Vacuum Bloch–Siegert shift in Landau polaritons with ultra-high cooperativity

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Vacuum Bloch-Siegert Shift in Landau Polaritons with Ultra-high Cooperativity

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A two-level system resonantly interacting with an ac magnetic or electric field constitutes the physical basis of diverse phenomena and technologies. However, Schrödinger's equation for this seemingly simple system can be solved exactly only under the rotating wave approximation, which neglects the counter-rotating field component. When the ac field is sufficiently strong, this approximation fails, leading to a resonance-frequency shift known as the Bloch-Siegert (BS) shift. Here, we report the vacuum BS shift, which is induced by the ultrastrong coupling of matter with the counter-rotating component of the vacuum fluctuation field in a cavity. Specifically, an ultra-high-mobility 2D electron gas inside a high-Q terahertz cavity in a quantising magnetic field revealed ultra-narrow Landau polaritons, which exhibited a vacuum BS shift up to $40\,\mathrm{GHz}$. This shift, clearly distinguishable from the photon-field self-interaction effect, represents a unique manifestation of a strong-field phenomenon without a strong field.

An electron spin in a static magnetic field naturally precesses in a right-hand circular motion about the field axis at the Larmor frequency. For an incoming frequency-matching ac magnetic field, the left-hand circularly polarised (LCP) component, which rotates in the opposite direction to the spin motion, goes into and out of phase with the spin twice every oscillation period. Hence, the cumulative effect of the LCP component of the ac field is negligible compared to the co-rotating right-hand circularly polarised (RCP) component. The approach to keep only the latter component is called the rotating wave approximation (RWA)[1], which significantly simplifies the problem of calculating field-spin interactions. After the Bloch equations were generalized from the magnetic resonance context to the realm of optical resonance in two-level systems[2], the RWA has been ubiquitously applied to various light-matter coupling problems so successfully that, in many cases, it is implicitly made without any justification.

When the applied ac field becomes sufficiently strong, the RWA breaks down, which results in a shift in the resonance frequency known as the Bloch-Siegert (BS) shift[3], stemming from interaction with the counter-rotating ac field component. The lowest-order term in the BS shift is on the order of Ω^2/ω_0, where Ω is the Rabi frequency and ω_0 is the unperturbed resonance frequency[1]. Higher-order terms of the BS shift have also been calculated quantum mechanically[4,5], which have to be taken into account in order to correctly estimate the magnetic moment values from magnetic resonance experiments[6]. Furthermore, recent studies on circuit quantum electrodynamics (QED) systems[7,8] have revealed frequency shifts that are equivalent to the BS shift in the case of cavity QED. More recently, a BS shift has been reported for a transition metal dichalcogenide system driven by circularly polarised sub-bandgap optical fields[9].

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Here, we report a vacuum BS shift in a solid-state cavity QED system, which occurs when the average photon number inside the cavity is much less than one. The shift in this case is caused by the ultra-strong coupling of matter with the counter-rotating component of the vacuum fluctuation field inside the cavity. Such a BS shift was predicted for atomic cavity QED systems[10], but an observation has been elusive due to the small dipole moments of atomic transitions. Excitations in solids are better candidates for observing a vacuum BS shift because of the possibility of entering the ultra-strong coupling (USC) regime, which is defined by $g/\omega_{\text{cav}} > 0.1$; here, g is the light-matter coupling rate, which is equal to half of the vacuum Rabi splitting (VRS), and ω_{cav} is the cavity photon frequency. For example, $g/\omega_{\text{cav}} = 1.43$ has been achieved using cyclotron resonance (CR) in a two-dimensional electron gas (2DEG) in semiconductor quantum wells (QWs)[11]. However, no clear demonstration of a vacuum BS shift has been made due to the typically broad linewidths caused by ultra-fast decoherence and lossy cavity designs as well as the co-existence of competing features in the USC regime[12–14]. In the context of circuit QED, several recent studies have demonstrated USC[15–19], notably achieving $g/\omega_{\text{cav}} = 1.34$ in one study[18]. A 'quantum' BS shift, similar to the vacuum BS shift, has also been reported for a circuit QED system[15]; however, the evidence presented was based on spectral fitting with a simplified Hamiltonian that was not derived from first principles.

