Longitudinal SDW order in a quasi-1D Ising-like quantum antiferromagnet

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From neutron diffraction measurements on a quasi-1D Ising-like Co2+ spin compound BaCo2V2O8, we observed an appearance of a novel type of incommensurate ordering in magnetic fields. This ordering is essentially different from the Néel-type ordering, which is expected for the classical system, and is caused by quantum fluctuation inherent in the quantum spin chain. A Tomonaga-Luttinger liquid (TLL) nature characteristic of the gapless quantum 1D system is responsible for the realization of the incommensurate ordering.

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The quasi-one-dimensional (1D) antiferromagnets, in which spin chains are coupled only weakly by interchain interactions, involve significant quantum fluctuation and often display exotic behavior. As expected for the classical models, the antiferromagnet in three-dimension typically develops a usual Néel order with antiparallel alignments of neighboring spins when it is cooled. Owing to the strong quantum fluctuation, however, qualitatively different situations from the classical models can appear for the quasi-1D antiferromagnets. Several exotic states have been discovered in the quasi-1D antiferromagnets, such as the valence-bond-solid ground state [1] or the spin-Peierls state [2]. In this letter, we show that in the system with Ising-like anisotropy in magnetic fields, the strong quantum fluctuation, stemming from non-commutative relations of the longitudinal spin component, expressed as $< S^z_i S^z_{i+1} > \approx (\text{const}) r^{-\eta_z}$. The incommensurate correlation of the longitudinal spin component, expressed as $< S^x_i S^x_{i+1} > \approx -m^2 \cos(2k_f r) r^{-\eta}$, is a distance between the spins. $\eta_z$ and $\eta$ are Tomonaga-Luttinger exponents, which satisfy a relation $\eta = 1$.

$H = J \sum_i \{ S^x_i S^x_{i+1} + \epsilon (S^y_i S^y_{i+1} + S^y_i S^y_{i+1}) \} - g\mu_B \sum_i S_i \cdot H$ (1)

where, $J$ ($>0$) is an antiferromagnetic exchange constant, $\epsilon$ an anisotropic parameter, $g$ a $g$-value, $\mu_B$ the Bohr magneton and $H$ the magnetic field. The system with $\epsilon < 1$, $\epsilon = 1$ and $\epsilon > 1$ corresponds to the Ising-like, Heisenberg and XY-like antiferromagnetic chain, respectively. The quantum fluctuation, stemming from non-commutative properties of spin operators, plays a crucial role in determining its ground state properties. Indeed, the exact quantum mechanical ground state of the Heisenberg chain with spin $S = 1/2$, found by Bethe in 1931 [3], is a spin liquid state with no order, showing that the quantum fluctuation in this case disturbs the system in taking a long range ordered state even at absolute zero Kelvin. After a theoretical finding that the spin chain with $S = 1/2$ can be represented by a pseudo-fermion model [4, 5], an important conclusion has been extracted [6, 7], i.e. the spin liquid state belongs to a universality class called a Tomonaga-Luttinger liquid (TLL). The TLL includes most 1D quantum systems having a gapless elementary
excitation with a linear energy dispersion, such as a linear chain conducting electron system. In general, a TLL has no long range order even at zero Kelvin, but it is in a quantum critical state with intrachain correlations of algebraic decay. A distinctive feature of the TLL in the spin chain is that two kinds of the correlation develop there as shown in Fig. 1. One is the staggered correlation of the transverse spin component perpendicular to the chain direction. The other is the incommensurate correlation of the longitudinal component parallel to the chain. One may associate the former with the usual Néel order, but the later has no classical analog and is peculiar to the quantum spin chain.

