Quantum and nonlinear effects in light transmitted through planar atomic arrays

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Understanding strong cooperative optical responses in dense and cold atomic ensembles is vital for fundamental science and emerging quantum technologies. Methodologies for characterizing light-induced quantum effects in such systems, however, are still lacking. Here we unambiguously identify significant quantum many-body effects, robust to position fluctuations and strong dipole-dipole interactions, in light scattered from planar atomic ensembles by comparing full quantum simulations with a semiclassical model neglecting quantum fluctuations. We find pronounced quantum effects at high atomic densities, light close to saturation intensity, and around subradiant resonances. Such conditions also maximize spin-spin correlations and entanglement between atoms, revealing the microscopic origin of light-induced quantum effects. In several regimes of interest, our approximate model reproduces light transmission remarkably well, permitting analysis of otherwise numerically inaccessible large ensembles, in which we observe many-body analogues of resonance power broadening, vacuum Rabi splitting, and significant suppression in cooperative reflection from atomic arrays.
light can mediate strong interactions between atoms, inducing strong position-dependent correlations, even in the limit of low light intensity, when the response (for the case of a simple level structure) is entirely classical. Such a correlated optical response can differ dramatically from that predicted by standard electrodynamics of continuous media, where resonant-light-induced dipole–dipole (DD) interactions between atoms are treated in an averaged sense. Beyond the limit of low light intensity, an isolated atom can scatter light quantum-mechanically (such as in the incoherent Mollow spectrum)\(^4\), and quantum effects in the interactions of light with dilute atomic ensembles have been utilized in, e.g., quantum information protocols\(^4\). In strongly interacting dense systems the possible role of ensembles have been utilized in, e.g., quantum information processing\(^5\–\(^8\). A particularly promising system to explore and utilize strong light-induced DD interactions is a regular planar array of scatterers, such as atoms. As shown both theoretically and experimentally, in the linear low-excitation limit these manifest a wealth of phenomena, e.g., subdiffraction features\(^9\–\(^10\), nontrivial topological phases\(^11\–\(^12\), transmission varying from complete reflection to full transparency\(^13\–\(^19\), narrow resonances, and subradiance\(^15\–\(^17\),\(^20\–\(^27\), as well as quantum technological applications\(^28\–\(^29\), and other collective effects\(^30\–\(^35\).

We show that we can identify quantum effects in the light transmitted through planar arrays and uniform-density disks of cold and dense atomic ensembles. Many-body quantum correlations are induced by light-atom coupling, which, surprisingly, survive even strong many-body resonant DD interactions and atomic position fluctuations. Specifically, comparing the correlated optical response determined using the quantum master equation (QME) to simulations neglecting any quantum fluctuations between atomic levels in different atoms [referred to as the "semiclassical" equations (SCEs)\(^36\)], we systematically identify light-established quantum effects between atoms in the transmitted light as a function of atom confinement, density, and driving intensity. The effect of many-body quantum fluctuations on the scattering manifests most prominently at high densities when the light is close to saturation intensity, and especially significantly in the vicinity of subradiant resonances. We find that these conditions also produce maximal spin–spin correlations and entanglement of formation in the underlying atomic system, further confirming the role of many-body quantum correlations and entanglement in observing a difference in light transmission between QME and SCEs models. Incorporating the single-atom quantum description of light emission into the semiclassical scattering, we can typically use SCEs also for incoherent scattering to qualitatively reproduce the full quantum scattering even in the regimes where quantum effects in coherent scattering are most pronounced, and elsewhere also quantitatively. SCEs therefore allow us to analyze cooperative transmission of light through large atomic arrays and disks beyond the limit of low light intensity, without needing to solve the full strongly-interacting quantum dynamics. Doing so, we find collective phenomena due to DD interactions that are many-body analogs of power broadening and vacuum Rabi splitting of atomic resonances in cavities\(^37\–\(^38\), and demonstrate a significant effect of intensity on the transmission that may ultimately restrict the utilization of atomic arrays as highly reflective cooperative mirrors.

