

# Nuclear spin relaxation in the SU(4) spin-pseudospin intertwined skyrmion regime in the $\nu = 1$ bilayer quantum Hall state

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We investigate the electron spin degree of freedom in imbalanced density  $\nu = 1$  bilayer quantum Hall states using the resistively detected nuclear-spin-lattice relaxation rate  $1/T_1$ . The values of  $1/T_1$  in imbalanced states increase rapidly in the vicinity of  $\nu = 1$ , similar to the phenomenon that is observed in the region in which skyrmion crystals are formed. Furthermore, the value of  $1/T_1$  in the back layer (the layer from which electrons are transferred to the front layer) also increases in the intermediate imbalanced state. These results indicate that the low-energy electron-spin mode, similar to the mode observed in skyrmion crystals, exists across the two layers. We suggest that such a mode arises in spin-pseudospin intertwined SU(4) skyrmions.

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## I. INTRODUCTION

Skyrmions, in which spins point in all directions normal to a sphere, have been of great interest in recent years, not only to the fundamental physics but also to future electronics because they are topologically stable. This topological object was originally introduced to describe a model for nucleons in pion field theory,<sup>1</sup> however, it now appears in spin structures in condensed matter physics such as chiral-lattice ferromagnets,<sup>2-5</sup> ordinary ferromagnets,<sup>6</sup> superfluid <sup>3</sup>He-A phase,<sup>7</sup> and quantum Hall states (QHSs).<sup>8-11</sup> More stimulating situations can arise when the system has an additional pseudospin degree of freedom. In this situation, both the spin and the pseudospin degrees of freedom can provide the system with a higher SU(4) symmetry for a combination of a spin and pseudospin pair of SU(2)'s. The spin and pseudospin intertwined skyrmions [SU(4) skyrmions] were considered by one of the authors in bilayer quantum Hall systems,<sup>12-15</sup> in which the layer degree of freedom can be regarded as pseudospin. Recently, the existence of SU(4) skyrmions has been predicted in graphene,<sup>16</sup> in which the valley degree of freedom is present as a pseudospin.

Skyrmion spin textures in quantum Hall systems in GaAs/AlGaAs heterostructures appear in the monolayer  $\nu = 1$  QHS as a quasiparticle in the spin-polarized ground state.<sup>8</sup> Here,  $\nu$  denotes the Landau level filling factor, which expresses the ratio of carrier densities to magnetic fields; thus  $\nu = 1$  represents that electrons and magnetic flux quanta have a one-to-one correspondence. Experimentally, optically pumped nuclear magnetoresonance measurements revealed that the state at filling factors slightly away from  $\nu = 1$  contains a finite density of skyrmions.<sup>9</sup> They are expected to crystallize, at least when they are sufficiently dilute.<sup>17,18</sup> It was also revealed that the low-energy spin-wave mode (Goldstone mode) in skyrmion crystals allows nuclear spins to relax more rapidly<sup>10,11,18</sup> after nuclear spins are polarized at  $\nu = 2/3$  QHS by a so-called flip-flop process between electron spins and nuclear spins.<sup>19,20</sup> Our aim in this work is to detect this fast nuclear-spin relaxation rate  $1/T_1$ , where  $T_1$  denotes the

nuclear-spin-lattice relaxation time, in the imbalanced bilayer QHS, where the degree of freedom of the layer can be regarded as the pseudospin. At the bilayer  $\nu = 1$  QHS when electron densities are equally distributed in each layer, the existence of pseudospin textures like a half-skyrmion (or meron) is plausible from a number of experiments<sup>21-24</sup> and theories.<sup>25,26</sup> It is intriguing when the density in each layer is imbalanced, because spin and pseudospin intertwined SU(4) skyrmions can emerge as a consequence of the interlayer and intralayer Coulomb interaction.

