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## Circularly Polarized Photoluminescence Probe of Density of States in GaAs/AlGaAs Quantum Hall Bilayers

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Polarized magneto-photoluminescence is employed to study the energies and occupancies of four lowest Landau levels in coupled quantum Hall GaAs/AlGaAs double quantum well. As a result, the magnetic field induced redistribution of charge over the Landau levels manifesting to the continuous formation of the charge density wave and the direct evidence for the symmetric-antisymmetric gap shrinkage at  $\nu = 3$  are found. The observed inter-level charge exchange causes depolarization of the ferromagnetic ground state.

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Owing to the quantization of the in-plane kinetic energy, the effects of electron-electron interaction may be managed by a strong perpendicular magnetic field in twodimensional electron gas. Further control of the interaction is provided by means of spin, subband or layer degrees of freedom in multi-component electron system. A simplest realization of a multiply layer system is a bilayer system. A possibility controlling the competition between intrawell and interwell electron-electron correlations by layer degree of freedom associated with the third dimension in quantum Hall bilayers causes rich variety of quantum phases predicted at odd filling factors in Refs.[1–4]. Some of them were observed in magnetotransport experiments in Refs. [5–9]. Moreover, depending on interlayer tunneling and layer separation a ferromagnetic, a symmetric spin-singlet and an antiferromagnetic canted phases were found at total even filling factor  $\nu = 2$  [10, 11]. Recently, formation of not uniform charge distribution in quantum Hall bilayers at partially populated filling factors was discussed in Refs. [4, 12, 13].

A bilayer electron system may be realized in double quantum well (DQW) semiconductor heterostructures, which consist of two closely spaced two-dimensional electron sheets separated by an insulating barrier. The interlayer coupling hybridizes the individual quantum well states into symmetric (S) and antisymmetric (AS) components, which are split by an energy  $\Delta_{SAS}$ . This splitting results in different emission energies from S and AS states, which can be independently explored by spectroscopic methods. Thus, in contrast to magnetotransport measurements, where an average response of occupied electron states is usually examined, photoluminescence (PL) may probe individual contributions of each state. Polarization resolved PL measurements provide with energy, spin and space distribution of electrons. A probe of the electron distribution is of particular interest because it may throw some light on a development of the charge-density-wave (CDW) in quantum Hall bilayers.

In DQW systems PL was used to study a structure of

energy levels and excitonic states [15–19]. To the best of our knowledge polarized PL was not yet employed. We focalized on examination of spin polarization and occupancy of individual electron states formed in the GaAs/AlGaAs/GaAs bilayer system in quantizing magnetic field in vicinity of the filling factors  $\nu = 2, 3$ . In strong magnetic field, when the cyclotron energy dominates, the electron energy spectrum of this system is composed of four closely spaced LLs resided in an energy interval about a few meV and it has spin and pseudospin (associated with layer) degrees of freedom. The interaction among electrons make them accommodating over the LLs to minimize the total energy and thus, resulting in a new ground state. Similar phenomenon was recently found in electron bilayers in the vicinity of the filling factor  $\nu = 1$  by Raman scattering of spin excitations (see for a review [20]), where the tunneling gap shrinkage was shown to occur due to partial population of the higher LL.

The selectively doped GaAs/AlGaAs/GaAs DQW structure was grown by molecular beam epitaxy. Two identical quantum wells of 14 nm width were separated by  $1.4 \ nm$  AlGaAs barrier. The total sheet electron density  $n_s = 9.2 \times 10^{11} cm^{-2}$  (corresponding to the Fermi energy about 17 meV) and the mobility  $\mu =$  $9.7 \times 10^5 cm^2/Vs$  were measured at the temperature T =1.4 K in Ref. [21]. The circularly polarized PL measurements were performed at base temperature  $T \simeq 300 \ mK$ with the laser excitation energy  $2.33 \ eV$  and the power lower than 1  $mW/cm^2$  and with the spectral resolution better than 1 meV. A linear polarizer and a quarter-wave plate were set in front of the sample in a <sup>3</sup>He cryostat. The magneto-PL was measured with the magnetic field perpendicular to the sample surface. The degree of the circular polarization (DCP) was determined according to the formula:

$$P_{\sigma} = \frac{I^{-} - I^{+}}{I^{-} + I^{+}} \tag{1}$$

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where  $I^+$ ,  $I^-$  are the integrated PL intensities measured in the  $\sigma^+$  and  $\sigma^-$  polarizations respectively. The PL circular polarization is associated with the polarization of the excitonic emission with effective g factor:  $g_{ex} =$  $g_e + g_h$ , where  $g_e$  and  $g_h$  stand for the electrons and for the holes, respectively [22]. The non-linear Zeeman splitting, caused by a complicated structure of the valence band in GaAs, determines the polarization of the holes measured in high magnetic field which monotonically varies with the magnetic field [23]. At the same time as the electron polarization exhibits sharp features attributed to the QH states [24].

