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Research Article

Sae R. Endo, Dasom Kim, Shuang Liang, Geon Lee, Sunghwan Kim, Alan Covarrubias-Morales, Minah Seo, Michael J. Manfra, Dukhyung Lee*, Motoaki Bamba and Junichiro Kono*

Cavity-mediated coupling between local and nonlocal modes in Landau polaritons

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Abstract: The multimode ultrastrong coupling (USC) regime has emerged as a novel platform for accessing previously inaccessible phenomena in cavity quantum

*Corresponding authors: Dukhyung Lee, School of Applied and Engineering Physics, Mohammed VI Polytechnic University (UM6P), Ben Guerir 43150, Morocco, E-mail: dukhyung.lee@um6p.ma.

https://orcid.org/0000-0003-1878-175X; and **Junichiro Kono**, Smalley–Curl Institute, Rice University, Houston, TX 77005, USA; Department of Electrical and Computer Engineering, Rice University, Houston, TX 77005, USA; Rice Advanced Materials Institute, Rice University, Houston, TX 77005, USA; Department of Physics and Astronomy, Rice University, Houston, TX 77005, USA; and Department of Materials Science and NanoEngineering, Rice University, Houston, TX 77005, USA,

E-mail: kono@rice.edu. https://orcid.org/0000-0002-4195-0577

Sae R. Endo, Smalley–Curl Institute, Rice University, Houston, TX 77005, USA; and Department of Applied Physics and Physico-Informatics, Keio University, Yokohama 223-8522, Japan.

https://orcid.org/0009-0006-9742-5014

Dasom Kim, Department of Electrical and Computer Engineering, Rice University, Houston, TX 77005, USA.

https://orcid.org/0000-0002-2331-4526

Shuang Liang and Michael J. Manfra, Department of Physics and Astronomy, Purdue University, West Lafayette, IN 47907, USA. https://orcid.org/0000-0002-5801-7521 (S. Liang). https://orcid.org/0000-0002-2049-8438 (M.J. Manfra)

Geon Lee, Sensor System Research Center, Korea Institute of Science and Technology (KIST), Seoul 02792, Republic of Korea.

https://orcid.org/0000-0002-8569-0260

Sunghwan Kim, Department of Physics, Ulsan National Institute of Science and Technology (UNIST), Ulsan 44919, Republic of Korea. https://orcid.org/0000-0002-0102-9169

Alan Covarrubias-Morales, Department of Electrical and Computer Engineering, Rice University, Houston, TX 77005, USA; and Applied Physics Graduate Program, Smalley–Curl Institute, Rice University, Houston, TX 77005, USA

Minah Seo, Department of Physics, Sogang University, Seoul, 04107, Republic of Korea. https://orcid.org/0000-0003-1290-9716

Motoaki Bamba, Department of Physics, Graduate School of Engineering Science, Yokohama National University, Yokohama 240-8501, Japan; and Institute for Multidisciplinary Sciences, Yokohama National University, Yokohama 240-8501, Japan. https://orcid.org/0000-0001-9811-0416

electrodynamics. Of particular interest are cavity-mediated correlations between local and nonlocal excitations, or equivalently, between modes at zero and finite in-plane momentum, which offer new opportunities for controlling light-matter interactions across space. However, direct experimental evidence of such interactions has remained elusive. Here, we demonstrate nonlocal multimode coupling in a Landau polariton system, where cavity photons simultaneously interact with the zero-momentum cyclotron resonance and finite-momentum magnetoplasmons of GaAs two-dimensional electron gas. Our slot cavities, with their subwavelength mode volumes, supply in-plane momentum components that enable the excitation of finite-momentum matter modes. Terahertz time-domain magnetospectroscopy measurements reveal a clear splitting of the upper-polariton branch, arising from hybridization between magnetoplasmon modes and the cavitycyclotron-resonance hybrids. Extracted coupling strengths confirm USC of the cyclotron resonance and strong coupling of the magnetoplasmon modes to the cavity field, respectively. The experimental results are well captured by the multimode Hopfield model and finite-element simulations. These findings establish a pathway for engineering multimode light-matter interactions involving zero- and finite-momentum matter modes in the USC regime.

