# Negatively Charged Quantum Well Polaritons in a GaAs/AlAs Microcavity: An Analog of Atoms in a Cavity

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The negatively charged exciton ($X^-$) is observed to strongly couple with the microcavity- (MC-)confined photons in a GaAs quantum well containing a two-dimensional electron gas with $0 < n_e \lesssim 3 \times 10^{10}$ cm$^{-2}$. This strong coupling results in a formation of charged polaritons. The coupling strength is found to depend on $\sqrt{n_e}$, in analogy to two-level atoms in a cavity. The analysis of the reflection and photoluminescence spectra shows that $X^-$ is strongly admixed with the neutral exciton via their coupling with the MC photons. The linewidth dependence on $n_e$ indicates that electron-polariton scattering is effective.

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Frequently, an analogy is made between the spectroscopic properties of quantum well (QW) excitons that are strongly coupled to microcavity- (MC-)confined photons and those of atoms coupled to confined photons in a metal cavity [1–3]. The reason is that both systems exhibit a normal mode (Rabi) splitting that can be described by the interaction of a two-level electronic oscillator at resonance with the confined photons [2–4]. However, there are basic differences between the atomic and semiconductor systems. While the coupling strength of two-level atoms can be increased by increasing their number in the cavity [1], the excitons coupling strength is determined by the density of QW unit cells and thus has a fixed value (under low exciton densities) [5]. Increasing the exciton density results only in an increased scattering rate and reduced oscillator strength. Consequently, a bleaching of the mixed modes is observed (under high exciton densities), indicating that the QW-exciton–MC-photon system transforms from the strong to the weak coupling regime [6]. It is noted, though, that varying the coupling strength of the QW-exciton–MC mode can be achieved by applying *external* electric and magnetic fields, since these vary the exciton dipole moment [4]. Also, the coupling strength can be increased by increasing the number of QW's embedded in the MC [4]. Finally, while atoms move freely in the cavity, excitons are QW elementary excitations that have an in-plane dispersion. This leads to dispersive exciton-MC coupled modes: the cavity polaritons [7]. Since these polaritons are electrically neutral, they cannot be transported in the QW plane, in contrast to ionized atoms.

It is known that in the presence of a low density two-dimensional electron gas (2DEG) ($0 < n_e < 5 \times 10^{10}$ cm$^{-2}$), a new elementary excitation appears in the optical spectra of GaAs QW's, namely, the negatively charged exciton $[X^- = (e1{:}hh1)1S + e1]$ [8,9]. $X^-$ is a bound complex of one heavy hole and two conduction electrons that coexists with the neutral $(e1{:}hh1)1S$ exciton ($X$), and is the semiconductor analog of the negative hydrogen ion. Its binding energy is $\sim$1.2 meV with respect to $X$ (in QW's with a width of $\sim$200 Å). The $X^-$ optical properties depend on $n_e$. Thus, if $X^-$ can interact strongly with the MC photons, it is expected that such a coupling will differ from the $X$–MC-photon coupling by being dependent on $n_e$ and that charged MC polaritons will be formed.

In this Letter we show that in photoexcited GaAs/AlAs QW's, embedded in a MC, $X^-$ is strongly coupled to the MC mode with an interaction strength that increases with $n_e$, in analogy to atoms in a cavity. We also show that since the $X$ and $X^-$ excitons are close in energy, they are strongly admixed via their interaction with the MC mode, leading to mixed modes of neutral and charged polaritons. The observed $X^-$–MC-mode coupling dependence on $n_e$ suggests that the cavity polaritons involving the $X^-$ have a large in-plane coherence length, similar to the $X$-cavity polaritons [5].

