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Molecular dynamics simulation of atom–atom collisional scattering

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Abstract. Molecular dynamics techniques have been used to simulate atom–atom collisional scattering of 0–5.5 eV/nucleon Ti atoms in bulk Ti. The recoiling atoms to be simulated were produced by the emission of MeV γ-rays after the thermal neutron capture reaction ^{48}Ti(n,γ)^{49}Ti. The information on the collisional scattering was deduced from Doppler broadened γ-ray lineshapes, produced in the decay of excited bound states in recoiling ^{49}Ti. The measured γ-ray lineshapes were simulated by combining the molecular dynamics calculation for the atomic collisions with calculations using the Monte Carlo method for the experimental conditions and decays of excited nuclear states. The dependence of the simulated γ-ray lineshapes on the interatomic potential is demonstrated.

1. Introduction

The interatomic potential between atoms has been studied during the past decades in several experimental and theoretical works [1]. The disagreement between potentials predicted by different models is apparent at radial separations greater than 1 Å. Recently, Ziegler *et al* [1] have constructed an analytical function to be used in the calculation of atom–atom collisional scatterings. The universal screening function derived on the basis of all experimental information is believed to yield reliable atom–atom potentials for calculation of energy losses in atomic collisions. However, at collision energies corresponding to radial separations of atoms greater than 1 Å the screening function could be tested only in very few cases [1]. More data are desirable to test different interatomic potentials. A new method, gamma-ray-induced Doppler broadening (GRID) developed very recently at the Institut Laue–Langevin for measurements of short nuclear lifetimes [2], offers a new experimental method to learn more about interatomic potentials inside bulk matter.

In the present work, the GRID data obtained for ultra-low velocity ($v/c = \beta \sim 10^{-4}$, where c is the velocity of light) Ti atoms slowing down in Ti substrate [3], have been simulated by molecular dynamics (MD) and Monte Carlo (MC) calculations. To our knowledge, this method has not previously been used to study slowing down of energetic ions. The aim of this paper is to describe the principles of the simulation and demonstrate the sensitivity of the simulated GRID data on different interatomic potentials.

In GRID measurements, de-excitation of a capture state by a γ-ray emission after thermal neutron capture imparts a recoil velocity to the nucleus. Secondary γ-rays

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emitted by the recoiling nucleus before it has slowed down in matter, are thus Doppler shifted. Since the primary γ-rays are emitted isotropically the observed γ-ray lineshape is Doppler broadened. The observed lineshape of the secondary γ-rays depends on the time elapsed between the formation of the excited nucleus and the γ-ray emission and also the slowing down process. For typical recoil velocities of the order of $\beta \sim 10^{-4}$, the energy loss mechanism is dominated by elastic scattering of recoiling atoms from the screened Coulomb potential of the substrate atoms.

Usually the slowing down of recoil atoms is analysed by the binary collision approximation (BCA). Collisions between the recoiling and target atoms are treated separately and only the asymptotic trajectory of the recoil is calculated. This approximation can, however, be applied only at relatively high energies, greater than 100 eV [1].

At ultra-low recoil velocities atomic collisions can not be approximated as isolated phenomena. The potentials of many target atoms may simultaneously effect the trajectory of a recoil. The most realistic method to simulate the atom–atom collisions is the MD calculations where the equations of motion of both the recoiling and target atoms are integrated numerically. In this method the crystalline structure of the slowing-down material and thermal motion of the atoms are included in the simulation in a realistic way.

The MC simulation of Doppler shifted γ-ray lineshapes developed in our laboratory [4] has been modified in the present work to correspond to the experimental conditions in the GRID measurements.

2. Experimental results

The GRID data simulated in this work have been obtained in the study of the reaction ^{48}Ti(n,γ)^{49}Ti [2, 3]. The capture state of the nucleus ^{49}Ti has an excitation energy of $E_x = 8.14$ MeV. The transitions studied were $3.26 \rightarrow 1.76$ MeV ($E_\gamma = 1499$ keV) and $3.18 \rightarrow 1.38$ MeV ($E_\gamma = 1793$ keV). The initial recoil velocities for the direct decays of the capture state to the states under study were 3.21×10^4 m s^{-1} and 3.26×10^4 m s^{-1}, respectively. The measured 1499 keV and 1793 keV lineshapes are shown in figures 1 and 2. The decay schemes of the capture state are illustrated in the inserts of figures 1 and 2. For more details of the experiments, see [2] and [3].

