Observation of vacuum-induced collective quantum beats

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Observation of vacuum-induced collective quantum beats

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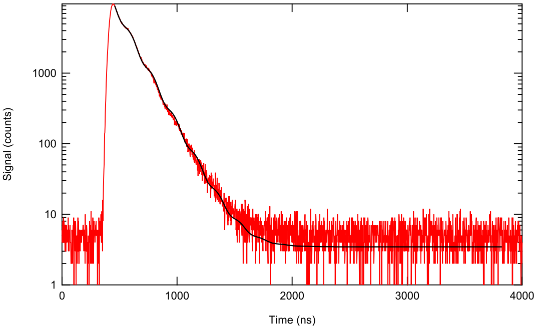
We demonstrate collectively enhanced vacuum-induced quantum beat dynamics from a three-level V-type atomic system. Exciting a dilute atomic gas of magneto-optically trapped ^{85}Rb atoms with a weak drive resonant on one of the transitions, we observe the forward-scattered field after a sudden shut-off of the laser. The subsequent radiative dynamics, measured for various optical depths of the atomic cloud, exhibits superradiant decay rates, as well as collectively enhanced quantum beats. Our work is also the first experimental illustration of quantum beats arising from atoms initially prepared in a single excited level as a result of the vacuum-induced coupling between excited levels.

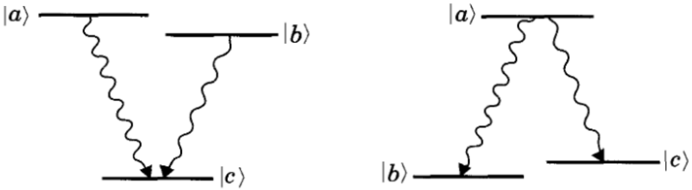
Quantum beats

- Oscillatory behavior in the intensity of radiation emitted by atomic/molecular systems in a superposition of (excited) states
- Ex: quantum beats in the decay $\left|5P_{3/2}\right\rangle \to \left|4S_{1/2}\right\rangle$ in ^{39}K



Simplest case is the three-level system. Two types: V and Λ.

- Semi-classical:



$$|\psi(t)\rangle = \alpha_a e^{-i\omega_a t}|a\rangle + \alpha_b e^{-i\omega_b t}|b\rangle + \alpha_c e^{-i\omega_c t}|c\rangle$$

Each atom contains two oscillating dipoles:

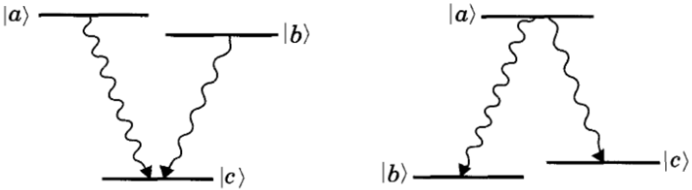
$$e\langle\psi(t)|\, r\,|\psi(t)\rangle = \begin{cases} \mathcal{P}_{ac}(\alpha_a^*\alpha_c)e^{i\nu_1 t} + \mathcal{P}_{bc}(\alpha_b^*\alpha_c)e^{i\nu_2 t} + c.c. & \text{if } V \\ \mathcal{P}_{ab}(\alpha_a^*\alpha_b)e^{i\nu_1 t} + \mathcal{P}_{ac}(\alpha_a^*\alpha_c)e^{i\nu_2 t} + c.c. & \text{if } \Lambda \end{cases}$$

Radiated field:

$$E^{(+)} = \mathcal{E}_1 e^{-i\nu_1 t} + \mathcal{E}_2 e^{-i\nu_2 t} \implies \left|E^{(+)}\right|^2 = \cdots + [\mathcal{E}_1^*\mathcal{E}_2 e^{i(\nu_1-\nu_2)t} + c.c.]$$

Simplest case is the three-level system. Two types: V and Λ.

- QED:



$$|\psi_V(t)\rangle = \sum_{i=a,b,c} \alpha_i |i,0\rangle + \alpha_1 |c, 1_{\nu_1}\rangle + \alpha_2 |c, 1_{\nu_2}\rangle$$

$$|\psi_\Lambda(t)\rangle = \sum_{i=a,b,c} \alpha'_i |i,0\rangle + \alpha'_1 |b, 1_{\nu_1}\rangle + \alpha'_2 |c, 1_{\nu_2}\rangle$$

With

$$E_j^{(-)}(t) \sim a_j^\dagger e^{i\nu_j t} \quad \text{and} \quad E_j^{(+)}(t) \sim a_j e^{-i\nu_j t}$$

