ESRF	Experiment title: Phonon-coupled metal-insulator transition as a function of temperature and pressure, and the magnetic phase diagram in $SmNiO_3$	Experiment number: HS-2082
Beam line: ID22N	Date of experiment: from: 24/09/2003 to: 10/10/2003	Date of report: 08/10/2004
Shifts: 21 (42/2)	Local contact(s): Alessandro BARLA and Rudolf RÜFFER	Received at ESRF:

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Report:

We have studied the changes in the samarium partial phonon density of states in the charge-transfer insulator $SmNiO_3$ using Nuclear Inelastic Scattering (NIS) in order to further elucidate the role of the strong electron-phonon interaction in the metal–insulator transition found in the entire $RNiO_3$ series. Furthermore the Sm magnetic moment has been measured both through the transition to the antiferromagnetic state of the nickel ions as well as through the ordering of the Sm ions using Nuclear Forward Scattering (NFS).

Amongst the larger family of charge-transfer insulators the rare earth nickelates provide ideal subjects for the study of a variety of solid state properties, due to the well-defined dependence in their case of these properties on the size of the rare-earth ion[1]. All display a MI transition (with the exception of R = La), and those heavier than Pr also show a transition to an antiferromagnetic state. The MI transition (T_{MI} for $SmNiO_3$ = 403K) is understood to arise from the tilting of the NiO_6 octohedra of the perovskite structure in order to optimise the R-O bond distances which results in the bending of the Ni-O-Ni bond[2,3]. These determine the Ni 3d and the O 2p orbital overlap and thus the valence bandwidth. A sufficiently large bandwidth causes the valence and conduction bands to overlap resulting in metallisation. The accompanying mechanisms of this are still not properly understood. The importance of a strong electron-phonon interaction at the transition has been confirmed[4] and was the main driving force behind this experiment.

Isotopically enriched $^{149}SmNiO_3$ was prepared using now standard techniques[5]. The measurements were carried out in 16-bunch mode at beam line ID22N. A new high resolution monochromator for the energy of the ^{149}Sm resonance (22.494 keV) was used which delivers a flux of $8 \cdot 10^7$ photons/s in a bandwidth of ~ 0.9 meV and is therefore well suited for both NIS and NFS studies[6]. The high temperature NIS spectra were measured for substantially more than one day per point, allowing three temperatures to be measured. For the magnetic studies all the required temperature points to be taken, with special emphasis on the regions where the Ni and Sm moments order.

The NIS results are shown in Figure 1. A clear softening of the low energy modes (around 8 K) is seen in the spectra at and above 350 K. Such changes well below T_{MI} have also been seen in a Raman study on $NdNiO_3$[7]. A theoretical study of the phonon dynamics to aid in the understanding of this result is now underway in collaboration with J.K. Dewhurst at the Karl Franzens Universität, Graz. Use will be made of a FP-LAPW basis code which is better suited to rare-earth compounds than usual pseudopotential methods[8].

NFS spectra have been taken over the full ranges of interest, with much improved statistics over the preliminary study, due to the use of the enriched sample. Clear ordering of the rare-earth sublattice is seen (see spectra in Figure 2). A full analysis of the data is underway.

As a final point it should be noted that this was a shared beam time, along with proposal HE-1594, which explains the long period over which the shifts were granted.

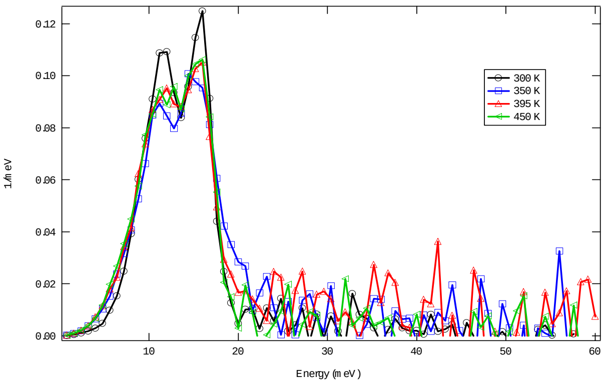

Figure 1: Samarium partial phonon density of states of $SmNiO_3$ as a function of temperature through the metal-insulator transition. Of particular interest is the softening at low energies (~8 K) well below the transition temperature (T_{MI}=400 K).

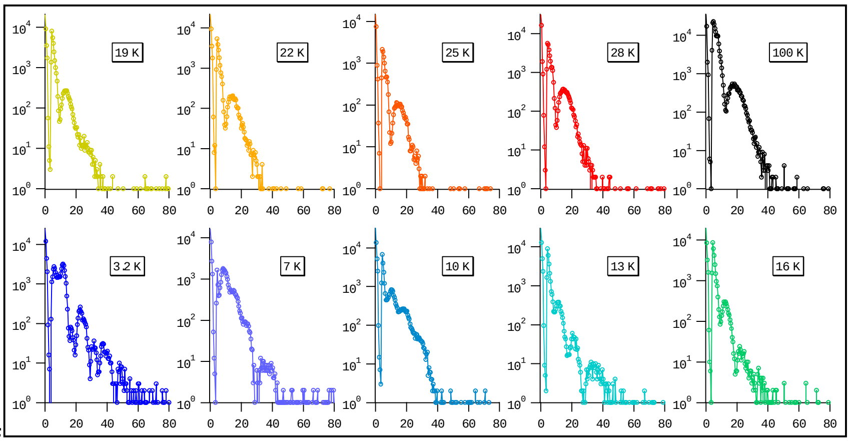

Figure 2: NFS data of $SmNiO_3$ at various temperatures showing the hyperfine beating due to the ordering of the Sm moments at low temperature.

References:

1) For an early review, see M.L. Medarde, J. Phys.: Condens. Matter, 9 (1997) 1679; see also J.A. Alonso et al., Phys. Rev. Lett., 82 (1999) 3871
2) P. Lacorre et al., J. Solid State Chem., 91 (1991) 225
3) J.B. Torrance et al., Phys. Rev. B, 45 (1992) 4414
4) M. Medarde et al., Phys. Rev. Lett., 80 (1998) 2397
5) see J.A. Alonso et al., J. Am. Chem. Soc., 121 (1999) 4754 and references there-in
6) A. Barla, J.P. Sanchez, Y. Haga, G. Lapertot, B.P. Doyle, O. Leupold, R. Rüffer, M.M. Abd-Elmeguid, R. Lengsdorf and J. Floquet, Phys. Rev. Lett., 92 (2004) 066401
7) M. Zaghrioui et al., J. Magn. Magn. Mater., 211 (2000) 238
8) http://physik.kfunigraz.ac.at/~kde/secret_garden/exciting.html