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The Application Of Multiple Scattering Theory (Mst) In Calculating The Deuterium Flux Permeating The Pd Thin Film

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The multiple-scattering theory[2] is applied to the de Broglie wave of deuterons inside the palladium film. The formalism for band structure calculation and the reflection and transmission calculations for finite slices is presented. The latter is based on a double-layer scheme which obtains the reflection and transmission matrix elements for the multiplayer slice from those of a single layer. With a relative simple model for the potential of palladium crystal lattice, we calculate the band structures of probability wave of deuterons propagating in the palladium, as well as the transmission coefficients through finite periodic slices. Selective resonant tunneling theory is adopted when obtaining the scattering matrix T. Our calculations consist with experimental results which can not be explained by diffusion theory.

1 Introduction

The behavioral features of deuteron particles in gas-loading D/Pd system has always been a very important subject in the research of condensed matter nuclear science.14 year pursuing in gas-loading experiments at Tsinghua University results in the discovery of the anomalous feature of the deuterium flux permeating the Pd thin film[1]. Instead of the monotonic feature of the deuterium flux, the peaky deuterium flux appears at certain temperature, which is higher than the boiling point of the heavy water. This is unexpected if the diffusion model is applied to this permeation process. Based on the conventional diffusion theory, the diffusion coefficient increases dramatically when the temperature of the palladium increases. However, we observed the peaky feature repeatedly at certain temperature. The resonant feature shown in the experiments implies that the deuteron particles should be described by the probability wave.

Therefore we introduced a general de Broglie wave function to characterize the interaction between the deuterium flux and the gas-loading D/Pd system. The phase factor of deuteron wave is the key for interference and resonance. The idea of the importance of the phase factor is also the core of selective resonant tunneling theory.

Multiple-scattering theory was applied to implement the above concept in calculating the deuterium flux permeating the gas-loading Pd thin film. Multiple-scattering theory (MST) usually known as KKR (Korringa, Kohn, and Rostoker) approach, was developed mainly for the calculation of electronic band structures, although it originated from the study of classical waves. It was widely used in the research of de Broglie wave, elastic wave, electromagnetic wave and so on. The main idea of MST is to separate the complicated potential distributed in the three-dimensional space into non-overlapped regions (this is quite clear for periodic structure but not so easy for disordered systems). Each region is taken as a single scatterer and the incident waves on this scatterer are composed of the scattered waves of other scatterers and the incident waves far away while its scattered waves become part of incident waves on other scatterers. Such transformation between incident waves and scattered waves of different scatterers are achieved through the vector structure constant G. That's the keystone of MST.

MST was developed into different kinds of equations when dealing with different problems. The concrete equations adopted in this paper were first brought forward by Modinos[3] in his work about the scattering problem of electromagnetic waves. Then Professor Zhengyou Liu[4] extended them for elastic waves and got excellent agreement with experimental data. The calculation was done in the following steps: (1) a three-dimensional crystal was divided into many periodically arranged two-dimensional layers and the scattering matrix of each layer was got; (2) the total scattering matrix of the crystal was formed by combining these one-layer matrix elements; (3) transmittance and reflectance can be calculated based on the scattering matrix. Since two-dimensional layers play an important role here, this method was also called layer MST theory.

2 Mathematical Equations of Layer MST Theory

For one layer, all the divided scatters are located on sites { $\vec{R}_n$ } of a two-dimensional lattice in the x-y plane, i.e.,

$$\vec{R}_n = n_1\vec{a}_1 + n_2\vec{a}_2 \tag{1}$$

where $\vec{a}_1, \vec{a}_2$ are primitive vectors in the x-y plane, and n_1, n_2 are integers. For reference, the z axis is assumed to point to the right of the x-y plane. A plane de Broglie wave incident on the scatters may be expressed in general as

$$U^{in\pm}(\vec{r}) = \sum_g u_g^{in\pm} \exp(i\vec{k}_g^{\pm} \cdot \vec{r}) \tag{2}$$

$$\vec{k}_g^{\pm} = (\vec{k}_{\parallel} + \vec{g}, \pm\sqrt{k^2 - \left|\vec{k}_{\parallel} + \vec{g}\right|^2}) \tag{3}$$

