

## NEUTRON INCOHERENT SCATTERING FROM KDP\*

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**Abstract**—The incoherent, inelastic neutron cross section is derived for  $\text{KH}_2\text{PO}_4$  following the model discussed by Tokunaga and Matsubara. This expression should give the dominant contribution of the proton tunneling motion to the cross section. Matsubara has given the coherent cross section for KDP. However the scattering in hydrogen is primarily incoherent.

### INTRODUCTION

IN A RECENT paper Tokunaga[1] derived the coherent, neutron cross section for the proton tunneling motion in  $\text{KH}_2\text{PO}_4$  (KDP). Neutron scattering from hydrogen is primarily incoherent (98 per cent). Since the dominant contribution in neutron scattering in this case is incoherent, we extend Tokunaga's calculations to include the incoherent cross section.

Several papers[2-5] describe KDP as having the hydrogens occupying sites in a double minimum potential well on the hydrogen bonds which connect adjacent  $\text{PO}_4$  groups. For a more detailed description the reader is referred to references [2-5]. Our calculations indicate that the proton tunneling between the two possible sites on the hydrogen bonds should be observed most easily in the incoherent cross section for a single crystal of KDP.

### CALCULATIONS

In the model proposed in reference [2], the proton field operators are taken to be

$$\psi(\mathbf{r}) = \sum_{v,l} b_{vl} \phi_{vl}(\mathbf{r}) \quad (1)$$

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where

$$b_{vl} = \begin{pmatrix} b_{vl1} \\ b_{vl2} \end{pmatrix} \quad (2)$$

and 'v' labels the hydrogen bonds, l labels the two minima in the potential. The  $\phi$ 's are taken to be orthonormal wave functions, localized at the potential minima. (We will later choose these to be harmonic oscillator wave functions.)

$b_{vlj}^\dagger$  is a fermion creation operator and the b's satisfy the anticommutation relations

$$[b_{vlj}, b_{v'l'j'}^\dagger]_+ = \delta_{vv'} \delta_{ll'} \delta_{jj'} \quad (3)$$

The condition that there is one proton on each bond is

$$\sum_{l,j} b_{vlj}^\dagger b_{vlj} = 1. \quad (4)$$

Using these relations the Hamiltonian is[2]

$$\begin{aligned} H = & -2\Omega \sum_{vj} (b_{v1j}^\dagger b_{v2j}^\dagger + b_{v2j}^\dagger b_{v1j}) \\ & - \frac{1}{2} \sum_{\substack{vj \\ v'j'}} J_{v'v} (b_{v1j}^\dagger b_{v1j'} - b_{v2j}^\dagger b_{v2j'}) \\ & \times (b_{v'1j'}^\dagger b_{v'1j'} + b_{v'2j'}^\dagger b_{v'2j'}). \end{aligned} \quad (5)$$

In the molecular field approximation[4], we can write H as

$$\begin{aligned} H = & - \sum_{vj} [2\Omega (b_{v1j}^\dagger b_{v2j} + b_{v2j}^\dagger b_{v1j}) \\ & + J\langle Z \rangle (b_{v1j}^\dagger b_{v1j} - b_{v2j}^\dagger b_{v2j})] \end{aligned} \quad (6)$$

where

$$\langle Z \rangle = \left\langle \sum_j (b_{v1j}^\dagger b_{v1j} - b_{v2j}^\dagger b_{v2j}) \right\rangle_T \quad (7)$$

Equation (6) is easily transformed into

$$H = \sum_{vj} \lambda_+ \alpha_{vj}^\dagger + \alpha_{vj} + \sum_{vj} \lambda_- \alpha_{vj}^\dagger - \alpha_{vj} \quad (8)$$

where

$$\lambda_\pm = \pm \sqrt{((2\Omega)^2 + (J\langle Z \rangle)^2)} \quad (9)$$

and

$$\alpha_{v\pm} = c_{1\pm} b_{v1} - c_{2\pm} b_{v2} \quad (10)$$

Noting that  $F_{ll'vv'}(\boldsymbol{\kappa}) = F_{lv}(\boldsymbol{\kappa})$  due to the orthonormality of the  $\phi_{lv}$  we get

$$\begin{aligned} \frac{d^2\sigma^{inc}}{d\Omega d\epsilon} &= \frac{k'}{k} |a_{inc}|^2 \sum_{v'} F_{lv}(-\boldsymbol{\kappa}) F_{lv'}(\boldsymbol{\kappa}) q_l^2 q_{l'}^2 \frac{1}{\mathcal{F}} \\ &\times \{ \delta(\epsilon) (e^{-\beta\lambda_+} p_1^2 p_2^2 + e^{\beta\lambda_+}) \\ &+ (e^{-\beta\lambda_+} \delta(\epsilon - 2\lambda_+) + e^{\beta\lambda_+} \delta(\epsilon + 2\lambda_+)) \\ &\times p_l p_{l'} \} \quad (16) \end{aligned}$$

$$\begin{aligned} c_{1\pm} &= \frac{J\langle Z \rangle \pm \sqrt{(J^2\langle Z \rangle^2 + (2\Omega)^2)}}{\sqrt{(2(J^2\langle Z \rangle^2 + (2\Omega)^2 \pm J\langle Z \rangle \sqrt{(J^2\langle Z \rangle^2 + (2\Omega)^2))}} \\ c_{2\pm} &= \frac{2\Omega}{\sqrt{(2(J^2\langle Z \rangle^2 + (2\Omega)^2 \mp J\langle Z \rangle \sqrt{(J^2\langle Z \rangle^2 + (2\Omega)^2))}} \quad (11) \end{aligned}$$

