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Superextensive electrical power from a quantum battery

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Abstract

Superextensivity, where the response of a physical system scales super-linearly with size, originates from collective quantum effects and provides a promising route to augment next-generation quantum technologies. While recent work has demonstrated superextensive behaviour in the coherent dynamics of quantum systems, these effects typically occur on short timescales, prohibiting their practical utility. In contrast, triggering steady-state superextensive effects in, for example, a generated electric current, remains unexplored despite the immediate impact on photovoltaic technologies. Here, we utilise a microcavity quantum battery as an experimental platform that superextensively captures light energy and converts it to an electric current via the incorporation of charge transport layers into the resonant microcavity. This architecture enables, for the first time, a complete quantum battery charge-discharge cycle. We demonstrate that strong light-matter coupling induced by the microcavity leads to superextensive scaling of the steady-state electrical discharging power under low-intensity, incoherent illumination. Our results provide the first experimental demonstration of superextensive light-to-charge conversion in steady-state, highlighting the feasibility of leveraging strong light-matter coupling for enhanced energy harvesting under low-light conditions.

Introduction

The deployment of quantum technologies, with unique and often counterintuitive advantages over their classical counterparts, presents an exciting prospect for both commercial applications and the advancement of foundational quantum physics^{1–3}. While quantum computing has historically garnered the most attention, other prospective quantum technologies are now emerging with distinct quantum advantages, such as quantum sensors^{4–6}, quantum simulators^{7,8} and quantum batteries (QBs)^{9–11}. In each embodiment, the quantum advantage stems from entanglement. In quantum batteries, entanglement serves to minimise the number of traversed states during charging¹², or trigger collective effects that increase the effective coupling between the battery and its energy source¹³. Consequently, they are theorised to

exhibit exotic properties such as a charging power that scales faster than the battery capacity (i.e., super-extensively)^{9,12,14,15} as well as increased bounds on energy density¹⁶ and storage lifetime¹⁷. Although there is a large body of literature on the theory of quantum batteries^{18–36} very few experiments exist^{37–41}. These experiments investigated isolated properties of the quantum battery only, and none demonstrated a fully functioning quantum battery.

Quantum batteries are an experimental testbed for thermodynamics beyond the classical limit, where bounds on energy transfer and heat generation may be overcome by quantum correlations and quantum fluctuations⁴². Superabsorption of energy from light, the time-reversal of superradiance, is an example of faster-than-classical energy-transfer that was recently demonstrated in quantum batteries comprised of cold-atoms³⁷ as well as organic molecules in an optical cavity³⁸. In the optical microcavity, superabsorption was facilitated by molecular correlations triggered by a collective coupling to the fundamental mode of the confined electromagnetic field.

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For N molecules, this coupling scaled as $\sqrt{N}$, so that the charging time scaled inversely as $1/\sqrt{N}$; a counter-intuitive result and prohibited in classical thermodynamics. In this work, we go beyond the device design in ref. ³⁸ by incorporating a mechanism to extract stored energy from the quantum battery as an electric current.

A rich literature describes enhanced light-harvesting properties and photocurrent generation in photodiodes when embedding them inside optical microcavities^{43–47}. Several mechanisms have been proposed to explain the enhanced performance of microcavity solar cells, including more efficient light trapping⁴⁸, the mitigation of disorder⁴⁹ and longer charge carrier lifetimes⁵⁰. However, a comprehensive justification for microcavity enhancement remains an open question. Less attention has been given to the role of microcavity-induced collective effects on electrical energy extraction since these quantum correlations are expected to decay rapidly at room temperature. Nevertheless, these mechanisms, central to the concept of superabsorption, have the potential to offer new insights and advancements in microcavity solar cell research.

Here, we engineer a superabsorbing quantum battery that is wirelessly charged by either a coherent or an incoherent light source and outputs superextensive electrical power. The quantum battery is constructed with a multi-layered microcavity design (Fig. 1a) that is tuned to

the resonant frequency of the ground to first-excited singlet transition of an absorber molecule, copper phthalocyanine (CuPc), to induce a strong light–matter coupling. Using ultrafast spectroscopy, we show that the charging rate scales superextensively with the number of absorber molecules. Upon charging, the energy is rapidly transferred to a metastable triplet state in CuPc, whose population persists for six orders of magnitude longer than the charging laser pulse. Electrical extraction is facilitated by charge transport layers that introduce both an energy gradient, which favours charge separation and transport, together with a charge blocking function that suppresses undesirable recombination. For a steady-state incoherent light source, the strong light–matter coupling facilitates an electrical power output that scales superextensively with the capacity of the battery. This new phenomenon is intimately related to the strong light–matter coupling underpinning the operation of the quantum battery but may find useful applications in other photovoltaic devices such as cavity-enhanced solar cells. By integrating superextensive charging, metastable energy storage, and superextensive electrical power output within a single device, our work establishes a prototypical framework for practical quantum batteries. Moreover, we report the first experimental observation of superextensive steady-state electrical discharging power, a

