Simulating Quantum Drude Oscillators on a photonic quantum computer

Matthieu Sarkis*
Department of Physics and Materials Science
University of Luxembourg, L-1511,
Luxembourg City, Luxembourg.

†

(Dated: March 14, 2023)

I. INTRODUCTION

Dispersion forces, also known as van der Waals (vdW) forces originate from the electromagnetic interaction between electrically neutral atoms or molecules which do not have permanent electric moments [1–4]. They are ever-present long-range forces between atoms or molecules arising from the zero-point fluctuations of the quantum electromagnetic field [5–9]. Their importance can be appreciated on both macro- and micro-scales; for instance, the first macroscopic signature of dispersion forces is the well-known correction to the equation of state of an ideal gas that led to the van der Waals equation [10]. Moreover, dispersion forces also influence the structure of liquids and solids such as the anomalies of water [11] as well as the macroscopic properties of macromolecules such as their structure [12], stability [13, 14], dynamics [15–17], and electric [18] and optical [19] responses. The most natural framework for the investigation of the response of matter subjected to such forces is quantum electrodynamics [20–24]. However, as widely shown in the literature, the inclusion of vdW dispersion interactions can be done by means of many-body methods [25–32]. Dispersion vdW interactions are often represented within the Lennard-Jones approach, namely, through a pairwise two-body interatomic potential [33–38] of the form C_6/R^6 (where R is the interatomic distance and C_6 a system-dependent constant). Among all the existing models in the literature, the many-body dispersion (MBD) framework has been undoubtedly proved to be an accurate approach [37, 39]. In the MBD framework, the drudonic response of valence drudons in atoms and molecules is supposed to be linear and this can be formally done through the introduction of the quantum Drude oscillator (QDO). A single QDO is coarse-grained quantum-mechanical model in which the properties of an atom are encompassed in a small number of parameters. The model consists in assimilating the atom to a point particle of mass m and electric charge $-q$ attached to a fixed (infinite mass) center of charge $+q$ by a harmonic spring characterized by a frequency ω. Molecules are then defined as a collection of QDOs in dipole-dipole interaction. For some specific choices of the matter system geometry, the quantum Hamiltonian can be exactly diagonalized. For instance, in a closed linear chain of molecules, one can analytically solve for the spectrum of the Hamiltonion [40]. In the case of a general geometry, instead, one can solve for the London-van der Waals interaction energy through a perturbative approach [41]. This simple model has been extensively used in various contexts, for instance in order to tackle the drudonic structure problem for isolated molecules, in particular long range interactions, as well as to study the impact of an ambient bath or of an external electric field on molecular properties [42, 43]. Though simple, through a numerical treatment this system was shown to capture long-range phenomena in large biomolecular systems [44]. By construction, the MBD framework relies on the dipole approximation of the drudon–drudon Coulomb interaction leaving aside any contribution coming from high-order terms. In the literature, dispersion forces have been addressed mostly for atomic dimers and small systems, via multipolar generalizations of the pairwise second-order perturbative approaches [33, 34, 38, 45]. In this Letter ... They did full coulomb FCI in [46]

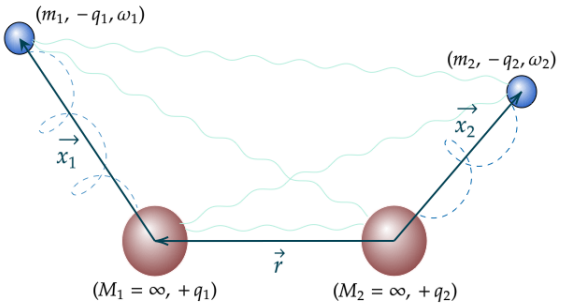

FIG. 1. a

* matthieu.sarkis@uni.lu
†

II. DEFINITION OF THE MODEL

A. Three-dimensional model

The Hamiltonian describing a system of N Quantum Drude Oscillators in three dimensions is given by:

$$H = \sum_{i=1}^{N} \left[\frac{\boldsymbol{p}_i^2}{2m_i} + \frac{1}{2} m_i \omega_i^2 \boldsymbol{x}_i^2 \right] + \sum_{i<j} V_{\mathrm{Coul}}\left(\boldsymbol{x}_i, \boldsymbol{x}_j\right) , \quad (1)$$

with the Coulomb interaction receiving contributions from interacting drudon-drudon and drudon-nucleus pairs:

$$\begin{aligned} \frac{V_{\mathrm{Coul}}\left(\boldsymbol{x}_i, \boldsymbol{x}_j\right)}{q_i q_j} = \frac{1}{r_{ij}} - \frac{1}{|\boldsymbol{r}_{ij} + \boldsymbol{x}_i|} - \frac{1}{|\boldsymbol{r}_{ij} - \boldsymbol{x}_j|} \\ + \frac{1}{|\boldsymbol{r}_{ij} - \boldsymbol{x}_j + \boldsymbol{x}_i|} . \end{aligned} \quad (2)$$

The usual approach consists in solving the theory in the multipolar expansion framework, in which the potential can be expressed as a power series in the inverse distance separating the two centers:

$$V_{\mathrm{Coul}}\left(\boldsymbol{x}_i, \boldsymbol{x}_j\right) = \sum_{n \geq 0} V_n\left(\boldsymbol{x}_i, \boldsymbol{x}_j\right) , \quad (3)$$

with the following scaling behavior in terms of the distance between the centers:

$$V_n\left(\boldsymbol{x}_i, \boldsymbol{x}_j\right) \propto r_{ij}^{-n-3} . \quad (4)$$

The potential V_0 corresponds then to the dipole-dipole interaction, and is at the core of the Many Body Dispersion (MBD) model. V_1 corresponds to the dipole-quadrupole interaction, and V_2 to the quadrupole-quadrupole and dipole-octupole interaction.

