

Optical bistability and laserlike emission in a semiconductor microcavity

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We report the experimental observation of hysteresis in the optical response of a semiconductor microcavity under normal mode coupling regime. A third resonance mode, spectrally lying at the bare cavity mode frequency, suddenly appears in the reflectivity and resonant Rayleigh scattering spectra when increasing the excitation power, resulting in a coexistence of three resonance modes. Bistability loops in both microcavity optical response are found at the energy of this third mode. Associated to the bistability, laserlike emission occurs at the bare cavity mode.

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I. INTRODUCTION

Bistability is a very general phenomenon involving a phase transition far from thermal equilibrium, which can be retrieved in many different fields of physics. Generally speaking, a bistable system has an output that, within a range of inputs, can assume two different values depending on the history of the system. Optical bistability (OB) requires that both the input and the output are optical signals. Therefore, in order to obtain OB, light and matter must be closely coupled together in a very intriguing way and this explains the large interest devoted to this topic. It can be shown that OB arises from the combination of nonlinear media and positive feedback mechanisms.¹ These requirements are achieved in Fabry-Perot (FP) interferometers filled with non-linear materials: The cavity field, which is enhanced by the resonance condition, strongly depends on the dielectric function of the medium inside the cavity producing the optical feedback.¹⁻⁷ Since the pioneering works in the 1970s,²⁻⁴ many different nonlinear mechanisms in FP systems have been addressed, such as absorption saturation,^{2,3} optical Kerr effect,^{4,5} and soliton formation⁷ by using gases,⁴ liquid crystals,⁷ or semiconductor material.^{5,6} All the above mentioned experimental observations have concerned the weak coupling (WC) regime, where the light-matter interaction inside the optical cavity can be considered as a small perturbation and the photon and matter modes are not mixed.

In the last decade great interest has been devoted to a very peculiar FP system, that is, quantum well semiconductor microcavity (MC) where high quality quantum well (QW) is embedded in high finesse FP cavity tuned at the QW exciton absorption. The sharpness of the exciton and cavity modes results in the normal mode (NM) coupling (often referred to as strong coupling). NM coupling means that the exciton mode and the cavity photon mode are in such close interaction to give rise to two mixed polariton states. The anticrossing behavior and the Rabi splitting between the upper and lower polaritons has been observed by Weisbuch *et al.*⁸ and then reported in a number of different systems and optical experiments.⁹ Among many interesting peculiarities of the NM-MC, in the last years particular attention has been devoted to the nonlinear properties. For strong carrier injection,

a rich assortment of optical non-linear effects has been so far reported.⁹⁻²⁴ On one side the attention was devoted to the nonlinearity associated to the reduction of the NM splitting when increasing the excitation power, producing the collapse of the polariton modes at high intensity and the crossover between NM and WC regime.¹⁰⁻¹² These effects have been interpreted in terms of bleaching of the exciton absorption¹⁰ and excitation induced dephasing¹¹ due to exciton-exciton interaction. Lately, it has been shown that the transition between NM and WC regimes can be much more complicated and that three modes can coexist in the optical response of the MC.^{13,14} The simultaneous presence of the two NMs and a third WC-like mode in single beam and pump-probe transmission experiments has been attributed to quantum correlation of the electron-hole polarization.¹³ More recently the coexistence of NM emission and lasing at the WC-like mode has been explained in terms of bound excitons.¹⁴ On the other side great interest has been raised by the peculiar properties of the nonlinear emission from NM-MC.¹²⁻¹⁹ Stimulated emission, giant amplification, and lasing, above some threshold in the external power have been observed both in nonresonant¹²⁻¹⁷ and in resonant excitation conditions with very peculiar angular resonance.¹⁸ Several mechanisms have been debated in order to interpret the experimental data,^{15,16} and finally the picture of parametric polariton amplifier has been largely accepted.¹⁹ As a matter of fact, the relevant role of the bosonic character of the MC polaritons has driven a strong effort for the realization of polariton condensation²⁰ and polariton lasers.²¹ Recently the relevance of other mechanisms, such as polariton collisional broadening,²² electron-electron,²³ and electron-polariton scattering²⁴ have been pointed out to be effective in limiting or even in improving the polariton condensation.

