Supplementary Information

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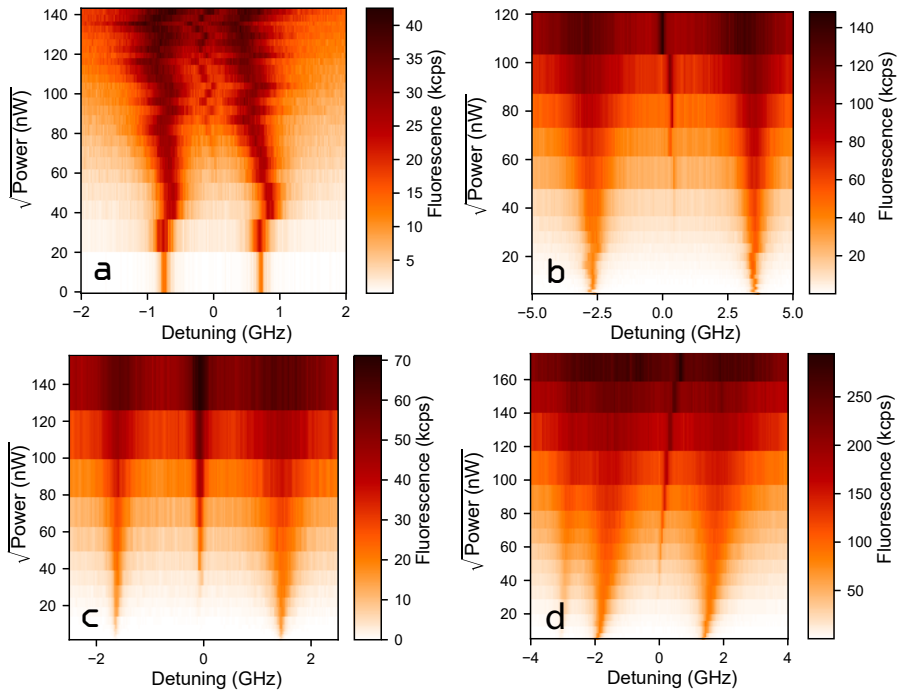

Fig. S1 Raw saturation traces Raw saturation traces for four pairs of coupled molecules. **(c)** corresponds to Fig.3(a-b) and **(d)** corresponds to Fig.3(c-d) from the main text.

When comparing to simulations, the saturation spectra were aligned to the average of the superradiant and subradiant frequencies. The raw saturation spectra are shown in Fig. S1. Note that Fig. S1(a) is spectrally unstable compared to the others. Fig. S1(d) appears to be relaxing slightly towards higher frequencies while being saturated, as it had just been frequency shifted. Resonances are seen to relax on the order of 1 GHz after 10s of GHz of shifting, but quickly stabilize.

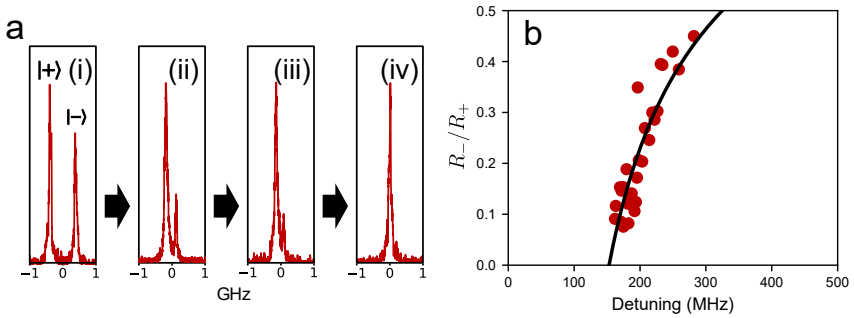

Fig. S2 Two interacting molecules tuning into resonance (a) A pair of coupled molecules being tuned into resonance with pump cycles of 100s of W for 10s of seconds, accompanied by the extinction of the subradiant peak. The molecules are in the J-aggregate orientation, so that the subradiant peak $|-\rangle$ is higher in energy. The molecules spontaneously tuned into resonance from (iii) to (iv) over the course of 10s of minutes without any additional pumping, and stayed resonant for over 35 hours of continuous monitoring. **b** Ratio of the heights of the peaks in **a** as a function of the separation of the detuning of the eigenstates. When the molecules are resonant, the eigenstates are separated by $2\,|J|$. A fit to a master equation simulation gives $J = -76\,\mathrm{MHz}$, $I/I_{\mathrm{sat}} = 0.53$.