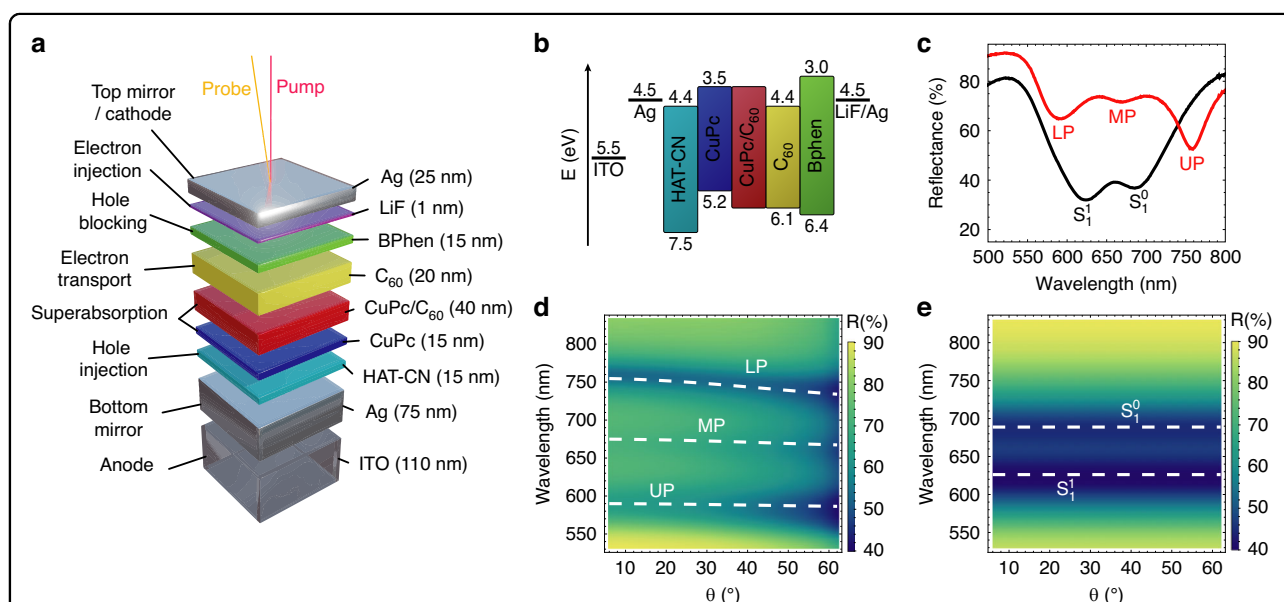


Fig. 1 Composition of the quantum battery tuned for strong light–matter coupling. **a** Schematic of the layered structure of an embodiment of the quantum battery, describing the function and composition of each component. Ultrafast pump and probe laser pulses are used to charge and measure the device. **b** Work functions and HOMO/LUMO energy levels of each layer of the quantum battery, defining an energy gradient for charge separation. **c** Steady-state reflectance spectrum at 22° incidence angle for the D5 quantum battery (red line) and the optical control device (black line), which has no top silver mirror. The control spectrum is characterised by transitions to singlet states S_1^0 and S_1^1 . In the quantum battery, these singlet state transitions are entangled with the photonic cavity mode to give rise to polaritons, as characterised by the upper (UP), middle (MP), and lower (LP) polariton states. **(d)** and **(e)** show the reflection spectra for the D5 quantum battery and its corresponding control as a function of incidence angle

previously unpredicted phenomenon with immediate relevance for low-light and always-charging energy storage systems enhanced by collective quantum effects.

Results

Device engineering

We fabricated eight quantum battery devices (labelled D1–D8) based on Fabry-Perot microcavities⁵¹ whose optical length was tuned to the absorption frequency of an ensemble of CuPc molecules. The number of CuPc absorbers N was adjusted in each device to span $N \approx 2.8 \times 10^{14}$ for the smallest battery to $N \approx 7.9 \times 10^{14}$ for the largest battery. To facilitate energy extraction from each device as an electric current, we incorporated a charge donor-acceptor combination of CuPc and fullerene (C_{60})^{52–54}, as well as a suitable choice of optically inert electron and hole blocking and transport materials that were integrated into the cavity⁵⁵. The schematic of the device (Fig. 1a) indicates the direction of laser irradiation (the detailed compositions of each device D1–D8 are shown in Fig. S1). For each device, we fabricated two otherwise identical structures without either the top or the bottom silver mirrors, to act as optical and electrical controls, respectively (Fig. S2). To minimise parameter variability, the controls were fabricated on the same substrate and fabrication run as the corresponding cavity devices D1–D8 (see “Materials and methods” section). Figure 1b schematically depicts the work functions and HOMO/LUMO energy levels for each layer of the cavity device. The layer composition induces an energy gradient within the cavity that facilitates charge separation of excitons created by superabsorption in the CuPc layer. Specifically, photoexcited singlet excitons (or polaritons) rapidly relax into triplet excitons, which diffuse toward the C_{60} interface. At the interface, interfacial charge transfer states can form before eventual separation into free charges⁵³. The hole and electron blocking layers present a kinetic barrier for charges propagating in the backwards direction, such that a photocurrent is generated and passed between the top silver cathode and the ITO anode of the device. The layer composition was chosen to maximise the electrical performance of the quantum battery without drastically detuning the fundamental frequency of the cavity from the CuPc Q-band.

Each CuPc molecule is a three-level system with the first-excited singlet S_1 separated from the ground state S_0 by ~ 2 eV and a lower-lying triplet state T_1 at approximately 1.2 eV above S_0 ⁵⁶. A Davydov splitting of the excited singlet arises from multipolar interactions between neighbouring chromophores, common to most phthalocyanine thin films⁵⁷. This feature is prominent in the reflection measurements of the optical no-cavity devices (black curve in Fig. 1c) and manifests as two weakly-split signals we label as S_1^0 and S_1^1 with

approximately equal oscillator strengths. In the presence of a cavity, these absorption bands are strongly hybridised with the fundamental mode of the confined photon field leading to the formation of upper (UP), middle (MP), and lower (LP) polariton states (red curve in Fig. 1c). By systematically varying the number of CuPc molecules in the mixed CuPc: C_{60} layer while keeping the cavity length fixed, we were able to tune the Rabi splitting between the polariton branches as a function of N . We estimate the value of N via stoichiometric analysis (Eq. S3) of the layer thicknesses, which are accurate to within ~ 1 nm. As a consequence of adjusting the CuPc concentration and layer thicknesses, the fundamental cavity frequency did not remain constant across the devices, leading to shifting asymmetries in the UP and LP reflectance bands (Fig. S3). Nevertheless, our theoretical model captured the optical response of each device very well (Fig. S5), from which it was possible to extract critical parameters of the device, such as the cavity detuning and the collective light–matter coupling. In addition, we analysed the spectral composition of the polaritonic branches of each device in Fig. S6 as a function of cavity detuning.