The PL emission in  $\sigma^+$  polarization is associated with the transition from the conduction band electron spin state  $m_i = -1/2$  to the electron valence band state with  $m_j = -3/2$ , while the valence band state with  $m_i = +3/2$  and the electron spin state  $m_i = +1/2$  are involved in the  $\sigma^-$  polarization. The polarized PL spectra of the DQW structure, measured as a function of the magnetic field are plotted in Figs.1(a,b). The low energy emission is due to the GaAs cap layer. Two close PL lines observed at a moderate magnetic field around 1.53 eV are assigned to the interband transitions between the S and AS states. The energy interval between them is composed of the  $\Delta_{SAS}$  gaps of the electrons and of the holes. The electron contribution dominates due to the smaller electron effective mass as compared to that of the holes. Thus obtained spectroscopic  $\Delta_{SAS} \simeq 3 \ meV$ is found somewhat smaller than the gap  $\Delta_{SAS} \simeq 4.2$ meV measured by magneto-transport experiments [21].

The spectra plotted in Figs.1(a,b) reveal oscillations of the PL intensities emitted by S and AS states. They are caused by the magnetic field induced changes in the occupied density of states of the LLs nearest to the Fermi energy: one emptying (the upper level) and another filling (the lower level). Consequently, the observed PL intensities are attributed to the filling factors of the LLs closest to the Fermi energy.

The details of the  $\sigma^+$  polarized PL line structure measured in the range of the high magnetic field where S and AS lines come together are shown in Fig.1(c). The evolution of the PL line shape with the magnetic field implies a superposition of two lines even in high magnetic field range. Different intensities of the S and AS lines make the resulting PL line shape asymmetric, depending on the magnetic field. A high signal-to-noise ratio allowed reproducing accurate spectral shapes. As shown in the figure, perfect fits are obtained with two Gaussian lines. As a result, the PL energy positions and integrated intensities were obtained and they are analyzed below.

The transport LL broadening gives the value 100 times smaller than the spectroscopic broadening. Thus the main contribution to the PL line width is due to the holes. For that reason, the observed overlapping between the PL lines does not points to an overlapping between corresponding LLs.

The obtained DCP of the S and AS emissions are shown in Fig.2(a) as functions of the magnetic field. The

observed polarizations reveal distinct peaks at the filling factors  $\nu = 3,7$  and  $\nu = 5$  for the AS and S PL lines, respectively, where the filling factors are determined by the magnetotransport measurements [21]. At higher excitation power these oscillations of the polarizations are found up to the filling factor  $\nu = 15$ . The polarization peaks correspond to the filling factor defined as  $\nu = 4N \pm 1$ , where + and - hold for the S and AS LLs, respectively. The contribution of the holes to the DCP can be estimated at the filling factors  $\nu = 4, 6, 8$ when the electrons are unpolarized. The polarization peaks measured with respect to the hole polarization correspond to roughly full spin-polarized odd quantum Hall states. It ought to be mentioned that the peaks of the S and AS polarizations take place at different filling factors. This is a manifestation of insignificant symmetricto-antisymmetric scattering. Accordingly, the S and AS electron states are uncoupled and emit independently. Similar conclusion was recently obtained in Ref. [25].

The Landau fan diagram for the conduction band of the DQW is demonstrated in Fig.2(b). The parameters used in the calculations are  $g_e = -2.6$  and the electron effective mass  $m = 0.068m_0$ . The best fit of the magnetic field values which match the filling factors corresponding to the polarization peaks was obtained with the sheet electron densities  $n_S = 4.8 \times 10^{11} \text{ cm}^{-2}$  and  $n_{AS} = 4.4 \times 10^{11} \text{ cm}^{-2}$  occupying the lowest S and AS levels, respectively. According to the calculations, the lowest spin-down S level and the lowest spin-up AS level cross around the filling factor  $\nu = 4$ . This crossing manifests itself to the magnetic field driven quantum phase transition from the spin-singlet symmetric phase to the spin polarized ferromagnetic one [10].