Following the history of the FP systems in the WC regime, this large variety of nonlinearities in NM-MC is, *a priori*, expected to produce different kinds of OBs. However, such a relevant aspect of the physics of NM-MC has not yet fully addressed. OB was observed in the early studies where the MC optical quality was not high enough to reach the NM coupling.^{5,6} There are few theoretical papers predicting optical bistability associated to the bleaching of the excitonic absorption.^{25,26} Only recently OB in NM-MC has been experimentally observed in the reflected intensity of the lower

polariton and it has been interpreted in terms of Kerr-like nonlinearity associated to the polariton-polariton interaction.²⁷

We report an experimental study of the optical nonlinearities of a NM-MC, by means of reflectivity, resonant Rayleigh scattering (RRS), and photoluminescence (PL) measurements, demonstrating the presence of a bistable response under resonant excitation conditions. The bistability occurs at the energy of the bare cavity mode and therefore it has to be related to a different nonlinear mechanism with respect to the findings of Ref. 27. The simultaneous presence of three modes is found above the bistability threshold, that is the two NMs and the WC mode coexist in the MC optical response. The comparison of the reflectivity with resonant secondary emission allows us to extract further physical information on the nonlinear mechanisms driving the OB. In particular, laserlike emission at the WC mode is observed at the bistability threshold. We discuss the experimental findings on the basis of the existing theoretical models for the MC nonlinearities.

The paper is organized as follows. In Sec. II we describe the sample and the experimental details. In Sec. III the experimental results are presented and discussed. Section IV contains the summary and concluding remarks.

II. SAMPLE AND EXPERIMENTAL DETAILS

The sample consists in a single 8.5 nm $\text{In}_{0.04}\text{Ga}_{0.96}\text{As}$ quantum well (QW) embedded in a λ GaAs cavity with 14 (16.5) pairs of $\lambda/4$ GaAs/AlAs layers for the top (bottom) distributed Bragg mirrors. The flux asymmetry during the epitaxial growth has given rise to a wedged MC structure with a rate of variation of the cavity modes along the direction of the gradient of the cavity thickness of the order of 11 meV/mm.

A tunable cw Ti-sapphire laser is used as excitation source, focused with a 15-cm lens to a spot of about 40 μm diameter. Different external incidence angles θ in the range $\theta=0^\circ-9^\circ$ with respect to the normal have been used and in all cases the RRS and the reflectivity spectra have been simultaneously detected. The emission at normal incidence has been also investigated by means of a flat field spectrometer coupled to a cooled Si-(charge-coupled device) camera. We used a computer controlled $\lambda/2$ plate, placed in front of a polarization analyzer, in order to finely tune the excitation power density. For the measurements with 150 fs pulsed excitation Ti-sapphire laser has been also used. Spatially resolved measurements have been performed by reimaging the laser spot on a 20 μm pinhole after magnification by a factor of 2. The sample was mounted in a He bath cryostat and, in all the measurements reported here the temperature was $T=1.8$ K. In the following the resonance condition on the MC will be described by $\delta(\theta)=E_{\text{cav}}(\theta)-E_{\text{exc}}$ where E_{exc} and $E_{\text{cav}}(\theta)$ are the energies of the QW exciton mode and the bare cavity mode at the excitation angle (θ), respectively. Note that, in order to stress the exciton-photon resonance condition at the experimentally used excitation angle θ , the detuning $\delta(\theta)$ is calculated by taking into account the dependence of $E_{\text{cav}}(\theta)$ on the excitation angle. A study of the optical characterization of the investigated MC has been reported

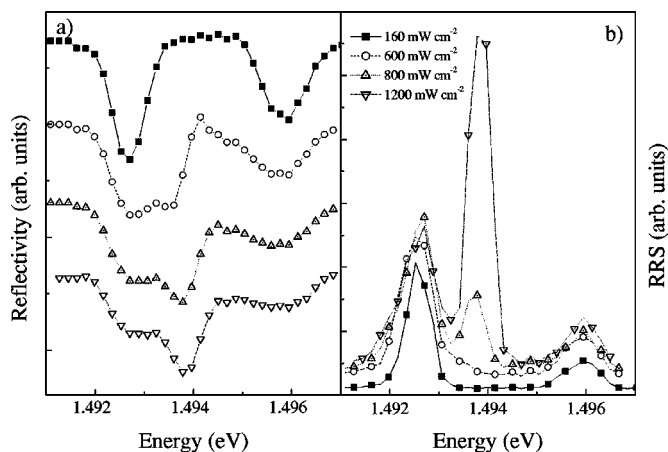


FIG. 1. (a) Reflectivity and (b) RRS spectra for different P_{exc} under cw excitation.

elsewhere.²⁸ Preliminary results on the OB have been reported in recent conference proceedings.²⁹