Keywords: multimode coupling; magnetoplasmons; Landau polaritons

1 Introduction

Vacuum—matter interactions in cavities are currently explored in diverse systems both for controlling material properties without any external driving field and for applications in quantum information technologies. The ultrastrong coupling (USC) of vacuum—matter arises when the vacuum Rabi frequency becomes a significant fraction of the bare frequencies of vacuum and matter at zero detuning (ω_0) , i.e., $g/\omega_0 \geq 0.1$, where g is the coupling strength

[1], [2]. This regime gives rise to exotic phenomena via the non-negligible contribution of the counter-rotating terms in the Hamiltonian, such as quantum squeezing in the ground state [3], [4], the vacuum Bloch-Siegert shift [5], the Dicke superradiant phase transition [6], [7], cavitymediated superconductivity [8], and modifications of the quantum Hall effect [9], [10]. The USC regime has been realized in Landau polaritons [5], [11]-[21], phonon polaritons [22]-[25], and magnon polaritons [7], [26], [27]. Among these systems, Landau polaritons offer exceptional tunability through externally applied magnetic fields.

Recently, attention has expanded toward the multimode USC regime, where a matter excitation ultrastrongly couples simultaneously with multiple cavity modes, or conversely, a single cavity mode interacts with multiple matter excitations. This regime is not only a natural extension of single-mode USC but also introduces fundamentally new physics due to the nontrivial interplay between cavity modes or between matter excitations [20], [25], [28]-[31]. For example, correlations between bare excitations emerge due to the significant contribution of the counter-rotating terms, which do not exist in the absence of ultrastrong light-matter interactions. Very recently, matter-mediated photon-photon correlations [31] and cavitymediated superthermal phonon-phonon correlations [25] have been realized by leveraging the large dipole moment of Landau-level transitions and the subwavelength light confinement of metamaterial cavities, respectively.

Furthermore, nonlocal, i.e., $k \neq 0$ aspects of USC are also drawing increasing interest [32], where k is the in-plane wave vector of matter excitation. In particular, the possibility of mediating long-range nonlocal correlations through the vacuum field opens a new direction in controlling collective matter excitations across space. Therefore, it would be particularly interesting to explore hybrid quantum states that bridge local and nonlocal matter excitations – coupling between k = 0 and finite k modes mediated by a cavity mode. While recent studies have explored multimode USC involving finite k excitations [19], [30], [32], [33], direct observation of mode-resolved anticrossings between individual modes has so far remained elusive. To further deepen our understanding of multimode USC, it is desirable to investigate systems where such interactions can be more clearly resolved.

Here, we report spectroscopic evidence for coupling between a local mode and a nonlocal mode, through simultaneous USC and strong coupling (SC) with a single cavity mode, in a Landau polariton system. We fabricated an array of slot cavities on a two-dimensional electron gas (2DEG), and its microstructure allowed cavity photons to couple

with finite-k plasmon modes of the 2DEG by providing in-plane momentum. In a magnetic field, we experimentally observed SC of magnetoplasmon (MP) modes and the cavity mode, as well as USC between the cyclotron resonance (CR) of the 2DEG and the cavity mode. This multimode hybridization resulted in a clear splitting of the upper-polariton (UP) branch, which can be described by the multimode Hopfield model. Simulated transmission spectra using the finiteelement method exhibited similar behavior, confirming the effect of finite-k MP modes. Our findings highlight a new platform where local and finite-k matter excitations can be coupled through a cavity mode, offering a perspective on momentum-resolved multimode interactions in the USC regime.

2 Results

Providing in-plane momentum for light-matter interactions has long been achieved using periodic lattice structures [34] – [39], via patterning 2DEGs [17], [19], [33], [40] – [45] or by confining light in a small mode volume [32], [46]. To implement this mechanism, we fabricated an array of slot cavities on a GaAs 2DEG using standard photolithography and lift-off techniques. The 2DEG was present about 45 nm below the slot, ensuring strong overlap with the electromagnetic fields of the cavity mode; see Figure 1(a). The electron density and mobility were $n_{\rm e} = 3.6 \times 10^{11} \, \rm cm^{-2}$ and $\mu_{\rm e} =$ 1.2×10^6 cm²/(V s), respectively, obtained through van der Pauw measurements. The electron cyclotron mass was found to be $m^* = 0.076m_0$ through terahertz (THz) magneto spectroscopy measurements, where m_0 is the free electron mass in vacuum. The loop length of slots was tuned to have a resonance frequency in the THz frequency range. The bare cavity frequency, ω_0 , was at 0.925 THz. The width of the slot, d, was 4 µm to confine light inside the cavity, providing cavity photons with finite in-plane momentum. Although the periodic structure also provided finite in-plane momentum, its frequency was too low compared with the cavity mode. Therefore, we consider only the k components generated by the individual slots.

