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CORRIGENDUM • OPEN ACCESS

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Corrigendum: Detection of motional ground state population using delayed pulses (2016 *New J. Phys.* 18 013037)

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For the numerical simulation of the STIRAP process, we modeled the system with only one excited state. This approximation reproduces the experimental results in the publication reasonably well. However, the description is incomplete, since it predicts improved state transfer efficiency for larger detunings, while the opposite effect is observed experimentally. This can be explained by Stark-shift cancellation from coupling to other magnetic substates in the excited states. Efficient population transfer is recovered by introducing an additional frequency chirp. We provide an improved theoretical model including all involved excited state levels of the hyperfine manifold and new versions of figures 4 and 5 to illustrate that even for 9.2 GHz detuning the improved model shows better agreement with the experimental results.

The Hamiltonian for an off-resonant STIRAP population transfer in the interaction picture for a three level system reads (see figure 1 for level definitions)

$$H^{\mathrm{STIRAP}} = \frac{\hbar\Omega_1(t)}{2}|e\rangle\langle\downarrow|\,\mathrm{e}^{-\mathrm{i}\Delta_1 t} + \frac{\hbar\Omega_2(t)}{2}|e\rangle\langle\uparrow|\,\mathrm{e}^{-\mathrm{i}\Delta_2 t} + \mathrm{h.c.}, \tag{1}$$

where $\Omega_{1,2}(t)$ is the Rabi frequency for resonant coupling of the two lasers and $\Delta_{1,2}$ is the frequency detuning for the respective laser. To fulfill the two-photon resonance condition, the detunings have to be equal, i.e. $\Delta_1 = \Delta_2 \equiv \Delta$. In the following we will assume temporally Gaussian shapes of the Rabi frequencies and define the pulses according to the notation in the main paper [1]:

$$\Omega_1(t) = \Omega_1 \exp\left(-\frac{(t-t_0)^2}{2\tau^2}\right) \tag{2}$$

$$\Omega_2(t) = \Omega_2 \exp\left(-\frac{(t-t_0-t_d)^2}{2\tau^2}\right). \tag{3}$$

The pulse shape is Gaussian, centered at t_0 or $t_0 + t_d$ and pulse width is defined as $t_w = 2\sqrt{2\ln 2}\,\tau$. The time-averaged effect of the STIRAP Hamiltonian can be understood by applying the effective Hamiltonian theory [2] which gives

$$\begin{aligned} H_{\mathrm{eff}}^{\mathrm{STIRAP}} = &-\frac{\hbar\Omega_1(t)^2}{4\Delta}(|e\rangle\langle e| - |\uparrow\rangle\langle\uparrow|) \\ &-\frac{\hbar\Omega_2(t)^2}{4\Delta}(|e\rangle\langle e| - |\downarrow\rangle\langle\downarrow|) \\ &+\frac{\hbar\Omega_1(t)\Omega_2(t)}{4\Delta}(|\uparrow\rangle\langle\downarrow| + |\downarrow\rangle\langle\uparrow|). \end{aligned} \tag{4}$$

The first two terms are time dependent AC-Stark shifts, and the third term describes a Raman coupling between the two qubit states $(|\uparrow\rangle, |\downarrow\rangle)$. The differential AC-Stark shift between the two qubit states $\frac{\hbar}{4\Delta}(\Omega_2(t) - \Omega_1(t))$ acts as a nearly linear sweep for the time period where the effective two-photon coupling $\Omega_2(t)\Omega_1(t)/\Delta$ has a significant contribution to the dynamics. The effective Hamiltonian pictures clearly illustrates the connection between off-resonant STIRAP and RAP and helps to understand additional complications that arise when more complicated substructure in the excited state is considered and large relative detunings are chosen to suppress off-resonant scattering. This scenario will be described in the following on the example of $^{25}Mg^+$.

