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Perspective

Towards deterministic quantum non-linear optics with Cu₂O Rydberg polaritonsHamid Ohadi^{a,*}, David Petrosyan^{b,c,*}, Matthew P.A. Jones^{d,*}, Pavlos G. Savvidis^{b,e,*}^aSUPA, School of Physics and Astronomy, University of St Andrews, St Andrews KY16 9SS, United Kingdom^bInstitute of Electronic Structure and Laser and Center for Quantum Science and Technologies, FORTH, Heraklion 70013, Greece^cA. Alikhanyan National Science Laboratory (YerPhI), Yerevan 0036, Armenia^dDepartment of Physics, Durham University, Durham DH1 3LE, United Kingdom^eDepartment of Physics, School of Science, Westlake University, Hangzhou 310014, China

Photons are ideal carriers of quantum information: they are fast, low-loss, and compatible with mature photonic infrastructure. Linear optical quantum computing exploits these properties but relies on probabilistic measurements and post-selection, incurring an exponential resource overhead that limits scalability [1]. Circumventing this bottleneck requires deterministic quantum logic gates, which in turn demand strong, controllable interactions between individual photons.

Photons do not interact in free space or in linear optical media. The standard route to attain effective photon–photon interactions is to hybridise photons with interacting matter excitations, forming polaritons. The matter component introduces an interaction-induced anharmonic level shift U into the ladder of polariton states: when one polariton occupies a mode, the next photon sees a shifted resonance (Fig. 1a). The key figure of merit is U/γ , where γ is the total polariton decoherence rate. For $U/\gamma \ll 1$ the nonlinearity is classical; for $U/\gamma \gtrsim 1$ the system enters the quantum nonlinear regime, enabling photon blockade, large conditional phase shifts, and deterministic photonic gates.

Atomic Rydberg systems have provided the clearest demonstration of this regime. Atoms excited to Rydberg states—electronic states with large principal quantum numbers $n \gg 1$ —possess unique properties [2]. Their lifetimes scale as $1/\gamma \propto n^3$ and transition dipole moments as $\mu \propto n^2$, leading to strong dipole–dipole ($U \propto n^4/r^3$) or van der Waals ($U \propto n^{11}/r^6$) interactions over long distances r . These interactions can be imprinted onto photons via electromagnetically induced transparency (EIT) [3]: photons hybridise with collective atomic excitations to form slowly propagating Rydberg polaritons that strongly interact with each other over distances of up to tens of micrometres [4]. This mechanism has produced photon blockade [5] and gates [6], establishing Rydberg EIT polaritons as the most advanced platform for quantum nonlinear optics to date [4]. Yet atomic implementations face low single-pass optical depth, requiring long interaction distances to

accumulate sufficient nonlinear phase, and are constrained by the need for laser cooling and trapping (Fig. 1d), making chip-scale integration technically demanding.

Solid-state platforms offer a chip-compatible alternative: micron-scale confinement, higher optical densities, and direct compatibility with semiconductor fabrication. Among solid-state candidates, Rydberg excitons in the direct-gap semiconductor Cu₂O combine a well-resolved hydrogenic (Rydberg) series extending beyond $n = 25$ [7] with the potential for on-chip integration. Creating Rydberg exciton–polaritons in microcavities [8] imprints Rydberg interactions onto photons, enabling nonlinear energy shifts that already surpass those of GaAs [9], the previously best-performing solid-state platform. Here we examine the physical basis and current status of this system, identify the specific problems hindering the attainment of the regime of nonlinear quantum optics, and outline a roadmap toward photon blockade and deterministic gate operation.

Rydberg polaritons in Cu₂O. Excitons in Cu₂O form a hydrogenic series with principal quantum numbers observable above $n = 25$ and spatial extents $x_n \propto n^2$ exceeding one micrometre at $n \approx 25$ (Fig. 1c) [7]. The dipole moments and the resulting van der Waals interactions of these states have similar n -dependence as their atomic counterparts [10]. Strong light–matter coupling in an optical microcavity hybridises photons and excitons into polaritons. Their excitonic component inherits Rydberg interactions $V(r) \propto r^{-6}$ whereas the photonic component enables easy addressability. When two polaritons occupy the same mode, their excitonic components interact and shift the two-polariton energy by $U \propto 1/V_{\text{mode}}$ (Fig. 1a), where V_{mode} is the effective mode volume. Hence, confining the optical mode to smaller volumes V_{mode} increases the effective interaction between the polaritons. Micropillar cavities lithographically defined from Cu₂O layers (Fig. 1e) can reduce mode volumes to the cubic-micrometre scale [11], strongly enhancing U .