Here the sign + means incident from the left of the plane (positive z), and – means incident from the right of the plane (negative z), $\vec{g}$ is one of the two-dimensional (2D) reciprocal lattice vectors in the plane of the scatters, and $\vec{k}_{\parallel}$ is the reduced wave vector in the 2D Brillouin zone of the reciprocal lattice (also the Bloch wave vector).

We should note that this expression of the incident wave means that it's no longer far away from the layer but already has been interacting with the 2D lattice. So its possible wave vectors of the same stationary state are not continuous but discrete in space due to the periodical boundary conditions of the 2D lattice. Because equation (2) gives a complete plane wave expression of wave function of the two-dimensional plane of scatters, the scattered wave can also be given out in the similar form:

$$U^{sc\pm}(\vec{r}) = \sum_g u_g^{sc\pm} \exp(i\vec{k}_g^{\pm} \cdot \vec{r}) \tag{4}$$

Then wave function on both sides of the plane, i.e. the solution of the Schrodinger equation can be shown as Fig.1

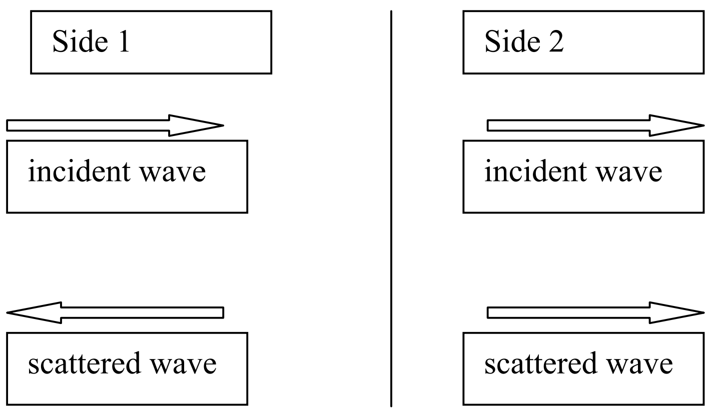


Figure 1. Wave function divided as the incident and scattered wave in both sides of one layer of scatters under the case that incident wave on the plane is along the positive z from side one.

As we can see in Fig 1, both the wave function on side 1 and side 2 are the sum of two parts: the incident wave and the scattered wave. This is similar to the one-dimensional case, while the only difference is that there are a group of differently directional wave vectors instead of one in the 1D case. So the job to solve the Schrodinger equation with complicated potential to get the wave function equals to calculate all the $u_g^{sc\pm}$ in equation (4), which means getting the plane wave form of scattered wave. We should note that g in equation (3) has a limit that $\sqrt{k^2 - \left|\vec{k}_{\|} + \vec{g}\right|^2}$ should be real.

Based on above discussions, we can express the problem in matrix-form (now assuming that incident wave from both sides):

$$\vec{U}_{sc}^{+} = \vec{M}^{++} \bullet \vec{U}_{in}^{+} + \vec{M}^{+-} \bullet \vec{U}_{in}^{-} \qquad (5)$$
$$\vec{U}_{sc}^{-} = \vec{M}^{-+} \bullet \vec{U}_{in}^{+} + \vec{M}^{--} \bullet \vec{U}_{in}^{-}$$

where $\vec{U}_{in}^{\pm} = \left\{ u_g^{in\pm} \right\}_g, \vec{U}_{sc}^{\pm} = \left\{ u_g^{sc\pm} \right\}_g, \vec{M}^{ss'} = \left\{ \vec{M}_{gg'}^{ss'} \right\}_g$ and tags s, s' denote the sign + or -. Thus once we get the scattering matrice $\vec{M}^{ss'}$, the problem of one-layer scatter is solved. To achieve this, transformations between plane waves and spherical waves are necessary because we all know the scattering calculation of single scatter is in the form of spherical waves.

