

Multiple quantum-well polaritons in organic-inorganic two-dimensional perovskite films with controlled well-layer number

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We report the observation of multiple quantum-well (MQW) polaritons in organic-inorganic metal halide two-dimensional (2D) perovskite films with precisely controlled well-layer numbers. Using the Langmuir-Blodgett method, we fabricated high-quality MQW structures that eliminate well-width fluctuations, enabling the study of MQW polaritons in the low layer number region. Optical reflection spectroscopy and transfer matrix calculations reveal that spectral changes with increasing well numbers originate from MQW-polariton effects, successfully demonstrating the correspondence between spectral structures and MQW-polariton modes. We identify the superradiant regime, where a single radiant mode dominates, followed by the sequential emergence of higher-order radiant modes. These findings provide crucial insights into the evolution of MQW polaritons toward three-dimensional polaritons and contribute to fundamental studies of ultimate 2D systems. Furthermore, our results offer valuable knowledge for the development of perovskite-based optical devices.

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I. INTRODUCTION

Excitonic properties in two-dimensional (2D) systems, such as quantum wells (QWs), have been extensively studied due to their strong light-matter interactions [1]. In multiple quantum wells (MQWs), excitons and exciton-polaritons—coupled modes of exciton and light—are typically confined within the QW planes. However, under ideal conditions, MQW polaritons can form along the direction perpendicular to the wells [2–12]. This occurs when the excitons with large oscillator strength couple optically to excitons in adjacent wells. Notably, this coupling is purely optical, not electronic, as the electronic wavefunctions in different wells do not spatially overlap. In such cases, MQW-polariton modes emerge with the same number of QWs (N_w) and can be classified as either radiant (bright) modes, which exhibit enhanced oscillator strength compared to a single QW, or dark modes, which have reduced oscillator strength.

The MQW polaritons discussed here arise not from the coupling between cavity photons and excitons, as in

conventional cavity polaritons, but from optical coupling among excitons located in different quantum wells. The study of such MQW polaritons is of significant interest for both fundamental physics and optical device applications, as it provides insight into the transition from 2D to three-dimensional (3D) optical properties [9–11]. While increasing the QW thickness (d_w) naturally drives the system from 2D toward 3D behavior, our focus here is on whether a similar dimensional crossover can be achieved by increasing N_w , while keeping both d_w and the well spacing (d) sufficiently small. Specifically, our interest lies in whether such a configuration leads to the emergence of a forbidden band, known as the longitudinal-transverse (LT) gap, characteristic of 3D polaritons [1]. In this context, it becomes crucial to understand how the radiant and dark modes of MQW polaritons evolve into the LT gap structure as N_w increases. An even more intriguing regime arises when d is much smaller than the exciton resonance wavelength, such that $kd \ll 1$ (where k is the resonance wavenumber), and N_w remains small enough to satisfy $N_w kd \lesssim 1$. In this so-called superradiant regime, theoretical models predict that the oscillator strength of a single radiant mode increases linearly with N_w , while all other modes remain dark [4]. As N_w increases further, additional radiant modes, specifically the second and third, begin to emerge sequentially. This regime is especially rich in physical phenomena and demands detailed experimental exploration.

However, experimental studies of MQW polaritons in this regime remain challenging. As noted in Ref. [4], the primary difficulty arises from the loss of optical coupling

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between excitons in different wells, a consequence of well-width fluctuations commonly found in typical QW structures. Specifically, in artificially fabricated QWs, d_w often varies due to imperfections in the fabrication process. Although narrow wells are favored to enhance the exciton oscillator strength, the confinement energy scales as $1/d_w^2$, making such systems highly sensitive to these fluctuations. When the exciton energies differ significantly between wells, the formation of MQW polaritons is inhibited.

