Resonant and nonresonant inelastic x-ray scattering in $CuGeO_3$

M.v. Zimmermann[1], J.P. Hill[1], C.-C. Kao[2], T. Gog[3], C. Venkataraman[3], A. Bommannavar[3], I. Tsukada[4], T. Masuda[4], and K. Uchinokura[4]
[1]*Dept. of Physics, Brookhaven National Laboratory, Upton, NY 11973 USA*
[2]*National Synchrotron Light Source (NSLS), Brookhaven National Laboratory, Upton, NY 11973 USA*
[3]*CMC-CAT, Argonne National Laboratory, Argonne, IL 60439 USA*
[4]*Dept. of Applied Physics, U. Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo, Japan*

Introduction

Transition metal oxides are presently the focus of much attention in condensed matter physics because of the diverse phenomena exhibited by these materials. Examples include antiferromagnetism, superconductivity, and colossal magnetoresistance. The origin of these phenomena lies in the strong electron correlations present in these materials, which place them between the well-understood limits of band insulators and simple metals. The presence of these correlations makes these materials hard to handle theoretically, and there is a need for more detailed experimental work, particularly with regard to the electronic structure and excitations.

Here, in contrast to other techniques, inelastic x-ray scattering can offer some advantages, including bulk sensitivity and lack of final-state effects. Unfortunately, the cross-section is small, limiting applicability of the technique. Recently, however, resonant enhancements were discovered in the hard x-ray regime on tuning the incident photon energy through an absorption edge [1–5]. While such enhancements greatly extend the materials that may be studied, the interpretation of the cross-section is problematic. In large part, this is because to date there have been no direct comparisons between resonant scattering and the well understood non-resonant limit.

We report here an inelastic x-ray scattering study of the insulating cuprate $CuGeO_3$. The purpose of this work is two fold: Firstly, to shed light on the electronic structure of a strongly correlated system and secondly, to explore the resonant cross-section in detail. In this latter regard, we report for the first time the observation of an excitation in both the resonant and nonresonant limits of the scattering.

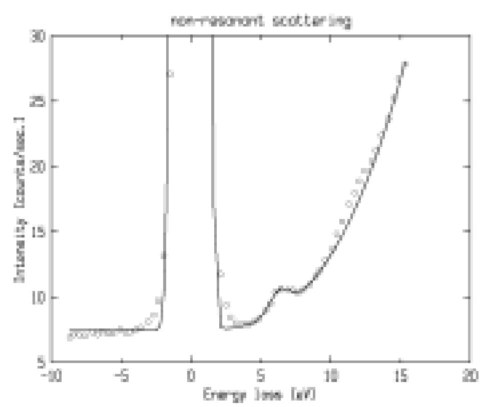
Methods and Materials

$CuGeO_3$ was chosen for this work because of its one-dimensional crystal structure (in contrast to previously studied two-dimensional cuprates) and because it has relatively low absorption, making it a good candidate to observe nonresonant scattering. It has previously attracted attention as the only inorganic spin Pierels compound–a dimerization of the Cu-O chains occurs at $T_{SP} = 14$ K. The chains run along the c-axis.

The experiments were performed at CMC-CAT's 9-ID beamline utilizing a cryogenically cooled Si(111) double-bounce monochromator together with an Al harmonic rejection mirror. The scattered radiation was collected and the energy analyzed with a spherically bent Si(553) analyzer. The energy bandwidth of the incident beam, measured with the analyzer in backscattering ($\theta_B = 88.5^o$), was 1.2 eV. At resonance ($\theta_B = 77.3^o$), finite size effects degraded the overall resolution to 1.8 eV (FWHM or full width half maximum). All data reported here were taken at room temperature with $q = 1.6\ A^{-1}$.