First of all, we should choose a central scatter (on or near the central axis of the plane of layer) as the origin of the coordinate. Then incident wave in equation (2) can be rewritten in the following form of spherical waves:

$$U_{in}(\vec{r}) = \sum_{lm} a_{lm} J_{lm}(\vec{r}) \qquad (6)$$

where

$$a_{lm} = \sum_{sg} u_g^{ins} \cdot A_{lm}^{gs} \qquad (7)$$

$$A_{lm}^{g\pm} = 4\pi i^l (-1)^m Y_{l-m}(\hat{\mathrm{k}}_{\mathrm{g}}^{\pm}) \qquad (8)$$

$$J_{lm}(\vec{r}) = j_l(kr) Y_{lm}(\hat{\mathrm{r}}) \qquad (9)$$

then the totally scattered waves by the layer is the sum of the scattered wave of all scatters:

$$U^{sc}(\vec{r}) = \sum_{ilm} b_{lm}^{i} H_{lm}^{i}(\vec{r}_i) \qquad (10)$$

sign i means the ith scatterer of the layer, $\vec{r}_i$ means the coordinate with the ith scatter's center as the origin.

According to the Bloch theorem,

$$b_{lm}^{i} = b_{lm} \exp(i\vec{k}_{\|} \cdot \vec{R}_i) \qquad (11)$$

so equation (10) can be transformed to:

$$U^{sc}(\vec{r}) = \sum_{lm} b_{lm} \sum_{\vec{R}} \exp(i\vec{k}_{\|} \cdot \vec{R}) H_{lm}(\vec{r} - \vec{R}) \qquad (12)$$

With $\boldsymbol{A} = \left\{ a_{lm} \right\}$, $\boldsymbol{B} = \left\{ b_{lm} \right\}$,and the definition of scattering matrix of single scatter as $\boldsymbol{T} = \left\{ t_{lml'm'} \right\}$, the vector structure factor of MST as :

$$G_{lml'm'}(\vec{R}) = 4\pi \sum i^{l'+l''-l} C_{l'm'l''m-m'}^{lm} h_{l''}(kR) Y_{l''m-m'}(\hat{R}) \qquad (13)$$

where

$$C_{l'm'l''m''}^{lm} = \iint_s Y_{lm}(\Omega) Y_{l'm'}^{*}(\Omega) Y_{l''m''}^{*}(\Omega) d\Omega \qquad (14)$$

thus we have the following form of $U^{sc}(\vec{r})$:

$$U^{sc}(\vec{r}) = \sum_{lm} b_{lm} \sum_{\vec{R}} \exp(i\vec{k}_{\|} \cdot \vec{R}) \sum_{l'm'} G_{lml'm'}(-\vec{R}) J_{lm}(\vec{r})$$
$$= \sum a'_{lm} J_{lm}(\vec{r}) \qquad (15)$$

Here,

$$a'_{lm} = \sum b_{l'm'} G_{l'm'lm}(\vec{k}_{\|}) \qquad (16)$$

and

$$G_{lml'm'}(\vec{k}_{\|}) = \sum_{\vec{R}} \exp(i\vec{k}_{\|} \cdot \vec{R}) G_{lml'm'}(-\vec{R}) \quad (17)$$

As we have mentioned above, the waves incident on each scatter of the layer are the sum of two parts: the incident waves as equation (6) expresses and the waves scattered by other scatters. So for the central scatter, we have the incident waves as:

$$\sum_{lm} (a_{lm} + a'_{lm}) J_{lm}(\vec{r}) \quad (18)$$

then the coefficients of scattered wave by the central scatter are:

$$b_{lm} = \sum_{l'm'} t_{lml'm'} (a_{l'm'} + a'_{l'm'}) \quad (19)$$

written in the matrix form:

$$\begin{aligned} B &= T(A + A') \\ &= T[A + G^{Tr}(k_{\|}) B] \end{aligned} \quad (20)$$

With an inverse transformation we have:

$$B = ZA \quad (21)$$

and

$$Z = [I - TG^{Tr}(k_{\|})]^{-1} T \quad (22)$$

where ***I*** is unit matrix.

