Hydrogen bonds in Al$_2$O$_3$ as dissipative two-level systems in superconducting qubits

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Dissipative two-level systems (TLS) have been a long-standing problem in glassy solids over the last fifty years, and have recently gained new relevance as sources of decoherence in quantum computing. Resonant absorption by TLSs in the dielectric poses a serious limitation to the performance of superconducting qubits; however, the microscopic nature of these systems has yet to be established. Based on first-principles calculations, we propose that hydrogen impurities in Al$_2$O$_3$ are the main source of TLS resonant absorption. Hydrogen is an ubiquitous impurity and can easily incorporate in Al$_2$O$_3$. We find that interstitial H in Al$_2$O$_3$ forms a hydrogen bond (O-H...O). At specific O-O distances, consistent with bond lengths found in amorphous Al$_2$O$_3$ or near Al$_2$O$_3$ surfaces or interfaces, the H atom feels a double well. Tunneling between two symmetric positions gives rise to resonant absorption in the range of 10 GHz, explaining the experimental observations. We also calculate the expected qubit-TLS coupling and find it to lie between 16 and 20 MHz, consistent with experimental measurements.

The existence of dissipative two-level systems (TLSs), typically observed in amorphous solids, is a long-standing problem in solid-state physics, but specific microscopic models have been lacking. A new impetus for uncovering their origins has emerged with the advance of qubits based on superconducting Josephson tunnel junctions. Dissipation in the qubit causes the excited $|1\rangle$ state to decay to the $|0\rangle$ ground state, leading to decoherence. Two-level systems (TLS) in the insulating layer have been reported to be a major source of energy loss; this mechanism seems to dominate other sources of decoherence.

Dielectric loss from TLS can be large in amorphous materials (see Pohl et al. for a detailed survey), and is thought to arise from random bonding of atoms. In superconducting qubits, TLSs are a source of decoherence in the tunnel barrier of the Josephson junction. Loss arises from absorption of microwave radiation by TLSs with an electric dipole moment. It can be modeled as atoms tunneling between two distinct positions, and has been shown to be important even for the surface oxide of superconducting metals (Al$_2$O$_3$). The effect of decoherence can be mitigated by the use of single-crystal Al$_2$O$_3$; a reduction in the density of spectral splittings of up to 80% has been observed. The defects have resonance frequencies on the order of 10 GHz, comparable to the qubit circuit; the coupling strengths and decoherence times are sufficiently large for coherent oscillations between the qubit and TLS. Despite extensive studies on the physics and effects of TLSs, their microscopic origin has remained unsettled.

Here we show that TLSs in Al$_2$O$_3$ can be attributed to hydrogen impurities that incorporate on interstitial sites. Hydrogen is an ubiquitous impurity, present in many growth and processing environments, and able to unintentionally incorporate in most materials, including the Al$_2$O$_3$ dielectric used in superconducting qubits. The specific characteristics of hydrogen that give rise to the TLS are related to its propensity for hydrogen bonding; by definition it is the only element that exhibits this type of chemical bond. Interstitial hydrogen in oxides exhibits a strong, mainly covalent bond with a primary O atom, with a typical O-H bond length of $\sim$1 Å, but can also interact with a next-nearest-neighbor (nnn) O atom, resulting in an O-H...O configuration. For suitable O-O distances, the interaction with the nnn O atom leads to quantum-mechanical tunneling between these neighboring sites. Tunneling of interstitial hydrogen between adjacent O sites has been observed in oxides such as KTaO$_3$, for which Spahr et al. reported tunneling rates in the 7–40 GHz range. We will demonstrate, based on first-principles theory, that hydrogen in Al$_2$O$_3$ can give rise to TLSs with tunneling frequencies that explain the dielectric loss observed in superconducting qubits.