**Results**

An appealing feature of light scattering from cold atoms\(^39\–\(^50\) is that light-mediated strong DD interactions can establish correlations between atoms at fluctuating positions, which are most simply described using atomic field operators for the ground and excited states \(\psi_{g,e}(r)\). Hence, \(\langle P_\theta(r) \rangle = \langle \psi_{g,e}^\dagger(r) d_{g,e} \psi_{g,e}(r) \rangle\) denotes the positive frequency component of the light-induced atomic polarization, where \(d_{g,e} = d_{g,e}^\dagger\) is the dipole matrix element, and the populations are \(\langle \psi_{g,e}^\dagger(r) \psi_{g,e}(r) \rangle\) and \(\langle \psi_{g,e}(r) \psi_{g,e}^\dagger(r) \rangle\). Because of the DD interactions, the polarization and populations also depend on two-body correlations \(\langle \psi_{g,e}^\dagger(r_1) \psi_{g,e}(r_2) \psi_{g,e}(r_3) \psi_{g,e}^\dagger(r_4) \rangle\), \(a, b, c, d \in \{g,e\}\), representing the correlations in the optical response of an atom at \(r\) given the presence of a second atom at \(r'\). These in turn depend on three-body correlations, etc., resulting in a hierarchy of equations of motion for correlation functions\(^51\–\(^52\).

In a cold, dense ensemble (Fig. 1) this hierarchy can significantly and nonperturbatively modify the scattering behavior even in the classical regime, invalidating attempts to truncate it\(^3\). This is a key ingredient in, e.g., Anderson localization of light, which has been a subject of considerable controversy and debate\(^53\–\(^54\).

**Quantum master equation.** A numerical device for solving this correlation function hierarchy is to treat the atoms as discrete point particles, meaning for a particular configuration of atomic positions \(\{r_1, \ldots, r_N\}\) that two-body correlation functions take the form

\[
\langle \psi_{g,e}^\dagger(r_1) \psi_{g,e}(r_2) \rangle = \sum_{\mu' = g,e} p_{ab,cde}^{(1)}(\mu') \delta(r_1 - r_2),
\]

where \(p_{ab,cde}^{(1)}\) denote correlation functions of the internal atomic energy levels only\(^36\). We then solve the internal atom dynamics at discrete positions, and the new correlation functions simply emerge from the \(N\)-body density matrix \(\rho \equiv \rho_{\{r_1, \ldots, r_N\}}\). This evolves according to QME:

\[
\dot{\rho} = -\frac{i}{\hbar} \left[ H_{\text{sys}}, \rho \right] - \sum_{\mu' = g,e} h\Omega_{\mu'} \hat{a}_{\mu'}^{(1)} \hat{a}_{\mu'}^{(1)\dagger} \rho - \sum_{\mu' = g,e} \sum_{\mu'' = g,e} \gamma_{\mu'\mu''} \left( 2\hat{a}_{\mu'}^{(1)} \hat{a}_{\mu''}^{(1)\dagger} \rho - \hat{a}_{\mu'}^{(1)\dagger} \hat{a}_{\mu''}^{(1)} \rho - \rho \hat{a}_{\mu'}^{(1)\dagger} \hat{a}_{\mu''}^{(1)} \right),
\]

where the collective scattering is represented by the dispersive \(\Omega_{\mu'}\) and dissipative \(\gamma_{\mu'\mu''}\) DD interactions, the single-atom half-width at half-maximum (HWHM) linewidth by \(\gamma_{\mu} = \gamma\), and \(\hat{a}_{\mu'}^{(1)} = (\hat{a}_{\mu'}^{(1)})^\dagger = |\mu\rangle_{\mu'}\langle \mu|\). For simplicity, we consider two-level
atoms and the Hamiltonian

$$H_{sys,j} = -\hbar \Delta \hat{\sigma}_{ee}^{(i)} - d_{ee} \cdot \mathbf{E}^{(e)}(\mathbf{r}_j) \hat{\sigma}_{ee}^{(i)}(\mathbf{r}_j) - d_{ge} \cdot \mathbf{E}^{(g)}(\mathbf{r}_j) \hat{\sigma}_{ee}^{(i)}(\mathbf{r}_j),$$

