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Method for assessing atomic sources of flicker noise in superconducting qubits

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Flicker noise causes decoherence in Josephson junction-based superconducting qubits, thus limiting their practical potential as building blocks for quantum computers. This is due to limited length and complexity of executable algorithms, and increased dependency on error-correcting measures. Therefore, identifying and subsiding the atomic sources of flicker noise are of great importance to the development of this technology. We developed a method that combines ab initio DFT calculations and quantum dynamics to model charge transport across a Josephson junction, by which it is possible to more accurately assess different defects as sources of flicker noise. We demonstrate the use of our method in an investigation of various atomic defects, including vacancies, trapping, and substitutions, in an Al|Al₂O₃|Al Josephson junction. This demonstration both reveals weaknesses in previous attempts to pinpoint the atomic sources of flicker noise and highlights new candidates.

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INTRODUCTION

Quantum computers (QC) are a topic for theoretical research for nearly 40 years, but only in the last decade, advancement in fabrication and design processes resulted in limited but functional QC processors. One such type of promising design is based on superconducting materials used for the fabrication of electrical components called Josephson junctions (JJ). Unfortunately, this type of qubits, as with most other types, suffers from short coherence time, which limits the performance and calculations these processors can execute. The coherence time is limited, not only but also, by an internal phenomenon called flicker noise, which can be mathematically explained by using two-level system (TLS) defects as “black box” objects. TLS are localized low-energy excitations that are widely believed to be the source of many anomalies in the properties of JJ. These TLSs can arise from defects in the material, and their collective behavior manifests itself as low-frequency flicker noise^{1–4}.

Understanding the source of flicker noise can have a tremendous contribution to the development of QC via hardware improvement, and not just by efforts of developing software solutions such as error-correction codes. Indeed, efforts were put into measuring flicker noise and modeling it phenomenologically⁵, as well as using density functional theory (DFT) simulations to identify the sources of the noise^{6–9}. But despite all of the efforts mentioned above, there is still neither clear understanding nor a widely accepted source of flicker noise in superconducting JJs. In these studies, DFT calculations were made on particular defects, including adsorbed O₂ molecules, but there has been no consistent comparison between different defect types. Furthermore, previous studies focus on noise originating from the surface as there is a large amount of surface on the metal–oxide capping layer of the entire circuit. There are theoretical^{6–9} and experimental^{10–12} ways to account for surface-related noise, but less so for the interior of JJ and new learnings can be achieved for interior-related noise that may act as a limiting factor.

Motivated to solve this conundrum, we developed a method, which allows to assess candidates of flicker noise under a single

framework. The foundations of our method rely on DFT calculations using Vienna Ab initio Simulation Program (VASP) as the first step, and quantum dynamics of wave-packet propagation through a JJ for charge-transport simulation as the second step. This method can have great impact on this research field by formalizing the search for the source, increasing consistency between different research groups using this method, and allowing the elimination of suspected sources by a more complete set of tools. Also, there are other efforts to study the current response in atomic models of JJ^{13–16}, which focuses on a specific type of defect or disorder at a time happening in the oxide barrier.

In this paper, we present our method through a demonstration focused on Al|Al₂O₃|Al-based JJ because of their prevalence in the fabrication of superconducting-based qubits^{17,18}, and in the supplementary information, we include more technical details alongside the scripts. We also include a discussion about the results emerging from this demonstration, show how and why previous work arrived at imprecise conclusions, and point out a new type of defects as candidates for the source of flicker noise.

The general idea of our method is to construct a defect-free structure serving as reference JJ and to characterize its structural and electrical properties. On the reference JJ, one can implement different types of defects, such as vacancies, adsorption, or substitution. This allows to examine the defects' impact on the JJ properties, and even to extract the noise's Fourier components and to compare them with flicker noise shape of 1/*f* at low frequency.