The process of quantum-mechanical tunneling of interstitial hydrogen is complicated by the strong interactions of hydrogen with the lattice, which lead to relaxations of the host. In its most stable configuration, the
interstitial H is primarily bonded to one O atom, with a larger distance to the \( nnn \) O (Fig. 1a). In this atomic configuration, the metastable minimum at the \( nnn \) O is significantly higher in energy, or even nonexistent, when compared to the ground-state minimum. To enable tunneling, the lattice must be brought into a symmetric coincidence configuration \(^{17,18}\) (Fig. 1b) where a double-well potential occurs and tunneling can take place (Fig. 1c). The three-dimensional (3D) potential-energy surface corresponding to the coincidence configuration determines the energy levels and tunnel splittings for the quantum-mechanical motion of the H atom. These energy levels and tunneling frequencies can be calculated by numerically solving Schrödinger’s equation. The determination of the coincidence configuration and the corresponding potential energy surface is an important aspect of the present work, discussed in detail below. We note that considering a purely adiabatic process, in which the host atoms assume their lowest-energy positions for each location of the H atom, is unrealistic due to the large mass difference between H and the other atoms. On the other hand, fixing the host atoms to positions corresponding to an unrelaxed lattice, or to those for the ground-state configuration of interstitial H, is unrealistic as well. We address this by employing a novel interpolation scheme between the ground state-minima at two adjacent O atoms, and we repeat this for different O-O distances in order to explore the properties of H in a wide range of possible coincidence configurations.

**Results**

Interstitial H in \( \alpha-Al_2O_3 \) can assume three different charge states: positive (\( H^+ \)), neutral (\( H^0 \)), and negative (\( H^- \)). These configurations are calculated within density functional theory (DFT) using a screened hybrid functional (see Methods). The use of a hybrid functional is absolutely essential for properly describing the interaction between H and the \( nnn \) O atom, and thus the tunneling frequencies of the associated TLS. DFT within the standard generalized gradient approximation (GGA) overestimates the strength of the interaction between H and the \( nnn \) O atom so that the calculated O-H frequency is too low\(^{19}\). For the neutral and negative charge states, the interstitial hydrogen occupies a position in between two Al atoms, as shown in Fig. 2a.b. \( H^+ \), on the other hand, bonds to an O atom with a bond length of 1.01 Å, and a distance of 1.70 Å to the \( nnn \) O (Fig. 1c, 3b). The relative stability of the different charge states depends on the Fermi-level position. The formation energy of an interstitial H in charge state \( q [E^f (H^q)] \) is calculated as \(^{20}\)

\[
E^f (H^q) = E_{tot} (H^q) - E_{tot}(Al_2O_3) - \mu_H + q\epsilon_F,
\]

where \( E_{tot} (H^q) \) is the total energy of the supercell containing an interstitial H atom in charge state \( q \), \( E_{tot}(Al_2O_3) \) is the total energy of the bulk supercell, \( \mu_H \) is the hydrogen chemical potential, and \( \epsilon_F \) is the Fermi level, referenced to the valence-band maximum (VBM). The value of \( \mu_H \) does not affect the relative stability of the different configurations, and we can set \( \mu_H \) equal to half the total energy of an isolated \( H_2 \) molecule.

The calculated formation energies as a function of \( \epsilon_F \) are shown in Fig. 3. We find that the donor state, \( H^+ \), is the stable charge state for Fermi levels up to 5.9 eV above the VBM; above that, the acceptor state, \( H^- \), is most stable. The neutral charge state, \( H^0 \), is always higher in energy than both the \( H^+ \) and \( H^- \), reflecting a negative-U character, as observed for H\(_i\) in many other semiconductors and insulators\(^{13}\). The position of the Fermi level is determined by charge neutrality; in an undoped \( \alpha-Al_2O_3 \) crystal, native defects lead to a Fermi-level position around the middle of the gap\(^{21}\). Under these conditions, Fig. 3 shows that interstitial hydrogen is most stable in the positive charge state, \( H^+ \).