One obvious limitation of the multipolar expansion is the lower bound it imposes on the interatomic distance. One can easily see that within the MBD (dipole-dipole) model by direct diagonalization of the quadratic Hamiltonian in terms of normal modes. In that case, one of the normal modes (the center of mass mode) develops a purely imaginary frequency at short range. Higher order physical effects are also neglected in the MBD model, motivating the study of the QDO model with full Coulomb interaction potential between its constituents.

Let us define the following dimensionless position and momententum operators associated to QDO i:

$$\boldsymbol{X}_i := \sqrt{\frac{m_i \omega_i}{\hbar}}\, \boldsymbol{x}_i , \qquad \boldsymbol{P}_i := \frac{\boldsymbol{p}_i}{\sqrt{2\hbar m_i \omega_i}} , \quad (5)$$

in terms of which the Hamiltonian reads

$$\begin{aligned} H = \sum_{i=1}^{N} \frac{\hbar \omega_i}{2} \left(\boldsymbol{X}_i^2 + \boldsymbol{P}_i^2 \right) \\ + \sum_{i<j} V_{\mathrm{Coul}} \left(\sqrt{\frac{\hbar}{m_i \omega_i}} \boldsymbol{X}_i, \sqrt{\frac{\hbar}{m_j \omega_j}} \boldsymbol{X}_j \right) \end{aligned} \quad (6)$$

One can define the $3N$ creation and annihilation operators

$$\boldsymbol{a}_i = \frac{\boldsymbol{X}_i + i\boldsymbol{P}_i}{\sqrt{2}} , \qquad \boldsymbol{a}_i^\dagger = \frac{\boldsymbol{X}_i - i\boldsymbol{P}_i}{\sqrt{2}} , \quad (7)$$

in terms of which the Hamiltonian reads

$$\begin{aligned} H = \sum_{i=1}^{N} \frac{\hbar \omega_i}{2} \left(\boldsymbol{a}_i^\dagger \cdot \boldsymbol{a}_i + \frac{3}{2} \right) \\ + \sum_{i<j} V_{\mathrm{Coul}} \left(\sqrt{\frac{\hbar}{m_i \omega_i}} \frac{\boldsymbol{a}_i + \boldsymbol{a}_i^\dagger}{\sqrt{2}}, \sqrt{\frac{\hbar}{m_j \omega_j}} \frac{\boldsymbol{a}_j + \boldsymbol{a}_j^\dagger}{\sqrt{2}} \right) . \end{aligned} \quad (8)$$

Let us from now on restrict the problem to a pair of QDOs ($N = 2$) for concreteness, though the following developements carry to bigger systems. We denote by d the interatomic distance.

B. One-dimensional case

In order to reduce the complexity of the problem, let us define one-dimensional instances of the QDO model as follows: we restrict the movement of the two drudons to be along a common axis (directed by a unit vector $\hat{\boldsymbol{e}}_\theta$) which form an angle $\theta \in [0, \pi/2]$ with respect to the vector $\boldsymbol{r}_{12}$ connecting the two nuclei. We therefore have a family of one-dimensional models which can be obtained from the full-fledged 3d model simply by setting to zero the contribution from the oscillator modes belonging to the plane perpendicular to $\hat{\boldsymbol{e}}_\theta$. Let us denote by (X, P) the remaining position and momentum degree of freedoms. As limiting cases, we obtain models in which the drudons are constrained to move either in the direction parallel to the axis separating the two nuclei ($\theta = 0$), or perpendicular to the latter ($\theta = \pi/2$). Those two models were studied in [47] in the dipole-dipole approximation. In that paper, the authors encode the states in the truncated Fock space of the system into the state of a set of qubits, and run VQE-type algorithms on IBQ quantum processors. However as we will see, the angle θ captures the competition between *existence of binding* (for small θ) and *smoothness* (for large θ), and interesting one-dimensional model actually sit at values of θ in the open segment $(0, \pi/2)$.

In the case of a generic angle θ and interatomic distance, the one-dimensional Coulomb potential reads:

$$\begin{aligned} \frac{V_{\mathrm{Coul}}^{\theta,d}(x_1, x_2)}{q_1 q_2} = \frac{1}{d} - \frac{1}{\sqrt{d^2 + 2d(\cos\theta) x_1 + x_1^2}} \\ - \frac{1}{\sqrt{d^2 - 2d(\cos\theta) x_2 + x_2^2}} \\ + \frac{1}{\sqrt{d^2 - 2d(\cos\theta)(x_2 - x_1) + (x_2 - x_1)^2}} . \end{aligned} \quad (9)$$

The model we consider is therefore given by

$$H = \frac{\hbar\omega_1}{2}\left(P_1^2 + X_1^2\right) + \frac{\hbar\omega_2}{2}\left(P_1^2 + X_1^2\right) + V_{\text{Coul}}^{\theta}\left(\sqrt{\frac{\hbar}{m_1\omega_1}}X_1, \sqrt{\frac{\hbar}{m_2\omega_2}}X_2\right). \tag{10}$$

For the simulation, we set $\hbar = 1$ as well as $m_i = q_i = \omega_i = 1$ for both QDOs.