The MC structure used in this study was grown by molecular beam epitaxy on a (001)-oriented GaAs substrate. It consists of an $Al_{0.1}Ga_{0.9}As$ $\lambda$-wide cavity with 15/25 periods of AlAs/$Al_{0.1}Ga_{0.9}As$ distributed Bragg reflectors (DBR) on the top and bottom sides, respectively. Embedded in the MC center is a mixed type-I–type-II QW structure (MTQW) [10] consisting of a single (102 Å)/(200 Å)/(102 Å) AlAs/GaAs/AlAs wide QW, clad on both sides by 26 Å/102 Å GaAs/AlAs narrow QW's. The MTQW structure is particularly suitable for this study since it has the following property: under laser photoexcitation with an energy below the narrow QW band gap energy, $E_{L1} < E_g(e1\text{-}hh1)_N$ (with intensity $I_{L1}$), only excitons are generated in the wide QW. However, for a laser energy $E_{L2} > E_g(e1\text{-}hh1)_N$, a 2DEG is generated in the wide QW (and a separate 2D hole gas in the narrow well). The 2DEG density is controlled with the laser intensity $I_{L2}$, and it varies approximately

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as $n_e(I_{L2}) \approx 6 \times 10^9 I_{L2}$ (mW/cm$^2$), where $I_{L2}$ is the intensity estimated to reach the MTQW [11].

Reflection and photoluminescence (PL) spectra were taken at $T = 6$ K, in the backscattering geometry and under excitation with a Ti-sapphire laser ($E_{L1} = 1.62$ eV) and with a He-Ne laser ($E_{L2} = 1.96$ eV). In this study, $n_e$ was varied in the range of $0 < n_e \leq 3 \times 10^{10}$ cm$^{-2}$. The spectra were monitored at different illumination spot positions on the sample surface. This resulted in different MC-mode detuning energies, $\delta = E_C - E_X$, where $E_C$ is the empty MC-mode energy and $E_X$ is the bare $X$ energy.

Figure 1(a) presents a series of PL spectra that were excited with $I_{L1} \approx 50$ mW/cm$^2$ and $I_{L2} \approx 0.5$ mW/cm$^2$ (corresponding to $n_e \approx 3 \times 10^9$ cm$^{-2}$), observed for various $\delta$ values, and are each normalized to its peak intensity. Only the low energy part is shown but the inset shows the entire PL spectrum, for $\delta = +3$ meV, and the notation used here for the four observed lines. The energies of the uncoupled neutral exciton $X$ and of the negatively charged exciton $X^-$ are marked in Fig. 1(a). For $\delta \gg 0$ the energy difference between the $L$ and $LM$ lines, $\Delta_{L,LM} \approx 1$ meV, is about equal to the $X^-$ binding energy that is measured in a separate sample with bare GaAs/AlAs MTQW's [11]. Upon increasing $n_e$ (by increasing $I_{L2}$) the intensity ratio $I_X/I_{X^-}$ decreases and line $L$ broadens, as is shown in Fig. 1(b). This dependence on $I_{L2}$ was observed for $X$ and $X^-$ in bare MTQW's [11]. Therefore, for $\delta \gg 0$, far from resonance with the MC mode, the $LM$ line is identified to be predominantly $X$-like and the $L$ line to be $X^-$-like. We note that since $X$ is neutral, it interacts weakly with the low density 2DEG while $X^-$ is charged and is more efficiently scattered by electrons. The peak energies of the four PL lines as a function of $\delta$ are shown in Fig. 2(a). For $\delta \gg 0$, the $H$ and $HM$ lines are predominantly MC-like mode and $X_{lh}[\equiv(e1{:}lh1)1S]$-like mode, respectively. As $\delta$ decreases, *three* anticrossings are observed corresponding to resonances of the MC mode with the $X_{lh}$, $X$, and $X^-$ excitons.

Figure 3 presents on-resonance ($\delta \sim 0$) reflection spectra. For $I_{L2} = 0$, only three lines are observed—a spectrum that is typical of undoped GaAs QW's embedded in a MC [4]. Upon increasing $I_{L2}$ [Figs. 3(b)–3(f)], a new line appears on the *high-energy side* of the lower mode. $\Delta_{L,LM}$ increases with increasing $I_{L2}$ and this reflects the increase in the $X^-$–MC-mode coupling strength as $n_e$ increases (see below). Furthermore, the linewidths' increase shows that all the mixed modes (around $\delta \sim 0$) are effectively scattered by the *free* electrons.

