Establishment of equilibrium of electrostatic potential by photo-irradiation in a GaAs quantum well at low temperature

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Abstract. We measured photoluminescence (PL) spectra from a 20-nm GaAs/AlGaAs quantum well (QW) grown on an n-type substrate by selectively exciting the GaAs at 2 K. We observed a two-stage change of PL spectra as a function of the total amount of photo-irradiation \((p \times t)\) after cooling down. This corresponds to the process of establishing the equilibrium of electrostatic potential between the sample surface and QW and between the QW and n-doped substrate.

1. Introduction

Photoluminescence (PL) spectra of semiconductor quantum wells (QWs) are very sensitive to both the existence of residual electrons and perpendicular electric fields [1, 2]. The electronic states, such as the surface electron states, affect the PL spectra by changing the perpendicular electric field electrostatically or causing a charge flow that changes the residual charge density in the QW. The PL is thus a sensitive probe of the establishment of the equilibrium of the surface states, the residual charges, and donor states under photo-irradiation. It is widely known that photo-irradiation changes the condition of the GaAs/AlGaAs heterostructure sample at low temperature. For example, photo-irradiation improves the electron mobility of two-dimensional electron gas [3]. This is understood as charge re-distribution by exciting the DX center in a Si-doped layer, the impurity states, and the surface state. For an undoped QW, the surface and impurity states are important because there is no intentionally doped layer near the QW. The frozen surface model describes the surface state at low temperature, while the mid gap pinning model describes it at high temperature [4]. Transport and optical measurements of back-gate undoped samples have shown that the surface potential becomes almost zero with photo-irradiation [1, 2, 3]. In this paper, we present the time evolution of PL spectra after cooling. The two-stage evolution of the spectra corresponds to the charge transfer between the surface and QW and between the QW and n-doped substrate.

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2. Experiment
We performed PL measurements at 2 K on a 20-nm-wide undope GaAs/Al$_{0.2}$Ga$_{0.8}$As QW grown by molecular beam epitaxy. The layer structure of the sample is shown in Fig. 1. The sample was set at the cold finger in the vacuum space of a 1 K-pot-equipped 4He cryostat. The sample was irradiated with a laser beam from a 400-μm-core multimode optical fiber. The opening end of the fiber was fixed 1 mm above the sample surface in the cold space (Fig. 1) and the other end was coupled to an optical system for laser input and photo-detection at room temperature. The laser wavelength was 800 nm, which selectively excites the GaAs layers. The present PL spectra were measured with a laser power of 0.8 μW (unless mentioned otherwise). We used a single-mode titanium-sapphire laser for excitation and a monochromator with a nitrogen-cooled charge-coupled-device (CCD) camera for detection.

The sample was cooled in the dark by capping the optical fiber, and we started the laser-irradiation and the PL measurements at 2 K at the same time $t = 0$. We loaded three-times-averaged 20-s-accumulated PL spectra every 60 s. After each cycle of time evolution measurement, we warmed up the sample to $T_{warm} > 100$ K and then cooled it again for the next measurement.

3. Results and discussion
A color-coded plot of a typical time evolution of the PL spectra is shown in Fig. 2(a). The lines at 1.5258 and 1.527 eV correspond to charged exciton $X^-$ and neutral exciton $X_0$, respectively. The PL spectrum changes with a time scale of minutes. We found two typical times, $t_1 \sim 40$ min and $t_2 \sim 375$ min, as depicted in Fig. 2(a). Typical spectrum profiles at $t = 0$, $t_1 < t < t_2$, and $t > t_2$ are shown in Fig. 2(b). The spectrum gradually changed during $0 < t < t_1$ and it became almost stable at $t = t_1$. The spectrum remained unchanged for a while, but abruptly changed again at $t = t_2$. The spectrum lineshape did not change after $t_2$. The two typical times, $t_1$ and $t_2$, indicate that the sample first reached the quasi equilibrium state at $t = t_1$ and then reached the equilibrium state at $t = t_2$. Among a number of measurements, when the sample was cooled from room temperature, the PL spectrum at $t = 0$ sometimes showed a larger linewidth than that shown in Fig. 2(b). However, such a wide $X^-$ spectrum always changed to the narrower peak with laser irradiation and followed the same evolution as in Fig. 2(a).

