Investigation of the Spectral Triplet in Strongly Coupled Quantum Dot–Nanocavity System

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We experimentally investigated the excitation power dependence of a strongly coupled quantum dot (QD)–photonic crystal nanocavity system by photoluminescence measurements. At a low excitation power regime, we observed a vacuum Rabi doublet emission at the QD–cavity resonance condition. With increasing excitation power, in addition to the doublet, a third emission peak appeared. This observed spectral change is unexpected from conventional atomic cavity quantum electrodynamics. The observations can be attributed to featured pumping processes in the semiconductor QD–cavity system.

SOLID-STATE cavity quantum electrodynamics (QED), based on semiconductor quantum dots (QDs), has been extensively studied as a key tool for quantum information processing.\textsuperscript{1–3} In these studies, single QDs are often considered as atomic two-level systems.\textsuperscript{4} However, recent experiments on coupled QD–cavity systems\textsuperscript{5–8} have reported several strange phenomena unexpected from conventional atomic cavity QED. One of the major oddities is the so-called non-resonant coupling, which describes strong photon feeding into the cavity mode from the QDs with large spectral detuning from the cavity resonance. Another peculiarity is triplet emission in the strong coupling regime at the resonance condition where vacuum Rabi doublet emission is expected. Both peculiar observations were first reported by Hennessy \textit{et al.}\textsuperscript{5} and considerable efforts have been made to understand them.

With regard to the mystery of non-resonant coupling, several groups have been investigating the mechanisms both experimentally\textsuperscript{6,7,9} and theoretically.\textsuperscript{10–12} In contrast, detailed experimental studies of the spectral triplet have not been conducted and there are only a few theoretical studies.\textsuperscript{12,13} Also, the effect of pumping processes on strongly coupled QD–cavity systems is little known even though photoluminescence (PL) measurements, in which collective carriers are injected around and inside the QDs, are a major experimental tool in semiconductor cavity QED. A theoretical model\textsuperscript{14,15} considering the effect of incoherent pumping on both the QD and the cavity mode has recently been introduced and applied to explain the pumping power dependence of the vacuum Rabi doublet emission;\textsuperscript{16} however, the spectral triplet was outside its scope. Deeper understanding of the peculiar observations is necessary for developing QD-based cavity QED systems for wide application in quantum information technology.

In this paper, we studied the excitation power dependence of a strongly coupled QD–cavity system in the resonance condition by micro-PL measurements. With increasing excitation power, a transition from vacuum Rabi doublet to triplet emission was observed. Quantum correlations of the emitted photons were also investigated and the degradation of the quantum nature along with the appearance of the third emission peak was observed. The spectral triplet is attributed to featured pumping processes in semiconductor cavity QED systems, including collective carrier injection inside the host material and incoherent cavity photon pumping by background oscillators.

The investigated sample was grown on a (100)-oriented GaAs substrate by molecular beam epitaxy. First, InAs QDs were grown on an 80-nm-thick GaAs layer on top of a 700-nm-thick $\text{Al}_{0.6}\text{Ga}_{0.4}\text{As}$ sacrificial layer. The QD density was $\sim 4 \times 10^{10}$ cm$^{-2}$. Then a partially covered island growth technique was applied to the QDs to obtain blue-shifted emission.\textsuperscript{17} Finally, the layer containing the QDs was buried in the middle of 160-nm-thick GaAs slab layer.

A two-dimensional air-bridged photonic crystal (PhC) was fabricated by forming a triangular array of air holes using a combination of electron beam lithography and etching processes. The lattice constant was $a$ (240 nm) and the radius was $r$ (0.26$a$). The cavity design employed here is a symmetrically modified H1, which is an extension of our previous work,\textsuperscript{18} and is shown in Fig. 1(a). The radius of the nearest-neighbor holes around the defect were shrunk to 0.23$a$ and shifted outward by $s_1 = 0.14a$. The second and third nearest-neighbor holes along the $\Gamma K$ direction and the first nearest holes along the $\Gamma M$ direction were also shifted by $s_2 = 0.025a$, $s_3 = 0.12a$, and $s_4 = 0.03a$, respectively. The average number of QDs inside each cavity is estimated to be 0.4.

Micro-PL measurements were performed at $3.3\;\text{K}$ with a temperature-controlled liquid helium cryostat. The excita-

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\includegraphics[width=0.5\textwidth]{Fig1.png}
\caption{(a) Schematic illustration of investigated H1-type photonic crystal nanocavity. (b) Observed spectra under various QD–cavity detuning $\delta$ at 3.3\,K.}
\end{figure}
tion source was a pulsed Ti:sapphire laser (80 MHz, 2 ps duration) oscillating at 848 nm, where the laser light is predominantly absorbed and creates carriers in the host material (GaAs). The excitation laser was focused onto the sample surface with a spot size of ~3 µm using a microscope objective (40×, NA = 0.6). The PL signal was collected using the same objective lens and sent to a 0.75 m grating spectrometer equipped with a cooled charge coupled device (spectral resolution = 0.016 nm) after passing through a half-wave plate and linear polarizer to effectively detect linearly polarized cavity resonant mode emission. To control the wavelength of the cavity mode, we employed a nitrogen gas deposition technique.\(^{19}\) For photon auto-/cross-correlation measurements, we used a Hanbury–Brown–Twiss setup consisting of a pair of single photon counters located after monochrometers serving as bandpass filters.