In this study, we report the observation of increases in the nuclear-spin relaxation rate  $1/T_1$  in the intermediate imbalanced density  $\nu = 1$  bilayer QHS, in which the existence of SU(4) skyrmion crystals has been predicted theoretically. When the density imbalance is expressed as  $\sigma = (n_f - n_b)/(n_f + n_b)$ , where  $n_f$  ( $n_b$ ) represents the electron density in the front (back) layer, our data reveal a strong enhancement of  $1/T_1$  in the vicinity of  $\nu = 1$  at intermediate  $\sigma$  values, similar to the result that is observed in the region in which skyrmion crystals exist. In addition, an enhancement of  $1/T_1$  at intermediate  $\sigma$  values is also observed in the back layer, from which electrons are transferred to the front layer, indicating that the low-energy electron-spin mode extends across the two layers. A possible attribution of this fast relaxation is the low-energy spin-wave mode that arises in the predicted crystal form of SU(4) skyrmions.

This paper is structured as follows: In Sec. II, we describe the experimental method and the procedure for measuring  $1/T_1$ . In Sec. III, we present the experimental results on  $1/T_1$ . In Sec. IV, we discuss our results. Comparisons with previous experiments are made and a possible example of quasiparticles to explain our results is discussed. Finally, we present our conclusions in Sec. V.

## II. EXPERIMENTAL METHOD

The sample used in this study consists of two 20-nm-wide GaAs quantum wells separated by a 1.5-nm-thick AlAs

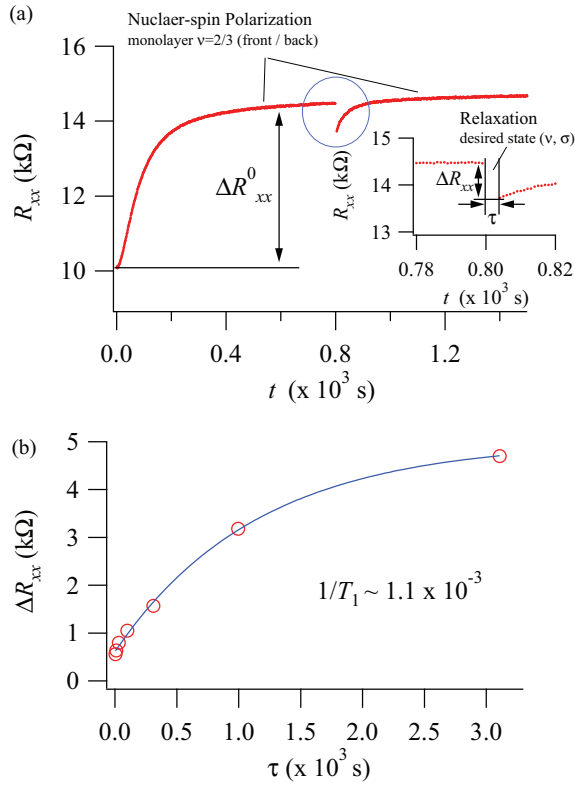


FIG. 1. (Color online)  $1/T_1$  measurement procedure using nuclear-spin polarization and relaxation in the monolayer  $\nu = 2/3$  QHS. (a) Typical example of time evolution of  $R_{xx}$ . Inset: Expanded view of the trace at which the magnitude of resistance changes is measured [open circle in the main figure] after nuclear-spin relaxation for  $\tau$  seconds. (b) Plot of  $\Delta R_{xx}$  as a function of  $\tau$ . The curve is the result of fitting.

barrier layer with a tunneling gap of approximately 5 K. The interlayer separation  $d$  of this sample is 21.5 nm. By adjusting the biases of the front  $V_f$  and back  $V_b$  gates, we can independently control the densities in the front  $n_f$  and back  $n_b$  layers.<sup>27</sup> Measurements of resistances are performed at 60 mK in a dilution refrigerator using a standard low-frequency lock-in technique. The low-temperature electron mobility is approximately  $2 \times 10^6$  cm<sup>2</sup>/V s at a density of  $1.0 \times 10^{11}$  cm<sup>-2</sup>.

