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Contrast between static- and mobile-impurity effects on Haldane-gap system Y_2BaNiO_5 studied by specific heat

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Abstract

The spin entropy estimated from Schottky-like peak in the temperature dependence of specific heat in magnetic field is well explained by the edge state model for both of static and mobile impurities in the title system. We found sharp contrast in exchange interaction between edge (and impurity) spins for these two types of impurities. In the case of static impurity, the result is consistent with a picture that two edge spins adjacent to an impurity interact with each other by small exchange constant. In the case of mobile impurity, on the other hand, the result supports a picture that impurity (carrier) spin and adjacent two edge spins behave freely without interaction between them, possibly due to the motion of the carrier.

Keywords: Haldane gap; Y_2BaNiO_5; Impurity effect; Specific heat

An one-dimensional $S = 1$ antiferromagnetic Heisenberg chain is known to show a gap (the so-called Haldane gap) in the spin excitation spectrum. The essence of its ground state is explained by the valence bond solid (VBS) state. One of the hallmarks of the VBS state is an edge state created by impurity doping, where edge spin appears at the edge of the finite chain cut by the impuriry. The edge spin has been confirmed in NENP by ESR experiments [1,2]. ESR detects excitation energy between eigenstates with different spin configurations. In Ref. [1], the ESR spectrum depends on the magnetic field direction, which is analyzed successfully by the introduction of an anisotropic exchange interaction between edge and impurity spins.

Y_2BaNiO_5 is a Haldane gap system with a large spin gap. This compound has an unique characteristic feature that it can be doped with two different types of impurities (up to high doping level): (i) static impurity, Mg, doped into Ni site on the chain, which is non-magnetic; (ii) mobile impurity, Ca (divalent), doped into Y (trivalent) site apart from the chain, which introduces a hole on the chain, although it is localized. In order to compare states created by these two types of impurities, we have studied the specific heat in magnetic fields of impurity doped Haldane-gap system Y_2BaNiO_5, instead of ESR often used thus far. The temperature dependence of the specific heat reflects the thermal distribution among eigenstates, which appears as a Schottky peak whose temperature roughly corresponds to the excitation energy.

In this study, we have used two single crystal samples grown by the traveling-solvent floating-zone (TSFZ) method [3]. One is static-impurity-doped $Y_2BaNi_{1-y}Mg_yO_5$ ($y = 0.04$), and the other is mobile-impurity-doped $Y_{2-x}Ca_xBaNiO_5$ ($x = 0.012$). Temperature-dependent specific heat in magnetic field, $C(T, H)$, was measured using the thermal relaxation method.

The results for static and mobile impurities are shown in Figs. 1 and 2. Since the results for $H \| c$ are similar to those of $H \| b$, the results only for $H \| a$ and b are shown, where the chain runs parallel to a-axis. The dashed lines show the lattice contribution to the specific heat, $C_{\text{lattice}}(T)$, estimated from the non-doped sample. With impurity doping, a Schottky-like peak appears for both types of impurities, indicating that spin degrees of freedom are induced by impurities. Entropy estimated by the integration of $(C(T) - C_{\text{lattice}}(T))/T$ up to

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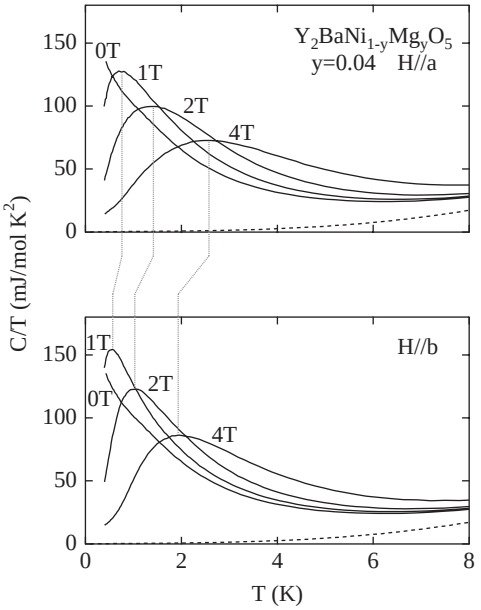

Fig. 1. The temperature dependence of $C(T,H)/T$ of static-impurity-doped $Y_2BaNi_{1-y}Mg_yO_5$ ($y = 0.04$) for $H\|a$ (chain) and b. The dashed lines show lattice contribution estimated from non-doped samples.

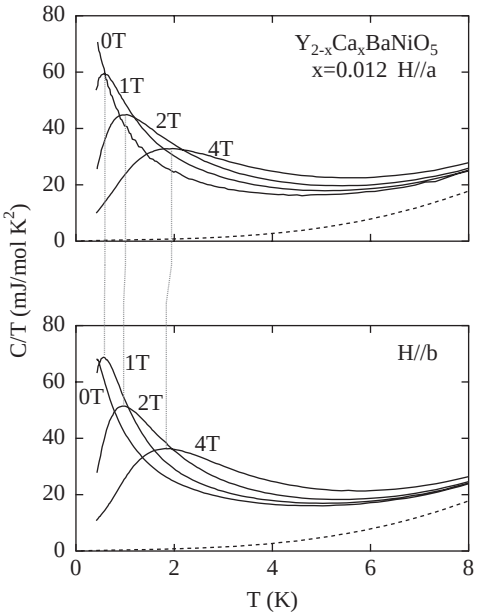

Fig. 2. The temperature dependence of $C(T,H)/T$ of mobile-impurity-doped $Y_{2-x}Ca_xBaNiO_5$ ($x = 0.012$) for $H\|a$ (chain) and b. The dashed lines show lattice contribution estimated from non-doped samples.

$T = 20$ K is 0.43 J/mol K for the Mg doped sample and 0.20 J/mol K for the Ca doped one. According to the edge spin model, spin entropy is $2 \times R \ln(2 \times 1/2 + 1) \times 4\% = 0.46$ J/mol K for the former, assuming two edge spins per impurity, and $(2+1) \times R \ln(2 \times 1/2 + 1) \times 1.2\% = 0.21$ J/mol K for the latter, assuming two edge spins and an impurity spin per impurity where R is gas constant. This good coincidence supports the edge spin picture. The result that carriers doped by Ca substitution show Schottky-like behavior with the reasonable edge spin entropy is consistent with the localization of carriers [3].

The peak position shifts to higher temperatures with increasing field for both types of impurities, reminiscent of Schottky anomaly. In the case of static impurity (Mg), the peak shift, i.e. excitation energy, is anisotropic, which is similar to the ESR result reported in Ref. [1]. Our rough fitting shows that this anisotropy can be explained by the introduction of an anisotropic ferromagnetic exchange interaction of $\sim$10 K between edge spins adjacent to the Mg site with two-site interval apart. Therefore, the static-impurity doping is understood by a picture of edge spins and interaction between them.

In the case of mobile impurity (Ca), on the other hand, the peak shift is isotropic including the results for $H\|c$ (not shown). This indicates that the interaction between impurity (carrier) spin and adjacent edge spins is almost negligible. It is natural to expect that exchange interaction between spins for the Ca case with one-site interval apart is stronger than that between spins for the Mg case with two-site interval apart. Therefore, our result for mobile impurity suggests that interaction is weakened by the motion of the carriers that can hop in the scale of localization length.

Our results have made clear the contrasted effects of static- and mobile-impurity doping on exchange interaction between edge (and impurity) spins in the Haldane-gap system.

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