The detailed calculation of an excitation probability P_e of a two-level atom by a travelling light pulse is desctibed in details in [1–3]. In particular, $P_e(t)$ is determined by dynamical coupling strength g(t) which is given by [1]

$$g(t) = \sqrt{\Gamma_p N}\,\xi(t). \tag{1}$$

Here, N is the average number of photons in a pulse with a coherent statistics, $\xi(t)$ is the normailzed temporal envelope mode function and Γ_p is the decay rate of an atom into a spatial mode subtended by an excitation pulse. Since in the theoretical description of experiments described below, Γ_p will be the only parameter, we briefly outline its evaluation for experimental settings.

We start with the expression of the electric field operator

$$\hat{\mathbf{E}}^{+}(\mathbf{r},t) = i\sum_{\lambda}\int d^3\mathbf{k}\sqrt{\frac{\hbar\omega_k}{(2\pi)^3\,2\epsilon_0}}\,\hat{a}_{\mathbf{k},\lambda}\,\epsilon_{\mathbf{k},\lambda}\,u_{\mathbf{k},\lambda}(\mathbf{r},t)\,e^{-i\omega_k t}, \tag{2}$$

where $\omega_k = c|\mathbf{k}|$, c is the vacuum speed of light, ϵ_0 is the permittivity of the vacuum, $u_{\mathbf{k},\lambda}$ are spatial mode functions, and $\epsilon_{\mathbf{k},\lambda}$ are unit polarization vectors $\lambda = 1,2$. A two-level atom, represented by a dipole operator $\hat{d} = |d|\hat{\sigma}_{x,y,z}\mathbf{e_d}$ interacts with a field at its location $\mathbf{r_a}$ with a coupling strength (dipole matrix element, Rabi frequency, interaction Hamiltonian, etc...) $g(\mathbf{r_a}) = (\hat{d}\cdot\hat{E}^{+})$

$$g(\mathbf{r_a}) = |d|\sqrt{\frac{\omega_k}{(2\pi)^3\,2\hbar\epsilon_0}}u_{\mathbf{k},\lambda}(\mathbf{e_d}\cdot\epsilon_{\mathbf{k},\lambda}), \tag{3}$$

where $\mathbf{e_d}$ is the unit dipole vector defining atomic transition symmetry. Using Wigner-Weisskopf approximation, one arrives to spontaneous decay rate Γ_p of an atom into pulse mode [4]

$$\Gamma_p = 2\pi\sum_{\lambda}\int d^3\mathbf{k}\,|g_{\mathbf{k},\lambda}(\mathbf{r_a})|^{\mathbf{2}}\,\delta(\omega_{\mathbf{k}} - \omega_{\mathbf{a}}). \tag{4}$$

Substitution of Eq. 3 into Eq. 4 and employing spherical coordinates yields

$$\begin{aligned}\Gamma_p &= \Gamma\cdot\left(\tfrac{3\Lambda}{8\pi}\right)\\ \Gamma &= \tfrac{1}{3\pi}\left(\tfrac{\omega_a}{c}\right)^3\tfrac{|d|^2}{\hbar\epsilon_0}\\ \Lambda &= \textstyle\int d\Omega\,|u_{\vec{k},\lambda}|^2\,|(\hat{e}_d\cdot\hat{\epsilon}_{\vec{k},\lambda})|^2\end{aligned} \tag{5}$$

Here Γ is the overall spontaneous decay rate [3]. The overlap integral $\Lambda \in \left[0, \frac{8\pi}{3}\right]$ shows the overlap between the spatial mode subtended by a pulse and atomic dipolar emission modes. Evaluation of this integral for different input mode functions was done in several recent works [5–7]. Since in our experiment the spatial mode function of the excitation pulse is a strongly focused Gaussian, we will follow results of work [7] to express the overlap integral in terms of the focusing strength $u := w_L/f$ defined through input beam waist w_L and focal distance of the coupling lens f

$$\Lambda = \frac{2\pi\,R_{sc}}{3} = \frac{\pi}{2u^3}e^{2/u^2}\left[\Gamma\left(-\frac{1}{4},\frac{1}{u^2}\right) + u\Gamma\left(\frac{1}{4},\frac{1}{u^2}\right)\right]^2 \tag{6}$$

For our experimental value $u = 0.22$, $\Gamma_p \approx 0.03\Gamma$ and coupling strength of equation 1 becomes

$$g(t) = \sqrt{0.03\Gamma\,N}\,\xi(t). \tag{7}$$

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