(3)

where $\mathbf{E}^e$ is the positive-frequency-component of the frequency $\omega = \omega_0$ laser field, detuned from the atomic resonant frequency $\omega_0$ by $\Delta = \omega - \omega_0$ and $\hat{\sigma}_{ee}^{(i)} = |\psi_{ee}^{(i)}(\mathbf{r}_j)$. We take the polarization of the incident field to be parallel to the orientation of the atomic dipoles. Spatial correlations are numerically synthesized by ensemble-averaging over stochastic realizations of atomic positions sampled from the density distribution. Solving Eq. (2) for large systems is numerically taxing, although few-atom ensembles already demonstrate many-body effects in their spectra.

**Semiclassical model.** In the limit of low light intensity, where the excited state population vanishes, the internal level correlations, such as those described by $\rho_{adbc}^{(i)}$ in Eq. (1), also vanish for two-level atoms. The stochastic electrodynamics simulations are then formally exact, reproducing the many-atom spatial correlations, which are identical to those occurring in the classical electrodynamics of coupled linear electric dipoles. Beyond the limit of low light intensity, the full dynamics of Eq. (2) can be greatly simplified by factorizing the internal atomic level correlation functions:

$$\rho_{adbc}^{(i)} \approx \rho_{ad}^{(i)} \rho_{bc}^{(i)}.$$  

(4)

Following the formalism of ref. 36 we then obtain coupled non-linear equations:

$$\frac{d}{dt} \rho_{ge}^{(i)} = (\Delta - \gamma) \rho_{ge}^{(i)} - \frac{i}{\hbar} \left( 2 \rho_{ee}^{(i)} - 1 \right) d_{ge} \cdot \mathbf{E}^e(\mathbf{r}_j) - i \left( 2 \rho_{ee}^{(i)} - 1 \right) \sum_{\ell \neq j} \left( \Omega_{\ell \ell} + i \gamma_{\ell \ell} \right) \rho_{ge}^{(i)}.$$ 

(5)

$$\frac{d}{dt} \rho_{ee}^{(i)} = -2 \gamma \rho_{ee}^{(i)} + \frac{2}{\hbar} \text{Im} \left[ \mathbf{E}^e(\mathbf{r}_j) \cdot \mathbf{d}_{ge} \rho_{ge}^{(i)} \right]$$

$$+ 2 \text{Im} \left[ \sum_{\ell \neq j} \rho_{ge}^{(i)} \left( \Omega_{\ell \ell} - i \gamma_{\ell \ell} \right) \rho_{ee}^{(i)} \right].$$

(6)

Note the relatively small number of equations 2N compared to the full quantum system size 2N. This formalism has been applied to the modeling of pumping of atoms in dense clouds, and has also been extended to cavity quantum electrodynamics (QED).

Spatially correlated scattering between different atoms is accounted for in Eqs. (5) and (6) via $\Delta \mathbf{E}^e$ and $\gamma_{\ell \ell}$ (for $\Omega_{\ell \ell} = \gamma_{\ell \ell} = 0$ they reduce to the independent-atom optical Bloch equations). In the limit of low light intensity the ensemble-averaged response of SCEs coincides with the exact classical electrodynamics; beyond this limit, the model incorporates nonlinear internal level dynamics of the atoms. However, because of the factorization in Eq. (4), they cannot account for many-body quantum entanglement between different atoms’ internal levels. Finding situations in which the predictions of SCEs observably differ from the full QME solution therefore identifies light-induced quantum effects in the transmitted light. Conversely, regimes where quantum fluctuations are minimal allow for the simulation of much larger systems than are accessible with QME, and also test the validity of related approaches in other contexts, based, e.g., on mean-field approximations, intensity expansions, or truncations of the correlations.