In addition to cavity confinement, ladder-type excitation via a two-step optical–THz scheme provides a powerful mechanism to enhance polariton nonlinearities [12]. By coupling low-lying exciton–polaritons to highly excited Rydberg states using coherent THz radiation, EIT can be implemented in the solid state (Fig. 1b).

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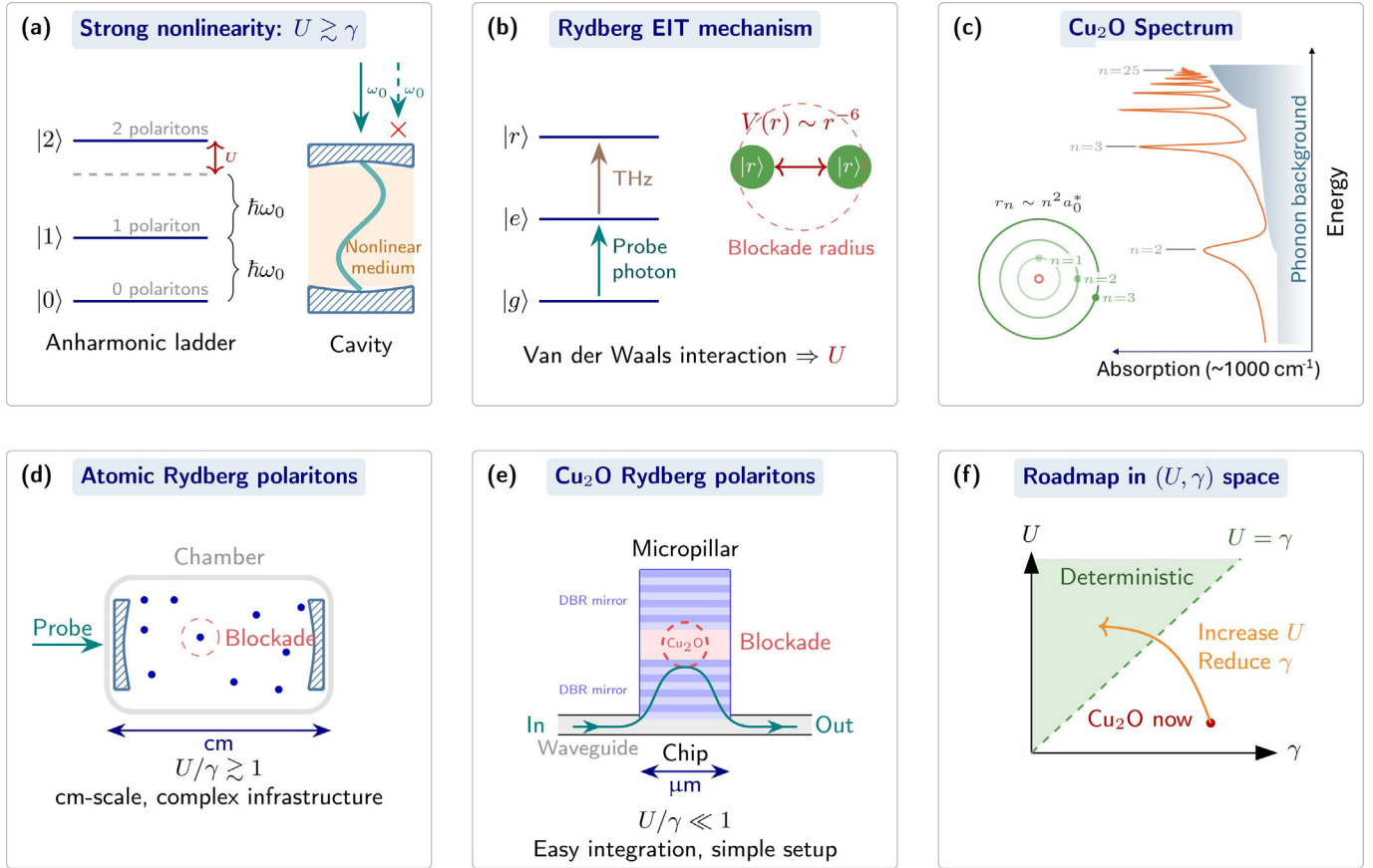