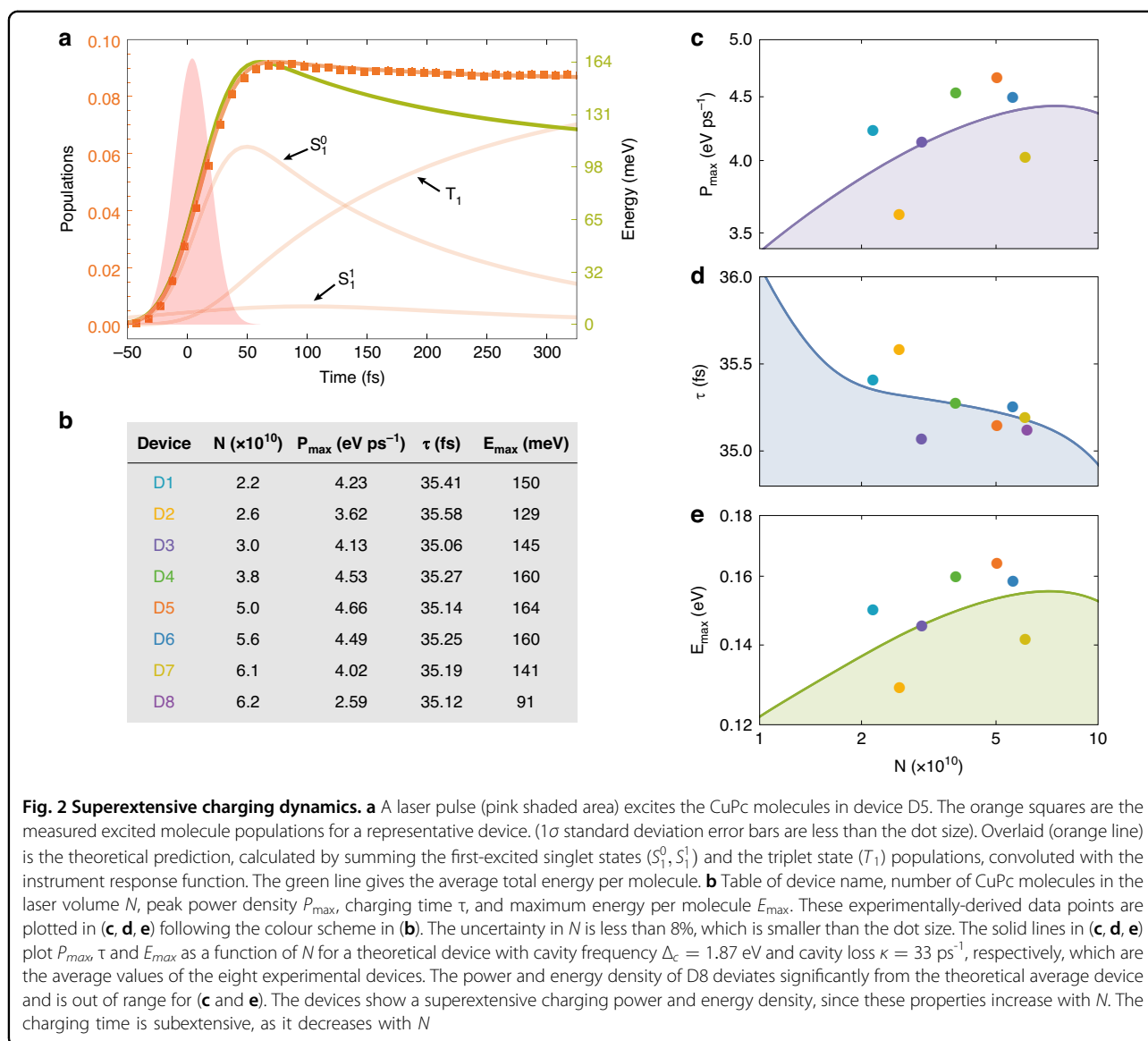
The reflectance spectrum as a function of incidence angle for the quantum battery is shown in Fig. 1d. Overlaid are the theoretically predicted three polaritonic states. In comparison, Fig. 1e shows the reflectance spectrum of the no-cavity control, which is overlaid with two Davydov-split electronic states. The molecular response to an incoherent light field does not show a dependence on the angle of incidence of the light field when it is not coupled to the cavity. That the molecule-cavity coupled response (Fig. 1d) is weakly angle dependent is thus a signature of exciton-photon hybridisation and further evidence of exciton-polaritons.

Superextensive charging with photons

In this section, we demonstrate the superextensive charging properties of our quantum battery devices. To reveal their dynamics, we utilise ultrafast transient spectroscopy, a technique that probes electronic and vibrational evolution in molecules and semiconductors on femtosecond time scales⁵⁸. In our experiments, a laser pump pulse with bandwidth ~ 31 nm and resonant with the LP branch induces excitations in the quantum battery, whose time-evolution is monitored by a delayed probe pulse resonant with the UP branch (see Figs. S7–S11). Using this scheme, we are able to monitor the temporal dynamics of the excited-state populations (see “Materials and methods” section). When comparing the ultrafast dynamics across different devices, we maintained a constant ratio between incident photons in the laser pulse and absorber molecules in the laser volume.