**Light-established quantum effects in transmitted light.** We begin by calculating the coherent (Fig. 2a–c) and incoherent (Fig. 2d–f) forward transmission, $T_{coh}$ and $T_{inc}$ (see “Methods” section), through planar square arrays and thin disks of N = 4 atoms (Fig. 1), and the corresponding relative differences (Fig. 3a–c) between quantum and semiclassical results (for the effects of a larger 3 × 3 array, see Supplementary Note 1). The array could be realized, e.g., by an optical lattice or dipole traps. Unless otherwise stated, we consider lattice spacing a = 0.25λ and disk radius R = 0.28λ. Physically, we calculate the far-field light intensity in the same mode as the driving field $\mathbf{E}^e(\mathbf{r})$, integrated over the polar angle $\sin \theta \leq 0.24$ (see “Methods” section). Light-induced position-dependent correlations between the atoms (see earlier “hierarchy of correlation functions”) exist also classically, since the DD interactions depend on the precise interatomic separations. These classical many-body position correlations are therefore present in both the full quantum $T_{coh}^{QM}$ and semiclassical $T_{coh}^{SC}$ transmissions, but all quantum effects have been neglected in $T_{coh}^{QM}$. Hence, we unambiguously identify quantum effects in the coherent transmission by the difference $T_{coh}^{QM} - T_{coh}^{SC}$.

Moreover, since the coherent scattering for a single atom is always purely classical, $T_{coh}^{QM} - T_{coh}^{SC}$ cannot depend on the single-atom response, and therefore represents quantum correlations solely due to many-body effects.

To obtain the incoherently scattered light $\langle \Delta \mathbf{E}_\ell(\mathbf{r}) \Delta \mathbf{E}_\ell^\ast(\mathbf{r}) \rangle$, we write the scattered light field as $\mathbf{E}_\ell = (\mathbf{E}_\ell^0 + \Delta \mathbf{E}_\ell)$, where $\Delta \mathbf{E}_\ell$ denotes the fluctuation. This yields the incoherent transmission (see “Methods” section), from which we also isolate quantum behavior by $T_{inc}^{QM} - T_{inc}^{SC}$, in which case all the quantum effects have again been systematically neglected in $T_{inc}^{QM}$. Although the coherent scattering of a single atom is classical, this is not the case for the incoherent emission. We can improve the semiclassical incoherent model, without increasing the computational complexity, by adding the single-atom quantum description of incoherent light emission for all the atoms. In a single stochastic realization of atomic positions, the incoherent scattering contribution to intensity from independent quantum-mechanical atoms $\propto \sum A_{\ell}(\hat{\sigma}_{ee}^{(i)} - \langle \hat{\sigma}_{ee}^{(i)} \rangle)^2$, where $A_{\ell}$ encapsulates the light propagation effects (see “Methods” section). Augmenting the semiclassical model with this single-atom quantum description integrated over the sample yields the incoherent transmission $T_{inc}^{AQ}$. The quantum effects of the incoherent signal solely due to many-body processes are then encapsulated in $T_{inc}^{AQ}$.

The optical depth of the coherent transmission (Figs. 2a–c, 3a, c) corresponds physically to the degree to which extinction of the incident laser field by the averaged scattered field acts to reduce the transmission. The incoherent fluctuations in the scattered field (Figs. 2d–f, 3b) then counteract this reduction. In Fig. 2b, we identify many-body quantum fluctuations in the coherent transmission ($T_{coh}^{QM} - T_{coh}^{SC}$) that increase with increasing DD interaction (Fig. 3a, c), reaching normalized residuals of over 10% at $\alpha = 0.25\lambda$ and $L = 1$ (when the dipole amplitudes are greatest), where $L = 2c_0 \lambda |\mathbf{E}^e(\mathbf{r} = 0)|^2$ is the peak incident laser intensity and $I_{sat} = 4\pi^2 \hbar c^3/3\lambda^4$ is the saturation intensity. Remarkably, even for a fully random disk quantum effects on the scattering do not wash out, but can produce residuals between the models of a few percent. On the other hand, there are also regimes where $T_{coh}^{SC}$ accurately describes transmitted light. For example, in Fig. 3c the difference between $T_{coh}^{QM}$ and $T_{coh}^{SC}$ is less than 5% for $\alpha > 0.44$ or $L_{sat} > 16$. 
In Fig. 3c we observe two distinct peaks in the quantum many-body signatures of the coherent scattering for $a = 0.25\lambda$. The peak at $\sqrt{I/I_{\text{sat}}} \approx 0.3$ results from a Fano interference between a narrow and an overlapping broad resonance (Fig. 3c inset), originating from an underlying highly subradiant (HWHM = 0.1$\lambda$) and superradiant (HWHM = 2.7$\lambda$) low light intensity eigenmode, respectively. The incident light couples to the phase-matched uniform superradiant eigenmode, but due to nonorthogonality of the non-Hermitian eigenmodes (Supplementary Note 3), the subradiant mode with rapid phase variation ("checkerboard" phase pattern, with dipoles at adjacent sites $\pi$ out-of-phase) becomes populated at the narrow resonance (Fig. 2a). Strikingly, around this narrow resonance quantum effects constitute over 30% of the coherent scattering signal. We observe in Fig. 3c that, with increasing lattice spacing, the narrow Fano peak at $\sqrt{I/I_{\text{sat}}} \approx 0.3$ disappears, as the corresponding eigenmode becomes less subradiant.

