Ultrafast Coherence Delocalization in Real Space Simulated by Polaritons

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Coherence delocalization has been investigated on a coupled-cavity molecular polariton platform in time, frequency, and spatial domains, enabled by ultrafast two-dimensional infrared hyperspectral imaging. Unidirectional coherence delocalization (coherence prepared in one cavity transferred to another cavity) has been observed in frequency and real space. This directionality is enabled by the dissipation of delocalized photon from high-energy to low-energy modes, described by Lindblad dynamics. Further experiments show that when coherences are directly prepared between polaritons from different cavities, only energetically nearby polaritons can form coherences that survive the long-range environmental fluctuation. Together with the Lindblad dynamics, this result implies that coherences delocalize through a one-step mechanism where photons transfer from one cavity to another, shedding light to coherence evolution in natural and artificial quantum systems. This new optical platform based on molecular vibrational polariton thus demonstrates a way of combining photon and molecular modes to simulate coherence dynamics in the infrared regime.

1. Introduction

Coherences, a result from the superpositions of wavefunctions, are key ingredients of quantum systems, such as artificial quantum simulation platforms, natural light-harvesting antennas, ballistic energy transfer and charge-transfer materials. To mediate non-diffusive energy transfer or transfer phase sensitive information, coherence propagates among different sites that are composed of distinct quantum states, which was shown by pioneering ultrafast spectroscopic studies by resolving their signals in the frequency domain. However, it remains to be a challenge to spatially track coherences in these systems, because coherence transfer or delocalization in molecular systems happens between atoms that are a few angstroms or nanometers away.

In the current report, we show that by taking advantage of the fast propagation of photon modes, molecular vibrational polaritons in the coupled dual cavities, as a potential qubit system, can overcome the intrinsic fast vibrational decoherence and indeed simulate coherence delocalization on micron spatial scales. When molecular vibrational modes are strongly coupled to the cavity modes, they form hybridized quasi-particles–molecular vibrational polaritons. Using a coupled dual-cavity polariton platform, we previously demonstrated that nonlinear interactions in the system can be delocalized, through exciting the dark state populations. However, it is insufficient to realize coherence transfer because, unlike dark states, polariton coherences are much shorter lived and are vulnerable to solvent fluctuations across spaces. In this work, using two-dimensional infrared (2D IR) microscopy, we show this molecular vibrational polariton platform can support the preparation of polariton coherence, and the coherence can delocalize and propagate in the coupled dual cavities. We learn these insights by time-resolving the polariton coherence spatial distribution. We prepare polaritons into specific quantum coherences in one cavity using a pair of pump laser pulses and then follow the coherence evolution by the probe pulse (Figure 1a), which generates 2D IR spectra. The 2D IR spectra show a unidirectional coherence delocalization, imaged by a home-built hyperspectral microscope. Spatial dynamics show the coherence delocalize from high to low-frequency cavities, but the reverse is unfavored, driven by non-Hermitian Lindbladian dynamics of photon delocalization and dissipation. To further understand the coherence spatial transport, we attempt to generate coherence directly between two neighboring cavities, referred as inter-cavity coherence. Only inter-cavity coherence between polaritons with sufficiently small energy separation could be prepared. These results indicate that the spatial environment fluctuation only does not destroy phase relations between states that are energetically close enough.
By combining the fast propagation of photon and nonlinearity of molecular vibrational modes, molecular vibrational polariton thus can bypass the fast decoherence in the liquid phase, opening a new optical material as a platform for simulating certain complex and chemistry-related quantum and classical phenomena under ambient conditions.

2. Results

2.1. Coupled Cavity Polaritons and their Spatial Distribution

The key component of the coherence propagation study was the coupled cavity which enabled coherence delocalization and non-linear interactions. The cavity was formed by a distributed-Bragg-reflector (DBR) with checkerboard patterns and another flat DBR, which created alternating cavities with different longitudinal lengths. In our experiment, the depth of cavity A and B were 12.5 and 12.7 µm, respectively and the lateral width of each cavity was 50 µm (Figure 1b). To enable nonlinearity, we injected 40 mM W(CO)₆ in hexane solution into the coupled cavity. As shown before, with a Rabi splitting of ≈40 cm⁻¹, which was significantly larger than the width of the cavity mode (≈10 cm⁻¹) and the vibrational modes (≈5 cm⁻¹), the system satisfied the criteria of strong light-matter interactions. Specifically, the asymmetric vibrational modes of the molecules at 1983 cm⁻¹ strongly coupled to the cavity that contained the molecules, and weakly coupled to the neighbor cavities by evanescent waves. Therefore, four polaritons (noted as UPᵢ and LPᵢ for polaritons in cavity i, where i = A or B) were formed (Figure 1c).

