NONLINEAR JUMP DIFFUSION MODEL APPLIED TO THE INCOHERENT THERMAL NEUTRON SCATTERING

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A "nonlinear" Multi-Sublattice Jump Diffusion model (MSJD) was developed. "Equations of motion" analogous to the collision term in the Boltzmann transport equation were derived. The model was applied in the theory of incoherent thermal neutron scattering. The incoherent double differential cross-section in the multi-phonon approximation is obtained in terms of the MSJD model. The model can be applied to systems where jump diffusion occurs.

1. Introduction

In the last years, neutron diffraction and inelastic neutron scattering techniques have used extensively to study both the structure and the proton dynamics in the transition--metal hydrides [1–10]. The investigation of the dynamic behaviour of hydrogen in metals by inelastic and quasi-elastic neutron scattering is favourized by the high and mainly incoherent scattering cross-section of the proton [7].

The first theory of quasi-elastic neutron scattering by hydrogen in metals has been developed by Chudley and Elliott [11] for primitive interstitial site lattices. This theory has been extended to non-primitive interstitial lattices by Blaesser and Peretti [12] and Rowe et al. [13].

Another method for calculating the quasi-elastic scattering law has been given by Gissler and Rother [14] who used a random flight technique. Recently, the influence of finite jump times has also been discussed [7].

Diffusion models based on quantum mechanics suggested by some authors (see e. g. [15]) could not be proved due to the fact that the corresponding scattering functions have not been evaluated.

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In the present paper we have extended our previous work [16] in the following points:

(1) The migration of impurity atoms is described by the nonlinear jump diffusion equations which only in special case transform into a system of linear jump equations [16].

(2) The multi-phonon formula for the incoherent two-fold differential cross-section has been obtained as a generalization of the quasi-elastic expression.

It should be emphasized here that the fit to experimental data obtained for hydrides of Group Vb metals, using the earlier published models, was not satisfactory [6].

2. *Basic formalism*

As shown by Van Hove [17], the incoherent, differential cross-section per atom, in the Born approximation, for the scattering of neutrons from a system of atoms into a unit solid angle Ω and unit range ε is given by the equation

$$\frac{d^2\sigma_{\text{inc}}}{d\Omega d\varepsilon} = \frac{a_{\text{inc}}^2 k'}{\hbar k_0} S(\vec{\kappa}, \omega) = \frac{a_{\text{inc}}^2 k'}{2\pi\hbar k_0} \int d\vec{r}dt \exp\left[i(\vec{\kappa}\vec{r} - \omega t)\right] G_s(\vec{r}, t). \tag{1}$$

Here a_{inc}^2 is the bound incoherent scattering length of atom; the incident neutrons have a wave number $\vec{k}_0$, the outgoing neutrons have the wave number $\vec{k}$; $\vec{\kappa}$ is the scattering vector, $\vec{\kappa} = \vec{k}_0 - \vec{k}'$; $\hbar\omega$ is the energy transfer during the scattering process, $\hbar\omega = \hbar^2(k_0^2 - k'^2)/2m$, m being the mass of the neutron. The self-correlation function $G_s(\vec{r}, t)$ gives in the classical approximation the probability density of finding at point $\vec{r}$ and time t the atom which was at the origin at time 0.

Assuming, after Chudley and Elliot [11], that the atomic motion is oscillatory about well-defined points between which the atom moves by diffusion, we write

$$G_s(\vec{r}, t) = \int d\vec{r}' G_s^{\text{D}}(\vec{r}', t) G_s^{\text{V}}(\vec{r} - \vec{r}', t), \tag{2}$$

where G_s^{D} describes the diffusive and G_s^{V} the vibrational motion. When writing Eq. (2), it has been assumed that the oscillatory and diffusive motions are completely uncorrelated, and that the jumps take place very rapidly so that the oscillatory motion proceeded for the whole time interval t. It may be shown, basing on the Schofield [18] theory, that the convolution formula (2) is fulfilled also for the quantum correlation functions.