The relaxed geometry of the \( H^+ \) configuration in \( Al_2O_3 \) at its equilibrium volume is asymmetric, with significant relaxations of the host atoms, particularly of the O atom to which it is bonded (Fig. 2c). The energy difference between the relaxed \( Al_2O_3 \) lattice containing an \( H^+ \) and the relaxed lattice with \( H^+ \) in its most stable configuration is defined as the self-trapping energy\(^{9,22,23}\), and is approximately 1.5 eV. One would expect this large energy difference to prohibit H from tunneling. In order to enable tunneling, a configuration must be created in which the hydrogen is equally likely to be bonded to either of the two neighboring O atoms. The lowest energy required to take a self-trapped configuration into a symmetric structure, i.e., the coincidence configuration, is the “coincidence energy” \( E_c \). The formation of a coincidence geometry may be assisted by lattice vibrations, or may occur in regions where the atomic arrangement deviates from that in the bulk crystal, such as near surfaces or interfaces, or in an amorphous phase, in which a range of O-O distances from around 2.4 Å to 2.8 Å are observed\(^{24–27}\).

An exact determination of the coincidence geometry would in principle require a self-consistent treatment of the quantum-mechanical hydrogen motion coupled to the host-atom relaxation\(^{28}\), something that is too computationally demanding to be performed in conjunction with a first-principles treatment of the electronic structure. Approximate methodologies for obtaining the coincidence geometry have therefore been developed. Here we determine the coincidence configuration by averaging the positions of the host atoms for two self-trapped configurations, corresponding to H bonded to adjacent O sites. This procedure produces a symmetric double well potential for the tunneling hydrogen atom. We then place a H atom (\( H^+ \)) in the structure formed by the host atoms in the coincidence geometry and allow the hydrogen to relax (keeping the host atoms fixed). The energy difference between the relaxed \( H^+ \) in the coincidence geometry and the \( H^+ \) in the most stable, self-trapped configuration is the coincidence energy \( E_c \).

We determined the coincidence configurations and coincidence energies for a range of different volumes of \( \alpha-Al_2O_3 \) from equilibrium to 6% isotopic (linear) strain. This variation in lattice parameters corresponds to O-O distances between 2.715 to 2.59 Å in the perfect crystal, a range which includes the O-O distances found in amorphous \( Al_2O_3 \)\(^{22–25}\). The corresponding O-O distance in the coincidence configuration varies in the range of 2.56 to 2.45 Å. The results are listed in Table 1 and shown in Fig. 4. The coincidence energies
vary from 0.36 to 0.25 eV, with a minimum at 4% compression, corresponding to an O-O distance (in the coincidence geometry) of 2.49 Å.

The solution of Schrödinger’s equation for the H atom in the 3D potential energy surface corresponding to the coincidence geometry produces wave functions and energies characteristic of a double-well potential. The tunnel splitting is calculated from the energy difference between the ground state and the first excited state, and leads to the tunneling frequencies listed in Table 1. In order to enable direct comparison to experiments, we have also calculated the qubit-TLS coupling strength for each possible configuration. The measured signal of a TLS in a phase qubit reflects the resonance between the qubit frequency (5.4 GHz, ref. 27) and for the capacitance and gate width, resulting in values between 16 and 20 MHz (Table 1).

**Discussion**

The results in Table 1 indicate that the tunneling frequencies are sensitive to the O-O distance, albeit not in a straightforward manner. Compression of the lattice initially makes little difference to the tunneling frequency, which remains around the value of 50 GHz calculated for the equilibrium volume (see Fig. 4). The higher values of $E_\text{c}$ (compared to those calculated at compressed volumes) suppress the possibility of tunneling in this regime. At 4.5% compression, corresponding to an O-O distance of 2.59 Å in the impurity-free crystal, the O-O distance in the coincidence configuration is 2.48 Å and the tunneling frequency is 16 GHz. Further compression of the lattice leads to a decrease in the potential barrier between the wells in the double well potential, and thus an increase in the tunneling frequency, away from the range observed in experiment. Such small O-O distances also have a decreased probability of occurring in actual samples.

