Supplementary Information: Limits on atomic qubit control from laser noise

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Supplementary Note 1

The shot noise limit

The shot noise of light is determined by the Poisson statistics of photons, which sets a maximum signal to noise ratio (SNR) of an N photon field of $N/\sqrt{N}$. For a detector of bandwidth B the SNR is given by [1]

$$\mathrm{SNR} = \frac{\bar{P}}{\delta P} = \sqrt{\frac{\bar{P}}{2\hbar\omega_{\mathrm{opt}} B}} \;, \tag{1}$$

where $\bar{P}$ is the average power and δP is the RMS power noise. The RMS noise can be related to the single-sided PSD of the laser RIN over a bandwidth B by Parseval's theorem as

$$\frac{\delta P}{\bar{P}} = \sqrt{\frac{1}{2\pi}\int_0^{2\pi B} S_{\mathrm{RIN}}(\omega) d\omega} \;, \tag{2}$$

such that the shot noise limit of the PSD, $S_{\mathrm{RIN}}^{\mathrm{SNL}}$, is given by

$$\frac{\delta P}{\bar{P}} = \sqrt{S_{\mathrm{RIN}}^{\mathrm{SNL}} B} \;, \tag{3}$$

$$\sqrt{\frac{2\hbar\omega_{\mathrm{opt}} B}{\bar{P}}} = \sqrt{S_{\mathrm{RIN}}^{\mathrm{SNL}} B} \;, \tag{4}$$

$$S_{\mathrm{RIN}}^{\mathrm{SNL}} = \frac{2\hbar\omega_{\mathrm{opt}}}{\bar{P}} \;, \tag{5}$$

where the final line is the one given in the main text.

The quantum noise limit

The fundamental floor of laser frequency noise is determined by the quantum limited noise floor of the phase coherence due to spontaneous emission [2]. The resulting laser (FWHM) linewidth, Γ_{QNL} was first derived by Schalow and Townes, and has been later refined to give the expression [2]

$$\Gamma_{\mathrm{QNL}} = \frac{h\omega_{\mathrm{opt}}\gamma_{\mathrm{c}}^2}{2\bar{P}} \;, \tag{6}$$

where we have made the assumption that the cavity bandwidth γ_{c} is much less than the atomic linewidth of the gain medium.

The laser linewidth is well defined for a pure white noise PSD as $\Gamma_{\mathrm{FWHM}} = S_{\omega_{\mathrm{opt}}}/(4\pi)$ such that the white noise spectrum from the Schalow-Townes linewidth is

$$S_{\omega_{\mathrm{opt}}}^{\mathrm{QNL}} = \frac{2\pi h\omega_{\mathrm{opt}}\gamma_{\mathrm{c}}^2}{\bar{P}} \;, \tag{7}$$

which is the expression given in the main text.

Supplementary Table 1: Typical laser intensity (left) and frequency (right) noise parameters for an external cavity diode laser (ECDL) [5–8], diode-pumped solid-state laser (DPSSL)[9–11], modelocked fibre laser (MLFL) [12]. For the MLFL, the frequency noise of interest is in the repetition rate, and no relaxation oscillations are present.

Parameter	ECDL	DPSSL	MLFL
$\omega_{\rm flk}$ (Hz)	$2\pi\times10^4$	$2\pi\times10^4$	$<2\pi\times10^3$
$A_{\rm wht}$ (dB/Hz)	SNL	-140 to -110	-140 to -110
$A_{\rm rlx}$ (dB)	10 to 30	10 to 30	0 to 10
$\omega_{\rm rlx}$ (Hz)	$2\pi\times10^9$	$2\pi\times10^5$	$2\pi\times10^4$

Parameter	ECDL	DPSSL	MLFL
$\omega_{\rm flk}$ (Hz)	$2\pi\times10^4$	$2\pi\times10^4$	$<2\pi\times10^3$
$A_{\rm wht}$ (dB/Hz)	20	-20	20
$A_{\rm rlx}$ (dB)	10 to 30	10 to 30	-
$\omega_{\rm rlx}$ (Hz)	$2\pi\times10^9$	$2\pi\times10^5$	-

Technical laser noise

The susceptibility of a laser to driving and environmental noise sources is given by its modulation transfer function [3]

$$H(\omega)=\frac{\omega_{\rm rlx}^2}{\omega_{\rm rlx}^2-\omega^2+i\omega\gamma}\ , \tag{8}$$

where $\gamma=\omega_{\rm rlx}/\sqrt{A_{\rm rlx}}$ characterises the damping of relaxation oscillations, where $A_{\rm rlx}$ is the amplitude of the relaxation oscillation peak with respect to the asymptotic noise below the oscillation peak.

The technical noise spectrum, for both frequency and intensity noise, can then be modelled as

$$S_j(\omega)=\frac{A_{\rm flk}^{(j)}}{\omega}|H(\omega)|^2+A_{\rm wht}^{(j)}|H(\omega)|^2+A_{\rm FNF}^{(j)}\ , \tag{9}$$

where j is either RIN or $\omega_{\rm opt}$, $A_{\rm FNF}^{(RIN)}=S_{\rm RIN}^{\rm SNL}$, and $A_{\rm FNF}^{(\omega_{\rm opt})}=S_{\omega_{\rm opt}}^{\rm QNL}$. The first term determines the amplitude of flicker noise $A_{\rm flk}^{(j)}=A_{\rm wht}^{(j)}\omega_{\rm flk}$ with corner frequency $\omega_{\rm flk}$ accepted by the transfer function. The second term determines the amplitude of white noise, $A_{\rm wht}^{(j)}$, accepted by the transfer function. While the last term is the fundamental noise floor of the PSD as outlined above.

When using modelocked lasers to driver hyperfine qubits using a beatnote between comb harmonics, the important PSD for determining LO frequency noise is not the optical one, but the PSD of the repetition rate, $\omega_{\rm rep}$. In this case, the modulation transfer function effectively becomes $H(\omega)=1$ as the relaxation dynamics of the gain medium do not limit the upper frequency limit of repetition rate noise. The PSD can then be modelled as

$$S_{\omega_{\rm rep}}(\omega)=\frac{A_{\rm flk}^{(j)}}{\omega}+A_{\rm wht}^{(j)}\ , \tag{10}$$

where we assume the fundamental noise floor of the beatenote linewidth is incorperated into the value of $A_{\rm wht}^{(j)}$. For repetition rate noise, there is an additional fundamental floor due to the quantum limit of the pulse-to-pulse timing. This floor appears in the phase noise spectrum $S_{\phi_{\rm rep}}(\omega)=S_{\omega_{\rm rep}}(\omega)/\omega^2$ and we refer the interested reader to Ref. [4] for further details.

The typical values of the laser noise model parameters vary between laser technologies, and between RIN and frequency noise. We list representative parameters present in the literature from simulations and measured laser sources in Table 1.

Supplementary Note 2

The technical noise present in free-running laser sources limits them from being directly used as qubit LOs. In order to prepare LOs for qubit control from laser radiation, the low-Fourier-frequency technical noise is typically actively suppressed using stabilisation loops. The stabilisation loops take as input an error signal of the laser frequency compared to a stable reference, and provide as output a correction signal back to the laser or to a frequency modulator in the laser beam path. We refer to these stabilised laser sources as LOs. In this section, we relate the laser frequency noise PSDs to the stabilised LO noise PSDs for different qubit types.

Optical qubits For the preparation of optical LOs, the stabilisation reference is typically an ultra-low-expansion cavity where the laser frequency is compared to the length of the cavity by monitoring transmission on a photodiode.

