Systematic Study of Magnetic Interactions in Insulating Cuprates
Yoshiaki Mizuno, Takami Tohyama and Sadamichi Maekawa

Abstract: The magnetic interactions in one-dimensional, two-dimensional (2D) and ladder cuprates are evaluated systematically by using small Cu-O clusters. We find that the superexchange interaction $J$ between nearest neighbor Cu spins strongly depends on Cu-O structure through the Madelung potential, and in 2D and ladder cuprates there is a four-spin interaction $J_{\text{cyc.}}$, with magnitude of 10 % of $J$. We show that $J_{\text{cyc.}}$ has a strong influence on the magnetic excitation in the high-energy region of 2D cuprates.

A variety of insulating cuprates affords us an opportunity to study the magnetic properties of low-dimensional systems. Recent experiments for insulating cuprates have revealed interesting characteristics of magnetic interactions: The superexchange interaction between nearest neighbor Cu spins $J$ remarkably depends on Cu-O network structure $J$, and additional interactions such as a four-spin (4S) interaction are important for ladder $J$ and two-dimensional (2D) cuprates $J$. These characteristics indicate the necessity to establish proper magnetic descriptions for the cuprates. In this paper we perform a systematic study of magnetic interactions for one-dimensional (1D), 2D, and ladder cuprates theoretically.

A starting model to describe the electronic states of cuprates is the $d$-$p$ model, in which hopping integrals between Cu$3d$ and O$2p$ orbitals ($T_{pd}$) and between O$2p$ orbitals ($T_{pp}$), an energy-level separation between the Cu$3d$ and O$2p$ orbitals ($\Delta$), and Coulomb interactions on Cu and O sites are taken into account. $T_{pd}$ and $T_{pp}$ are obtained by considering not only the bond length dependence but also the effect of the Madelung potential around Cu and O ions. We find that the potential enhances the magnitudes of $T_{pd}$ and $T_{pp}$ in the 1D cuprates as compared with those in the 2D ones $J$. In the two-leg ladder compounds such as SrCu$_2$O$_3$, $T_{pp}$ along the leg of the ladder is enhanced by the Madelung potential due to adjacent two-leg ladders. These enhancements play an important role in the dependence of $J$ on the dimensionality. The $\Delta$ is determined from the difference in the Madelung potential between Cu and O sites.

The magnetic interactions are evaluated by mapping the lowest several eigenstates of small clusters (Cu$_5$O$_7$, Cu$_4$O$_{12}$, and Cu$_6$O$_{17}$) for the $d$-$p$ model onto those of the corresponding Heisenberg-type model $J$. For 2D systems, we take into account not only $J$, but also a diagonal interaction $J_{\text{diag}}$ and 4S interaction $J_{\text{cyc.}}$ in the model: $H = J \sum_{(i,j)} S_i \cdot S_j + J_{\text{diag}} \sum_{(i,j)} S_i \cdot S_j + J_{\text{cyc.}} \sum \text{plaquette}(P_{ijkl} + P_{ijkl}^{-1})$, where $S_i$ is a spin operator at site $i$, and $J_{\text{cyc.}}$ is defined as the coefficient of the 4S cyclic permutation operators $P_{ijkl}$ and $P_{ijkl}^{-1}$, which can be rewritten by using the two-spin interaction $(S_i \cdot S_j)$ and the four-spin interactions $(S_i \cdot S_j)(S_k \cdot S_l)$. For ladder systems, we distinguish between the nearest neighbor interactions along the leg ($J_{\text{leg.}}$) and along the rung ($J_{\text{rung}}$) of the ladder.

The calculated results are summarized in Table 1, where we take La$_2$CuO$_4$, SrCu$_2$O$_3$, and Sr$_2$CuO$_3$ as typical systems of 2D, ladder and 1D cuprates, respectively (see Refs. 1 and 5 for the parameters used in the calculations). We find that $J$ in the 1D cuprate is larger than that in the 2D one. This is caused by the enhancement of the hopping integrals in 1D cuprates as mentioned above. For 2D cuprates, we obtain $J$ to be $\sim$0.15 eV, consistent with the experimental values $J$. In addition, we find that $J_{\text{cyc.}}$ is 7 % of $J$, while $J_{\text{diag}}$ is zero. These results are consistent with a previous cluster calculation $J$, and a re-
Table 1
The magnetic interactions obtained by mapping the eigenstates of Cu2O7, Cu4O12, and Cu6O17 clusters for Sr2CuO3, La2CuO4, and SrCu2O3, respectively, onto the corresponding Heisenberg-type models. The numbers in parentheses denote the deviation in the last significant digit.

| Material    | J [eV] | J_{diag} [eV] | J_{cyc} [eV] |
|-------------|--------|---------------|--------------|
| Sr2CuO3     | 0.17   | --            | --           |
| La2CuO4     | 0.146(1) | 0.00(0)     | 0.011(1)     |
| SrCu2O3     | J_{leg}: 0.195(5) | 0.003(2)   | 0.018(2)  |
|             | J_{rung}: 0.15(2)         |              |             |

Figure 1. The magnetic excitation spectra of a 4×4 Heisenberg model with J and J_{cyc}.

Next, in order to examine the effect of J_{cyc} on the magnetic excitation, we calculate the dynamical spin-correlation function S(q,ω) for 2D cuprates. Figure 1 shows the dispersion and the intensity of S(q,ω) for a 4×4 Heisenberg model with J=0.146 eV and J_{cyc}=0.011 eV. For comparison, the result for J_{cyc}=0 are also shown. We find that the intensity is not sensitive to J_{cyc}, while the dispersion is strongly suppressed in the high-energy region. In particular, it is worth noting that ω(q) at q=(π/2,π/2) becomes smaller than that at q=(π,0). This is in contrast with the case of J_{cyc}=0, in which the magnetic zone boundary (q=(π,0)→(π/2,π/2)) has a flat dispersion. Therefore, it is desirable that inelastic neutron-scattering experiments in the wide energy region be performed to verify the role of J_{cyc}, that is, the suppression of the dispersion at (π/2,π/2).

In summary, we have evaluated the magnetic interactions in various cuprates systematically. We have shown that an ionic nature inherent in insulating cuprates is important for the material dependence of J. We found that J_{cyc} is ~10 % of J, and greatly influences the magnetic excitation spectra in 2D cuprates.

This work was supported by a Grant-in-Aid for Scientific Research on Priority Areas from the Ministry of Education, Science, Sports and Culture of Japan, CREST and NEDO. The parts of the numerical calculation were performed in the Supercomputer Center in ISSP, University of Tokyo, and the supercomputing facilities in IMR, Tohoku University.

REFERENCES
1. Y. Mizuno, T. Tohyama and S. Maekawa, Phys. Rev. B 58, R14713 (1999).
2. M. Matsuda, K. Katsumata, R. S. Eccleston, S. Brehmer and H.-J. Mikeska, unpublished.
3. J. Lorenzana, J. Eroles and S. Sorella, Phys. Rev. Lett. 83, 5122 (1999).
4. Y. Mizuno, T. Tohyama and S. Maekawa, J. Low Temp. Phys. 117 389 (1999).
5. H. J. Schmidt and Y. Kuramoto, Physica C 167, 263 (1990).
6. J.C. Johnston, Phys. Rev. B 54, 13009 (1996).