The detailed calculation of an excitation probability $\mathrm{P_e}$ of a two-level atom by a travelling light pulse in free space is described in [1–3]. In particular, $\mathrm{P_e(t)}$ 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 photon number in the coherent state pulse, $\xi(t)$ is the normalized temporal envelope function, and Γ_p is the atomic decay rate into the excitation pulse mode. The explicit formula of Γ_p can be found in Ref.[1] and expressed in terms of the total spontaneous decay rate Γ as

$$\begin{aligned} \Gamma_p &= \Gamma \cdot \left(\tfrac{3\Lambda}{8\pi}\right), \\ \Gamma &= \tfrac{1}{3\pi}\left(\tfrac{\omega_a}{c}\right)^3 \tfrac{d^2}{\hbar\epsilon_0}, \end{aligned} \tag{2}$$

where c is the vacuum speed of light, ϵ_0 is the permittivity of the vacuum, ω_a is the atomic transition frequency, and d is the scalar atomic dipole momentum. The single parameter $\Lambda \in \left[0, \frac{8\pi}{3}\right]$, which describes the spatial mode overlap, is obtained by weighting the atomic dipole pattern in the solid angle covered by the pulse

$$\Lambda = \sum_\lambda \int d\Omega\, |u_{\mathbf{k},\lambda}|^2\, |(\hat{e}_d \cdot \hat{\epsilon}_{\mathbf{k},\lambda})|^2, \tag{3}$$

where $u_{\mathbf{k},\lambda}$ are field spatial mode functions, $\epsilon_{\mathbf{k},\lambda}(\lambda = 1, 2)$ are unit polarization vectors, and $\mathbf{e_d}$ is the unit atomic dipole vector. Evaluation of the spatial overlap factor with different input mode functions has been done in several recent works [4–6]. Since in our experiment the spatial mode function of the excitation pulse is a strongly focused Gaussian, we will follow results of work [6] to express the overlap integral in terms of the scattering ratio R_{sc}, which further depends on the focusing strength $u := w_L/f$ as

$$\Lambda = \frac{R_{sc}}{4}\frac{8\pi}{3} = \frac{\pi}{2u^3} e^{2/u^2}\left[\Gamma\left(-\frac{1}{4}, \frac{1}{u^2}\right) + u\Gamma\left(\frac{1}{4}, \frac{1}{u^2}\right)\right]^2, \tag{4}$$

where w_L is the input beam waist, f is the focal distance of the coupling lens, and $\Gamma(a, b) = \int_b^\infty t^{a-1} e^{-t} dt$ is the incomplete gamma function. For our experimental value $u = 0.22$, we have $\Gamma_p \approx 0.03\Gamma$ and thus Eq. (1) becomes

$$g(t) = \sqrt{0.03\Gamma N}\,\xi(t). \tag{5}$$

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