$1/T_1$  is measured by using the remarkable property of the  $\nu = 2/3$  QHS. The  $\nu = 2/3$  QHS, which can be interpreted as a  $\nu = 2$  composite fermion QHS, has two competing composite-fermion Landau levels with different spin configurations. Therefore, the degeneracy of spin-polarized and unpolarized ground states induces fluctuations in electron spins which change the nuclear-spin polarization via hyperfine coupling and increase additional inhomogeneous sources of scattering.<sup>20</sup> A large current promotes this process and increases the longitudinal resistance, which is proportional to the nuclear-spin polarization.<sup>10</sup> The following procedure is used to measure  $1/T_1$  and Fig. 1 shows a typical data set as obtained by this procedure. First, the nuclear-spin polarization in the host materials (GaAs quantum well) is pumped by a relatively large current ( $=50$  nA) in the monolayer  $\nu = 2/3$  regime (at  $\nu = 0.62$ ) that induces an enhancement in the

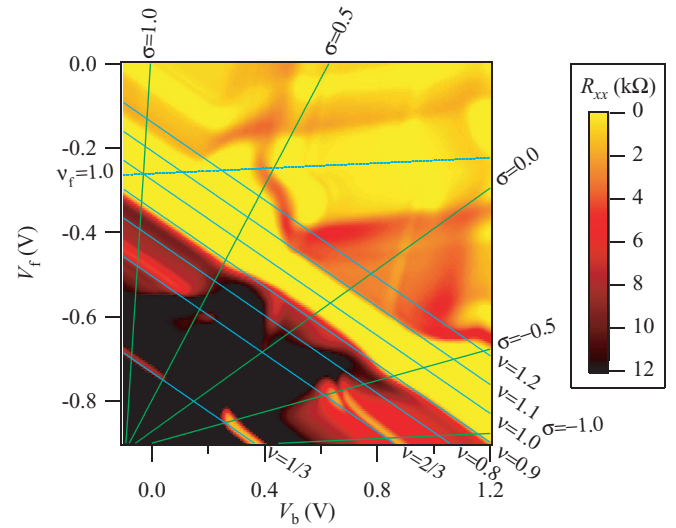


FIG. 2. (Color online) Image plot of  $R_{xx}$  as a function of the front and back gate voltages,  $V_f$  and  $V_b$ , at  $B = 6.0$  T.

longitudinal resistance  $R_{xx}$  until it saturates [Fig. 1(a)] at 6.0 T. Then the gate voltages are tuned to the desired density state (expressed in terms of  $\nu$  and  $\sigma$ ) for a given period of time  $\tau$ , during which nuclear spins are allowed to relax to equilibrium [inset in Fig. 1(a)]. Subsequently, the gate voltages are restored to the first state (monolayer  $\nu = 2/3$ ), and the magnitude of the change in resistance  $\Delta R_{xx}$  due to the relaxation in nuclear-spin polarization is measured and plotted against  $\tau$  [Fig. 1(b)]. By repeating the above procedure with increasing  $\tau$ , we derive the relation between  $\Delta R_{xx}$  and  $\tau$  and fit it with  $\Delta R_{xx}/\Delta R_{xx}^0 = 1 - \exp(-\tau/T_1)$ . Here,  $\Delta R_{xx}^0$  is the induced enhancement in resistance from its initial value in the monolayer  $\nu = 2/3$  state. We can select either the front layer or the back layer as the first monolayer  $\nu = 2/3$  state and thereby independently obtain the values of  $1/T_1$  in each layer.

### III. RESULTS

Figure 2 shows an image plot of  $R_{xx}$  at  $B = 6.0$  T as a function of  $V_f$  and  $V_b$ . Bright (yellow) regions indicate that the values of  $R_{xx}$  are small, therefore they correspond to QHS. Several values of  $\nu$  and density imbalance parameter  $\sigma$  are indicated by thin lines. Among those lines is one denoted  $\nu_f = 1.0$ , where  $\nu_f$  represents the filling factor in the front layer. The QHS that is formed mainly by the electrons in the front layer lies along this line.<sup>28</sup> We can see a large QHS region that corresponds to  $\nu = 1$  extending from  $\sigma = 1.0$  (monolayer system in the front layer) to  $\sigma = -1.0$  (monolayer system in the back layer) at the center of Fig. 2. The QHS collapses for the lower-filling side of the  $\nu = 1$  QHS. We note that the densities still change outside of the  $\sigma = \pm 1.0$  lines after the system becomes a monolayer system by the gate voltages. The  $\nu = 1$  QHS region becomes wider for  $\sigma \gtrsim 0.5$  and  $\sigma \lesssim -0.5$ , indicating an increase in stability. This is attributed to the  $\nu_f = 1$  QHS that merges into the  $\nu = 1$  QHS. We see that this additional expanded region of  $\nu = 1$  QHS affects the  $1/T_1$  behavior in Fig. 3. The calculated value of  $d/\ell_B$  ( $\ell_B \equiv \sqrt{\hbar/eB}$ ), which represents the ratio of the interlayer Coulomb energy to the intralayer Coulomb energy, is approximately 2.1