The error signal is fed back onto the laser frequency through any of a variety of modulation techniques. The LO PSD therefore becomes

$$S_{\omega_{\rm LO}}(\omega) = \left|\frac{1}{1+G(\omega)}\right|^2 S_{\omega_{\rm opt}}(\omega) \ , \tag{11}$$

where $G(\omega)$ is the gain transfer function of the servo loop and $G(\omega_{\rm srv}) = 1$ defines the *servo bandwidth*, $\omega_{\rm srv}$. The ultimate frequency stability of the optical LO is limited by the thermal stability of the reference cavity[13]. At room temperature, these thermal fluctuations limit stabilised laser linewidths to approximately 1 Hz. The thermal fluctuations can be reduced by cryogenically cooling the cavity to 4 K, which limits laser linewidths to approximately 10 mHz [14].

Hyperfine qubits: CW lasers For the preparation of a microwave LO from two CW lasers, the lasers must be phase-locked together with an offset frequency equal to the qubit frequency. The phase-locking can either be performed actively, by monitoring the beatnote signal between the two lasers and referencing to a microwave oscillator [15], or passively, by injection locking the secondary laser with frequency-shifted light from the primary laser [16]. In both cases, the resulting LO PSD then becomes

$$S_{\omega_{\rm LO}}(\omega) = \left|\frac{1}{1+G(\omega)}\right|^2 (S_{\omega_{\rm opt,1}}(\omega) + S_{\omega_{\rm opt,2}}(\omega)) \ , \tag{12}$$

where $S_{\omega_{\rm opt,1}}(\omega)$ and $S_{\omega_{\rm opt,2}}(\omega)$ are the frequency noise PSDs of the primary and secondary laser respectively. For active phase locking, the ultimate stability is limited by microwave oscillator stability [17], and $\omega_{\rm srv}$ is limited by the loop delay. For tightly controlled servo-loops with sufficient gain, beatnote linewidths < 1 mHz have been demonstrated for phase-locked DPSSLs [18–20], due to the inherent stability of microwave oscillators. For passive phase locking, the stability is limited by the interferometric stability between the lasers, and $\omega_{\rm srv}$ is limited by the cavity bandwidth, $\gamma_{\rm c}$, of the laser sources [21]. A beatnote linewidth of 42 mHz has been measured for a pair of passively injected locked diode lasers [16].

Hyperfine qubits: ML lasers LOs derived from modelocked lasers differ from the other examples highlighted above, as the optical frequency noise, $S_{\omega_{\rm opt}}(\omega)$, does not contribute to $S_{\omega_{\rm LO}}(\omega)$ [4]. The LO noise is determined by the timing jitter in the pulse train, or equivalently, the frequency noise on the repetition rate, $S_{\omega_{\rm rep}}(\omega)$. As the repetition rate, $\omega_{\rm rep}$, is typically much less than the qubit frequency, $\omega_{\rm qbt}$, the LO is generated from a higher-order harmonic, m, such that $\omega_{\rm LO} = m\omega_{\rm rep} + \omega_{\rm aom}$ where $\omega_{\rm aom}$ is the frequency imparted on an AOM in one beam path. Therefore, the LO PSD is a combination of noise in the laser, and noise in the AOM, $S_{\omega_{\rm LO}}(\omega) = m^2 S_{\omega_{\rm rep}}(\omega) + S_{\omega_{\rm aom}}(\omega)$. The stabilisation of the beatnote can be performed by modulating the AOM frequency, which is typically performed with low bandwidth to remove the effect of repetition rate drift over time due to thermal effects [22–25]. The LO frequency noise then becomes

$$S_{\omega_{\rm LO}}(\omega) = \left|\frac{m}{1+G(\omega)}\right|^2 S_{\omega_{\rm rep}}(\omega) + S_{\omega_{\rm aom}}(\omega) \ , \tag{13}$$

where we have assumed a feed forward stabilisation loop that does not monitor the excess noise from the AOM and only stabilises the $m^{\rm th}$ harmonic. $G_m(\omega)$ is the loop transfer function unique to harmonic m. The LO stability is then limited by the reference clock stability of the loop.

Cascaded qubits In the case of a LO for driving a two-photon optical transition, the two lasers are typically of different wavelength. Therefore, they cannot be phase-locked are their noise spectra are uncorrelated. If the two lasers have frequencies of $\omega_{\rm opt,1}$ and $\omega_{\rm opt,2}$, then $\omega_{\rm LO} = \omega_{\rm opt,1} + \omega_{\rm opt,2}$ such that the frequency noise PSD of the LO is given by

$$S_{\omega_{\rm LO}}(\omega) = \left|\frac{1}{1+G_1(\omega)}\right|^2 S_{\omega_{\rm opt},1}(\omega) + \left|\frac{1}{1+G_2(\omega)}\right|^2 S_{\omega_{\rm opt},2}(\omega) \ , \tag{14}$$

where $G_{1,2}$ are the separate loop gains of the separate servo loops for each laser.

To relate the above expressions to parameters used in the text, recall the simple model used in the main text for a servoed noise PSD

$$S_{\rm srv}(\omega) = \begin{cases} h_{\rm a} & \text{for } \omega < \omega_{\rm srv} \\ h_{\rm b} & \text{for } \omega > \omega_{\rm srv} \end{cases} \ . \tag{15}$$

where $h_{\rm b} = A^{(j)}_{\rm wht}$ is the residual free-running white noise outside the stabilisation loop. The value of $h_{\rm a}$ is determined by the higher of the noise with the stabilisation loop (determined by the gain transfer function, $G(\omega)$) or the fundamental noise floor of the servo. For the purposes of this manuscript, we make the assumption that the servo loop has been designed such that the noise spectrum takes the form of white noise below the servo bandwidth. We also make the assumption in the case of cascaded qubits, that is the gain transfer functions are identical for the two laser sources such that $G(\omega) = G_1(\omega) + G_2(\omega)$. Under this approximation, Equation 14 reduces to Equation 12 and becomes identical to that of phase-locked lasers addressing a hyperfine qubit.

Supplementary Note 3

Consider the expression for fidelity in terms of the quantum operation filter function, $F_z^{(u)}(\omega)$, and the control noise power spectral density, $S_z(\omega)$

$$\mathcal{F}^{(u)}(\tau) = \frac{1}{2}\left\{1 + \exp[-\chi^{(u)}(\tau)]\right\} , \tag{16}$$

with

$$\chi^{(u)}(\tau) = \frac{1}{\pi}\int_0^\infty \frac{d\omega}{\omega^2} S_z(\omega) F_z^{(u)}(\omega) . \tag{17}$$

Now consider the simple model PSD of frequency noise on the local oscillator that drives the qubit transition:

$$S_{\omega_{\rm LO}}(\omega) = \begin{cases} h_{\rm a} & \text{for } \omega < \omega_{\rm srv} , \\ h_{\rm b} & \text{for } \omega > \omega_{\rm srv} , \end{cases} \tag{18}$$

such that $h_{\rm a}$ is a constant white noise amplitude within the servo bandwidth, $\omega_{\rm srv}$, and $h_{\rm b}$ is the residual free-running white noise amplitude outside the servo bandwidth. The relation between the PSD in the Z basis and the LO frequency noise is

$$S_z(\omega) = \frac{1}{4} S_{\omega_{\rm LO}}(\omega) . \tag{19}$$

The filter function (FF) is dependent on the exact quantum operation being performed, however to first order it has the structure of a high pass filter, with a flat acceptance band above the cut-on frequency, $\omega_{\rm cut}$, and a fall off in the filter below $\omega_{\rm cut}$ such that it can be represented by the piecewise function

$$F_z(\omega) = \begin{cases} c_a^{(u)}\omega^n & \text{for } \omega < \omega_{\rm cut} , \\ c_b^{(u)} & \text{for } \omega > \omega_{\rm cut} , \end{cases} \tag{20}$$

where the order, n, of the filter function controls how efficiently low frequency noise can be suppressed.