Figure 1(b) shows a series of micro-PL spectra of the investigated coupled QD–cavity system at various detuning \(\delta\) from -0.3 to 0.3 nm. The excitation power was 0.6 µW (measured before the objective lens). Each spectrum is normalized so that its integrated PL intensity is equal to unity. The observed cavity mode (C) originated from one of the fundamental dipole-like modes. The quality factor of the mode was 11,400 (116 µeV), measured with a high pumping power of 25 µW at \(\delta = -0.6\) nm. The other dipole-like mode, split due to fabrication errors, was at 5 nm shorter than C (not shown). Throughout the cavity scan, the two emission peaks could be seen and they exhibited anti-crossing, a signature of strong coupling. The strong coupling behavior is also confirmed by mixing of the line widths and intensities near resonance. From the smallest splitting, a vacuum Rabi splitting of ~194 µeV was extracted.

Next, we studied the pumping power dependence of the strongly coupled QD–cavity system at \(\delta \sim 0\). Figure 2(a) shows normalized micro-PL spectra under various pumping powers. At the lowest excitation power of 0.05 µW, the dominant contribution from QD–cavity polariton emission, indicated by blue and red curves, can be seen. With increasing pumping power, an additional emission peak, indicated by the green curve, became prominent and showed dominant emission at a high pumping power of 3 µW. The transition from doublet to triplet emission was not expected from the conventional atomic cavity QED model. This is because strongly coupled two-level atom–cavity systems at resonance do not possess their eigenstates at the bare cavity frequency even with incoherent cavity pumping coming from other sources.\(^{15}\)

The three panels in Fig. 2(b) show the integrated PL intensities (top), peak positions (middle), and linewidths (bottom) of the three peaks of the QD–cavity system at resonance under various pumping powers. These properties were extracted by fitting the spectra with three Lorentzian curves. We assumed a fixed cavity position for fitting the data taken with excitation powers below 0.8 µW. The polariton intensities show linear increase under weak excitation (~0.4 µW) and saturation at higher pumping powers, which is the same behavior as single QDs without strong coupling with a cavity.\(^{20}\) The intensity of the third emission peak also increases linearly below a power of 0.4 µW and forms an s-shaped curve around the polariton saturation. We consider that this anomalous power dependence does not arise from lasing because the characteristic linewidth behavior of a laser is not present. The linewidths of the third peak remain close to the intrinsic linewidth of the cavity, ~116 µeV, at any pumping power. This fact leads us to the same conclusion as Hennessy et al.\(^{5}\) that the third emission peak arises from the bare cavity mode. The observed agreement of the polarization between the third emission peak and the bare cavity mode also supports this conclusion. Moreover, the closeness of the measured linewidth values to that of the bare cavity suggests that absorption by oscillators inside the cavity, including the resonant QD, is weakened or off when photon emission from the third peak occurs. Thus, it is appropriate to consider that at that time, the QD is driven to what we call pumped states,\(^{21}\) which are all-inclusive excitonic states other than the cavity-resonant one. The pumped states are formed by capturing or releasing processes in the QD and have weak or no interactions with the cavity mode due to the formation of dark states or sufficient spectral shifts. Thus, when the QD is in the pumped states, the third peak can appear. We mention the invariance of the peak positions and linewidths of the polariton modes against increasing pumping power, which implies that there are few pump-induced decoherence effects\(^{22}\) in our sample. We also note that pulsed excitation is not essential. Under continuous-wave laser pumping at the same wavelength, the transition from the doublet to the triplet was also observed in a similar manner.

To understand photon sources of the third peak, we measured the quantum correlation between photons emitted from the QD–cavity system. We measured the second-order intensity correlation function for the sum of the three emission peaks \(g^2(\tau) = \langle I(t)I(t + \tau)\rangle/I(t)^2\), where \(I(t)\) is the emission intensity of the photons at time \(t\). In particular, we paid attention to \(g^2(0)\), which is obtained by normalizing the counts in the peak with zero delay (\(\tau = 0\)) to the averaged
counts of the other peaks. An example of the measured coincidence histogram is shown in the inset of Fig. 3. The histogram was measured with band-pass filtering centered at 930.2 nm (bandwidth 0.25 nm). The obtained histogram was measured with band-pass filtering centered at 930.2 nm (bandwidth 0.25 nm). The obtained histogram was measured with band-pass filtering centered at 930.2 nm (bandwidth 0.25 nm). The obtained histogram was measured with band-pass filtering centered at 930.2 nm (bandwidth 0.25 nm). The obtained histogram was measured with band-pass filtering centered at 930.2 nm (bandwidth 0.25 nm).

In summary, we have experimentally investigated a strongly coupled QD–cavity system and observed a transition from doublet to triplet emission along with an increase in the excitation power. The third emission peak is assigned to the bare cavity mode excited by background oscillators with Poissonian photon statistics. We provide a possible explanation for the observations by a combination of the pumped QD state and incoherent cavity pumping, which are essential characteristics of semiconductor quantum dot systems. We consider that the origin of the cavity pumping is photon feeding from the non-resonantly coupled excited states instead of background oscillators. We believe that this effect is minor for our case, because we could not find other prominent excitonic emission peaks around the target QD.

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We also mention that our explanation based on a combination of non(weak)-interacting QD states and cavity pumping is similar to the recent theoretical proposal by Yamaguchi et al., in which exciton complexes are considered. They considered that the origin of the cavity pumping is photon feeding from the non-resonantly coupled excited states instead of background oscillators. We believe that this effect is minor for our case, because we could not find other prominent excitonic emission peaks around the target QD.

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