RESULTS

From bulk to interface structure

For the construction of our defect-free reference JJ structure, denoted as JJ_{ref} from this point onward, we used bulk Al and α -Al₂O₃ as building blocks. As initial geometry before structural optimization we used a single FCC unit cell with lattice parameter $a = 4.05\text{\AA}$ ¹⁹ for the Al metal caps, and for the α -Al₂O₃, we used a six-layered hexagonal unit cell with lattice

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Fig. 1 Post-relaxation reference structure, with red and gray atoms are O and Al respectively, and yellow cross marks interface Al atoms. **a** Side view showing epitaxial growth. **b** Top view showing parallel lattice directions of the metal cap and oxide.

parameters of $a = b = 4.759\text{\AA}$ and $c = 12.991\text{\AA}$ ²⁰. Previous work^{21–23} showed that the most stable oxide termination at the interface is with O atoms, the Al–O bond length is $1.76\text{\AA}$ at the interface, and the preferred orientation for epitaxial continuation at the interface is $\text{Al}(111)[101]||\text{Al}_2\text{O}_3(0001)[1000]$. Figure 1 shows the calculated geometry of a JJ_{ref} from both side and top views.

The DFT calculation also yields the potential energy that the charge carriers need to cross through in a JJ, which served as the potential energy element in the charge-transport Hamiltonian, as will be explained later. Figure 2 shows the one-dimensional Kohn–Sham potential curve of JJ_{ref} along the z axis.

Defect implementation

The defects we introduced to JJ_{ref} in our demonstration can be divided into three main categories: vacancy defects at strategic locations in the vicinity of the interface, trapping defects of an atom or a molecule in penetration sites near the interface, and substitution defect of one atom by another. We chose those defects based on two criteria: either there exists previous theoretical work that found those defects as plausible explanation for flicker noise^{6,7}, or there is a high probability for these defects to form as part of the fabrication process, mainly shadow evaporation and Dolan bridge techniques^{8,9,24,25}. The defects were implemented on the relaxed JJ_{ref} structure, and their initial geometry before a second relaxation is shown in Fig. 3.

This part alone was sufficient to show that previous attempts⁷ at explaining flicker noise reached incomplete conclusions, since they focused on cleaved oxide rather than a complete JJ structure. Our method shows that adsorbed O_2 molecules are not stable as free rotors when the metal cap is also taken into consideration, rather, they tend to dissociate in favor of local epitaxial growth of the oxide.

We recognize the apparent limitation that JJ_{ref} has dimensions much smaller than an actual JJ, but this is reconciled by the fact that we were able to converge our results for different system sizes. For future research, there are no technical limitations, aside from time and compute power, to extend the model to any arbitrary size. In addition to that, we used an entirely crystalline oxide as the JJ barrier, which is an ideal state compared with the usually amorphous oxide grown in the fabrication step. The crystalline and amorphous barriers may yield different behavior of charge transport, yet our method can handle amorphous layers with no special requirements or changes needed as long as an amorphous layer can be constructed and modeled^{8,26–28}.

As long as one can build the unit cells and generate the relevant information required for the algorithm to run using DFT/DFT + U/HSE/etc. for superconductors, our method that includes subsequent dynamical calculations is general enough and transferable to other materials.

Fig. 2 The potential for the charge carriers along the z axis in the reference JJ $\text{Al}/\text{Al}_2\text{O}_3/\text{Al}$ structure. The vertical dashed black lines indicate the interfaces locations.

Phonon-vibration modes

Phonon-vibration modes (PVM) play a crucial role in the formation of Cooper pairs (CP), as described by Bardeen–Cooper–Schrieffer (BCS) theory. Therefore, our method also includes the calculation of each defect's PVM, which encompass information about the participating atoms, the displacement magnitude along the cartesian axes, and the vibration frequency. This information is then used to add time dependency to the charge-transport Hamiltonian in the following steps.

Figure 4 shows a comparison of the calculated PVM spectra for all of the models shown in Fig. 3 while focusing on frequencies at the interface close to the defect. We see that the PVM frequencies are spread uniformly across the range of frequencies even when considering PVM resulting from atoms far from the interface. Figure 4 shows also three outlying frequencies marked by the red rectangle, which belong to the trapped H atom and OH molecule models. We used the free software wxDragon²⁹ to animate these PVM in order to understand their origin, and found that these elements resulted from the H atom vibrating in its interstitial volume. This finding was the first indication that this type of defect leads to a different behavior and characteristics of JJ.

