

# Supplementary Information for Applications of Quantum Computing for Investigations of Electronic Transitions in Phenylsulfonyl-carbazole TADF Emitters

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## Supplementary methods

Detailed procedure for the practical application of quantum state tomography for VQD simulations on an IBM Quantum device:

(i) Compute the ground state  $|\Psi_0\rangle$  by VQE to minimize the cost function

$$C_0(\theta) = \langle \Psi(\theta) | H | \Psi(\theta) \rangle \quad (1)$$

with respect to the parametrized Ansätze state

$$|\Psi(\theta)\rangle = U(\theta) |0\rangle \quad (2)$$

where  $U(\theta)$  is a unitary transformation with parameter  $\theta$ . Ideally, this procedure produces the purified ground state

$$|\Psi_0\rangle = |\Psi(\theta_*)\rangle = U(\theta_*) |0\rangle \quad (3)$$

In practice, the readout error mitigation technique can be used to compute  $C_0(\theta)$  and thereby find  $\theta_*$ .

(ii) The ground state obtained in step (i) is then used to find the first excited state with VQD by minimizing the following modified cost function:

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$$\langle \Psi(\theta) | H | \Psi(\theta) \rangle + \beta_0 |\langle \Psi(\theta) | \Psi_0 \rangle|^2 \quad (4)$$

with a suitably chosen weighting parameter,  $\beta_0$  as described in ref. <sup>6</sup>. The second term denotes a state that minimizes the energy under the constraint that  $|\Psi(\theta)\rangle$  is orthogonal to the reference state  $|\Psi(\theta_*)\rangle$ . Ideally, the minimizer of  $C_1(\theta)$  is the 1st excited state. On the other hand, the final state of a quantum operation  $U(\theta_*)$  implemented on current noisy quantum devices, must be a mixed state  $\rho_0$ , rather than the pure state  $|\Psi(\theta_*)\rangle$ , and thus the following alternative cost function can be considered:

$$\langle \Psi(\theta) | H | \Psi(\theta) \rangle + \beta_1 \langle \Psi(\theta) | \rho_0 | \Psi(\theta) \rangle \quad (5)$$

where  $\rho_0$  is found via state tomography (without readout error mitigation). Note that instead of  $\rho_0$ , we have defined the cost function in this manuscript by using the largest eigenvector of  $\rho_0$ , denoted by  $|\Psi'_0\rangle$ , such that the cost function is given by

$C'_1(\theta) = \langle \Psi(\theta) | H | \Psi(\theta) \rangle + \beta_0 |\langle \Psi(\theta) | \Psi'_0 \rangle|^2$  The (approximate) first excited state is then given by the minimized  $C'_1(\theta)$ . The procedure adopted for the investigations described in this manuscript have compared the minimizers of  $C_1(\theta)$  and  $C'_1(\theta)$ .

(iii) The second excited state is computed on a quantum device by utilizing a process similar to that used for the ground state. First, the final state of the mixed first excited state that minimizes  $C'_1(\theta)$  is purified by constructing the density matrix via state tomography. The largest eigenvector of this matrix, denoted by  $|\Psi'_1\rangle$ , is used to minimize the cost function under the constraint that  $|\Psi(\theta)\rangle$  is orthogonal to two reference states  $|\Psi'_0\rangle$  and  $|\Psi'_1\rangle$ .

$$C'_2(\theta) = \langle \Psi(\theta) | H | \Psi(\theta) \rangle + \beta_0 |\langle \Psi(\theta) | \Psi'_0 \rangle|^2 + \beta_1 |\langle \Psi(\theta) | \Psi'_1 \rangle|^2 \quad (6)$$

This procedure can then be repeated to compute higher excited states.

## Supplementary Tables

**Supplementary Table 1.** Energy deviations, in *mHa*, for simulations performed on a quantum device in comparison to FCI.

### Ground state energy deviations for VQE simulations in comparison to FCI

| <i>TADF emitter</i> | <i>unmitigated</i> | <i>readout mitigated</i> | <i>state tomography mitigated</i> |
|---------------------|--------------------|--------------------------|-----------------------------------|
| PSPCz               | 13                 | 6                        | 1                                 |
| 2F-PSPCz            | 12                 | 3                        | 1                                 |
| 4F-PSPCz            | 11                 | 2                        | 1                                 |

### Excited state energy deviations for qEOM-VQE simulations in comparison to FCI

| <i>TADF emitter</i> | <i>State</i>   | <i>VQE: unmitigated/<br/>qEOM: unmitigated</i> | <i>VQE: readout mitigated/<br/>qEOM: unmitigated</i> | <i>VQE: readout mitigated/<br/>qEOM: readout mitigated</i> | <i>VQE: state tomography mitigated/<br/>qEOM: readout mitigated</i> |
|---------------------|----------------|------------------------------------------------|------------------------------------------------------|------------------------------------------------------------|---------------------------------------------------------------------|
| PSPCz               | T <sub>1</sub> | 3                                              | 3                                                    | -1                                                         | 0                                                                   |
|                     | S <sub>1</sub> | 1                                              | 3                                                    | 1                                                          | -1                                                                  |
| 2F-PSPCz            | T <sub>1</sub> | 5                                              | 5                                                    | -2                                                         | 0                                                                   |
|                     | S <sub>1</sub> | 3                                              | 3                                                    | -2                                                         | 1                                                                   |
| 4F-PSPCz            | T <sub>1</sub> | 1                                              | -2                                                   | -5                                                         | 0                                                                   |
|                     | S <sub>1</sub> | 17                                             | 19                                                   | 1                                                          | 3                                                                   |

### Excited state energy deviations for VQD simulations in comparison to FCI

| <i>TADF emitter</i> | <i>State</i>   | <i>overlap using the unpurified reference state/<br/>unmitigated molecular Hamiltonian</i> | <i>overlap using purified reference state from state tomography/<br/>unmitigated molecular Hamiltonian</i> | <i>overlap using purified reference state from state tomography/<br/>mitigated molecular Hamiltonian</i> |
|---------------------|----------------|--------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------|
| PSPCz               | T <sub>1</sub> | 21                                                                                         | 8                                                                                                          | 0                                                                                                        |
|                     | S <sub>1</sub> | -11                                                                                        | -6                                                                                                         | 1                                                                                                        |
| 2F-PSPCz            | T <sub>1</sub> | 23                                                                                         | 8                                                                                                          | 1                                                                                                        |
|                     | S <sub>1</sub> | 88                                                                                         | 12                                                                                                         | 2                                                                                                        |
| 4F-PSPCz            | T <sub>1</sub> | 79                                                                                         | 2                                                                                                          | -1                                                                                                       |
|                     | S <sub>1</sub> | 82                                                                                         | 15                                                                                                         | 4                                                                                                        |

**Supplementary Table 2.** Coefficients of the Pauli terms in the molecular Hamiltonian for PSPCz, 2F-PSPCz, 4F-PSPCZ described in the section II (Methods) of the main text.

| Coefficients of the Pauli terms | PSPCz     | 2F-PSPCz  | 4F-PSPCz  |
|---------------------------------|-----------|-----------|-----------|
| $h_{II}$                        | -0.518418 | -0.553646 | -0.540009 |
| $h_{ZI}$                        | -0.136555 | -0.126735 | -0.181261 |
| $h_{ZZ}$                        | -0.025866 | -0.034240 | -0.083888 |
| $h_{XI}$                        | -0.000296 | 0.012806  | 0.000951  |
| $h_{XX}$                        | 0.015725  | 0.011196  | 0.000107  |