Fig. S2 shows a pair of molecules being tuned into resonance, separate from the pair shown in the main text. After tuning onto resonance and measuring the extinction of the subradiant peak, the spectrum was measured continuously for 35 hours without any sign of the resonant molecules becoming detuned.

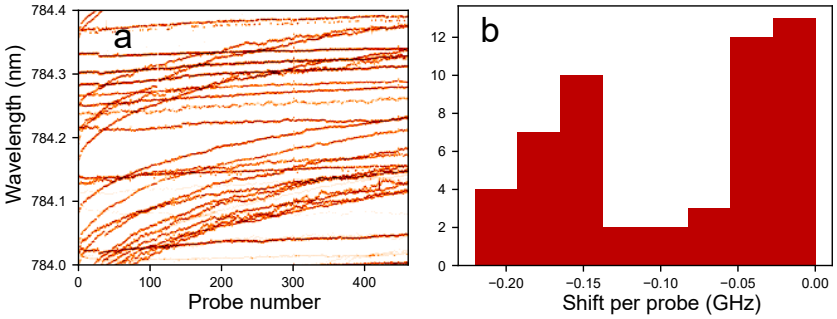

Fig. S3 Frequency shifting dynamics (a) Many molecules shifting as a nanocrystal is pumped with 1 mW of light for 10 seconds between probes. **(b)** A histogram of initial tuning rates is bimodal, with roughly half of the resonances very stable.

Fig. S3 shows many molecules in a 200 GHz range tuning while a nanocrystal undergoes pump and probe cycles. Interestingly, half of the resonances shifted at a much higher rate than the others. The molecules may have been embedded in two separate nanocrystals that were stuck together, so that they could not be spectrally distinguished.

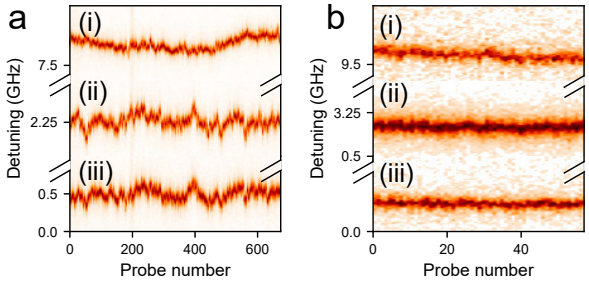

Fig. S4 Spectral wandering of interacting molecules (a) The resonant frequencies of three molecules tracked over the course of 5 hours. Molecules (ii) and (iii) are coupled with $J = -76\,\mathrm{MHz}$. A group of resonances in a separate nanocrystal are monitored for thirty minutes in **(b)**, where resonances (ii) and (iii) are coupled with $J = 1020\,\mathrm{MHz}$. The spectral wandering of these resonances is limited by our measurement uncertainty of 5 MHz. This demonstrates that strongly coupled molecules are capable of extremely high spectral stability.

Because laser-induced charge mobilization in nanocrystals appears to spontaneously decrease inhomogeneous broadening for pairs of nearby molecules, it is expected that the nearby molecules will share a similar electronic environment. This is confirmed in Fig. S4(a), where the spectral stability of two coupled molecules is analyzed over the course of 5 hours. The molecules, which have an interaction strength of $J = 1020\,\mathrm{MHz}$, have clearly correlated spectral wandering.

Because the excitation is shared over two molecules with detuning $\Delta \approx 1.75\,\mathrm{GHz}$ and $J = 76\,\mathrm{MHz}$, the eigenstates are expected to share only 0.2% of their spectral wandering at this detuning. By comparing the spectral wandering of the resonances and the spectral wandering of their frequency difference, we find that roughly 40% is shared. This indicates that nearby molecules that tune onto resonance as charges are mobilized have a generally similar electronic environment, and that the decrease in inhomogeneous broadening is not purely random.

Importantly, the majority of interacting molecules are quite stable. Fig. S4(b) displays a pair of coupled molecules with stability limited by our measurement uncertainty of $\sim 10\,\mathrm{MHz}$. The laser frequency was monitored by a HighFinesse Wavemeter WS6 with a nominal absolute accuracy of 600 MHz, but we simultaneously monitored the resonant frequency of a temperature stabilized cavity (Quantaser FPE001C) to determine our relative frequency uncertainty over the course of the measurement.