III. RESULTS AND DISCUSSION

Let us first describe the main features of the observed MC nonlinear response. We report in Fig. 1 the reflectivity and the RRS spectra obtained by tuning the cw laser at different excitation power densities, $\theta=4^\circ$ and $\delta(4^\circ)=0$. At low excitation power P_{exc} , NM coupling regime is found with the two polariton resonances separated by a Rabi splitting of 3.3 meV. Increasing the excitation power the spectra are strongly modified. First the two NMs broaden, due to excitation induced dephasing, and their separation slightly reduces. Then a third peak, spectrally lying in between the two normal modes, suddenly appears. Finally, for very high excitation power only the third peak survives in the MC optical spectra just in the middle of the two normal modes, corresponding to the resonance of the bare cavity. The main features of this phenomenology are well known in the literature as due to a transition from NM to WC regime. This transition has been studied by changing the excitation density, the temperature, or applying external electric fields.⁹⁻¹² Reducing the exciton oscillator strength, the field-matter interaction weakens and eventually the two NMs collapse to the uncoupled cavity mode. However, for intermediate power, we observe the simultaneous presence in the spectra of the two polariton modes and of the WC-like mode. Measurements of the reflected beam after spatial filtering by a pinhole to select only the central part show that, in this power range, the presence of three resonances is not due to the spatial inhomogeneity.

The coexistence of three modes in the optical spectra of the MC cannot be directly inferred by cw measurement, since the modes are not simultaneously excited. In order to clarify the interplay between these modes we have also performed reflectivity measurements with fs laser pulses. The reflected beam has been spatially resolved to avoid spatial inhomogeneity. Results are reported in Fig. 2 for different excitation powers and $\theta=0$. On top of the laser pulse the MC resonances are clearly resolved (see also the inset). The two

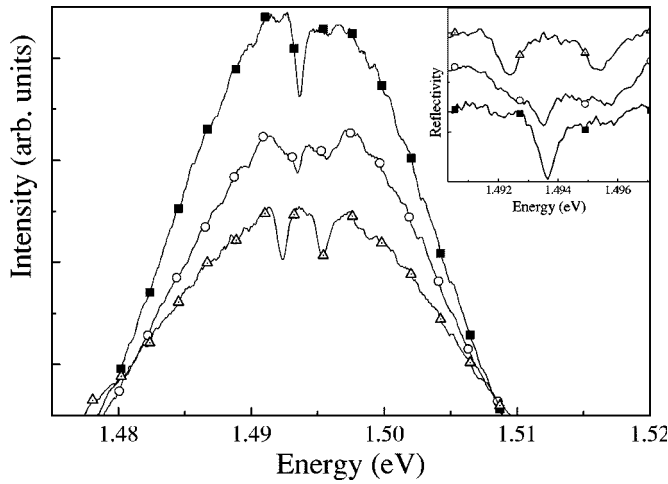


FIG. 2. Reflected intensity for different P_{exc} under fs pulse excitation. The inset shows the reflectivity spectrum obtained by dividing the experimental data by a Gaussian profile of the pulse spectrum.

NMs found at low excitation power collapse in the WC mode at the highest intensity. However for intermediated excitation power the simultaneous coexistence of three mode is observed. Similar findings were found in transmission spectra in Refs. 13; we will discuss them later on.

In order to observe bistability, we have carefully measured the power dependence of the reflected and RRS signals for different fixed laser energies and different angles of incidence θ . No bistability behavior is observed at the energy of the NM resonances, contrary to the findings of Ref. 27. Instead when exciting at the energy of the third peak clear OB loops are observed. The reflected and RRS signals, for resonant excitation at the energy of the third peak and for three different external angles of incidence, are reported in Fig. 3, for both increasing and decreasing power. Hysteresis loops are observed in all the cases. It is worth stressing that the previous findings in OB (Refs. 4–7 and 27) were based on the observation of the primary emission (reflected or transmitted beam); here we show that OB can be monitored also in the resonant secondary emission. The jumps in the optical response occur at a higher external power, when increasing P_{exc} , with respect to the case in which P_{exc} is decreased. For external power in between the two thresholds, the optical response of the MC is bistable and depends on the history of the MC photo injection. Comparing the hysteresis loops, we note that, as far as the angular dependence is concerned, the most relevant experimental observations are that both the OB threshold and the width of the OB loop decrease with increasing the incidence angle.

