

Supplementary materials for acoustic spectral hole-burning in a two-level system ensemble

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I. ADDITIONAL ANALYSIS OF THE SPECTRAL HOLE

In the reference [1] a model for the loss and frequency shift in two-tone spectroscopy is introduced that assumes a uniform (average) coupling strength of TLSs to the resonant modes. For the frequency shift and loss, respectively, they derive

$$\Delta\omega_r = -\frac{\Gamma_0}{2} \frac{(\Delta/\Gamma_2) \tilde{n}}{\sqrt{1+\tilde{n}} \left[(\Delta/\Gamma_2)^2 + (1\sqrt{1+\tilde{n}})^2 \right]}, \quad (\text{S1})$$

$$\frac{\Gamma_{int}}{\Gamma_0} = 1 - \frac{\tilde{n} (1 + \sqrt{1+\tilde{n}})}{\sqrt{1+\tilde{n}} \left[(\Delta/\Gamma_2)^2 + (1\sqrt{1+\tilde{n}})^2 \right]}. \quad (\text{S2})$$

Here, $\tilde{n} = n/n_c$ is the average phonon number scaled with the critical phonon number. The maximal resonator damping due to TLS is set by Γ_0 and Γ_2 represents the intrinsic TLS linewidth. This is related to the effective Rabi frequency via [1]

$$\Omega = \Gamma_2 \sqrt{1+\tilde{n}} \approx \Gamma_2 \sqrt{\tilde{n}}. \quad (\text{S3})$$

Fits of our two-tone spectroscopy data to Eqs. S1-S2 are shown in Supplementary Figs. 1-2. The average and critical phonon numbers are *not* free parameters in this fit, but determined from the single mode loss saturation measurement (See Fig. 2 of the main text). This implies that the scaling of Ω with power will appear as a power dependence of the extracted Γ_2 value when traces are fit independently. Fits for different pump powers do not yield a uniform Γ_2 , but a power dependent behavior shown in Supplementary Fig. 3. This power dependence of Γ_2 is approximately

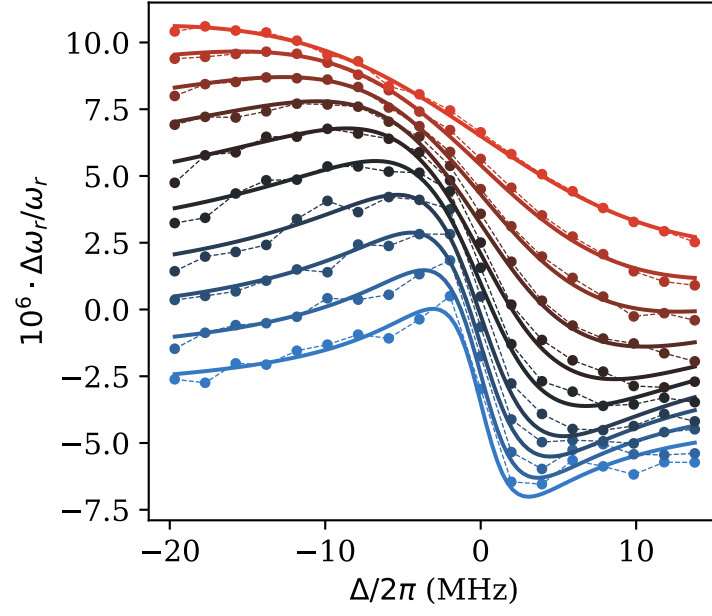
$$\Gamma_2 = \Gamma_{2,0} n^{-0.25}. \quad (\text{S4})$$

Inserting this power dependence we get a scaling of the effective Rabi frequency with pump phonon number as

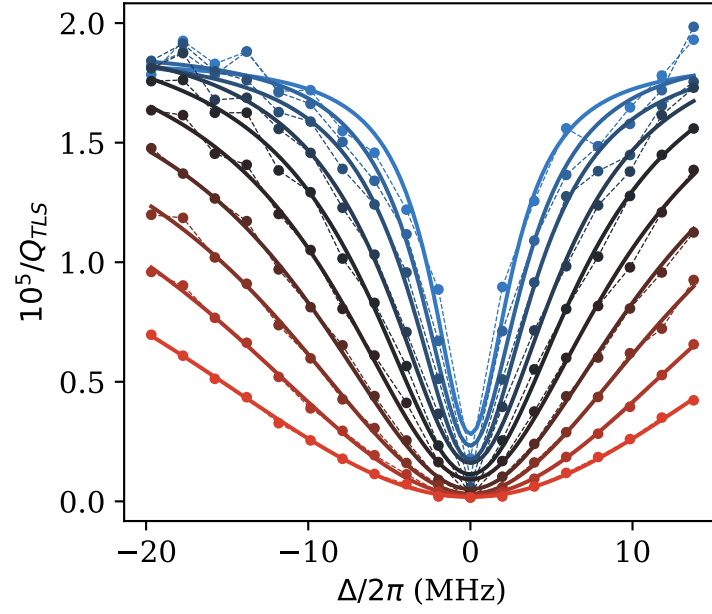
$$\Omega \propto n^{0.25}. \quad (\text{S5})$$

This behavior is similar to what we obtain in the analysis based on expressions derived in [2].

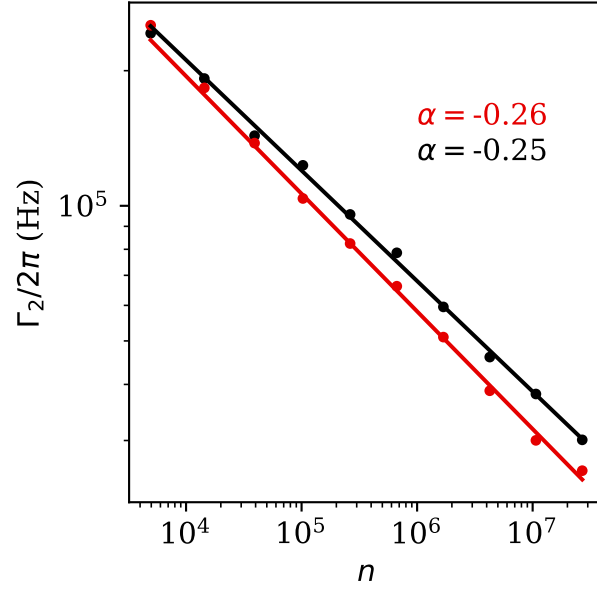
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- [1] Capelle, T. *et al.* Probing a two-level system bath via the frequency shift of an off-resonantly driven cavity. *Phys. Rev. Applied* **13**, 034022 (2020).
 [2] Kirsh, N., Svetitsky, E., Burin, A. L., Schechter, M. & Katz, N. Revealing the nonlinear response of a tunneling two-level system ensemble using coupled modes. *Physical Review Materials* **1**, 012601 (2017).



Supplementary Figure 1. Relative frequency shift measured in two-tone spectroscopy with fits to Eq. S1



Supplementary Figure 2. Loss rate measured in two-tone spectroscopy with fits to Eq. S2



Supplementary Figure 3. Intrinsic TLS linewidth extracted from two-tone spectroscopy measurements using fits to Eqs. S1-S2. The presence of a power dependence indicates a scaling of the effective Rabi frequency with phonon number slower than square-root. Values extracted for the loss (frequency shift) are shown in black (red).