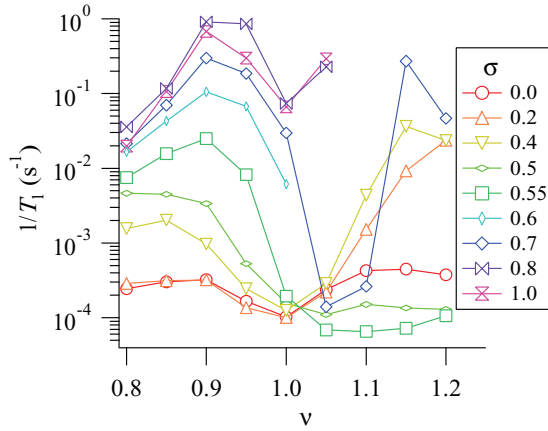


FIG. 3. (Color online)  $1/T_1$  as a function of  $\nu$  around the  $\nu = 1$  QHS for various values of  $\sigma$ , from 0 to 1.

at  $\sigma = 0$ . Subsequently, we measure  $1/T_1$  along the lines of  $\sigma$  (for  $\nu$  dependence) and  $\nu$  (for  $\sigma$  dependence).

Figure 3 shows a plot of  $1/T_1$  measured in the front layer for each value of  $\sigma$ , as a function of  $\nu$ . At the monolayer point, the  $1/T_1$  value increases in the vicinity of either side of  $\nu = 1$  because the Goldstone mode in skyrmion crystals has a low-energy spin excitation mode that exchanges the angular momentum with the host nuclear spins via a hyperfine interaction.<sup>10,11,29</sup> When  $\sigma$  is decreased,  $1/T_1$  values continuously decrease to the values at  $\sigma = 0$  for the  $\nu < 1.0$  region, while maintaining the maximum value at  $\nu = 0.9$  for most values of  $\sigma$  even at  $\sigma = 0$ . Whereas  $1/T_1$  values for the  $\nu > 1.0$  region do not change continuously,  $1/T_1$  values at, for example,  $\sigma = 0.55$  are lower than those at  $\sigma = 0$ . This asymmetric behavior can be explained easily by reviewing Fig. 2. The lower relaxation rate is ascribable to the stable QHS region, which is broadened to larger values of  $\nu$  especially at around  $\sigma = 0.6$ , on account of the superposition of  $\nu_f = 1$  and  $\nu = 1$  QHSs. Moreover, as  $\sigma$  is increased,  $1/T_1$  increases rapidly for most values of  $\sigma$ . For example,  $1/T_1$  for  $\sigma = 0.55$  increases by approximately two orders of magnitude at  $\nu = 0.9$ . This strongly suggests the existence of low-energy electron-spin fluctuations of quasiparticles with structures similar to that of the skyrmion crystal at  $\sigma = 1$  and of continuous evolution in the range from  $\sigma = 0$  to 1.

In order to confirm the spatial distributions of the spin texture across the two layers, we investigate  $1/T_1$  for *both* layers. Figure 4 shows  $1/T_1$  as a function of  $\sigma$  for the front and back layers at  $\nu = 0.9$  and 1.0. As  $\sigma$  is increased (i.e., the electrons in the back layer are brought to the front layer),  $1/T_1$  in the front layer increases significantly and approaches the fast monolayer relaxation rate. However, in the back layer, nuclear relaxation by electrons is supposed to be suppressed monotonously. However, in the back layer as well,  $1/T_1$  at  $\nu = 0.9$  increases and takes a maximum value at  $\sigma = 0.55$ , somewhat mimicking the rate in the front layer, although it then decreases to as low as the value observed at  $\sigma = 0$  due to the absence of electrons. At  $\sigma = 0.55$ , the filling factor of the back layer is approximately 1/4, and no QHS with this filling factor is observed in this sample; nor is the liquid state of composite fermion with four flux quanta probable.