The Fourier transform of Eq. (2) is

$$S(\vec{\kappa}, \omega) = \int d\omega' S^{\text{D}}(\vec{\kappa}, \omega') S^{\text{V}}(\vec{\kappa}, \omega - \omega'). \tag{3}$$

Then the incoherent vibrational scattering law S^{V} is expanded into a sum of terms involving zero, one, two, etc. phonons, a multi-phonon formula [19] was obtained

$$S^{\text{V}}(\vec{\kappa}, \omega - \omega') = \sum_n \sum_{m=0}^{n} S_{n,m}^{\text{V}}(\vec{\kappa}, \omega - \omega'), \tag{4}$$

where $n = 0, 1, 2, \ldots$, indicate the n-phonon process, and m is the number of phonons created in the system. Basing on the general form of $S^{V}_{n,m}(\vec{\kappa}, \omega-\omega')$ [19] we finally obtain the incoherent two-fold differential cross-section, $d^2\sigma_{inc}/d\Omega d\varepsilon$, in the form

$$\frac{d^2\sigma_{inc}}{d\Omega d\varepsilon} = \frac{a_{inc}^2 k'}{\hbar k_0} \exp(-2W) \sum_n \left(\frac{\hbar}{2m_H N_1}\right)^n \sum_{m=0}^{n} \sum_{\lambda_1,\ldots,\lambda_n} \frac{1}{(n-m)!m!}$$

$$\times \overline{\prod_{p=1}^{n} |\vec{\kappa}\vec{e}^{\lambda_p}|^2} \frac{\exp(-\hbar\omega_{\lambda_p}/2k_B T)}{\omega_{\lambda_p}[1-\exp(-\hbar\omega_{\lambda_p}/2k_B T)]} \exp\left(-\sum_{s=1}^{n-m} \omega_{\lambda_s} + \sum_{s=n-m+1}^{n} \omega_{\lambda_s}\right)$$

$$\times S^{D}(\vec{\kappa}, \omega - \sum_{s=1}^{n-m} \omega_{\lambda_s} + \sum_{s=n-m+1}^{n} \omega_{\lambda_s}), \tag{5}$$

where $\exp(-2W)$ is the Debye-Waller factor, m_H is the mass of the hydrogen atom, N_1 is the number of the primitive cells in the crystal, $\vec{e}^{\lambda_p}$ is the polarization vector, the symbol $\overline{(\ldots)}$ denotes an average over all sublattices; ω_{λ_s} are the phonon frequencies, k_B is the Boltzmann constant, T is the temperature.

It should be noticed that for crystals with cubic symmetry this formula simplifies slightly; for a detailed description see for example Ref. [19].

One can see that the phonon line (5) is constructed by superposition of the respectively shifted diffusion scattering laws S^{D}.

For calculating the diffusion scattering law S^{D}, the Multi-Sublattice Jump Diffusion model [16] was developed in terms of which we directly constructed the diffusion self-correlation function G_s^{D}.

3. Physical foundations of the model

The jump diffusion theory for weakly interacted particles can be characterized, as in paper [16], by the following assumptions:

(1) Impurity particles, when trapped at any equilibrium sites for a finite mean residence time, perform rapid jumps to any neighbours with varying in space and even asymmetric jump times.

(2) Equilibrium sites form a lattice which, in general, is not of the Bravais type.

(3) In one equilibrium position only one particle may reside.The next postulate, suggested in paper [16], plays the principal role in the present theory. So we have

(4) The hopping probability can be influenced by the presence of other particles, so, for example, the occupation of the neighbouring sites, in general, cannot be excluded.

It deserves noting that postulates (3) and (4) are responsible for nonlinear form of the "equations of motion" presented in section below.

4. *Mathematical formulation*

4.1. "Equations of Motion"

Let us now apply the nonhomogeneous Markov chains with continuous time [20] to the jump diffusion problem. We can write immediately

$$\mathscr{P}_a^{\alpha}(\vec{l}, t+\tau') = \sum_{\beta,\vec{l}'} \mathscr{P}_a^{\beta}(\vec{l}', t) V_a^{\beta\alpha}(\vec{l}', \vec{l}; t, t+\tau'), \tag{6}$$