To address these challenges, we utilize “natural QWs” instead of artificially engineered ones—specifically, 2D organic-inorganic perovskites with the general formula $(C_nH_{2n+1}NH_3)_2PbX_4$ ($X = I, Br, Cl$) [13–15]. These materials self-assemble into ideal QW structures without well-width fluctuations: the well layer comprises a single monolayer of corner-sharing $[PbX_6]^{4-}$ octahedra, with no regions containing two or more layers. Moreover, the dielectric constant of the barrier layer is significantly lower than that of the well layer, leading to the so-called image charge effect (ICE) [13–17], which results in strong exciton confinement within the well layers. These excitons exhibit remarkably large binding energies of 360–480 meV [15,17] and high oscillator strengths of 0.5–0.7 per formula unit [14]. Owing to these properties, the excitonic behavior of such materials has been widely studied [17–23], and they are of growing interest for light-emitting devices [24–26] and solar cell applications [27–29]. Additionally, the large bandgap offset between the well and barrier layers suppresses electronic coupling between wells [13–15], ensuring that only optical coupling occurs, an ideal condition for MQW polariton studies. Indeed, research related to MQW polaritons in 2D perovskites has been recently reported [30]. However, this pioneering work did not involve control of N_w , leaving detailed analyses of MQW-polariton modes still to be addressed.

In this study, we employed the Langmuir-Blodgett (LB) method to precisely control N_w in the 2D perovskite $(C_{18}H_{37}NH_3)_2PbI_4$ [31]. Because both d and the total film thickness are smaller than the exciton resonance wavelength, these samples are well suited for investigating MQW polariton effects in the regime where $N_w kd \lesssim 1$. We measured reflection spectra at various N_w values and compared the spectra obtained at 20 K and room temperature (RT) with those calculated using the transfer matrix method. The observed evolution of the reflection spectra with increasing N_w was well reproduced by the MQW-polariton model, providing the first clear experimental demonstration of MQW polaritons in a system with controlled N_w in the $N_w kd \lesssim 1$ regime. Furthermore, we examined the relationship between the spectral features and MQW-polariton modes.

Although a few studies have explored non-Bragg conditions [32–35], most previous experimental investigations of MQW polaritons have focused on the Bragg condition ($kd = \pi$) [36–44], where the effects are enhanced and manifest as photonic crystal features. In contrast, we successfully observed MQW polaritons in the $kd \ll 1$ regime with precisely controlled N_w . This was achieved by utilizing 2D perovskites, which naturally form QWs, and by adopting the LB method—rarely used in perovskite fabrication—to accurately control N_w . Our findings not only illuminate the optical properties that bridge the 2D and 3D regimes, but also

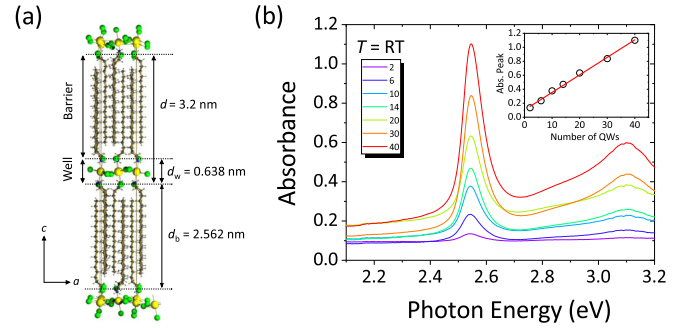


FIG. 1. (a) Sample structure of $(C_{18}H_{37}NH_3)_2PbI_4$ and (b) the QW number dependence of absorption spectra at RT. The inset shows the QW number dependence of absorbance peak at 2.54 eV.

advance the development of perovskite-based optoelectronic technologies.

II. EXPERIMENTAL

A film of the organic-inorganic 2D perovskite material $(C_{18}H_{37}NH_3)_2PbI_4$ was fabricated on quartz glass substrates using the LB method and intercalation techniques [31]. Octadecylamine (ODA) was deposited onto the substrate via the LB method, and samples with varying numbers of ODA layers were subsequently immersed in an aqueous PbI_2/HI solution for intercalation. Figure 1(a) illustrates the sample structure after intercalation. The final structure consisted of organic barrier layers with a width of $d_b = 2.562$ nm and inorganic well layers with a width of $d_w = 0.638$ nm. Further details of the sample preparation and fundamental properties have been reported in Ref. [31].

The number of QWs (N_w), i.e., the number of inorganic layers in each sample, was determined from the total film thickness, estimated via white-light interference using a Keyence VK-X3000 laser scanning microscope (Fig. S1 of the Supplemental Material [45]). The results confirm the successful fabrication of samples with different N_w , ranging from $N_w = 2$ to 40, with each sample exhibiting a layer-thickness fluctuation of approximately one layer (3.2 nm). Figure 1(b) presents the RT absorption spectra for seven samples with different N_w (see Fig. S2 of the Supplemental Material [45] for the spectra measured at 20 K). Exciton absorption peaks are clearly observed at 2.54 eV, with absorbance increasing linearly with N_w .