Experimentally, a range of TLS tunneling frequencies have been reported, centered around 10 GHz$^{-28}$. To further corroborate our model, we can also compare our calculated qubit-TLS coupling constants $S_{\text{Max}}$ to experiment. Reported values range from 25 MHz to 45 MHz$^{-28}$. The agreement with our calculated values (16–20 MHz, Table 1) further validates our claim that hydrogen is responsible for TLS decoherence in the dielectric layer.

The presence of hydrogen in dielectrics is highly plausible, particularly when they are deposited or grown using hydrogen-containing precursors. Hydrogen has been observed with secondary ion mass spectrometry (SIMS) in atomic-layer-deposited oxides$^{30}$. Unfortunately, accurate experimental detection of hydrogen is fraught with difficulty, at least at the low concentrations and in the small volumes that are relevant for qubits. A decrease in loss has been reported in the case of crystalline dielectrics$^{36}$. This is consistent with our results for crystalline $\text{x-Al}_2\text{O}_3$ at equilibrium volume, in which the tunneling frequencies are well outside the range that is relevant for the qubits. However, even in devices that use crystalline dielectrics, there is still a range of O-O distances at surfaces and interfaces$^{11}$, so TLS losses are never completely eliminated.

The results reported here are distinct from recent work by Holder et al.$^{12}$, also based on hybrid density functional theory, which considered the rotor motion of a H atom in $\text{x-Al}_2\text{O}_3$, either as an interstitial or in the Al vacancy. They concluded that interstitial hydrogen could not be responsible for TLSs, based on a value for the calculated tunneling frequency that was much higher than experiment. They did find one defect displaying frequencies in the GHz range, namely a complex of H with an Al vacancy in the $+1$ charge state. However, this complex is stable only if the Fermi level is within 1.2 eV of the VBM, which is highly unlikely (essentially impossible) in $\text{x-Al}_2\text{O}_3$, a wide-band-gap insulator. The frequency calculated for $H_{i+}^+$ in Ref. 12 ($\sim$240 GHz) is significantly higher than the values reported here, likely stemming from their highly simplified 1D model. Our own tests have indicated that a full mapping of the 3D potential energy.
surface is essential in Al₂O₃, and that 1D models do not match the full results. In addition, the sixfold or threefold degeneracies assumed in the rotor model of Ref. 12 are extremely unlikely to occur in real materials—even in crystalline solids. In contrast, the two-fold degeneracy inherent in our hydrogen-related double-well systems is highly plausible and fits within the general framework of two-level systems.

In summary, we have proposed a microscopic model for TLSs in Al₂O₃ based on interstitial hydrogen in an O-H...O hydrogen-bonding configuration. Hybrid DFT calculations produce frequencies that are in the range reported for TLSs known to be responsible for the main loss in superconducting qubits, and calculated qubit-TLS coupling parameters are close to experimental values. We suggest that hydrogen could be responsible for TLSs in other materials as well, given its ubiquity. As the only element to exhibit “hydrogen bonding”, it stands out as a candidate for TLSs in oxides, in which a range of suitable O-O distances occur. The low migration barriers observed (and calculated) for interstitial H⁺ suggest that barriers between equivalent sites can be low enough to lead to significant tunneling and hence double-well systems.

### Methods

Our calculations are based on density functional theory (DFT)

with a characteristic screening length of 10 Å. In the short-range region, the exchange potential of Perdew, Burke and Ernzerhof (PBE)

The impurity calculations were performed using a supercell containing 120 atoms; this is a $2^p \times 2^q \times 2^r$ multiplier of the 30-atom unit cell of $\alpha$-Al₂O₃. We used a $2^p \times 2^q \times 2^r$ Monkhorst-Pack k-point mesh for the integrations over the Brillouin zone, and a cutoff of 500 eV for the plane-wave basis set. Our HSE calculations for $\alpha$-Al₂O₃ produce lattice parameters $a = b = 4.74$ Å and $c = 12.95$ Å, in very good agreement with the experimental values, $a = b = 4.76$ Å and $c = 12.99$ Å.

