Comment on "Dispersion Interaction between Two Hydrogen Atoms in a Static Electric Field"

For the noncovalent interaction between two ground-state H atoms, modeled as two-level systems, under static electric fields, Fiscelli *et al.* [1] obtained leading contributions to the interaction energy scaling with interatomic distance as $r^{-3}(r^{-4})$ for nonretarded (retarded) regime:

$$E_{\rm int}^{\perp} \simeq \frac{9\alpha^2\mathcal{E}\mathcal{E}'}{4\pi\epsilon_0}\begin{cases}\frac{1}{2r^3},\\ \frac{4}{\pi k_0 r^4};\end{cases} \qquad E_{\rm int}^{\parallel} \simeq -\frac{9\alpha^2\mathcal{E}\mathcal{E}'}{4\pi\epsilon_0}\begin{cases}\frac{1}{r^3},\\ \frac{4}{\pi k_0 r^4}.\end{cases} \tag{1}$$

The static field $\mathcal{E}/\mathcal{E}'$ at atom A/B is assumed to be either perpendicular ($E_{\rm int}^{\perp}$) or parallel ($E_{\rm int}^{\parallel}$) to $\boldsymbol{r}$, whereas α is the atomic polarizability and $k_0 = 2|E_2 - E_1|/\hbar c$, where $E_2 - E_1 = E_1/4 - E_1$ is the energy difference between the first two levels of hydrogen [1]. Given the different scaling for the retarded and nonretarded regimes, the authors interpreted this field-induced interaction as "dispersion."

In their Comment, Abrantes *et al.* [2] interpreted the results of Ref. [1] as a combination of dispersion and electrostatic interactions, employing a classical picture without referring to quantum electrodynamics (QED) used by Fiscelli *et al.* [1], who still argued in their Reply [3] that Eq. (1) corresponds to the dispersion interaction between fluctuating dipoles upon exchanging one virtual photon.

By using second-order perturbation theory in QED with properly orthogonalized atomic states, we show that the resulting interaction between two hydrogen atoms in static fields corresponds to a field-induced electrostatic energy scaling as r^{-3} for any r. Our derivation settles recent conflicting discussions in Refs. [1–4] and proves that the QED second-order interaction between two atoms in static electric fields has a purely electrostatic origin.

Unperturbed states in QED perturbation theory must satisfy the closure relation $\sum_n |n\rangle\langle n| = \mathbb{1}$ [5]. Following the approach of Ref. [1], we obtain the eigenstates of a two-level hydrogen in the static field $\mathcal{E}$ as

$$|g\rangle = c_0\left[(1-\gamma^2\mathcal{E}^2)|100\rangle - \frac{(3/2)^6}{\sqrt{2}}\gamma^2\mathcal{E}^2|200\rangle - \sqrt{2}\gamma\mathcal{E}|210\rangle\right],$$

$$|e^{\pm}\rangle = \frac{1}{\sqrt{2}}\{|210\rangle \pm |200\rangle\}, \text{ with } \gamma = 2^9 q_e a_0/3^6 E_1, \tag{2}$$

using the second and zeroth order of perturbation theory for the ground ($|g\rangle$) and excited ($|e^{\pm}\rangle$) states, respectively. Here, $|nlm\rangle$ represent eigenstates of an isolated H atom. Since $c_0 \approx 1$ [6], Eq. (2) agrees with Eqs. (5) and (7) of Ref. [1], where the wave functions can be written as $|\psi\rangle = |g_A\rangle|g_B\rangle|0_{\mathbf{k}\lambda}\rangle$ and $|\tilde{I}_0^{\pm,\pm}\rangle = |e_A^{\pm}\rangle|e_B^{\pm}\rangle$, respectively. Since the states in Eq. (2) do not satisfy the closure relation, we apply the Gram–Schmidt orthonormalization procedure

$$|u_1\rangle = |g\rangle, \qquad |u_2\rangle = c_1\{|e^+\rangle - \langle e^+|g\rangle|g\rangle\},$$
$$|u_3\rangle = c_2\{|e^-\rangle - \langle e^-|g\rangle|g\rangle - \langle e^-|u_2\rangle|g\rangle\}, \tag{3}$$

which yields states (with normalization factors c_1, c_2) obeying the closure relation. Now evaluating Eq. (6) of Ref. [1] with these states, we get $E_{\rm int}^{\perp/\parallel} \propto r^{-3}$ for any r.

This finding becomes clear with our detailed analysis [7] based on quantum mechanics and QED. Atom A acquires a field-induced dipole $\alpha\boldsymbol{\mathcal{E}}$ when coupled to the vacuum field and emits a virtual photon. Then, Atom B couples to the vacuum field via its field-induced dipole $\alpha\boldsymbol{\mathcal{E}}'$ and absorbs a virtual photon. This photon exchange corresponds to two time-ordered diagrams [7] similar to the electrostatic interaction between permanent dipoles [8]. Performing sums over the field-dressed atomic states and polarization of the vacuum field, as well as integrals over frequencies of the exchanged virtual photons, gives $E_{\rm int} \propto \alpha^2\mathcal{E}\mathcal{E}'/4\pi\epsilon_0 r^3$. Consistent with nonretarded quantum mechanics [7], this expression describes the field-induced dipole-dipole electrostatic interaction.

Moreover, the Planck constant does not enter Eq. (1), which is obvious for the nonretarded regime. For the retarded regime, $\hbar$ is also eliminated by using $k_0 = 2\omega/c$, where ω is the transition frequency. Hence, this interaction is not dispersion but field-induced electrostatics [7].

Independently of our Letter, the issue with unperturbed states of Ref. [1] was also recognized by Hu *et al.* [4], following the QED formalism in Refs. [9,10]. Nevertheless, we highlight the idea of Fiscelli *et al.* [1] to influence molecular interactions by static electric fields that provided a strong motivation for our research [7].

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