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Inelastic neutron scattering from $Tl_2CaBa_2Cu_2O_8$

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Abstract. Coherent inelastic neutron scattering measurements have been carried out on the high temperature superconductors $Tl_2CaBa_2Cu_2O_8$ (Tl-2122, $T_c = 107$ K) and $YBa_2Cu_3O_7$ (Y-123, $T_c = 92$ K), at the Dhruva reactor at Trombay. The density of phonon states in Tl-2122 is enhanced at 6–17 meV and reduced at 40–70 meV compared to that in Y-123.

Keywords. Inelastic neutron scattering; phonon density of states; high temperature superconductors; $Tl_2CaBa_2Cu_2O_8$; $YBa_2Cu_3O_7$; lattice dynamics.

1. Introduction

While the role of phonons in superconductivity of the new ceramic materials continues to be an open question, the lattice dynamics is of fundamental importance in understanding the interatomic interactions and many related properties in these solids. We have undertaken to investigate and compare the phonon spectra in $Tl_2CaBa_2Cu_2O_8$ (Tl-2122, $T_c = 107$ K) and $YBa_2Cu_3O_7$ (Y-123, $T_c = 92$ K), by the technique of coherent inelastic neutron scattering. While the data in Tl-2122 perhaps represent the first reported phonon spectra in this material, those in Y-123 are in reasonable agreement with earlier reports (see e.g. Renker *et al* 1988; Natkaniec *et al* 1988). It appears that the density of phonon states in Tl-2122 is enhanced at low energies of 6–17 meV and reduced at 40–70 meV as compared to that in Y-123.

There are very few cases when medium flux reactors have been used to obtain density of phonon states of coherent neutron scatterers, particularly those with the complexity of high-T_c ceramics. We have recently demonstrated (Chaplot *et al* 1989) the feasibility of such work at the Dhruva reactor, which has a maximum neutron flux of $1{\cdot}8 \times 10^{14}$ neutrons/cm^2/s, by carrying out experiments on tetracyanoethylene. We have used the triple axis spectrometer which is provided with the fully automatic computer control data collection and operation system.

2. Experimental

The details of the experimental technique of inelastic neutron scattering and data analysis are similar to those in the work on tetracyanoethylene (Chaplot *et al* 1989). The incident neutron energy was varied using a copper (111) monochromator; neutrons of fixed-scattered energy (E_f) were counted in different scans for $E_f = 14{\cdot}8$, 30 and 50 meV using a pyrolytic graphite (002) analyser and in the constant momentum transfer mode.

Bulk samples of single phase Tl-2122 were synthesized by a modified matrix reaction method starting from a stoichiometric composition (Sequeira *et al* 1988 and references therein). The matrix used was $CaBa_2Cu_2O_5$ which was prepared by reacting a mixture of $BaCO_3$, $CaCO_3$ and CuO at 1120 K for 24 h, followed by heating at 1200 K for 48 h with several intermediate grindings. The pellets of mixture of the matrix and Tl_2O_3 powder were wrapped in a Pd–Ag alloy foil to avoid Tl-losses, and reacted at 1200 K for 15 min according to a quick reaction method, avoiding any moisture during synthesis. The X-ray diffraction pattern confirmed the sample to be the pure Tl-2122 single phase (Hewat *et al* 1988). Also the superconducting transition temperature corresponding to the zero electrical resistance state was confirmed to be 107 K. Sequeira *et al* (1988) reported a neutron diffraction study on this material giving the composition as $Tl_{1.81}Ca_{0.93}Ba_2Cu_2O_{7.86}$. About 20 g of the Tl-2122 sample in aluminium container was used in our inelastic neutron scattering experiments.

The measurements of Y-123 were made using a smaller sample of about 10 g, which was synthesized by the oxalates coprecipitation method (Phatak *et al* unpublished). The composition of the sample to be $YBa_2Cu_3O_{6.89}$ and its single phase nature were ascertained respectively by chemical analysis and X-ray diffraction methods.