The polaritons were imaged using a 12x hyperspectral 1D IR imaging, by which a vertical line of an image was projected so that its signals along the vertical axis revealed the vertical spatial pattern and the horizontal dispersion corresponded to the linear IR spectra (Figure 1d). The image in Figure 2a confirmed that two polaritons were formed in each cavity and they were vertically displaced from each other, reflecting the spatial separation between cavity A and B. The polaritons also diffused into the neighboring modes, indicating delocalization (see Supporting Information S1 for details).

To properly understand the polariton mode distribution, we simulated the cavity modes by the wave equation of electric fields where the coupled-cavity unit cell was modeled using parameters from experiments and assuming periodic boundary conditions. Then we simulated the strong coupling between the cavity and molecular modes using the Tavis-Cummings model (see Materials and Methods and Supporting Information S9). The simulated spectra (Figure 2b, top) showed four polariton peaks with the corresponding hyperspectral image (Figure 2b, bottom) revealing the polaritons from cavity A resided in the upper region whereas the ones from cavity B were in the lower region. Figure 2c plotted the polariton spatial distribution simulated from the model, which showed that polaritons were mostly localized in their own cavities. However, the UPᵢ and LPᵢ states had distributions in cavity B. As shown later, the propagation of UPᵢ and LPᵢ into cavity B were crucial for coherence delocalization.

2.2. Polariton Coherence Delocalization in Space

We next imaged the spatial coherent transport by an ultrafast hyperspectral IR imaging setup. This instrument was done
by adding a pump pulse to enable a 2D IR pulse sequence (Figure 1a) in the hyperspectral IR microscopy (Figure 1d). This setup allowed up to five-dimension imaging (3D in frequency and 2D in space). In this study, we used it to spatially resolve 2D IR signals and coherence spatial distribution.

Because the four polariton states could generate many pathways that contributed to 2D IR signals when broadband pulses were used, to avoid unnecessary signals and focus on coherence delocalization only, we applied tailored pulse sequences to create specific coherences.\[34,45,46\] For example, we truncated the first two pulses in the frequency domain so that it only created the initial coherence $|UP_i\rangle\langle LP_i|$, where $i$ represented cavity A or B. Then, we scanned $t_2$ (the time delay between the second pump pulse and probe pulse, Figure 1a) to monitor the coherence evolution. This process can be represented diagrammatically by the double side Feynman diagram\[47\] (Figure 3a). The representative spectra of all four initial coherences were listed in Figure 3b. It was noticeable that when $|UP_A\rangle\langle LP_A|$ or $|LP_A\rangle\langle UP_A|$ were created, there were also cross peaks at $\omega_3 = \omega_{UPB}$ and $\omega_{LPB}$ (grey shaded areas in Figure 3b), whereas such cross peaks were negligible when the other coherences were prepared.

When the initial coherence was $|UP_A\rangle\langle LP_A|$, strong oscillating signals appeared at $\omega_3 = \omega_{UPA}$ and $\omega_{LPB}$, (indicated by the white dashed lines in Figure 3c) and $\omega_3 = \omega_{UPB}$ and $\omega_{LPB}$ (black dashed line in Figure 3c). All spectral features oscillated at $\approx 40 \text{ cm}^{-1}$, corresponding to the Rabi frequency. Because we excluded the possibility of exciting polaritons in cavity B by the tail of the pump spectra (see Supporting Information S5), the oscillating signals at $\omega_3 = \omega_{UPB}$ and $\omega_{LPB}$ suggested that coherence delocalized to cavity B upon being generated in cavity A (Feynman pathway illustrated in Figure 3a). In contrast, when initial coherence $|UP_B\rangle\langle LP_B|$ was prepared, there was no noticeable delocalization to either $UP_A$ or $LP_A$ (for example, lack of coherence oscillation in the trace indicated by the white dashed lines in Figure 3d). This unidirectional delocalization was robust, regardless of whether $|UP_A\rangle\langle LP_A|$ or $|LP_A\rangle\langle UP_A|$ was created. However, neither $|LP_B\rangle\langle UP_B|$ nor $|UP_B\rangle\langle LP_B|$ could transfer to cavity A (see Supporting Information S6). Such unidirectional delocalization became relaxed, when preparing and probing polariton coherences at higher in-plane momentum ($k||$) (additional experiments at higher $k||$ are shown in Supporting Information S7).