Possible band profiles for the three time regimes are shown schematically in Fig. 3. Residual charges in the barrier and QW layers bend the band slightly. At $t = 0$, the potential of the surface is lifted because of the mid-gap pinning [4]. With laser irradiation, the electrons trapped in the surface state escape to the substrate through the QW layer. The electrostatic potential between the surface and QW reaches equilibrium at $t = t_1$ and then that between the QW
Figure 2. (a) Color-coded plot of the time evolution of the PL spectra. (b) PL spectral profiles.

and substrate reaches equilibrium at \( t = t_2 \). The difference between \( t_1 \) and \( t_2 \) could be due to the difference in the thickness and the structure of the front- and back-barrier layers. This explanation is consistent with the fact that the photo-irradiation reduces the surface potential to \( \phi_s \sim 0 \) [1, 2, 3]. However, it should be noted that we excited the GaAs band gap, not the band gaps of the barrier layers.

Let us discuss the details of the PL data in terms of the band profiles. At \( t < t_1 \), the \( X^- \) intensity gradually decreases and blue-shifts with time, accompanying an increase in the \( X^0 \) intensity. The intensity change indicates the reduction of the density of the residual charges, which had been frozen in the QW, by their flowing out to the substrate. In addition, the blue shift of \( X^- \) energy is attributed to the quantum confined Stark effect (QCSE). The observed shift at \( t < t_1 \) was 0.2 meV. The perpendicular electric field decreases because of the charge transfer and the potentials of the surface and the QW approach equilibrium. The QCSE shift of this sample is given by \(-1.34 \times \Delta V^2 \) meV, where \( \Delta V \) (V) is the potential difference between the surface and n-doped substrate, which was obtained from the PL measurements on a double-gated sample made from this wafer. Assuming the band profile to be linear, the surface pinning potential \( \phi_s \) of 0.6 V [3] gives the QCSE shift of 0.48 meV. Although the observed shift of 0.2 meV is smaller than 0.48 meV, this could be due to the reduction of the perpendicular electric field in the QW caused by the re-distribution of the residual charges in the barrier and QW layers.

The abrupt increase of \( X^- \) at \( t = t_2 \) is attributed to the increase in the number of residual charges in the QW, which corresponds to the conduction band of the QW reaching the Fermi energy of the n-doped substrate. However, the electron density is smaller than \( 10^{10} \) cm\(^{-2} \), which is estimated from the PL lineshape by comparing it with that of the known electron density [6].

We measured the PL time evolution with different laser powers \( p \) from 0.1 to 100 \( \mu \)W. Figure 4 shows \( t_1 \) and \( t_2 \) as functions of \( p \). Both \( t_1 \) and \( t_2 \) were proportional to \( p^{-1} \), which indicates that the PL evolution depends on the total amount of photo-irradiation, \( p \times t \). The number density of the absorbed photons is estimated to be \( 2.0(1.3) \times 10^{13} \) s\(^{-1} \)\( \mu \)W\(^{-1} \)cm\(^{-2} \) in the surface (QW), where the 3%(2%) of the irradiated photons are estimated to be absorbed in the cap (QW) layer [5]. On the other hand, the residual charge density in the surface state is estimated
to be $C\phi_s=5\times10^{10}\text{cm}^{-2}$, where we assumed the surface pinning potential $\phi_s=0.6$ V and used the geometrical capacitance $C$. The number of surface charges is much smaller than that of absorbed photons at $t < t_2$.

We performed several additional experiments. (1) We measured the recovering temperature by changing $T_{\text{warm}}$ from 70 to 135 K. The PL spectrum did not recover at $T_{\text{warm}} < 90$ K and recovered to the initial state when the sample was warmed above 100 K. This recovery temperature is consistent with the previous reports [3, 4]. (2) We confirmed that the observed PL change is not due to the irradiation by high-energy photons in our laser beam. We put an additional blue filter in the laser line to cut the high-energy laser beam, especially the 532-nm laser of the pump beam, but $t_1$ and $t_2$ did not change. (3) We irradiated a sample with a laser with a lower energy than the GaAs band gap at 850 nm (135 $\mu$W; 5-minute duration at $t < t_1$). We found that the PL spectrum (measured with 800-nm wavelength excitation) changed after the 850-nm irradiation. However, the sample did not reach the final state. This suggests that the charge transfer is induced even if the excitation energy is lower than the GaAs band gap; however, the efficiency is different. (4) The two-stage change disappeared when the electrostatic potential of both the surface and substrate was defined by preparing a metallic gate on the sample surface and an ohmic contact to the n-type substrate.

In summary, we measured the time evolution of PL spectra of an undoped GaAs QW at 2 K by selectively exciting the GaAs layers. We observed a two-stage change in the spectra as a function of the total amount of photo-irradiation ($p \times t$) after cooling. Our measurements indicate that the laser irradiation establishes the potential equilibrium of the undoped GaAs QW with the surface and substrate by charge transfer even though the laser does not excite the barrier layer.

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