III. PHOTONIC CIRCUIT

The circuit implements a unitary $U(\theta)$ acting on an input reference state (the Fock vacuum for instance) that we simply take to be the vacuum state $|0\rangle$. The state prepared by the circuit is therefore given by

$$|\psi(\theta)\rangle = U(\theta)|0\rangle\,. \tag{11}$$

In the dipolar approximation, namely for the Hamiltonians $H_{(1,1)}$, $H_{(1,2)}$ and $H_{(1,3)}$, we expect that using a Gaussian state would be enough. The circuit is therefore composed of at most quadratic optical components (squeezing operations for our ansatz). However for the other models, non-Gaussian operation should be added in the end of each layer in the ansatz circuit.

One the ansatz state $|\psi(\theta)\rangle$ has been produced, one should extract the value of the energy in that state, namely compute the value of

$$\langle\psi(\theta)|H|\psi(\theta)\rangle \tag{12}$$

To be specific, let us take the model H, and let us denote by angular brackets the expectation in state $|\psi(\theta)\rangle$. One has

$$\langle H\rangle = \sum_{i=1}^{N}\frac{\hbar\omega_i}{2}\left(\langle n_{i,x}\rangle + \langle n_{i,y}\rangle + \langle n_{i,z}\rangle + \frac{3}{2}\right) + \sum_{i<j}\left\langle V_{\text{Coul}}\left(\sqrt{\frac{\hbar}{m_i\omega_i}}\boldsymbol{X}_i, \sqrt{\frac{\hbar}{m_j\omega_j}}\boldsymbol{X}_j\right)\right\rangle \tag{13}$$

by linearity of the expectation. On the second line one has to compute something of the form $\langle f(X_{i,\alpha})\rangle$, where $(X_{i,\alpha})$ denotes collectively the position quadrature of all the photon channels (including QDO and spatial index). One therefore needs to extract the statistics of quadratures by preparing and measuring the state $|\psi(\theta)\rangle$ in the quadrature basis. Once the joint density ρ of $(X_{i,\alpha})$ in the state $|\psi(\theta)\rangle$ is known, one can compute

$$\langle f(X_{i,\alpha})\rangle = \int_{\mathbb{R}^{3N}} f(x_{i,\alpha})\rho(x_{i,\alpha})\prod_{i,\alpha}\mathrm{d}x_{i,\alpha}\,. \tag{14}$$

There is a complication related to the fact that strawberryfields does not allow to access the joint law of the position quadratures. We therefore have to extract it by ourselves. Let us first suppose that there are K photon modes. The statevector is represented in the Fock basis as follows:

$$|\psi\rangle = \sum_{n_1,\dots,n_{2K}=0}^{\infty}\alpha_{n_1\dots n_{2K}}|n_1\rangle\otimes\cdots\otimes|n_{2K}\rangle\,. \tag{15}$$

The modes labeled by $(n_1,\dots,n_K)$ correspond to QDO_1, while the modes $(n_{K+1},\dots,n_{2K})$ are attached to QDO_2. The amplitude of a specific tuple of the quadratures $(X_1,\dots,X_K)$ is therefore given by:

$$\langle X_1,\dots,X_{2K}|\psi\rangle = \sum_{n_1,\dots,n_{2K}=0}^{\infty}\alpha_{n_1\dots n_{2K}}\prod_{i=1}^{2K}\frac{e^{-\sum_{i=1}^{2K}\frac{X_i^2}{2}}H_{n_i}(X_i)}{\sqrt{\pi^{1/2}2^{n_i}n_i!}}\,,$$

in terms of the Hermite polynomials. The joint law of the quadratures in the state $|\psi\rangle$ is therefore given by

$$\rho(X_1,\dots,X_{2K}) = \sum_{\substack{n_1,\dots,n_{2K}\\ m_1,\dots,m_{2K}}}\alpha_{n_1\dots n_{2K}}\alpha^*_{m_1\dots m_{2K}} \times\prod_{i=1}^{2K}\frac{e^{-X_i^2}H_{n_i}(X_i)H_{m_i}(X_i)}{\sqrt{\pi^{1/2}2^{n_i}n_i!}\sqrt{\pi^{1/2}2^{m_i}m_i!}} \tag{16}$$

After extracting as well the mean photon numbers $\langle n_{i,\alpha}\rangle$, one obtains $\langle H\rangle$.

IV. VARIATIONAL ALGORITHM

We define the following loss function:

$$\mathcal{C}(\theta) := \langle\psi(\theta)|H|\psi(\theta)\rangle \tag{17}$$

In order to compute this loss, one therefore has to measure both the photon number operator on each channel, as well as the position quadrature operator on each channel, as described in the previous section.

Algorithm 1: Extract distribution of observable

Parameters: statevector $|\psi\rangle$, observable $\mathcal{O}$, shots $M \in \mathbb{N}$
Result: Probability distribution of $\mathcal{O}$ in state $|\psi\rangle$

for $m = 1$ *to* $\dim(spec(\mathcal{O}))$ **do**
  Initialize $N_m \leftarrow 0$;
for $j = 1$ *to* M **do**
  Measure the state $|\psi$ in the basis $\mathcal{O}$, obtain the eigenvalue o_m, set $N_m \leftarrow N_m + 1$;
for $m = 1$ *to* $\dim(spec(\mathcal{O}))$ **do**
  Normalize $N_m \leftarrow N_m/M$;
return $(N_m)_m$.

Algorithm 2: Computation of the loss

Parameters: $M \in \mathbb{N}$
Result: Value of the loss $\mathcal{C}$

Initialize $\mathcal{C} \leftarrow 0$;
Get the position quadratures distribution with alg. (1);
Get the photon numbers distribution with alg. (1);
Compute the loss $\mathcal{C}$ using eq. (13);
return $\mathcal{C}$.