In order to analyze these reflection spectra, the interaction between the three QW excitons and the MC mode was treated by the linear dispersion model (LDM) [12] using a standard transfer matrix method [13]. The optical response of the wide QW was modeled by three damped Lorentzian oscillators that represent the three types of QW excitons ($X_{lh}, X, X^-$). The calculation included the residual, below band gap absorption of the $Al_{0.1}Ga_{0.9}As$ layers [14]. The energies and linewidths of the different oscillators were extracted from the off-resonance reflection spectra. The coupling strengths of $X$ and $X_{lh}$ with the MC mode were extracted from the reflection spectra with $I_{L2} = 0$. From the fitting of the reflection spectra with $I_{L2} > 0$, we find that the $X$ and $X_{lh}$ coupling strengths are not affected by the presence of a low density 2DEG. The calculated reflection fit well the experimental ones, as shown in Fig. 3, and the difference between them is in the increase of the $X^-$–MC-mode coupling strength and in the damping factors.

The experimental PL energy dependence on $\delta$ of the four cavity polaritons (with $k_\parallel \approx 0$) [Fig. 2(a)] is analyzed by diagonalizing the excitons–MC-photon Hamiltonian [15]:

$$H = \begin{bmatrix} (E_C - i\gamma_C) & V_{lh} & V_X & V_{X^-} \\ V_{lh}^* & (E_{lh} - i\gamma_{lh}) & 0 & 0 \\ V_X^* & 0 & (E_X - i\gamma_X) & 0 \\ V_{X^-}^* & 0 & 0 & (E_{X^-} - i\gamma_{X^-}) \end{bmatrix}. \tag{1}$$

Here $E_C(\gamma_C)$, $E_{lh}(\gamma_{lh})$, $E_X(\gamma_X)$, and $E_{X^-}(\gamma_{X^-})$ are the energies (linewidths) of the bare MC mode, $X_{lh}$, $X$, and $X^-$ excitons, respectively. The parameters for this calculation were obtained from the PL spectra in the same manner as those obtained in fitting the reflection spectra. The calculated energies are shown by solid lines in Fig. 2(a). Good agreement with the experimental energies (for $I_{L2} = 0.5$ mW/cm$^2$) is obtained using $2V_{X^-} = 0.9 \pm 0.3$ meV. This is compared to the $X$–MC-photon coupling strength: $2V_X \cong 3.8$ meV. Similar values are also obtained from the fit of the corresponding reflection spectrum.

The wave function ($\Psi_n$) of each of the four mixed modes is an admixture of the four bare components,

$$|\Psi_n\rangle = \alpha_{n,C}|C\rangle + \alpha_{n,lh}|X_{lh}\rangle + \alpha_{n,X}|X\rangle + \alpha_{n,X^-}|X^-\rangle, \tag{2}$$

where $n = H, HM, LM,$ and $L$ is the mixed mode index. $|\alpha_{n,C}|^2$, $|\alpha_{n,lh}|^2$, $|\alpha_{n,X}|^2$, and $|\alpha_{n,X^-}|^2$ are the MC-photon, $X_{lh}$, $X$, and $X^-$ fractions in the $n$th mixed mode wave function. Figures 2(b)–2(e) show these calculated fractions as a function of $\delta$ for each mixed mode. The $H$ and $HM$ modes' dependence on $\delta$ is similar to that of the mixed MC-$X_{lh}$ mode, as observed for GaAs QW's in a MC [4,16], with the $HM$-mode turning into a mixed MC-$X$ mode for $\delta < +2$ meV. The two lowest modes are different. As