where $\mathscr{P}_a^{\alpha}(\vec{l}, t+\tau')$ is the probability of finding the a-th particle at time $t+\tau'$ at site α, in the primitive cell with a Bravais vector $\vec{l}$; $V_a^{\beta\alpha}(\vec{l}', \vec{l}; t, t+\tau')$ is the conditional probability of finding the a-th particle at site $(\alpha, \vec{l})$ at $t+\tau'$, respectively, when the same particle is at $(\beta, \vec{l}')$ at time t; $\alpha, \beta = 1, 2, \ldots, M$, M being the number of equilibrium positions in the unit cell of the host lattice; $a = 1, 2, \ldots, N$, N being the total number of impurity particles. For small τ' we can write the conditional probability in the form:

$$V_a^{\beta\alpha}(\vec{l}', \vec{l}; t, t+\tau') = W^{\beta\alpha}(\vec{l}'-\vec{l}; \tau')\,[1-\sum_{b\neq a} \mathscr{P}_b^{\alpha}(\vec{l}, t+\tau')]; \quad (\beta, \vec{l}') \neq (\alpha, \vec{l}), \tag{7}$$

where $W^{\beta\alpha}(\vec{l}'-\vec{l}, \tau')$ describes the one particle migration and is also the conditional probability of finding the particle at site $(\alpha, \vec{l})$ after time τ' later when, respectively, the same particle is at $(\beta, \vec{l}')$.

Notice that in Eq. (7) the expression in square brackets has the sense of probability that the site (α, l) is not occupied at time $t+\tau'$, whereas it should be a conditional probability that the position $(\alpha, \vec{l})$ is not occupied at time $t+$when the a-th particle is at time t in $(\beta, \vec{l}')$. However, it can be asumed that in the case of diffussion in the macroscopic crystal, and for weakly interacting particles, these two kinds of probabilities are equal with good approximation.

When writing the identity (7), the occupation of the equilibrium positions lying between the initial $(\beta, \vec{l}')$ and final $(\alpha, \vec{l}')$ sites was ignored.

Using Eqs (6) and (7) and applying the normalization conditions for $V_a^{\alpha\beta}$ functions, we finally obtain, after simple transformations, the jump diffusion "equations of motion".

$$\frac{\partial}{\partial t} \mathscr{P}_a^{\alpha}(\vec{l}, t) = \sum_{\beta,\vec{l}'} \left\{ \frac{\mathscr{P}_a^{\beta}(\vec{l}', t)\,[1-\sum_{b\neq a} \mathscr{P}_b^{\alpha}(\vec{l}, t)]}{\tau_{\beta\alpha}(\vec{l}'-\vec{l})} - \frac{\mathscr{P}_a^{\alpha}(\vec{l}, t)\,[1-\sum_{b\neq a} \mathscr{P}_b^{\beta}(\vec{l}', t)]}{\tau_{\alpha\beta}(\vec{l}-\vec{l}')} \right\},$$

$$\alpha = 1, \ldots, M; \quad a = 1, \ldots, N, \tag{8}$$

where the jump times $\tau_{\beta\alpha}$ are

$$\tau_{\beta\alpha}(\vec{l}'-\vec{l}) = \left[\lim_{\tau'\to 0} \frac{W_{\beta\alpha}(\vec{l}'-\vec{l}; \tau')}{\tau'} \right]^{-1}. \tag{8a}$$

It should be noticed that Eqs (8) are a system of coupled nonlinear rate equations, so the solution is not as simple as in the previous "linear" jump models [3, 16].

Eqs (8) are analogous to the collision term of the Boltzmann transport equation [21] and have a well understood physical interpretation.

Furthermore, if the jump times are symmetric in space, i.e. $\tau_{\beta\alpha}(\vec{l}'-\vec{l}) = \tau_{\alpha\beta}(\vec{l}-\vec{l}')$. Eqs (8) reduce to the system of linear equations of the form

$$\frac{\partial}{\partial t}\mathscr{P}_a^{\alpha}(\vec{l},t) = \sum_{\beta,\vec{l}'} \frac{1}{\tau_{\beta\alpha}(\vec{l}'-\vec{l})}[\mathscr{P}_a^{\beta}(\vec{l}',t) - \mathscr{P}_a^{\alpha}(\vec{l},t)], \quad \alpha = 1,\ldots,M; \quad a = 1,\ldots,N. \tag{8b}$$

The last system of linear equations was presented in our earlier raport [22].