In Fig. 2e, f, on the other hand, we see the incoherent transmission is almost entirely dominated by quantum fluctuations when $I \gtrsim I_{\text{sat}}$ ($T_{\text{inc}}^{\text{SC}} \to 0$). However, once we incorporate the single-atom quantum description into the scattering and therefore transmission $T_{\text{inc}}^{\text{SACQ}}$, the difference becomes much smaller and the many-body quantum fluctuations are, as with the coherent scattering, maximal around $I \sim I_{\text{sat}}$. Hence, using the improved model $T_{\text{inc}}^{\text{SACQ}}$, it is possible, even for incoherent transmission, to obtain excellent qualitative, and frequently quantitative agreement with the full quantum scattering (see Supplementary Note 4 for further demonstration of the different roles of quantum fluctuations in the incoherent scattering).

**Spin correlations and entanglement.** Up until now, we have identified many-body quantum effects via the transmitted light. These originate from the light-induced quantum correlations between internal levels of different atoms that do not satisfy the factorization assumption [given in Eq. (4)] of SCEs. In Fig. 4 we explicitly show these induced spin-spin correlations, and in Fig. 5a–c we show the many-body entanglement of formation (for a pair of atoms, using the formalism of ref. 67) — see Supplementary Note 5. The spin–spin correlations and entanglement exhibit behavior qualitatively similar to the quantum many-body correlations observed in the light scattering. As in Fig. 3c, both the correlations (Fig. 4) and entanglement (Fig. 5a–c) are maximal at $I \sim I_{\text{sat}}$, where the intensity at which this peak occurs and the peak’s amplitude decrease for increasing atomic spacing. The correlations and entanglement also both manifest linesplitting for $I \gtrsim I_{\text{sat}}$, corresponding to when the atomic excited state population starts to increase. This is consistent with Fig. 2, where the observed quantum effects in transmitted light change from being maximal close to zero detuning when $I \sim I_{\text{sat}}$ to being maximal off resonance when $I \gg I_{\text{sat}}$. The purity of the atomic state (Fig. 5d) decreases to the limiting value 1/4 (Supplementary Note 5) for increasing $I$ and is less sensitive to the array spacing than the entanglement $E$.

