Supporting information for: Probing the superfluid response of
para-hydrogen with a sulfur dioxide dopant

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FIG. S1. Potential energy surfaces. (a) SO\textsubscript{2}-pH\textsubscript{2} angular potential energy surface (PES) at \( r = 6.38 \) \( a_0 \), the intermolecular distance with potential minimum of \(-114.90\) cm\(^{-1}\). The symbol “S” marks the saddle point on the contour plot of the PES. The yellow dashed circle marks the low-energy trough of the PES; (b) H\textsubscript{2}O-pH\textsubscript{2} angular potential energy surface at \( r = 6.35 \) \( a_0 \), the intermolecular distance with potential minimum of \(-97.56\) cm\(^{-1}\). Due to the \( C_2v \) symmetry of the SO\textsubscript{2} and H\textsubscript{2}O, only the first quadrants, i.e. \( \chi \in [0^\circ,90^\circ] \), of the potential energy surfaces are plotted. The angular coordinates are defined beside the respective PESs. The axes are also labelled following the \( abc \) convention of the principal-axis frame.
FIG. S2. Density isosurface of $p\text{H}_2$ of some $\text{SO}_2(p\text{H}_2)_N$ clusters in the $\text{SO}_2$ MFF. In (a), (b), and (c), the solid surface represents the density values of 30% of the maximum density of each cluster, while the transparent represents the density value of 7%. In (d), the value of the surface is 45% of the highest density of the cluster. Note that the two lobes of density distribution in $\text{SO}_2(p\text{H}_2)_1$ originate from the $C_{2v}$ symmetry of $\text{SO}_2$.

FIG. S3. SFF and MFF superfluid fractions of $^4\text{He}$ particles in CO($^4\text{He})_N$ clusters. The MFF superfluid fractions are calculated along an axis perpendicular to the CO axis.
SII. TECHNICAL DETAILS OF THE PIMC SIMULATIONS OF SO$_2$(pH$_2$)$_N$ CLUSTERS

We employ the path-integral Monte Carlo (PIMC) method$^1$ to simulate SO$_2$(pH$_2$)$_N$ clusters. The Hamiltonian operator for this system is

$$\hat{H} = \hat{T}_{\text{SO}_2} + \hat{R}_{\text{SO}_2} + \sum_{i=1}^{N} \hat{T}_{\text{pH}_2}^i + \sum_{i<j}^{N} \hat{V}_{\text{pH}_2-\text{pH}_2}(r_{ij}) + \hat{V}_{\text{SO}_2-\text{pH}_2}(\tilde{r}_i, \Omega),$$

(1)

where $\hat{T}$ and $\hat{R}$ stand for translational and rotational kinetic energy operators for the subscript species, $\hat{V}$ potential energy operator for the subscript pairs, $r_{ij}$ and $\tilde{r}_i$ distances between the $i$th and $j$th pH$_2$ particles and between the $i$th pH$_2$ and SO$_2$, and $\Omega$ the three Euler angles of SO$_2$. Pairwise additive potential is used. The pH$_2$pH$_2$ interaction is described by the isotropic version of the H$_2$-H$_2$ interaction potential developed by Patkowski and co-workers$^2$. The SO$_2$pH$_2$ potential is introduced in the next section. The worm algorithm was used to sample bosonic exchange$^3$. The approach of Noya and co-worker was used to construct the asymmetric top rotor PIMC density matrix$^4,5$. We restrict the rotational states of SO$_2$ to be symmetric upon the exchange of the two $^{16}$O atoms. The number of translational and rotational imaginary time slices were chosen to be 1024 and 256. With these numbers of slices, we obtained the energy of $-74.91 \pm 0.03$ K for the SO$_2$pH$_2$ dimer at $T = 0.37$ K, compared to the value of $-74.64$ K from our benchmark calculation with the discrete variable representation (DVR) basis.$^6$ All PIMC simulations were conducted for the low temperature of 0.37 K, the typical temperature for pH$_2$ to exhibit superfluidity.$^7$ The total number of Monte Carlo steps for each simulation was of the order of $10^8$ to achieve satisfactory error bars.

The usage of different numbers of translational and rotational imaginary time slices was briefly introduced in Refs. 8 and 9. We here give more details of this method using a simplest example, a rotor in an external field. Let us assume that the rotation of the rotor is less quantum than its translation and the imaginary time interval $\tau$ is short enough to have converged Trotter factorization approximation$^{10,11}$ for the rotational propagator. The translation, however, needs half shorter imaginary time interval $\frac{\tau}{2}$. Our derivation starts from the primitive PIMC propagator

$$\rho^\tau(r, \Omega; r', \Omega') = \langle r | e^{\tau\hat{V}(\Omega)} e^{-\tau\hat{T}} e^{-\tau\hat{R}} | r' \rangle = \langle r | e^{-\tau\hat{V}(\Omega)} e^{-\tau\hat{T}} | r' \rangle \langle \Omega | e^{-\tau\hat{R}} | \Omega' \rangle.$$  