4.2. Formal solution of the jump diffusion equations

Eqs (8) may formally be solved by the method of successive iterations. The "equations of motion" for any given a-th particle in the N-th step of the approximation scheme have the form

$$\frac{\partial}{\partial t}\overset{\mathscr{N}}{\mathscr{P}}{}_a^{\alpha}(\vec{l},t) = \sum_{\beta,\vec{l}'}\left\{\frac{\overset{\mathscr{N}}{\mathscr{P}}{}_a^{\beta}(\vec{l}',t)\left[1-\sum_{b\neq a}\overset{\mathscr{N}-1}{\mathscr{P}}{}_b^{\alpha}(\vec{l},t)\right]}{\tau_{\beta\alpha}(\vec{l}'-\vec{l})} - \frac{\overset{\mathscr{N}}{\mathscr{P}}{}_a^{\alpha}(\vec{l},t)\left[1-\sum_{b\neq a}\overset{\mathscr{N}-1}{\mathscr{P}}{}_b^{\beta}(\vec{l}',t)\right]}{\tau_{\alpha\beta}(\vec{l}-\vec{l}')}\right\},$$

$$\alpha = 1,\ldots,M; \quad a = 1,\ldots,N. \tag{9}$$

This is a system of coupled linear rate equations for functions with time-dependent coefficients. To solve these equations we rewrite them in the matrix form

$$\frac{\partial}{\partial t}\overset{\mathscr{N}}{\boldsymbol{\mathscr{P}}}{}_a(t) = \overset{\mathscr{N}-1}{\boldsymbol{T}}{}^a(t)\overset{\mathscr{N}}{\boldsymbol{\mathscr{P}}}{}_a(t), \tag{10}$$

where $\overset{\mathscr{N}}{\boldsymbol{\mathscr{P}}}{}_a(t)$ is the multi-vector whose components are $\overset{\mathscr{N}}{\mathscr{P}}{}_a^{\alpha}(\vec{l},t)$ and $\overset{\mathscr{N}-1}{\boldsymbol{T}}{}^a(t)$ is the "transition matrix" with elements

$$\overset{\mathscr{N}-1}{T}{}^a_{\alpha\beta}(\vec{l},\vec{l}';t) = \begin{cases} \dfrac{1-\sum_{b\neq a}\overset{\mathscr{N}-1}{\mathscr{P}}{}_b(\vec{l},t)}{\tau_{\beta\alpha}(\vec{l}'-\vec{l})}; & (\beta,\vec{l}') \neq (\alpha,\vec{l}), \\ \sum\limits_{\substack{(\alpha',\vec{l}'')\neq \\ (\alpha,\vec{l})}} \dfrac{1-\sum_{b\neq a}\overset{\mathscr{N}-1}{\mathscr{P}}{}_b^{\alpha'}(\vec{l}'',t)}{\tau_{\alpha\alpha'}(\vec{l}-\vec{l}')}; & (\beta,\vec{l}') = (\alpha,\vec{l}). \end{cases} \tag{10a}$$

Since, in practice, the particle jumps only between near-neighbour sites, it is readily shown that in each row and column of the matrix $\overset{\mathscr{N}-1}{\boldsymbol{T}}{}^a(t)$ only the numerous elements are essentially nonvanishing.

The solution of Eq. (10) may formally be written in terms of the evolution operator.

$$\overset{\mathscr{N}-1}{\boldsymbol{U}}{}^a(t|0) = \mathscr{T}\exp\left[\int_0^{|t|}\overset{\mathscr{N}-1}{\boldsymbol{T}}{}^a(t')dt'\right], \tag{10b}$$

where $\mathscr{T}$ is used to indicate time ordering. Operator $\overset{\mathscr{N}-1}{U^a}(t, |0)$ depends only on $|t|$ because the "backward evolution" in time from a given state is governed by the same equation as the "forward evolution". By definition

$$\overset{\mathscr{N}-1}{U^a}(t|0)\overset{\mathscr{N}}{\mathscr{P}_a}(0) = \overset{\mathscr{N}}{\mathscr{P}_a}(t). \tag{10c}$$

According to Eqs (10b) and (10c) we determine the final form of $\overset{\mathscr{N}}{\mathscr{P}^{\alpha}_a}(\vec{l}, t)$ functions

$$\overset{\mathscr{N}}{\mathscr{P}_a}(t) = \mathscr{T} \exp\left[\int_0^{|t|} \overset{\mathscr{N}-1}{T^a}(t')dt'\right]\mathscr{P}_a(0). \tag{11}$$

In Eq. (11) it has been tacitly assumed that we can stop the iteration procedure, with good approximation, on the $\mathscr{N}$-th step and therefore we put $\overset{\mathscr{N}}{\mathscr{P}_a}(0) \approx \mathscr{P}_a(0)$.