We study a variety of O-O interatomic distances (representative of those occurring in the amorphous phase or near surfaces or interfaces) by varying the volume of the crystalline solid, up to a total volumetric compression of 6%. The physics of the TLS studied here is determined by the local environment of the hydrogen atom, for which the presence (or absence) of long-range order is irrelevant. Our results for crystalline $\alpha$-Al₂O₃ therefore also apply to amorphous Al₂O₃, in which the relevant O-O interatomic distances occur, as evidenced by pair-correlation distributions.

The 3D potential energy surface for the H atom in the host lattice (fixed to the coincidence configuration) was determined by calculating the total energy for each point on a grid with a spacing of 0.24 Å in three spatial directions. This grid was then interpolated using the energy gradients obtained from the forces acting on the H atom, leading to a smooth function. To numerically solve the Schrödinger’s equation for the quantum-mechanical motion of H, the wave functions were expanded in a plane-wave basis set with a cutoff energy of 800 eV; this cutoff was determined based on convergence tests for the tunnel splittings.

The qubit-TLS coupling $S_{\text{Max}}$ can be calculated as:

$$ S_{\text{Max}} = \frac{2\pi}{\hbar} \sqrt{2 \epsilon M_0 2C}, $$

### Table 1 | Calculated TLS parameters for H in Al₂O₃.

Coincidence energies $E_c$, tunnel splittings $\Delta$, tunneling frequencies $\nu$, and coupling constants $S_{\text{Max}}$ for two-level systems associated with interstitial hydrogen in $\alpha$-Al₂O₃. Values are listed for various O-O distances $d_{O-O}$ ranging from 2.56 Å to 2.45 Å, corresponding to volume compression by the specified amounts.

| Compression | $d_{O-O}$ (Å) | $E_c$ (eV) | $\Delta$ (eV) | $\nu$ (GHz) | $S_{\text{Max}}$ (MHz) |
|-------------|---------------|------------|--------------|-------------|---------------------|
| 0%          | 2.56          | 0.36       | $2.20 \times 10^{-4}$ | 53          | 19.8               |
| 1%          | 2.55          | 0.35       | $2.09 \times 10^{-4}$ | 50          | 18.3               |
| 2%          | 2.52          | 0.33       | $2.48 \times 10^{-4}$ | 59          | 19.2               |
| 3%          | 2.51          | 0.32       | $2.17 \times 10^{-4}$ | 53          | 17.1               |
| 3.5%        | 2.50          | 0.29       | $1.38 \times 10^{-4}$ | 33          | 16.7               |
| 4%          | 2.49          | 0.25       | $1.07 \times 10^{-4}$ | 26          | 16.4               |
| 4.5%        | 2.48          | 0.28       | $6.7 \times 10^{-5}$  | 16          | 16.4               |
| 5%          | 2.47          | 0.28       | $4.12 \times 10^{-4}$ | 90          | 16.3               |
| 6%          | 2.45          | 0.26       | $1.57 \times 10^{-3}$ | 360         | 16.0               |

**Figure 4** | Tunneling frequency as a function of O-O distance. Calculated tunneling frequencies and coincidence energies are shown for O-O distances ranging from 2.45 to 2.56 Å in the coincidence geometry. Tunneling frequencies are shown in red, coincidence energies in blue.
where \( x \) is the barrier thickness, \( p \) is the effective dipole moment of the charge in the double-well system, \( C \) is the capacitance, and \( E_{\text{ab}} \) is the qubit splitting energy. We assume a junction width \( x = 2 \text{ nm} \), a capacitance \( C = 850 \text{ fF} \), and a qubit splitting energy 5.4 GHz, corresponding to representative experimental values\(^{22,23}\). The dipole moment is calculated as the product of the effective charge around the hydrogen atom in the coincidence configuration and the distance between the symmetric potential wells, determined from first-principles calculations.

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Author contributions
L.G., H.A.F. and C.G.V.d.W. designed and performed all research. L.G., A.J. and C.G.V.d.W. wrote the main manuscript text.

Additional information
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