1 Synthesis

Anthracene nanocrystals doped with dibenzoterrylene (DBT) molecules were synthesized according to a procedure outlined in Ref. [1, 2]. A $1:10^4$ Mixture of DBT/anthracene was prepared by adding 10 µL of 0.2 mM DBT in toluene to 10 mL of 5 mM anthracene in acetone (DBT synthesized by Symeres, zone-refined anthracene from MilliporeSigma. Toluene was OmniSolv grade and Acetone was HPLC grade, both from MilliporeSigma). 500 µL of the DBT/anthracene solution was added dropwise to 10 mL of milli-Q water and sonicated at 50° C for 30 minutes while the crystals formed. After growth, the solution of nanocrystals was cooled with a ice bath. Solution was drop cast onto Si substrates and dried in a desiccator. The sample was spin-coated with 3% PVA to prevent sublimation of the anthracene. The resulting nanocrystals were between 200 nm and 1 µm in diameter and around 100 nm in thickness. The nanocrystals had tens to hundreds of DBT molecules with resonances distributed between 783 nm and 785 nm.

2 Measurement

We cooled DBT-doped anthracene nanocrystals to 2.7 K in a Montana Cryostation. We located nanocrystals with 200-300 fluorescing DBT molecules and pumped with 10 mW of 785 nm laser light focused to a 1.5 μm-waist spot for 30 minutes each. We tuned a continuous-wave tunable TiSaph laser (MSquared SolsTis-SRX) that was polarization-aligned to the molecules through the inhomogeneous broadening. Increments of 20 GHz were scanned with intensities ranging from I_{sat} to $100I_{sat}$. We filtered out the Stokes-shifted fluorescence with 800-nm long pass filters and counted the photons on an avalanche photodiode (Excelitas SPCM-900-14-FC). Using this method on twenty-five nanocrystals, we found ten clear signatures of interacting pairs of molecules, of which we subjected four pairs to further analysis. Because the resonances were not monitored continuously while pumping, it is possible that some came into resonance and were not identified as interacting because the subradiant state had already extinguished. We performed $g^{(2)}(\tau)$ measurements with resonant light correlated with a time digitizer (FAST ComTec MCS6A). To perform lifetime measurements, we drove molecules into steady-state with resonant light and attenuated with two fiber-integrated Jenoptik electrooptical amplitude modulators (AM906b and AM830) at a rate of 1 MHz.

3 Simulation

We simulated dipole-dipole interactions with numerical solutions of the master equation in Qutip [3]. In the rotating reference frame of the laser, the evolution of the density matrix is

$$\frac{d}{dt}\rho = -i[H,\rho] + \mathcal{L}_{\mathrm{dd}}[\rho] + \mathcal{L}_{\mathrm{deph}}[\rho] + \mathcal{L}_{\mathrm{ind}}[\rho] \tag{1}$$

with Hamiltonian

$$H = \sum_i \Delta_i \sigma_i^\dagger \sigma_i + \sum_{ij} J_{ij} \sigma_i^\dagger \sigma_j + \sum_i [\Omega_R(\mathbf{r_i})\sigma_\mathbf{i} + \text{h.c.}]. \tag{2}$$

Here Δ_i is the detuning of the ith dipole from the laser. The Lindbladians are

$$\mathcal{L}_{\text{dd}}[p] = \sum_{ij} \alpha\Gamma_{ij} \left\{ \sigma_j \rho \sigma_i^\dagger - \frac{1}{2} \left[\sigma_i^\dagger \sigma_j, \rho \right] \right\} \tag{3}$$

$$\mathcal{L}_{\text{deph}}[p] = \sum_i \frac{1}{2\pi T_2} \left\{ (\sigma_i^z)\, \rho\, (\sigma_i^z)^\dagger - \frac{1}{2} \left[(\sigma_i^z)^\dagger (\sigma_i^z), \rho \right] \right\} \tag{4}$$

$$\mathcal{L}_{\text{ind}}[p] = \sum_i (1-\alpha)\Gamma_0 \left\{ \sigma_i \rho \sigma_i^\dagger - \frac{1}{2} \left[\sigma_i^\dagger \sigma_i, \rho \right] \right\} \tag{5}$$

Here, α is the Franck-Condon/Debye-Waller factor and T_2 is the dephasing time.

To write the dipole-dipole Lindbladian $\mathcal{L}_{\text{dd}}[\rho]$ as a sum over one index, we can diagonalize the matrix $\Gamma_{ik} = \sum_j U_{ij}\lambda_j U_{jk}^\star$ to obtain collective operators $\mathcal{O}_i = \sum_j U_{ji}\sigma_j$ with decay rates $\sqrt{\alpha\lambda_j}$.

$$\mathcal{L}[p] = \sum_i \sqrt{\alpha\lambda_j} \left\{ \mathcal{O}_i \rho \mathcal{O}_i^\dagger - \frac{1}{2} \left[\mathcal{O}_i^\dagger \mathcal{O}_i, \rho \right] \right\} \tag{6}$$

References

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