To investigate MQW-polariton effects, we acquired normal-incidence reflection spectra of each sample around the exciton resonance at 20 K and RT using a halogen lamp as the light source and examined the dependence of spectral features on N_w .

III. RESULTS AND DISCUSSION

Figure 2 shows the dependence of the reflection spectra on N_w at 20 K and RT. While some perovskite materials undergo structural phase transitions between RT and low temperatures [46,47], our sample does not exhibit such a transition (Fig. S3 of the Supplemental Material [45]), indicating that it is well suited for investigating excitonic properties. At 20 K,

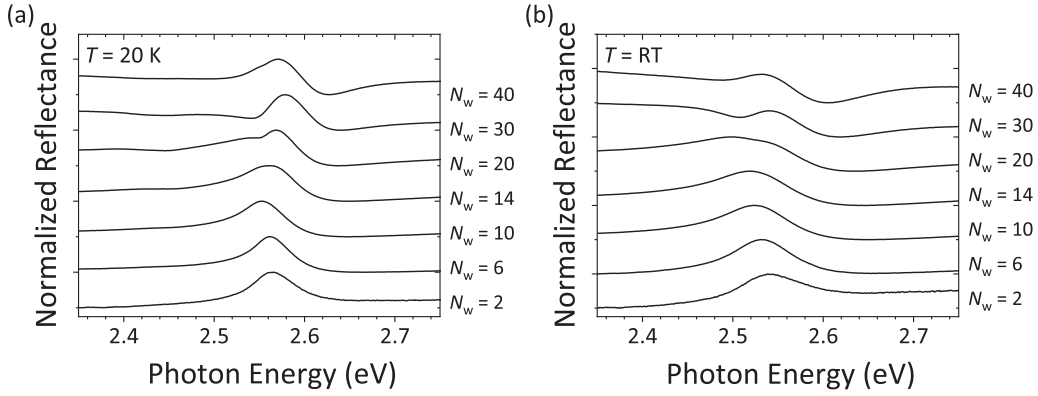


FIG. 2. Dependence of the reflection spectra on the number of QWs at (a) 20 K and (b) RT.

the spectrum for $N_w = 2$ exhibits a single peak at the exciton resonance energy of 2.57 eV. As N_w increases, the spectra broaden, and their overall structure changes. At RT, similar spectral changes with increasing N_w are also observed.

To determine whether the observed spectral dependence on N_w is attributable to MQW polaritons, we simulated the reflection spectra using the transfer matrix method [2,37]. The simulations were performed at normal incidence, taking into account light enters the sample from vacuum to replicate the actual experimental conditions. The equations used in the calculations are provided in the Supplemental Material [45]. Importantly, our model incorporates the quantum-well thickness (d_w) and the difference in background dielectric constants between the well and the barrier [37]. However, the overall trend in the reflection spectra remains essentially unchanged even when these factors are excluded (see Fig. S4 of the Supplemental Material [45]). The parameters used to calculate the reflection spectra are summarized in Table I. The exciton resonance energy ω_0 was determined from the peak position in the reflection spectrum for $N_w = 2$ at 20 K and RT. The background dielectric constants of the barriers (ϵ_b) and wells (ϵ_w) were adopted from Ref. [14], while the radiative relaxation rate in a single QW (Γ_0) and the nonradiative relaxation rate (γ) were adjusted to match the experimental results. The reflection spectra arising from MQW polaritons involve complex contributions from a total of N_w radiant and dark modes, yet the spectra can be reproduced using only two parameters, Γ_0 and γ . If a single set of two parameters successfully reproduces all experimental data, it confirms that the observed spectra originate from MQW polaritons.

