Random Ordering in Solid Hydrogen Films: NMR studies

Kiho Kim (University of South Alabama), J. R. Bodart (Chipola College), N.S. Sullivan (UF, Physics)

Introduction

NMR studies of molecular hydrogen in monolayer films adsorbed on hexagonal boron nitride for ortho-H_2 concentrations 48<x[o-H_2]<69% and temperatures ($0.2 \leq T \leq 1K$) provide information about the ordering of the ortho molecules in the temperature and concentration region where frustration and reduced dimensionality favor random ordering. The observed NMR line shapes were distinct from those reported for bulk samples. This difference is attributed to the substrate crystal field which is not negligible compared to the electrostatic quadrupole-quadrupole interactions between two adjacent H_2 molecules. We report calculated line shapes assuming random ordering in the plane parallel to the film.

Results and Discussion

The calculated NMR line shapes are based on two hypotheses: (i) The fluctuations of the molecular orientation are very fast compared to the intramolecular dipolar frequency, and (ii), in the absence of interactions that break time-reversal symmetry, the dipole moments are all zero. With these assumptions, the NMR spectrum is given by a sum of Pake-powder spectra $\Pi(\sigma,\omega)$ for each value of σ that occurs in the distribution $P(\sigma)$ where $-0.5 < \sigma < 1$. The resulting NMR line shape $S(\omega)$ is given by:

$S(\omega) = \int_{-1/2}^{1} d\sigma P(\sigma)\Pi_{\pm}(\sigma,\omega)$, where $\Pi_{\pm}$ denotes the well-known Pake doublet, with $\Pi_{+} = \frac{1}{2\sqrt{3}}\frac{1}{|\sigma|}\frac{1}{\sqrt{1-\frac{\omega}{\sigma}}}$ for $-2 < \frac{\omega}{\sigma} < 1$, and $\Pi_{-} = \frac{1}{2\sqrt{3}}\frac{1}{|\sigma|}\frac{1}{\sqrt{1+\frac{\omega}{\sigma}}}$ for $-1 < \frac{\omega}{\sigma} < 2$. The simplest choice for $P(\sigma)$ is a uniform distribution, and the resulting line shapes are shown in Figure 1 for a range of local order parameters 0.23<σ<0.33.

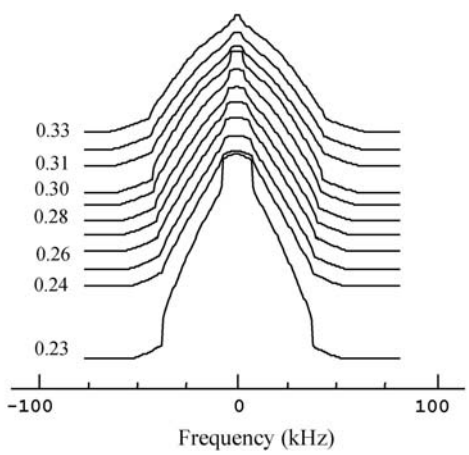

Figure 1. Calculated NMR lineshapes for P(σ) = constant, following convolution with a Gaussiam broadening function with an rms width of 0.39 kHz.

Conclusions

The calculated NMR line shapes for a broad structureless probability distribution of orientational order parameters provides a good fit to the observed line shapes. In contrast, a triangular function P(σ) ~ σ is deduced from NMR studies for bulk H_2.

References

[1] Kim, K et al., AIP Conf. Proceed. **850**, 64-65 (2006).