3. Results

The inelastically-scattered neutron intensity is proportional to the dynamical structure factor $S(Q, E)$ of the material, where Q and E denote the wave vector and energy transfer respectively. $S(Q, E)$ can be converted to the neutron-weighted phonon density of states $g^{(n)}(E)$, which however has some Q-dependence due to coherent scattering. So far we have obtained reasonably good data corresponding to $Q = 6\,\text{Å}^{-1}$ in both Tl-2122 and Y-123, and we are in the process of obtaining more data from several different constant-Q scans for Q in the range 4–7 Å^{-1}.

The present data of $6\,\text{Å}^{-1}$ given in figure 1 are quite useful for making a good first comparison of the phonon spectra in the two compounds. The data analysis has been carried out according to Chaplot *et al* (1989), and the resulting phonon density

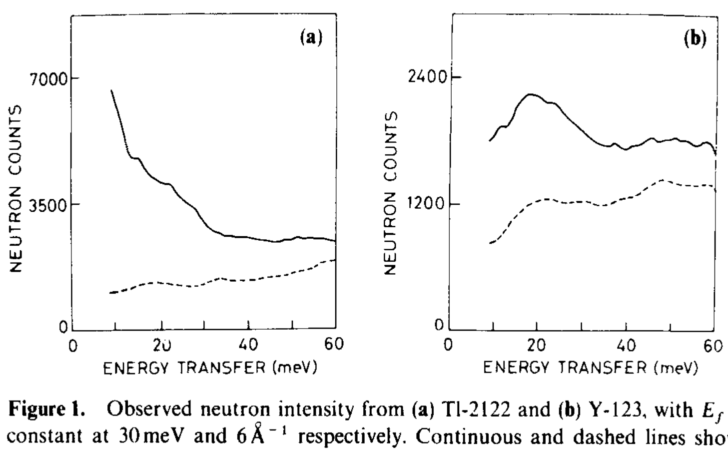

Figure 1. Observed neutron intensity from (**a**) Tl-2122 and (**b**) Y-123, with E_f and Q held constant at 30 meV and $6\,\text{Å}^{-1}$ respectively. Continuous and dashed lines show raw data and background contribution respectively.

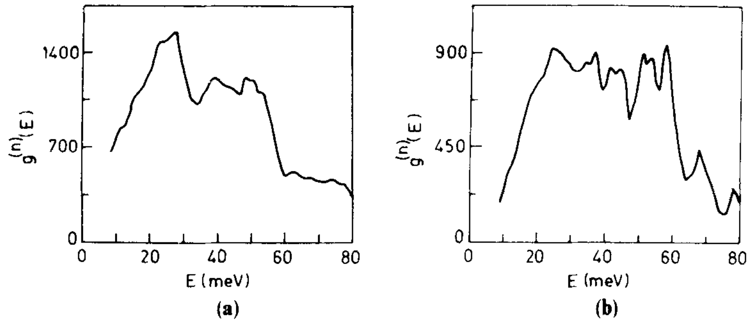

Figure 2. Neutron-weighted phonon density of states for (**a**) Tl-2122 and (**b**) Y-123 for $Q = 6\,\text{Å}^{-1}$. The energy resolution varied from 5 to 25 meV in the range of E from 0 to 80 meV.

of states are given in figure 2. A small multiphonon contribution is expected to be similar for the two compounds. These neutron-scattering cross-section weighted spectra have the largest contribution from the oxygen atoms since the weightage factors $4\pi b^2/m$(b = scattering length and m = mass) are as follows.

Tl	Ca	Y	Ba	Cu	O
4·8	7·5	8·5	2·5	11·8	26·5 (10^{-2} barn/amu).

The observed density of states in figure 2 in the low-energy region below 17 meV is considerably enhanced for Tl-2122 compared to that for Y-123. This is more clearly evident in the raw intensity data in figure 1. On the other hand, the density is reduced at energies above 40 meV for Tl-2122. These important differences should form a basis for evaluating any lattice-dynamical models for these materials.

Considering that our results refer to $Q = 6\,\text{Å}^{-1}$ only and do not involve Q-averaging, the results on Y-123 are in fair agreement with earlier experimental data of Renker *et al* (1988) and Natkaniec *et al* (1988), and also the lattice-dynamical calculations of the neutron-weighted density of states of Chaplot (1988) and Chaplot (1990). Similar calculations for Tl-2122 are not available to the best of our knowledge.

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