Fig. 1. Photon blockade, Rydberg polariton mechanism, and platform comparison. (a) Anharmonic ladder of polariton levels: the two-polariton state $|2\rangle$ is shifted by U relative to the harmonic level (dashed), blocking a second photon from entering the cavity. (b) Rydberg EIT mechanism: THz coupling on the transition to $|e\rangle \rightarrow |r\rangle$ transfers van der Waals interactions $V(r) \sim r^{-6}$ between the excitonic states $|r\rangle$ to the polariton mode, leading to the nonlinear shift U . (c) Cu_2O absorption spectrum showing the discrete Rydberg exciton series above the phonon background. (d) Atomic Rydberg polaritons achieve $U/\gamma \gtrsim 1$ but require cm-scale apparatus and laser cooling and trapping. (e) Cu_2O micropillar on chip offers μm -scale confinement and waveguide integration, currently having $U/\gamma \ll 1$. (f) Roadmap in (U, γ) space towards the deterministic quantum non-linear optics regime (shaded).

In this regime, polaritons acquire a large Rydberg excitonic component while remaining optically addressable.

Current Cu_2O Rydberg polariton devices operate at $U/\gamma \lesssim 10^{-3}$ [9], which is still three orders of magnitude below the strong interaction regime (Fig. 1f), but it already surpassed the nonlinear energy shifts in optimised GaAs devices [9], which is remarkable given the relative novelty of this platform. Currently U is limited by large optical mode volumes in existing microcavity designs and by the moderate exciton quantum numbers achievable at the polariton resonance; whereas γ is dominated by exciton-phonon coupling, which generates a broad absorption continuum superimposed with the discrete Rydberg resonances [13], and by material disorder and cavity scattering losses. While neither shortcoming appears fundamental to the material system, overcoming them is far from trivial: phonon-assisted decoherence at high n , disorder and charged-defect broadening in synthetic Cu_2O , strong coupling in nanostructured devices, and THz ladder EIT in a microcavity all remain unresolved, and progress requires simultaneously increasing U and reducing γ while performing crystal growth, cavity engineering, phonon mitigation, and THz driving.

Roadmap. In Fig. 2 we summarize the roadmap toward achieving $U/\gamma \gtrsim 1$, organised by the objectives (increase U , reduce γ) and timescale (near-term, long-term). Near-term strategies will exploit gains achievable with current fabrication capabilities, while long-term strategies will require new techniques, such as THz ladder dressing and phononic bandgap engineering, which are theoretically well-founded but not yet realised in Cu_2O .

Increasing the polariton nonlinearity U . The intrinsic interaction strength grows rapidly with the principal quantum number n of the exciton state. Current Cu_2O polariton devices reach $n = 7$ [9]; increasing this to $n = 25$ would increase U by a factor of $(25/7)^{4.4} \approx 270$ based on the experimentally observed scaling $n^{4.4 \pm 1.8}$ [9]. But higher- n states also experience stronger phonon-assisted absorption, which increases γ and partially offsets the gain in U/γ . This limitation can be mitigated by EIT.

Optical confinement offers a complementary near-term route for enhancing U . Advanced mechanical thinning of bulk Cu_2O and thin-film growth [14] enable micropillar fabrication with mode volumes of the order of r_b^3 , where r_b is the blockade radius of the Rydberg excitation, i.e., the distance at which the interaction-induced Rydberg level shift is equal to or larger than the excitation linewidths. Nanofabrication of photonic crystal cavities offers sub-micron confinement and could push V_{mode} below $1 \mu\text{m}^3$ as a longer-term target.