To derive the dependence of the control error on the parameters of the simple model PSD, we substitute Equations 18 and 20 into Equation 17 such that

$$\chi^{(u)}(\tau) = \frac{1}{4\pi}\left[\int_0^{\omega_{\rm srv}} \frac{d\omega}{\omega^2} h_{\rm a} F_z^{(u)}(\omega) + \int_{\omega_{\rm srv}}^\infty \frac{d\omega}{\omega^2} h_{\rm b} F_z^{(u)}(\omega)\right] , \tag{21}$$

$$= \frac{1}{4\pi}\left[\int_0^{\omega_{\rm cut}} \frac{d\omega}{\omega^2} h_{\rm a} c_a^{(u)}\omega^n + \int_{\omega_{\rm cut}}^{\omega_{\rm srv}} \frac{d\omega}{\omega^2} h_{\rm a} c_b^{(u)} + \int_{\omega_{\rm srv}}^\infty \frac{d\omega}{\omega^2} h_{\rm b} c_b^{(u)}\right] , \tag{22}$$

$$= \frac{1}{4\pi}\left[h_{\rm a} c_a^{(u)} \frac{\omega_{\rm cut}^{n-1}}{n-1} + h_{\rm a} c_b^{(u)}\left(\frac{1}{\omega_{\rm cut}} - \frac{1}{\omega_{\rm srv}}\right) + h_{\rm b} c_b^{(u)} \frac{1}{\omega_{\rm srv}}\right] , \tag{23}$$

$$= \frac{1}{4\pi}\left[h_{\rm a}\left(c_a^{(u)} \frac{\omega_{\rm cut}^{n-1}}{n-1} + \frac{c_b^{(u)}}{\omega_{\rm cut}}\right) + c_b^{(u)} \frac{h_{\rm b} - h_{\rm a}}{\omega_{\rm srv}}\right] . \tag{24}$$

$$\tag{25}$$

The first term in the final equation is the interaction of the noise within the stabilisation loop ($h_{\rm a}$) with the cut-off frequency of the quantum operation. The second term determines the amount of noise coming from outside the servo and its interaction with the qubit. Increasing the servo bandwidth to infinity leaves only the effect of the stabilised noise. The point at which these two terms are equal is given by

$$h_{\rm a}\left(c_a^{(u)}\frac{\omega_{\rm cut}^{n-1}}{n-1}+\frac{c_b^{(u)}}{\omega_{\rm cut}}\right)=c_b^{(u)}\frac{h_{\rm b}-h_{\rm a}}{\omega_{\rm srv}}\ , \tag{26}$$

$$\omega_{\rm srv}=\frac{c_b^{(u)}}{c_a^{(u)}\frac{\omega_{\rm cut}^{n-1}}{n-1}+\frac{c_b^{(u)}}{\omega_{\rm cut}}}\left(\frac{h_{\rm b}}{h_{\rm a}}-1\right)\ . \tag{27}$$

Using the continuity condition $h_{\rm b}^{(u)}=h_{\rm a}^{(u)}\omega_{\rm cut}^{n}$ we find

$$\begin{aligned}\omega_{\rm srv}&=\frac{c_a^{(u)}\omega_{\rm cut}^{n}}{c_a^{(u)}\frac{\omega_{\rm cut}^{n-1}}{n-1}+c_a^{(u)}\omega_{\rm cut}^{n-1}}\left(\frac{h_{\rm b}}{h_{\rm a}}-1\right)\ , &(28)\\ &=\frac{\omega_{\rm cut}^{n}}{\frac{\omega_{\rm cut}^{n-1}}{n-1}+\omega_{\rm cut}^{n-1}}\left(\frac{h_{\rm b}}{h_{\rm a}}-1\right)\ , &(29)\\ &=\frac{(n-1)\omega_{\rm cut}}{n}\left(\frac{h_{\rm b}}{h_{\rm a}}-1\right)\ , &(30)\\ & &(31)\end{aligned}$$

which for a primitive π-pulse ($n=2$ and $\omega_{\rm cut}=\Omega$, where $\Omega=\pi/\tau$ is the Rabi frequency) is

$$\omega_s^{(\pi)}=\frac{\Omega}{2}\left(\frac{h_{\rm b}}{h_{\rm a}}-1\right)\ . \tag{32}$$

To investigate the fidelity dependence, we impose the continuity condition on χ, which is better understood in terms of $c_b^{(u)}$ such that

$$\begin{aligned}\chi^{(u)}(\tau)&=\frac{1}{4\pi}\left[h_{\rm a}\left(c_a^{(u)}\frac{\omega_{\rm cut}^{n-1}}{n-1}+\frac{c_b^{(u)}}{\omega_{\rm cut}}\right)+c_b^{(u)}\frac{h_{\rm b}-h_{\rm a}}{\omega_{\rm srv}}\right]\ , &(33)\\ &=\frac{c_b^{(u)}}{4\pi}\left[\frac{n}{n-1}\frac{h_{\rm a}}{\omega_{\rm cut}}+\frac{h_{\rm b}-h_{\rm a}}{\omega_{\rm srv}}\right]\ , &(34)\end{aligned}$$

From this expression we can easily recover the specific result for the π pulse, as well as the general requirement on the servo bandwidth.

For the regime when $\omega_{\rm srv}<\Omega$ we follow a similar derivation

$$\begin{aligned}\chi^{(u)}(\tau)&=\frac{1}{4\pi}\left[\int_0^{\omega_{\rm srv}}\frac{d\omega}{\omega^2}h_{\rm a}F_z^{(u)}(\omega)+\int_{\omega_{\rm srv}}^{\infty}\frac{d\omega}{\omega^2}h_{\rm b}F_z^{(u)}(\omega)\right]\ , &(35)\\ &=\frac{1}{4\pi}\left[\int_0^{\omega_{\rm srv}}\frac{d\omega}{\omega^2}h_{\rm a}c_a^{(u)}\omega^n+\int_{\omega_{\rm srv}}^{\omega_{\rm cut}}\frac{d\omega}{\omega^2}h_{\rm b}c_a^{(u)}\omega^n+\int_{\omega_{\rm cut}}^{\infty}\frac{d\omega}{\omega^2}h_{\rm b}c_b^{(u)}\right]\ , &(36)\\ & &(37)\end{aligned}$$

which after some rearranging gets to

$$\chi^{(u)}(\tau)=\frac{c_b^{(u)}h_{\rm b}}{4\pi(n-1)}\left[\frac{n}{\omega_{\rm cut}}-\frac{\omega_{\rm srv}^{n-1}}{\omega_{\rm cut}^{n}}\left(1-\frac{h_{\rm a}}{h_{\rm b}}\right)\right]\approx\frac{nc_b^{(u)}h_{\rm b}}{4\pi(n-1)\omega_{\rm cut}}\ , \tag{38}$$

For a primitive pulse this becomes $\chi^{(u)}(\tau)\approx 2h_{\rm b}/(\pi\Omega)$ which is just the expected error from a constant white noise source of magnitude $h_{\rm b}$.

From the equation for the servo bandwidth we can view it as a reciprocal relationship between the bandwidth and the free-running noise, $h_{\rm b}$. For a given Fourier frequency, ω, this sets a maximum value of $h_{\rm b}$ that the PSD can have at that point in order for it not to adversely affect the qubit fidelity. To get an expression of this boundary line we first recall the condition on the servo bandwidth

$$\omega_{\rm srv} \geq \frac{(n-1)\omega_{\rm cut}}{n}\left(\frac{h_{\rm b}}{h_{\rm a}}-1\right) \ . \tag{39}$$

We can reframe this expression by acknowledging that it sets a maximum value of $h_{\rm b}$ at $\omega_{\rm srv}$ in order to satisfy the condition

$$\max(h_{\rm b}) = h_{\rm a}\left(\frac{n\omega_{\rm srv}}{(n-1)\omega_{\rm cut}}+1\right) \ , \tag{40}$$

which can be rewritten

$$S_{\omega_{\rm LO}}(\omega_{\rm srv}) = h_{\rm a}\left(\frac{n\omega_{\rm srv}}{(n-1)\omega_{\rm cut}}+1\right) \ , \tag{41}$$

This line of varying $\omega_{\rm srv}$ can be plotted on a Fourier frequency space plot (by replacing $\omega_{\rm srv}$ with ω) to divide the spectrum into the region that contributes to the fidelity and a region that doesn't.