The jumps in the optical response of the MC are associated to the sudden appearance of the third resonance. This is strikingly demonstrated by the analysis of the reflectivity and

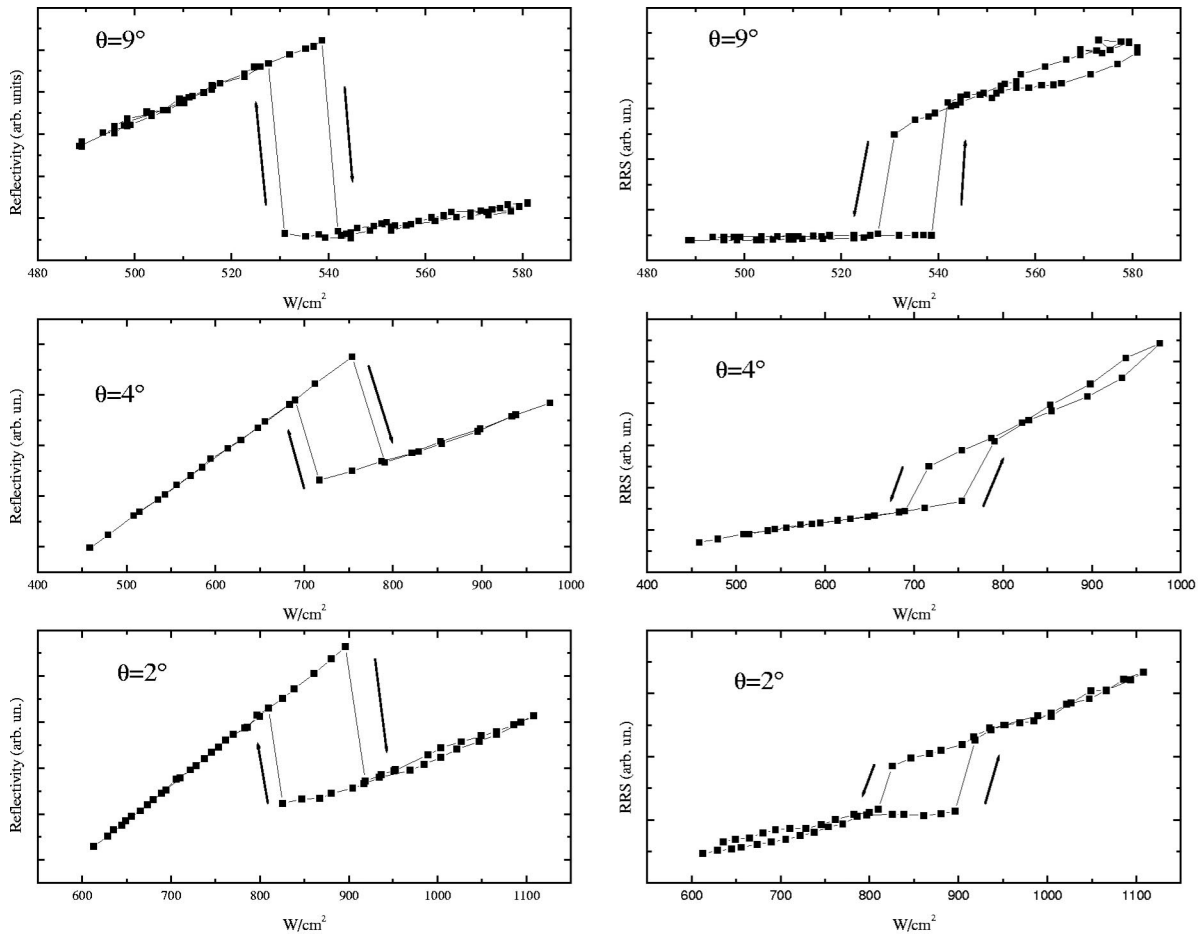


FIG. 3. Bistability loops of the reflected and RRS signals measured at the third peak for three different angles of incidence θ .

