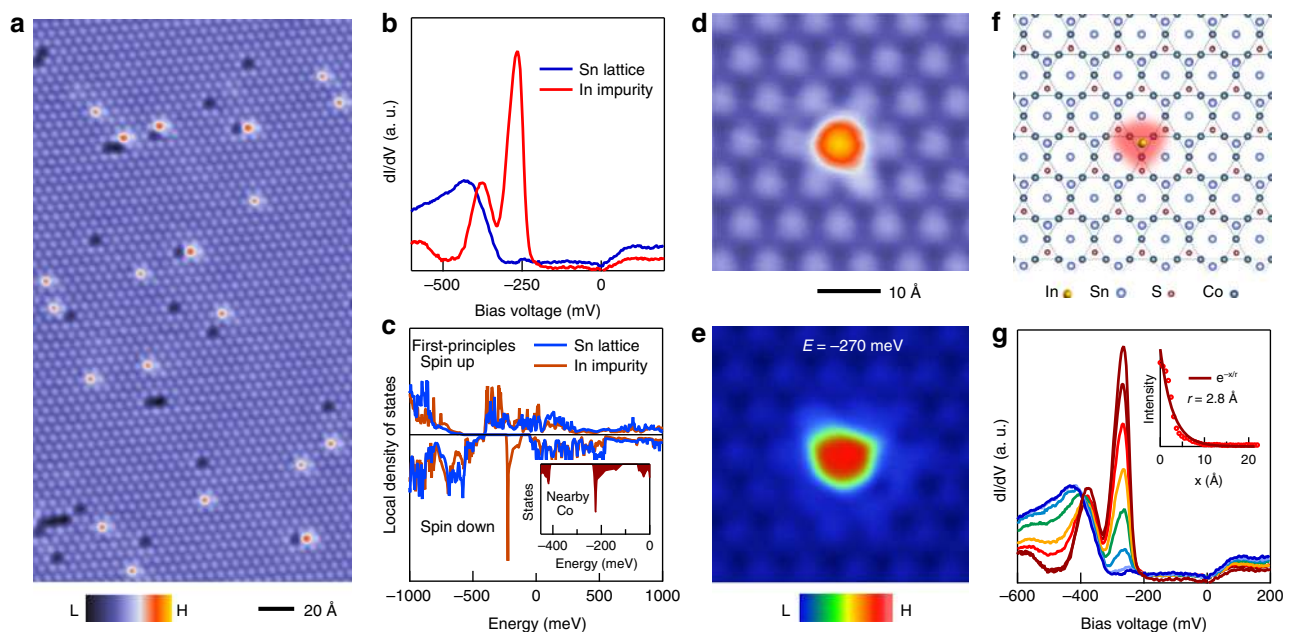


Fig. 1 Engineered atomic impurity state in a topological magnet. **a** Atomically-resolved topographic image of Sn layer of 1% In doped $\text{Co}_3\text{Sn}_2\text{S}_2$. **b** Differential conductance spectrums taken on the Sn lattice (blue) and at the In impurity (red), respectively. **c** First-principles calculation¹³ of the spin-resolved local density of states of the Sn lattice (blue) and an In impurity (red), which shows a magnetic impurity resonance. The inset shows the spin-down states of the Co atom closest to the In impurity. **d** Topographic image of an isolated impurity. **e** Corresponding differential conductance map taken at $E = -270$ meV (resonance energy). **f** Correlation between the atomic structure and the pattern in the differential conductance map. **g** Differential conductance spectra taken across the surface with spatial variation from the center of the In impurity (dark red) to far away (blue). The inset shows the exponential fit to the spatial decay of the impurity resonance.

real-space feature, we probe the local electronic structure for an isolated In impurity, as shown in Fig. 1d. The corresponding dI/dV map at the impurity resonance energy in Fig. 1e shows a localized pattern bound to the impurity site (Fig. 1e). The bound state couples with three nearby Co atoms in the underlying kagome lattice, as illustrated in Fig. 1f. This is consistent with the first-principles calculation that the nearby magnetic Co atoms also feature such resonance state (inset of Fig. 1c), supporting the Co-In coupling (See Supplementary Note 1). Figure 1g shows the representative dI/dV curves measured with increasing distance from the impurity, demonstrating the bound state decaying in intensity without detectable energy dispersion or splitting. An exponential fit to the decay yields a characteristic length scale of 2.8 Å (inset of Fig. 1g). Therefore, these systematic characterizations reveal that the nonmagnetic In impurity couples with the underlying magnetic kagome lattice to introduce a striking localized bound state.