To examine whether polariton coherence indeed was delocalized from cavity A to B, we spatially-resolved the coherence dynamics. After first creating $|UP_A\rangle\langle LP_A|$, the oscillating signal at $\omega_3 = \omega_{UPA}$ and $\omega_{LPB}$ was localized in cavity A region (Figure 4a and b). However, the same initial coherence also created the beating patterns at $\omega_3 = \omega_{UPB}$ and $\omega_{LPB}$ that centered at cavity B, at a slight delayed time (Figure 4c,d). Therefore, these time-resolved hyperspectral images showed clear evidence that when coherence in cavity A was created, it delocalized into cavity B. Similar results were obtained when $|LP_A\rangle\langle UP_A|$ was created.

### 2.3. Mechanism of Unidirectional Coherence Delocalization and the Importance of Non-Hermitian Dynamics

This unidirectional delocalization could be understood by the delocalization and dissipation of photon modes. The
Figure 3. Intercavity coherence delocalization dynamics measured by 2D IR. a) A representative Feynman diagram showing the polariton coherence transfer. b) Four 2D IR spectra with four different initial polariton coherence, from top to bottom: $|\text{UP}_A\rangle|\text{LP}_A\rangle$, $|\text{UP}_B\rangle|\text{LP}_B\rangle$, $|\text{LP}_A\rangle|\text{UP}_A\rangle$, and $|\text{LP}_B\rangle|\text{UP}_B\rangle$, where the 2D IR cross-peaks indicating the coherence transfer are labeled in grey shaded area. 2D IR dynamics with initial coherence of c) $|\text{UP}_A\rangle|\text{LP}_A\rangle$ and (d) $|\text{UP}_B\rangle|\text{LP}_B\rangle$, the former shows clear coherence delocalization from cavity A to B, whereas the latter does not.

Figure 4. Spatially-resolved coherence dynamics obtained from 2D IR spectral cut and their corresponding linear transmission images (left). Initial states are all $|\text{UP}_A\rangle|\text{LP}_A\rangle$ and the probed states are a) UP$_A$, b) LP$_A$, c) LP$_B$ and d) UP$_B$. 
transfer of polariton coherence was driven by the propagation of photons from one cavity to another, supported by the associated ultrafast time scale. As photon transfer to another cavity, its energy should be conserved following the relations of \( k_i^2 + (n\pi/d_i)^2 = \omega^2/c^2 = k_f^2 + (n\pi/d_f)^2 \), where \( c \) was the velocity of light in the cavity, \( d_i \) and \( k_i \) were the cavity thickness and in-plane momentum in cavity \( i \). Because \( d_A < d_B \), the lowest \( (k_A \rightarrow 0) \) photon mode in cavity \( B \) could not propagate to cavity \( A \), since the propagation of photon required \( k_A \) to be imaginary, implying attenuation of waves. Instead, when the lowest \( (k_A \rightarrow 0) \) photon mode in cavity \( A \) could hybridize with higher momentum modes in cavity \( B \), as \( k_B \) remained real and finite, supporting photon propagation from cavity \( A \) to \( B \). Thus, the same principle that caused total internal reflections drove the unidirectional delocalization of photon.

However, the unidirectional photon delocalization alone was still insufficient to explain the transition of polariton in the frequency space, as the photon propagation was a Hermitian dynamic that preserved energy. Non-Hermitian energy dissipation dynamics also played an important role. As photon propagated from cavity \( A \) to cavity \( B \), its momentum increased, which allowed the photon to couple to the dark modes of molecular vibration in cavity \( B \). The dark mode then decayed to the lowest momentum bright mode in cavity \( B \) by dissipating its energy to the environment (i.e., solvents), which ensures the unidirectional transfer.