As a finite-k matter excitation, we had the plasma oscillations of the 2DEG. The 2D plasma frequency in the longwavelength limit is given by [47]

$$\omega_{\rm p}(k) = \sqrt{\frac{ke^2n_{\rm e}}{2m^*\varepsilon_0\varepsilon_{\rm r}}},\tag{1}$$

where ε_0 , ε_r , and e are the vacuum permittivity, the effective dielectric constant of the surrounding medium ($\varepsilon_r = 6.98$), and the elementary charge, respectively; see Figure 1(b)

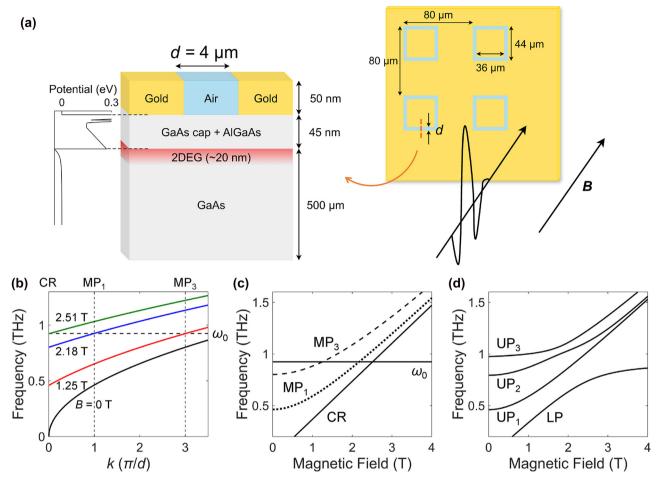


Figure 1: A multimode coupled system involving local and nonlocal matter excitations, and a cavity mode. (a) Schematic of the GaAs 2DEG-slot system. 2DEG: two-dimensional electron gas. (b) Dispersion relations of plasmons at 0 T (black) and magnetoplasmons at 1.25 T (red), 2.18 T (blue), and 2.51 T (green), corresponding to zero-detuning magnetic fields of MP₁, MP₂, and CR to ω_0 , ω_0 ; bare cavity frequency, (c), (d) Illustration of the multimode coupling of CR, MP₁, and MP₃ to the cavity mode. (c) Uncoupled case. (d) Coupled case. The multimode coupling leads to the formation of one lower polariton (LP) and three upper polaritons (UP₁, UP₂, and UP₃).

(black solid line). The slot can provide the in-plane momentum $k = n\pi/d$, where n is restricted to odd integers due to the symmetry of the electric field [19], [41]. While higherorder terms are possible, they are typically weaker and more difficult to observe. Based on our experimental results, we focus on the lowest two components, corresponding to the n = 1 and 3 modes. However, even these modes alone do not provide sufficient momentum to match the plasma resonance to the cavity frequency ω_0 , resulting in finite detuning, i.e., there is no intersection between the cavity mode and the n = 1 and 3 MP modes at $k \le 3\pi/d$.

Another matter excitation in our system was the CR of the 2DEG, arising from transitions between adjacent Landau levels at k = 0, which can be tuned by a perpendicular magnetic field, B, through the cyclotron frequency $\omega_c = eB/m^*$. In the presence of B, the 2D plasma excitations evolve into

MPs, whose dispersion is given by [48], [49]

$$\omega_{\text{MP}}^2(k) = \omega_{\text{p}}^2(k) + \omega_{\text{c}}^2. \tag{2}$$

As shown in Figure 1(b) (colored lines), the entire dispersion curve blue-shifts as the magnetic field increases. The colored curves represent the MP dispersion at various magnetic field strengths, with the red (at 1.25 T), blue (at 2.18 T), and green lines (at 2.51 T) indicating the zerodetuning conditions to the cavity mode. These field-tuned resonances can satisfy the momentum matching condition for n = 3 mode (MP₃), n = 1 mode (MP₁), and CR, respectively, enabling the exploration of hybridization of k = 0and $k \neq 0$ matter excitations through the cavity mode.

Figure 1(c) plots the bare matter excitation frequencies and the cavity frequency as a function of B. In this uncoupled situation, the CR and MP modes cross the cavity mode, showing three zero-detuning points, as expected from Figure 1(b). However, when the CR and MP modes are coupled with the cavity mode, they are expected to exhibit an anticrossing behavior at each zero-detuning point; see Figure 1(d). The multimode coupling gives rise to one lower polariton (LP) and three upper polariton branches (UP₁, UP₂, and UP₃).