Magnetic nature of single impurity resonance. To probe the magnetic nature of the impurity bound state, we perform tunneling experiments with a spin-polarized Ni tip under weak magnetic fields^{26–29}. The bulk crystal has a coercive field $B_C \sim 0.3$ T, and Ni tip is a soft magnet with a $B_C \ll 0.1$ T that can be easily flipped by reversing the magnetic field²⁹. We measure the tunneling signal of the impurity state while sequentially applying fields along the c -axis of +0.5 T, +0.1 T, –0.1 T, –0.5 T, –0.1 T, and +0.1 T to systematically flip the magnetization of the tip and sample (Fig. 2a). This sequence allows us to perform spin-polarized measurements of the impurity. The +0.5 T field polarizes both the sample and tip, aligning the spin of the tip and anti-aligning the spin of the impurity state, due to the spin-down nature of the impurity state. A +0.1 T field does not change the polarization of either the tip or impurity state. Flipping the field to –0.1 T also flips the spin of the tip, leaving the spin of the impurity state unchanged (down). Here, with both tip and impurity state spins aligned down, we observe an intensity increment of the tunneling signal. Next, we further decrease the field to –0.5 T, which flips up the spin of the impurity state with a corresponding reduction of the tunneling signal. Last, by sequentially changing the field to –0.1 T and +0.1 T, we flip the spin of the tip (down) and again observe an increase in the intensity. Our progressive field manipulation strongly supports that impurity state features spin-down polarization tied to

the bistable magnetic bulk, consistent with the first-principles calculation.

To further determine the effective moment of this magnetic polarized state, we probe the state by applying a strong external magnetic field ($|B| \gg B_C$) along the c -axis with a nonmagnetic tip. Under the field, a spin-up/spin-down band hosting an intrinsic magnetic moment of +1/–1 Bohr magneton will exhibit a Zeeman shift to lower/higher energies in a rate of 0.058 meV T^{-1} . Moreover, when the magnetism of the system is polarized with an applied field, the spin-polarized state will always shift to the same energy direction regardless of the relative field orientation¹¹ (top inset schematic in Fig. 2b), which was also experimentally observed for the 8 T and –8 T data (Fig. 2b). The positive energy shift indicates the state has a negative magnetic moment, calculated to be $-5\mu_B$ (or a Landé g factor of 10) based on a shift rate of 0.275 meV T^{-1} (right inset of Fig. 2b). This large value is beyond the spin Zeeman effects ($\sim 1\mu_B$) and indicates the additional negatively polarized orbital magnetization. The anomalous Zeeman effect with an unusual moment or g factor has been observed in the electronic bands of kagome magnets^{9,11,17}, which is often linked to the Berry phase physics associated with magnetism and spin-orbit coupling^{9,11,30,31}. Since the In impurity couples with the Co atoms in the kagome lattice, the higher orbital angular momentum of the hybridized In-Co orbital can contribute to the large effective moment. We note that the effective moment of $-5\mu_B$ represents the diamagnetic response induced by the In impurity, and should not be thought as the local magnetic moment of the impurity atom.

Interacting impurity states induced quantized orbitals. Having characterized magnetic resonance state from the isolated impurity, we further probe the coupled impurity states to understand how they interact with each other through extensive imaging and spectroscopy investigation with a nonmagnetic tip. In Fig. 3a, we present the evolution of the impurity bound state with increasing perturbation strength caused by a second nearby impurity. We find that with decreasing spatial separation, the bound state progressively decreases in intensity and finally splits into two subpeaks. Figure 3b further compares three cases with one isolated impurity, two neighboring impurities, and three neighboring impurities, respectively. We find the quantized number of split impurity states matches with the coupled impurity number, highlighting their atomic-scale quantum-level