**Large ensembles.** When conditions are such that quantum effects on the light scattering are minimal, we can neglect quantum fluctuations, employing SCEs [Eqs. (5) and (6)] to analyze the coherent transmission through much larger ensembles, for which the full QME is inaccessible. In Fig. 6a–c we show how the
transmission lineshapes of a 10 × 10 array differ significantly from the Lorentzian of independent atoms. For a single atom the linewidth is power broadened γ_{PB}(I) = γ \sqrt{1 + I/I_{sat}}. For spacings a ≥ 0.3λ, the coherent optical depth for a 10 × 10 interacting array can also be fitted well to a single Lorentzian (ignoring the small structure present at low light intensity due to interfering eigenmodes, Fig. 6a), with a linewidth which also scales with intensity. If the linewidth at low light intensity is superradiant (e.g., a = 1.1λ) or subradiant (e.g., a = 0.8λ), it will also be correspondingly larger or smaller than γ_{PB} for high intensities (Fig. 6c). For smaller spacings (e.g., a = 0.25λ), at high intensity the lineshape splits into a double resonance, producing a dip or "hole burning" (Fig. 6b). This dip is analogous to vacuum Rabi splitting, where the interatomic DD coupling has now taken the role of the cavity coupling. In cavity QED, the mirrors create images of an atom inside the cavity, mimicking a periodic array, and the resonance doublet can be understood as a splitting of the excited state (Supplementary Note 6). While the dip in Fig. 6b only occurs at sufficiently high density, it can interestingly still exist even in the fully random ensemble. With increasing light intensity, the incoherent scattering lineshape tends to that of the independent atom (Supplementary Note 7).
Fig. 6 Semiclassically evaluated optical depth, resonance Rabi splitting, power broadening, and maximum extinction for 100 atoms. a, b From bottom to top: semiclassical coherent optical depth $\Delta \gamma$, for a $10 \times 10$ square array with lattice constant $a = 0.25\lambda$ and normally distributed position fluctuations (rms width $\sigma_x = 0.0, 0.1, 0.2$, respectively); and 100 atoms randomly distributed within a radius $R = 1.4\lambda$ (peak density $10^5$ thin disk. The standard sampling errors (shaded regions) are too small to see. For clarity each line is offset from that below it by 0.5. Fluctuations in thin disk. The standard sampling errors (shaded regions) are too small to see. It suggests that optical quantum information processing in atomic ensembles need not necessarily be restricted to dilute systems. Subradiant resonance narrowing has now been experimentally observed in the transmitted light through an optical lattice of atoms in a Mott-insulator state in the classical limit of low light intensity. Several of our findings could also be verified in this setup by increasing the intensity of the incident light. The presence, even in uniform ensembles, of many-body effects attracting considerable interest further relaxes the conditions necessary for their experimental observation.

Methods

Dynamics and correlation functions. We simulate the optical response of $N$-atom ensembles by stochastically sampling fixed positions $\{r_1, ..., r_N\}$ of stationary atoms, as the atomic center-of-mass dynamics are assumed negligible. In the full quantum dynamics, for each stochastic realization we solve the equations of motion for the $N$-atom density matrix $\rho_{\{r_1, ..., r_N\}}$ with the atoms at fixed positions $\{r_1, ..., r_N\}$, obeying QME [Eq. (2)]. In the Hamiltonian in Eq. (3) we use slowly-varying field amplitudes and atomic variables where the rapid rotation at the laser frequency has been factored out by substitutions $\mathcal{E} \rightarrow \mathcal{E}_c$, $\mathcal{E}_c \rightarrow \mathcal{E}_c t$, etc. The collective coupling matrices $\Omega_c$ and $\gamma_c$ resulting, respectively, in collective resonance shift and linewidths in Eq. (2) (see Supplementary Note 3), are the real and imaginary parts of the dipole radiation kernel $G(r)$:

$$\frac{1}{\hbar} \delta_{d} \left[ \mathcal{G}(r_i - r_j)\mathcal{d}_{ij} \right] = \Omega_{ij} + i\gamma_{ij},$$

where

$$\mathcal{G}(r) = \frac{\mathcal{E}_c^*}{4\pi} \left\{ (n \times \mathbf{d}) \times n \mathbf{d} \right\} \frac{e^{i\mathbf{d}\cdot(\mathbf{r}_i - \mathbf{r}_j)}}{kr} + \frac{1}{3} \mathbf{d} \cdot \frac{1}{kr} \mathbf{d} \frac{e^{i\mathbf{d}\cdot(\mathbf{r}_i - \mathbf{r}_j)}}{kr^3}$$

is the electric field amplitude for an oscillating electric dipole $\mathbf{d}$ at the origin and $n = r/r$. Note that we typically drop the contact interaction term.