Figure 3 shows the dependence of the reflection spectra on N_w , calculated using $\Gamma_0 = 1$ meV and $\gamma = 15$ meV (20 K) and 25 meV (RT). These reflectance spectra are averaged over ± 1

layer because of the one-layer fluctuation in our samples, as previously mentioned. To compare the experimental and calculated reflection spectra, we differentiated Figs. 2 and 3 with respect to photon energy. This differentiation clarifies the fine structure of the reflectance and reduces experimental artifacts, such as reflections from the cryostat window. Figure 4 shows the first derivative of the reflection spectra from Figs. 2 and 3. The N_w -dependent behavior of the experimental reflection spectra at 20 K and RT agrees well with the calculated spectra, confirming that MQW polaritons formed in our samples and affect the reflection spectra. These results demonstrate, for the first time, the successful observation of MQW polaritons in the intriguing regime where $kd \ll 1$ and $N_w kd \lesssim 1$, made possible by the use of natural QWs with precisely controlled N_w and by calculations specifically tailored to the experimental configuration. Notably, by increasing γ while keeping the temperature-independent Γ_0 constant, we also reproduced the RT spectra, indicating that the MQW-polariton effect persists at RT.

We examine whether the value of $\Gamma_0 = 1$ meV, which reproduces the experimental results well, is physically reasonable. Since Γ_0 is proportional to the oscillator strength per unit area [4], we estimated its value for our sample using reported values of relevant physical parameters (see the Supplemental Material for calculation details [45]). The estimated Γ_0 lies in the range of approximately 1–2 meV, supporting the conclusion that adopting $\Gamma_0 = 1$ meV is both reasonable and consistent with physical expectations.

To further examine the relationship between MQW-polariton modes and the spectral features, we calculated the MQW-polariton modes according to the method in Ref. [2]. For simplicity, these mode calculations approximate the well width as infinitesimally thin and assume identical background dielectric constants for the well and barrier. In contrast, the reflection spectra in Fig. 3 were calculated considering both the actual well thickness and dielectric properties. While absolute energy values cannot be directly compared, it is sufficient to discuss the relationship between MQW-polariton modes and reflection spectra.

Figure 5 shows the dependence of mode energies on N_w . The real part of the polariton mode energy represents the energy shift [Fig. 5(a)], while the imaginary part represents the radiative relaxation rate [Fig. 5(b)]. Modes with imaginary parts $> \Gamma_0$ are radiant modes, whereas those with imaginary

TABLE I. Parameters used in the calculation.

Exciton resonance energy, ω_0	2.57, 2.54 eV
Nonradiative relaxation rate, γ	15, 25 meV
Radiative relaxation rate in a single QW, Γ_0	1 meV
Background dielectric constant in the barrier, ϵ_b	2.1
Background dielectric constant in the well, ϵ_w	6.1
Width of barrier layers, d_b	2.562 nm
Width of well layers, d_w	0.638 nm

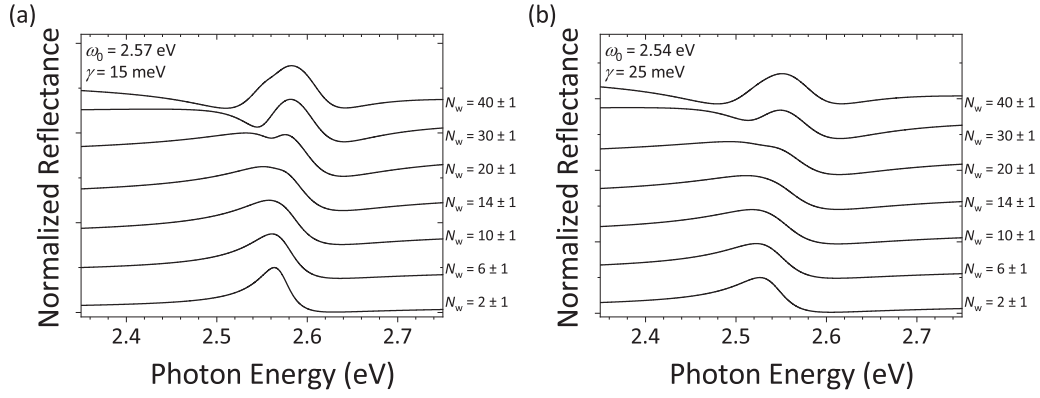


FIG. 3. QW-number dependence of calculated reflection spectra for (a) $\omega_0 = 2.57$ eV, $\gamma = 15$ meV and (b) $\omega_0 = 2.54$ eV, $\gamma = 25$ meV.