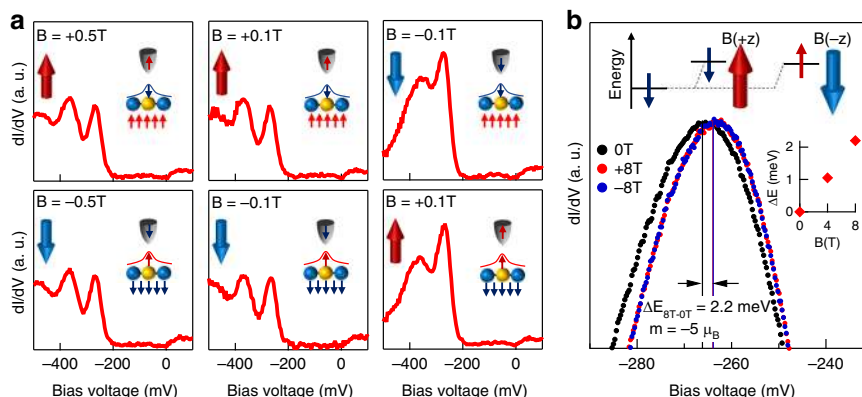


Fig. 2 Magnetic nature of the impurity bound state. **a** Dependence of the impurity state with Ni tip under a weak magnetic field. We apply +0.5 T, +0.1 T, –0.1 T, –0.5 T, –0.1 T, +0.1 T fields to systematically flip the magnetization of the tip and sample. The inset schematics illustrate the respective spins of the tip and the impurity state that is anti-aligned with bulk magnetization direction. **b** Dependence of the impurity state with a strong magnetic field. Under both +8 T and –8 T, the peak exhibits a magnetization-polarized Zeeman energy shift of 2.2 meV, which amounts to an effective moment of $-5\mu_B$. The inset data shows the energy shift for different magnetic field magnitudes. Inset schematic illustrates the magnetization-polarized Zeeman effect. The applied field aligns the spins of the impurity state in the same orientation, hence +z and -z orientation fields lead the energy to shift in the same direction.

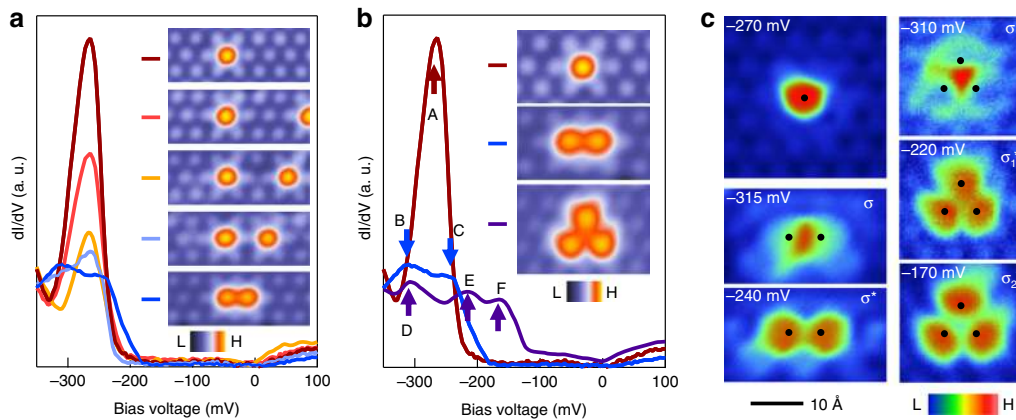


Fig. 3 Interacting impurity states induced quantized orbitals. **a** Differential conductance spectra taken at the central impurity with perturbations of varying strengths from a second impurity. Inset: respective topographic images for impurity configurations. Note the images are acquired by finding different surface locations. **b** Local impurity state with coupling to different numbers of impurities. The arrows highlight the quantized splitting with additional interacting impurity numbers. Note the images are acquired by finding different surface locations. **c** dI/dV maps at the respective bound state energies in **(b)**. dI/dV maps taken at $V = -270$ mV for a single impurity; -315 mV (bonding state σ) and -240 mV (antibonding state σ^*) for a double impurity, respectively; -310 mV (bonding state σ), -220 mV (antibonding state σ_1^*) and -170 mV (antibonding state σ_2^*) for a triple impurity, respectively. The black dots mark the center of impurities.

coupling. Differential conductance maps at these corresponding splitting energies demonstrate their distinct orbital hybridizations (Fig. 3c). For two neighboring impurities, the dI/dV maps show a bonding (σ) and antibonding (σ^*) orbital formation⁴, consistent with the quantum coupling of doubly degenerate states. For three impurities, the dI/dV maps show the formation of one bonding (σ) and two antibonding (σ_1^* , σ_2^*) orbitals, an unusual situation in which we discuss below.

