論文の内容の要旨

Investigation of Quantum Critical Phenomena in Quasi-one-dimensional Magnets by Low-temperature Magnetization and Specific-heat Measurements

(極低温磁化・比熱測定で探る擬一次元磁性体の量子臨界現象)

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This dissertation presents an experimental study of quantum critical phenomena at magnetic-field-induced quantum critical points (QCPs) in three (quasi-)one-dimensional (1D) quantum magnets by means of dc magnetization and specific-heat measurements at low temperatures. This dissertation covers three related topics (Chapters 3-5), as well as the background theory behind the experimental results (Chapter 1).

Experimental details used in the present work are described in Chapter 2. Dc magnetization measurements were performed by a Faraday-force technique. After an explanation of the principle of the method, an attempt to improve the sensitivity of the measurements, which was essential for the present work, is described. Specific-heat measurements were performed by a standard quasi-adiabatic heat-pulse method and a relaxation method.

In Chapter 3, the quantum criticality of the magnetization of $Cu(C_4H_4N_2)(NO_3)_2$, or CuPzN for short, near the saturation field $H_s \sim 14\,T$ is discussed on the basis of the phenomenological theory for 1D free fermion gas. In our previous study, we proved that CuPzN is a practically perfect 1D spin-1/2 Heisenberg antiferromagnet by comparison of our magnetization data at the base temperature of 80 mK with numerical calculations and the phenomenological theory. In this dissertation, we improve the cooling efficiency of the samples in the dilution refrigerator and achieve magnetization measurements at the lowest temperature of 39 mK, which is comparable to the energy scale of the interchain interactions of CuPzN $J' \sim 0.046\,K$. Much better agreement of the magnetization curve with well-known Bethe ansatz calculations at 0 K is obtained at 39 mK, implying that the interchain interactions are irrelevant near H_s even at this temperature. Applying a universal relation derived from 1D free fermion theory, all the magnetization curve near H_s in the temperature range from 39 mK to 350 mK are well scaled into a single curve. A small deviation of the curve from the theoretical prediction would be attributed to an effect of the repulsive interaction between the fermions. These facts confirm the strong one dimensionality of CuPzN and establishes the universality of the phenomenological theory for 1D free fermion gas near H_s in real magnets.

Chapters 4 and 5 deal with the critical exponent ν of the 3D ordering temperature near the critical field H_c, $T \propto |H_\mathrm{c}(T) - H_\mathrm{c}(0)|^{\nu}$. The compounds we focused on are spin-1/2 ferromagnetic-leg (FM-leg) ladders, in which leg interactions J_leg are ferromagnetic and rung interactions J_rung are antiferromagnetic. These new types of ladder compounds have been synthesized recently. We precisely determine the phase boundaries of two spin-1/2 FM-leg ladder compounds, 3-Br-4-F-V [3-(3-bromo-4-fluorophenyl)-1,5-diphenylverdazyl] and 3-I-V [3-(3-iodophenyl)-1,5-diphenylverdazyl], which consist of verdazyl-radical-based molecules, from temperature dependences of the magnetization ($M(T)$) and the specific heat ($C(T)$). The obtained critical exponents ν are discussed in the light of Bose-Einstein condensation (BEC) universality and quasi-one dimensionality.

3-Br-4-F-V is a strong-rung type ($|J_\mathrm{rung}/J_\mathrm{leg}| > 1$) FM-leg ladder and has two field-induced QCPs: the lower critical field $H_{c1} \sim 5\,\mathrm{T}$, where a spin gap is destroyed, and the saturation field $H_{c2} \sim 9\,\mathrm{T}$. We demonstrate that a crossover temperature can be determined from the broad peaks and dips in $M(T)$, and it moves in proportion to the magnetic field near the QCPs. This behavior could arise from the quasi-one dimensionality of 3-Br-4-F-V. A systematic change of the 3D ordering temperatures T_c is defined from anomalies in the temperature derivative of $\chi(T) = M(T)/H$, $d\chi(T)/dT$, and $C(T)$. Anomalies in $d(T\chi(T))/dT$ are also discussed to confirm that the difference of the definitions of T_c does not affect the critical exponent ν. The critical exponents ν obtained from a temperature-window technique are in good agreement with the three-dimensional (3D) BEC universality, $\nu = 2/3$, near both of the QCPs in the limit $T \to 0$. No sample dependence is observed for the critical exponents. These results have proven that the verdazyl-radical-based FM-leg ladders are promising as a new model system to study BEC physics.

By contrast, 3-I-V is a strong-leg type ($|J_\mathrm{rung}/J_\mathrm{leg}| < 1$) FM-leg ladder antiferromagnet ($T_\mathrm{N} =$ 1.4 K) and has only one field-induced QCP, the saturation field $H_c \sim 5.5\,\mathrm{T}$. In a preceding study, a nontrivial phase was reported to exist near H_{c2} on the basis of the magnetization ($M(H)$) and the specific heat ($C(H)$) measurements. We firstly reexamine this nontrivial phase near H_{c2}. The second-order field derivative of $M(H)$, d^2M/d^2H, shows a peak indicating the saturation and a shoulder-like anomaly, but $C(H)$ shows only a single sharp peak, different from the previous report. We consider that the broad peak of $C(H)$ in the previous data stemmed not from double phase boundary but from a collapse of the sharp peak due to sample inhomogeneity. The 3D ordering phase boundary is defined from cusp-like anomalies in $\chi(T)$ and peaks in $C(T)$. Anomalies in $d(T\chi(T))/dT$ and $d\chi(T)/dT$ are also discussed to check whether the difference of the definitions of T_c would affect the critical exponent ν. The critical exponent ν obtained from a sliding-window technique shows the nontrivial critical exponent $\nu = 1$, different from the conventional 3D BEC exponent $\nu = 2/3$, as slightly moving away from the QCP. The $\nu = 1$ region is common to all the definitions of T_c. The nontrivial critical exponent could be attributed to the 1D nature of the strong-leg-type ferromagnetic-leg ladder and the frustrations of the intra- and inter-ladder interactions.

The difference of the critical exponents in the FM-leg ladder compounds derived from low dimensionality and frustration would provide novel insight into BEC physics beyond the conventional 3D BEC universality in other quasi-1D quantum magnets.