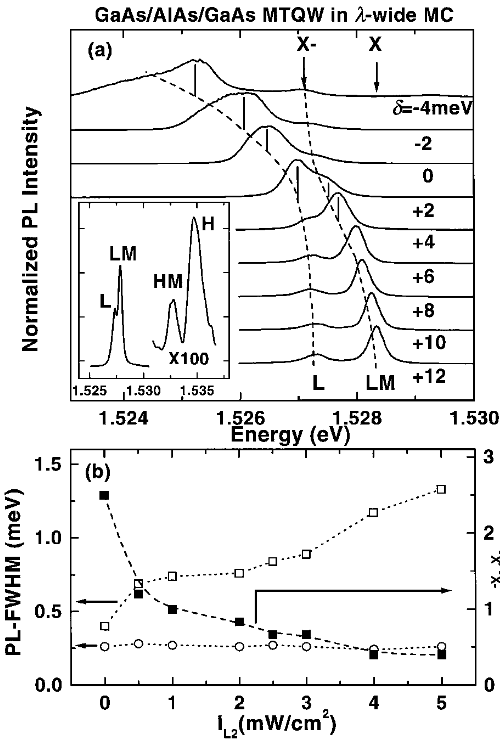


FIG. 1. (a) Photoluminescence spectra excited with $I_{L1} \approx 50\ \mathrm{mW/cm^2}$ and $I_{L2} \approx 0.5\ \mathrm{mW/cm^2}$ and monitored at various points on the sample (various detuning energies $\delta$). The down-pointing arrows show the bare $X$ and $X^-$ energies. The dashed lines are guides to the eye (with bars indicating the peak energies). The inset shows the full PL spectrum. (b) Linewidth (open circles: LM mode; open squares: L mode) and $X/X^-$ intensities ratio (full squares) as a function of $I_{L2}$, measured for $\delta = +3$ meV.

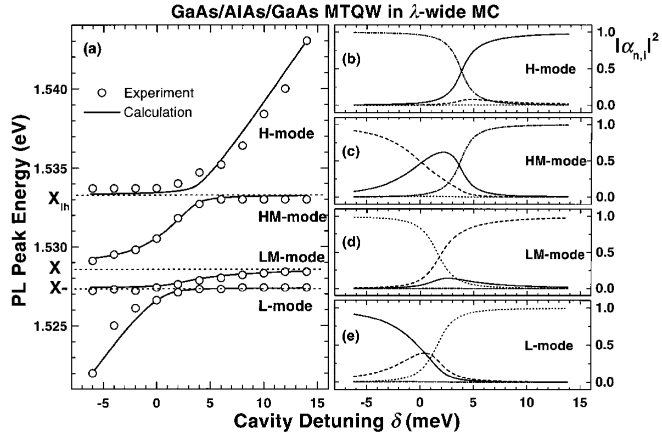


FIG. 2. (a) Peak energies of the PL lines as a function of the detuning energy measured for excitation intensities $I_{L1} \approx 50\ \mathrm{mW/cm^2}$ and $I_{L2} \approx 0.5\ \mathrm{mW/cm^2}$. The solid lines are the calculated energies of the mixed modes (denoted by $L$, $LM$, $HM$, and $H$). The horizontal dotted lines mark the bare exciton energies. The right panels (b)–(e) present the relative fractions of each of the bare modes: MC-photon (solid lines), $X_{lh}$ (dash-dotted lines), $X$ (dashed lines), $X^-$ (dotted lines) in the mixed polariton modes.