The excited-state population of a representative quantum battery device is shown in Fig. 2a (orange squares),





where each data point is an average of 25,000 measurements. The ultrafast differential reflectance signals for all other devices are reported in Fig. S12. Superimposed on these experimental data points is the theoretical total excited population of CuPc, which is calculated by summing the population of the first-excited singlet states (S_1^0, S_1^1) and the triplet state (T_1) (see “Materials and methods” section). Our ultrafast model utilised the cavity frequencies and collective couplings for each device that were derived from the steady-state reflectance experiments, thereby ensuring consistency in the theoretical description of both sets of experiments and in the derivation of the excited-state population dynamics. There is good agreement between the experimental data and the theoretically predicted populations. Using the known energy values of the singlet and triplet states⁵³, we

calculate the total energy of the system from their excited populations (green line in Fig. 2a).

For each device, Fig. 2b summarises the estimated number of CuPc absorbers N in the pump laser cross section, the maximum stored energy per absorber ($E_{\max}$), the time to reach half maximum energy beginning from $1/e^2$ of the Gaussian laser pulse (τ), and the peak charging power density ($P_{\max} = E_{\max}/\tau$). These values are plotted in Fig. 2c–e. Changes in the device composition as N was varied unavoidably led to slight differences in the cavity resonances and Q factors, but do not affect the overall interpretation of our results. To address whether the superextensive trends stemmed from non-linearities in the cavity resonances and Q factors rather than the collective coupling, we also plotted (solid lines in Fig. 2c–e) the theoretical predictions for hypothetical devices with a



fixed cavity resonance ($\Delta_c = 1.87$ eV) and fixed photon leakage rate ($\kappa = 33$ ps⁻¹) obtained from averaging these parameters over the eight devices. Since the predictions for the hypothetical devices still show a superextensive scaling as a function of N , we can conclude that nonlinearities in the cavity resonances and Q factors do not dominantly contribute to the superextensive scaling observed in devices D1–D8. Due to these variances, the experimental data points deviate from the plot of a theoretical device with averaged cavity properties. In particular, D8 deviates the most from these average values, since this device exhibits the largest detuning from the Davydov resonances, explaining its relatively low $E_{\max}$ and $P_{\max}$ (which fall out of the range of the Fig. 2c–e plots). Nevertheless, a superextensive increase in the charging power and energy density is seen, accompanied by a subextensive charging time. At this point, recall that a property such as energy density, which is classified as an intensive property according to classical thermodynamics, would remain constant with increasing N in conventional energy storage devices provided that the device composition remains fixed.

Stabilisation of stored energy

In this section, we demonstrate the metastability of the stored energy in our quantum battery devices. We measure their long-time decay dynamics using a spectrally broad supercontinuum probe with a spectral width spanning 500–1000 nm. The transient reflectance spectra for all devices are reported in Fig. S12, with a representative spectrum of D5 given in Fig. 3a. The positive (red) signals in the transient reflectance spectra result from ground-state bleaching, and the negative (blue) differential reflectance from excited-state absorption. Each cavity device exhibits differential reflectance signals that persist for tens of nanoseconds, showing that the excited state remains populated for six orders of magnitude longer than the device charging time (Figs. S13 and S14). These long excited-state lifetimes originate from intramolecular relaxation of the singlet excitations to metastable triplet excitations, as shown in Fig. 3b. At approximately 200 fs, intersystem crossing (ISC) becomes the dominant singlet relaxation mechanism, forming metastable triplet excitations with typical lifetimes on the order of 10–50 ns. This is three orders of magnitude longer than the equivalent state-of-the-art in microcavity quantum batteries operating at room temperature³⁸. Notably, the charge retention time measured here is significantly shorter than NMR-based quantum batteries, which also operate at room temperature and have been shown to remain charged for up to two minutes⁴¹. The efficiency of this conversion mechanism is enhanced by spin-orbit coupling of the Cu²⁺ central metal ion that works to invert the spin of the excited phthalocyanine

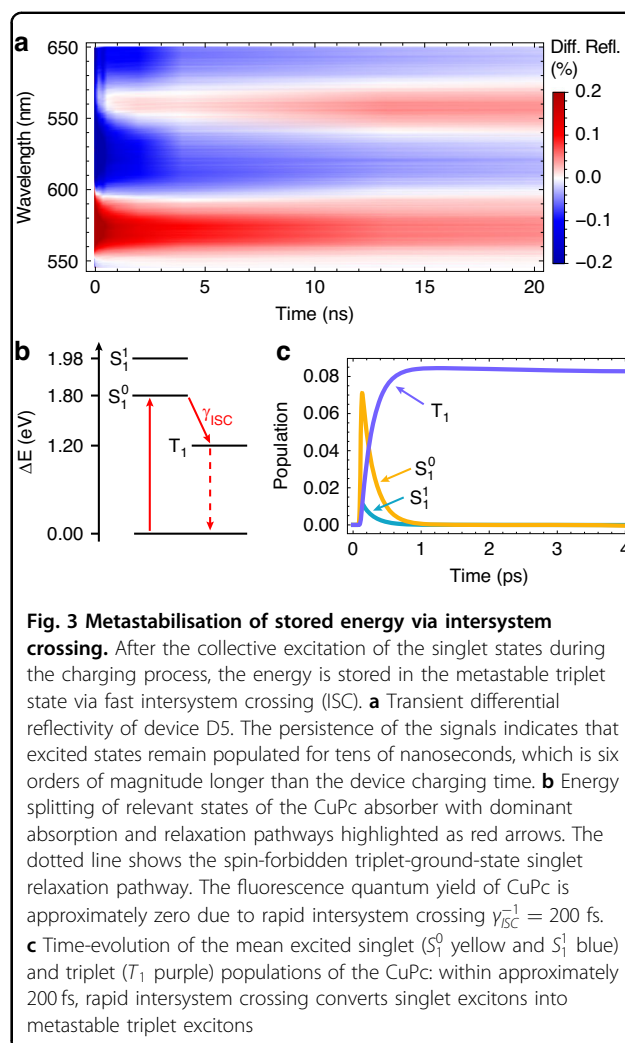


Fig. 3 Metastabilisation of stored energy via intersystem crossing.