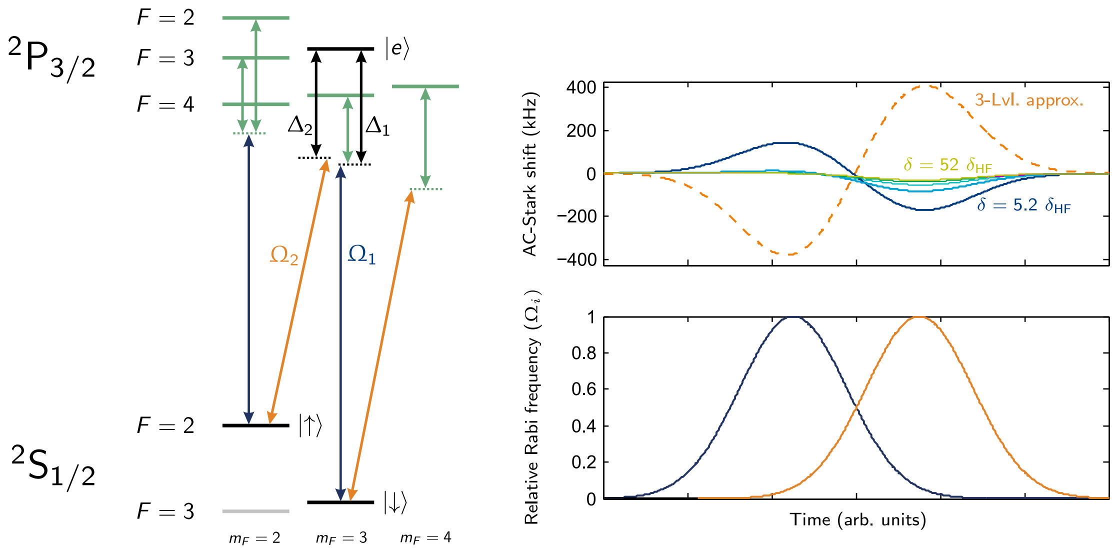

Figure 1. Reduced level scheme of $^{25}Mg^+$. The two qubit states $|\uparrow\rangle = |^2S_{1/2}, F = 2, m_F = 2\rangle$ and $|\downarrow\rangle = |^2S_{1/2}, F = 3, m_F = 3\rangle$ are coupled via the excited state $|e\rangle = |^2P_{3/2}, F = 3, m_F = 3\rangle$. Four additional substates of the $^2P_{3/2}$ manifold have to be taken into account, since they contribute in terms of an AC-Stark shift, that influences the Population transfer. The right panel shows the temporal behavior of the differential AC-Stark shift between states $|\downarrow\rangle$ and $|\uparrow\rangle$ for two delayed pulsed with Gaussian pulse shape as shown in the lower panel and a two-photon Rabi frequency of 104 kHz.

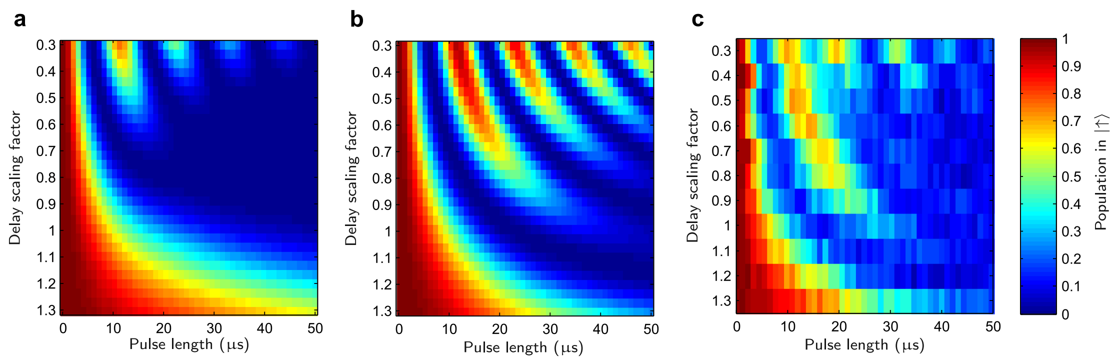

Figure 2. Transfer efficiency for different delay scaling factors and pulse length for carrier transitions for a ground state cooled ion. (a) Simulation using the three level approximation and (b) the full Hamiltonian as well as (c) experimental data for the STIRAP transfer. Red corresponds to no and blue to complete transfer.