To justify our understanding, we further simulated the 2D IR spectra based on the Lindblad dynamic of the density matrix with a non-Hermitian term that described the photon decay from cavity \( A \) to cavity \( B \). Although the phase twist in the experimental data was not reproduced in the simulation, likely due to the inherent inhomogeneity of the polariton modes in experiments, the simulated spectrum in Figure 5a captured the salient features (such as peak position and intensities) of the experimental one. For example, the cross peaks that originated from coherence delocalization from cavity \( A \) to \( B \) was much more pronounced than the ones from the other direction. The simulated coherence dynamics in Figure 5b showed that the oscillation frequency of the \( |{UP_A}\rangle|{UP_B}\rangle \) coherence was locked with that of \( |{LP_A}\rangle|{LP_B}\rangle \). However, if we turned off the non-Hermitian dissipation term in the Lindblad dynamics, the simulation in Figure 5c showed that the coherent transfer was no longer stable, and more importantly, the oscillation frequencies were no longer locked together (see dynamic cuts in Figure S12, Supporting Information). Because the energy was conserved in the absence of dissipation, such that each polariton mode would beat independently according to their detuning from the central frequency. The observation of the frequency locking behavior in the experiment strongly supported the importance of non-Hermitian dynamics in understanding the unidirectional coherence transfer.

2.4. Intercavity Polariton Coherences Across Space

We analyzed further the extent of the robustness of coherence across cavities. The intercavity distance was 50 µm, and solvent motion across this large distance was disordered and uncorrelated. It was unclear how robust the phase could be against the fluctuation of solvents in real space. To gain insight into this question, we prepared coherences between polaritons in different cavities directly, e.g., \( |{UP_A}\rangle|{UP_B}\rangle \) or \( |{LP_A}\rangle|{LP_B}\rangle \) (Figure 6a,b). When either \( |{UP_A}\rangle|{UP_B}\rangle \) or \( |{LP_A}\rangle|{LP_B}\rangle \) was created, we observed a very slow oscillation at \( \omega_0 = 10 \, \text{cm}^{-1} \) (a period of \( 3 \approx 4 \, \text{ps} \)), which agreed with the energy difference between the corresponding states (Figure 6c,d). The nonlinear oscillating signal was not only seen in the original excited polariton state, but also the other polariton from the same cavity. For instance, when \( |{UP_A}\rangle|{UP_B}\rangle \) were prepared, oscillating signals appeared at \( UPA \) and \( UPB \), and at \( LPA \) and \( LPB \) transitions. This was not surprising as the nonlinear interaction was strong between polaritons in the same cavity. We noted the oscillating signal appeared at an even higher frequency than \( \omega_0 = \omega_{UPA} \) could be from polaritons of high order cavity modes, which was out of the scope of the current work. The existence of intercavity coherence such as \( |{UP_A}\rangle|{UP_B}\rangle \) and \( |{LP_A}\rangle|{LP_B}\rangle \) suggested that they could survive environmental variations between cavities.

A sharp contrast was when \( |{UP_A}\rangle|{LP_B}\rangle \) or \( |{LP_A}\rangle|{UP_B}\rangle \) was prepared, no nonlinear signal and coherence dynamics were observed (Figure 6e). This result indicated that such coherences did not survive the solvent variation across cavities. The energy separation between \( UPA \) and \( LPB \) was close to 50 cm\(^{-1}\) and between \( UPB \) and \( LPA \) was about 30 cm\(^{-1}\), significantly larger than the 10 cm\(^{-1}\) when the intercavity coherences were observed. This result agreed with that in general high-frequency coherence was more vulnerable to fluctuations than low-frequency ones. Considering that neither \( |{UP_A}\rangle|{LP_B}\rangle \) nor \( |{LP_B}\rangle|{LP_A}\rangle \) could be created directly using laser pulses, the coherences would be destroyed if \( |{LP_B}\rangle|{LP_A}\rangle \) transferred to \( |{UP_A}\rangle|{LP_B}\rangle \) through these intermediate pathways (i.e., \( |{LP_B}\rangle|{LP_A}\rangle \) or \( |{LP_B}\rangle|{LP_B}\rangle \)). This result suggested that multi-step coherence transfer across the cavity was less likely. Instead, the coherence transfer was facilitated through a photon delocalization and dissipation process, where both the bra and ket states of the coherence finished transfer in one step.