After the collective excitation of the singlet states during the charging process, the energy is stored in the metastable triplet state via fast intersystem crossing (ISC). **a** Transient differential reflectivity of device D5. The persistence of the signals indicates that excited states remain populated for tens of nanoseconds, which is six orders of magnitude longer than the device charging time. **b** Energy splitting of relevant states of the CuPc absorber with dominant absorption and relaxation pathways highlighted as red arrows. The dotted line shows the spin-forbidden triplet-ground-state singlet relaxation pathway. The fluorescence quantum yield of CuPc is approximately zero due to rapid intersystem crossing $\gamma_{ISC}^{-1} = 200$ fs. **c** Time-evolution of the mean excited singlet (S_1^0 yellow and S_1^1 blue) and triplet (T_1 purple) populations of the CuPc: within approximately 200 fs, rapid intersystem crossing converts singlet excitons into metastable triplet excitons

electron⁵⁹. Thus, the system is blocked from relaxing from the triplet T_1 to the singlet ground state S_0 by Pauli exclusion, resulting in energy metastabilisation in the quantum battery. In Fig. 3c, we plot the long-time evolution of the excited-state CuPc populations.

Superextensive discharging of electrical power

Controlled extraction of stored energy is a necessary feature of batteries. To power a small electronic device, for example, it is desirable for the extracted energy to be available as electrical work. Here we demonstrate the enhanced electrical energy output of our quantum batteries. In Fig. 4a, b, we plot the ratio of ejected electrons to incident photons, or external quantum efficiency (EQE), for the cavity and no-cavity controls. A comparison of the two figures shows that the cavity devices have a threefold enhancement over the no-cavity controls. The polariton branches are also visible in the EQE measurements of the cavity. Despite the modest quantum efficiency enhancement observed so far, the cavity quantum battery design



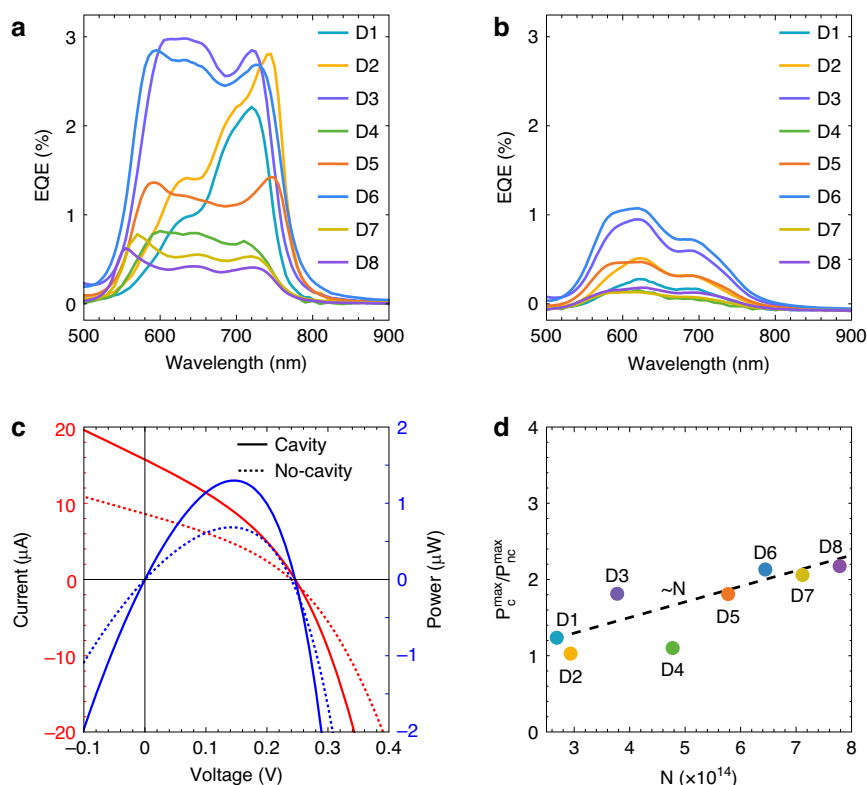


Fig. 4 Energy extraction from the quantum battery as electrical work. A comparison of the external quantum efficiency of the (a) quantum batteries and their (b) control devices shows a threefold photon-to-charge conversion enhancement in the former. **c** Current (red lines) and discharging power (blue lines) shown as a function of voltage for a representative device D5 (solid lines) and its control (dotted lines) under constant illumination at 625 nm. **d** Plots the ratio of the peak discharging power of the quantum batteries and their control devices under constant illumination at 625 nm. This ratio increases with N , indicating that the discharge power of the quantum battery scales superextensively

opens a promising avenue toward significantly improved photon-to-charge conversion, complementing and extending conventional materials-optimisation strategies.

In addition to the EQE, we investigated the discharging power of the devices (Fig. S16). Figure 4c plots current-voltage (I - V) curves for the cavity and no-cavity control of a representative device D5 (red lines), shown with their steady-state discharging power (blue lines). The optimal operating point for energy extraction is at the peak of the discharge power curve. Our devices displayed reasonable maximal discharging power densities between ~ 10 and $40 \mu\text{Wcm}^{-2}$ when compared to high-performance micro-supercapacitors⁶⁰, reported to show power densities of 30.2 and $176.5 \mu\text{Wcm}^{-2}$. We investigated the ratio of cavity to no-cavity peak discharging power, $P_c^{\max}/P_{nc}^{\max}$; as the cavity and no-cavity controls were fabricated on the same substrate, this ratio accounts for variations between fabrication runs and for the change in carrier dynamics of CuPc due to differing concentrations of C_{60} across the devices. In Fig. 4d, we observe a linear increase in $P_c^{\max}/P_{nc}^{\max}$ with N , indicating superextensive discharging power in the cavity quantum battery devices. In comparison, we point out that a constant discharging power ratio

greater than one and independent of N would indicate a non-superextensive enhancement. This superextensive discharging power has not previously been reported and suggests a collective enhancement stemming from the strong light-matter coupling in the quantum batteries.