Now substituting equation 21 and 22 into equation 12, and we can get the spherical-wave form of scattered wave by the two-dimensional layer. Nevertheless we still need to find the plane wave form. Use has been made of the following equation:

$$\begin{aligned} &\sum_{\vec{R}} \exp(i\vec{k}_{\|} \cdot \vec{R}) H_{lm}(\vec{r} - \vec{R}) \\ &= \sum_{\vec{R}} \exp(i\vec{k}_{\|} \cdot \vec{R}) h_{lm}(k|\vec{r} - \vec{R}|) Y_{lm}(\hat{r} - \hat{R}) \\ &= \frac{2\pi}{Sk} \sum_{g} \frac{Y_{lm}(\hat{k})}{(k^2 - |\vec{k}_{\|} + \vec{g}|^2)^{1/2}} \exp(i\vec{k} \cdot \vec{r}) \end{aligned} \quad (23)$$

where $S = A_0 \sqrt{l(l+1)} i^l$, and A_0 is the area of the 2D unit cell.

Giving the definition of the following parameter:

$$B_{lm}^{g\pm} = \frac{2\pi}{Sk} \frac{Y_{lm}(\hat{k}_g^{\pm})}{(k^2 - |\vec{k}_{\|} + \vec{g}|^2)^{1/2}} \quad (24)$$

we have the coefficients in equation (4)

$$u_g^{sc\pm} = \sum_g b_{lm} B_{lm}^{g\pm} \quad (25)$$

Substituting equation (8) and (21) into (25),

$$u_g^{scs} = \sum_{g'} M_{gg'}^{ss'} \cdot u_{g'}^{scs'} \quad (26)$$

where

$$M_{gg'}^{ss'} = \sum_{lml'm'} B_{lm}^{gs} Z_{lml'm'} A_{l'm'}^{g's'} \quad (27)$$

Thus we obtain the plane wave form of the scattered waves, and as discussed above, the total wave function on both sides of the layer are the sum of incident waves and scattered waves:

$$\vec{U}^{+}(1) = \vec{U}_{in}^{+}, \vec{U}^{-}(1) = \vec{U}_{sc}^{-} + \vec{U}_{in}^{-} \quad (28)$$

$$\vec{U}^{+}(2) = \vec{U}_{in}^{+} + \vec{U}_{sc}^{+}, \vec{U}^{-}(2) = \vec{U}_{in}^{-} \quad (29)$$

substituting equation (5) into (28) and (29),

$$\vec{U}^{+}(2) = (I + \vec{M}^{++}) \bullet \vec{U}^{+}(1) + \vec{M}^{+-} \bullet \vec{U}^{-}(2)$$
$$\vec{U}^{-}(1) = \vec{M}^{-+} \bullet \vec{U}^{+}(1) + (I + \vec{M}^{--}) \bullet \vec{U}^{-}(2) \tag{30}$$