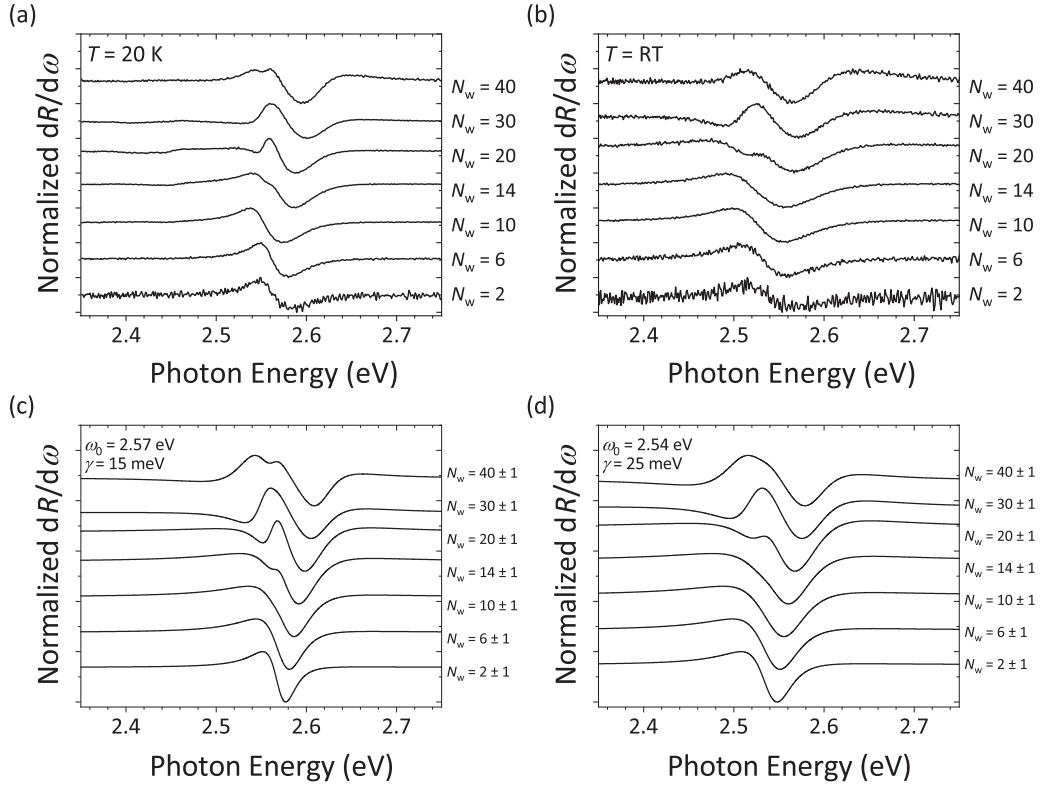


FIG. 4. Dependence of (a), (b) the experimental and (c), (d) the calculated reflection spectra on the number of QWs, differentiated from Figs. 2 and 3.

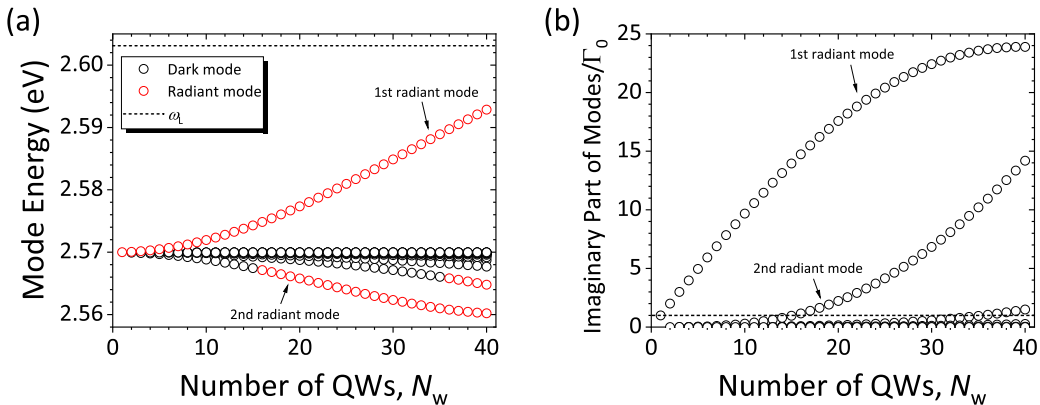


FIG. 5. QW-number dependence of the (a) real and (b) imaginary parts of MQW mode energies.