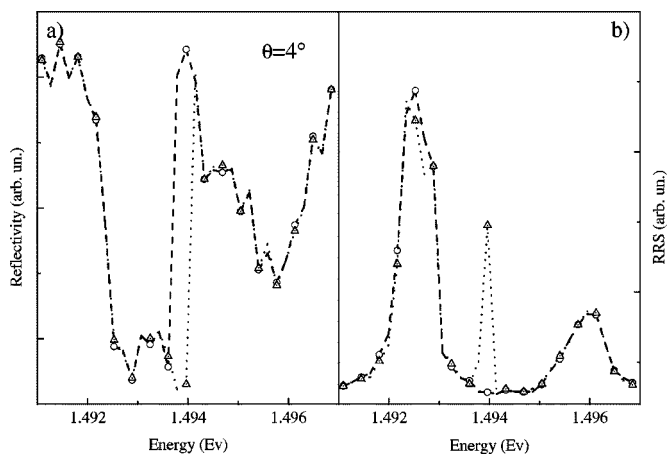


FIG. 4. (a) Reflectivity and (b) RRS spectra measured at $P_{\text{exc}}=750 \text{ W cm}^{-2}$ and $\theta=4^\circ$ angle of incidence. Different symbols refer to the two different bistable responses.

RRS spectra. Fixing P_{exc} between the two bistability thresholds we find that the third peak is randomly present in the spectra. The bistable reflectivity and RRS spectra obtained at the same excitation power are reported in Figs. 4(a) and 4(b), respectively. In both cases the two spectra are almost identical except for the appearance of the third resonance. In fact, when scanning the laser frequency for performing the spectra, the history of the MC photo injection is not well defined and the two MC bistable states are therefore randomly observed. The two bistable states of the NM-MC are the presence or the lack of the WC peak in the optical response. It is worth noticing that the sudden decrease of the reflected power is associated to a strong increase of the RRS signal. In other words, part of the photons that enter into the cavity, as a consequence of the photoinduced transparency of the MC associated to the OB threshold, are elastically scattered into different directions by the structural disorder inside the MC. This involves dephasing-free scattering mechanisms which are in competition with exciton absorption. From a phenomenological point of view, this also means that the RRS becomes strongly nonlinear with respect to the external excitation power. Finally note that the appearance in the RRS spectra of a well separated third peak spectrally lying between the two NMs is much more clear with respect to reflectivity spectra, due to the lack of background in RRS measurement.

We have also investigated the NM-MC emission at normal incidence at the onset of the OB when using an excitation angle of $\theta=8^\circ$. A white light from a halogen lamp has been used to probe the reflectivity spectrum at normal incidence ($\theta=0$) exactly at the same spot on the sample. Comparison between the reflectivity spectrum obtained by scanning the laser wavelength ($\theta=8^\circ$) and the reflectivity spectrum of the white light (normal incidence) is reported in Fig. 5(a), where the arrows indicate the energy of the bare cavity modes in the two cases. The resonance condition was $\delta(8^\circ)=0$ for the excitation at $\theta=8^\circ$ which corresponds to $\delta(0)=-1.6 \text{ meV}$ at normal incidence. The inset reports (in semilogarithmic scale) the NM-MC emission spectrum. Both the lower polariton (1.4918 eV) and the upper polariton

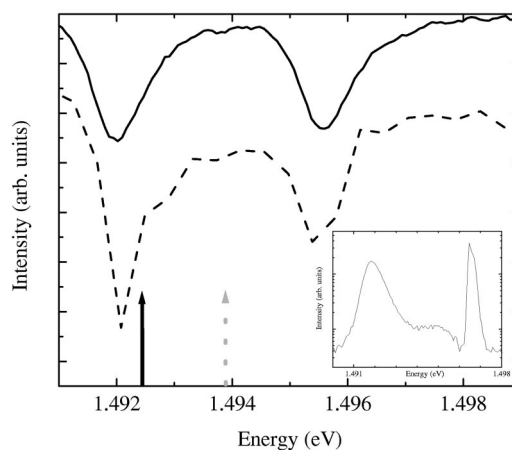


FIG. 5. Comparison between reflectivity at normal incidence (solid line) and at $\theta=8^\circ$ angle of incidence (dashed line). The solid (dotted) arrow shows the energy of the WC mode at normal ($\theta=8^\circ$) incidence. The inset reports the PL emission at normal incidence after laser excitation at 1.4965 eV and $\theta=8^\circ$.

(1.4952 eV) modes are observed in the emission spectrum, while the sharp peak at 1.4965 eV is the elastic scattering of the laser excitation.