For a π-pulse we obtain

$$S_{\omega_{\rm LO}}(\omega_{\rm srv}) = h_{\rm a}\left(\frac{2\omega_{\rm srv}}{\Omega}+1\right) \ . \tag{42}$$

Supplemetary Note 4

Consider the time-varying electric field of a local oscillator at frequency $\omega_{\rm LO}$

$$E(t) = E_0\cos(\phi_{\rm LO}(t)) \tag{43}$$

where $\phi_{\rm LO}(t) = \int \omega_{\rm LO} dt$. Modulating the LO at a Fourier frequency ω_j with an amplitude of $\delta\omega_{\rm LO}^{(j)}$ transforms the LO frequency as

$$\omega_{\rm LO} \rightarrow \omega_{\rm LO} + \delta\omega_{\rm LO}^{(j)}\cos(\omega_j t) \ . \tag{44}$$

The noisy local oscillator field is now

$$E(t) = E_0\cos\left[\omega_{\rm LO}t + \frac{\delta\omega_{\rm LO}^{(j)}}{\omega_j}\sin(\omega_j t)\right] \ . \tag{45}$$

We define the *modulation index* at Fourier frequency ω_j as

$$\beta_j = \frac{\delta\omega_{\rm LO}^{(j)}}{\omega_j} = \frac{\delta\nu_{\rm LO}^{(j)}}{f_j} \ , \tag{46}$$

where $\omega_j = 2\pi f_j$ and $\delta\omega_{\rm LO}^{(j)} = 2\pi\delta\nu_{\rm LO}^{(j)}$.

The intensity of the local oscillator in the Fourier domain is given by the Fourier transform of the absolute square of the electric field [26]

$$I(\omega) \propto F[|E(t)|^2] \propto \sum_{n=-\infty}^{n=+\infty} |J_n(\beta_j)|^2\delta(\omega = \omega_{\rm LO} \pm \omega_j n) \ , \tag{47}$$

where $J_n(\beta_j)$ are the Bessel functions of order n. Note that for $\beta_j < 1$ then the dominant noise term is $n = 1$ and the modulation causes single sidebands around the carrier. For $\beta_j > 1$ a spectrum of sidebands is created such that the noise is broadened. As $\beta \propto 1/\omega$, this excess noise occurs at low frequencies, with the importance at higher frequencies diminishing. Further, when $\beta_j < 1$, $|J_n(\beta_j)|^2 \propto \beta_j^2$ such that the magnitude of the sideband falls off rapidly in Fourier frequency with a $1/\omega^2$ dependence.

Now consider that $\delta\omega_{\mathrm{LO}}^{(j)}$ is just one noise component in a spectrum of noise given by the PSD $S_{\omega_{\mathrm{LO}}}(\omega)$, the amplitude at ω_j can be calculated from the RMS power

$$(\delta\omega_{\mathrm{LO}}^{(j)})^2 = 2(\delta\omega_{\mathrm{LO,rms}}^{(j)})^2 = \frac{1}{\pi}\int_0^{\omega_j} S_{\omega_{\mathrm{LO}}}(\omega)d\omega \ , \tag{48}$$

To maintain completeness, also note the relation for $\delta\nu_{\mathrm{LO}}^{(j)}$

$$(\delta\nu_{\mathrm{LO}}^{(j)})^2 = 2\int_0^{f_j} S_{\nu_{\mathrm{LO}}}(f)df \ . \tag{49}$$

For purely Lorentzian lineshapes, the power spectral density takes a constant value $S_{\omega_{\mathrm{LO}}}(\omega_j) = h_{\omega_{\mathrm{LO}}}^{(j)}$, $S_{\nu_{\mathrm{LO}}}(f_j) = h_{\nu_{\mathrm{LO}}}^{(j)}$ such that

$$(\delta\omega_{\mathrm{LO}}^{(j)})^2 = \frac{1}{\pi}h_{\omega_{\mathrm{LO}}}^{(j)}\omega_j \tag{50}$$

$$(\delta\nu_{\mathrm{LO}}^{(j)})^2 = 2h_{\nu_{\mathrm{LO}}}^{(j)} f_j \ , \tag{51}$$

and the modulation index can be given in terms of the value of the PSD at ω_j [26]

$$\beta_j = \sqrt{\frac{h_{\omega_{\mathrm{LO}}}^{(j)}}{\pi\omega_j}} = \sqrt{\frac{2h_{\nu_{\mathrm{LO}}}^{(j)}}{f_j}} \ , \tag{52}$$

Rearranging we find $h_{\omega_{\mathrm{LO}}}^{(j)} = (2\pi)^2 h_{\nu_{\mathrm{LO}}}^{(j)}$. More generally it can be shown that $S_{\omega_{\mathrm{LO}}}(f) = (2\pi)^2 S_{\nu_{\mathrm{LO}}}(f)$ [27].

The laser lineshape, $S(\nu)$, where ν is a "physical" frequency as would be measured on a spectrum analyzer, can be calculated as the Fourier transform of the autocorrelation function, $\Gamma(\tau)$ of the time-varying electric field [28]

$$S(\nu) = F[\Gamma(\tau)] \ , \tag{53}$$

where

$$\Gamma(\tau) = E^*(t)E(t+\tau) = E_0^2 \mathrm{e}^{\mathrm{i}\omega_{\mathrm{LO}}\tau}\exp\left[-2\int_0^\infty \frac{S_{\nu_{\mathrm{LO}}}(f)}{f^2}\sin^2(\pi f\tau)df\right] \ . \tag{54}$$

The width of the lineshape is governed by how quickly the exponential term decays, and therefore the linewidth is determined by

$$H(\tau) = \exp\left[-2\int_0^\infty \frac{S_{\nu_{\mathrm{LO}}}(f)}{f^2}\sin^2(\pi f\tau)df\right] \ . \tag{55}$$

For simplicity, assume a white noise spectrum (model of an ideal laser source), $S_{\nu_{\mathrm{LO}}}(f) = h_0$ such that

$$H(\tau) = \exp\left[-2\int_0^\infty \frac{h_0}{f^2}\sin^2(\pi f\tau)df\right] \tag{56}$$

$$= \exp\left[-\frac{1}{2h_0}\int_0^\infty \frac{4h_0^2}{f^2}\sin^2(\pi f\tau)df\right] \tag{57}$$

$$= \exp\left[-\frac{1}{2h_0}\int_0^\infty \beta^4\sin^2(\pi f\tau)df\right] \tag{58}$$

where we have refactored in terms of the modulation index. The FWHM linewidth is determined by the condition $H(\tau) = 1/2$. For this constant white noise spectrum, the FWHM linewidth is given by [29]

$$\Gamma_{\mathrm{FWHM}} = \pi h_0 \ , \tag{59}$$

which for the corresponding PSD, $S_{\omega_{\mathrm{LO}}}(f) = h_0'$, is

$$\Gamma_{\mathrm{FWHM}} = \frac{\pi h_0}{(2\pi)^2} = \frac{h_0'}{4\pi} \ . \tag{60}$$

To further illustrate, consider a white noise PSD that is low-pass filtered at f_c

$$S_{\nu_{\mathrm{LO}}} = \begin{cases} h_0 & \text{for } f < f_c \ , \\ 0 & \text{for } f > f_c \ . \end{cases} \tag{61}$$

The linewidth for such a spectrum can be found to be approximated by [28]

$$\Gamma_{\mathrm{FWHM}} = h_0 \frac{\sqrt{8\ln(2) f_c / h_0}}{\left[1 + \left(\frac{8\ln(2) f_c}{\pi^2 h_0}\right)^2\right]^{1/4}} \ , \tag{62}$$

which we can rewrite by noting that $f_c/h_0 = 2/\beta_c^2$ where β_c is the modulation index at the cut off frequency such that

$$\Gamma_{\mathrm{FWHM}} = h_0 \frac{\sqrt{16\ln(2)/\beta_c^2}}{\left[1 + \left(\frac{16\ln(2)}{\pi^2 \beta_c^2}\right)^2\right]^{1/4}} \ , \tag{63}$$

For $\beta_c > 1$, as the cut-off frequency increases (decreasing values of β_c) the linewidth increases. At $\beta_c = 1$ the linewidth asymptotes to a constant value of $\Gamma_{\mathrm{FWHM}} = \pi h_0$. Therefore, noise of low modulation index ($\beta < 1$) does not contribute to the linewidth of the laser radiation.