Algorithm 3: Training of the parameterized photonic circuit

Parameters: $N_{\text{steps}} \in \mathbb{N}$, initial parameters $\theta_0 \in \mathbb{R}^K$, learning rate $\eta \in \mathbb{R}_+$
Result: Optimized hyperparameters $\theta \in \mathbb{R}^K$

Initialize hyperparameters $\theta \leftarrow \theta_0$;
for $i = 1$ *to* N_{steps} **do**
  Compute the loss $\mathcal{C}$ with alg. (2);
  Compute the gradient $\nabla_\theta \mathcal{C}$ with shift rule and alg. (2);
  Update the parameters $\theta \leftarrow \theta - \eta \nabla_\theta \mathcal{C}$;
  end for
return θ.

V. RESULTS

We gather here the results of the simulations. We focus on the case of 2 QDOs. In particular we plot the profile of the binding energy as a function of the distance between the two nuclei. The binding energy is simply defined as the ground state energy of the interacting system to which one substract the ground state energy of the uninteracting system, namely that of a pair of free harmonic oscillators in this Drude model.

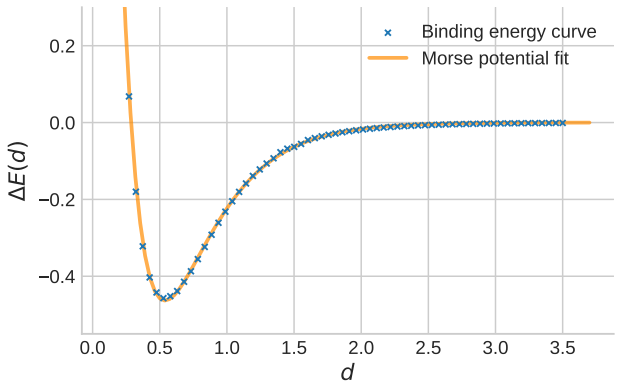

FIG. 2. a

VI. QUANTUM MUTUAL INFORMATION

Let us give the expression of the partial densitity matrix associated to QDO_1. The total state of the system, we recall, is expressed in the Fock basis as

$$|\psi\rangle = \sum_{n_1,\dots,n_{2K}=0}^{\infty} \alpha_{n_1\dots n_{2K}} |n_1\rangle \otimes \cdots \otimes |n_{2K}\rangle \,, \tag{18}$$

leading to the following expression for the density matrix:

$$\rho = \sum_{\substack{n_1,\dots,n_{2K} \\ m_1,\dots,m_{2K}}} \alpha^*_{m_1\dots m_{2K}} \alpha_{n_1\dots n_{2K}} |n_1\rangle\langle m_1| \otimes \cdots \otimes |n_{2K}\rangle\langle m_{2K}| \,.$$

The partial trace associated to QDO_1 is therefore given by:

$$\rho_1 = \sum_{\substack{n_1,\dots,n_K \\ m_1,\dots,m_K \\ l_1,\dots,l_K}} \alpha^*_{m_1\dots m_K l_1,\dots,l_K} \alpha_{n_1\dots n_K l_1,\dots,l_K} |n_1\rangle\langle m_1| \otimes \cdots \otimes |n_K\rangle\langle m_K| \,. \tag{19}$$

Since the state of the total system is pure, the von Neumann entropy of the total density matrix is zero. QDO number 2 can then be interpreted as purifying the system composed solely of QDO number 1. The two QDO therefore have identical von Neumann entropy $S(\rho_1)$, the entanglement entropy. The quantum mutual information of the system is therefore given by

$$I(1:2) = S(\rho_1) + S(\rho_2) - S(\rho) = 2S(\rho_1) \,, \tag{20}$$

with the von Neumann entropy being defined as

$$S(\rho) = -\text{Tr}\rho \log \rho \,. \tag{21}$$

The profile of the quantum mutual information as a function of the interatomic distance is provided in fig. (...)

