Wigner Crystallization of Single Photons in Cold Rydberg Ensemble – Supplemental Material

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EFFECTIVE HAMILTONIAN

In this supplementary we derive the effective Hamiltonian (1) of the main text in one spatial dimension and explicitly establish its conditions of validity. The treatment is based on the formalism developed in [1]. The interaction of probe and control fields with the three-level atoms shown in Fig. 1 of the main text in the absence of Rydberg interactions can be described by the following atom-light coupling Hamiltonian in a rotating frame

$$\hat{H} = \int \mathrm{d}^3\mathbf{r}\Big\{ \Delta\hat{\Sigma}_{ee}(\mathbf{r}) + \delta\hat{\Sigma}_{rr}(\mathbf{r}) + \Omega\hat{\Sigma}_{re}\mathrm{e}^{i\mathbf{q}_c\mathbf{r}} + g\sqrt{n}\hat{\mathcal{E}}(\mathbf{r})\hat{\Sigma}_{eg}(\mathbf{r})\mathrm{e}^{i\mathbf{q}_p\mathbf{r}} + \mathrm{H.a.}\Big\}, \tag{1}$$

where all quantities are defined as in the main text and $\hat{\Sigma}_{\mu\nu}(\mathbf{r}) \equiv \sum_{j\in\Delta V} |\mu\rangle_{jj}\langle\nu|/\sqrt{\Delta N}$ are continuous atomic flip operators defined on a small volume $\Delta V(\mathbf{r})$ centered around position $\mathbf{r}$ containing $\Delta N \gg 1$ atoms. Assuming that all atoms are initially prepared in the ground state $|g\rangle$ and considering weak probe fields, i.e. a photon density much less than the atom density, we can treat the light-atom coupling perturbatively. Consequently, in lowest order of the atom-field coupling g we find the Heisenberg-Langevin equations for the atomic operators

$$\frac{\partial}{\partial t}\hat{\Sigma}_{ge} = -(i\Delta+\gamma)\hat{\Sigma}_{ge} + \hat{F}_{ge} + ig\sqrt{n}\,\hat{\mathcal{E}}\mathrm{e}^{i\mathbf{q}_p\mathbf{r}} + i\Omega^*\,\hat{\Sigma}_{gr}\mathrm{e}^{-i\mathbf{q}_c\mathbf{r}}, \tag{2}$$

$$\frac{\partial}{\partial t}\hat{\Sigma}_{gr} = -i\delta\hat{\Sigma}_{gr} + i\Omega\,\hat{\Sigma}_{ge}\mathrm{e}^{i\mathbf{q}_c\mathbf{r}}. \tag{3}$$

Here the $\hat{F}_{ge}$ is a delta-correlated Langevin noise operator associated with the decay from the intermediate (excited) state $|\mathrm{e}\rangle$ which is necessary to preserve commutation relations [2]. One easily verifies that the correlation functions of the Langevin operators are proportional to the population in the excited state $|e\rangle$. In the linear response and for sufficiently small two-photon detuning δ this population is small and we can safely ignore the noise operators in the following. If need be these operators can be re-introduced by hand using the fluctuation-dissipation theorem. To arrive at a closed description of the atom-field system we also need the equation of motion for the slowly varying probe-field envelope $\hat{\mathcal{E}}(\mathbf{r},t)$. Restricting ourselves to a one-dimensional problem the dynamics of the probe field is described by a truncated wave-equation in paraxial approximation

$$\left[\frac{\partial}{\partial t} + c\frac{\partial}{\partial z}\right]\hat{\mathcal{E}}(z,t) = ig\sqrt{n}\,\hat{\Sigma}_{ge}(z,t)\mathrm{e}^{i\mathbf{q}_p\mathbf{r}}. \tag{4}$$

Transforming Eqs. (2-4) into Fourier space according to $f(z,t) = \int \mathrm{d}k\, e^{-ikz} f(k,t)$ yields the following matrix equations

$$\frac{d}{dt}\mathbf{X} = -i\mathsf{H}\mathbf{X} \tag{5}$$

where $\mathbf{X}^\top = \{\hat{\mathcal{E}}, \hat{\Sigma}_{gr}\mathrm{e}^{i(\mathbf{q}_p+\mathbf{q}_c)\mathbf{r}}, \hat{\Sigma}_{ge}\mathrm{e}^{i\mathbf{q}_p\mathbf{r}}\}$ and the Hamiltonian matrix reads

$$\mathsf{H} = \begin{bmatrix} -kc & 0 & -g\sqrt{n} \\ 0 & \delta & -\Omega \\ -g\sqrt{n} & -\Omega^* & \Delta - i\gamma \end{bmatrix}.$$