By using the expression for β_c with $\beta_c = 1$ we find that the cut-off frequency that corresponds to this roll off

$$f_c^* = 2h_0 \ , \tag{64}$$

such that the β-separation line is give by

$$S_{\nu_{\mathrm{LO}}}(f) = f/2 \ , \tag{65}$$

$$S_{\omega_{\mathrm{LO}}}(\omega) = \pi\omega \ , \tag{66}$$

which is the β-separation line used in the main text and is valid for white noise PSDs. Domenico et al. numerically found the more general relation $S_{\nu_{\mathrm{LO}}}(f) = 8\ln(2)/\pi^2 f \approx 0.56 f$ [28], and we use the form in Equation 66 for simplicity as it does not affect our results. Domenico et al. show that the β-separation line correctly predicts the transition from free-running linewidth to narrowed linewidth in the simple model used in the main text [28].

Supplementary Note 5

In order to estimate the shot noise limited intensity error for an optical qubit, we must come up with a conversion from Rabi frequency of the quadrupole transition to the optical laser power. We follow the derivation from reference [30]. By solving the Hamiltonian for a quadrupolar transition driven by a laser, we get the resultant Rabi frequency of

$$\Omega = \left| \frac{eE}{\hbar} \langle g | \hat{r}_i \hat{r}_j | e \rangle \epsilon_i n_j \right| \ , \tag{67}$$

where e is the electron charge, E is the electric field of the light field, $\langle g|$ is the lower ground state, $|e\rangle$ is the upper excited state, $\hat{r}_i$ is the ith component of the position operator for the valence electron, ϵ_i is the polarization vector, and n_i is the unit vector. We can rewrite the matrix elements in terms of Racah tensors as follows:

$$\langle g | \hat{r}_i \hat{r}_j | e \rangle \epsilon_i n_j = \langle g || r^2 C^{(2)} || e \rangle \sum_{q=-2}^{2} \begin{pmatrix} J & 2 & J' \\ -m & q & m' \end{pmatrix} c_{ij}^{(q)} \epsilon_i n_j \ , \tag{68}$$

where the braket is now the fully reduced matrix element of the transition, the first factor in the summation is the three-j symbol, J and J' are the ground and excited state total angular momenta, and $c_{ij}^{(q)}$ are the Racah tensor matrices. The reduced matrix element is related to the total rate of decay from the excited state by

$$A_{ge} = \frac{c\alpha k^5}{15(2J'+1)} \left| \langle g || r C^{(2)} || e \rangle \right|^2 \ , \tag{69}$$

where c is the speed of light in vacuum, α is the fine-structure constant, and k is the wavevector of the laser. Following the convention of James [31] we define a relative coupling strength of a transition as

$$\sigma = \sqrt{\frac{15(2J'+1)}{4}} \sum_{q=-2}^{2} \begin{pmatrix} J & 2 & J' \\ -m & q & m' \end{pmatrix} c_{ij}^{(q)} \epsilon_i n_j \ , \tag{70}$$

for an isotope with no nuclear spin ($I = 0$) and

$$\sigma = \sqrt{\frac{15(2J'+1)}{4}} \sqrt{(2F'+1)(2J+1)} \begin{Bmatrix} J & J' & 2 \\ F' & F & I \end{Bmatrix} \sum_{q=-2}^{2} \begin{pmatrix} J & 2 & J' \\ -m & q & m' \end{pmatrix} c_{ij}^{(q)} \epsilon_i n_j \ , \tag{71}$$

Performing a numerical search over all quadrupole transitions with $0 \leq I \leq 9/2$, $0 \leq J \leq 2$ and $1 < J' < 5/2$ over all polarizations finds $\sigma < 1.06$. The resulting on-resonant Rabi frequency is given by

$$\Omega = \frac{e|E|}{\hbar} \sqrt{\frac{A_{ge}\lambda^3}{8\pi^3 c\alpha}} \sigma \ , \tag{72}$$

where λ is the wavelength of the light. From the relationship between electric field and laser power $E = \sqrt{2P/c\epsilon_0 \pi w_0^2}$ (ϵ_0 is the permittivity of free space and w_0 is the beam waist), we can write the final result of laser power as a function of Rabi frequency

$$P = \frac{4\pi^4 \epsilon_0 \alpha}{{}^2 A_{ge}\lambda^3} \left(\frac{\hbar c w_0 \Omega}{e} \right)^2 = \frac{\pi^3 \hbar c}{\sigma^2 A_{ge} \lambda^3} w_0^2 \Omega^2 \ . \tag{73}$$

Supplementary Note 6

The fidelity decay constant due to servoed intensity noise for a $\pi-$pulse is given by (see main text)

$$\chi^{(\pi)} = \frac{\kappa \Omega^2}{\pi} \left[\frac{\pi h_{\mathrm{a}}}{\Omega} + \frac{h_{\mathrm{b}} - h_{\mathrm{a}}}{\omega_{\mathrm{srv}}} \right] \ , \tag{74}$$

where $\kappa = 1$ for Raman transitions and $\kappa = 1/4$ for optical transitions. To ensure that this error is below the error from spontaneous emission we set $\epsilon_{\mathrm{SE}} > \chi^{(\pi)}/2$ and get the requirement on the servo bandwidth

$$\omega_{\mathrm{srv}} > \left(\frac{2\pi\epsilon_{\mathrm{SE}}}{\kappa \Omega^2 h_{\mathrm{a}}} - \frac{\pi}{\Omega} \right)^{-1} \left(\frac{h_{\mathrm{b}}}{h_{\mathrm{a}}} - 1 \right) \ , \tag{75}$$

and therefore, for ω_{srv} to be positive and finite we set the requirement

$$\frac{2\pi\epsilon_{\mathrm{SE}}}{\kappa \Omega^2 h_{\mathrm{a}}} > \frac{\pi}{\Omega} \ , \tag{76}$$

which places the constraint

$$h_{\mathrm{a}} < \frac{2\epsilon_{\mathrm{SE}}}{\kappa \Omega} \ . \tag{77}$$

The ultimate lowest limit on the value of h_{a} is given by the shot noise limit, which recall takes the form

$$S_{\mathrm{RIN}}^{\mathrm{SNL}}(\omega) = \frac{2\hbar\omega_{\mathrm{opt}}}{\bar{P}} \ , \tag{78}$$

which in terms of wavelength of the laser light, λ_{O} sets the limit

$$h_{\mathrm{a}} \geq \frac{2hc}{\lambda_{\mathrm{O}} \bar{P}} \ , \tag{79}$$

As the shot noise limit is the lowest possible intensity noise for a laser source, it corresponds to the lowest possible qubit error. Therefore the task is now to find when the SNL is below the SE floor

$$\frac{2hc}{\lambda_{\mathrm{O}}\bar{P}} < \frac{2\epsilon_{\mathrm{SE}}}{\kappa\Omega} \;, \tag{80}$$

To simplify this expression further, we use the fact that for a given Rabi frequency, there is a fixed required laser intensity at the atom position. The exact relation depends on atomic and laser parameters, and is different for optical and Raman transitions.