In our experiments, electron CR in a Landau-quantised high-mobility 2DEG in a GaAs QW coupled ultra-strongly with vacuum photons in a high-quality-factor terahertz (THz) photonic crystal cavity (PCC). In this Landau-polariton system, we simultaneously achieved $g/\omega_{\text{cav}} = 0.36$ and an ultrahigh cooperativity $C \equiv 4g^2/\kappa\gamma = 3513$, where κ and γ are the photon and matter decay

rates, respectively. We found that the probe light polarisation state plays a critical role in exploring USC physics in this ultra-high-cooperativity system. As shown in Fig. 1a, the resonant co-rotating coupling of electrons with CR-active (CRA) circularly polarised radiation leads to the extensively studied VRS. On the other hand, the counter-rotating coupling of electrons with the CR-inactive (CRI) mode leads to the time-reversed partner of the VRS, namely, the vacuum BS shift, as we identified for the first time. Here, the separation of the VRS and the vacuum BS shift to two time-reversed polarisation states is analogous to the physics of valley-exclusive optical Stark shift and dynamical BS shift observed in WS_2 (Ref. 9); see Supplementary Information Section 8.

Experimental scheme

Our scheme of probing the VRS and vacuum BS shift is depicted in Fig. 1b. The energy versus magnetic field (B) relations measured with RCP and LCP probes are mirror symmetric about the $B = 0$ axis. Note that a negative B means a change in polarity compared to a positive B. When we use a linearly polarised THz probe, i.e., a 50%-50% mixture of RCP and LCP radiation, in the $B > 0$ region, the RCP component shows the VRS, while the vacuum BS shift appears for the LCP component due to the state repulsion[9] between the cavity mode (black dashed line) and the CR at negative B fields (blue dashed line). When an RCP THz probe is used, one can separately observe the VRS and the vacuum BS shift in the $B > 0$ and $B < 0$ regions, respectively. We demonstrated that the vacuum BS shift exclusively appears in the CRI mode dispersion and is distinguished from the other unique signature of the USC regime, i.e., the photon-field self-interaction effect due to the A^2 (or diamagnetic) terms in the Hamiltonian[20].

In our cavity QED setup, a 2DEG membrane containing 10 GaAs QWs with a total electron density $n = 3.2 \times 10^{12}\,\text{cm}^{-2}$ was placed in a 1D THz PCC. As shown in Fig. 1c, the cavity consisted of five silicon wafers whose thicknesses from left to right were $50\,\mu\text{m}$, $50\,\mu\text{m}$, $100\,\mu\text{m}$, $50\,\mu\text{m}$, and $50\,\mu\text{m}$, respectively. Equal spacings of $195\,\mu\text{m}$ were created in between, forming a spatially alternating refractive index. Since the thickness of the central silicon layer was twice as thick as the other layers, it created defect cavity modes within the photonic band gaps that would have resulted for a perfect photonic crystal structure. Figure 1d shows an experimental transmittance spectrum for a bare cavity without a 2DEG, exhibiting two sharp cavity modes with a full-width-at-half-maximum (FWHM) of $2.2\,\text{GHz}$ and $2.4\,\text{GHz}$, respectively. For each cavity mode, the electric field maximum overlapped with the 2DEG layer to ensure maximum coupling; see Fig. 1c for the electric field amplitude distribution for the first cavity mode. We varied B to tune the cyclotron frequency of the 2DEG, $\omega_{\text{c}} = eB/m^*$, where $m^* = 0.067m_0$ is the electron effective mass; see Fig. 1e for 2DEG magneto-transmittance spectra in free space, showing the linear evolution with B of a superradiance-broadened CR line[21]. The USC of CR with cavity photons occurs when ω_{c} is tuned to coincide with a photonic mode frequency $\omega_{\text{cav}}^{n_z}$, where n_z is the mode index.