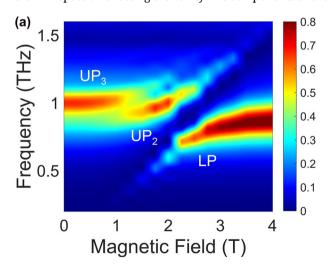
To confirm our model, we performed THz time-domain magnetospectroscopy measurements on the coupled system. THz pulses were generated and detected using InGaAs photoconductive antennas fiber-coupled to an Er-doped fiber laser with a center wavelength of 1,550 nm and repetition rate of 80 MHz. The electric field strength was sufficiently low to prevent any nonlinear effects. Measurements were conducted at 1.6 K in a magneto-optical cryostat operating in the Faraday geometry. Time-domain signals of the samples and reference (bare GaAs 2DEG) were Fourier transformed to obtain transmission spectra from 0.2 THz to 1.6 THz. Further details of the experimental setup are provided in the Supplementary Materials. Figure 2(a) shows a color map of transmittance as a function of frequency and magnetic field. At around 2.50 T, an anticrossing behavior is observed, arising from the USC between the cavity mode and the CR of the 2DEG at k = 0. At around 1.25 T, an additional splitting of the UP branch is clearly observed. This is evidence of SC of the MP3 with the cavity mode having $k = 3\pi/d$, induced by the confinement in the 4 µm slot. The split upper polaritons can be interpreted as UP2 and UP3 in Figure 1(d). UP₁ is absent in the experimental data due to the THz spectra reflecting the cavity-like components of the

polaritons, as well as the proximity of UP₁ to the CR at finite fields.

To theoretically explain the coupling between a cavity mode, CR, and MP modes, we introduce a multimode Hopfield model. By adding the MP modes to the full Hamiltonian of Landau polaritons presented in ref. [5] while considering only a single cavity mode, the multimode Hamiltonian can be written as

$$\begin{split} \frac{\widehat{H}}{\hbar} &= \sum_{\xi=\pm} \omega_0 \hat{a}_{\xi}^{\dagger} \hat{a}_{\xi} + \omega_c \hat{b}^{\dagger} \hat{b} + \sum_n \omega_{\text{MP}_n} \hat{c}_n^{\dagger} \hat{c}_n \\ &+ i \left[\left(\overline{g} \hat{b}^{\dagger} + \sum_n \overline{g}_n \hat{c}_n^{\dagger} \right) (\hat{a}_+ + \hat{a}_-^{\dagger}) \right. \\ &- \left(\overline{g} \hat{b} + \sum_n \overline{g}_n \hat{c}_n \right) (\hat{a}_- + \hat{a}_+^{\dagger}) \right] \\ &+ D \left(\hat{a}_- + \hat{a}_+^{\dagger} \right) (\hat{a}_+ + \hat{a}_-^{\dagger}), \end{split} \tag{3}$$

where \hbar is the reduced Planck constant, \hat{a}^{\dagger} (\hat{a}) in the first term is the creation (annihilation) operator for the cavity photons of CR-active ($\xi=+$) and CR-inactive ($\xi=-$) circularly polarized modes, the second term represents the energy of the collective CR excitation of the 2DEG, \hat{b}^{\dagger} (\hat{b}) being its creation (annihilation) operator, MP modes of different orders n=1,3 are introduced in the third term, where \hat{c}_n^{\dagger} (\hat{c}_n) and ω_{MP_n} are the corresponding creation (annihilation) operator and resonance angular frequency, and the fourth term is the light–matter interaction term considering the coupling of CR and all MP modes with the single cavity mode.



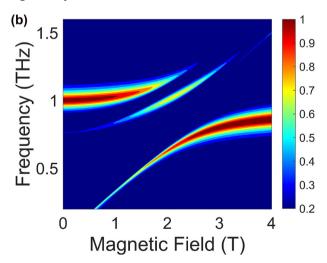


Figure 2: Spectroscopic evidence for the multimode coupling. (a) Experimental color map of transmittance as a function of frequency and magnetic field. The upper polariton is split into two parts, corresponding to UP₂ and UP₃ in Figure 1(d). (b) Theoretical color map of transmittance based on the multimode Hopfield model.