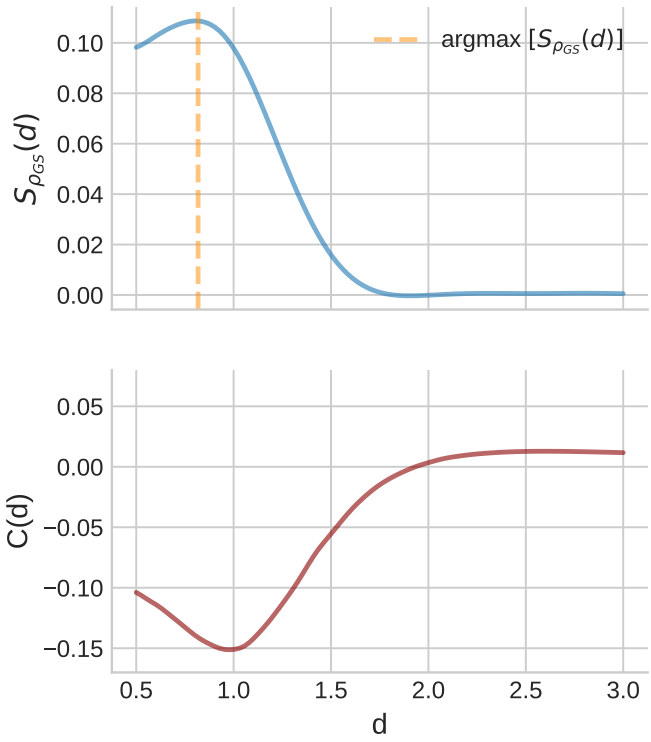

FIG. 3. a

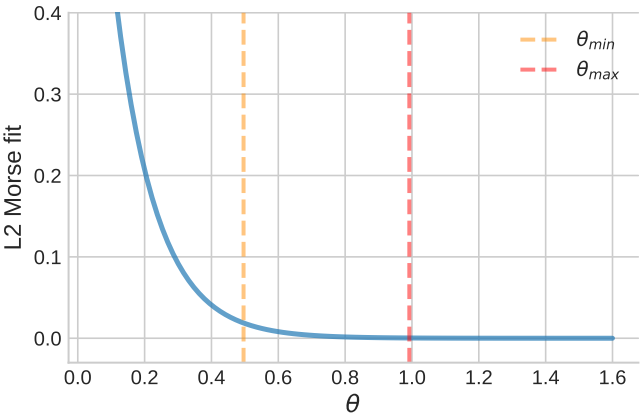

FIG. 4. a

VII. CONCLUSION

ACKNOWLEDGMENTS

We would like to thank Dahvyd Wing and Kyunghoon Han for important discussions.

CODE AVAILABILITY

The reader will find an open source python code accompanying this paper at github repository.

[1] H. Margenau and N. R. Kestner, *Theory of Intermolecular Forces: International Series of Monographs in Natural Philosophy*. Elsevier, 2013, vol. 18.

[2] I. G. Kaplan, *Intermolecular interactions: physical picture, computational methods and model potentials*. John Wiley & Sons, 2006.

[3] A. Stone, *The theory of intermolecular forces*. oUP oxford, 2013.

[4] J. O. Hirschfelder, *Intermolecular Forces, Volume 12*. John Wiley & Sons, 2009, vol. 12.

[5] H. B. Casimir and D. Polder, "The influence of retardation on the london-van der waals forces," *Physical Review*, vol. 73, no. 4, p. 360, 1948.

[6] S. Y. Buhmann, *Dispersion Forces I: Macroscopic quantum electrodynamics and ground-state Casimir, Casimir–Polder and van der Waals forces*. Springer, 2013, vol. 247.

[7] S. Y. Buhmann and D.-G. Welsch, "Dispersion forces in macroscopic quantum electrodynamics," *Progress in quantum electronics*, vol. 31, no. 2, pp. 51–130, 2007.

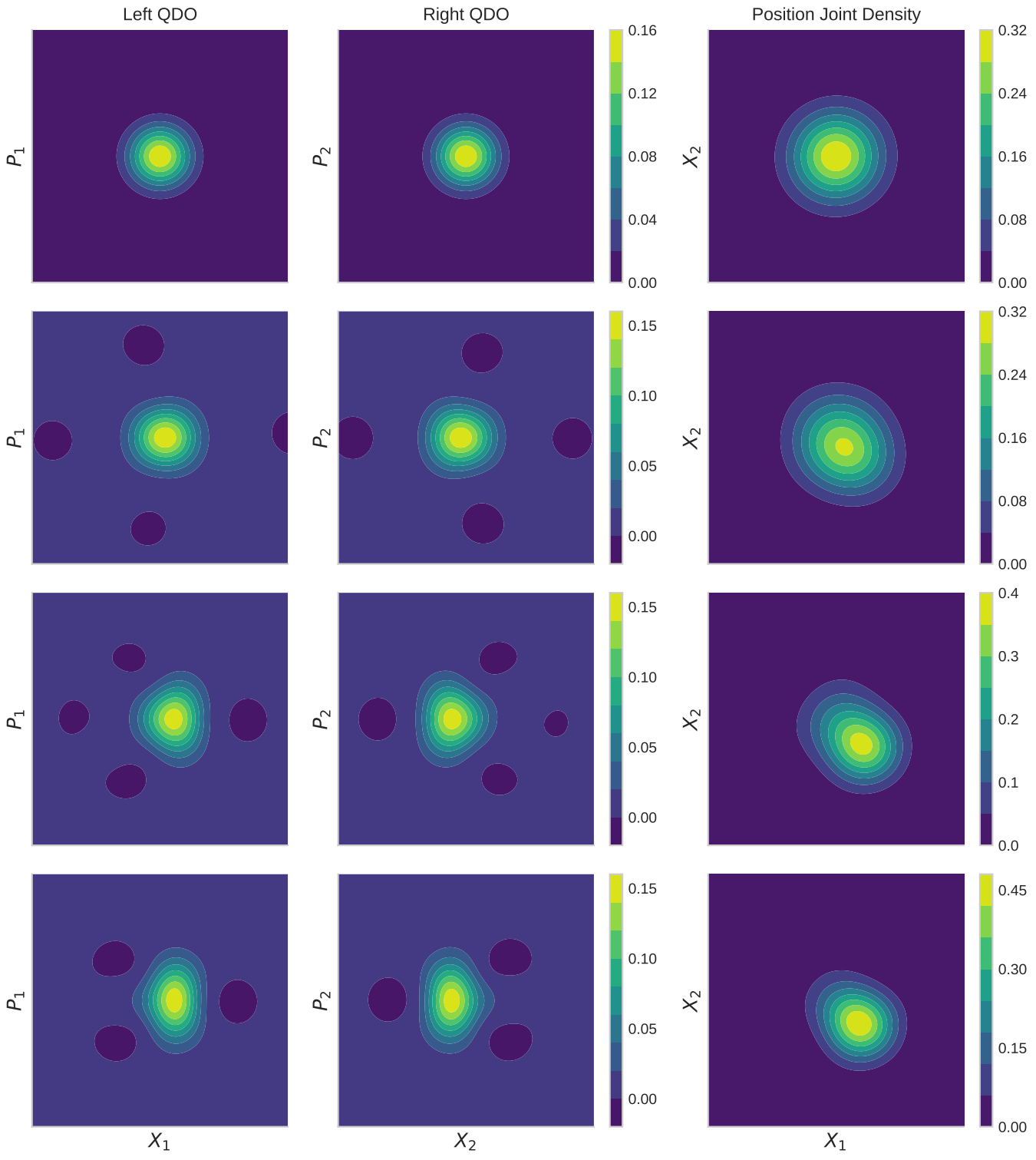

FIG. 5. a

[8] G. Compagno, R. Passante, and F. Persico, *Atom-field interactions and dressed atoms*, 1995.