The EIT mechanism provides the most powerful long-term lever for increasing U . In this scheme, coherent THz radiation couples the low- n exciton state $|e\rangle$ to a high- n Rydberg state $|r\rangle$, opening a narrow transparency window for the probe photon and imprinting strong Rydberg interactions onto the polariton, without requiring the optical photon to be resonant with the transition $|g\rangle \rightarrow |r\rangle$. This transfers the experimentally observed $U \propto n^{4.4}$ interaction scaling to the polariton mode [12]. THz ladder dressing in Cu_2O microcavities has been theoretically proposed [12] but not yet demonstrated. However, experiments have demonstrated the dressing

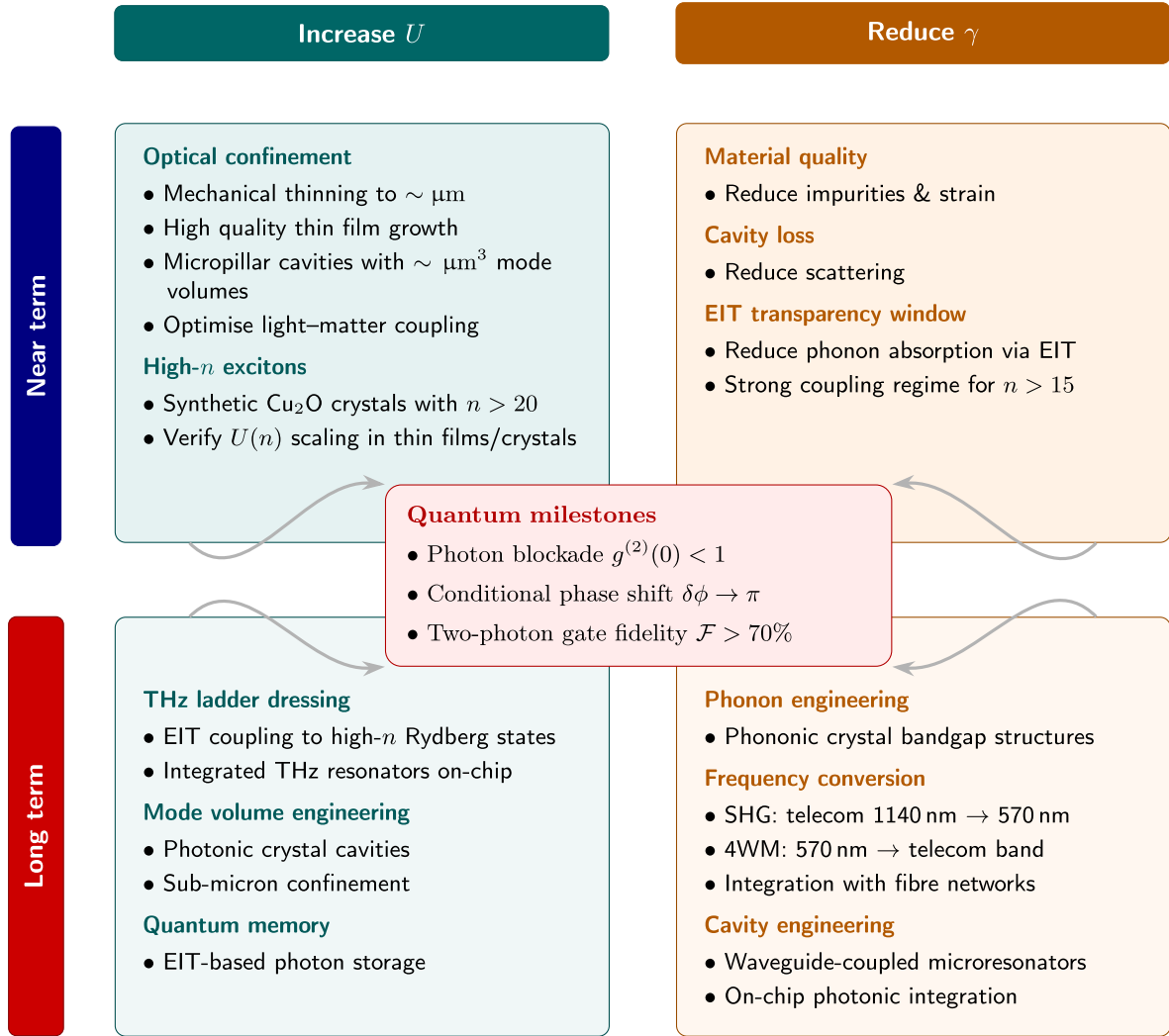


Fig. 2. Roadmap toward deterministic quantum nonlinear optics with Cu_2O Rydberg exciton-polaritons, organised by objective and timescale. Near-term strategies focus on achievable gains through optical confinement, material quality, and cavity engineering. Long-term strategies exploit THz ladder dressing to transfer strong Rydberg-state interactions to the polariton mode, phononic bandgap engineering to suppress decoherence, and frequency conversion for telecom integration. All four quadrants converge on the central quantum milestones of photon blockade and deterministic gate operation.

of high- n Rydberg states using microwave radiation up to 20 GHz [15]. The next steps are to extend these results to microcavities and to the THz region to fully exploit the strong optical coupling of the low- n intermediate state ($|e\rangle$ Fig. 1b) and the EIT ladder scheme. On-chip integration of THz resonators with optical micropillars would enhance dressing efficiency and reduce the THz power required for coherent coupling. THz quantum cascade lasers operate at low temperatures, making this approach compatible with the cryogenic infrastructure already required for high- n states.