For unpolarized neutrons, the cross section is

$$\begin{aligned} \frac{d^2\sigma}{d\Omega d\epsilon} &= \frac{k'}{2\pi\hbar k} \int_{-\infty}^{\infty} dt \\ &\times e^{i\epsilon t/\hbar} \{ |a_{inc}|^2 \langle \boldsymbol{\rho}(-\boldsymbol{\kappa}) \cdot \boldsymbol{\rho}(\boldsymbol{\kappa}, t) \rangle_T \\ &+ |a_{coh}|^2 \langle \rho_0(-\boldsymbol{\kappa}) \rho_0(\boldsymbol{\kappa}, t) \rangle_T \} \quad (12) \end{aligned}$$

where

$$a_{mc} = a_+ - a_-$$

$$\boldsymbol{\rho}(\boldsymbol{\kappa}) = \int d^3r \boldsymbol{\psi}^\dagger(\boldsymbol{r}) e^{-i\boldsymbol{\kappa} \cdot \boldsymbol{r}} \mathbf{J} \boldsymbol{\psi}(\boldsymbol{r}) \quad (13)$$

$\mathbf{J}$  is the spin of the target nuclei. The components of  $\mathbf{J}$  are the Pauli matrices, and

$$\rho_0(\boldsymbol{\kappa}) = \int d^3r \psi^\dagger(\boldsymbol{r}) e^{-i\boldsymbol{\kappa} \cdot \boldsymbol{r}} \psi(\boldsymbol{r}). \quad (14)$$

The second term on the right-hand side of (12) is discussed in some detail by Matsubara[1]. Consequently, we will concern ourselves exclusively with the first term, the incoherent part.

Using (1) we see that

$$\boldsymbol{\rho}(\boldsymbol{\kappa}) = \sum_{\substack{vv' \\ ll'}} F_{ll'vv'}(\boldsymbol{\kappa}) b_{l'v'}^\dagger \mathbf{J} b_{lv}. \quad (15)$$

where

$$\begin{aligned} F_{lv} &= \int d^3r e^{-i\boldsymbol{\kappa} \cdot \boldsymbol{r}} |\phi_{lv}|^2 \\ p_1 &= -\frac{J\langle Z \rangle + \lambda_+}{2\Omega} \\ p_2 &= -\frac{J\langle Z \rangle - \lambda_+}{2\Omega} \\ q_1 &= \frac{2\Omega}{[2(\lambda_+^2 - J\langle Z \rangle \lambda_+)]^{1/2}} \\ q_2 &= -\frac{(J\langle Z \rangle - \lambda_+)}{[2(\lambda_+^2 - J\langle Z \rangle \lambda_+)]^{1/2}}. \quad (17) \end{aligned}$$

$\mathcal{F} = 2 \cosh \beta\lambda_+$  is the partition function per proton[2].

### DISCUSSION

We can see the properties of neutron scattering from the hydrogen tunneling motions more clearly by taking an explicit model for the proton wave functions. First we define  $\mathbf{d}_v$  with magnitude  $d$  and direction  $\Omega_v$  to be the vector distance between the potential minima on the  $v$ th bond.

We now take the  $\phi$ 's to be harmonic oscillator ground state wave functions with

characteristic frequencies  $\omega_d$  for motions along the bond and  $\omega_r$  for motions perpendicular to the bond. With this model we have

$$F_{1v} = e(-(\boldsymbol{\kappa} \cdot \boldsymbol{\Omega}_v)^2/4 a^2 - (\boldsymbol{\kappa} \times \boldsymbol{\Omega}_v)^2/4 b^2)$$

$$F_{2v} = e^{i\boldsymbol{\kappa} \cdot \mathbf{d}_v} F_{1v} \quad (18)$$

where

$$a^2 = m\omega_d/\hbar$$

$$b^2 = m\omega_r/\hbar.$$

These  $F$ 's give the inelastic part of (16) as

$$\frac{d^2\sigma_{inc}}{d\Omega d\epsilon} = \frac{k'}{k} |a_{inc}|^2 \frac{1}{\mathcal{F}} \{ e^{-\beta\lambda_+} \delta(\epsilon - 2\lambda_+) + e^{\beta\lambda_+} \delta(\epsilon + 2\lambda_+) \}$$

$$(x) \sum_v |F_{1v}|^2 \frac{2\Omega^2}{\lambda_+^2} (1 - \cos \boldsymbol{\kappa} \cdot \mathbf{d}_v). \quad (19)$$

We can see from (19) that the inelastic cross section vanishes for  $\boldsymbol{\kappa}$  oriented perpendicularly to  $\mathbf{d}_v$ . Such an orientation can be made to within  $3^\circ$  by keeping  $\boldsymbol{\kappa}$  along the  $c$  axis of the crystal. This effect along with the diffraction effect from the factor  $(1 - \cos \boldsymbol{\kappa} \cdot \mathbf{d}_v)$  should serve to distinguish the contribution of the tunneling motion from the other motions in the crystal.

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