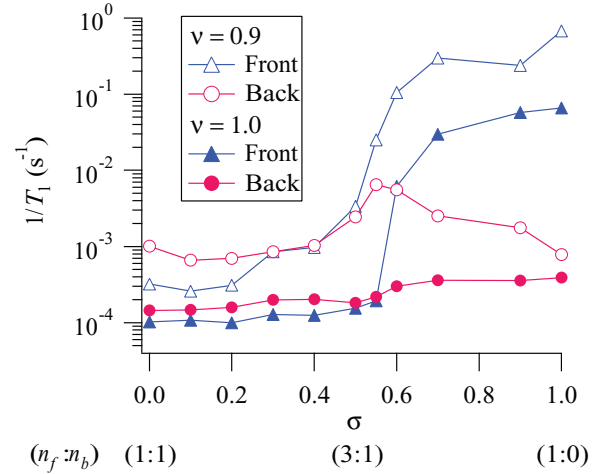


FIG. 4. (Color online) Data set for  $1/T_1$  as a function of  $\sigma$  for the front and back layers at  $\nu = 0.9$  and  $\nu = 1.0$ .

Therefore, the maximum in  $1/T_1$  at  $\nu = 0.9$  in the back layer strongly indicates that the low-energy spin excitation mode occurs across the two layers and that the quasiparticles have interlayer charge distributions due to the finite value of the tunneling energy.

#### IV. DISCUSSION

As mentioned, the quasiparticle excitation at  $\sigma = 0$  in pseudospin textures (bimerons), in which a low-energy spin mode is considered to be absent. However, our observation at  $\sigma = 0$  as well as the corresponding observation of Kumada *et al.*<sup>30,31</sup> show a small maximum in  $1/T_1$  at  $\nu = 0.9$ . They attribute this to the spin fluctuations in the compressible puddles in the incompressible (QHS) back ground.<sup>32</sup> (We discuss the spin degree of freedom further later.) Careful comparisons reveal the difference from the previous data<sup>30,31</sup> that were measured at 0.3 K. The values of  $1/T_1$  for  $\sigma = 0$  in our data are an order smaller than in the previous work.<sup>30,31</sup> We attribute this to suppression of the thermal fluctuation in spins because our experiment was performed at a lower temperature (60 mK). However, at the monolayer limit ( $\sigma = 1$ ), the values of  $1/T_1$  become slightly larger than the values in previous observations.<sup>10,30</sup> According to the heat capacity measurement near  $\nu = 1$  QHS,<sup>33</sup> a sharp peak was observed at approximately 36 mK, suggesting the transition from liquid to solid state of skyrmions. Our data were taken at a temperature much closer to this value. Therefore, skyrmions are in a more favorable situation for forming a local structure similar to that of a solid state, especially in the presence of potential variations by the intrinsic impurities. This favors fast nuclear relaxation because the wavelength of the spin mode becomes longer, although the size of each skyrmion is smaller in our experiment due to the higher magnetic field. Between  $\sigma = 0$  and  $\sigma = 1$ , we see a behavior of  $1/T_1$  similar to that at  $\sigma = 1$  in Fig. 3. The similarity and the continuous change observable in the range  $\sigma = 1$  to  $\sigma = 0$  suggests a continuous transition from skyrmions to bimerons, which was also observed in the activation energy measurement.<sup>34</sup> However, skyrmions do not support the interlayer charge distribution, and on the contrary, bimerons do not support the collective spin

mode. Therefore, we need quasiparticles that possess properties of both skyrmions and bimerons to explain the result in Fig. 4. Such quasiparticles may be SU(4) skyrmions that intertwine spin and pseudospin textures and also have the interlayer charge distribution.<sup>12</sup> Although it is controversial whether the spin and pseudospin entangled textures can be stabilized as the energy minima for charged excitations, this has been argued to occur in the intermediate charge imbalance state.<sup>15</sup>