Beat note term:

$$\langle\psi_V(t)|\, E_1^{(-)}(t)E_2^{(+)}(t)\,|\psi_V(t)\rangle \sim \langle 1_{\nu_1}0_{\nu_2}|\, a_1^\dagger a_2\, |0_{\nu_1}1_{\nu_2}\rangle\, e^{i(\nu_1-\nu_2)t}\, \underbrace{\langle c|c\rangle}_{1}$$

$$\langle\psi_\Lambda(t)|\, E_1^{(-)}(t)E_2^{(+)}(t)\,|\psi_\Lambda(t)\rangle \sim \langle 1_{\nu_1}0_{\nu_2}|\, a_1^\dagger a_2\, |0_{\nu_1}1_{\nu_2}\rangle\, e^{i(\nu_1-\nu_2)t}\, \underbrace{\langle c|b\rangle}_{0}$$

- Quantum beats can exist in V-type systems, but not in Λ-type
- This is not explained by semi-classical theory.

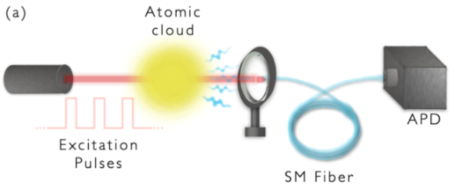
Textbooks: Quantum beats require a coherent superposition initially

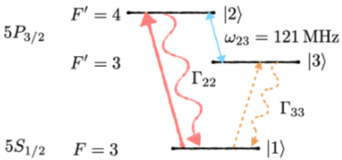
"Observation of..." by Han et al.

Experimentally demonstrates 2 new aspects:

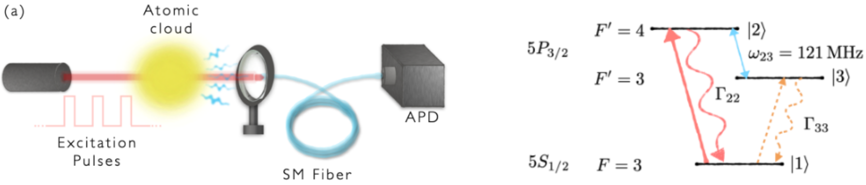
- **Vacuum-induced quantum beats:** observed quantum beats without an initial superposition of excited levels
- **Collective effect:** enhanced beat amplitudes and decay rates

Experiment





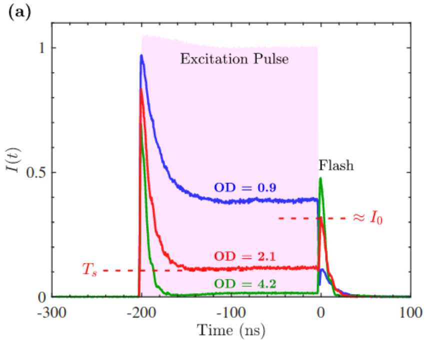
- $\sim 10^8$ ^{85}Rb atoms in a MOT, $\rho\lambda^3 \ll 1$
- V-type system, well-separated. $\Gamma_{22} = 2\pi \cdot 6.1$ MHz, $\Gamma_{33} = 5/9\Gamma_{22}$
- $\Gamma_{23} \approx \sqrt{\Gamma_{22}\Gamma_{33}}$: 2nd order coupling between $|2\rangle, |3\rangle$
- $v = 120$ nm/μs $\implies$ negligible to 780 nm over $1/\Gamma_{22} \approx 26$ ns



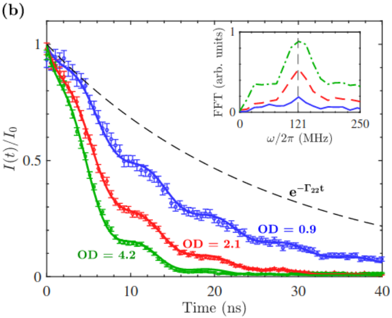
- Lin. pol. 780 nm light (D_2) drives $|1\rangle \to |2\rangle$. 200 ns on, 800 ns off.
- $>$ 30 dB extinction ratio, 3.5 ns fall time (2 EOMs in series)
- $I_x \ll I_s = 3.9$ mW/cm^2 $\implies$ single-excitation in $|2\rangle$
- Forward-scattered photons collected by SM fiber into APD
- Histograms for ∼30 mins, new shot every 2 ms $\implies$ 2×10^8 shots