3. Principles of the simulations

For the reproduction of the measured γ-ray lineshapes the collisional scattering of Ti atoms was simulated using constant energy and volume MD method. The MD cell comprised 216 atoms. Periodic boundary conditions were applied in all three dimensions. Atoms were placed in an HCP array with initial velocities to yield the desired temperature of 1000 K. The velocities corresponded to non-correlated atomic vibrations. The equations of motion of the atoms were solved numerically using the algorithm by Beeman [5] with a time step of 0.5 fs.

At the beginning of a simulation event, an isotropic recoil velocity was imparted to one of the lattice atoms. The simulation was continued until the excited state in the recoiling ^{49}Ti nucleus emitted the γ-ray corresponding to the lineshape of interest.

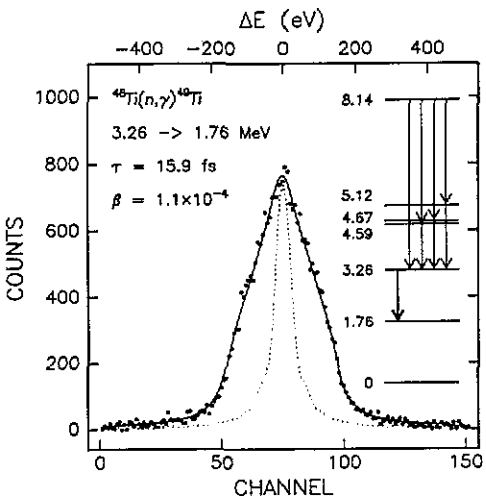

Figure 1. The experimental (full circles) and simulated (full curve) γ-ray lineshapes for the 1499 keV transition. The universal potential of Ziegler *et al* [1] (ZBL) used in the simulation resulted in the lifetime value of 15.9 fs. The dotted line is the instrumental response function of the spectrometer. Also shown is the decay scheme of the capture state to the 3.26 MeV state.

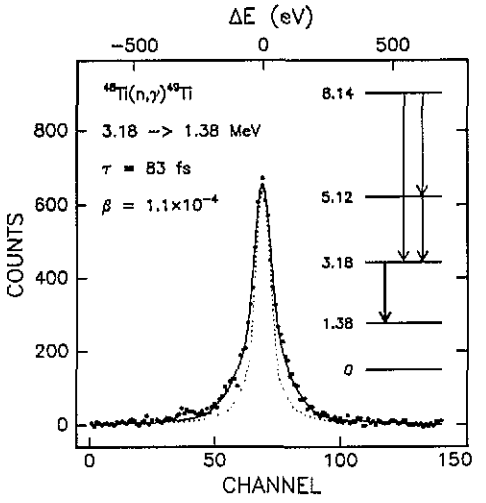

Figure 2. As figure 1 but for the 1793 keV transition. The lifetime value of 83 fs was obtained from the simulation with the ZBL potential [1].

The final velocity vector of the nucleus was stored for the use in the MC calculation of the γ-ray lineshape. About 4000 such events were calculated for each lineshape.

In addition to the direct decay, the capture state can decay to the state under study via intermediate states. In these cases the recoil velocity is imparted to the atom in several steps. The effect of these cascade γ-rays to the velocity of the recoiling atom was taken into account by the MC method. In the simulations of the experimental γ-ray lineshapes, different decay branches were weighted by the branching ratios taken from [6].