Reducing the polariton decoherence rate γ . There are three complementary strategies for reducing γ : suppressing phonon-assisted absorption via EIT, engineering vibrational bandgaps with phononic crystals, and improving cavity and material quality to reduce scattering and inhomogeneous broadening.

Exciton-phonon coupling is the dominant decoherence channel in Cu_2O . It generates a broad absorption continuum that increases the polariton linewidth and suppresses U/γ even when U is large. EIT directly suppresses this loss: coherent coupling to a long-lived Rydberg state creates a narrow transparency window in the phonon-broadened continuum, selectively reducing absorption at the polariton resonance [16]. The achievable suppression is governed

by the competition between the discrete excitonic and phonon-continuum excitation pathways, quantified by the ratio $\beta = g_p\Omega/(g\Omega_s)$ [16], where g_p and g are the probe-field couplings to the discrete exciton state and to the phonon-dressed continuum, respectively, and Ω_s and Ω are the corresponding control-field Rabi frequencies coupling these two types of states to the long-lived Rydberg level: at $\beta = 1$ the two pathways share a common dark state and absorption vanishes on two-photon resonance, while for $\beta \gg 1$ the background is suppressed to a residual set by the Rydberg-state linewidth. Since β is a material constant of Cu_2O that has not yet been measured, its experimental determination is an early priority. This mechanism links the near-term and long-term strategies, since the same THz dressing that increases U simultaneously reduces γ .

Phononic crystal structures open vibrational bandgaps at frequencies resonant with the exciton transitions, passively suppressing phonon-assisted absorption. At the material level, reducing impurities, strain, and Kirkendall voids from the oxidation process [17] suppresses inhomogeneous broadening. High- Q distributed Bragg reflector cavities with reflectivities exceeding 99.9%—routinely achieved in III-V systems and transferable to the Cu_2O platform—minimise photon leakage and further reduce γ .