One should note that all the plane-wave expansions, including the incident and scattered waves, are referred to the central scatterer in the plane. If we shift the center of expansion by $-\vec{a}_3/2$ for waves on side 1 and $\vec{a}_3/2$ for waves on side 2, where $\vec{a}_3$ is the translation vector of the two-dimensional plane in forming a three dimensional crystal, then

$$\vec{U}^{+}(2) = \vec{Q}^{++} \bullet \vec{U}^{+}(1) + \vec{Q}^{+-} \bullet \vec{U}^{-}(2)$$
$$\vec{U}^{-}(1) = \vec{Q}^{-+} \bullet \vec{U}^{+}(1) + \vec{Q}^{--} \bullet \vec{U}^{-}(2) \tag{31}$$

where

$$\vec{Q}^{ss'} = \phi^{s}\phi^{s'}\delta_{ss'} + \phi^{s}\vec{M}^{ss'}\phi^{s'} \tag{32}$$

and the definition of ϕ^s is:

$$\phi^s = \begin{bmatrix} \exp(si\vec{k}^s_{g1} \cdot \vec{a}_3/2) & & \\ & \ddots & \\ & & \exp(si\vec{k}^s_{gN} \cdot \vec{a}_3/2) \end{bmatrix} \tag{33}$$

Given the Q matrix of single layer, we can combine the matrix elements of two successive single layers to obtain those of the double layer, then combine those of one single-layer and one double-layer to obtain those of the triple layers; then, of whatever number of layers. The notation used is as follows. The matrices $\vec{Q}^{++}, \vec{Q}^{+-}, \vec{Q}^{-+}, \vec{Q}^{--}$ for the first layer (not mean to single layer, but may be numbers of layers) are denoted as $\vec{Q}_{11}(\mathrm{I}), \vec{Q}_{12}(\mathrm{I}), \vec{Q}_{21}(\mathrm{I}), \vec{Q}_{22}(\mathrm{I})$, respectively, the corresponding matrices for the second layer are denoted by $\vec{Q}_{11}(\Pi), \vec{Q}_{12}(\Pi), \vec{Q}_{21}(\Pi), \vec{Q}_{22}(\Pi)$. All matrices refer to the same k and $\vec{k}_{\parallel}$. It is easily shown that

$$\boldsymbol{Q}_{11}(\mathrm{I}\Pi) = \boldsymbol{Q}_{11}(\Pi)[\boldsymbol{I} - \boldsymbol{Q}_{21}(\mathrm{I})\boldsymbol{Q}_{12}(\Pi)]^{-1}\boldsymbol{Q}_{11}(\mathrm{I})$$
$$\boldsymbol{Q}_{12}(\mathrm{I}\Pi)$$
$$= \boldsymbol{Q}_{11}(\Pi)\boldsymbol{Q}_{12}(\mathrm{I})[\boldsymbol{I} - \boldsymbol{Q}_{21}(\Pi)\boldsymbol{Q}_{12}(\mathrm{I})]^{-1}\boldsymbol{Q}_{22}(\Pi) + \boldsymbol{Q}_{12}(\Pi)$$
$$\boldsymbol{Q}_{21}(\mathbf{III}) = \boldsymbol{Q}_{22}(\mathrm{I})\boldsymbol{Q}_{21}(\Pi)[\boldsymbol{I} - \boldsymbol{Q}_{12}(\mathrm{I})\boldsymbol{Q}_{21}(\Pi)]^{-1}\boldsymbol{Q}_{11}(\mathrm{I}) + \boldsymbol{Q}_{21}(\mathrm{I})$$
$$\boldsymbol{Q}_{22}(\mathrm{I}\Pi) = \boldsymbol{Q}_{22}(\mathrm{I})[\boldsymbol{I} - \boldsymbol{Q}_{12}(\Pi)\boldsymbol{Q}_{21}(\mathrm{I})]^{-1}\boldsymbol{Q}_{22}(\Pi) \tag{34}$$

where the argument $\mathrm{I}\Pi$ refers to the newly formed crystal of which layers equal to the sum of the above two. So we can calculate the rate of transmission and the rate of reflection for crystal of any number of layers using the following equations:

$$T = \frac{\sum_g u_g^{trn} \cdot u_g^{trn*} k_{gz}}{\sum_g u_g^{inc} \cdot u_g^{inc*} k_{gz}}$$

$$R = \frac{\sum_g u_g^{ref} \cdot u_g^{ref*} k_{gz}}{\sum_g u_g^{inc} \cdot u_g^{inc*} k_{gz}} \tag{35}$$

where $k_{gz} = \sqrt{k^2 - \left|\vec{k}_{\parallel} + \vec{g}\right|^2}$.