mentioned above, for $\delta \gg 0$, the $LM$ mode is $X$-like and the $L$ mode is $X^-$-like. As $\delta$ is decreased, the two modes show a mixing of the bare components. For $\delta \cong 2$ meV, the $L$ mode is an almost equal admixture of the MC photon with $X$ and $X^-$ excitons (with a slightly higher fraction of $X$). The $LM$ mode, on the other hand, is an almost pure admixture of $X$ and $X^-$ with only a small fraction of the MC photon. *This is then a strong admixture of two elementary electronic excitations that are very different in nature:* $X$ is neutral and bosonlike, $X^-$ is charged and fermionlike. This admixture is induced by their strong interaction with the MC photon and is enhanced by the small energy difference between $X$ and $X^-$, as compared to their coupling strengths with the MC mode. For $\delta \cong 0$, the $LM$ mode has a higher fraction of $X^-$ than the $L$ mode has. $L$ is then mostly a mixture of $X$ and MC photon. This explains the observation (Fig. 3) that at resonance, the $LM$ line appears on the *high energy side* of the $L$ line (while for a bare QW, $X^-$ is *below* $X$).

We now analyze, in two *independent* ways, the dependence of $V_{X^-}$ on $I_{L2}$ as extracted from the reflection spectra [e.g., Figs. 3(a)–3(f)]: (1) Using the coupling Hamiltonian [Eq. (1)], we calculate the $\Delta_{L,LM}$ dependence on $V_{X^-}$ [inset, Fig. 4(a)]. This allows us to obtain, from the measured $\Delta_{L,LM}$ of Fig. 4(a), the dependence of $V_{X^-}$ on $I_{L2}$ [circles in Fig. 4(b)]. Note that the error bar for $I_{L2} = 0$ indicates that there is always a residual density of electrons in the MTQW [17]. The $2V_{X^-}(I_{L2})$ dependence is fitted to the expression $2V_{X^-} = \beta\sqrt{I_{L2}}$, where $\beta$ is a fitting parameter $[\beta = 0.92 \pm 0.06\ \mathrm{meV/(mW/cm^2)^{1/2}}]$.

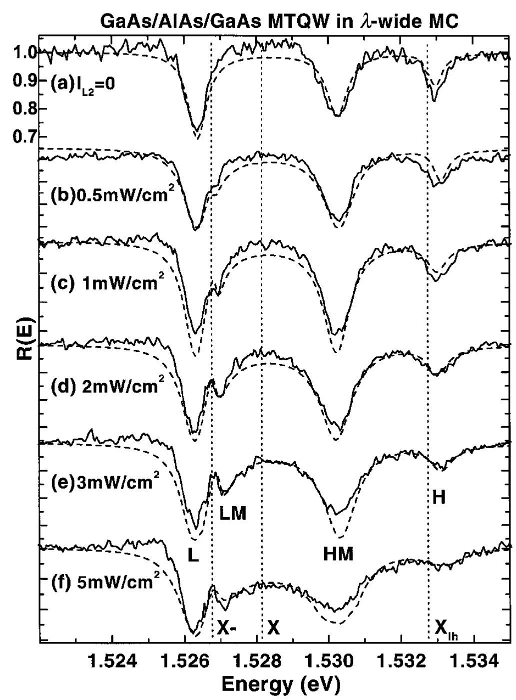


FIG. 3. Reflection spectra measured for $\delta \sim 0$ and for several laser intensities ($I_{L2}$) that generate a 2DEG with $0 < n_e \le 3 \times 10^{10}\ \mathrm{cm^{-2}}$. Solid lines: experimental; dashed lines: calculated spectra. The vertical dotted lines mark the bare exciton energies.