Charge transport

In order to study the noise characteristics that each defect generates, we simulated current flowing through the JJ. In our demonstration, we apply several approximations to simplify the calculations without impairing the usefulness of our modeling environment. The approximations used were:

1. Using the one-dimensional Kohn–Sham potential curve solved for fermions by DFT, and not the standard CP potential as a function of the phase difference. This was required since DFT is usually not implemented for modeling bosonic particles and can be further justified by the fact that CP is not the only charge carrier that can conduct current. On the other hand, this approximation helps simplify the

Fig. 3 The atomic models investigated in our demonstration, where the purple ellipses indicate the defect location. **a** Reference structure. **b** Al vacancy in the metal cap. **c** Al vacancy in the oxide. **d** O vacancy in the oxide, **e** trapped H, **f** trapped OH, **g** trapped O_2 , and **h** Al for O substitution.

structure of the Hamiltonian while ignoring conduction parallel to the interface³⁰.

- Using a Gaussian wavefunction with initial momentum and spatial width to model the CP particle. This was inspired by the CP characteristic coherence length as described by Pippard's relation^{31–33}.
- Using PVM to vibrate the potential curve as a mean to add time dependency to the Hamiltonian³⁴.

The charge-transport calculations were executed according to the algorithm described by the flowchart shown in Fig. 5. This algorithm is based on our earlier method for calculating charge

transport through functional interfaces^{35,36} and goes beyond our previous methodology by the inclusion of the phonon vibrations in the potential energy operator, through adding time-dependent vibrations to our Hamiltonian.

The inputs to the first step came from our DFT calculations and consisted of the potential curve, PVM, and Fermi energy. The second step included initialization of the Gaussian wavefunction of the form

$$|\psi(t=0)\rangle = \frac{1}{A} e^{-ik_z z} e^{-\frac{(z-z_0)^2}{2\sigma_0^2}} \quad (1)$$

Fig. 4 PVM frequencies in descending order with outlying frequencies marked in red rectangle. Models (a–h) in the legend coincide with the order presented in Fig. 3.

Fig. 5 Schematic representation of the algorithm used for calculating charge transport through a JJ. The algorithm collects inputs about the modeling environment, then it initializes the wavefunction. Following that, a loop will run to calculate the current system state and propagate the system in space and time according to the Hamiltonian dynamics.

Here z_0 and k_f are the initial position of the Gaussian center and the wavefunction's momentum along the z axis respectively, σ_0 is the spatial width of the Gaussian, and A is a normalization factor. The third step was to vibrate the potential curve by expanding and contracting it along the chemical bonds' projection on the z axis, while keeping the potential energy

values fixed, according to:

$$z_i(\tau) = z_i(0) + \sum_{PVM} \text{Mag}_j \sin(2\pi f_j \tau) \sin\left(\frac{2\pi}{\lambda_j} z\right) \quad (2)$$

Here z_i is the location of the i^{th} discrete-potential curve data point, and Mag_j , f_j , and λ_j are the j^{th} PVM magnitude, frequency, and wavelength, respectively. The fourth step was to use the vibrated potential curve as the potential energy term in the time-dependent Schrödinger equation, and to calculate the probability current density through the interface. The fifth step was to advance the system by a time step using the second-order split-operator method^{37,38}. The updated wavefunction is then fed back as a loop to the third step until halt. Figure 6 shows an example for the initial and final wavefunction's states in the charge-transport calculations.

We integrated over the calculated probability current densities to get the probability current, which, according to Ginzburg–Landau theory, is proportional to the actual current flow through the JJ by a scaling factor equal to a CP charge³⁹. Figure 7 shows the overlay of the calculated probability currents through the interface as a function of time. Figure 7 also shows that model (c) probability current decreases after about 2.5 s. The reason for that, and with greater detail in the supplementary information, is the dramatic change in the shape of the potential curve, which resulted in backward reflection through the interface counted with a negative sign for directionality.