The microscopic origin of superextensive electrical discharging arises from the polaritonic dressing of transitions to the excited singlet states of CuPc induced by the microcavity. As the energy of the dressed state scales with the number of absorbers as $\sqrt{N}$, the open-circuit voltage also grows according to $\sqrt{N}$ (see Fig. S17). This allows the photocurrent to persist to higher applied voltages, resulting in a maximum discharging power that scales as $P_c^{\max} \sim N^2$. Using a novel quantum battery model that accounts for charge transfer between redox states of the chromophores (see Supplementary Section 4), we identified a crossover between linear and superlinear regimes of electrical discharging power that occurs precisely when the quantum battery transitions into the strong-coupling regime (Fig. S19). Techniques such as multidimensional coherent spectroscopy⁶¹ and ultrafast photocurrent measurements⁶² may offer further insights into the microscopic mechanisms driving this enhancement.



Discussion

Our results reveal that strong light–matter coupling not only facilitates superextensive charging dynamics in microcavity quantum batteries but also enables super-extensive electrical discharging in the steady-state—a regime typically unexplored for quantum enhancements based on collective effects. This behaviour emerges from the interplay between rapid superabsorption, moderated by dephasing mechanisms that suppress superradiant decay, and the efficient conversion of excitons into free charge carriers via intersystem crossing. Crucially, this cascade supports a steady-state light-to-charge conversion process whose power output scales super-linearly with system size, even under low-intensity, incoherent illumination. This constitutes the first experimental realisation of steady-state superextensive electrical power, marking a significant step toward practical quantum-enhanced energy harvesting.

We have studied the operation of our devices at room temperature under ambient conditions as the most applicable regime for future quantum battery technologies. Temperature-dependent electrical discharge studies offer a further avenue to probe the influence of thermal noise on the superextensive light-to-charge conversion mechanism. The role of hybrid charge-phonon polaron states in charge transport may also be further elucidated by temperature-dependent discharging experiments in which the contributions from lattice vibrations may be quenched at low temperatures. Future studies could employ time-resolved XPS measurements as a direct probe of the charge separation in CuPc:C₆₀ complexes⁶³. Additionally, this technique may be exploited to study superextensive photocurrents that stem from polaron-polariton quantum states⁶⁴.

Until now, the study of quantum batteries has been predominantly a theoretical endeavour with scarce experimental verification. As the first experimental demonstration of a full operational cycle of a quantum battery (from superabsorption and energy metastabilisation of light energy to superextensive energy extraction as an electric current), our device represents a decisive step forward in the development of quantum battery technologies and offers an intriguing platform for the exploration of foundational photophysics at the frontier of ultrafast light-to-charge conversion and quantum thermodynamics.

Materials and methods

Device fabrication

ITO-coated glass substrates (Asahi Glass Co. Ltd., Japan) were cleaned with isopropyl alcohol and deionised water in an ultrasonic bath. The substrates were then subjected to a UV–ozone treatment in a UV–ozone cleaner (Novascan PSD Pro) to obtain an oxygen-rich ITO surface, increasing the work function of ITO. A 75 nm

thick Ag (Sigma-Aldrich) bottom mirror layer was then deposited on the ITO layer, followed by a 15 nm hole-injection HAT-CN (Luminescence Technology Co.) layer under high vacuum. From then on, all layers up to the Ag top mirror layer were deposited using a thermal evaporation method under high vacuum ($<1.0 \times 10^{-4}$ Pa). Next, a 15 nm CuPc layer, a mixed layer of CuPc:C₆₀ (Luminescence Technology Co.), and a C₆₀ layer were sequentially deposited (the thicknesses of these layers varied for each device). A 15 nm BPhen (Luminescence Technology Co.) and a 1 nm LiF (Sigma-Aldrich) layer were then deposited as electron transport and injection layers, respectively. Finally, a 25 nm Ag (Sigma-Aldrich Solutions) top mirror layer was deposited on the LiF layer. The devices were encapsulated with cover glass and a desiccant, and then sealed with a UV-curable epoxy resin. The final device is $\sim 0.5 \times 2$ cm² and has four separated 2×5 mm² regions of cavity, and two 2×5 mm² regions of the corresponding no-cavity controls. Further fabrication details can be found in the Supplementary Information.

Steady-state spectroscopy

Angle-resolved reflectometry measurements were performed with an Agilent Cary 7000 UV-Visible-NIR spectrophotometer with Universal Measurement Accessory (UMA) and xenon lamp source. Measurements across the devices were baseline-corrected and performed with standardised spot size and lamp intensity. Fixed-angle reflectometry and transmission measurements were performed with an Agilent Cary 5000 UV-Visible-NIR spectrophotometer with Diffuse Reflectance Accessory (DRA) and xenon lamp source. Measurements across the devices were baseline-corrected and performed with standardised spot size and lamp intensity. Reflectance measurements were collected at 8° angle of incidence to enable the collection of a specular and diffuse reflectance signal.