In the range of low magnetic field ( $\nu \ge 3$ ) our data agree well with a single-electron model. The PL peak energies associated with the energies of the LLs behave linearly with the magnetic field. At  $\nu = 4$  the emissions due to the S and AS levels exhibit minimum and maximum, respectively. In the neighborhood of the filling  $\nu = 3$  the upper spin-down AS LL becomes empty and the electrons are transferred to the low-lying spin-down S level. Such redistribution of the electrons between the S and AS levels indeed is evident in the changes of the corresponding PL intensities ( $\sigma_S^+$  and  $\sigma_{AS}^+$ ) shown in Fig.3(b) and the resulting crossing between them at  $\nu = 3.2$  proves the symmetric-antisimmetric level crossing. Also, in accord with a single-electron model, the states with odd filling factors are found fully polarized.

The following features observed in the high magnetic field ( $\nu < 3$ ) are inconsistent with a single-electron model: (i) the nonlinear dependencies of the LL energies on the magnetic field shown in Fig3(a); (ii) the integrated intensity of the  $\sigma_{AS}^+$  emission shown in Fig.3(b) is found increasing up to the filling  $\nu = 2$ , while for noninteracting electrons it must vanish already at  $\nu = 3$ ; (iii) in the vicinity of  $\nu = 2$  the observed polarization is due to the holes, while the electron system is unpolarized [Fig.3(d)]. However, as shown in Fig.2(b), at  $\nu = 2$  the electrons occupy two identically polarized S and AS lowest LLs and therefore, the electron system must be completely polarized. The expected single-electron behaviors of the  $\sigma_{AS}^+$ integrated intensity and of the AS polarization are shown in Figs.3(b,d) by dashed lines. We attribute the mentioned discrepancies to the effects of interaction among electrons.

The crossing between the spin-down S and the spin-up AS levels favors the low energy electron excitations across the  $\Delta_{SAS}$  gap in the ferromagnetic phase. According to Ref.[26], such excitations may result in a magnetic-fielddriven  $\Delta_{SAS}$  gap collapse and consequently, in a destruction of the quantum Hall effect, firstly demonstrated at  $\nu = 1$  and  $\nu = 3$  in Ref.[5]. Indeed, as shown in Fig.1(a), at the filling factor  $\nu \leq 3$  the  $\sigma^+$  polarized S and AS lines merge to a single asymmetric PL line. This directly demonstrates the  $\Delta_{SAS}$  gap decreasing with the magnetic field. On the contrary, the PL measured in  $\sigma^{-}$ polarization depicted in Fig.1(b) reveals separated S and AS lines even in highest available magnetic field. As follows from Fig.2(b), in the ferromagnetic phase the lowest energy excitations are possible only across the  $\Delta_{SAS}$  gap which separates the filled S and empty AS spin-down LLs. The gap for the spin-up excitations is larger than the  $\Delta_{SAS}$  gap and therefore, no low energy  $\sigma^-$  polarized excitations are realizable.

The phase diagram which determines conditions for the  $\Delta_{SAS}$  collapse at  $\nu = \text{odd}$  sets up the ratio d/l as a function of dimensionless energy  $\varepsilon = \Delta_{SAS}/(e^2/\epsilon l)$ , where d is the interwell spacing,  $l = \sqrt{\hbar c/eB}$  is the magnetic length,  $\epsilon$  is the dielectric constant [26]. In the studied here DQW  $d/l \simeq 2.2$ . For such ratio the theory predicts the  $\Delta_{SAS}$  collapse at  $\varepsilon < 0.1$ , while in our sample  $\varepsilon \simeq 0.1$ . These calculations do not account for LL mixing, which may lead to a larger suppression of the  $\Delta_{SAS}$  excitation gap due to intralayer correlations and as a result, to larger critical value of  $\varepsilon$ . Thus, a manifestation of electron-electron interaction is presented at  $\nu = 3$  in the observed shrinkage of the  $\Delta_{SAS}$  gap.

Further evidence for the electron-electron interaction is the integrated  $\sigma_{AS}^+$  PL intensity increasing with the increasing magnetic field at  $\nu < 3$ . As stated above, the shape of the  $\sigma^+$  PL line in high magnetic field implies a superposition of two lines each attributed to the emissions from S ( $\sigma_S^+$ ) and AS ( $\sigma_{AS}^+$ ) spin-down LLs. A proportion between the  $\sigma_S^+$  and  $\sigma_{AS}^+$  integrated PL intensities is defined by the relative occupancy of the S and AS levels located in the neighborhood of the Fermi energy. The relative S and AS PL intensities calculated as  $I_{S(AS)}/(I_S + I_{AS})$  for both polarizations are shown in Figs.3(b,c). Remarkably, the balance in the occupancy of the S and AS LLs drastically changes at the filling  $\nu < 3$ when the  $\sigma_{AS}^+$  intensity increases instead of vanish. This results in depolarization of the  $\nu = 2$  ferromagnetic state. The crossing points at  $\nu = 3.2$  and  $\nu = 5/2$ , when the integrated PL intensities of the S and AS lines become equal, imply identical occupancies of the corresponding LLs. This is directly seen in Fig.1(c) at the filling factors related to the fits obtained with two equal Gaussian lines.