Experimental results

Figure 2a shows linearly polarised THz transmittance spectra for the cavity containing the 2DEG at various B in the first-order anti-crossing region (around ω_{cav}^1). This experiment maps out the solid lines in the positive B region of Fig. 1b. Three branches of transmission peaks are well

resolved in the spectra. Two of them are ascribed to the Landau polaritons arising from the CRA circularly polarised mode that resonantly coupled with CR in a co-rotating manner through the optical conductivity $\sigma_{\mathrm{CRA}}(\omega) = (ne^2/m^*)/[\gamma - i(\omega - \omega_c)]$, which has a peak at $\omega = \omega_c$. They are labeled 'lower polariton (LP)' and 'upper polariton (UP)', respectively, based on their frequencies. We see anti-crossing of the LP and UP peaks with a large splitting, $\omega_{\mathrm{UP}} - \omega_{\mathrm{LP}}$, at zero detuning ($\Delta = \omega_{\mathrm{cav}}^1 - \omega_c = 0$). However, the value of the coupling strength g is not exactly equal to $(\omega_{\mathrm{UP}} - \omega_{\mathrm{LP}})/2$ at $\Delta = 0$ due to the coupling of CR with other photonic modes, for example, the second-order cavity mode in the photonic band gap or transmission modes in the pass bands. We determined $g/2\pi = 150.1\,\mathrm{GHz}$ and $g/\omega_{\mathrm{cav}}^1 = 0.36$ for the first-order anti-crossing through theoretical estimation using device parameters (see a brief summary described below and more details in Supplementary Information Section 3).

The middle branch seen in Fig. 2a is the CRI circularly polarised mode, which, in free space, is not absorbed by the 2DEG, because the conductivity $\sigma_{\mathrm{CRI}}(\omega) = (ne^2/m^*)/[\gamma - i(\omega + \omega_c)]$ does not show a peak for $\omega > 0$. However, because of USC, this mode frequency ω_{CRI} red-shifts with B through counter-rotating coupling with CR. As we will show below, the shift of the CRI mode with B is the vacuum BS shift. In the limit of an infinite B, the CRI mode and the CRA-LP converge onto the true cavity mode frequency ω_{cav}^1 of the system. The asymptotic frequency is also confirmed from the transmittance peak shown in Fig. 1d for the cavity without the 2DEG. The CRI mode closes the 'polaritonic gap' that appears in the CRA dispersion (see Supplementary Information Section 4); such an effect was not observed in previous studies[22,23] because the CRI mode was not probed. The minisplitting feature observed in the CRI mode is due to back reflections

from the cryostat windows; see Supplementary Information Section 6.

As shown in Figs. 2b-2d, we determined the line-widths of the UP and LP peaks at $\Delta = 0$ and the CRI mode at $3\,\mathrm{T}$ to be $5.5\,\mathrm{GHz}$, $4.8\,\mathrm{GHz}$, and $4.8\,\mathrm{GHz}$, respectively. It was previously shown that, for systems in the USC regime, UP and LP line-widths are not necessarily equal to each other[24] or simply given by $(\kappa + \gamma)/2$. In our case, as described in Supplementary Information Section 5, we obtained $\kappa/2\pi = 4.5\,\mathrm{GHz}$ and $\gamma/2\pi = 5.7\,\mathrm{GHz}$. Here, $\kappa/2\pi$ is larger than the $2.2\,\mathrm{GHz}$ line-width determined for a bare cavity in Fig. 1d due to the loss introduced by the 2DEG. On the other hand, the intrinsic CR decay rate $\gamma/2\pi$ is 60 times smaller than the linewidth of CR for the same sample in free space (inset to Fig. 1e) due to the suppression of super-radiance[14]. The calculated cooperativity $C = 4g^2/\kappa\gamma = 3513$ is the highest reported for an intraband cavity QED system.

Figure 2e shows the transmittance spectra measured with an RCP THz probe. This experiment maps out the two red solid RCP lines in Fig. 1b in both the positive and negative B regions. The RCP THz probe beam was generated using a THz achromatic quarter wave plate[25], whose detailed characteristics are shown in Supplementary Information Section 7. In the positive B region, an RCP beam corresponds to the CRA mode, so we observed the CRA-UP and CRA-LP branches (red curves) that are identical to the CRA-UP and CRA-LP peaks in Fig. 2a. The CRI mode, which is absent in the positive B region, appears in the negative B region (blue curves) as an extension of the UP; however, the same RCP probe light now counter-rotates with the CR because the helicity of the electron CR motion changes. The vacuum BS shift is again clearly observed in the

CRI mode. Our RCP THz spectra unambiguously demonstrates that the VRS and vacuum BS shift are a time-reversed pair in the Landau polariton system, with distinct optical polarisation selection rules.