For two-photon transitions, the power is given by[32]

$$\begin{aligned}
P_{2\gamma} &= \frac{4\pi}{3\epsilon_{\mathrm{SE}}} w_0^2 \hbar c k_{3/2}^3 \Omega \;, &(81)\\
&= \frac{4\pi}{3\epsilon_{\mathrm{SE}}} w_0^2 \hbar c \left(\frac{2\pi}{\lambda_{3/2}}\right)^3 \Omega \;, &(82)\\
&= \frac{32\pi^4}{3\epsilon_{\mathrm{SE}}\lambda_{3/2}^3} w_0^2 \hbar c \Omega \;, &(83)\\
&= \frac{16\pi^3}{3\epsilon_{\mathrm{SE}}\lambda_{3/2}^3} w_0^2 h c \Omega \;. &(84)
\end{aligned}$$

We now substitute this expression into Equation 80 with $\kappa = 1$ to find

$$\begin{aligned}
\frac{2hc}{\lambda_{\mathrm{O}} \frac{16\pi^3}{3\epsilon_{\mathrm{SE},2\gamma}\lambda_{3/2}^3} w_0^2 h c \Omega} &< \frac{2\epsilon_{\mathrm{SE}}}{\Omega} \;, &(85)\\
\frac{1}{\lambda_{\mathrm{O}} \frac{8\pi^3}{3\epsilon_{\mathrm{SE},2\gamma}\lambda_{3/2}^3} w_0^2 \Omega} &< \frac{2\epsilon_{\mathrm{SE}}}{\Omega} \;, &(86)\\
\frac{3\epsilon_{\mathrm{SE}}\lambda_{3/2}^3}{\lambda_{\mathrm{O}} 8\pi^3 w_0^2 \Omega} &< \frac{2\epsilon_{\mathrm{SE}}}{\Omega} \;, &(87)\\
\frac{\lambda_{3/2}^3}{\lambda_{\mathrm{O}} 8\pi^3 w_0^2} &< \frac{2}{3} \;, &(88)\\
& &(89)
\end{aligned}$$

Now making the assumption that $\lambda_{3/2} \approx \lambda_{\mathrm{O}}$, which sets a stricter bound because $\lambda_{3/2} > \lambda_{\mathrm{O}}$, such that

$$\begin{aligned}
\frac{\lambda_O^2}{8\pi^3 w_0^2} &< \frac{2}{3} \;, &(90)\\
\frac{\lambda_O^2}{\pi^2 w_0^2} &< \frac{16\pi}{3} \;, &(91)\\
& &(92)
\end{aligned}$$

and we recognise that $\mathrm{NA} = \lambda_{\mathrm{O}}/(\pi w_0)$ such that

$$\begin{aligned}
\mathrm{NA}^2 &< \frac{16\pi}{3} \;, &(93)\\
\mathrm{NA} &< 4\sqrt{\frac{\pi}{3}} \approx 4 \;, &(94)\\
& &(95)
\end{aligned}$$

and therefore, for two-photon transitions, the shot noise limit of light is always below the SE floor up to the Abbe limit of $\mathrm{NA} = 1$.

For optical qubits the required power is given by

$$P_{1\gamma} = \frac{\pi^3 \hbar c}{\sigma^2 \Gamma \lambda_{\mathrm{O}}^3} w_0^2 \Omega^2 \ , \tag{96}$$

$$= \frac{\pi^2 h \tau_e c}{2\sigma^2 \lambda_{\mathrm{O}}^3} w_0^2 \Omega^2 \ , \tag{97}$$

$$= \frac{\pi^2 h c}{2\sigma^2 \lambda_{\mathrm{O}}^3} w_0^2 \Omega \frac{3\pi}{8} \left(\frac{8\tau_e \Omega}{3\pi} \right) \ , \tag{98}$$

$$= \frac{3\pi^3 h c}{16\sigma^2 \lambda_{\mathrm{O}}^3 \epsilon_{\mathrm{SE}}} w_0^2 \Omega \ , \tag{99}$$

$$\tag{100}$$

where we have used the relation $\epsilon_{\mathrm{SE}} = 3\pi/(8\Omega\tau_e)$. The condition on the noise to be below the SE floor then becomes

$$\frac{2hc}{\lambda_{\mathrm{O}} \frac{3\pi^3 hc}{16\sigma^2 \lambda_{\mathrm{O}}^3 \epsilon_{\mathrm{SE}}} w_0^2 \Omega} < \frac{8\epsilon_{\mathrm{SE}}}{\Omega} \ , \tag{101}$$

$$\frac{2\sigma^2}{\frac{3\pi^3}{16\lambda_{\mathrm{O}}^2 \epsilon_{\mathrm{SE}}} w_0^2 \Omega} < \frac{8\epsilon_{\mathrm{SE}}}{\Omega} \ , \tag{102}$$

$$\frac{\sigma^2}{\frac{3\pi^3}{16\lambda_{\mathrm{O}}^2} w_0^2} < 4 \ , \tag{103}$$

$$\frac{\lambda_{\mathrm{O}}^2}{\pi^2 w_0^2} < \frac{12\pi}{16\sigma^2} \ , \tag{104}$$

$$\mathrm{NA}^2 < \frac{12\pi}{16\sigma^2} \ , \tag{105}$$

$$\mathrm{NA} < \sqrt{\frac{12\pi}{16}} \frac{1}{\sigma} \ , \tag{106}$$

such that for a maximum found value of $\sigma = 1.06$, $\mathrm{NA} < 1.44$ which is always satisfied as it is above the Abbe limit of $\mathrm{NA} = 1$.

Supplementary Note 7

We plot typical values of the SE floor as given by Equations (15) and (16) of the main text for typical atomic species in Supplementary Figure 1.

Supplementary Note 8

Given an LO generated from a modelocked laser, the AC Stark shift imparted on the qubit splitting is given by [33]

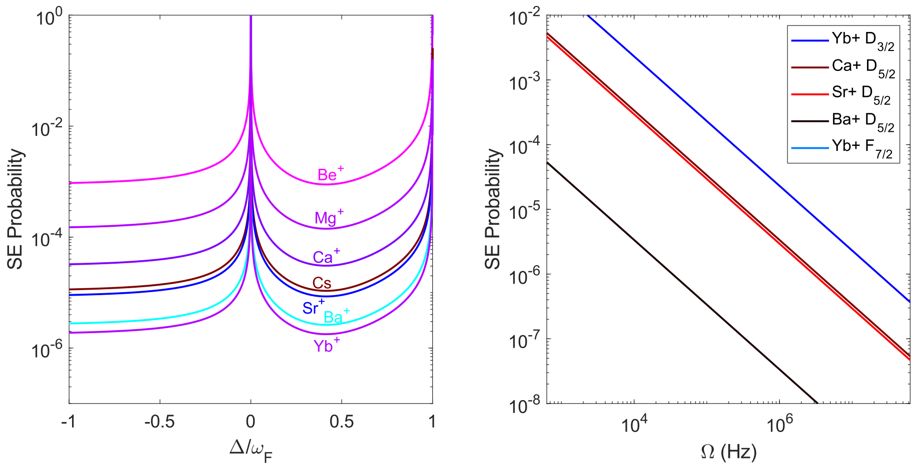
$$\Delta_{\mathrm{AC}}^{(\mathrm{fc})} = \frac{\Omega_0^2}{2\omega_{\mathrm{rep}}} \left[3.412\omega_{\mathrm{qbt}}\tau_p + \mathrm{sech}^2(\omega_{\mathrm{qbt}}\tau_p/2) \left(\frac{1}{\sigma} + \frac{1}{1+2\sigma} + \frac{1}{1-2\sigma} \right) \right] \ , \tag{107}$$

where $\sigma = [\omega_{\mathrm{aom}} (\mathrm{mod}\ \omega_{\mathrm{rep}})]/\omega_{\mathrm{rep}}$ and takes the values from 0 to 1 (with $\sigma \neq 0.5$), and $\Omega = \Omega_0 \mathrm{sech}(\frac{\omega_{\mathrm{qbt}}\tau_p}{2})$. Away from the values $\sigma = \{0, 0.5, 1\}$ the value of $\Delta_{\mathrm{AC}}^{(\mathrm{fc})}$ is relatively constant and we present typical values of $\Delta_{\mathrm{AC}}^{(\mathrm{fc})}/\Omega^2 = \mu_{\mathrm{fc}}$ in Supplementary Table 2 for $\sigma = 0.75$.