The TLL nature which appears in the $S = 1/2$ Ising-like antiferromagnetic spin chain, brings the curious ordering found in this study. The Ising-like chain shows an interesting quantum phase transition in magnetic fields. Different from the Heisenberg system, the ground state of the Ising-like chain has a long range order at zero magnetic field, because the Ising-like anisotropy stabilizes the Néel state. The external field, however, induces the strong quantum fluctuation, that drives the system into the TLL phase above a certain critical field. The idea underlying our results concerns with the real quasi-1D compounds, in which interactions between the chains inevitably exist. The interchain interactions make the TLL phase unstable, and can thereby lead the system into the long range ordering at a finite temperature. On this situation, between two kinds of the correlation mentioned above, the most dominant one will grow rapidly and build up the order. An important point for the Ising-like chain is that the incommensurate correlation is enhanced above the critical field, in contrast to the Heisenberg system, for which the staggered correlation is always dominant in a magnetic field. Thus, we propose that a density-wave-like incommensurate ordering appears for the quasi 1D Ising-like antiferromagnet in the field-induced region, when the temperature is lowered enough to make the interchain interaction relevant. In this ordered state, the spins align to be collinear along the chain direction with modulation of those amplitude, characterized by the incommensurate wave number $2k_F$. This ordering is essentially different from that expected for the classical Ising-like antiferromagnet with no quantum fluctuation. The field-induced transition in the classical case is a spin-flop type, and the Néel order of the transverse component, which relates to the breaking of rotational symmetry around the field, appears in the field-induced region as well as the Heisenberg antiferromagnet. On the other hand, the incommensurate ordering discussed here occurs as a consequence of the breaking of quasi-continuous translation symmetry. According to the pseudo-fermion model, this incommensurate ordering is regarded as a charge density wave (CDW) ordering for the pseudo-fermion. The modulation wave number $2k_F$ of this incommensurate ordering can be easily tuned by applying magnetic fields, since it is determined by the magnetization per spin $m$ as $2k_F = \pi(1 - 2m)$. This means that the periodicity of the incommensurate structure varies continuously with the field strength. This property is also different from that of the density-wave ordering in the conducting electron systems, of which $2k_F$ depends on the electron occupation number and is therefore little affected by external perturbations. The density-wave ordering in the conducting electron systems is arisen by the nesting of Fermi surface, whereas the incommensurate ordering in the quasi 1D Ising-like $XXZ$ case is entirely due to the quantum fluctuation inherent in the system.

In order to find this curious ordering, neutron diffraction measurements, which provide direct information about the spin structure, are particularly useful. For this measurement, we adopt the Co$^{2+}$ spin system BaCo$\text{V}_2\text{O}_8$, in which magnetic Co-O chains are running along the crystallographic $c$-axis. Recently, this compound was revealed to be a good realization of quasi-1D $S = 1/2$ Ising-like antiferromagnet with the transition field $H_c \approx 3.9$ T that can be achieved in current neutron facilities. The field-temperature phase diagram of BaCo$\text{V}_2\text{O}_8$, obtained from our thermodynamic measurements, are depicted in Fig. 2. BaCo$\text{V}_2\text{O}_8$ undergoes Néel ordering at $T_N = 5.4$ K at zero magnetic field, but the ordered temperature is rapidly lowered by the external field along the chain, which corresponds the easy axis direction. The suppression of the Néel order by magnetic fields can be understood by an appearance of the TLL nature in the field, which was mentioned for the Ising-like chain before. However, we recently found that at very low temperatures below 1.8 K, another ordered phase emerges in the field-induced region above $H_c \approx 3.9$ T. In the ordered phase in the field-induced region, we expect a realization of the incommensurate spin structure.

We have performed the neutron diffraction measurements in the following condition. A single crystal of BaCo$\text{V}_2\text{O}_8$, having a shape of a plate with $6 \times 6 \times 26$...
In neutron scattering measurements, only field dependence of the scan profile of the (4 0 3) axis and at temperature below 2 K. Figure 3 shows the was conducted in magnetic fields up to 5 T along the horizontal collimator sequence was guide-80'-80'-open. The fixed incident neutron energy was 13.7 meV. The horizontal collimator was used for the field generation. The pair superconducting magnet manufactured by Oxford Instruments, UK, was used for the field generation. The measurement were conducted in horizontal fields up to 5 T. A split-pair superconducting magnet manufactured by Oxford Instruments, UK, was used for the field generation. The fixed incident neutron energy was 13.7 meV. The horizontal collimator was guide-80'-80'-open. The cooling of the sample was achieved by a 3He-4He dilution refrigerator.

FIG. 3: Magnetic field dependence of neutron diffraction profiles of (4 0 l) scan measured at temperature $T = 0.85 \text{K}$ (a), 1.5 K (b) and 2 K (c). Solid lines are the results of fits to Gaussian functions.