We confirmed that Figs. 2a and e yield the same result if the CRI mode in Figs. 2a is folded about the $B = 0$ axis to the negative B region. Below, we present all the dispersion data in a way that is similar to Fig. 2e, meaning that we measured the mode frequencies with a linearly polarised probe at positive B fields, but the CRI mode was folded to the negative B region to show what we obtain when the probe is RCP. We extracted the positions of the UP, LP, and CRI peaks in the first and second anti-crossing regions. The dispersions are plotted in Fig. 3, together with simulation results obtained using a semiclassical transfer-matrix method with experimental material and cavity structure parameters. See Supplementary Information Section 2 for details on the simulations. Excellent agreement between experiment and simulation is achieved.

Theoretical analysis

We developed a quantum model, derived from the first principles (i.e., physical laws), within the electric-dipole approximation. The full Hamiltonian of the interacting CR-cavity-photon system is

$$\hat{H} = \hat{H}_{\mathrm{CR}} + \hat{H}_{\mathrm{cav}} + \hat{H}_{\mathrm{int}} + \hat{H}_{A^2} \, , \tag{1}$$

where $\hat{H}_{\mathrm{CR}}$, $\hat{H}_{\mathrm{cav}}$, $\hat{H}_{\mathrm{int}}$, and $\hat{H}_{A^2}$ represent the Hamiltonian for the 2DEG CR, cavity photons, CR-cavity-photon interaction, and photon-field self-interaction, respectively. These different con-

tributions are expressed, respectively, as

$$\hat{H}_{\text{CR}} = \hbar\omega_{\text{c}}\hat{b}^{\dagger}\hat{b} \,, \tag{2a}$$

$$\hat{H}_{\text{cav}} = \sum_{n_z=1}^{\infty} \sum_{\xi=\pm} \hbar\omega_{\text{cav}}^{n_z}\hat{a}_{n_z,\xi}^{\dagger}\hat{a}_{n_z,\xi} \,, \tag{2b}$$

$$\hat{H}_{\text{int}} = \sum_{n_z=1}^{\infty} i\hbar\bar{g}_{n_z} \left[\hat{b}^{\dagger}(\hat{a}_{n_z,+} + \hat{a}_{n_z,-}^{\dagger}) - \hat{b}(\hat{a}_{n_z,-} + \hat{a}_{n_z,+}^{\dagger}) \right] \,, \tag{2c}$$

$$\hat{H}_{A^2} = \sum_{n_z=1}^{\infty} \sum_{n'_z=1}^{\infty} \frac{\hbar\bar{g}_{n_z}\bar{g}_{n'_z}}{\omega_{\text{c}}}(\hat{a}_{n_z,-} + \hat{a}_{n_z,+}^{\dagger})(\hat{a}_{n'_z,+} + \hat{a}_{n'_z,-}^{\dagger}) \,, \tag{2d}$$

where $\hat{b}^{\dagger}$ ($\hat{b}$) is the creation (annihilation) operator for the collective CR excitation of the electrons in the highest Landau level, which satisfy $[\hat{b}, \hat{b}^{\dagger}] = 1$, $\hat{a}_{n_z,\xi}^{\dagger}$ ($\hat{a}_{n_z,\xi}$) represents the creation (annihilation) operator for cavity photons of the n_z-th cavity mode, where ξ is the polarisation index, with '+' and '−' representing the CRA and CRI circularly polarised modes, respectively, $\bar{g}_{n_z} = g_{n_z}\sqrt{\omega_{\text{c}}/\omega_{\text{cav}}^{n_z}}$ is the coupling rate of CR with the n_z-th cavity mode, where $g_{n_z} = \sqrt{e^2 n/(\varepsilon_0\varepsilon_{\text{cav}}m^*L_{n_z})}$ is the coupling rate at zero detuning, L_{n_z} is the effective cavity length, and ε_{cav} is the permittivity of the cavity filling material. A detailed derivation of the Hamiltonian is given in Supplementary Information Section 3.