Once $\rho_{\{r_1, ..., r_{N-1}\}}$ is known, the one-body $\rho_{\{r\}}^0$ (5th atom), two-body $\rho_{\{r_2, r_3\}}^0$ (5th and 6th atoms), etc., expectation values for this stochastic realization are given by:

$$\rho_{\{r\}}^0 = \left\langle \mathbf{\hat{P}}_r^0 \right\rangle = \text{Tr} \left( \mathbf{\hat{P}}_r^0 \rho_{\{r_1, ..., r_{N-1}\}} \right),$$

$$\rho_{\{r_2, r_3\}}^0 = 1 - \rho_{\{r\}}^0 = \left\langle \mathbf{\hat{P}}_r^0 \right\rangle = \text{Tr} \left( \mathbf{\hat{P}}_r^0 \rho_{\{r_1, ..., r_{N-1}\}} \right),$$

and so forth. Here $\rho_{\{r\}}^0$ represents the correlations $\langle \mathbf{\hat{P}}_r^0 \rangle$ of Eq. (14), and corresponds to the matrix element $(\mathbf{r}_i | \mathbf{r}_j)$ of the one-body density matrix $\rho_{\{r\}}$.
In each stochastic realization, the N-atom configuration of positions \( \{r_1, \ldots, r_N \} \) is obtained by sampling from a joint probability distribution \( P(r_1, \ldots, r_N) \), taken to be the initial distribution of stationary atoms. Ensemble-averaging over many such realizations then transforms the expectation values \( \langle \hat{b}_i(t) \rangle, \langle \hat{b}_i^\dagger(t) \rangle \), etc., to spatial correlation functions for the atoms at any given time \( t \):

\[
\left\langle \hat{b}_i(t) \hat{b}_j^\dagger(t) \right\rangle = \int d^3r_1 \ldots d^3r_N \left\langle \hat{b}_i(r_1) \hat{b}_j^\dagger(r_2) \right\rangle P(r_1, \ldots, r_N),
\]

and so forth for higher-order correlations. The atomic correlation functions for a single realization of fixed atomic positions are given in terms of \( \rho_{ab} \) and \( \rho_{ab\hat{b}^\dagger\hat{a}^\dagger} \) by:

\[
\left\langle \hat{b}_i(t) \hat{b}_j^\dagger(t) \right\rangle = \sum_{\nu} \rho_{ab}(t) \delta(r_i - r_j),
\]

and through solving the coupled dynamics between the light and atoms for each stochastic run and ensemble-averaging over many such realizations that we establish the light-induced spatial correlations between atoms.\(^{3,2}\)

For a single, isolated atom at the laser focus, the solutions to Eqs. (5), (6) (i.e., the optical Bloch equations) in the steady state are:

\[
\rho_{ab} = \frac{d_{ab} \cdot E^+(0)}{\hbar} \frac{\Delta - \gamma E}{\Delta + \gamma^2 (1 + I/I_{\text{sat}})},
\]

where \( I/I_{\text{sat}} = 2d_{ab} E^+(0)/|\gamma| \). The effective linewidth in the denominators of Eqs. (16) and (17) is now power broadened, i.e., \( \gamma_{\text{eff}} = \gamma \sqrt{1 + I/I_{\text{sat}}} \).

We compare the full quantum solution of QME [Eq. (23)] with SCES Eqs. (5) and (6) for the one-body terms \( \rho_{ab}(t) \) based on the factorization \( \rho_{ab\hat{b}^\dagger\hat{a}^\dagger} \approx \rho_{ab\hat{b}^\dagger\hat{a}^\dagger}(t) \), which neglects quantum fluctuations. In terms of the stochastic sampling procedure, we express this semiclassical factorization as:

\[
\left\langle \hat{b}_i(t) \hat{b}_j^\dagger(t) \right\rangle_{\text{SC}} = \sum_{\nu} \left\langle \hat{b}_i(r) \hat{b}_j^\dagger(r) \right\rangle_{\nu},
\]

where \( \nu \) is the term present when the annihilation operators refer twice to the same atom. Despite the factorization of the internal atomic correlation functions, we generally have:

\[
\left\langle \hat{b}_i(t) \hat{b}_j^\dagger(t) \right\rangle_{\text{SC}} = \sum_{\nu} \left\langle \hat{b}_i(r) \hat{b}_j^\dagger(r) \right\rangle_{\nu},
\]

as the fluctuations of the atomic positions that are included in SCES approach can result in strong light-induced correlations.