The dependence of the simulated γ-ray lineshape on the interatomic potential was studied by using three potentials. An universal potential (ZBL) widely used in stopping power calculations, has been derived by Ziegler *et al* [1] by fitting an universal screening function to the experimental stopping power data

$$V_{\mathrm{ZBL}}(r) = \frac{Z_1 Z_2 e^2}{r}\phi(r/a_U) \tag{1}$$

where the screening function is

$$\phi(x) = 0.1818\mathrm{e}^{-3.2x} + 0.5099\mathrm{e}^{-0.9423x} + 0.2802\mathrm{e}^{-0.4028x} + 0.02817\mathrm{e}^{-0.2016x} \tag{2}$$

the screening length

$$a_U = \frac{0.8854 a_0}{Z_1^{0.23} + Z_2^{0.23}} \tag{3}$$

and the Bohr radius $a_0 = 0.529$ Å. A pair potential of Born–Mayer form (BM) for Ti–Ti has been obtained by Abrahamson by fitting an exponential function to the results of Thomas–Fermi–Dirac calculation [7]:

$$V_{\mathrm{BM}}(r) = Ae^{-r/B} \tag{4}$$

where $A = 9352.6$ eV and $B = 0.2779$ Å. A new method to calculate interatomic interactions based on the effective-medium theory (EMT) has recently been developed by Jacobsen *et al* [8]. The lineshapes were also simulated using a pair potential (EMT) calculated for Ti–Ti by this method [9].

The lattice sites of the atoms corresponding to their equilibrium positions, were obtained by adding an attractive potential to the three repulsive potentials. The attractive part was described by the Morse potential:

$$V_{\mathrm{M}}(r) = D\{\exp[-2\alpha(r - r_0)] - 2\exp[-\alpha(r - r_0)]\}. \tag{5}$$

The parameters $D = 0.43$ eV and $\alpha = 1.5$ Å^{-1} were taken from [10], calculated for Cr. The position of potential minimum was the nearest-neighbour distance of titanium $r_0 = 2.90$ Å. The calculated lineshapes were found to be insensitive to the exact form of the attractive potential.

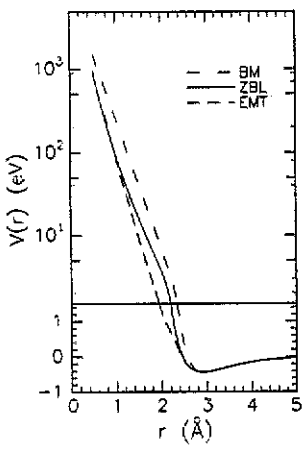

Figure 3. Pair potentials for Ti–Ti used in the simulations. Full curve, universal potential of Ziegler *et al* [1] (ZBL); broken curve, potential calculated with the effective medium theory [9] (EMT); chain-dot curve, Born–Mayer potential of Abrahamson [7] (BM). Note the change between the linear and logarithmic scales at +1.5 eV.

The total interatomic potentials used in the simulations, were obtained by adding the repulsive potential to the attractive potential $V_{\mathrm{M}}(r)$

$$V(r) = \big(1 - f(r)\big)V_{\mathrm{rep}}(r) + f(r)V_{\mathrm{M}}(r)\,. \tag{6}$$

The attenuation factor $f(r) = \{1+\exp[-b(r-r_1)]\}^{-1}$ was used to obtain a continuous function for the total potentials. The values of b and r_1 were chosen so that the splining did not affect the repulsive potentials above a threshold value $V(r) = V_0$ and that the attractive potential V_M was not changed at interatomic distances near equilibrium

separation. The value of V_0 was determined as the minimum value of the interatomic potential which was obtained to affect the simulated γ-ray lineshape. The value of V_0 determined by the instrumental resolution was obtained to be 4 eV. The total interatomic potentials are shown in figure 3. The lineshape calculations by using only the repulsive potentials (BM, ZBL and EMT) for scattering of the recoiling atom resulted in identical lineshapes with those obtained with the splining described above. The cut-off radius of 5.0 Å was used in the MD calculations.

