

**Sadhukhan and Tkatchenko Reply:** In our Letter [1], we investigated the interaction between a pair of 3D Drude oscillators (charge-separated, overall-neutral quantum harmonic oscillators), coupled by a Coulomb potential in reduced spatial dimensions, utilizing a novel perturbation expansion based on Ref. [2] and correlated dipolar oscillator states. The coupled oscillator Hamiltonian models the instantaneous, quantum-mechanical electronic fluctuations (not the permanent deformations of the electron density), therefore being a model for electron correlation via the adiabatic connection fluctuation-dissipation theorem [3]. We have shown that as a result the repulsive interaction between van der Waals (vdW) dimers arises from dynamic electron correlation effects in confined environments and can only be realized when going beyond the widely used dipole coupling between the oscillators. Our results do not require any permanent multipole moments.

In the preceding Comment [4], Podszwa and Jansen (PJ) criticize our interpretation of the repulsive interaction as “rather the interaction between the static quadrupole moments of 1D, 2D, and anisotropic 3D QHOs”, claiming our result involves only one oscillator’s parameters and postulated that the expression will contain the product of two independent oscillator’s parameters. Here, we show that their classical interpretation disagrees with our full quantum-mechanical treatment of two dissimilar Coulomb-coupled oscillators, proving that the repulsive interaction arises from electronic correlations.

We generalize our expression [1] for two 1D oscillators with different charges ( $q_{A/B}$ ), masses ( $m_{A/B}$ ), and frequencies ( $\omega_{A/B}$ ). Diagonalization of the dipole-coupled potential matrix in coordinates  $\zeta_{A/B} = \sqrt{m_{A/B}}x_{1/2}$  (see Supplemental Material of Ref. [1]) yields the frequencies of two uncoupled oscillators as

$$\omega_{\pm} = \omega_{\text{eff}} \sqrt{1 \pm \sqrt{1 - \frac{(1 - \gamma^2)\omega_A^2\omega_B^2}{\omega_{\text{eff}}^4}}}$$

$$\omega_{\text{eff}} = \sqrt{\frac{\omega_A^2 + \omega_B^2}{2}} \quad (1)$$

where  $\gamma' = (-2q_Aq_B/\sqrt{m_A m_B}\omega_A\omega_B R^3)$ . This leads to [5]

$$\epsilon_1^{AB} = 2 \frac{\alpha_2^{AB} \hbar \omega_{\text{eff}}}{R^5}; \quad \alpha_2^{AB} = \frac{3\tilde{q}^2 \hbar}{4\omega_{\text{eff}}^3}; \quad \tilde{q} = \sqrt{\frac{q_A q_B}{m_A m_B}},$$

where  $\omega_A = \omega_B = \omega$  recovers results from Refs. [1,6], without the frequency dependence claimed by PJ [7]. Additionally, for  $N$  arbitrary oscillators, the repulsive energy [1] cannot be expressed in terms of oscillator densities only, highlighting its quantum-mechanical origin. This inconsistency in PJ’s expression arises since it describes interaction between negative charge densities. Considering both positive and negative charges in each

oscillator and treating the oscillator Hamiltonian as an electronic one, the electrostatic energy becomes (dropping prefactors)

$$E_{es} = \frac{q_A q_B}{R} + \frac{\Theta_A \Theta_B}{R^5} + \frac{\Theta_A q_B}{R^3} + \frac{\Theta_B q_A}{R^3}, \quad (2)$$

implying a long-range, repulsive  $1/R$  interaction between positive charges, contradicting our quantum-mechanical derivation.

Such contradictions arise as they calculate the parallel component of the quadrupole moment of a QDO (Eq. (2.13) of Ref. [8]) as  $\Theta_{\parallel} = (\hbar/2m\omega)$ , which is independent of charge  $q$ . For a Drude oscillator to model permanent electrostatics, one needs to unphysically equate electronic charge  $e$  to  $q$  (see Ref. [2]).

Similarly, PJ’s analysis of oscillators confined in 2D and quasi-3D spaces disagrees with our analytical results [1]. Our result for the quasi-3D confinement scales as  $R^{-5}$ , contradicting PJ.

Feynman’s [9] famous *a posteriori* electrostatic picture helps to understand the vdW force’s distance dependence, where the source of the electron density modification are quantum-mechanical fluctuations on distant atoms. Similarly, PJ’s model accounts for the distance dependence, albeit with incorrect prefactors.

To clarify confusion, we elaborate on what we mean by confinement. In absence of any permanent electron density deformation, an applied inhomogeneous electric field changes the polarizability tensor of molecules embedded in a material (nanotube, 2D nanostructure, semi-infinite surface, or other matter). The 3D molecule possesses anisotropic polarizabilities due to the imposed confinement, modeled in Ref. [1], by breaking the rotational symmetry of the 3D vacuum. Consequently, 3D oscillators interact by a modified Coulomb potential. Implementation on top of the microscopic DFT + MBD Hamiltonian [10,11] will generalize [1] for arbitrary materials, where the underlying DFT functional fully describes electrostatic interactions.

We conclude that our original derivation and interpretation [1] of the repulsion between vdW dimers, arising from dynamic correlation, are correct.

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