[9] R. Passante, "Dispersion interactions between neutral atoms and the quantum electrodynamical vacuum," *Symmetry*, vol. 10, no. 12, p. 735, 2018.

[10] P. W. Milonni, *The quantum vacuum: an introduction to quantum electrodynamics*. Academic press, 2013.

[11] R. Schmid, "Recent advances in the description of the structure of water, the hydrophobic effect, and the like-dissolves-like rule," *Monatshefte für Chemie/Chemical*

Monthly, vol. 132, no. 11, pp. 1295–1326, 2001.

[12] J. Hoja, H.-Y. Ko, M. A. Neumann, R. Car, R. A. DiStasio Jr, and A. Tkatchenko, "Reliable and practical computational description of molecular crystal polymorphs," *Science Advances*, vol. 5, no. 1, p. eaau3338, 2019.

[13] J. Hoja and A. Tkatchenko, "First-principles stability ranking of molecular crystal polymorphs with the dft+ mbd approach," *Faraday Discussions*, vol. 211, pp. 253–274, 2018.

[14] M. Mortazavi, J. G. Brandenburg, R. J. Maurer, and A. Tkatchenko, "Structure and stability of molecular crystals with many-body dispersion-inclusive density functional tight binding," *The journal of physical chemistry letters*, vol. 9, no. 2, pp. 399–405, 2018.

[15] M. Stöhr and A. Tkatchenko, "Quantum mechanics of proteins in explicit water: The role of plasmon-like solute-solvent interactions," *Science advances*, vol. 5, no. 12, p. eaax0024, 2019.

[16] A. M. Reilly and A. Tkatchenko, "Role of dispersion interactions in the polymorphism and entropic stabilization of the aspirin crystal," *Physical review letters*, vol. 113, no. 5, p. 055701, 2014.

[17] M. Galante and A. Tkatchenko, "Anisotropic van der waals dispersion forces in polymers: Structural symmetry breaking leads to enhanced conformational search," *arXiv preprint arXiv:2110.06646*, 2021.

[18] A. Kleshchonok and A. Tkatchenko, "Tailoring van der waals dispersion interactions with external electric charges," *Nature communications*, vol. 9, no. 1, pp. 1–9, 2018.

[19] A. Ambrosetti, P. Umari, P. L. Silvestrelli, J. Elliott, and A. Tkatchenko, "Optical van-der-waals forces in molecules: from electronic bethe-salpeter calculations to the many-body dispersion model," *Nature communications*, vol. 13, no. 1, pp. 1–8, 2022.

[20] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Photons and Atoms-Introduction to Quantum Electrodynamics*, 1997.

[21] ——, *Atom-photon interactions: basic processes and applications*, 1998.

[22] G. Preparata, *QED Coherence in matter*. World Scientific, 1995.

[23] A. Salam, *Molecular quantum electrodynamics: long-range intermolecular interactions*. John Wiley & Sons, 2009.

[24] D. Craig, "T. thirunamachandran in molecular quantum electrodynamics," 1998.

[25] D. Richardson, "Dispersion contribution of two-atom interaction energy: Multipole interactions," *Journal of Physics A: Mathematical and General*, vol. 8, no. 11, p. 1828, 1975.

[26] J. Mahanty and B. Ninham, "Dispersion contributions to surface energy," *The Journal of Chemical Physics*, vol. 59, no. 11, pp. 6157–6162, 1973.

[27] L. Woods, D. A. R. Dalvit, A. Tkatchenko, P. Rodriguez-Lopez, A. W. Rodriguez, and R. Podgornik, "Materials perspective on casimir and van der waals interactions," *Reviews of Modern Physics*, vol. 88, no. 4, p. 045003, 2016.

[28] A. Tkatchenko, "Current understanding of van der waals effects in realistic materials," *Advanced Functional Materials*, vol. 25, no. 13, pp. 2054–2061, 2015.

[29] X. Ren, P. Rinke, C. Joas, and M. Scheffler, "Random-phase approximation and its applications in computational chemistry and materials science," *Journal of Materials Science*, vol. 47, no. 21, pp. 7447–7471, 2012.

[30] J. Harl and G. Kresse, "Accurate bulk properties from approximate many-body techniques," *Physical review letters*, vol. 103, no. 5, p. 056401, 2009.

[31] J. F. Dobson and T. Gould, "Calculation of dispersion energies," *Journal of Physics: Condensed Matter*, vol. 24, no. 7, p. 073201, 2012.

[32] V. A. Parsegian, *Van der Waals forces: a handbook for biologists, chemists, engineers, and physicists*. Cambridge university press, 2005.

[33] A. D. Becke and E. R. Johnson, "A simple effective potential for exchange," *The Journal of chemical physics*, vol. 124, no. 22, p. 221101, 2006.

[34] ——, "Exchange-hole dipole moment and the dispersion interaction: High-order dispersion coefficients," *The Journal of chemical physics*, vol. 124, no. 1, p. 014104, 2006.

