Electronic Excitations in a One-Dimensional Cuprate, $CuGeO_3$

M. V. Zimmermann,[1] J. P. Hill,[1] C.-C. Kao,[2] T. Ruf[3], T. Gog,[4]
C. Venkataraman,[4] I. Tsukada,[5] K. Uchinokura[5]

[1] *Department of Physics, Brookhaven National Laboratory, Upton, NY, U.S.A.*
[2] *National Synchrotron Light Source, Brookhaven National Laboratory, Upton, NY, U.S.A.*
[3] *Max Planck Institute, Stuttgart, Germany*
[4] *CMC-CAT, Advanced Photon Source, Argonne National Laboratory, IL, U.S.A.*
[5] *ISSP, Tokyo, Japan*

Introduction

Understanding the electronic properties of the transition metal oxides in general, and the cuprates in particular, remains at the center of condensed matter research today. Key questions include the roles played by the strong electron correlations and the reduced dimensionality common to these materials. In elucidating these effects, studies of the electronic excitations are particularly valuable, providing stringent tests of the various theoretical approaches. Here we report inelastic x-ray scattering studies of the electronic excitation spectrum of a one-dimensional cuprate, $CuGeO_3$.

Methods and Materials

Inelastic x-ray scattering has a number of strengths in this endeavor. There are no final-state effects or problems with charging, and it offers bulk-like penetration and a large range of accessible momentum and energy transfers. Resonant enhancements[1] have allowed studies to be carried out on a number of cuprates, utilizing the Cu K-edge.[2-4]

$CuGeO_3$ is a charge transfer insulator. Its crystal structure consists of CuO_4 units arranged in one-dimensional edge-sharing chains running along the c-axis. At temperatures below T_{SP} = 14K, the Cu^{2+} spins drive a spin-Pierels transition in which the lattice is dimerized along the c-axis.

We have carried out studies at 9IDB at CMC-CAT. The optics consisteed of a Si(111) monochromator, a Si(333) secondary monochromator and a flat Al mirror. A Ge(733) spherically bent analyzer crystal (R = 1 m) was used to collect the scattered radiation. The overall resolution was 0.3 eV (FWHM).

Results

An energy loss scan measured at resonance is shown in Fig. 1. These data were taken at room temperature, with the momentum transfer set to the antiferromagnetic zone center, and along the chain direction, q = (0,0,1.5). Three excitations are observed, with excitation energies of 1.7 eV, 3.8 eV, and 6.5 eV, respectively.

On the basis of theoretical calculations,[5] we identify the feature at 1.7 eV with an optically forbidden *d-d* excitation. In the cuprates, such orbital excitations are localized. The sharp feature at 3.8 eV is just below the optical gap in this material, and we tentatively ascribe this to a charge-transfer exciton. Finally, the feature at 6.5 eV appears to be similar to features observed in other cuprates[2,3,4] and we identify this with a charge transfer excitation to the antibonding state, i.e. $3d^9$ to $3d^{10}\underline{L}$.

We have also measured the dispersion (q-dependence) of the excitations, shown in Fig. 2.

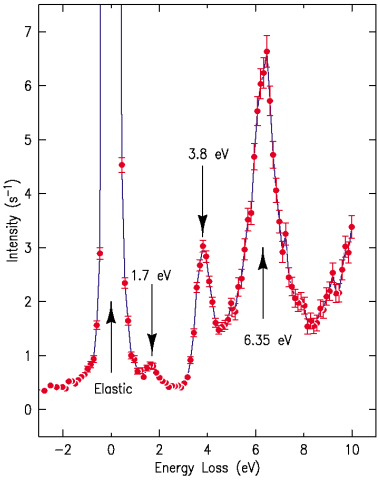

FIG. 1. Excitation spectrum of $CuGeO_3$. Data were taken at room temperature, with the momentum transfer aligned along the chain axis.

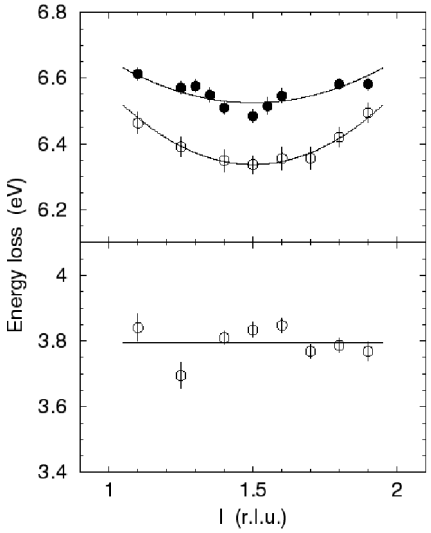

FIG. 2. Dispersion of the 6.5-eV feature and 3.8-eV feature. Data were taken at room temperature (open circles) and 10K (closed circles).

Discussion

The observation of dispersion in the 6.5-eV feature suggests that this excitation is propagating along the chain direction in a correlated hopping process. At room temperature, the system is paramagnetic, and there are essentially no magnetic correlations from one CuO_4 plaquette to the next. On cooling below TSP, the

system dimerizes and antiferromagnetic correlations are set up between nearest neighbor copper sites along the chain. The results above suggest that these magnetic correlations inhibit the propagation of the excitation, making it more localized. In addition, the excitation is pushed to higher energies. While some of this shift results from thermal contraction, the shift appears to be too large to be explained by such effects, suggesting that magnetic correlations also play a role in determining the excitation energy.

Acknowledgments

Discussions with G.A. Sawatzky are gratefully acknowledged. We thank BESSERC-CAT and UNI-CAT for giving us generous access to their equipment pool and CMC-CAT for their support. Work performed at Brookhaven National Laboratory was through the support of U.S. Department of Energy, Division of Materials Science, under contract no. DE-AC02-98CH10886. Use of the Advanced Photon Source was through the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under contract No. W-31-109-ENG-38.

References

[1] C.-C. Kao et al., Phys. Rev. B **54**, 16361 (1996).

[2] J.P. Hill et al., Phys. Rev. Lett. **80**, 4967 (1998).

[3] P. Abbamonte et al., Phys. Rev. Lett. **83**, 860 (1999).

[4] Z. Hasan et al., Science (2000).

[5] C. de Graag and R. Broer, Phys. Rev. B **62**, 702 (2000).