Supplementary material: connection with the minimal coupling Hamiltonian

So far the regime of ultrastrong light-matter coupling has been discussed in the standard representation through the use of the minimal coupling Hamiltonian, which relies on the vector potential rather on the electric field intensity [1]. In this supplementary material we discuss briefly the connection between the two viewpoints.

We start with the full Hamiltonian of the system, expressed in the Power-Zienau-Woolley (PZW) representation:

\[ H = \hbar \omega_c (a^\dagger a + 1/2) + \hbar \omega_{12} b^\dagger b + i \hbar \omega_p \frac{\omega_c}{\omega_{12}} (a - a^\dagger)(b + b^\dagger) + \hbar \omega_p^2 \frac{\omega_c}{4\omega_{12}} (b + b^\dagger)^2 \]  

(1)

We then perform on (1) the inverse PZW unitary transformation [2], which in our case can be expressed as:

\[ T = \exp \left\{ -i \frac{\chi}{\omega_{12}} (a + a^\dagger)(b + b^\dagger) \right\} \]  

(2)

Here we have introduced the light-matter coupling constant \( \chi \) in the standard representation [1, 3]:

\[ \chi = \frac{\omega_p^2}{2} \sqrt{\frac{\omega_{12}}{\omega_c}} \]  

(3)

This leads to the following transformed Hamiltonian:

\[ T^\dagger HT = \hbar \omega_c (a^\dagger a + 1/2) + \hbar \omega_{12} b^\dagger b + i \hbar \chi (a + a^\dagger)(b - b^\dagger) + \hbar \chi^2 \frac{\omega_c}{\omega_{12}} (a + a^\dagger)^2 + (1 - f_w) \frac{\hbar \omega_p^2}{4\omega_{12}} (b + b^\dagger)^2 \]  

(4)

The first three terms of (4) coincide with the Hopfield Hamiltonian [1, 4], in which all the relevant features of the ultrastrong coupling are present, namely the quadratic term \((a + a^\dagger)^2\) and the anti-resonant terms \((ab; a^\dagger b^\dagger)\) [1]. Note that the vector potential \( A \) is proportional to the sum of the operators \( A \propto (a + a^\dagger) \), whereas the in the PZW representation, the electric displacement \( D \) is provided by the difference \( D \propto (a - a^\dagger) \), which explains the difference in the coupling terms between (1) and (4).

The last term in (4), proportional to \((b + b^\dagger)^2\), describes a local field correction due to the multilayered structuring of our system (see Fig. 1(c) in the main text), and it vanishes for a homogeneous media \( f_w = 1 \). This term can be seen as an effective dipole-dipole interaction, analogous to the one introduced by Hopfield in his seminal paper of 1958 [4]. In our case, it naturally leads to the depolarization shift of the intersubband transition, observed in multipass absorption measurements. The later correspond to the weak light-matter coupling regime, where the intersubband transition is probed by a light mode with an extension that is very large compared to the thickness of the quantum well medium \( f_w \rightarrow 0 \).

---