Here, \overline{g} and \overline{g}_n are the coupling strengths of CR and the n-th MP mode with the cavity mode, respectively. Using the zero-detuning coupling strengths, g and g_n , which are independent of the external B, \overline{g} and \overline{g}_n can be written as

$$\overline{g} = \sqrt{\frac{\omega_{\rm c}}{\omega_0}} g,\tag{4}$$

$$\overline{g}_n = \sqrt{\frac{\omega_{\text{MP}_n}}{\omega_0}} g_n. \tag{5}$$

The coefficient D of the diamagnetic term is

$$D = \frac{\overline{g}^2}{\omega_{\rm c}} + \sum_n \frac{\overline{g}_n^2}{\omega_{\rm MP_n}} = \frac{g^2 + \sum_n g_n^2}{\omega_0}.$$
 (6)

The coupling strengths appear in the relative permittivity as

$$\varepsilon(\omega) = \varepsilon_{\text{bg}} - \frac{\omega_{\text{pl}}^2}{\omega \left(\omega - \omega_{\text{c}} + \frac{i}{\tau}\right)} - \sum_{n} \frac{\omega_{\text{pl}_n}^2}{\omega \left(\omega - \omega_{\text{MP}_n} + \frac{i}{\tau_n}\right)},\tag{7}$$

where the plasma frequencies of CR and n-th MP mode, ω_{nl} and $\omega_{\rm pl}$, have relations with the *B*-independent coupling strengths as

$$g^2 = \frac{\omega_{\rm pl}^2 d_{\rm QW}}{\varepsilon_{\rm bg} L_{\rm eff}},\tag{8}$$

$$g_n^2 = \frac{\omega_{\rm pl_n}^2 d_{\rm QW}}{\varepsilon_{\rm bg} L_{\rm eff}}.$$
 (9)

Here, $\varepsilon_{\rm bg} = \varepsilon_{\rm r}$ is the background relative permittivity, au (τ_n) is the lifetime of CR (MP $_n$), $d_{\rm QW}$ is the thickness of the quantum well, and $L_{\rm eff}$ is the effective cavity length. Using the above relative permittivity, the transmission spectra can be obtained through the transfer-matrix method.

On the other hand, the polariton dispersion can be calculated from the B-dependent coupling strengths. The Heisenberg equations for the operators \hat{a}_{+} , \hat{a}_{-}^{\dagger} , b, and c_n form a closed set describing the CR-active polariton modes, while those of \hat{a}_{-} , \hat{a}_{+}^{\dagger} , b^{\dagger} , and c_{n}^{\dagger} describe the CRinactive modes. The CR-active polariton annihilation operator is given by the Bogoliubov transformation, $\hat{p}_+ = w\hat{a}_+ + \hat{a}_+$ $y\hat{a}_{-}^{\dagger} + x\hat{b} + \sum_{n} x_{n}\hat{c}_{n}$. In the same manner as performed by Ciuti et al. [3], the polariton frequencies and the coefficients are determined by solving the eigenvalue problem of the

following matrix:

$$\begin{pmatrix} \omega_0 + D & -D & i\overline{g} & i\overline{g}_1 & \dots \\ D & -\omega_0 - D & i\overline{g} & i\overline{g}_1 & \dots \\ -i\overline{g} & i\overline{g} & \omega_c & 0 & \dots \\ -i\overline{g}_1 & i\overline{g}_1 & 0 & \omega_{MP_1} & \dots \\ \vdots & \vdots & \vdots & \vdots & \ddots \end{pmatrix}$$
(10)

Theoretical transmission spectra for the multimode Landau polariton system, calculated using the transfermatrix method with the relative permittivity $\varepsilon(\omega)$ derived in Eq. (7), are shown in Figure 2(b). The resonator structure is simplified to a Fabry-Pérot cavity; details of the calculation setup are provided in the Supplementary Materials. The splitting in the UP branch is observed and captures the key feature of the experimental data. We emphasize that the MP₁ interacts with the cavity-cyclotron-resonance hybrids at only high frequencies and cannot account for the splitting of the upper polariton at around 1 THz without including the MP3 in the model; see Supplementary Materials. The discrepancy in linewidth compared to the experimental spectra is associated with the difference in cavity geometry, with the slots having a reduced O-factor compared to an ideal Fabry-Pérot cavity. From fitting to the experimental peak frequencies, we obtained the normalized coupling strengths of $g/\omega_0 = 0.18$ and $g_1/\omega_0 =$ $g_3/\omega_0=$ 0.084, confirming the USC of the CR and the SC of MP modes with the cavity mode. Figure 3 shows the calculated polariton frequencies (black solid lines) overlaid with the peak frequencies extracted from the experiments (red dots). Theoretical predictions match the experimental results well. We note that the assumption $g_1 = g_3$ is generally not valid but is enough to explain the experimental behavior.