(2)
Here we use \( r \) and \( \Omega \) to label the centre of mass position and orientation of the rotor, and \( \hat{V}, \hat{R}, \) and \( \hat{T} \) for its potential, rotational, and translational kinetic operators. Due to the locality of \( \hat{V} \), \( e^{-\tau\hat{V}} \) can directly act on the bra \( \langle \Omega | \) and result in an operator \( e^{-\tau\hat{V}(\Omega)} \) that only acts on the \( r \) space. To improve the accuracy of the translational propagator in the first bracket, we can further split it into two propagators with imaginary time interval \( \frac{\tau}{2} \):

\[
\rho^\tau (r, \Omega; r', \Omega') = \langle r | e^{-\tau\hat{V}(\Omega)} e^{-\tau\hat{T}} | r' \rangle \langle \Omega | e^{-\tau\hat{R}} | \Omega' \rangle
\]

\[
= \langle r | e^{-\frac{\tau}{2}\hat{V}(\Omega)} e^{-\frac{\tau}{2}\hat{V}(\Omega)} e^{-\frac{\tau}{2}\hat{T}} e^{-\frac{\tau}{2}\hat{T}} | r' \rangle \langle \Omega | e^{-\tau\hat{R}} | \Omega' \rangle
\]

\[
\approx \langle r | e^{-\frac{\tau}{2}\hat{V}(\Omega)} e^{-\frac{\tau}{2}\hat{T}} e^{-\frac{\tau}{2}\hat{V}(\Omega)} e^{-\frac{\tau}{2}\hat{T}} | r' \rangle \langle \Omega | e^{-\tau\hat{R}} | \Omega' \rangle
\]

\[
= \int d\rho'' \langle r | e^{-\frac{\tau}{2}\hat{V}(\rho)} e^{-\frac{\tau}{2}\hat{T}} | r'' \rangle \langle r'' | e^{-\frac{\tau}{2}\hat{V}(\rho)} e^{-\frac{\tau}{2}\hat{T}} | r' \rangle \langle \Omega | e^{-\tau\hat{R}} | \Omega' \rangle
\]

\[
= \int d\rho'' e^{-\frac{\tau}{2}V(r,\rho)} \langle r | e^{-\frac{\tau}{2}\hat{T}} | r'' \rangle e^{-\frac{\tau}{2}V(r',\rho)} \langle r'' | e^{-\frac{\tau}{2}\hat{T}} | r' \rangle \langle \Omega | e^{-\tau\hat{R}} | \Omega' \rangle. \quad (3)
\]

This newly inserted integral is to represent the resolution of identity in the \( r \) space. It can be performed through Monte Carlo sampling of \( \rho'' \). With this propagator, a closed path would have the number of translational slices twice of that of rotational. Centre of mass coordinates in the pair of adjacent translational slices \( 2k - 1 \) and \( 2k \) share the same orientation in the rotational slice \( k \) in calculating their respective potential propagators. This derivation can be straightforwardly generalized to more complicated situations.

**SIII. THE SO\(_2\)-PH\(_2\) POTENTIAL**

Our SO\(_2\)-PH\(_2\) potential is based on the 5-D potential from Spielfiedel et al.\(^{12}\) We followed exactly the same scheme in obtaining the H\(_2\)O-pH\(_2\) 3-D potential\(^{13}\) in Fig. S1(b) to obtain the more economic SO\(_2\)-pH\(_2\) 3-D potential, with pH\(_2\) being treated as a point-like particle. Since SO\(_2\) is a slower rotor than H\(_2\)O, this adiabatic-hindered-rotor approximation should work even better. This 3-D potential is employed in our PIMC simulations for SO\(_2\)(pH\(_2\))\(_N\) clusters. Fig. S1(a) shows two unsurmountable high peaks in the PES in the symmetrically unique azimuthal angle (\( \chi \)) region. These barriers will pushed the surrounding pH\(_2\) particles to follow the rotation of SO\(_2\). The minimum of the SO\(_2\)-pH\(_2\) PES has a depth of \(-114.90\) cm\(^{-1}\), deeper than that (-97.56 cm\(^{-1}\)) of the H\(_2\)O-pH\(_2\) PES. This suggests that despite the formidable repulsive peaks, several pH\(_2\) particles can still be attached firmly to SO\(_2\). These firstly attached pH\(_2\) particles will then attract more pH\(_2\) particles based on the
$pH_2-pH_2$ interaction. After all, the anisotropy of the SO$_2-pH_2$ PES is evident, especially when it is compared with the flat H$_2$O-$pH_2$ PES in Fig. S1(b).

**SIV. CALCULATION OF EFFECTIVE ROTATIONAL CONSTANTS**

The formula to calculate the effective rotational constant along the $a$-axis is

$$A_{\text{eff}} = \frac{\hbar^2}{2I_{\text{eff}}} = \frac{\hbar^2}{2 \left( I_{SO_2}^{SO_2} + I_{a}^{pH_2} \right)} = \frac{\hbar^2}{2 \left( I_{SO_2}^{SO_2} + I_{cl}^{pH_2} (1 - f_a^s) \right)}. \quad (4)$$

The other rotational constants are calculated using the same formula. If $f_a^s$ is close but not equal to unity, then a substantial $I_{cl}^{pH_2}$ can still make a non-negligible contribution to the denominator and suppress $A_{\text{eff}}$ to be lower than the $A$ of a free rotor. This explains the coexistence of the close-to-one $f_a^{s,B,E}$ and substantially suppressed $A_{\text{eff}}^{B,E}$ for SO$_2(pH_2)_{12-16}$ in Figs. 1(a) and 3(a). A turnaround increase of the effective rotational constant with the number of bosons around usually requires a substantial increase of superfluid fraction. This is because the added bosons usually monotonically increase the classical moment of inertia and only an increase in the superfluid fraction can make the denominator smaller. This is reflected by the turnaround of $A_{\text{eff}}^{B,E}$ from $N = 3$ to 4 in Fig. 2(a) and the sharp peak of $f_s^{a,B,E}$ in Fig. 1(a).

**SV. TECHNICAL DETAILS OF THE PIMC SIMULATIONS OF CO$(^4$He)$_N$ CLUSTERS**

The Hamiltonian operator for the simulations is similar to Eq. (1), with CO being the rotor and $^4$He being the boson. PES from Chuaqui et al.$^{14}$ is employed. Temperature is chosen to be 0.5 K. 512 and 256 translational and rotational beads are used.