Discussion

Geometrically, the three neighboring impurities have C_{3v} symmetry, which would form a doubly degenerate orbital state⁷ σ^* protected by the mirror symmetry. The mirror symmetry operation, however, would transfer spin-up to spin-down. On the other hand, the magnetic resonances induced by different non-magnetic In impurities are expected to be of the same spin-polarization direction, which is locked by the ferromagnetic ordering of the underlying magnetic Co kagome lattice perturbed by these impurities (See Supplementary Note 2). Therefore, the combination of the same spin-polarization direction of the degenerate impurity states and the atomic spin-orbit coupling naturally breaks the mirror symmetry, leading to the splitting of σ^* (see “Methods” for theoretical modeling). Such splitting is analogous to the splitting of the bulk topological nodal line or magnetic Weyl fermion line^{32,33}, which is protected by the crystalline mirror symmetry.

In conclusion, we report the first STM/S studies of the non-magnetic impurity behavior in a topological magnet. Associated with the atomic nonmagnetic impurities, we find an intense spin-orbit polarized bound state with an unusual magnetic moment and quantized energy splitting under impurity-impurity interaction. The striking spin-orbit quantum states revealed here can advance the understanding of the magnetic and transport behaviors of doped topological magnets, and call for new perspectives on the interplay between magnetism and spin-orbit coupling at the atomic scale. The discrete quantum level of interacting impurities resembles that of a quantum dot, which is critical to nanophotonics and quantum information processing. With the single-atom precision, the atomic quantum dot has a high level of digital fidelity⁷. The interacting spin-orbit polarized quantum

impurity involves multiple degrees of freedom, including charge, spin, and orbital, the quantum control of which in the magnetic bistable platform can provide a useful guideline for the development of spin-orbit entangled quantum technology. Note added in proof: with completion of this work, we became aware of Ref. 29, which reported another kind of impurity resonance in $\text{Co}_3\text{Sn}_2\text{S}_2$ exhibiting negative magnetism as well.

Methods

STM/S measurement. Single crystals²² of $\text{Co}_3\text{S}_{2-x}\text{In}_x\text{Sn}_2$ up to $1.5 \text{ mm} \times 1.5 \text{ mm} \times 0.3 \text{ mm}$ were used in this study. Samples were cleaved mechanically in situ at 77 K in ultra-high vacuum conditions, and then immediately inserted into the STM head, already at He4 base at 4.2 K. The magnetic field was applied under zero-field cooling, after which we carefully approached the tip to locate the same atomic-scale area for tunneling spectroscopy^{11,13}. Tunneling conductance spectra were obtained using standard lock-in amplifier techniques with a root mean square oscillation voltage of 0.2 meV–5 meV and a lock-in frequency of 973 Hz. The topographic images were taken with tunneling junction set up: $V = -300 \sim -500$ mV, $I = 100$ pA, the conductance maps were taken with tunneling junction set up: $V = -100 \sim -500$ mV, $I = 200$ pA, and the tunneling spectra were taken with junction set up: $V = -600$ mV, $I = 300$ pA. Commercial STM Ir/Pt tip (nonmagnetic) and STM Ni tip (soft magnet) tips were used in this study. To study the impurity-impurity interaction, we checked the topographic images for over 3000 impurities to obtain the cases in Fig. 3 in the 1% In doped samples.