The vacuum BS shift results from the counter-rotating coupling of CR and the cavity vacuum field. In the full Hamiltonian, Eq. (1), counter-rotating coupling appears in $\hat{H}_{\text{int}}$ as products of the CR operators ($\hat{b}^{\dagger}$ and $\hat{b}$) with the CRI mode photon operators ($\hat{a}_{n_z,-}^{\dagger}$ and $\hat{a}_{n_z,-}$); see Eq. (2c). These products are known as the counter-rotating terms (CRTs). On the other hand, the terms in $\hat{H}_{A^2}$, given in Eq. (2d), affect both the CRA and CRI modes. When the CR-photon interaction enters the USC regime, both the CRTs and the A^2 terms play nontrivial roles in the polariton physics. We found that only by measuring the CRI mode can one distinguish the CRT contribution, which

exclusively leads to the vacuum BS shift.

By solving the equations of motion of the full Hamiltonian, taking into account only the first-order cavity mode, we found that the eigen-frequencies of the CRA and CRI modes satisfy

$$\frac{\omega_{\mathrm{cav}}^1}{\omega} = \left\{ 1 - \frac{2g_1{}^2}{\omega(\omega - \omega_{\mathrm{c}})} \right\}^{1/2}, \tag{3}$$

and

$$\frac{\omega_{\mathrm{cav}}^1}{\omega} = \left\{ 1 - \frac{2g_1{}^2}{\omega(\omega + \omega_{\mathrm{c}})} \right\}^{1/2}, \tag{4}$$

respectively. Equations (3) and (4) correspond to the dispersion relations of the system; the left-hand side is proportional to k_1/ω, where $k_1 = \omega_{\mathrm{cav}}^1 \sqrt{\varepsilon_{\mathrm{cav}}}/c$ is the confinement wavenumber, and the right-hand side is the effective refractive index. The B-dependent solutions to Eqs. (3) and (4) can qualitatively reproduce essential features of the experimental CRA and CRI mode dispersions in Fig. 2a, Fig. 2e, and Fig. 3. The positive solution to Eq. (4) shows a vacuum BS shift, $\Delta\omega_{\mathrm{BS}}$, as a function of B, whereas the solutions to Eqs. (3) and (4) in the $B \to 0$ limit give $\omega = \sqrt{(\omega_{\mathrm{cav}}^1)^2 + 2g_1{}^2}$, indicating a frequency blue-shift $\Delta\omega_{A^2}(B = 0) = \sqrt{(\omega_{\mathrm{cav}}^1)^2 + 2g_1{}^2} - \omega_{\mathrm{cav}}^1$ due to the A^2 terms. Note that $\Delta\omega_{A^2}(B = 0)$ is known as the polaritonic gap[22,23,26]. In Supplementary Information Section 3.4, we confirmed that the conclusions above are immune to photon and CR losses added to the system.

Furthermore, we simulated polariton spectra for a total of four cases, where we selectively removed the effective contributions from the CRTs and the A^2 terms from the full Hamiltonian; see Figs. 4a-d for the results plotted together with experimental data and Supplementary Information Section 3 for more details. The simulation takes into account all photonic modes that are interacting

with CR. From the perfect agreement between experiment and theory shown in Fig. 4a, deviations appear when either the CRTs or the A^2 terms are removed. By comparing Figs. 4a and b, we can confirm that the A^2 terms produce an overall blue-shift for both polariton branches and the CRI mode. On the other hand, by comparing Figs. 4a and c, we can confirm that the CRTs only affect the CRI mode, producing a vacuum BS shift as a function of B. The shift can be quantitatively evaluated by $\Delta\omega_{\mathrm{BS}}(B) = \omega_{\mathrm{CRI}}(B) - \omega_{\mathrm{CRI}}(B = 0)$, where both of the CRI mode frequencies on the right are experimentally measured.

The unique capability of switching on and off the CRTs and the A^2 terms in the simulations allows us to quantitatively determine the experimentally observed $\Delta\omega_{\mathrm{BS}}(B)$ and $\Delta\omega_{A^2}(B)$. As shown in Fig. 4e, in the CRI mode dispersion, $\Delta\omega_{\mathrm{BS}}(B)$ monotonically increases with B in magnitude and reaches $-37\,\mathrm{GHz}$ at $B = 5\,\mathrm{T}$. On the other hand, as shown in Fig. 4f, both the CRA and CRI mode dispersions blue-shift entirely due to the A^2 terms, leading to positive $\Delta\omega_{A^2}(B)$ shifts for all three branches. Note that $\Delta\omega_{A^2}(B)$ is larger for modes that are more photon-like, which is consistent with the fact that the A^2 terms represent the photon-field self-interaction effect. As the CRA-LP and CRA-UP branches exchange their relative weights between photon nature and CR nature as a function of B, their $\Delta\omega_{A^2}(B)$ shifts show a monotonic increase and decrease with B, respectively. The CRI mode is mainly photon-like, thus its $\Delta\omega_{A^2}(B)$ shift remains large, only slightly increasing with B.