Changing the basis to a description in terms of dark- and bright-polaritons $\mathbf{Y}^\top = \{\hat{\Psi}, \hat{\Phi}, \hat{\Sigma}_{ge}\mathrm{e}^{i\mathbf{q}_p\mathbf{r}}\}$ via $\hat{\Psi} = \cos\theta\hat{\mathcal{E}} - \sin\theta\hat{\Sigma}_{gr}\mathrm{e}^{i(\mathbf{q}_p+\mathbf{q}_c)\mathbf{r}}$ and $\hat{\Phi} = \sin\theta\mathcal{E} + \cos\theta\hat{\Sigma}_{gr}\mathrm{e}^{i(\mathbf{q}_p+\mathbf{q}_c)\mathbf{r}}$ yields the equation of motion $\partial_t\mathbf{Y} = -i\mathsf{H}'\mathbf{Y}$ with

$$\mathsf{H}' = \begin{bmatrix} \delta\sin^2\theta - kc\cos^2\theta & -\sin\theta\cos\theta(\delta+kc) & 0 \\ -\sin\theta\cos\theta(\delta+kc) & \delta\cos^2\theta - kc\sin^2\theta & -\Omega_e \\ 0 & -\Omega_e & \Delta - i\gamma \end{bmatrix}.$$

Assuming that the time evolution is slow compared to $|\Delta - i\gamma|$ we can adiabatically eliminate the optical polarization $\hat{\Sigma}_{ge}$ which yields the coupled equations for bright and dark polaritons

$$\frac{d}{dt}\begin{bmatrix}\hat{\Psi}\\ \hat{\Phi}\end{bmatrix} = -i\mathsf{H}''\begin{bmatrix}\hat{\Psi}\\ \hat{\Phi}\end{bmatrix} \tag{6}$$

with

$$\mathsf{H}'' = \begin{bmatrix} \delta\sin^2\theta - kc\cos^2\theta & -\sin\theta\cos\theta(\delta+kc) \\ -\sin\theta\cos\theta(\delta+kc) & \delta\cos^2\theta - kc\sin^2\theta - \frac{\Omega_e^2}{\Delta-i\gamma} \end{bmatrix}.$$

For $\Delta > 0$ and under slow-light conditions, i.e. $\sin^2\theta \gg \cos^2\theta$, one recognizes that the off-diagonal coupling terms are small compared to the difference of the diagonal elements. Under these conditions the dynamics of dark and bright polaritons approximately separates and one can treat their cross coupling perturbatively. Within this perturbative treatment the

effective equation of motion of the dark polariton $\hat{\Psi}$ up to second order of the off-diagonal coupling is given by

$$\frac{d}{dt}\hat{\Psi} = -i\left(\delta\sin^2\theta - kc\cos^2\theta\right)\hat{\Psi} \\ -i\frac{\sin^2\theta\cos^2\theta(\delta+kc)^2}{(\delta+kc)(\sin^2\theta-\cos^2\theta)+\frac{\Omega_e^2}{\Delta-i\gamma}}\hat{\Psi}.$$

To arrive at an even more simplified but more transparent equation we assume $\delta \geq 0$ and require $\delta + kc \ll \Omega_e^2/|\Delta|$ for all relevant values of k, which implies in particular

$$0 \leq \delta \ll \frac{\Omega_e^2}{|\Delta|}, \tag{7}$$

where we used $|\Delta| \gg \gamma$. In this limit we find that the dynamics of the DSPs is described by

$$\frac{d}{dt}\hat{\Psi} = -i\delta\left(1+\frac{\delta\Delta\cos^2\theta}{\Omega_e^2}\right)\hat{\Psi} + ikv_{\rm g}\left(1-2\frac{\delta\Delta}{\Omega_e^2}\right)\hat{\Psi} \\ -i\frac{v_{\rm g}c\Delta}{\Omega_e^2}k^2\hat{\Psi}, \tag{8}$$

where we approximated $\sin^2\theta \approx 1$. The first term on the right hand side describes an energy offset due to a finite two-photon detuning. The second term accounts for the propagation with group velocity $v_{\rm g} = c\cos^2\theta$. The third term describes the quadratic dispersion with effective mass $m_\parallel^{-1} = c^2\cos^2\theta|\Delta|/\Omega_e^2 \approx v_{\rm g}L_{\rm abs}|\Delta|/\gamma$.

Condition (7) also determines the validity of the interaction Hamiltonian (3) of the main text. As already pointed out in the main text, the Rydberg interactions effectively induce a space-dependent two-photon detuning [3] which combined with eq. (7) leads to the critical minimal distance $a_{\rm c} = (C_\alpha\gamma/\Omega_e^2)^{1/\alpha}$.

We can also interpret the validity of the perturbation theory as a condition of a maximal k-value until which a separation into DSPs with slow-light dispersion and fast moving BSPs is valid. This condition reads

$$|kc| \ll \frac{\Omega_e^2}{|\Delta|} \tag{9}$$

leading to $|k_{\rm max}| = \Omega_e^2/|\Delta|c$. Plugging this into the zeroth order dispersion relation of the DSP leads to the maximal energy $\omega_{\rm max} = v_{\rm g}k_{\rm max} = \Omega^2/|\Delta|$, which in the end determines the maximal temperature of the DSP gas.