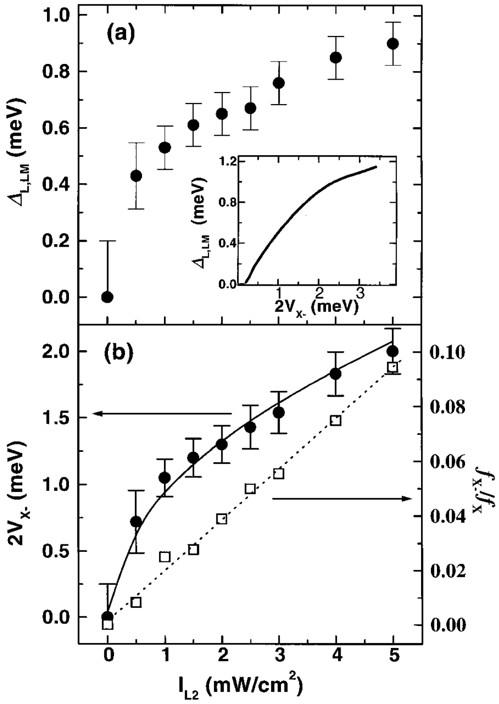


FIG. 4. (a) The energy splitting between the $L$ and $LM$ modes measured for $\delta \sim 0$ and for $I_{L2}$ values that generate a 2DEG density in the range $0 < n_e \leq 3 \times 10^{10}$ cm$^{-2}$. The inset shows the calculated energy splitting as a function of the $X^-$–MC-mode coupling strength [using Eq. (1)]. (b) The circles are the $V_{X^-}$ values extracted from the measured and the calculated $\Delta_{L,LM}$ values of Fig. 4(a). The solid line is the fitted $\beta I_{L2}^{1/2}$ dependence with $\beta = 0.92$ meV/(mW/cm$^2$)$^{1/2}$. The squares are the ratio between the oscillator strength (per unit area) of the charged and neutral excitons as obtained from the linear dispersion model fitting of the reflection spectra (Fig. 3).

(2) The $X^-$ oscillator strengths per unit area $(f_{X^-})$ that were previously extracted from the LDM fitting are plotted for each value of $I_{L2}$ in Fig. 4(b), and normalized to the constant $X$ oscillator strength $(f_X)$. Clearly, $f_{X^-} \propto I_{L2}$, and since $V_{X^-} \propto \sqrt{f_{X^-}}$ [12,18], we observe again that $V_{X^-} \propto \sqrt{I_{L2}}$. Since $n_e \propto I_{L2}$, we conclude that $V_{X^-} \propto \sqrt{n_e}$. This relation is in a complete analogy with that observed for $N \gg 1$ atoms in a metallic cavity, where the strength of the collective coupling to the confined photon is proportional to $\sqrt{N}$.

In order to explain this result, we assume that *all* the electrons in the 2DEG take an equally active role in the $X^-$ creation. Then, the possible number of equivalent $X^-$ excitations per unit area is determined by the density of the 2DEG. In an analogous description of two-level atoms, if all these $X^-$ excitations are coherent, the single-excitation coupling strength is enhanced by a factor of $\sqrt{n_e}$. Using a solid state description, one views the $X^-$ as an oscillator composed of a hole and two electrons. Then, since the MC photon can create only an electron-hole pair, the other $X^-$ electron must come from the free electron gas. Hence, the density of such oscillators is determined by $n_e$. Since the oscillator strength $f_{X^-}$ is proportional to the density of the oscillators, i.e., $f_{X^-} \propto n_e$, and since $V_{X^-} \propto \sqrt{f_{X^-}}$, we find that $V_{X^-} \propto \sqrt{n_e}$.

In summary, negatively charged excitons in GaAs QW's that contain a low density 2DEG are observed to strongly couple to the MC-confined photons, thus forming *charged polariton* modes. An analysis of the reflection and the PL spectra reveals several distinct properties of the mixed excitons–MC-photon modes: (a) $X^-$ is coupled to the MC mode with $V_{X^-} \propto \sqrt{n_e}$, while the neutral exciton coupling strength is independent of its density. (b) The neutral and charged excitons are strongly admixed by their interaction with the MC mode. (c) The square root dependence of $X^-$–MC-mode coupling on $n_e$ provides an experimental evidence to the large in-plane coherence length of the resulting polaritons, so that they are delocalized over a distribution of many electrons. (d) The mixed modes are effectively scattered by *free* electrons.

It is interesting to note that the formation of charged polaritons might be detected by polaritonic transport under an applied dc or low frequency electric field. This would constitute an electric charge motion with very high velocities due to the very small polariton mass [4].

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