In general for the atomic distribution before the light is turned on, we have:

\[
\left\langle \hat{b}_i(t) \hat{b}_j^\dagger(t) \right\rangle = \frac{1}{\hbar} \frac{d_{ab}}{d_{ab}} \frac{\Delta - \gamma E}{\Delta + \gamma^2 (1 + I/I_{\text{sat}})},
\]

and the incoherently scattered light intensity in Eq. (25) vanishes. Unlike the coherent scattering, the incoherent scattering for a single atom therefore depends on whether we treat it in a quantum or semiclassical manner.

Generalizing to the many-atom case, Eq. (24) now becomes:

\[
\left\langle \hat{b}_i(t) \hat{b}_j^\dagger(t) \right\rangle_{\nu} = \frac{1}{\hbar} \frac{d_{ab}}{d_{ab}} \frac{\Delta - \gamma E}{\Delta + \gamma^2 (1 + I/I_{\text{sat}})},
\]

where, as in Eq. (20), \( G(r - r') \) acts on \( \hat{P} \cdot \hat{R} \) and likewise \( G(r - r') \) on \( \hat{P} \cdot \hat{R} \). When calculating the full quantum solution, the correlation functions are evaluated using the solution to QME [Eq. (23)] and by ensemble-averaging over many realizations of atomic positions. However, we can also introduce another basis version of the single-atom semiclassical approximation [Eq. (26)] to light scattering:

\[
\left\langle \hat{b}_i(t) \hat{b}_j^\dagger(t) \right\rangle_{\text{SC}} = \frac{1}{\hbar} \frac{d_{ab}}{d_{ab}} \frac{\Delta - \gamma E}{\Delta + \gamma^2 (1 + I/I_{\text{sat}})},
\]

and substituting this back into Eq. (27) to give the semiclassical scattered field:

\[
\left\langle \hat{b}_i(t) \hat{b}_j^\dagger(t) \right\rangle_{\text{SC}} = \frac{1}{\hbar} \frac{d_{ab}}{d_{ab}} \frac{\Delta - \gamma E}{\Delta + \gamma^2 (1 + I/I_{\text{sat}})},
\]

Deriving the semiclassical scattered field in Eq. (29) corresponds to a systematic way of neglecting all quantum fluctuations when the atomic response is first calculated from SCES [Eqs. (5) and (6)]. Hence, comparing the scattered light of Eq. (29) with the equivalent full quantum solution of Eq. (27) provides a signature for quantum effects in the collective atomic response. Alternatively, if our goal is to determine a computationally efficient and accurate approximation to the full quantum solution, we can instead try to improve the semiclassical approximation. A simple way to achieve this without increasing computational demands is to include the single-atom quantum description to incoherent scattering [Eq. (25)] integrated over the extent of the sample, which is sufficient in a number of cases to capture the leading quantum contributions.

We begin this procedure by placing the atomic operators in Eq. (27) in the normal order. This yields for the expectation term on the right hand side of

**Scattered light.** The total electric field operator \( \hat{E}^\dagger(r) = \hat{E}^\dagger(r) + \hat{E}_i^\dagger(r) \) is the sum of the laser field and the fields scattered from all atoms:

\[
\langle \hat{E}_i^\dagger(r) \rangle = \int d^3R \langle G(r - R) \rangle \hat{P}(R).
\]
The data presented in this study are available in the Durham University repository with the identifier https://doi.org/10.15128/r1zc77sq127. The codes used to generate these data are available in the Zenodo repository with the identifier https://doi.org/10.5281/ZENODO.3924698.

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Author contributions
R.J.B. performed the numerical simulations with assistance from M.D.L. J.R. and R.J.B. developed the theory and derivations. R.J.B., S.A.G., and J.R. contributed to the manuscript text and R.J.B. generated the figures.

Competing interests
The authors declare no competing interests.

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