Supplementary Note 9

The use of two lasers to perform Raman transitions leads to stark shifts due to off-resonant scattering with the intermediate state. This enters into the Hamiltonian as an effective dephasing term (see Methods)

$$\mathcal{H}_{\mathrm{AC}} = \frac{1}{2} \Delta_{\mathrm{AC}} \sigma_z \ . \tag{108}$$



Supplementary Figure 1: **Spontaneous emission probability for a selection of atomic qubits.** (a) SE probability for hyperfine qubits as a function of detuning, Δ, from the resonant transition. It can be seen that the heavier the element, the lower the minimum SE floor is. (b) SE probability for optical qubits as a function of Rabi frequency. The SE floor is minimised for faster operation times and longer transition lifetimes.

Supplementary Table 2: Typical values of the AC Stark shift proportionality constant, $\mu_{\rm fc}$, for $\sigma = 0.75$. The presented values are within an order of magnitude of those for other values of σ unless a harmonic of $\omega_{\rm rep}$ or $2\omega_{\rm rep}$ is close to $\omega_{\rm qbt}$. The value of σ in typical experiments is kept away from these resonances to allow for frequency tuning with the AOM frequency.

Atom	$\omega_{\rm qbt}/2\pi$ (GHz)	$\mu_{\rm fc}/\Omega_{2\gamma}^2$ (Hz^{-1})
^{9}Be^{+}	1.25	2.7×10^{-9}
^{25}Mg^{+}	2.75	2.6×10^{-9}
^{43}Ca^{+}	3.26	2.9×10^{-9}
^{87}Sr^{+}	5.00	3.3×10^{-9}
^{133}Cs	9.19	4.5×10^{-9}
^{133}Ba^{+}	9.93	4.3×10^{-9}
^{171}Yb^{+}	12.64	4.9×10^{-9}

The expression for $\Delta_{\rm AC}$ depends on what laser source is used. For two CW beams of equal intensity the detuning is given by [34]

$$\Delta_{\rm AC}^{(CW)} = \mu_{\rm cw}\Omega_{2\gamma} \ , \tag{109}$$

where c is a proportionality constant that depends on the detuning from the transition, the fine structure splitting of the upper state, the polarisation of the addressing beams and the optical frequency of the intermediate transition [34]. With the relation $\Omega \propto I$ where I is the intensity in each beam, we find

$$\Delta_{\rm AC}^{(CW)} = \mu_{\rm cw}' I \ , \tag{110}$$

For a laser beam of fluctuating intensity, δI, the corresponding variations in the Stark shift (to first order) are given by

$$\delta\Delta_{\rm AC}^{(CW)} = \mu_{\rm cw}'\delta I = \Delta_{\rm AC}^{(cw)}\frac{\delta I}{I} = \mu_{\rm cw}\Omega_{2\gamma}\frac{\delta I}{I} \ , \tag{111}$$

such that

$$S_{\Delta_{\rm AC}^{(cw)}} = \left(\Delta_{\rm AC}^{(cw)}\right)^2 S_{\rm RIN}(\omega) = \left(\mu_{\rm cw}\Omega_{2\gamma}\right)^2 S_{\rm RIN}(\omega) \ . \tag{112}$$

The Stark shift noise then translates into noise on the Z quadrature as

$$S_z(\omega) = \frac{1}{4}S_{\Delta_{\rm AC}^{(cw)}} = \frac{1}{4}\left(\mu_{\rm cw}\Omega_{2\gamma}\right)^2 S_{\rm RIN}(\omega) \ . \tag{113}$$

To determine when CW Stark shift noise errors are below Rabi noise errors, we set the condition

$$\frac{1}{\pi}\int_0^\infty \frac{d\omega}{\omega^2} S_z(\omega)F_z(\omega) < \frac{1}{\pi}\int_0^\infty \frac{d\omega}{\omega^2} S_\theta(\omega)F_\theta(\omega)\ , \tag{114}$$

where $S_\theta(\omega) = 1/4\Omega_{2\gamma}^2 S_{\mathrm{RIN}}(\omega)$ such that

$$\frac{1}{\pi}\int_0^\infty \frac{d\omega}{\omega^2}\left(\frac{1}{4}\left(\mu_{\mathrm{cw}}\Omega_{2\gamma}\right)^2 S_{\mathrm{RIN}}(\omega)\right) F_z(\omega) < \frac{1}{\pi}\int_0^\infty \frac{d\omega}{\omega^2}\left(\frac{1}{4}\Omega_{2\gamma}^2 S_{\mathrm{RIN}}(\omega)\right) F_\theta(\omega)\ . \tag{115}$$

Without loss of generality, we let $S_{\mathrm{RIN}}(\omega) = h_0$ where h_0 is a constant (white) noise amplitude, and use the piecewise expressions for the filter functions (see Methods) to find

$$\mu_{\mathrm{cw}} < \sqrt{\frac{\pi^2+4}{8}}\ . \tag{116}$$

When using a frequency comb, an additional Stark shift comes from the off-resonant coupling of all the comb lines to the intermediate state. For equal intensities in each beam, the Stark shift takes the form (Equation 107)

$$\Delta_{\mathrm{AC}}^{(fc)} = \mu_{\mathrm{fc}}\Omega_{2\gamma}^2\ , \tag{117}$$

With the relation $\Omega \propto I$ where I is the intensity in each beam, we find

$$\Delta_{\mathrm{AC}}^{(CW)} = \mu_{\mathrm{fc}}' I^2\ , \tag{118}$$

For a laser beam of fluctuating intensity, δI, the corresponding variations in the Stark shift (to first order) are given by

$$\delta\Delta_{\mathrm{AC}}^{(fc)} = 2\mu_{\mathrm{cw}}' I\delta I = 2\Delta_{\mathrm{AC}}^{(fc)}\frac{\delta I}{I} = 2\mu_{\mathrm{fc}}\Omega_{2\gamma}\frac{\delta I}{I}\ , \tag{119}$$

such that

$$S_{\Delta_{\mathrm{AC}}^{(fc)}} = \left(2\Delta_{\mathrm{AC}}^{(fc)}\right)^2 S_{\mathrm{RIN}}(\omega) = 4\left(\mu_{\mathrm{fc}}\Omega_{2\gamma}^2\right)^2 S_{\mathrm{RIN}}(\omega)\ , \tag{120}$$

The Stark shift noise then translates into noise on the Z quadrature as

$$S_z(\omega) = \frac{1}{4}S_{\Delta_{\mathrm{AC}}^{(cw)}} = \left(\mu_{\mathrm{fc}}\Omega_{2\gamma}^2\right)^2 S_{\mathrm{RIN}}(\omega)\ , \tag{121}$$

where typical values of μ_{fc} are given in Section .

