

# Supporting Information

## Dipole-Oriented Molecular Solids Can Undergo a Phase Change and Still Maintain Electrical Polarization

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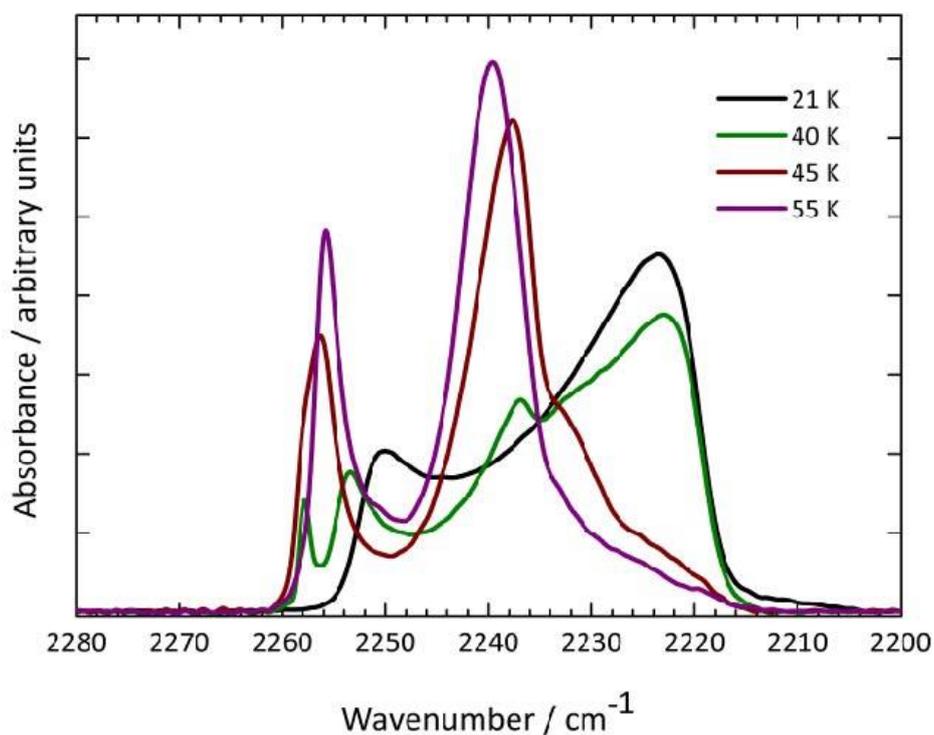
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**SP - Figure 1.** Reflection absorption infrared spectroscopy (RAIRS) data for films of nitrous oxide deposited at different temperatures on a silica substrate. All spectra were recorded at the temperature of deposition. Only data for the  $\nu_{\text{NN}}$  peak are shown. Deposition at 21 K gives rise to two broad peaks, with maxima at  $2252 \text{ cm}^{-1}$  and  $2225 \text{ cm}^{-1}$  and these are assigned as the longitudinal optical (LO) and transverse optical (TO) phonon modes respectively. Thus the spectrum recorded at 21 K relates to the low temperature phase of nitrous oxide as described in the main body of text. Deposition at 55 K gives rise to a spectrum which relates to the high temperature phase of nitrous oxide, with the LO mode peak at  $2256 \text{ cm}^{-1}$  and the TO mode peak at  $2240 \text{ cm}^{-1}$ . Deposition at 40 K gives rise to a film whose spectrum contains elements related to both the low temperature and the high temperature phases. And deposition at 45 K gives rise to a film which predominantly shows peaks related to the high temperature phase. From this data set it can be concluded that the phase of nitrous oxide grown can be selected by deposition above or below 45 K. Deposition below 45 K gives the low temperature phase of nitrous oxide while deposition above 45 K gives the high temperature phase. This is consistent with the surface potential measurements presented in Fig. 4a in the main body of text and more precisely with the temperature regime in which it is necessary to introduce a change to the spontelectric model parameters. The RAIRS data that resulted from annealing experiments, Fig 2 in the main body of text, indicate that the phase change occurs upon annealing above 48 K.