When all the scatters are elastic; the conservation of the particles requires:

$$T+R=1 \tag{36}$$

If absorption is introduced, i.e. nuclear reactions happen; then, $T+R<1$ and $\xi=1-T-R$, ξ is defined as the rate of absorption.

3 Model for Calculation and the Preliminary Results

In the preliminary calculation of the deuterium flux permeating the thin Pd film, the following physical model was assumed:

The scatterers' crystal is the Pd-deuteride thin film with loading ratio 1:1; and all the deuterons inside the Pd crystal are located at the octahedral positions, such that the deuterons themselves form an fcc crystal with the same lattice constant as the Pd (3.89A). The scattering effect of Pd nucleus was not considered for this calculation. The interaction between injected deuteron and the electrons in the crystal is described by a square potential well V_2 , which is distributed in a spherical shell around each deuteron target. The inner and outer radius of this shell are a_1 and a_2, respectively. The depth of the square potential well V_2 is –10 eV (a_1=0.32A, a_2=0.665A). A square potential barrier, V_1 ,was assumed in the region $a_0< r <a_1$ to represent the Coulomb barrier between deuterons. The height of this barrier is 30 keV. When the distance between two deuterons is smaller than a_0 (a_0=4.4fm), the square nuclear potential well plays a role whose real part V_{0r} is -63.04MeV as shown by figure 2.

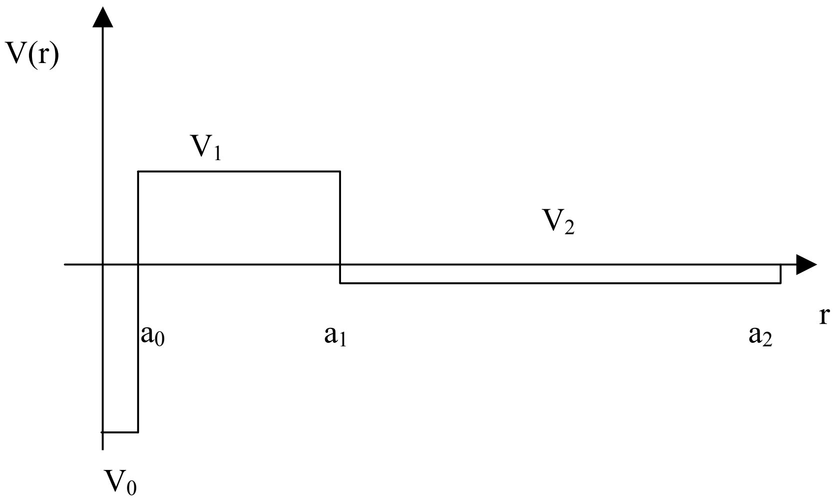


Figure 2. The potential model of single scatterer which models deuterons in Pd crystal.

For the incident deuteron wave, we assumed the eigenvalue of its energy is:

$$E=k_bT_D \tag{37}$$

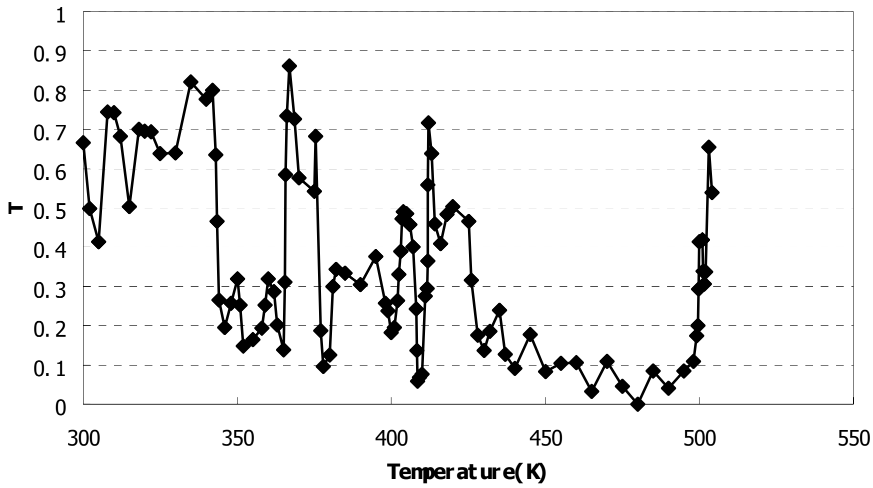
where T_D is the temperature of deuterium gas injected from one side of the Pd film, and k_b is the Boltzmann constant. As we have mentioned above, the incident wave vector should satisfy the periodical boundary conditions of the two-dimensional lattice(equation (2)).

Equation (37) shows that, the energy of incident wave changes with the temperature of the system, so does the incident wave vector and the vector structure factor G of gas-loading D/Pd system.

Using the abovementioned physical model, we obtained the following results with MST:

Based on abovementioned calculation using MST, we obtained some conclusions as follows:

The rate of transmission and the rate reflection of deuteron wave permeating thin Pd film are quite distinct from those from diffusion model.



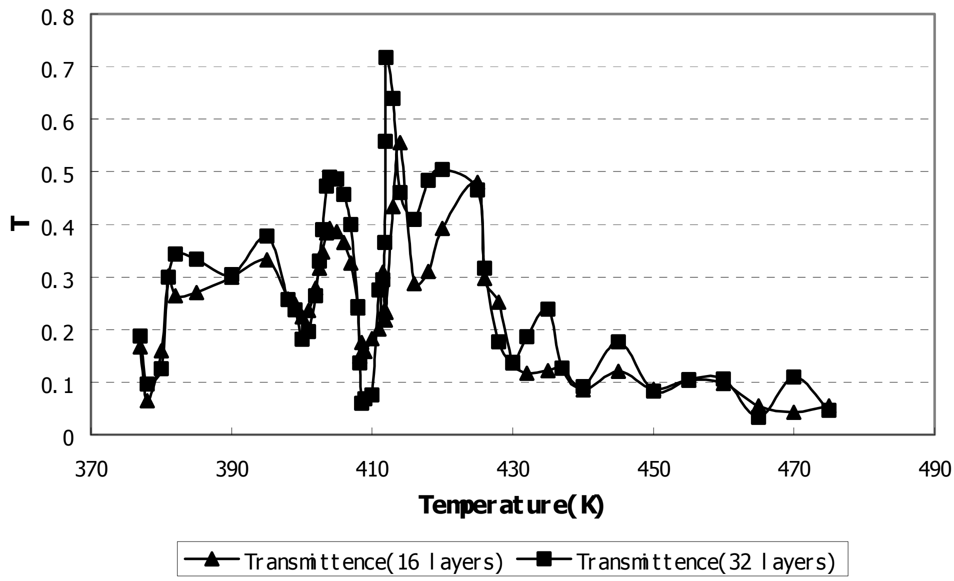


Figure 4. Rate of transmission of deuteron wave through 32-layer and 16-layer Pd crystal as a function of temperature (370K~490K, to compare with the following experimental data).

The peaky feature in the results of MST calculation are similar to those in the experimental observation.