We further validated our experimental observation and theoretical model using three-dimensional finiteelement simulations in COMSOL. Figure 4 shows a color map of transmittance simulated with $m^* = 0.070m_0$ and $\varepsilon_{\rm r}=9.77$. Compared with the experimental transmittance in Figure 2(a), the UP branch is split into three parts, separated not only by MP₃ but also by MP₅, resulting in UP₄. Such influence of the MP₅ could not be observed experimentally due to the weak magnitude. Incorporating the MP₅ contribution to the multimode Hopfield model, theoretical fits to the polariton frequencies are calculated, shown in black dashed lines in Figure 4; see Supplementary Materials for more detail. The obtained coupling strengths are $g/\omega_0=0.13$ and $g_1/\omega_0=g_3/\omega_0=g_5/\omega_0=0.062$. The slight deviation from the experimentally extracted values likely originates from uncertainties in the sample parameters and the strong

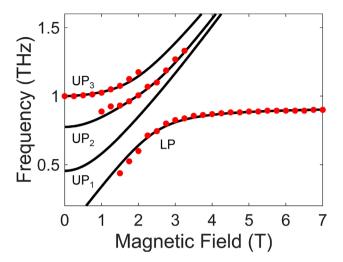


Figure 3: Theoretical analysis of the multimode coupled system. Theoretical polariton frequencies based on the multimode Hopfield model. Red dots are extracted peak frequencies from experiment.

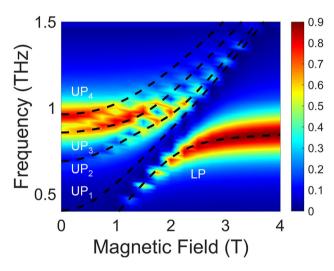


Figure 4: Finite-element simulation of the multimode coupled system. Simulated color map of transmittance as a function of frequency and magnetic field using COMSOL. The black dashed lines are theoretical fits to the polariton frequencies based on the multimode Hopfield model. An additional upper polariton mode, denoted UP_4 , appears due to the influence of the MP_5 mode.

effect of the MP₅ mode. Overall, the simulation is consistent with both the experimental and theoretical analysis of the multimode coupling.

3 Discussion

In conclusion, we experimentally explored the coupling between two distinct matter modes mediated by a cavity mode in an ultrastrongly coupled Landau polariton system. Owing to the small-mode-volume slots, cavity photons could interact with both the CR at k = 0 and MP modes at finite k. Such coexistence of CR and MP excitations has been discussed in previous studies using 2DEG stripes and periodic metasurfaces [20], [30], [33]. However, the multimode interaction demonstrated here could not be accessed due to the excitation geometry and frequency mismatch. In this work, the MP modes affected the Landau polariton through multimode hybridization and led to a splitting of the UP branch. From fitting, we obtained a normalized coupling strength of $g/\omega_0=$ 0.18 for CR, and $g_1/\omega_0=$ $g_3/\omega_0=0.084$ for MP modes with an in-plane wave vector of $k = \pi/d$ and $k = 3\pi/d$, showing the USC of CR and SC of MP modes to the cavity mode. The experimental results were well explained by the extended multimode Hopfield model and three-dimensional finite-element simulations. Our work establishes a foundation for controlling cavitymediated nonlocal correlations between matter excitations in the USC regime, unlocking new possibilities for tailoring light-matter interactions.

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Author contributions: JK supervised the project. SRE, DK, and AC-M performed all THz measurements. SRE and DK analyzed the experimental data under the guidance of JK. DK built the THz magnetospectroscopy setup. SL grew the quantum well under the guidance of MJM. GL, SK, and DK designed and fabricated the slot cavities under the guidance of MS. MB developed the theoretical model. DL performed numerical simulations and analyzed the experimental data. SRE, DK, MB, and JK wrote the manuscript. All authors discussed the results and commented on the manuscript. All authors have accepted responsibility for the entire content of this manuscript and consented to its submission to the

journal, reviewed all the results and approved the final version of the manuscript.

Conflict of interest: Authors state no conflicts of interest. Data availability: The datasets generated and/or analyzed during the current study are available from the corresponding authors upon reasonable request.

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