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:

$$H = \hbar\omega_c(a^\dagger a + 1/2) + \hbar\omega_{12} b^\dagger b + \frac{i\hbar\omega_P}{2}\sqrt{f_w \frac{\omega_c}{\omega_{12}}}(a - a^\dagger)(b + b^\dagger) + \frac{\hbar\omega_P^2}{4\omega_{12}}(b + b^\dagger)^2 \tag{1}$$

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

$$T = \exp\Big\{ -i\frac{\chi}{\omega_{12}}(a + a^\dagger)(b + b^\dagger)\Big\} \tag{2}$$

Here we have introduced the light-matter coupling constant χ in the standard representation [1, 3]:

$$\chi = \frac{\omega_P}{2}\sqrt{f_w \frac{\omega_{12}}{\omega_c}} \tag{3}$$

This leads to the following transformed Hamiltonian:

$$T^\dagger H T = \hbar\omega_c(a^\dagger a + 1/2) + \hbar\omega_{12} b^\dagger b + i\hbar\chi(a + a^\dagger)(b - b^\dagger) + \frac{\hbar\chi^2}{\omega_{12}}(a + a^\dagger)^2 + (1 - f_w)\frac{\hbar\omega_P^2}{4\omega_{12}}(b + b^\dagger)^2 \tag{4}$$

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 $\mathbf{A}$ is proportional to the sum of the operators $\mathbf{A} \propto (a + a^\dagger)$, whereas the in the PZW representation, the electric displacement $\mathbf{D}$ is provided by the difference $\mathbf{D} \propto (a - a^\dagger)$, 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 intersubdand transition is probed by a light mode with an extension that is very large compared to the thickness of the quantum well medium ($f_w \to 0$).

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