Ultrafast spectroscopy

Femtosecond transient reflection spectroscopy was used to probe the depletion of a common ground state between the LP and UP. We used two-colour pump-probe spectroscopy to circumvent pump scatter, which can obscure degenerate pump-probe measurements. The pump-probe measurement produces a differential reflectivity

$$\frac{\Delta R}{R} = \frac{R_{ON} - R_{OFF}}{R_{OFF}}$$

where R_{ON} (R_{OFF}) is the probe reflectivity with (without) a pump pulse. Since an increase in probe reflectivity in response to the pump is associated with the quantum system absorbing energy (i.e., by inducing a resonant transition $i \rightarrow f$ in the quantum battery), $\Delta R/R$ is proportional to the population P_i of the initial state i ³⁸. A



probe wavelength resonant with the ground to upper polariton transition was used, such that an increase in differential reflectivity provides a time-resolved measure of the ground-state population or, conversely, of the sum of all excited-state populations. We pump and probe at the lower and upper polaritons, respectively, specifically not to trigger relaxation dynamics between the polariton branches during the time-evolution of the experiment.

Both two-colour pump-probe and pump-supercontinuum-probe measurements were performed on the quantum battery devices in reflection geometry, at room temperature, in air. The two-colour femtosecond laser pulses were generated by two NOPAs (Light Conversion, Orpheus-N-2H and Orpheus-N-3H). The NOPAs were pumped by a Yb:KGW laser amplifier (Light Conversion Pharos, 1030 nm, 180 fs, 100 μ J pulses). A secondary identical amplifier is seeded by the oscillator of the first amplifier (Pharos Duo, Light Conversion). The output of the secondary amplifier was focused onto a 2 mm sapphire crystal to generate a broadband supercontinuum which spanned 500–1000 nm. Amplifier pulses were selected at a repetition rate of 33.3 kHz for all measurements. The two-colour pump-probe was used to measure the sub-100 fs dynamics, while the pump-supercontinuum-probe was used to measure spectrally broad long-time dynamics.

We determined and optimised the pulse durations using auto- and cross-correlation measurements (Figs. S6 and S7). The pump pulses from the 2H-NOPA had an average pulse width (Gaussian FWHM) of 34 ± 1 fs. The probe pulses from the 3H-NOPA had an average pulse width of 32 ± 3 fs across all measurements. We used the measured cross-correlation widths (~ 50 fs) for the instrument response function in the theoretical model. The pump and probe beams were focused onto the QB devices with an almost collinear geometry ($\sim 4^\circ$ between beam paths) with a FWHM spot diameter of 32 μ m for the pump, and 42 μ m for the probe. We have checked that the relative sizing between the pump and probe does not change the dynamics of the $\Delta R/R$ signal. The QB devices were rotated 25° to the incoming pump and probe pulses so the reflected probe beam could be spatially isolated and re-collimated. The probe was then directed into a spectrometer with a high-speed CCD (Entwicklungsbuero Stresing, FLC3030) triggered by the laser amplifier. To measure the $\Delta R/R$ signal, the pump was modulated by a mechanical chopper operating at 16.67 kHz (SciTec Instruments, 310CD). To scan the $\Delta R/R$ signal as a function of time, the probe beam was sent through optical retroreflector delay lines, which extended to 8 ns for the supercontinuum probe pulses (Newport, DL325) or to 0.8 ns for the NOPA probe pulses (Newport, ESP300). Longer supercontinuum probe delay times out to ~ 16 μ s were achieved by electronically triggering the secondary amplifier from later oscillator pulses (period of 13.3 ns) relative to the first amplifier. After each

pump pulse is delivered to the devices, they show a $\Delta R/R$ signal out to ~ 10 ns (Fig. S10), and the next pump pulse arrives ~ 66 μ s later.

Since the chopper modulates at half the repetition rate of the pump laser, sequential probe-only and pump-probe pulses are incident on the devices, thus enabling the acquisition of shot-to-shot $\Delta R/R$. The two-colour pump-probe measurements were scanned over the probe delay of -5 to 10 ps, with -200 to 400 fs in steps of 10 fs. At each probe delay time, 5000 shot-to-shot $\Delta R/R$ spectra are averaged. Measurements across the entire probe delay were repeated 5 times under identical experimental conditions, and the average of these measurements was taken, with the 1σ standard deviation as the error bars.

To compare the ultrafast dynamics across different devices, it was important to keep a constant ratio of incident photons to absorber molecules in the laser volume. To achieve this, the fluence and wavelength of the pump-probe pulses were varied to account for the steady-state reflection spectrum of each device (see Supplementary Information). The fluence of the pump as a function of $\Delta R/R$ intensity followed a linear dependence for all fluences used in the experiments (see Supplementary Information), ensuring higher-order effects induced by the pump are negligible in the measurements.

External quantum efficiency measurements

External quantum efficiency of the devices was calculated as the ratio of the number of photons absorbed by the device to the number of photons that participate in generating a photocurrent. This is given by $\hbar\omega I/eP$, where I is the photocurrent, $\hbar\omega$ is the photon energy, P is the power of incident photons, and e is the electron charge.

External quantum efficiency spectroscopy was performed at the *RMIT Nanostructures Laboratory* using a PV Measurements QEXL quantum efficiency instrument with Xenon arc lamp and monochromator, calibrated with a Si photodiode at 300–1100 nm. Internal quantum efficiency was calculated by dividing the experimentally measured external quantum efficiency by the absorption at each wavelength, obtained by fixed-angle UV-Visible-NIR reflectance and transmission spectroscopy on an Agilent Cary 5000 UV-Vis-NIR spectrophotometer with DRA and xenon arc lamp.

Discharging power measurements

I-V measurements were collected at the *RMIT Nanostructures Laboratory* using a Keithley 2636B SYSTEM SourceMeter® and a collimated 625 nm LED source (ThorLabs M625L4) at 10 mW/cm², with a resolution of 0.01 V. Measurements proceeded by first illuminating the device with the LED source, then varying the bias voltage from 1.5 V to -1.5 V. The power of each device was



calculated at the point where the product of current and voltage is at a maximum, i.e., the maximum power point, according to $P = IV$.