The MC emission spectrum is dramatically modified at the onset of the OB. This is shown in Fig. 6 where emission spectra are reported for three different excitation power. Here the laser is tuned at 1.4938 eV, corresponding to the WC mode with 8° angle of incidence, which is the energy at which OB is observed. The lowest P_{exc} in Fig. 6 is just below the threshold of the OB ($P_{\text{exc}}=535 \text{ W cm}^{-2}$) and the spectrum only shows the emission at the NMs and the Rayleigh scattering from the laser. The next spectrum is taken with an excitation power just above the OB threshold ($P_{\text{exc}}=550 \text{ W cm}^{-2}$) and, in fact, the intensity of the laser peak is almost unchanged. However the emission spectrum is

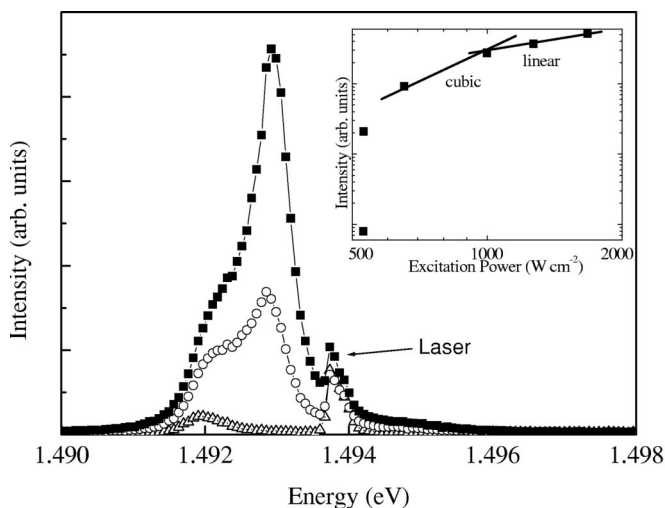


FIG. 6. Emission spectra at normal incidence after laser excitation with $\theta=8^\circ$ angle of incidence at 1.4938 eV (WC mode energy at 8°) for three different excitation powers: 545 W cm^{-2} (triangles), 552 W cm^{-2} (circles), and 680 W cm^{-2} (squares), respectively. The inset shows the power dependence of the WC emission.

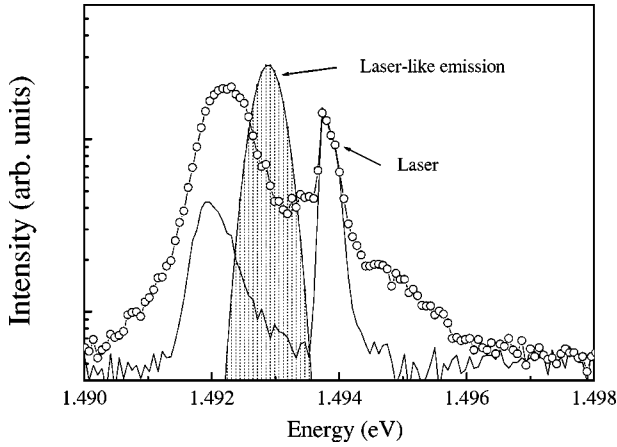


FIG. 7. Emission spectra at normal incidence after laser excitation with $\theta=8^\circ$ angle of incidence at 1.4938 eV (WC mode energy at 8°). Solid line corresponds to 550 W cm^{-2} (just below the bistability threshold). The experimental data above the threshold (552 W cm^{-2}) are reported by showing the Gaussian fit (dashed region) to the laserlike emission and the spectrum (dots) obtained by subtracting this fit to the experimental data.

completely different, with the presence of a third peak between the two NMs. The last spectrum corresponds to $P_{\text{exc}}=680 \text{ W cm}^{-2}$ and here the third peak largely dominates the MC emission. In the inset of Fig. 6 the excitation power dependence of this emission is reported. At the OB threshold the emission jumps abruptly, denoting a strong stimulated emission associated to the observed bistability in the MC optical response. Then it increases nonlinearly and tends to saturate to a linear power dependence around 1 KW cm^{-2} . The peak energy of this emission slightly shifts increasing the excitation power and saturates at the WC mode energy for normal incidence. Let us note that similar sudden jumps of the MC emission are commonly denominated in the literature as laserlike emission¹⁰ or lasing action;¹⁴ in the following we also will use this terminology for a better comparison with previous findings.

A further important information is that the coexistence of the NMs and WC mode is retrieved in the emission spectra. This is more clearly shown in Fig. 7 where the two spectra just below and just above the OB onset are reported in semilogarithmic scale. The spectrum above the threshold has been decomposed in the stimulated emission and in the NM-PL, the latter obtained by subtracting a Gaussian fit of the stimulated emission from the experimental spectrum. Comparing the NM-PL before and after the threshold, we find that even the emission at the NM is strongly enhanced above the threshold. This clearly discards the possibility that the presence of a Gaussian laser profile plays a relevant role in the coexistence of the three modes. We also conclude that the appearance of the third peak in the MC emission spectrum produces an efficient injection at the NMs.