With the calculated currents through the interfaces of the different structures in hand, we turned to extract the noise addition to the reference signal. This was achieved by subtracting the baseline JJ_{ref} current from the other defected models leaving only the deviations from the baseline, i.e., the noise.

With respect to the halting criteria mentioned above, in our study we chose the timestep at which the probability current through JJ_{ref} reached a saturation.

Noise power spectral density

For the extracted noise, we calculated the power spectral density (PSD) shown as the overlay in Fig. 8 as a function of the Fourier frequency components.

With the calculated PSD we were able to check the fit to flicker noise characteristic shape of $S_x(f) \propto \frac{1}{f}^5$, i.e., $\beta = 1$ in the general definition of noise bucketing $S_x(f) \propto f^{-\beta}$. Table 1 summarizes the fit coefficients and their match to flicker noise definition, where we chose to use the percentage of deviation and R^2 as the statistical measures to quantify the match of each model. This calculation shows that the presence of H atoms in interstitial sites generate the noise with best fit to flicker noise, compared with the rest of the defects in our demonstration, positioning this type of defect as a valid candidate to be the atomic source of flicker noise.

DISCUSSION

To summarize, the purpose of this paper is to showcase the use of our method for assessing atomic sources of flicker noise in JJ-based superconducting qubits. Our method takes a bottom-to-top approach to assess flicker noise sources, i.e., start from the basic building blocks of the atomic structure, from them, construct a full JJ that is then used to investigate the structural and electrical characteristics of these models. By using our proposed method, it is possible to narrow down suspected sources for flicker noise by means of structural and electrical characterization of a JJ, as well as by analyzing the noise characteristics arising from charge-transport modeling through the JJ.

The demonstration of application of our method on a $\text{Al}|\text{Al}_2\text{O}_3|$ Al-based JJ showed the strengths of our method twofold. The first comes from the fact that our method considers a complete JJ structure rather than just a cleaved oxide interface. This is of great

Fig. 6 Charge transport across a JJ. The black curve is the potential curve, and the red and blue curves are the real and absolute value of the wavefunction. **a** Initial state. **b** Final state after reaching a halt.

Fig. 7 Probability current through the JJ interface as a function of time. The calculations end after reaching the halting criteria. Models (a–h) in the legend coincide with the order presented in Fig. 3.

Fig. 8 Current noise PSD focused on the low-frequency regime that corresponds with flicker noise. Models (b–h) in the legend coincide with the order presented in Fig. 3.

Table 1. Summary of β fit coefficients and deviation from flicker noise. Models (a)–(h) coincide with the order presented in Fig. 3.

Model	B fit	Deviation (%)	R^2
(b)	1.213	21.27	0.958
(c)	0.902	−9.79	0.972
(d)	0.908	−9.22	0.939
(e)	1.054	5.39	0.987
(f)	0.990	−1.03	0.989
(g)	1.086	8.56	0.902
(h)	0.911	−8.85	0.926

importance since it allows to learn about the true stability of a defect. As a result of this, we were able to show that previous results about the nature of adsorbed O_2 molecules were inaccurate. Instead of behaving as a free rotor on a cleaved oxide interface inducing magnetic flux noise, when considering the metal caps as well the defect's stability changes, and an oxide growth is rather preferred, making this type of defect less likely to continue existing and generate flicker noise.

The second comes from the fact it is a more robust way to identify new candidates being the atomic source of flicker noise. We were able to show that the presence of trapped H atoms, in the form of a single atom or OH molecule, in interstitial sites in the vicinity of the interface, generate noise with Fourier component with very good match to the $1/f$ characteristic shape of flicker noise.

We believe that using our method will advance the search for the atomic sources of flicker noise, promoting the development of this technology.

On top of that, by choosing to implement our method as a standard for future research, there will be opportunities for implementing the method to other systems and opening the door for other research directions in the field.

DATA AVAILABILITY

The authors declare that the data supporting the findings of this study are available within the paper and its supplementary information files.

CODE AVAILABILITY

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AUTHOR CONTRIBUTIONS

M.C.T. supervised the project. A.R. performed the calculations. Both authors contributed to writing the paper.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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