Theoretical model

The reflectance spectra were modelled using a coupled oscillator Hamiltonian

$$H_{CO} = \Delta_c a^\dagger a + \Delta_1 X_{11} + \Delta_2 X_{22} + g_{co}(a^\dagger X_{01} + X_{10}a + a^\dagger X_{02} + X_{20}a) \quad (1)$$

where $a^{(\dagger)}$ annihilates (creates) excitations of the confined photon field with frequency Δ_c and $X_{\alpha\beta} = |\alpha\rangle\langle\beta|$ are Hilbert operators that work on the low-lying electronic levels of the CuPc absorber. The indices $\alpha, \beta \in \{0, 1, 2\}$ enumerate the ground S_0 , first-excited S_1^0 and second-excited S_1^1 levels, respectively, with energies 0, Δ_1 and Δ_2 as in Fig. 3b. Since we do not probe the steady-state reflectance of our devices in the near infrared, the triplet state dynamics do not contribute to the simulated reflectance, thus we do not consider their population in our coupled oscillator model. The collective light–matter coupling constant g_{co} quantifies the interaction of the CuPc ensemble electric dipole with the quantised cavity radiation field and is related to the bare coupling g via $g_{co} = \sqrt{N}g$. The steady-state reflectance of the cavity was simulated within a Fermi golden rule approach

$$R \approx 1 - A = 1 - I_0 \sum_{\mu} \left| \langle \phi_{\mu} | a^\dagger | \phi_0 \rangle \right|^2 e^{-(\epsilon_{\mu} - \epsilon_0 - \nu)^2 / 2\sigma} \quad (2)$$

involving the coupled oscillator eigenstates $|\phi_{\mu}\rangle$ with corresponding energies ϵ_{μ} that depend upon the microscopic parameters of the Hamiltonian (1). The intensity I_0 and combined homogeneous and inhomogeneous broadening σ are fitting parameters, and ν is the frequency of incident photons external to the quantum battery.

The pertinent dynamics of the ultrafast charging experiments are captured by the Tavis–Cummings Hamiltonian

$$H(t) = (\Delta_c - \nu)a^\dagger a + i\eta(t)(a^\dagger - a) + \sum_{j=1}^N \left((\Delta_1 - \nu)X_{11}^{(j)} + (\Delta_2 - \nu)X_{22}^{(j)} + \Delta_T X_{TT}^{(j)} \right) + g \sum_{j=1}^N \left(a^\dagger X_{01}^{(j)} + X_{10}^{(j)}a + a^\dagger X_{02}^{(j)} + X_{20}^{(j)}a \right) \quad (3)$$

written in the rotating frame of the laser. We assume a Gaussian temporal profile $\eta(t) = \exp[-((t - t_0)/2\sigma)^2]$

$/\sigma\sqrt{2\pi}$ for the pulse envelope and FWHM of 34 fs. The dissipative interactions between the cavity quantum battery and its environment are accounted for with the Lindblad master equation

$$\dot{\rho} = -i[H(t), \rho] + k\mathcal{L}[a] + \sum_{j=1}^N \left(\gamma^- \left(\mathcal{L}[X_{01}^{(j)}] + \mathcal{L}[X_{02}^{(j)}] \right) + \gamma_T^- \mathcal{L}[X_{0T}^{(j)}] + \gamma^z \left(\mathcal{L}[X_{11}^{(j)}] + \mathcal{L}[X_{22}^{(j)}] \right) + \gamma_{ISC} \left(\mathcal{L}[X_{T1}^{(j)}] + \mathcal{L}[X_{T2}^{(j)}] \right) \right) \quad (4)$$

where $\mathcal{L}[O] = O\rho O^\dagger - \frac{1}{2}\{O^\dagger O, \rho\}$ are Lindblad jump operators. In Eq. (4), we account for the rate of photon loss from the cavity κ , the rate of non-radiative relaxation from the singlet γ^- and triplet γ_T^- states, intersystem crossing γ_{ISC} and singlet dephasing γ^z .

The ultrafast transient reflectance experiments provide a measure of the summed excited-state CuPc populations $\Delta R/R \propto \langle X_{TT} \rangle + \langle X_{11} \rangle + \langle X_{22} \rangle$. The populations $\langle X_{\alpha\alpha} \rangle$ for a molecule in the ensemble are obtained by numerically solving a hierarchical set of coupled equations of motion for the molecular and cavity degrees of freedom, which, in general, do not close. We enforce the closure of these equations by approximating the time-evolution of three-body correlators as $\langle ABC \rangle = \langle AB \rangle \langle C \rangle + \langle AC \rangle \langle B \rangle + \langle A \rangle \langle BC \rangle - 2\langle A \rangle \langle B \rangle \langle C \rangle$, thus capturing the pertinent dynamics with corrections scaling as $O(1/N)$. By solving for the populations $\langle X_{\alpha\alpha} \rangle$, we obtain the time-evolution of the quantum battery energy density $E(t) = (\Delta_1 \langle X_{11} \rangle + \Delta_2 \langle X_{22} \rangle + \Delta_T \langle X_{TT} \rangle)$, the charging time τ (defined by $E(\tau) = E_{\max}/2$) and the maximum charging power $P_{\max} = E_{\max}/\tau$, where $E_{\max}$ is the maximum energy density stored in the device.

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Author contributions

J.Q.Q. conceived and managed the project. K.H. developed the theoretical model and theoretical analysis. D.T., J.v.E., and D.G. performed and interpreted steady-state reflectance and electrical measurements on the devices. J.B.M. and T.A.S. performed the ultrafast measurements. T.H. and C.J.D. fabricated the devices. All authors contributed to the discussion of the results and writing the manuscript.

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Data availability

The data that support the findings of this study are available from the corresponding author on request.

Conflict of interest

The authors declare no competing interests.

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