For $^{25}Mg^+$ the total interaction Hamiltonian for the STIRAP sequence reads

$$\begin{aligned} H^{\rm STIRAP} &= \hbar \sum_{F,mF} \frac{\Omega_{1,\downarrow}^{F,mF}(t)}{2} |e_{F,m_F}\rangle\langle\downarrow| \, \mathrm{e}^{-\mathrm{i}\Delta_1^{F,m_F}t} \\ &+ \hbar \sum_{F,mF} \frac{\Omega_{1,\uparrow}^{F,mF}(t)}{2} |e_{F,m_F}\rangle\langle\uparrow| \, \mathrm{e}^{-\mathrm{i}\Delta_1^{F,m_F}t} \\ &+ \hbar \sum_{F,mF} \frac{\Omega_{2,\uparrow}^{F,mF}(t)}{2} |e_{F,m_F}\rangle\langle\uparrow| \, \mathrm{e}^{-\mathrm{i}\Delta_2^{F,m_F}t} \\ &+ \hbar \sum_{F,mF} \frac{\Omega_{2,\downarrow}^{F,mF}(t)}{2} |e_{F,m_F}\rangle\langle\downarrow| \, \mathrm{e}^{-\mathrm{i}\Delta_2^{F,m_F}t}, \end{aligned} \tag{5}$$

where $\Delta_i^{F,\,mF}$ is the detuning of laser i from the excited state $|e_{F,m_F}\rangle = |^2P_{3/2}, F, m_F\rangle$ and the Rabi frequency for coupling of laser i to this state is denoted by

$$\Omega_{i,\updownarrow}^{F,mF} = (-1)^{F_\updownarrow - m_{F,\updownarrow}} \begin{pmatrix} F_\updownarrow & 1 & F \\ -m_{F,\updownarrow} & q_i & m_F \end{pmatrix} \Omega_{\updownarrow,F} \tag{6}$$

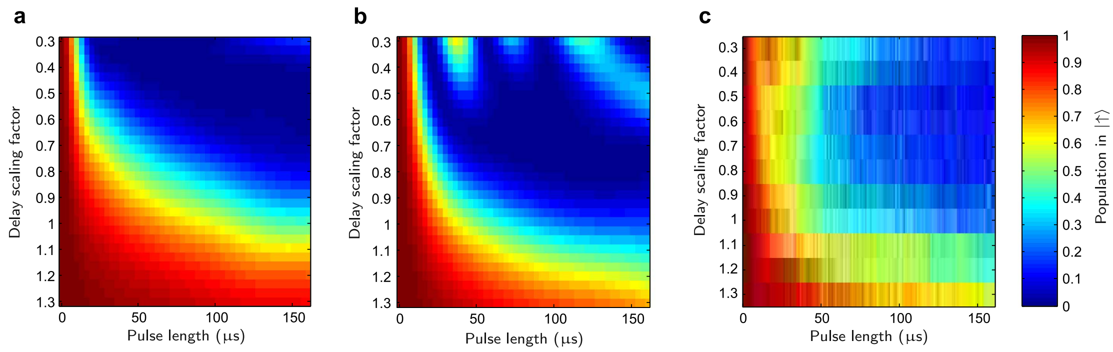

Figure 3. Transfer efficiency for different delay scaling factors and pulse length for blue sideband transitions for a ground state cooled ion. (a) Simulation using the three level approximation and (b) the full Hamiltonian as well as (c) experimental data for the STIRAP transfer. Red corresponds to no and blue to complete transfer.

with the Wigner 3-j symbol, the reduced Rabi frequency $\Omega_{\downarrow,F} = \frac{E\langle^2S_{1/2}, F_\downarrow \parallel \mathbf{d} \parallel^2 P_{3/2}, F\rangle}{\hbar}$ and the laser light polarization q_i. E is the electric field corresponding to the light and $\langle^2S_{1/2},\ F_i\|\mathbf{d}\|^2P_{3/2},\ F\rangle$ is the reduced dipole operator[5]. In total, seven states contribute to the process as can be seen in the partial level scheme in figure 1. Note that coupling to the $^2P_{1/2}$ state has been neglected due to the large detuning. Deriving the effective Hamiltonian [2] and neglecting all terms that couple only off-resonantly different excited substates gives