According to the theory, there are three Goldstone modes for the liquid state of SU(4) skyrmions.<sup>13</sup> In addition to spin-wave and pseudospin-wave modes, the SU(4) skyrmion liquid has a gapless collective mode involving spin and pseudospin fluctuations. Recent theoretical studies have suggested that SU(4) skyrmions form crystals in the intermediate imbalance region of the  $\nu = 1$  bilayer QHS.<sup>35,36</sup> Collective modes in crystals of SU(4) skyrmions have been studied extensively by Côté *et al.*<sup>36</sup> According to their analysis, the U(1) gapless mode that is responsible for the fast nuclear-spin relaxation still exists even in the presence of tunneling energy, Zeeman energy, and electrical bias between the two layers. Our experiment satisfies the above conditions except the temperature. However, taking into account that the temperature is close to the liquid-to-solid transition point,<sup>33</sup> the possibility of the U(1) gapless mode in the SU(4) skyrmion crystal cannot be excluded.

It is also intriguing that  $1/T_1$  at  $\nu = 1.0$  in the front layer increases rapidly at  $\sigma = 0.6$  in Fig. 4. Although the values are smaller than the values at  $\nu = 0.9$ , this is indicative of spin fluctuations at a finite temperature. It is noteworthy that there is no maximum around  $\sigma = 0.6$  in the behavior of  $1/T_1$  in the back layer at exactly  $\nu = 1.0$ . This difference suggests that the origin for increasing  $1/T_1$  is not the low-energy spin waves across the layers, but local skyrmions in compressible puddles in the front layer, since  $\nu_f$  can approach 0.8 or 0.9 in these puddles for  $\sigma \gtrsim 0.6$ .

Finally, we comment on the spin degree of freedom in the  $\nu = 1$  balanced bilayer QHS ( $\sigma = 0$ ). In earlier studies pertaining to the  $\nu = 1$  bilayer QHS, the spin degree of freedom was often neglected because of the simple assumption that a strong magnetic field leads to the generation of fully polarized spins. However, recently conducted experiments<sup>30,31,37</sup> have indicated the possible involvement of the spin degree of freedom in quasiparticle excitation in the  $\nu = 1$  bilayer QHS. Our data on  $\sigma = 0$  in Fig. 3 as well as previous experiments<sup>30,31</sup> show that  $1/T_1$  increases as  $\nu$  deviates from 1, suggesting that electron spins are included in the quasiparticle excitation of the balanced bilayer QHS. The small increase in  $1/T_1$  at  $\nu = 0.9$  in this state might be ascribable to the compressible puddles

in the incompressible background.<sup>30–32</sup> Since the monolayer  $\nu = 1/2$  state is regarded as a Fermi liquid of composite fermions<sup>38</sup> with opposite spin levels that are almost degenerate, electron spins can fluctuate without any change in energy.<sup>10</sup> If the number of compressible puddles increases when  $\nu$  deviates from  $\nu = 1$ ,  $1/T_1$  gradually increases, as shown in Fig. 3. There has been, however, a suggestion<sup>39</sup> that a lattice of entangled SU(4) skyrmions has a lower energy than a lattice of spin-polarized bimerons at  $\sigma = 0$  if  $d/\ell_B$  is large enough and the Zeeman coupling is not too large. Therefore a low-energy spin mode can occur as  $\nu$  away from 1. Our experiment satisfies these conditions, and we find an experimental fact that implies entanglement at  $\sigma = 0$ , namely, the continuous change shown in Fig. 3 in the range from  $\sigma = 1$  to  $\sigma = 0$ .

## V. CONCLUSIONS

In conclusion, we have investigated the nuclear-spin relaxation rate  $1/T_1$ , which has a strong relation with a low-energy spin-wave mode in crystals of quasiparticles. We have found that the values of  $1/T_1$  at intermediate imbalanced states increase by two orders of magnitude in the vicinity of the  $\nu = 1$  QHS. Furthermore, the values of  $1/T_1$  for the back layer also increase in intermediate density states. These results indicate that a low-energy spin-wave mode similar to the one in the skyrmion crystal at the monolayer  $\nu = 1$  QHS exists across the layers and that the quasiparticles have interlayer charge distributions. The SU(4) skyrmion, which has a spin-pseudospin intertwined texture, can be considered an example of such a quasiparticle compared with the theories on SU(4) skyrmions. In addition, in view of the application to nuclear-spin engineering, our results suggest that we might be able to control the nuclear-spin relaxation rate via controlling the bias voltage between two layers.

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