Data

Raw histograms, taken at various $OD = -\ln T_s$



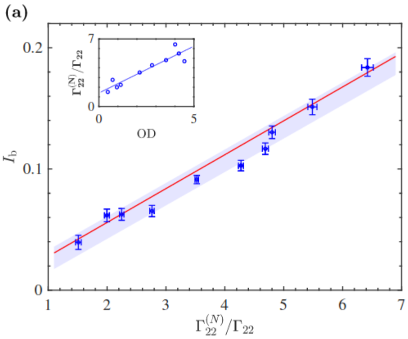
- ^{85}Rb cloud is initially transparent
- Transmission decays to $T_s = e^{-OD}$, from destructive interference
- "Flash" after driving field is switched off, amplitude $\propto OD$



$$I(t)/I_0 = e^{-\Gamma_{22}^{(N)}t} + I_b e^{-\Gamma_{\text{avg}}^{(N)}t}\sin(\omega_{23}t+\phi), \qquad \begin{cases} \Gamma_{ij}^{(N)} = (1+Nf)\Gamma_{ij} \\ \Gamma_{\text{avg}}^{(N)} = (\Gamma_{22}^{(N)}+\Gamma_{33}^{(N)})/2 \end{cases}$$

- From FFT, $f_{\text{osc}} \approx 121$ MHz $= \omega_{23}/2\pi$, as expected
- Enhanced decay rates $\implies$ collective effect
- Fit parameters $I_b, \Gamma_{22}^{(N)}, \phi$ quantify collective effects

Collective effects:



- Enhanced decay rates: $\Gamma_{22}^{(N)}/\Gamma_{22} = 1.0(1) \cdot OD + 1.4(4)$
- Enhanced beat amplitude, in good agreement with

$$I_b = \frac{\Gamma_{33}^{(N)}}{\omega_{23}} \approx \frac{5}{9}\frac{\Gamma_{22}^{(N)}}{\omega_{23}}$$

Model

$$\mathcal{H}_{\mathrm{A}} = \sum_{m=1}^{N} \sum_{j=2,3} \hbar\omega_{j1}\, \hat{\sigma}_{m,j}^{+}\hat{\sigma}_{m,j}^{-}$$

$$\mathcal{H}_{\mathrm{F}} = \sum_{k} \hbar\omega_{k}\, \hat{a}_{k}^{\dagger}\hat{a}_{k}$$

$$\mathcal{H}_{\mathrm{AD}} = -\sum_{m=1}^{N} \sum_{j=2,3} \hbar\Omega_{j}^{m} \left(\hat{\sigma}_{m,j}^{+} e^{-i\omega_{D} t} + \hat{\sigma}_{m,j}^{-} e^{i\omega_{D} t}\right)$$

$$\mathcal{H}_{\mathrm{AF}} = -\sum_{m=1}^{N} \sum_{j=2,3} \sum_{k} \hbar g_{m,j}(\omega_{k}) \left(\hat{\sigma}_{m,j}^{+}\hat{a}_{k} + \hat{\sigma}_{m,j}^{-}\hat{a}_{k}^{\dagger}\right) \quad \text{(RWA)}$$

Model: driven dynamics

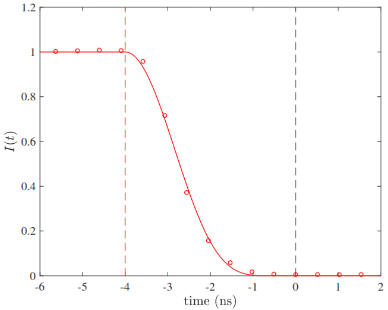
Using experimental parameters to numerically solve for ρ_A

- $\rho_{33,s} \approx 0$
- $\rho_{22,s} \approx 10^{-10}$
- $\rho_{11,s} \approx 1$
- Only non-zero coherence: $|\rho_{12}|^2 \approx 10^{-5}$

$\implies$ Well within the single-excitation regime

Model: driven dynamics

Broad spectral component of turn-off may excite to $|2\rangle$, $|3\rangle$



$$\rho_{33} = \rho_{22} = \rho_{23} = 0, \rho_{11} = 1, \rho_{12} = 10^{-5}, \rho_{13} \approx 10^{-6}$$

$\Longrightarrow$ Laser turn-off produces no significant excitation

$\Longrightarrow$ Wait 0.5 ns after turn-off to avoid residual drive & transient effects.