3. Discussion

Taking advantage of photon delocalization and molecular non-linearity, the coupled dual-cavity polariton was used to gain insights into coherence delocalization in real spaces, through a time-resolved hyperspectral IR microscope. The present study shed light on coherent propagation in quantum systems. First, in a system which was at the bottom of its dispersion curve (i.e., in-plane momentum was zero), downhill coherence transfer was much favored than uphill. However, such preference was less strict at higher momentum space or when momentum conservation could be relaxed. Second, all coherences oscillated at the same frequency as the initial coherence, which could only be modeled by a non-Hermitian Lindbladian dynamics, suggesting energy dissipation and photon delocalization both played critical roles in coherence transfer. Last, coherence could only be maintained between cavities when the energy difference between two quantum states was small enough to survive environmental variations in space. Thus, it was not likely that the coherence delocalized through any intermediate superposition states, with energy larger than 10 cm\(^{-1}\). Instead, a one-step
coherence transfer mechanism through single photon delocalization and dissipation was proposed. Such one-step mechanism suggested that in other coherence transfer systems, such as light-harvesting complex, the coherence delocalization could also depend on the spread of wavefunctions with the highest propagation speed.

The insights learned in this study could be applied when designing coherence transfer in new artificial quantum systems or explaining coherence evolutions in natural quantum phenomena. The present work also opened molecular vibrational polaritons as a new optical system for quantum simulation at ambient conditions. Molecular modes were often not considered for simulating coherent dynamics, as the stochastic processes in liquid phase at room temperature causes decoherence in the ultrafast timescale. The previous work on plasmon polaritons have demonstrated the precise control over the molecular nonlinearity at a nano-scale distance and femtosecond time scale. However, when strongly coupled to the cavity photons, whose velocity allow them to spread the coherence at a long distance before it decays, the molecular vibrational modes can be used to understand certain ultrafast processes despite still being open-dissipative. This advance can be further developed for simulating complex processes complementing the capability of existing quantum simulation platforms, by developing advanced photonic structures, taking advantage of intrinsic molecular dynamics, such as energy transfer and isomerization, and multiple molecular chromophores. The ability to spatially image coherence also...

Figure 5. Simulated spectra and coherence dynamics of the coupled cavity polariton systems. a) Simulated 2D IR spectrum of dual-cavity strong coupled system. Simulated 2D IR dynamics of initial coherence of $|UP_A\rangle\langle LP_A|$ and $|UP_B\rangle\langle LP_B|$ of b) a non-Hermitian system and c) a Hermitian system. Non-Hermitian Hamiltonian is necessary to reproduce experimental results.
Figure 6. Direct creation of intercavity coherence (superpositions between polaritons in different cavities). a) illustration of |UPA⟩⟨UPB| formation as an example of intercavity coherence. b) Feynman diagram showing the creation of intercavity coherence (left) and possible transfer pathways (right). Direct intercavity coherences appear with initial states of c) |UPA⟩⟨UPB| and d) |LPB⟩⟨LPB|, whereas e) |UPB⟩⟨LPB| shows nearly no signature of coherences.

4. Experimental Section

Preparation of Dual-Cavity Polariton System: In order to generate two pairs of polaritons, a dual-cavity system has been developed with checkerboard patterns of 50 µm lateral dimension (see SI S1 for more details). The W(CO)₆ (Sigma-Aldrich)/coupled-cavity system was prepared by sandwiching W(CO)₆/hexane solution by a dual-cavity mirror and a flat dielectric CaF₂ mirror, separated by a 12.5 µm Teflon spacer. The W(CO)₆/hexane solution was nearly saturated concentration (40 mM).

Hyperspectral 2D IR Microscopy: The 2D IR signal was sent through a pair of CaF₂ lenses which projected the IR images onto the entrance of the spectrograph slit. The slit allowed a vertical cut of the image to enter the spectrograph that relayed a one-to-one ratio image vertically and dispersed the spectra of each point along the vertical slice horizontally. In this way, the horizontal axis of hyperspectral images reported the spectra and the vertical axis reflected the spatial distribution of the signals (Figure 1d).