4.3. Self-correlation function and neutron scattering cross-section

The diffussion self-correlation function and its Fourier transform, incoherent, two-fold differential cross-section will now be introduced in the model.

Suppose we may choose an a-th particle which fulfills the initial conditions

$$\mathscr{P}^{\alpha}_a(\vec{l}, 0) = \delta_{\alpha\alpha_0}\delta_{\vec{l}\vec{l}_0}, \qquad \alpha = 1, \ldots, M, \tag{12}$$

i.e. that the diffusing a-th particle is initially at site α_0 of the cell at the origin. The solution $\mathscr{P}^{\alpha}_a(\vec{l}, t)$ corresponding to these particular initial conditions henceforth will be called $\mathscr{P}^{\alpha,\alpha_0}_a(\vec{l}, t)$. Thus, the diffusion, self-correlation function can be defined as

$$G_s^{D,\alpha_0}(\vec{r}-\vec{\mathscr{R}}_{\alpha_0}, t) = \frac{1}{N}\sum_a \sum_{\alpha,\vec{l}} \mathscr{P}^{\alpha,\alpha_0}_a(\vec{l}, t)\delta(\vec{r}-\vec{l}-\vec{\mathscr{R}}_{\alpha}), \tag{13}$$

where $\vec{\mathscr{R}}_{\alpha}$ is the position vector of the α site within the unit cell. It is remarkable that $G_s^{D,\alpha_0}(\vec{r}-\vec{\mathscr{R}}_{\alpha_0}, t)$ is the conditional probability density function that the atom which is in position $\vec{\mathscr{R}}_{\alpha_0}$ at time $t = 0$ will be at a later time t in position $\vec{r}$.

Using the definition (1) and the first result of Eq. (13), we find the double differential cross-section for incoherent thermal neutron scattering

$$\left(\frac{d^2\sigma_{\text{inc}}}{d\Omega d\varepsilon}\right)_{\alpha_0} = \frac{a^2_{\text{inc}}k'}{2\pi\hbar k_0} e^{-2W}\frac{1}{N}\sum_a \sum_{\alpha,\vec{l}} \int_{-\infty}^{\infty} dt \mathscr{P}^{\alpha,\alpha_0}_a(\vec{l}, t)$$

$$\times \exp\{i[\vec{\kappa}(\vec{l}+\vec{\mathscr{R}}_{\alpha}-\vec{\mathscr{R}}_{\alpha_0})-\omega t]\}\sum_n \sum_{m=0}^{n} \sum_{\lambda_1,\ldots,\lambda_n} A^{n,m}_{\lambda_1,\ldots,\lambda_n} \exp\left[it\left(\sum_{s=1}^{n-m}\omega_{\lambda_s} - \sum_{s=n-m+1}^{n}\omega_{\lambda_s}\right)\right], \tag{14}$$

where $A^{n,m}_{\lambda_1,\ldots,\lambda_n}$ are the coefficients the form of which can be derived by simple comparison of Eqs (14) and (5). It should be noted that the value $(d^2\sigma_{\text{inc}}/d\Omega d\varepsilon)_{\alpha_0}$ is not directly measured in the experiment.

We now construct the double differential cross-section for incoherent thermal neutron scattering directly measured in the experiment

$$\frac{d^2\sigma_{\text{inc}}}{d\Omega d\varepsilon} = \sum_{\alpha_0=1}^{M} \pi_{\alpha_0} \left(\frac{d^2\sigma_{\text{inc}}}{d\Omega d\varepsilon}\right)_{\alpha_0} = \frac{a_{\text{inc}}^2 k'}{2\pi\hbar k_0} e^{-2W} \frac{1}{N} \sum_a \sum_{\alpha,\vec{l}} \sum_{\alpha_0} \pi_{\alpha_0}$$