Results

In the first experiments, a search was carried out for the non-resonant signal. The final energy was held fixed at 8.77 keV (well below the Cu K-edge) to maximize the sample volume, and the energy loss was scanned by varying the incident energy. The results are shown in Figure 1, in which the scattered intensity is plotted as a function of energy loss. The a-axis was parallel to q and the polarization vector, ε, parallel to the c-axis. The sampling time was nine minutes per data point. A small peak is observed at 6.4 eV on top of the strong tail of the oxygen plasmon at ~35 eV. The 6.4 eV feature is a charge transfer excitation (i.e., from Cu 3d to O 2p). The fit to a Gaussian including a sloping background (solid line) reveals an intensity of 1.7 counts s^{-1} and width of 1.57 eV FWHM, slightly broader than the



elastic line (FWHM = 1.21 eV).

Figure 1: Inelastic x-ray scattering in $CuGeO_3$, far from any absorption edges.

The incident photon energy was then tuned to the Cu K-edge, and energy loss scans taken at different incident energies by scanning the final energy (Figure 2). The peak at ~6.5 eV is dramatically enhanced, with a maximum intensity of ~220 counts s^{-1}. These data allow us to quantify the resonant enhancement of the inelastic cross-section for the first time. After correcting for absorption, we estimate the enhancement to be a factor of 285.

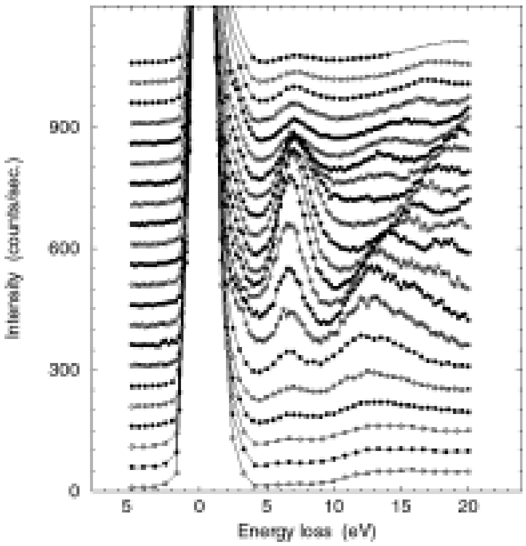


Figure 2: Inelastic scattering in the vicinity of the Cu K-edge. Scans were taken at incident energies 8.98–9.002 keV and offset vertically for clarity.

The position of the enhancement is strongly dependent on the relative orientation of the incident polarization and the Cu-O chains. This is illustrated in Figure 3 in which the intensity at 6.5 eV energy loss is plotted as a function of incident photon energy. A peak is observed at 8.989 keV when ε is parallel to the b-axis, while the peak is at 9.000 keV when ε is parallel to the c-axis.

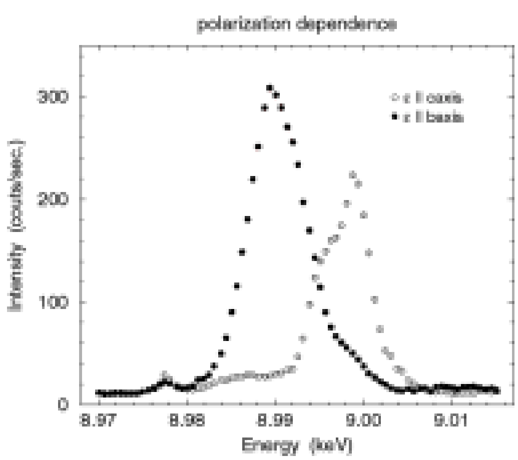
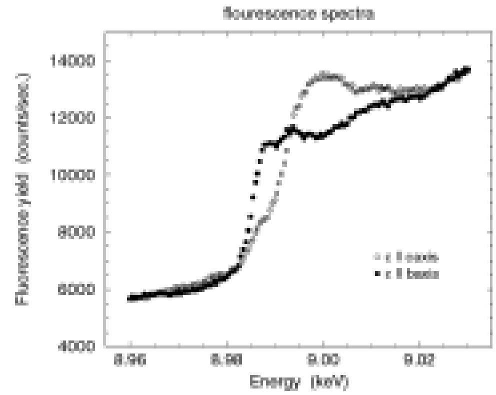


Figure 3: Incident energy dependence of the 6.5 eV energy loss feature for two different relative orientations of the incident polarization.