We attribute the rise in the  $\sigma_{AS}^+$  PL intensity observed at  $\nu < 3$  to not uniform charge distribution caused by formation of the coherent CDW. In such case, when the upper LL is partially populated, the uniform uncorrelated spin-polarized electron liquid is unstable against formation of CDW [4, 12, 13]. The parameters of the bilayer electron system studied here correspond to the boundary between the uniform ground state and the region where CDW is expected [12]. Such CDW takes a form of spatial domains with integer filling factors where the regions of high electron density in one well are nearest to regions of low electron density in the other [4]. The data shown in Fig.3(b) straightforwardly demonstrate the enhanced occupancy of the upper antisymmetric LL with simultaneous depopulation of the lower LL, which is caused by the continuous development of CDW. In this case the ground state is formed by the electron-rich regions in one layer (LL with enhanced occupancy) which align directly above the hole-rich regions (depopulated LL) in the another [13].

In addition, the integrated PL intensities from the completely filled lowest spin-up LLs ( $\sigma_s^-$  and  $\sigma_{AS}^-$ ) shown in Fig.3(c) also depend on the magnetic field, although not so drastically as the intensities from the LLs nearest to the Fermi energy. This is an experimental evidence that these LLs also take part of the CDW and cannot be considered as completely inert, even though in theory they are believed to be unaffected by interaction. As pointed out in Ref.[27], such situation takes place if two quantum wells are close enough that they begin interact one another.

It is worth adding, that a re-distribution of the heavy holes over the S and AS levels may cause the PL depolarization observed at  $\nu < 3$ . However, in order to correlate with the filling factors such effects should be associated with some kind of the electron-hole exchange interaction. No evidences of such interaction were found.

In conclusion, the circularly polarized photoluminescence measured in the GaAs/AlGaAs coupled double quantum well in quantizing magnetic field is presented as an effective tool separating responses of the S and AS LLs. The considerable reduction of the  $\Delta_{SAS}$  gap is observed at the filling factor  $\nu = 3$  and shown to occur due to the lowest energy symmetric-to-antisymmetric electron excitations. The changes in the integrated PL intensities associated with the occupancy of the S and AS electron states reveal a formation of the charge-densitywave in the range of the magnetic field where the  $\Delta_{SAS}$ gap is reduced. The inversion of the PL intensities due to the symmetric and antisymmetric LLs observed between the filling factors  $\nu = 3$  and  $\nu = 2$  is explained as a result of the magnetic field driven inter-Landau level charge transfer which causes the depolarization of the ferromagnetic ground state. The interpretation along this line manifests to the continuous formation of the chargedensity-wave in bilayer electron system.

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Fig.1. (Color online) (a) and (b) Polarized photoluminescence intensities of the DQW structure measured as a function of the magnetic field. Inset shows the energy structure of four lowest Landau levels and the relevant optical transitions at the filling  $\nu < 4$ . The details of the  $\sigma^+$  polarized spectra in the vicinity of the filling factor  $\nu = 3$  are shown in panel (c): thick black lines are the experimental spectra, white dashed lines show the best fits with two Gaussian line (thin red lines). Thicknesses of the experimental spectra lines are deliberately magnified for clear representation of the fitting results.

Fig.2. (Color online) (a) Polarizations of the photoluminescence lines assigned to the symmetric (open blue circles) and antisymmetric (closed red circles) transitions measured as functions of the magnetic field. (b) Calculated energy diagram of the Landau levels in the studied double-quantum well structure; the up and down arrows shown at right axis correspond to electrons with spin-up and down, respectively.

Fig.3. (Color online) (a) Peak energies of the  $\sigma^+$  polarized symmetric (open circles) and antisymmetric (closed circles) PL lines. (b) and (c) show relative integrated PL intensities of the symmetric (S) and antisymmetric (AS) lines as functions of the filling factor in  $\sigma^+$  and  $\sigma^-$  polarizations, respectively. (d) Polarizations of the PL lines assigned to the symmetric (open blue circles) and antisymmetric (closed red circles) transitions measured as functions of the filling factor. Dashed lines in (b) and (d) show the single-electron behavior of the  $\sigma^+_{AS}$ integrated intensity and the polarization expected for AS emission.



Figure 1 LN13208 19Jun2012



Figure 2 LN13208 19Jun2012



Figure 3 LN13208 19Jun2012