Go to interaction representation w.r.t $\mathcal{H}_A + \mathcal{H}_F$

$$\widetilde{\mathcal{H}}_{AF} = -\sum_{m=1}^{N} \sum_{j=2,3} \sum_{k} \hbar g_j(\omega_k) \left(\hat{\sigma}^{+}_{m,j} \hat{a}_k e^{i(\omega_{j1}-\omega_k)t} + \hat{\sigma}^{-}_{m,j} \hat{a}^{\dagger}_k e^{-i(\omega_{j1}-\omega_k)t} \right)$$

Initially there is one excitation in $|2\rangle$ symmetrically:

$$|\psi(0)\rangle = \frac{1}{\sqrt{N}} \sum_{m=1}^{N} \hat{\sigma}^{+}_{m,2} |11\ldots1\rangle |\{0\}\rangle$$

$$\implies |\psi(t)\rangle = \left(\sum_{m=1}^{N} \sum_{j=2,3} c_{m,j}(t) \hat{\sigma}^{+}_{m,j} + \sum_{k} c_{\omega_k}(t) \hat{a}^{\dagger}_k \right) |11\ldots1\rangle |\{0\}\rangle$$

Same IC, so $c_{m,j}(t) = c_j(t) \implies$ solve for $c_2(t), c_3(t)$

Model: relaxation/quantum beat dynamics

$$c_2(t) = \frac{1}{\sqrt{N}} \left[e^{-\Gamma_{22}^{(N)} t/2} - \left(\frac{\Gamma_{23}^{(N)}}{2\omega_{23}} \right)^2 \frac{\delta^*}{\delta} e^{-\Gamma_{33}^{(N)} t/2} e^{i\omega_{23} t} \right]$$

$$c_3(t) = -\frac{i\Gamma_{32}^{(N)}}{2\sqrt{N}\delta} \left[e^{-\Gamma_{22}^{(N)} t/2} e^{-i\omega_{23} t} - e^{-\Gamma_{33}^{(N)} t/2} \right]$$

- Decay rate is N times that of an individual atom (forward)
- Superradiance w.r.t both $|2\rangle$ (prep.) and $|3\rangle$ (vacuum-induced)
- Contribution of $|3\rangle$ creates quantum beats with frequency ω_{23}

Model: field intensity

Light intensity:

$$I(x,t) = \frac{\epsilon_0 c}{2} \langle \psi(t)| \hat{E}^{+}(x,t)\hat{E}(x,t) |\psi(t)\rangle$$

E-field operator:

$$\hat{E}(x,t) = \int_{\mathbb{R}} dk\, E_k \hat{a}_k e^{ikx} e^{-i\omega_k t}$$

Plugging in $|\psi(t)\rangle$ and eliminating x...

$$\frac{I(t)}{I_0} = e^{-\Gamma_{22}^{(N)} t} + \underbrace{\left(\Gamma_{33}^{(N)}/2\omega_{23}\right)^2}_{\ll 1} e^{-\Gamma_{33}^{(N)} t} + \underbrace{\left(\Gamma_{33}^{(N)}/\omega_{23}\right)}_{I_b} e^{-\Gamma_{\text{avg}}^{(N)} t} \sin(\omega_{23} t + \phi)$$

$$= e^{-(1+Nf)\Gamma_{22} t} + I_b e^{-\Gamma_{\text{avg}}^{(N)} t} \sin(\omega_{23} t + \phi)$$

Conclusion

Summary:

- Demonstrated collective quantum beats in spontaneous emission process without initial superposition of excited states
- Observed
  - enhanced decay rates
  - enhanced quantum beat amplitudes

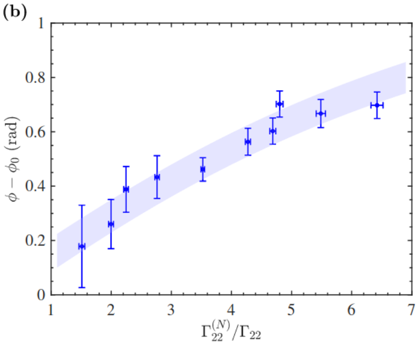
  Good agreement with theoretical model and previous studies.

Applications:

- Tool in precision spectroscopy: enhancing signals
- Combined with waveguide optics to study interactions between distant atomic ensembles (e.g. ONF experiment at JQI)

Extras: The phase term

Expect: $\phi = \arctan\left(\Gamma_{22}^{(N)}/\omega_{23}\right)$



Fit to $\phi = \arctan\left(\eta \cdot \Gamma_{22}^{(N)}/\Gamma_{22}\right) + \phi_0$. Found $\eta \approx 3 \times \Gamma_{22}/\omega_{23}$ and $\phi_0 \neq 0$.
$\Longrightarrow$ Possibly due to transience and non-equilibrium dynamics during switch off, causing OD-dependent phase delay not captured by the model.

References

- Scully, Marlan O., and M. Suhail Zubairy. "Quantum optics." (1999): 648-648.
- Han, Hyok Sang, et al. "Observation of vacuum-induced collective quantum beats." arXiv preprint arXiv:2102.11982 (2021).