Finally estimating the typical k-value of the system via the inverse characteristic length scale, i.e. $k \sim 1/L_{\rm char}$ we can rewrite condition (9) and obtain

$$\frac{L_{\rm abs}}{L_{\rm char}} \leq \frac{\gamma}{|\Delta|}, \tag{10}$$

which is just condition (2) of the main text with the appproximation $\sin\theta \approx 1$.

TIME-DEPENDENT LUTTINGER LIQUID THEORY

In this section we provide some details of the light storage protocol introduced in the main text and, using time dependent Luttinger liquid (LL) theory, derive an expression for the density-density correlation function.

Decomposition of the LL Hamiltonian (4) from the main text into bosonic momentum modes $\hat{b}_p, \hat{b}_p^\dagger$ [4] leads to

$$\hat{H} = \frac{u(t)}{2}\sum_{p\neq 0}|p|\left[w(t)\hat{b}_p^\dagger\hat{b}_p - \frac{g(t)}{2}\left(\hat{b}_p^\dagger\hat{b}_{-p}^\dagger + \hat{b}_{-p}\hat{b}p\right)\right], \tag{11}$$

where $u(t) = \pi\rho_0/m(t)K(t)$ is the speed of sound and $g(t),\ w(t)$ are given by the LL parameter $K(t)$ as $w(t) = K(t)+1/K(t),\ g(t) = K(t)-1/K(t)$. The time-dependence of the bosonic operators is given by Heisenberg equations of motion following from eq.(11). To solve these we perform a Bogoliubov transformation relating the time-dependent operators $\hat{b}_p^\dagger(t),\ \hat{b}_p(t)$ to time-independent ones thereby mapping the time-dependence to the coefficients [5]

$$\hat{b}_p(t) = u_p(t)\hat{b}_p(0) + v_p^*(t)\hat{b}_p^\dagger(0). \tag{12}$$

This yields coupled differential equations for the coefficients

$$i\partial_t\begin{pmatrix}u_p(t)\\v_p(t)\end{pmatrix} = \frac{\pi\rho_0}{2m(t)}|p|\begin{pmatrix}w(t) & -g(t)\\ g(t) & -w(t)\end{pmatrix}\begin{pmatrix}u_p(t)\\v_p(t)\end{pmatrix}. \tag{13}$$

To solve these equations we can diagonalize the coupling matrix on the right hand side. The corresponding transformation matrix is itself time-dependent thus leading leading to an off-diagonal coupling $\sim\dot{K}(t)/K(t)$ in the transformed equations which cannot be neglected. However, if we perform a subsequent diagonalization we get an off-diagonal coupling proportional to

$$\frac{\rm d}{{\rm d}t}\left(\frac{\dot{K}(t)}{u(t)K(t)}\right). \tag{14}$$

Since the time-dependence of both, $K(t)$ and $u(t)$, is given by the Polariton mass $m(t)$, we can choose the time-dependence such that the off-diagonal coupling vanishes. For the choice

$$m(t) = m_0\frac{e^{2x(t)}-1}{e^{2x(0)}-1}, \quad x(t) = {\rm acosh}(t/\tau + C), \tag{15}$$

with $m_0 = m(0)$ and $2C = K(0) + 1/K(0)$ expression (14) vanishes. Inverting the transformations gives an analytic solution for the coefficients $u_p(t),\ v_p^*(t)$. Using (12) and the fact that the time-independent Hamiltonian for $t \leq 0$ can be diagonalized lets us now compute arbitrary correlation functions. In particular we calculate

$$\langle[\phi(z)-\phi(0)]^2\rangle = \int_0^\infty {\rm d}p\, e^{-\alpha p}\frac{1-\cos pz}{p} \\ \times\langle(\hat{b}_p^\dagger(t)+\hat{b}_{-p}(t))(\hat{b}_{-p}^\dagger(t)+\hat{b}_p(t))\rangle \\ = K(t)\big[\ln(z/\alpha) + I(z,t)\big]. \tag{16}$$

Here α is a high momentum cutoff introduced to treat divergences which we choose as the smallest length scale $\sim 1/\rho_0$ in our system.

$$I(z,t) = \int_0^\infty \mathrm{d}p \frac{1-\cos pz}{p} \frac{\cos\xi(t) - 1 - \sqrt{l_0^2p^2-1}\sin\xi(t)}{1-l_0^2p^2},$$

where $\xi(t) = \sqrt{l_0^2p^2-1}\ln(K(t)/K(0))$ and can only be evaluated numerically. The expression (16) allows us to write the oscillatory part of the density-density correlations as follows

$$\langle\rho(z,t)\rho(0,t)\rangle_{\mathrm{osc}} \sim \cos(2\pi\rho_0 z)\left(\frac{1}{\rho_0 z}\right)^{2K(t)} e^{-K(t)I(z,t)}. \tag{17}$$

We see that the algebraic decay with exponent $\sim K(t)$ corresponding to the adiabatic quench gets modified by the exponential $I(z,t)$ which leads to a crossover at length scales $z > l_0$.

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