$$\times \int_{-\infty}^{\infty} dt \mathscr{P}_a^{\alpha,\alpha_0}(\vec{l}, t) \exp\{i[\vec{\kappa}(\vec{l} + \vec{\mathscr{R}}_\alpha - \vec{\mathscr{R}}_{\alpha_0}) - \omega t]\}$$

$$\times \sum_n \sum_{m=0}^{n} \sum_{\lambda_1,\ldots,\lambda_n} A_{\lambda_1,\ldots,\lambda_n}^{n,m} \exp\left[it\left(\sum_{s=1}^{n-m} \omega_{\lambda_s} - \sum_{s=n-m+1}^{n} \omega_{\lambda_s}\right)\right], \tag{15}$$

where occupation probabilities π_{α_0} fulfill the normalization condition of the form $\sum_{\alpha_0=1}^{M} \pi_{\alpha_0} = 1$. $d^2\sigma_{\text{inc}}/d\Omega d\varepsilon$ was obtained by averaging $(d^2\sigma_{\text{inc}}/d\Omega d\varepsilon)_{\alpha_a}$ over all possible initial conditions.

Finally, in the $\mathscr{N}$-th step of the approximation scheme the term $d^2\sigma_{\text{inc}}/d\Omega d\varepsilon$ has the form

$$\left(\frac{d^2\sigma_{\text{inc}}}{d\Omega d\varepsilon}\right)_{\mathscr{N}} = \frac{a_{\text{inc}}^2 k'}{2\pi\hbar k_0} e^{-2W} \frac{1}{N} \sum_a \varepsilon^{\text{T}}(\vec{\kappa}) \int_{-\infty}^{\infty} dt \mathscr{T} \exp$$

$$\times \left[\int_0^{|t|} \boldsymbol{T}^{\mathscr{N}-1}(t')dt'\right] \sum_{\alpha_0=1}^{M} \pi_{\alpha_0} \exp(-i\vec{\kappa}\vec{\mathscr{R}}_{\alpha_0}) \mathscr{P}_a^{\alpha_0}(0) \exp(-i\omega t) \sum_n \sum_{m=0}^{n}$$

$$\sum_{\lambda_1,\ldots,\lambda_n} A_{\lambda_1,\ldots,\lambda_n}^{n,m} \exp\left[it\left(\sum_{s=1}^{n-m} \omega_{\lambda_s} - \sum_{s=n-m+1}^{n} \omega_{\lambda_s}\right)\right]. \tag{16}$$

Let us note that the time evolution operator is simply related to the so-called intermediate scattering function [3], $I_{\text{s}}^{\mathscr{N}}(\vec{\kappa}, t)$, defined in terms of the MSJD model as

$$I_{\text{s}}^{\mathscr{N}}(\vec{\kappa}, t) = \varepsilon^{\text{T}}(\vec{\kappa}) \mathscr{T} \exp\left[\int_0^{|t|} \boldsymbol{T}^{\mathscr{N}-1}(t')dt'\right] \sum_{\alpha_0=1}^{M} \pi_{\alpha_0} e^{-i\vec{\kappa}\vec{\mathscr{R}}_{\alpha_0}} \mathscr{P}_a^{\alpha_0}(0). \tag{17}$$

The intermediate scattering function is directly connected with the velocity auto-correlation function [3].

5. *Final remarks*

In the present paper we derived:

(1) The general form of the multi-phonon formula, for the incoherent scattering, as a superposition of respectively shifted diffussion scattering laws $S_{\text{inc}}^{\text{D}}$.

(2) The nonlinear jump diffusion equations for any concentrations of impurity atoms, and briefly studied general consequences of the application of a formal solution to the theory of an incoherent thermal neutron scattering.

In this paper we assume for simplicity that the diffusive and vibrational motions of the impurity particle are completely uncorrelated. In a subsequent paper [23] we will discuss the situation where the diffusive and oscillatory degrees of freedom are correlated.

We want to emphasize that the nonlinear "equations of motion" provided the extended viewpoint on the continuous-time random lattice walk problem.

It may be proved [23] that for small and great concentrations of impurity particles the iteration procedure is fast convergent. Then, under the circumstances, the nonlinear "equations of motion" (8) transform into the system of linear equations, briefly discussed in our earlier paper [16].

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