Taken together, an order-of-magnitude budget indicates that no single improvement can lead to three orders of magnitude gain in U/γ , but their combination can. Reducing V_{mode} from the $\sim 30 \mu\text{m}^3$ of current planar cavities—set by the $\sim 30 \mu\text{m}$ cavity thickness at near-diffraction-limited spot areas of $\sim 1 \mu\text{m}^2$ —to the $\sim 1 \mu\text{m}^3$ of thinned micropillars is a geometric gain of up to ~ 30 in U , bounded below by the blockade volume r_b^3 and requiring no THz infrastructure. Raising the principal quantum number from $n = 7$ to $n = 20$ – 25 increases the underlying exciton–exciton interaction by a further factor of $\approx 10^2$ based on the measured $n^{4.4 \pm 1.8}$ scaling [9]; the fraction of this gain inherited by the polariton under THz dressing is, however, set by the Rydberg admixture of the dressed state, which grows with the THz Rabi frequency, while the $|e\rangle \rightarrow |r\rangle$ coupling strength is expected to fall as $\sim n^{-3/2}$ [16]. The realised enhancement therefore depends on the THz field strengths achievable in practice—motivating resonant enhancement with on-chip THz resonators—and, together with the phonon-suppression parameter β introduced above, constitutes an experimentally undetermined quantity of the roadmap. Reductions in γ provide additional benefit: EIT suppression of the phonon-assisted background is bounded above by full transparency on two-photon resonance but is governed by β , while improved mirrors and material quality contribute a further factor of a few. These factors are not fully independent—the confinement gain saturates at the blockade volume, and the interaction transfer and loss suppression are both set by the THz coupling—but the gap closes through their combination. Resolving the two THz-related unknowns, the achievable dressing Rabi frequency and the pathway ratio β , is thus an early experimental priority.

Synthetic crystal growth. Scalable quantum photonics with Cu_2O requires synthetic crystals: natural crystals cannot be thinned, patterned, or integrated on chip in a reproducible way. Several synthetic routes are under development, all based on oxidation of copper. Floating-zone growth produces bulk-like crystals in which Rydberg excitons up to $n = 15$ have been observed [18], and thermal oxidation of free-standing Cu foils produces millimetre-scale grains [17]; both require mechanical thinning before device integration. Oxidation of Cu films deposited on transparent substrates yields thin films that are immediately compatible with micropillar and waveguide fabrication, with Rydberg excitons up to $n = 8$ already demonstrated in CMOS-compatible material [14]. Across all routes, the gap to natural crystals ($n > 30$) reflects charged point defect density—now identified by DFT as oxygen and copper interstitials rather than the previously assumed vacancies [19]—which broaden exciton lines and suppress the maximum observable n . Reducing these defects through controlled growth conditions is therefore the principal material's challenge on the path to scalable Cu_2O devices.

Device architecture and telecom integration. Waveguide-coupled micropillar cavities and microresonators provide a platform for integrating nonlinear elements with interferometers and detectors. On-chip THz resonators resonantly enhance the Rydberg dressing efficiency and are compatible with optical confinement structures. Rydberg excitons up to $n = 25$ are accessible [7] at 4K; higher- n states require dilution-refrigerator temperatures. This is not a platform-specific barrier: superconducting single-photon detectors, already standard in quantum photonic experiments, demand comparable cooling, so cryogenic infrastructure is a shared requirement rather than an additional overhead.

Second-harmonic generation converts telecom photons at 1140nm up to the Cu_2O resonance at 570nm, providing a route to inject telecom-band input into the nonlinear cavity; bulk conversion at this wavelength has been demonstrated [20]. The reverse direction—converting Cu_2O emission at 570nm into the telecom O-band for fibre transmission—is achievable via four-wave

mixing (4WM). But neither process has yet been demonstrated at the single-photon level in waveguide-integrated nonlinear crystals.

Experimental milestones and quantum benchmarks. Three sequential measurements define the quantum benchmark hierarchy. Observation of antibunched emission with suppressed second-order correlations $g^{(2)}(0) < 1$ would mark entry into the quantum nonlinear regime; a stringent demonstration of photon blockade further requires pulsed correlation measurements with $g^{(2)}(0) \ll 0.5$ under resonant coherent driving, supported by spectroscopy of the two-polariton resonance shifted by U , to exclude interference-based (unconventional) antibunching and classical filtering artefacts. Interferometric measurement of a conditional phase shift $\delta\phi$ approaching π would demonstrate the two-photon interaction strength required for gate operation, and a controlled-phase gate with fidelity $\mathcal{F} > 1/\sqrt{2}$ would establish operation above the classical limit. Each milestone maps onto a characteristic interaction-to-decoherence ratio—blockade requires $U/\gamma \gtrsim 1$, a π phase shift $U/\gamma \sim \pi$, and a strictly higher ratio for gate operation—but these are indicative thresholds for a single-mode cavity model. The gate performance will actually depend on the precise protocol and will be affected by decoherence during the photon interaction or storage time, spatial mode-matching, input-output loss, or pulse bandwidth relative to the cavity linewidth.