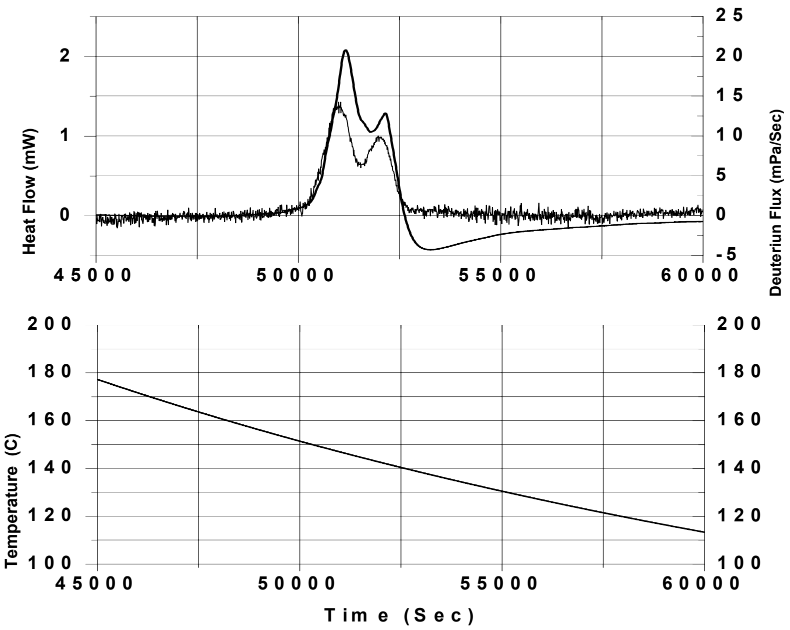


Figure 5. Experimental data of deuterium flux permeating the thin Pd film.

In fact, the most important distinction of our theory from diffusion theory is that the incident deuteron particles in Multiple Scattering Theory can still keep the phase factor of the de Broglie wave function. So the interference of the wave may cause the anomalous permeation of deuterium flux through thin Pd film. However, under some special conditions, our theory would possibly have conclusions close to that of diffusion theory: for example, if the Pd film is much too thick or the periodical feature of the crystal is so poor that almost near to an disordered system, it's hard for the de Broglie wave to interfere because the phase factor of the wave function is chaotic. In this sense, we can say that diffusion model is a special case of our theory.

M. Fleischmann and J. Giudice pointed out that the deuterons inside the palladium crystal lattice act like a QED object which might be described by a single wave function[5,6]. The present work is one step along this line.

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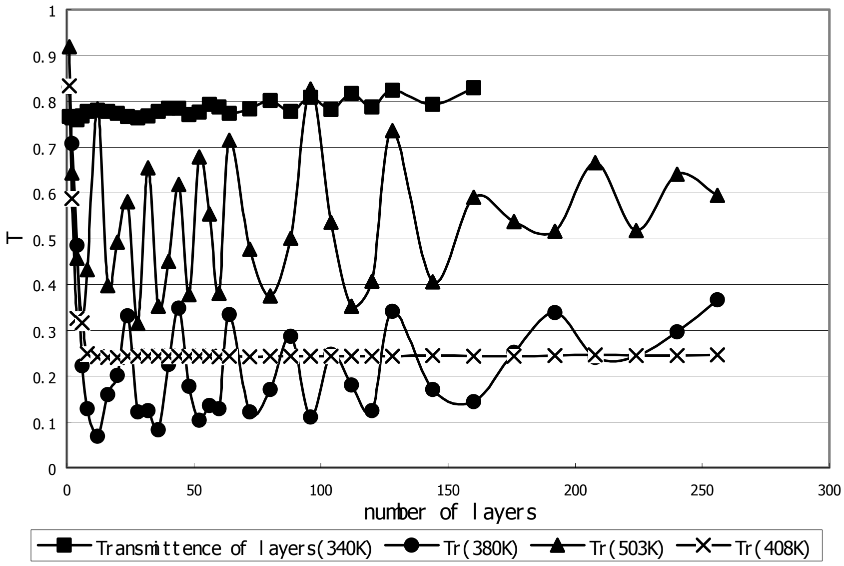

Figure 6. Rate of transmission of deuteron wave through different number of layers between 1 and 256 as the function of the number of Pd crystal layers at certain temperatures.

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