$$H_{\text{eff}}^{\text{STIRAP}} = -\hbar\sum_{F,mF}\left[\frac{\Omega_{1,\uparrow}^{F,mF}(t)^2}{4\Delta_1^{F,mF}} + \frac{\Omega_{2\uparrow}^{F,mF}(t)^2}{4\Delta_2^{F,mF}}\right](|F,\ m_F\rangle\langle e_{F,m_F}| - |\uparrow\rangle\langle\uparrow|)$$
$$-\hbar\sum_{e_{F,mF}}\left[\frac{\Omega_{1,\downarrow}^{F,mF}(t)^2}{4\Delta_1^{F,mF}} + \frac{\Omega_{2\downarrow}^{F,mF}(t)^2}{4\Delta_2^{F,mF}}\right](|F,\ m_F\rangle\langle F,\ m_F| - |\downarrow\rangle\langle\downarrow|)$$
$$+\frac{\hbar\Omega_{1,\downarrow}^{3,3}(t)\Omega_{2,\uparrow}^{3,3}(t)}{4\Delta}(|\uparrow\rangle\langle\downarrow| + |\downarrow\rangle\langle\uparrow|). \tag{7}$$

The additional couplings compared to the three level case do not influence the coherent population transfer term (last term in equation (7)) but affects the AC-Stark shift terms. For large detunings, the detuning of all states become similar, $\Delta_1 \approx \Delta_2$, and can thus be factored out of the sums in equation (7). Therefore, the differential AC-Stark shift $\delta_{|\downarrow\rangle,|\uparrow\rangle}$ between $|\downarrow\rangle$ and $|\uparrow\rangle$ is proportional to

$$\delta_{|\downarrow\rangle,|\uparrow\rangle} \propto \sum_{F,mF}[\Omega_{1,\uparrow}^{F,mF}(t)^2 + \Omega_{2\uparrow}^{F,mF}(t)^2] \tag{8}$$
$$-\sum_{F,mF}[\Omega_{1,\downarrow}^{F,mF}(t)^2 + \Omega_{2\downarrow}^{F,mF}(t)^2]. \tag{9}$$

Inserting the coupling coefficients for $^{25}Mg^+$ we get

$$\delta_{|\downarrow\rangle,|\uparrow\rangle} \propto -\frac{11}{144}\Omega_{\downarrow,2}(t). \tag{10}$$

However, considering only a three level system gives

$$\delta_{|\downarrow\rangle,|\uparrow\rangle} \propto -\frac{5}{48}\Omega_{\downarrow,1}(t) + \frac{1}{9}\Omega_{\downarrow,2}(t). \tag{11}$$

It is obvious that for the model considering the full magnetic substructure and large detunings from the excited state, the AC-Stark shift is no longer a linear sweep and is strongly reduced compared to the three level case. The right panel in figure 1 shows the AC-Stark shift over time for different detunings, where $\delta = 5.2\delta_{\text{HF}}$ corresponds to the detuning used for the experimental results in the main paper. Already for this detuning the actual AC-Stark shift is smaller compared to the oversimplified three-level-model which results in a different state transfer dynamic. The smaller frequency sweep range due to the AC-Stark shift increases the area in figures 4 and 5 in the main paper, where coherent Rabi flopping between the states occurs. An updated version of these plots and the comparison with the old theory and experimental results is shown in figures 2 and 3.

[5] Note, there are two different conventions for the reduced dipole operator. Here, the notation used by Edmonds [3] (and also [4]) is used. The reduced dipole operator given above, can be further reduced by introducing the 6-j symbol $\langle^2S_{1/2},\ F_i\|\mathbf{d}\|^2P_{3/2},\ F\rangle = (-1)^{J'+I+F+1}\sqrt{(2F_i+1)(2F+1)}\begin{Bmatrix} J'=1/2 & F_i & I=5/2 \\ F & J''=3/2 & 1 \end{Bmatrix}\langle^2S_{1/2}\|\mathbf{d}\|^2P_{3/2}\rangle$, where the further reduced dipole matrix element can be derived from the electronic lifetime of the state τ by $\frac{1}{\tau} = \frac{\omega_0^3}{3\pi\varepsilon_0\hbar c^3}\frac{1}{2J'+1}|\langle^2S_{1/2}\|\mathbf{d}\|^2P_{3/2}\rangle|$.

In the original manuscript we claim that STIRAP population transfer will benefit from increased detuning. This statement is incorrect. The right panel of figure 1 shows that for increasing detuning the AC-Stark shifts from the different levels almost cancel during population transfer. As a consequence, STIRAP is no longer adiabatic and population transfer becomes inefficient. By adding an additional frequency chirp to the pulses, this effect can be compensated and efficient state transfer is recovered. This represents an implementation of RAP [5, 6]. All other conclusions and results of the main paper are unaffected.

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