Discussion

Our ability to quantitatively determine the separate contributions from $\Delta\omega_{\mathrm{BS}}$ and $\Delta\omega_{A^2}$ in our experimental data is crucial for further explorations of phenomena expected to occur in the USC regime. The CRTs and A^2 terms control the realizability of many theoretical predictions. Specifically, the CRTs are crucial for producing Schrödinger-cat states[18,27] and extra-cavity vacuum photon emissions[28]; the A^2 terms play a decisive role in preventing the occurrence of super-radiant quantum phase transitions[29–33]. Our work will thus enable quantitative evaluation of separate contributions to provide future possibilities to engineer them at will. This capability will help construct best-designed quantum systems for specific applications of the phenomenon of ultra-strong light-matter coupling.

References

1. Allen, L. & Eberly, J. H. *Optical Resonance and Two-Level Atoms* (John Wiley & Sons, 1975).

2. Feynman, R. P., Vernon, F. L. & Hellwarth, R. W. Geometrical representation of the Schrödinger equation for solving maser problems. *J. Appl. Phys.* **28**, 49–52 (1957).

3. Bloch, F. & Siegert, A. Magnetic resonance for nonrotating fields. *Phys. Rev.* **57**, 522–527 (1940).

4. Shirley, J. H. Solution of the Schrödinger equation with a Hamiltonian periodic in time. *Phys. Rev.* **138**, B979–B987 (1965).

5. Cohen-Tannoudji, C., Dupont-Roc, J. & Fabre, C. A quantum calculation of the higher order terms in the Bloch-Siegert shift. *J. Phys. B* **6**, L214 (1973).

6. Abragam, A. *Principles of Nuclear Magnetism* (Oxford University Press, 1961).

7. Tuorila, J. *et al.* Stark effect and generalized Bloch-Siegert shift in a strongly driven two-level system. *Phys. Rev. Lett.* **105**, 257003 (2010).

8. Pietikäinen, I. *et al.* Observation of the Bloch-Siegert shift in a driven quantum-to-classical transition. *Phys. Rev. B* **96**, 020501 (2017).

9. Sie, E. J. *et al.* Large, valley-exclusive Bloch-Siegert shift in monolayer WS_2. *Science* **355**, 1066–1069 (2017).

10. De Zela, F., Solano, E. & Gago, A. Micromaser without the rotating-wave approximation: The Bloch-Siegert shift and related effects. *Opt. Commun.* **142**, 106 – 118 (1997).

11. Bayer, A. *et al.* Terahertz light–matter interaction beyond unity coupling strength. *Nano Lett.* **17**, 6340–6344 (2017).

12. Anappara, A. A. *et al.* Signatures of the ultrastrong light-matter coupling regime. *Phys. Rev. B* **79**, 201303(R) (2009).

13. Muravev, V. M., Andreev, I. V., Kukushkin, I. V., Schmult, S. & Dietsche, W. Observation of hybrid plasmon-photon modes in microwave transmission of coplanar microresonators. *Phys. Rev. B* **83**, 075309 (2011).

14. Zhang, Q. *et al.* Collective non-perturbative coupling of 2D electrons with high-quality-factor terahertz cavity photons. *Nat. Phys.* **12**, 1005–1011 (2016).

15. Forn-Díaz, P. *et al.* Observation of the Bloch-Siegert shift in a qubit-oscillator system in the ultrastrong coupling regime. *Phys. Rev. Lett.* **105**, 237001 (2010).

16. Niemczyk, T. *et al.* Circuit quantum electrodynamics in the ultrastrong-coupling regime. *Nat. Phys.* **6**, 772–776 (2010).

17. Forn-Díaz, P., Romero, G., Harmans, C. J. P. M., Solano, E. & Mooij, J. E. Broken selection rule in the quantum Rabi model. *Sci. Rep.* **6**, 26720 (2016).

18. Yoshihara, F. *et al.* Superconducting qubit-oscillator circuit beyond the ultrastrong-coupling regime. *Nat. Phys.* **13**, 44–47 (2017).