Theoretical Modeling: The cavity mode was simulated by solving the wave equation of electric field \( E(r) \) in the cavity

\[
c^2 \left( -\nabla^2 \alpha + \frac{n_e}{\alpha(r)} \right) E(r) = \alpha^2 E(r)
\]

where \( d(r) \) is the cavity thickness at position \( r = (x,y) \), \( c \) is the speed of light in the hexane solution inside the cavity, and \( n_e \) is the excitation mode quantum number in the perpendicular direction. Best fit to the observation data indicates that \( n = 4 \). The bright mode solutions of the wave equation were focused, which correspond to modes with zero quasi-momentum over the cavity lattice and s-wave symmetry inside the cavity. Two such modes at \( \omega_B = 1998.2 \text{ cm}^{-1} \) (dominantly in cavity A) and \( \omega_A = 1971.4 \text{ cm}^{-1} \) (dominantly in cavity B) was found. Based on these two cavity photon modes, the polariton modes can be modeled by the Tavis-Cummings model described by the following Hamiltonian

\[
H = \sum_{i=A,B} \omega_i \rho_i + \omega_0 \sigma_i^+ \sigma_i^- + g (\sigma_i^+ \rho_i^+ + \text{h.c.})
\]

where \( \sigma_i \) denotes the photon annihilation operator of the \( i \)th cavity mode and \( \sigma_i^+ \) denotes the raising/lowering operator of the (collective) vibration mode that couples to the corresponding photon mode. Dark modes would be omitted in the model. The best fit to the experimental observation shows that the light-matter coupling strength was around \( g = 18.7 \text{ cm}^{-1} \). Using this parameter, the model produces polariton modes at \( \omega_{2PA} = 1970.0 \text{ cm}^{-1} \), \( \omega_{2PB} = 1957.4 \text{ cm}^{-1} \), and \( \omega_{5PA} = 1957.4 \text{ cm}^{-1} \), matching the experimental observation nicely. The pump/probe laser can be modeled by the perturbation was assumed.

\[
V = \sum_{i=A,B} \mu_i \left( a_i^+ a_i^0 + a_i^0 a_i^+ \right)
\]

where \( \mu_i \) characterizes coupling strength between the laser mode and the cavity mode. To simulate the pump-probe dynamics, the Lindblad dynamics of the polariton was consider, under which the density matrix \( \rho \) of the system evolves by

\[
\frac{\partial}{\partial t} \rho = \mathcal{L}[\rho] = [H, \rho] + \sum_{m} \left( F_m \rho F_m^\dagger - \frac{1}{2} F_m^\dagger F_m \rho - \frac{1}{2} \rho F_m^\dagger F_m \right)
\]

where the Lindblad operator \( F_m \) enables us to describe the dissipation of the photon and vibration modes by \( F_{1,2,3,4,5,6} = \sqrt{\frac{1}{2}} \sigma_i \rho \sqrt{\frac{1}{2}} \sigma_i^+ \), as well as the non-Hermitian photon transfer from high-energy to low-energy modes \( F_7 = \sqrt{\frac{1}{2}} \sigma_i^+ \rho \sigma_i \). Starting from the initial vacuum state \( \rho = |0\rangle\langle 0| \), the pump/probe laser acts by \( \rho \rightarrow \rho + \sum_{m} \left( F_m \rho F_m^\dagger - \frac{1}{2} F_m^\dagger F_m \rho - \frac{1}{2} \rho F_m^\dagger F_m \right) \). Finally, emission amplitude is given by Tr V \( \rho \). Within this formalism, the 2D IR spectroscopy result could be simulated and compared it with the experiment. It turns out that the \( F_7 \) non-Hermitian photon transfer process plays important role in understanding the coherence transfer.
between cavities. Additional theoretical details can be found in the Supporting Information S9.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Author Contributions
W.X. conceived the original idea. W.X. and Y.Z.Y. developed the theoretical model in this work. B.X. conducted 2D IR experiments and analyzed the data. Z.Y. contributed to the experiments. B.X., Y.Z.Y. and W.X. wrote the manuscript.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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2D infrared spectra, coherence, hyperspectral imaging, molecular vibrational polaritons, quantum simulation, vibrational strong coupling

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