[35] S. Grimme, J. Antony, S. Ehrlich, and H. Krieg, "A consistent and accurate ab initio parametrization of density functional dispersion correction (dft-d) for the 94 elements h-pu," *The Journal of chemical physics*, vol. 132, no. 15, p. 154104, 2010.

[36] S. Grimme, "Semiempirical gga-type density functional constructed with a long-range dispersion correction," *Journal of computational chemistry*, vol. 27, no. 15, pp. 1787–1799, 2006.

[37] A. Tkatchenko, R. A. DiStasio Jr, R. Car, and M. Scheffler, "Accurate and efficient method for many-body van der waals interactions," *Physical review letters*, vol. 108, no. 23, p. 236402, 2012.

[38] D. Massa, A. Ambrosetti, and P. L. Silvestrelli, "Many-body van der waals interactions beyond the dipole approximation," *The Journal of Chemical Physics*, vol. 154, no. 22, p. 224115, 2021.

[39] A. Ambrosetti, A. M. Reilly, R. A. DiStasio Jr, and A. Tkatchenko, "Long-range correlation energy calculated from coupled atomic response functions," *The Journal of chemical physics*, vol. 140, no. 18, p. 18A508, 2014.

[40] W. L. Bade and J. G. Kirkwood, "Drude-model calculation of dispersion forces. ii. the linear lattice," *The Journal of Chemical Physics*, vol. 27, no. 6, pp. 1284–1288, 1957. [Online]. Available: https://doi.org/10.1063/1.1743992

[41] W. L. Bade, "Drude-model calculation of dispersion forces. i. general theory," *The Journal of Chemical Physics*, vol. 27, no. 6, pp. 1280–1284, 1957. [Online]. Available: https://doi.org/10.1063/1.1743991

[42] M. R. Karimpour, D. V. Fedorov, and A. Tkatchenko, "Quantum framework for describing retarded and nonretarded molecular interactions in external electric fields," *Physical Review Research*, vol. 4, no. 1, jan 2022. [Online]. Available: https://doi.org/10.1103%2Fphysrevresearch.4.013011

[43] ——, "Comprehensive Quantum Framework for Describing Retarded and Non-Retarded Molecular Interactions in External Electric Fields," 2021.

[44] M. Gori, P. Kurian, and A. Tkatchenko, "Second quantization approach to many-body dispersion interactions," 2022. [Online]. Available: https://arxiv.org/abs/2205.11549

[45] D. Massa, A. Ambrosetti, and P. L. Silvestrelli, "Beyond-dipole van der waals contributions within the many-body dispersion framework," *Electronic Structure*, vol. 3, no. 4,