19. Forn-Diaz, P. *et al.* Ultrastrong coupling of a single artificial atom to an electromagnetic continuum in the nonperturbative regime. *Nat. Phys.* **13**, 39–43 (2017).

20. Hagenmüller, D., De Liberato, S. & Ciuti, C. Ultrastrong coupling between a cavity resonator and the cyclotron transition of a two-dimensional electron gas in the case of an integer filling factor. *Phys. Rev. B* **81**, 235303 (2010).

21. Zhang, Q. *et al.* Superradiant decay of cyclotron resonance of two-dimensional electron gases. *Phys. Rev. Lett.* **113**, 047601 (2014).

22. Maissen, C. *et al.* Ultrastrong coupling in the near field of complementary split-ring resonators. *Phys. Rev. B* **90**, 205309 (2014).

23. Maissen, C., Scalari, G., Beck, M. & Faist, J. Asymmetry in polariton dispersion as function of light and matter frequencies in the ultrastrong coupling regime. *New J. Phys.* **19**, 043022 (2017).

24. Bamba, M. & Ogawa, T. System-environment coupling derived by Maxwell's boundary conditions from the weak to the ultrastrong light-matter-coupling regime. *Phys. Rev. A* **88**, 013814 (2013).

25. Kawada, Y. *et al.* Achromatic prism-type wave plate for broadband terahertz pulses. *Opt. Lett.* **39**, 2794–2797 (2014).

26. Scalari, G. *et al.* Ultrastrong coupling of the cyclotron transition of a 2D electron gas to a THz metamaterial. *Science* **335**, 1323–1326 (2012).

27. Ashhab, S. & Nori, F. Qubit-oscillator systems in the ultrastrong-coupling regime and their potential for preparing nonclassical states. *Phys. Rev. A* **81**, 042311 (2010).

28. Ciuti, C., Bastard, G. & Carusotto, I. Quantum vacuum properties of the intersubband cavity polariton field. *Phys. Rev. B* **72**, 115303 (2005).

29. Rzażewski, K., Wódkiewicz, K. & Żakowicz, W. Phase transitions, two-level atoms, and the A^2 term. *Phys. Rev. Lett.* **35**, 432–434 (1975).

30. Nataf, P. & Ciuti, C. No-go theorem for superradiant quantum phase transitions in cavity QED and counter-example in circuit QED. *Nat. Commun.* **1**, 72 (2010).

31. Bamba, M. & Ogawa, T. Stability of polarizable materials against superradiant phase transition. *Phys. Rev. A* **90**, 063825 (2014).

32. Hagenmüller, D. & Ciuti, C. Cavity QED of the graphene cyclotron transition. *Phys. Rev. Lett.* **109**, 267403 (2012).

33. Chirolli, L., Polini, M., Giovannetti, V. & MacDonald, A. H. Drude weight, cyclotron resonance, and the Dicke model of graphene cavity QED. *Phys. Rev. Lett.* **109**, 267404 (2012).

Author contributions: X.L. fabricated THz cavity devices, performed all measurements, analyzed all experimental data, and performed semiclassical simulations under the supervision and guidance of Q.Z. and J.K. M.B. performed quantum mechanical and semiclassical calculations. S.F., G.C.G., and M.J.M. grew the 2DEG sample. Q.Z., W.G., M.L., and K.Y assisted X.L. with cavity sample preparation and measurements. X.L., M.B., and J.K. wrote the manuscript. All authors discussed the results and commented on the manuscript.

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Figure captions:

Fig. 1. Cyclotron resonance (CR) cavity QED setup. a, Schematic diagram for the two circularly polarised photon fields interacting with the CR of a 2D electron gas (2DEG) in a cavity. The co-rotating CRA mode exhibits vacuum Rabi splitting phenomena, while the counter-rotating CRI mode exhibits the vacuum Bloch-Siegert shift. **b**, Schematic diagram for simultaneously probing the VRS and vacuum BS shift in a Landau polariton system. **c**, Schematic diagram for the experimental CR-cavity structure. CR couples with THz cavity photons at coupling rate g. κ and γ are the photon and matter decay rates, respectively. The black curve with a red shade shows the electric field distribution of the first cavity mode. **d**, Transmittance spectrum for the cavity without a 2DEG, showing the first and second cavity modes with FWHMs of $2.2\,$GHz and $2.4\,$GHz, respectively. **e**, Transmittance spectra for the 2DEG in free space as a function of frequency and B. Inset: transmittance spectrum at $3\,$T, showing a CR linewidth of $340\,$GHz fit by a Lorentzian function.