Simarily to the CW case, we find the requirement for the frequency comb Stark shift error to be below the Rabi noise error as

$$\frac{1}{\pi}\int_0^\infty \frac{d\omega}{\omega^2}\left(\left(\mu_{\mathrm{cw}}\Omega_{2\gamma}^2\right)^2 S_{\mathrm{RIN}}(\omega)\right) F_z(\omega) < \frac{1}{\pi}\int_0^\infty \frac{d\omega}{\omega^2}\left(\frac{1}{4}\Omega_{2\gamma}^2 S_{\mathrm{RIN}}(\omega)\right) F_\theta(\omega)\ . \tag{122}$$

such that for a white noise spectrum of intensity noise and the piecewise approximation of the filter functions

$$\mu_{\mathrm{fc}} < \sqrt{\frac{\pi^2+4}{32\Omega_{2\gamma}^2}}\ . \tag{123}$$

Supplementary Note 10

Each component of the filter functions is defined by

$$F_{ij}(\omega) = \sum_{k=x,y,z} R_{jk}(\omega)R_{ik}^*(\omega)\ , \tag{124}$$

where $R_{ij}(\omega)$ is the local control matrix in the Fourier frequency domain. $R_{ij}(\omega)$ is evaluated from $R_{ij}(\omega) = -i\omega \int_0^\tau \frac{1}{2}\mathrm{Tr}\left[\hat{U}^\dagger(t)\,\hat{\sigma}_i\hat{U}(t)\,\hat{\sigma}_j\right]\mathrm{e}^{\mathrm{i}\omega t}dt$, where $\hat{U}(t)$ is the unitary operator of the gate operation, $\hat{\sigma}_i$ is the Pauli operator and τ is the gate time.

For a simple unitary operation of the form, $\hat{U}(t) = \exp\left(-i\frac{\Omega}{2}t\hat{\sigma}_\theta\right) = \cos\left(\frac{\Omega}{2}t\right)\hat{I} - i\sin\left(\frac{\Omega}{2}t\right)\hat{\sigma}_\theta$, from [35], the local control matrices are given by

$$\begin{aligned}
R_{xx}(\omega) &= \cos^2(\theta)\left[1-\mathrm{e}^{\mathrm{i}\omega\tau}\right] + \frac{\omega\sin^2(\theta)}{\omega^2-\Omega^2}V(\omega)\\
R_{xy}(\omega) &= \frac{1}{2}\sin(2\theta)\left[1-\mathrm{e}^{\mathrm{i}\omega\tau}\right] - \frac{\omega\sin(2\theta)}{2(\omega^2-\Omega^2)}V(\omega)\\
R_{xz}(\omega) &= -\frac{\omega}{\omega^2-\Omega^2}\sin(\theta)\,B(\omega)\\
R_{yx}(\omega) &= \frac{1}{2}\sin(2\theta)\left[1-\mathrm{e}^{\mathrm{i}\omega\tau}\right] - \frac{\omega\sin(2\theta)}{2(\omega^2-\Omega^2)}V(\omega)\\
R_{yy}(\omega) &= \sin^2(\theta)\left[1-\mathrm{e}^{\mathrm{i}\omega\tau}\right] + \frac{\omega\cos^2(\theta)}{\omega^2-\Omega^2}V(\omega)\\
R_{yz}(\omega) &= \frac{\omega}{\omega^2-\Omega^2}\cos(\theta)\,B(\omega)\\
R_{zx}(\omega) &= \frac{\omega}{\omega^2-\Omega^2}\sin(\theta)\,B(\omega)\\
R_{zy}(\omega) &= -\frac{\omega}{\omega^2-\Omega^2}\cos(\theta)\,B(\omega)\\
R_{zz}(\omega) &= \frac{\omega}{\omega^2-\Omega^2}V(\omega)\ ,
\end{aligned} \tag{125}$$

where Ω is the Rabi frequency, θ is the spin phase of the gate which defines $\hat{\sigma}_\theta = \cos(\theta)\,\hat{\sigma}_x + \sin(\theta)\,\hat{\sigma}_y$, $V(\omega) = i\Omega\mathrm{e}^{\mathrm{i}\omega\tau}\sin(\Omega\tau) - \omega\mathrm{e}^{\mathrm{i}\omega\tau}\cos(\Omega\tau) + \omega$, $B(\omega) = i\Omega\mathrm{e}^{\mathrm{i}\omega\tau}\cos(\Omega\tau) + \omega\mathrm{e}^{\mathrm{i}\omega\tau}\sin(\Omega\tau) - i\Omega$.

To evaluate the error contribution from the cross-correlation of the intensity and frequency noise, we need to evaluate F_{xz} and F_{yz}. Starting with F_{xz}, we have

$$\begin{aligned}
F_{xz} &= R_{xx}R_{zx}^* + R_{xy}R_{zy}^* + R_{xz}R_{zz}^*\\
&= \frac{\omega^2}{\omega^2-\Omega^2}\sin(\phi)\left(VB^* - BV^*\right)\ .
\end{aligned} \tag{126}$$

Similarly, for F_{yz}, we have

$$F_{yz} = -\frac{\omega^2}{\left(\omega^2-\Omega^2\right)^2}\cos(\phi)\left(VB^* - BV^*\right)\ . \tag{127}$$

Now we take a look at the noise field due to laser intensity fluctuations, $\beta_\theta(t)$. For a general θ, $\beta_x(t)$ and $\beta_y(t)$ are non-zero. However, they come from the same source, which is the laser intensity noise. Thus, we can write $\beta_x(t)$ and $\beta_y(t)$ as

$$\begin{aligned}
\beta_x(t) &= \cos(\theta)\,\beta_\theta(t)\\
\beta_y(t) &= \sin(\theta)\,\beta_\theta(t)\ .
\end{aligned} \tag{128}$$

The noise PSD, $S_{ij}(\omega)$, is related to the real time noise profile, $\beta_i(t)$, by

$$\langle\beta_i(t_1)\beta_j(t_2)\rangle = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega S_{ij}(\omega)\mathrm{e}^{\mathrm{i}\omega(t_2-t_1)}\ . \tag{129}$$

Thus, $S_{xz}(\omega)$ and $S_{yz}(\omega)$ can be written as

$$\begin{aligned}
S_{xz}(\omega) &= \cos(\theta)\,S_{\theta z}(\omega)\\
S_{yz}(\omega) &= \sin(\theta)\,S_{\theta z}(\omega)\ ,
\end{aligned} \tag{130}$$

where $\langle \beta_\theta(t_1)\beta_z(t_2)\rangle = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega S_{\theta z}(\omega)e^{i\omega(t_2-t_1)}$. Now, looking at the gate error, we have

$$\begin{aligned}\chi^{(u)}(\tau) &= \sum_j \frac{1}{\pi}\int_0^\infty \frac{d\omega}{\omega^2} S_j(\omega) F_j^{(u)}(\omega) \\ &= \frac{1}{\pi}\int_0^\infty \sum_{m,n=x,y,z} \frac{d\omega}{\omega^2} S_{mn}(\omega) F_{mn}^{(u)}(\omega)\ .\end{aligned} \tag{131}$$

In the summation, $\sum_{m,n=x,y,z} S_{mn}(\omega)F_{mn}(\omega)$, the summation of the cross-correlation terms give

$$\begin{aligned}&S_{xz}(\omega)F_{xz} + S_{yz}(\omega)F_{yz} \\ &= \cos(\theta)\, S_{\theta z}(\omega)F_{xz} + \sin(\theta)\, S_{\theta z}(\omega)F_{yz} \\ &= S_{\theta z}\left(\frac{\omega^2}{\omega^2-\Omega^2}\cos(\theta)\sin(\theta)\,(VB^* - BV^*) - \frac{\omega^2}{\left(\omega^2-\Omega^2\right)^2}\cos(\theta)\sin(\theta)\,(VB^* - BV^*)\right) \\ &= 0\end{aligned} \tag{132}$$

Thus, the contribution of the cross-correlation of the intensity and frequency noise to the gate fidelity is zero, up to the first order in the Magnus expansion used to derive the filter function theory. Alternatively, we can define the x-axis to be the gate phase angle, which means that $\theta = 0$. Then, we only need to consider F_{xz} since $\beta_y(t) = 0$. With $\theta = 0$, $F_{xz} = 0$, and thus cross-correlation of the intensity and frequency noise are irrelevant.

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