p. 044002, 2021.
[46] M. Sadhukhan and F. R. Manby, "Quantum mechanics of drude oscillators with full coulomb interaction," *Physical Review B*, vol. 94, no. 11, p. 115106, 2016.
[47] L. W. Anderson, M. Kiffner, P. K. Barkoutsos, I. Tavernelli, J. Crain, and D. Jaksch, "Coarse-grained intermolecular interactions on quantum processors," *Physical Review A*, vol. 105, no. 6, p. 062409, 2022.
[48] B. L. Blaney, B. BL, and E. GE, "Van der waals molecules." 1976.
[49] A. Jones, "Quantum drude oscillators for accurate many-body intermolecular forces," 2010.
[50] A. P. Jones, J. Crain, V. P. Sokhan, T. W. Whitfield, and G. J. Martyna, "Quantum drude oscillator model of atoms and molecules: Many-body polarization and dispersion interactions for atomistic simulation," *Phys. Rev. B*, vol. 87, p. 144103, Apr 2013. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevB.87.144103
[51] K. Tang and J. P. Toennies, "An improved simple model for the van der waals potential based on universal damping functions for the dispersion coefficients," *The Journal of chemical physics*, vol. 80, no. 8, pp. 3726–3741, 1984.
[52] N. M. Tubman and J. McMinis, "Renyi entanglement entropy of molecules: Interaction effects and signatures of bonding," *arXiv preprint arXiv:1204.4731*, 2012.
[53] S.-H. Zhang and Z.-Y. Yan, "Frequency dependence of the entanglement entropy production in a system of coupled driven nonlinear oscillators," *Entropy*, vol. 21, no. 9, p. 889, 2019.
[54] S. Lloyd and S. L. Braunstein, "Quantum computation over continuous variables," *Physical Review Letters*, vol. 82, no. 8, p. 1784, 1999.
[55] P. Van Loock, W. Munro, K. Nemoto, T. Spiller, T. Ladd, S. L. Braunstein, and G. Milburn, "Hybrid quantum computation in quantum optics," *Physical Review A*, vol. 78, no. 2, p. 022303, 2008.
[56] N. Killoran, J. Izaac, N. Quesada, V. Bergholm, M. Amy, and C. Weedbrook, "Strawberry fields: A software platform for photonic quantum computing," *Quantum*, vol. 3, p. 129, 2019.
[57] M. A. Nielsen and I. Chuang, "Quantum computation and quantum information," 2002.
[58] A. K. Pati, S. L. Braunstein, and S. Lloyd, "Quantum searching with continuous variables," *arXiv preprint quant-ph/0002082*, 2000.
[59] A. K. Pati and S. L. Braunstein, "Deutsch-jozsa algorithm for continuous variables," *Quantum Information with Continuous Variables*, pp. 31–36, 2003.
[60] S. L. Braunstein and P. Van Loock, "Quantum information with continuous variables," *Reviews of modern physics*, vol. 77, no. 2, p. 513, 2005.
[61] U. L. Andersen, G. Leuchs, and C. Silberhorn, "Continuous-variable quantum information processing," *Laser & Photonics Reviews*, vol. 4, no. 3, pp. 337–354, 2010.
[62] S. J. Lomonaco Jr and L. H. Kauffman, "A continuous variable shor algorithm," *arXiv preprint quant-ph/0210141*, 2002.
[63] S. L. Braunstein, "Error correction for continuous quantum variables," *Physical Review Letters*, vol. 80, no. 18, p. 4084, 1998.
[64] J. M. Arrazola, V. Bergholm, K. Brádler, T. R. Bromley, M. J. Collins, I. Dhand, A. Fumagalli, T. Gerrits, A. Goussev, L. G. Helt *et al.*, "Quantum circuits with many photons on a programmable nanophotonic chip," *Nature*, vol. 591, no. 7848, pp. 54–60, 2021.
[65] M. Tillmann, B. Dakić, R. Heilmann, S. Nolte, A. Szameit, and P. Walther, "Experimental boson sampling," *Nature photonics*, vol. 7, no. 7, pp. 540–544, 2013.
[66] H.-S. Zhong, H. Wang, Y.-H. Deng, M.-C. Chen, L.-C. Peng, Y.-H. Luo, J. Qin, D. Wu, X. Ding, Y. Hu *et al.*, "Quantum computational advantage using photons," *Science*, vol. 370, no. 6523, pp. 1460–1463, 2020.
[67] T. R. Bromley, J. M. Arrazola, S. Jahangiri, J. Izaac, N. Quesada, A. D. Gran, M. Schuld, J. Swinarton, Z. Zabaneh, and N. Killoran, "Applications of near-term photonic quantum computers: software and algorithms," *Quantum Science and Technology*, vol. 5, no. 3, p. 034010, 2020.
[68] M. Schuld and F. Petruccione, *Machine learning with quantum computers*. Springer, 2021.
[69] N. Killoran, T. R. Bromley, J. M. Arrazola, M. Schuld, N. Quesada, and S. Lloyd, "Continuous-variable quantum neural networks," *Physical Review Research*, vol. 1, no. 3, p. 033063, 2019.
[70] J. M. Arrazola, T. R. Bromley, J. Izaac, C. R. Myers, K. Brádler, and N. Killoran, "Machine learning method for state preparation and gate synthesis on photonic quantum computers," *Quantum Science and Technology*, vol. 4, no. 2, p. 024004, 2019.
[71] Y. Enomoto, K. Anai, K. Udagawa, and S. Takeda, "Continuous-variable quantum approximate optimization on a programmable photonic quantum processor," *arXiv preprint arXiv:2206.07214*, 2022.
[72] M. Cerezo, A. Arrasmith, R. Babbush, S. C. Benjamin, S. Endo, K. Fujii, J. R. McClean, K. Mitarai, X. Yuan, L. Cincio *et al.*, "Variational quantum algorithms," *Nature Reviews Physics*, vol. 3, no. 9, pp. 625–644, 2021.
[73] A. Peruzzo, J. McClean, P. Shadbolt, M.-H. Yung, X.-Q. Zhou, P. J. Love, A. Aspuru-Guzik, and J. L. O'brien, "A variational eigenvalue solver on a photonic quantum processor," *Nature communications*, vol. 5, no. 1, p. 4213, 2014.
[74] K. Mitarai, M. Negoro, M. Kitagawa, and K. Fujii, "Quantum circuit learning," *Physical Review A*, vol. 98, no. 3, p. 032309, 2018.
[75] K. Marshall, R. Pooser, G. Siopsis, and C. Weedbrook, "Quantum simulation of quantum field theory using continuous variables," *Physical Review A*, vol. 92, no. 6, p. 063825, 2015.
[76] K. Yeter-Aydeniz, E. Moschandreou, and G. Siopsis, "Quantum imaginary-time evolution algorithm for quantum field theories with continuous variables," *Physical Review A*, vol. 105, no. 1, p. 012412, 2022.
[77] O. Kiss, F. Tacchino, S. Vallecorsa, and I. Tavernelli, "Quantum neural networks force fields generation," *Machine Learning: Science and Technology*, vol. 3, no. 3, p. 035004, 2022.
[78] J. Apanavicius, Y. Feng, Y. Flores, M. Hassan, and M. McGuigan, "Morse potential on a quantum computer for molecules and supersymmetric quantum mechanics," *arXiv preprint arXiv:2102.05102*, 2021.
[79] H. Wang, S. Ashhab, and F. Nori, "Efficient quantum algorithm for preparing molecular-system-like states on a quantum computer," *Physical Review A*, vol. 79, no. 4, p. 042335, 2009.
[80] Y. Nam, J.-S. Chen, N. C. Pisenti, K. Wright, C. Delaney, D. Maslov, K. R. Brown, S. Allen, J. M. Amini,

J. Apisdorf *et al.*, "Ground-state energy estimation of the water molecule on a trapped-ion quantum computer," *npj Quantum Information*, vol. 6, no. 1, p. 33, 2020.

[81] J. Romero, R. Babbush, J. R. McClean, C. Hempel, P. J. Love, and A. Aspuru-Guzik, "Strategies for quantum computing molecular energies using the unitary coupled cluster ansatz," *Quantum Science and Technology*, vol. 4, no. 1, p. 014008, 2018.

[82] J. I. Colless, V. V. Ramasesh, D. Dahlen, M. S. Blok, M. E. Kimchi-Schwartz, J. R. McClean, J. Carter, W. A. de Jong, and I. Siddiqi, "Computation of molecular spectra on a quantum processor with an error-resilient algorithm," *Physical Review X*, vol. 8, no. 1, p. 011021, 2018.