In order to elucidate the origin of this polarization dependence, we have measured the fluorescence yield for the two polarizations (Figure 4). When the polarization of the incident beam is parallel to the b-axis, two features are observed in the absorption spectrum, one at 8.989 keV and the other at 8.994 keV. These have been previously



identified as arising from transitions into the $4p_y$

Figure 4: Polarization-dependent fluorescence spectra for $CuGeO_3$.

antibonding state [6]. The splitting arises from the well-screened and poorly screened final states, $|\underline{c}3d^{10}\underline{L}4p_y>$ and $|\underline{c}3d^9\, 4p_y>$, respectively. Conversely, if the polarization is aligned along the c-axis, a transition into $4p_z$ takes place. Here, the features due to the well-screened and the poorly screened states are barely resolved at 8.999 keV and 9.004 keV.

Comparison of Figure 3 and Figure 4 reveals that for polarization parallel to the b-axis, the resonance at 8.989 keV is due to transitions into the well-screened $4p_y$ antibonding state. The width of the resonant enhancement is about 8 eV, significantly broader than the Cu $\underline{1s}$ lifetime (1.6 eV), suggesting that the poorly screened state might also contribute to the enhancement. Similarly, when the polarization is along the c-axis, the maximum enhancement is observed at 8.999 keV and is again associated with the well-screened $4p_z$ state.

The observation of a resonant enhancement associated with the well-screened intermediate states in one-dimensional $CuGeO_3$ is in contrast to observations in the two-dimensional $NdCuO_4$ for which only the poorly screened intermediate states gave rise to resonant enhancements. It was argued that in the latter case, nonlocal effects suppressed the resonance associated with these intermediate states [2, 6], and it expected that such nonlocal effects would be less effective in a one-dimensional material.

Finally, we note that the constant energy loss scan in Figure 3 shows also a small enhancement of the scattered intensity at 8978 eV, the energy of the pre-edge transition from the 1s into the 3d states.

Discussion

We have probed the charge transfer excitation in $CuGeO_3$ with inelastic x-ray scattering. For the first time, the same excitation has been seen on and off resonance. We find a resonant enhancement of ~300. The enhancement arises primarily through transitions with the well-screened 4p intermediate states, in contrast to previous observations in $NdCuO_4$.

Acknowledgments

Work supported at Brookhaven by the U.S. Department of Energy, Basic Energy Sciences, Office of Science, under Contract No. DE-AC02-98CH10886. Use of the Advanced Photon Source was supported by the U.S. Department of Energy, Basic Energy Sciences, Office of Science, under Contract No. W-31-109-Eng-38.

References

[1] C.-C. Kao, W.A.L. Caliebe, J.B. Hastings, and J.-M. Gillet, *Phys. Rev. B* **54,** 16361 (1996).
[2] J.P. Hill, C.-C. Kao, W.A.L. Caliebe, M. Matsubara, A. Kotani, J.L. Peng, and R.L. Greene, *Phys. Rev. Lett.* **80,** 4967 (1998).
[3] P. Abbamonte, C.A. Burns, E.D. Isaacs, P.M. Platzman, L.L. Miller, S.W. Cheong, and M.V. Klein, *Phys. Rev. Lett.* **83**, 860 (1999).
[4] P.M. Platzman and E.D. Isaacs, *Phys. Rev. B* **57**, 11107 (1998).
[5] K. Hamalainen, J.P. Hill, S. Huotari, C.-C. Kao, L.E. Berman, A. Kotani, T. Ide, J.L. Peng, and R.L. Greene, *Phys. Rev. B* **61,** 1836 (2000).
[6] D.Z. Cruz, M. Abbate, H. Tolentino, P.J. Schilling, E. Morikawa, A. Fujimori, and J. Akimitsu, *Phys. Rev. B* **59,** 12450 (1999).