Let us now discuss the data presented. The OB here reported is certainly of a different kind with respect to the recent findings in Ref. 27. In Ref. 27 the hysteresis was observed at the lower polariton mode, which undergoes an

energy shift towards negative cavity-exciton detuning at high optical pumping. This effect was explained in terms of polariton Kerr-like nonlinearity within a fully polaritonic picture. A similar picture cannot be applied to our data because we find OB at the WC mode. More generally we believe that the appearance of the third peak is behind any polaritonic model. It is also worth stressing that different kinds of OB are expected in NM-MC, depending on which nonlinear mechanism dominates under the experimental excitation conditions and MC parameters (i.e., Rabi splitting, mode broadenings, etc.). As already stated previously, in our case the general features of the observed non-linearity possess some similarity to the transition from normal mode to weak coupling, reported in the literature in a number of different experimental conditions.⁹⁻¹² A theoretical model for OB in NM-MC associated to this transition was reported few years ago in Ref. 25 and therefore we compare our data with those predictions. In Ref. 25 OB was predicted to occur on the wing of the two polariton modes, assuming that the exciton absorption α saturates, for increasing excitonic density N_{exc} , following the relationship:

$$\alpha(N_{\text{exc}}) = \alpha_o / (1 + N_{\text{exc}}/N_s), \quad (1)$$

where α_o is the unsaturated exciton absorption and N_s is the saturation density, which is of the order of the inverse of the exciton Bohr area. This simple saturation formula, which is due to both phase space filling and exchange effects,^{30,31} is commonly accepted to nicely reproduce the exciton bleaching in bulk and confined semiconductor structures.^{30,31} Within this assumption, the OB is predicted only on the wing of the polariton resonances and only in the case of quite sharp NM resonances.²⁵ Considering realistic broadenings in order to reproduce the experimental spectra of our NM-MC, we find that the model of Ref. 25 does not predict any bistability. In any case, this model does not predict a bistable behavior at the WC mode even in case of very small broadenings and therefore does not agree at all with our experimental data.

As a matter of fact the model of Ref. 25 predicts a soft OB. This result, not even commented in Ref. 25, is surprising since the saturation of the absorption is well known to give OB exactly at the bare cavity mode in atomiclike systems³ and also MC in WC regime.^{5,6} We believe that the reason for this discrepancy is related to the fact that Eq. (1) gives rise to a quite smooth nonlinearity of the excitonic absorption as a function of the internal field intensity I . Assuming steady state excitation, it follows that $N_{\text{exc}} = \alpha(N_{\text{exc}})I\tau$, with τ the exciton recombination time and $\alpha(N_{\text{exc}})$ given by Eq. (1). Then it is quite simple to show that the dependence of the exciton absorption on the field intensity I is given by $\alpha(I) = 2\alpha_o / [1 + (1 + 8I/I_s)^{0.5}]$, where $I_s = 2N_s / (\alpha_o\tau)$. On the contrary, in the case of old results for WC-MCs,^{3,5} the theoretical description used for the absorption saturation was the derived from a two level system, which gives

$$\alpha(I) = \alpha_o / (1 + I/I_s). \quad (2)$$

This formula, which produces (for $I > I_s$) a stronger nonlinearity than the one associated with Eq. (1), describes the

fact that the bleaching of the atomic transition does not occur when most of the atoms are in the excited state, as required by the Pauli exclusion principle, but instead when absorption and stimulated emission balance. A similar requirement is not usually included for the case of exciton bleaching in semiconductor structures. Assuming Eq. (2) instead of Eq. (1) within the same approximation used in Ref. 25 we have found that OB occurs at the WC mode for realistic values of the broadening of the exciton and cavity modes. We also remark that our experimental observation of a strong stimulated emission at the bistability onset suggests that the “atomiclike” Eq. (2) can be more appropriate than the standard Eq. (1) for exciton bleaching in the case of NM-MC. However, simple models for exciton bleaching, both using Eqs. (1) and (2), predict a collapse of the two normal modes into the WC mode and not the coexistence of three resonances in the optical response of the MC.