Now let us turn to our neutron diffraction investigation of the spin structure in BaCo$_2$V$_2$O$_8$. The measurement was conducted in magnetic fields up to 5 T along the $c$-axis and at temperature below 2 K. Figure 3 shows the field dependence of the scan profile of the (4 0 3) reflection in the fields. In neutron scattering measurements, only the component of the magnetization that is perpendicular to the scattering vector contributes to the scattering intensity [21]. Thus, to detect the magnetic ordered component parallel to the chain direction, we adopt the $l$-scan around (4 0 3). In fact, other reflections such as (4 0 1) are more suitable to detect the magnetic component along the $c$ axis. However, these reflections do not satisfy the configurational condition of our measurement system, which is restricted by the windows of the superconducting magnet. At zero magnetic field, a peak at (4 0 3), which corresponds to the Néel order of the magnetic moments along the chain, is observed. According to the extinction rule, the (4 0 3) nuclear reflection in I41/acd space group is prohibited. Thus, the observed (4 0 3) peak is purely magnetic, and we confirmed a disappearance of the peak above $T_N = 5.4$ K. The (4 0 3) peak gradually diminishes with increasing the field up to 3.75 T, and then a sudden change of the scan profile occurs around the transition field $H_c$. In the field-induced region above $H_c$, two peaks at positions incommensurate with the underlying lattice appear at temperatures below 1.5 K. The sudden change of the scan profile reflects the fact that the transition at $H_c$ is weakly first order as suggested from our previous thermodynamic measurements [12]. A tiny peak at (4 0 3) remains in the field-induced region, but its origin is not clear at the moment. The positions of the peaks are plotted in Fig. 3. The difference between the peak positions in field ascending and descending processes is small. The incommensurate peaks shift continuously in such a way that a distance between the two peaks increases with increasing the field. Slight temperature dependence of the peak positions is found for $H > H_c$. The line width for all the peaks, observed in the ordered region, is within a resolution limit of our apparatus, and we confirm that

FIG. 4: Magnetic field dependence of the peak position of the observed neutron scan profile. The data are extracted from the least-square fits to the neutron scan profile. Inset shows the field dependence of a normalized incommensurate modulation $2k_F/\pi$. The theoretical curve is obtained by the calculation based on the Bethe ansatz exact theory.
the incommensurate peak, observed from the h-scan, is also resolution-limited. The resolution-limited peaks correspond to the development of the long range order. The scan profiles, observed at 4.5 T, show that the Bragg peaks change to broad diffuse ones at 1.5 K, that is just above the ordering temperature, and then disappear with increasing the temperature. Our observation unambiguously demonstrates a realization of the incommensurate ordering in the field-induced phase of BaCo$_2$V$_2$O$_8$. In our experimental accuracy, no peak is found in the l-scan profile around (0 0 1) at which the neutron cross section includes solely transverse component of magnetic moment. The result indicates the density-wave-like ordering with collinear spin alignments along the chain directions in the field-induced phase. Satellite reflections, coming from a higher harmonic Fourier components of the incommensurate modulation for the ordered structure, are not observed in the scan profile of the (4 0 l) reflection. Thus, the incommensurate modulation is suggested not to be square-wave like but is close to proper sinusoidal. In the inset of Fig. 4, we plot the field dependence of a normal-field-induced phase. Satellite reflections, coming from a higher harmonic Fourier components of the incommensurate modulation for the ordered structure, are not observed in the scan profile of the (4 0 l) reflection. Thus, the incommensurate modulation is suggested not to be square-wave-like but is close to proper sinusoidal. In the inset of Fig. 4, we plot the field dependence of a normalized incommensurate modulation $2k_F/\pi$ along the c-axis, which is given by a relation $2k_F/\pi = 1 - \delta$. Taking into account the fact that four Co$^{2+}$ ions are included in a chemical unit of BaCo$_2$V$_2$O$_8$ along the chain, the $\delta$ is obtained from 1/8 of a distance between two incommensurate peaks. As anticipated, the $2k_F/\pi$ continuously decreases with increasing the field above $H_c$. The experimental result slightly deviates from the theoretical prediction $2k_F = \pi(1 - 2m)$ with increasing field as shown in the inset of Fig. 4. The theoretical curve is obtained by the calculation based on the Bethe ansatz exact theory with the parameters $J/k_B = 65$ K, $\epsilon = 0.46$ and $g = 6.2$, which are estimated from the magnetization curve. The observed two incommensurate peaks reflect an existence of two kinds of domain for the ordered structure. The left and right peaks correspond to the domain with the modulation wave vector pointing parallel and antiparallel to the field direction in the chain, respectively. The difference of the intensity between the two peaks probably originates from the imbalance in two domains caused by the external field.

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