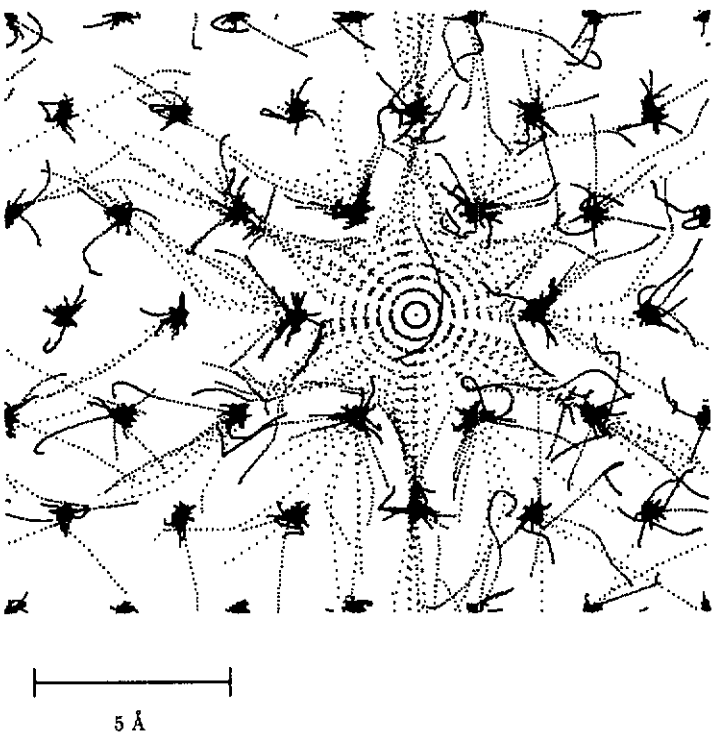

Figure 4. Superimposed trajectories of atoms in the (001) plane from 200 simulation events. In every event the atom in the middle of the picture was given a recoil velocity of 3.21×10^4 m s^{-1} with a random direction in the (001) plane. Time difference between dots of the trajectories is 1 fs. The mean lifetime of the excited state of the recoiling nucleus was 15.9 fs.

In the calculations periodic boundary conditions were applied in all three dimensions. This implies that the whole energy of the recoiling atom is confined to the MD cell during a simulation event. Due to relatively short simulation times the boundary conditions and the size of the MD cell should not effect the results. This was checked by calculating some lineshapes with a MD cell of 512 atoms. No disagreement was found between the lineshapes calculated with the MD cells of 216 and 512 atoms. In the beginning of the simulation the initial velocities of the atoms in the MD cell corresponded to non-correlated atomic vibrations. Due to the facts that the velocity imparted to the recoiling atom is orders of magnitude larger than thermal velocities of the other atoms and that the simulated lineshapes are insensitive to the exact form

of the attractive potential near the minimum and to the position of the minimum, the assumption of random initial velocities does not affect the simulated lineshapes. Whether the recoil velocity was imparted to the recoiling atom immediately in the beginning of the simulation or after the system had been simulated for 500 fs did not affect the calculated lineshapes. Figure 4 illustrates the MD simulations of the trajectories of recoiling and lattice atoms.

The electronic energy loss of the recoiling ^{49}Ti atoms [1] was included in the calculations. It was, however, found to have a negligible effect on the lineshapes.

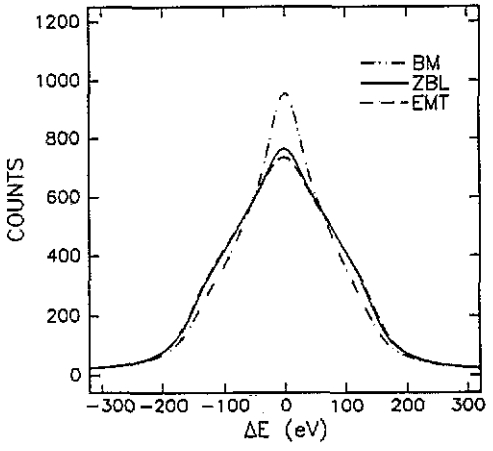

Figure 5. *The simulated lineshapes for the 1499 keV transition obtained with* interatomic potentials illustrated in figure 3. In all cases the lifetime of the 3.26 MeV state was 15.9 fs.

4. Comparison of experimental and simulated γ-ray lineshapes

In order to demonstrate the dependence of the simulated γ-ray lineshapes on the interatomic potential, the results obtained with the ZBL potential are compared to those obtained with the BM and EMT potentials.