Before addressing this key aspect of our experimental findings, let us discuss the previous observations on laserlike emission in NM-MC. Stimulated polariton scattering, a clear signature of the bosonic character of the polariton, has been observed for resonant excitation at the “magic angles,” where the phase matching conditions are fulfilled.^{18,19} Within these conditions, strong nonlinear emission and parametric polariton amplification have been reported.¹⁸ These observations have suggested to exploiting the possibility of polariton laser²⁰ and even polariton Bose-Einstein condensation.²¹ At the same time a fully bosonic picture of the MC polariton is questionable at very high density where the fermionic nature of the electron and hole forming the exciton should play a role.¹⁶ For example it was found that, at least for GaAs based MC and nonresonant excitation, the onset of the optical gain corresponds to transition from strong to weak coupling regime.¹² This result is strictly linked to the physics of vertical cavity surface emitting lasers where the emission of QW excitons is enhanced by the WC resonance with the cavity mode, but NM coupling is not reached. More recently laserlike emission at the WC cavity mode without the collapse of the NM has been observed,¹⁴ under nonresonant excitation condition. Note also that at high excitation power, the NM to WC transition was eventually observed in Ref. 14. The coexistence of low threshold lasing at the WC mode and NM emission has been attributed, in Ref. 14, to the presence of low density localized states. This localized excitons are assumed to be weakly coupled to the cavity mode and to be optically inverted at high excitation, leading to the appearance of stimulated emission.¹⁴

The findings in Ref. 14 concerning the lasing at the WC, the coexistence of three modes and the final transition to WC regime are, at a first glance, quite similar to ours, even if they use nonresonant excitation and do not observe OB. Nevertheless, in the reports of Ref. 14, the threshold is much smoother with respect to our observation, possibly due to the lack of OB in case of nonresonant excitation. In addition we observe that NM emission is enhanced at the OB threshold, which is not present in the findings in Ref. 14. A final and very important difference is the fact that we do observe the third peak also in the reflectivity spectrum demonstrating a

strong modification of the dielectric response of the NM-MC at the OB and laserlike threshold. We believe that this observation rules out the possibility of using the extrinsic picture of localized exciton proposed in Ref. 14 to explain our results. This data also shows the relevance of measuring and comparing reflectivity and secondary emission under the same experimental condition.

The simultaneous presence of the normal modes and the WC-like resonance in the intrinsic optical response of a NM-MC has been reported in Ref. 13 by means single beam transmission spectra and pump and probe experiments. When increasing the excitation power, besides the two NMs, a third peak emerged in the spectrum and grew with intensity. The third peak showed a WC character and, at high excitation intensity, the spectra collapsed in a single peak denoting the transition to WC regime. The observation was explained within a fully quantum theory based on a fermionic picture, leading to intraband correlation between different photon modes. The experimental data of Ref. 13 concerning the observation of the coexistence of NMs and WC mode in the intrinsic optical response of the MC are in qualitative agreement with our findings. In addition the enhancement of the NM-PL at the threshold for the appearance, reported in Fig. 7, of the third peak denotes a coupling between different cavity modes, which is at the basis of the theory of Ref. 13. Unfortunately the theoretical model reported in Ref. 13 is quite complicated and it is not simple to guess if OB and stimulated emission can be attributed to this non-linearity.

IV. CONCLUSIONS

We have reported a detailed analysis of optical nonlinearity in NM-MC. Four are the main features of our experimental results: (i) the simultaneous presence of NMs and WC mode in the MC optical response, including reflectivity; (ii) OB at the WC mode; (iii) stimulated emission at the WC mode simultaneously to the OB onset; and (iv) enhancement of the NM emission at the WC laserlike threshold. Most of these aspects can be referred to previous findings and possibly attributed to existing theoretical predictions. The simultaneous presence of the NMs and WC mode is fully explained within the model presented in Ref. 13. The coupling between different photon modes arising from quantum correlation predicted by the theory of Ref. 13 could also account for the enhancement of the NM emission at the threshold. OB could be possibly inferred to the exciton bleaching assuming that Eq. (2) describes, in agreement with the very simple atomiclike models,³ the absorption saturation. The stimulated emission at the WC mode without the collapse of the NMs agrees with the findings of Ref. 14 and following their interpretation it could be attributed to the inversion of a population of localized exciton states. Obviously a theory that includes all points (i) to (iv) in a unified picture is needed for a comprehensive understanding and this is a challenging task for theoreticians. As a matter of fact the variety of nonlinear effects in NM-MC is very rich but also quite complicated and puzzling. A very large assortment of experimental results exists so far in the literature and

even though many explanations have been given, several interesting and challenging problems still wait for final answers. We definitely hope that our experimental results will trigger further theoretical efforts to produce a better understanding of the very fascinating world of nonlinear effects in NM-MCs.

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