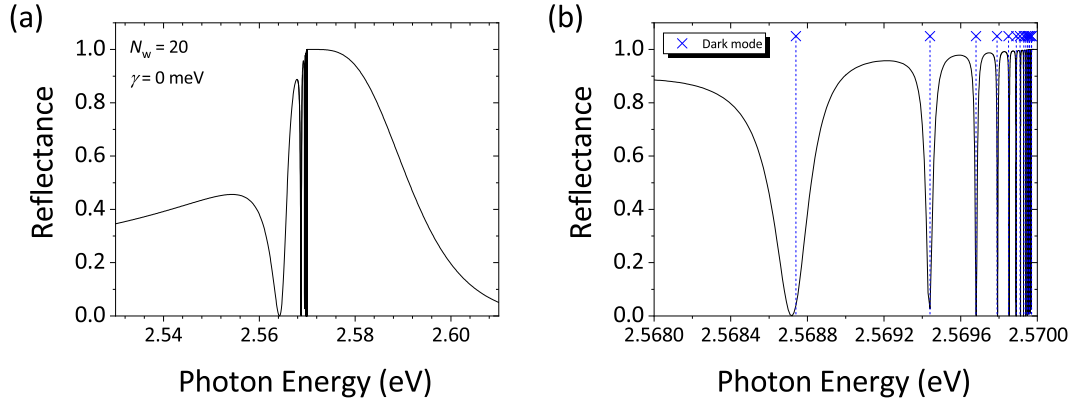


FIG. 6. (a) Calculated reflectance spectra and (b) enlarged view of the region near ω_0 for $N_w = 20$ and $\gamma = 0$ meV. The blue dotted lines are the energy position of dark modes.

parts $< \Gamma_0$ are dark modes. Although N_w -layer samples contain N_w polariton modes, most dark modes are not visible in Fig. 5 due to their small energy shifts and radiative relaxation rates. In fact, most dark modes have an imaginary part of less than $\Gamma_0/10^2$ (Fig. S5 of the Supplemental Material [45] shows the logarithmic scale). Figure 5(b) shows that the imaginary part of a single radiant mode grows proportionally to N_w until $N_w \approx 16$, indicating the superradiant regime [4]. At $N_w = 16$, a second radiant mode emerges on the low-energy side of ω_0 , leading to interference structures in the reflectance spectra for $N_w = 20$ (see Figs. 2–4). At $N_w = 36$, a third radiant mode appears, further complicating the reflection spectra for $N_w = 40$. These bright modes primarily determine the overall spectral structure. Thus, we identified the relationship between the radiant modes and spectral features, successfully confirming the intriguing sequential emergence of radiant modes and the corresponding spectral evolution.

Although the radiant modes mainly govern the spectra, we also discuss the behavior of dark modes. Since dark-mode effects are blurred by the nonradiative relaxation rate γ , we calculated the reflection spectrum for $\gamma = 0$. Figure 6 presents the reflection spectrum for $N_w = 20$ with $\gamma = 0$, omitting well thickness and dielectric constant differences for better correlation with MQW-polariton modes. (Figure S6 of the Supplemental Material [45] provides a relationship between reflection spectra and MQW modes with these factors included.) As shown in Fig. 6, in addition to the large dip near 2.565 eV caused by interference between two radiant modes, sharp dips appear near ω_0 . Figure 6(b) (enlarged view) reveals that these dips align precisely with dark mode energies. This unique feature of dark modes arises from their negligible interaction with electromagnetic waves. While these fine structures disappear as γ increases, dark modes play a crucial role in forming upper- and lower-branch polaritons for $N_w \gg 1$. The emergence of the LT gap as N_w increases, bridging to the 3D polariton, is shown in Fig. S7 of the Supplemental Material [45].