The best fit of the experimental 1499 keV γ-ray lineshape to a lineshape simulated with the ZBL potential, was obtained with the lifetime value of 15.9 ± 1.6 fs for the 3.26 MeV state, figure 1. The lineshapes produced with the three potentials are shown in figure 5 for the lifetime value of 15.9 fs. The BM potential is more repulsive than the ZBL potential. The narrower lineshape corresponds to a faster slowing down of the recoiling atoms than obtained with the ZBL potential. The best fit of the lineshape simulated with the BM potential to the experimental γ-ray lineshape, yielded a lifetime value of 9.6 ± 0.5 fs. The EMT potential is less repulsive at distances 1.0–2.0 Å than the ZBL potential. This small difference affects the lineshape and resulted in a longer lifetime value of 18.1 ± 0.7 fs than obtained with the ZBL potential.

The experimental and simulated lineshapes for the 1793 keV transition are shown in figure 2. The best fit between the experimental lineshape and lineshape simulated with the ZBL potential, was obtained with the lifetime value of 83 ± 4 fs. The simulated lineshapes for the 1793 keV transition with different interatomic potentials are shown in figure 6. The lifetime value used was 83 fs. Also in this case the differences between

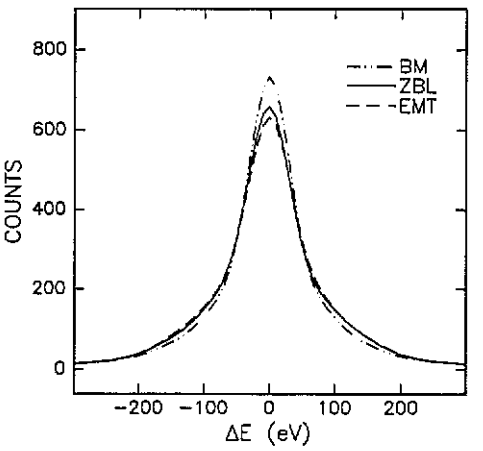

Figure 6. As figure 5 but for the 1793 keV transition. The lifetime value of 83 fs for the 3.18 MeV state was used in the simulations.

the potentials have similar effects on the lineshapes as in the case of the 1499 keV γ-ray lineshape. The reason behind the fact that the differences in the lineshapes are not as strong as in the case of the 1499 keV transition, is the longer lifetime value. The lifetime values obtained in the best fits of the experimental and simulated lineshapes are 54 ± 3 fs and 96 ± 3 fs for the BM and EMT potential, respectively.

5. Discussion

In order to draw conclusions from the accuracy of the ZBL, BM and EMT potentials to describe the Ti–Ti interaction, the accurate and reliable lifetime values of the 3.18 MeV and 3.26 MeV states should be known. The lifetime values reported in the literature for the 3.26 MeV level are 15.4 ± 0.4 fs [3] and 16.8 ± 0.4 fs [11]. These values have been obtained from the analysis of the same GRID data as used in the present work. In the previous studies, the analysis of the stopping process was based on BCA of hard-sphere atoms. The interaction was described with a BM potential.

The lifetime values reported in the literature for the 3.18 MeV level are 95 ± 6 fs [3], 98 ± 6 fs [11] and 110^{+48}_{-36} fs [12]. The first two values were obtained from GRID data with the stopping process described by BCA of hard-sphere atoms. The interaction was described with a BM potential. The last value was obtained in a conventional Doppler shift attenuation analysis of the ^{48}Ti(d,p)^{49}Ti reaction data obtained at considerably higher recoil velocities than those studied in the present work. However, no experimental confirmation of the stopping power has been given in [12]. The uncertainty due to the stopping power was included in the large error limits given .

Thus, at the moment there are no lifetime values available in the literature for the 3.18 MeV or 3.26 MeV states which could be used as reference values for the deduction of the interatomic potential in the Ti–Ti interaction.

In conclusion, the present work describes a new techique to test interatomic potentials and shows that by combining the MD and MC methods for simulation of GRID data different interatomic potentials can be tested accurately. In order to deduce the optimal interatomic potential an accurate and reliable lifetime standard is needed.

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