Evidence for surface identification. STM studies of $\text{Co}_3\text{Sn}_2\text{S}_2$ often encounter two kinds of surfaces, one dominated by adatom defects and the other dominated by vacancy defects. Here we discuss our evidence for their assignment¹¹ as S surface and Sn surface, respectively. First, as illustrated in Fig. 4a, based on crystalline symmetry, when S and Sn surfaces meet at a step edge, the Sn surface will be just above the S surface. This experimental evidence is directly provided in Fig. 4b, where we observe the vacancy surface is above the adatom surface. Second, this identification provides a natural explanation for the origin of the surface defects. The Sn vacancy in the S surface and Sn adatom on the S surface are simultaneously created due to incomplete cleaving, as seen in the experiment (Fig. 4b). Third, we show that the first-principles calculation of the surface dependent local density of states matches the experimental data (Fig. 4c). Last, we observe In dopant in the vacancy surface in the main paper, and In is known to substitute the Sn atom both experimentally and theoretically^{20–22}.

First-Principles calculations. First-principles calculations were performed in the density functional theory^{34,35} framework as implemented in the Vienna Ab initio Simulation Package^{36,37}. Generalized gradient approximation in Perdew–Burke–Ernzerhof functional³⁸ was applied to describe electron exchange–correlation interaction with the projector augmented wave potentials³⁹. The supercell consists of a periodically repeating 2×2 slab with a thickness of twice the bulk and a vacuum space of ~ 14 Å along the z -direction. The slab is cleaved to reveal the

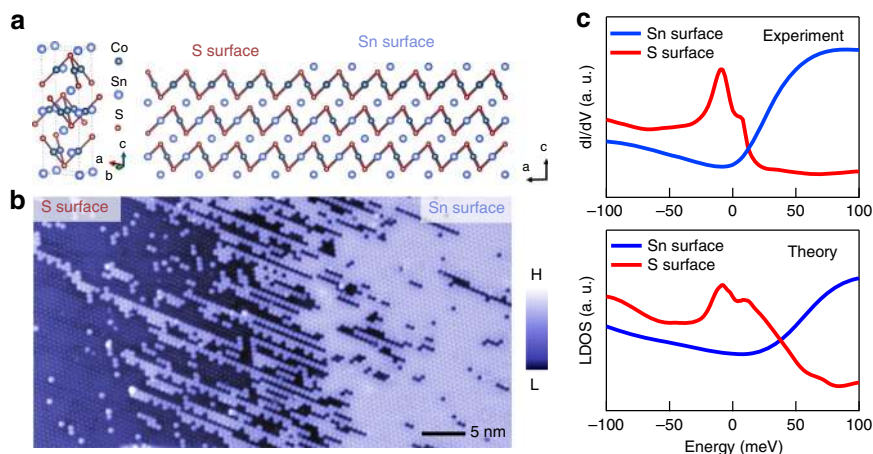


Fig. 4 Extended evidence for surface identification in $\text{Co}_3\text{Sn}_2\text{S}_2$. **a** Crystal structure of $\text{Co}_3\text{Sn}_2\text{S}_2$ (left) and the cleaving surface illustration (right). **b** Atomically resolved topographic image of the boundary between S surface and Sn surface. The S surface smoothly evolves into the Sn surface with increasing coverage of Sn adatom. **c** Comparison between spatially averaged surface dependent dI/dV curves with the first-principles calculations.

Sn-terminating surface, and one surface Sn atom is replaced with In to simulate dilute doping ($\sim 2.3\%$). The energy cutoff was set at 400 eV and the energies in self-consistent calculations were converged until 10^{-4} eV. The Brillouin zone was sampled using a $6 \times 6 \times 1$ Monkhorst-Pack⁴⁰ grid.

Data availability

All relevant data are available from the corresponding authors upon reasonable request.

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Author contributions

J.-X.Y., N.S., Y.J., and S.S.Z. conducted the scanning tunneling microscopy experiments in consultation with M.Z.H.; H.Z., S.J., Q.W., and H.Lei synthesized and characterized the magnetic and transport properties of samples; G.M., H.O.M.S., G.C., B.M.A., Z.-Q.H., F.-C.C., H.Lin, and Z.W. carried out the theoretical analysis in consultation with J.-X.Y. and M.Z.H.; Z.C., Z.G., Y.L., M.L., I.B., X.Y., Q.Z., and T.A.C. contributed to the calibration of the measurement; J.-X.Y., N.S., Y.J., and M.Z.H. performed the data analysis and figure development and wrote the paper with contributions from all

authors; M.Z.H. supervised the project. All authors discussed the results, interpretation, and conclusion.

Competing interests

The authors declare no competing interests.

Additional information

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