It should be noted that, even for $N_w = 40$, the high-energy shift of the first radiant mode does not reach the longitudinal frequency ω_L in the 3D case. The frequency ω_L marks the onset of the upper polariton branch, and is given by $\omega_L = \omega_{LT}^{MQW} + \omega_0$. Using $\omega_{LT}^{MQW} = 2\Gamma_0/kd$ [4,10], an approximation that neglects the well width, we find that the

effective LT gap energy leads to $\omega_{LT}^{MQW} = 33$ meV. When the well width and dielectric constant are taken into account [Fig. S7(b) of the Supplemental Material [45]], ω_{LT}^{MQW} increases to approximately 47 meV. These estimated LT gap energy values are consistent with those previously reported for single crystals of $(C_nH_{2n+1}NH_3)_2PbI_4$ ($n = 4-10$) [14]. In Fig. 5(a), the energy position of ω_L is shown for the case where $\omega_{LT}^{MQW} = 33$ meV. As clearly seen, within the N_w range examined in this study, the shift of the first radiant mode does not reach ω_L . This indicates that the system remains in a regime where N_w is still significantly smaller than the number required to form the LT gap.

We now discuss the impact of inhomogeneous broadening. At low temperatures, the phase relaxation time for this class of materials is approximately 0.2 ps [48], corresponding to a homogeneous broadening of $\gamma \approx 3.9$ meV. However, the value $\gamma = 15$ meV used to reproduce the experimental results at 15 K is significantly larger, indicating the presence of inhomogeneous broadening. One possible origin of this broadening is in-plane fluctuations of the ICE [13,15,16]. Disorder in the organic layers induces fluctuations in the dielectric environment experienced by excitons in the well, which may lead to inhomogeneous broadening. Thus, the value $\gamma = 15$ meV can be interpreted as effectively incorporating the effects of inhomogeneous broadening. Although this value greatly exceeds $\Gamma_0 = 1$ meV, it is known that the exciton-photon coupling strength is unaffected by inhomogeneous broadening [49], meaning that MQW polariton effects can still emerge. For these effects to become clearly visible despite the broadening, the condition $N_w\Gamma_0 > \gamma$ must be satisfied, i.e., $N_w > 15$. In our case, $N_w = 15$ corresponds approximately to the emergence of the second radiant mode, which was successfully observed as changes in the reflection spectra. Interestingly, even at RT, where inhomogeneous broadening is more pronounced and $\gamma = 25$ meV was used in the simulations, distinct spectral features still begin to appear from $N_w = 20$. To explore this phenomenon further, time-resolved spectroscopy could be a promising next step, as it provides enhanced sensitivity to inhomogeneous broadening [7,34,35]. Nonetheless, what is particularly meaningful in this study is the clear identification of MQW polaritons using simple reflection spectroscopy.

Finally, we compare our results with MQW polaritons in traditional inorganic MQW structures. In high-quality GaAs-based MQWs, extremely narrow inhomogeneous broadening on the order of 0.1 meV has been achieved, and MQW-polariton effects under the Bragg condition—i.e., observation of a single superradiant mode—have been reported [39–41]. In contrast, we observe MQW polaritons not at the Bragg condition but in the superradiant regime with $kd \ll 1$. Although the inhomogeneous broadening in our samples is roughly two orders of magnitude larger than that in these inorganic MQWs, the radiative relaxation rate Γ_0 is also larger by about one to two orders of magnitude, reflecting the large oscillator strength of excitons in 2D perovskites. Consequently, by increasing N_w , we have successfully observed clear MQW-polariton effects. This result is particularly significant, as it highlights the unique role of 2D perovskite excitons in enabling access to regimes of MQW-polariton physics.

IV. SUMMARY

We successfully observed MQW polaritons in the low layer number region of organic-inorganic 2D films with precisely controlled N_w . Using the LB method, we fabricated high-quality MQW structures that eliminate well-width fluctuations, making them ideal for studying MQW polaritons.

Our optical measurements and transfer matrix calculations confirm the correspondence between spectral structures and MQW-polariton modes, revealing the superradiant regime and the emergence of higher-order radiant modes as N_w increases. Furthermore, we confirmed that MQW-polariton effects persist even at RT. These results suggest that MQW polaritons not only are of fundamental interest in bridging 2D and 3D exciton-polariton physics but also hold potential for practical applications in perovskite-based optical and photonic devices. Moreover, our findings contribute to the broader exploration of ultimate 2D systems, where MQW effects have recently been observed even in optical phonons [50].

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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