Fig. 2. Landau polaritons in the ultrastrong coupling regime with ultrahigh cooperativity. a, Linearly polarised THz transmittance spectra in the B region where CR couples with the first-order cavity mode. The CRA mode splits into the LP and UP branches, exhibiting anti-crossing behavior. The CRI mode redshifts with B. The cavity mode frequency without the 2DEG (ω_{cav}^1) and the CR frequency in free space (ω_c) are shown as dashed red and black lines, respectively. Lorentzian fits for **b**, the CRI mode at $3\,$T, **c**, the UP peak at $\Delta = 0$, and **d**, the

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LP peak at $\Delta = 0$. The obtained FWHM values are indicated on the graphs. **e**, RCP THz transmittance spectra in both the positive and negative B regions.

Fig. 3. Landau polariton dispersions as a function of B. Experimental polariton peak positions (open circles) in the first and second anti-crossing regions are plotted on top of a simulated transmittance color contour map, showing excellent agreement. The cavity mode frequencies and the CR frequency in free space are shown as dashed red and white lines, respectively. The CRI mode is the CRA-UP branch in the negative B regime.

Fig. 4. Distinction between the vacuum Bloch-Siegert shift due to the counter-rotating terms (CRTs) and the shift due to the A^2 terms in the ultrastrong coupling regime. Simulated spectra **a**, with both the CRTs and the A^2 terms (full Hamiltonian), **b**, with the CRTs but without the A^2 terms, **c**, without the CRTs but with the A^2 terms, and **d**, without the CRTs and A^2 terms. Each graph includes experimental peak positions as open circles. **e**, The vacuum Bloch-Siegert shift ($\Delta\omega_{\mathrm{BS}}$) due to the CRTs as a function of B for the CRI, CRA-UP, and CRA-LP peaks. **f**, The frequency shift due to the A^2 terms ($\Delta\omega_{A^2}$) as a function of B for the CRI, CRA-UP, and CRA-LP peaks.

Methods:

THz spectroscopy. We performed transmission time-domain THz magneto-spectroscopy measurements in the Faraday geometry. THz pulses were generated from a nonlinear (110) zinc telluride crystal pumped by a Ti: sapphire-based regenerative amplifier (1 kHz, 0.9 mJ, 775 nm, 200 fs, Clark-MXR, Inc., CPA-2001) via optical rectification. Another small portion of the

laser beam was split for probing THz radiation. The probe beam was delayed by a mechanical stage, and incident onto the detection zinc telluride crystal together with the transmitted THz beam for probing THz electric field via the electro-optic sampling method. In RCP THz transmittance experiments, we used an achromatic THz quarter wave plate to convert the linearly polarised THz beam into a circularly polarised beam.

In THz time-domain spectroscopy, the THz electric field measured as a function of time can provide both amplitude and phase information. Here, for obtaining cavity transmittance spectra, we first measured a THz wave passing through free space as a reference, then measured under the same conditions, the cavity sample. Power transmittance is defined as the square of the ratio between Fourier-transformed THz signal intensity spectra of the cavity sample and the reference $T = |E_{\text{cavity}}(\omega)/E_{\text{reference}}(\omega)|^2$.

Cavity sample fabrication. In order to integrate the QW sample with a 1D terahertz (THz) photonic crystal cavity, the GaAs substrate was removed by etching. We used a mixture solution of citric acid (1 g $C_6H_8O_7$ − 1 mg DI H_2O) and hydrogen peroxide (30% H_2O_2 and 70% H_2O by volume) to etch away the GaAs substrate until the AlAs etch stop layer was reached. The optimized volume ratio between citric acid solution and hydrogen peroxide was 3:1. After etching, the 2.3-μm-thick QW membrane was transfered onto the cavity center defect layer and then, together with other layers, assembled into the cavity.

The sample was mounted in a liquid helium cryostat with a variable temperature range of 1.4-300 K. A magnetic field up to 10 T was applied to the sample along the propagation direction of the incident THz wave (Faraday geometry).

Data availability. The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.