Among the possible gate architectures, such as cavity blockade, travelling-photon phase gates, and stored-photon gates, we envisage a two-stage path built on the same hardware. The near-term milestone is cavity blockade: a single waveguide-coupled microcavity operated as a photon turnstile, for which the relevant requirements are $U/\gamma \gtrsim 1$ and efficient input-output coupling. The long-term target is a Mach–Zehnder controlled-phase gate: dual-rail photonic qubits propagate through an on-chip interferometer with the nonlinear microcavity in one arm providing the interaction-induced phase approaching π for $U/\gamma \gtrsim \pi$ when both photons simultaneously transit the cavity. This converts the conditional-phase measurement of the second milestone directly into the gate. The architecture requires an over-coupled, low-loss cavity, photon bandwidths well within the cavity linewidth, temporal overlap of the two photons, and a path-balanced interferometer—all compatible with integrated photonics—but, crucially, no photon storage. A single-pass travelling-photon gate without cavity enhancement is disfavoured, since micrometre-scale films cannot provide the required optical depth.

While the Mach–Zehnder architecture requires only temporal overlap of the two photons rather than storage, a quantum memory would substantially extend its reach. EIT-based photon storage, in which a photon is mapped onto a collective Rydberg coherence and retrieved on demand, provides a natural mechanism within the Cu_2O platform: the same Rydberg coupling used for ladder dressing can be adapted for storage, relaxing the simultaneity requirement, extending the effective interaction time—and thus the accumulated nonlinear phase—and enabling synchronisation between gates in larger circuits. An alternative storage route is provided by coherently mapping the excitation onto long-lived dark states, such as the lowest-energy 1S paraexciton or crystal defect states [20], which bypass the phonon-limited polariton lifetime entirely. Demonstrating either form of storage in Cu_2O is therefore a desirable long-term capability, contingent on first realising THz ladder dressing in a microcavity geometry.

Conclusions. Cu_2O Rydberg exciton–polaritons already deliver nonlinear energy shifts exceeding those of optimised GaAs devices [9]—a system with decades of platform development behind it—despite operating three orders of magnitude below the quantum nonlinear regime. No known fundamental material limit sets this

gap, and concrete strategies exist to close it—higher- n excitons, micropillar cavities with small mode volume $V_{\text{mode}} \lesssim 1 \mu\text{m}^3$, THz ladder dressing, and phonon engineering—although several of these, in particular THz ladder EIT in a cavity geometry, remain to be demonstrated. Of these, suppressing phonon-assisted decoherence is the most urgent priority: exciton–phonon coupling currently limits γ regardless of gains in U , and EIT-based phonon suppression is the one mechanism that simultaneously reduces γ and enables higher- n operation without additional fabrication overhead. Given the pace of recent progress—from the first Rydberg exciton–polaritons in 2022 [8] to nonlinear shifts surpassing GaAs in 2024 [9]—and given that concrete strategies have been identified for each element of the remaining gap, photon blockade in Cu_2O is a realistic medium-term target and deterministic gate operation a credible long-term one.

Beyond lab demonstrations, industrial adoption of Cu_2O as a quantum photonic platform rests on two additional prerequisites. Scalable synthetic crystal growth, particularly thin-film routes that eliminate mechanical thinning, must advance from individual device demonstrations to reproducible wafer-scale fabrication. Telecom-band frequency conversion, linking the Cu_2O operating wavelength to the fibre O-band, is required for integration with existing quantum network infrastructure. Neither is a fundamental obstacle: the crystal growth trajectory is well-defined, and the nonlinear optical processes for frequency conversion are established in other material systems. Together they define the engineering path from photon blockade to deployable deterministic quantum photonic gates.

Conflict of interest

The authors declare that they have no conflict of interest. During the language polishing and image generating of this study, Anthropic Claude Opus 4.7 was used. All generated content has been verified by the authors, who take full responsibility for the authenticity of the final data and the originality of the conclusions. No AI was used to generate or modify any original data or experimental images.

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