Slowing down of 3-eV Sm ions in Eu compounds

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(Received 20 June 1995)

Slowing down of ions at ultralow velocities has been studied in the case of 0.02-eV/amu ^{152}Sm recoils in EuF_2, EuF_3, $EuCl_3$, and Eu_2O_3. The recoiling atoms were produced in β decay of ^{152}Eu. The Doppler-broadened 842- and 963-keV γ-rays emitted by the recoiling ^{152}Sm nuclei were detected by a crystal spectrometer. The experimental γ-ray line shapes were simulated by molecular-dynamics technique using empirically derived interatomic pair potentials. The lifetime of 28 ± 6 fs was deduced for the first excited state at 963 keV in ^{152}Sm.

Experimental information concerning the slowing down of atoms in the energy region 1–10 eV in solid materials is very scarce in the literature. Nuclear resonance fluorescence measurements have been applied in the literature in studies of atomic collisions at such low energies.[1–8] The slowing-down models used in these studies are based on the harmonic approximation of lattice vibrations. These models are inadequate in two respects. First, anharmonic effects should be taken into account at the eV energy range. Second, the electron shell rearrangement process after the nuclear decay produces a highly charged recoiling ion. Using bulk material properties (e.g., Debye temperature or phonon density of states) to describe the slowing down of such an ion is questionable.

In this study, we have investigated the slowing down of an ion inside a potential well and the effect of the charge state of the recoiling atom on the slowing-down process. The high resolution of the double flat crystal γ spectrometer GAMS4 installed at the Institut Laue-Langevin was utilized. It offered a unique possibility to experimentally study the slowing-down process at very low energies. This was realized via measurements of Doppler shifts of γ rays emitted by the nuclei of the slowing-down atoms.[9]

The slowing down is simulated by molecular dynamics (MD) calculations[10] where details of the recoil process can easily be incorporated and anharmonic effects are naturally included. We simulate the slowing down of a 3.0-eV Sm ion created by neutrino emission after K electron capture from the 0^- isomeric state in ^{152}Eu in four compounds, namely, in EuF_2, EuF_3, $EuCl_3$, and Eu_2O_3. The Doppler-broadened γ-ray line shapes of the 963- and 841-keV transitions deexciting the 963-keV excited state in ^{152}Sm are used to deduce the lifetime of this state. The experimental neutrino-induced Doppler-broadening (NID) data were measured at the Institut Laue-Langevin using the GAMS4 crystal spectrometer.[9] Examples of experimental NID γ-ray line shapes are shown in Fig. 1.

The MD simulation method used in this work is largely the same as the one used in Refs. 11–13, namely, constant volume and energy calculations with periodic boundary conditions.

The target materials were assumed to be crystalline. The crystal structures, selected according to the phase data[14] for the experimental temperatures (see below) were fluorite,

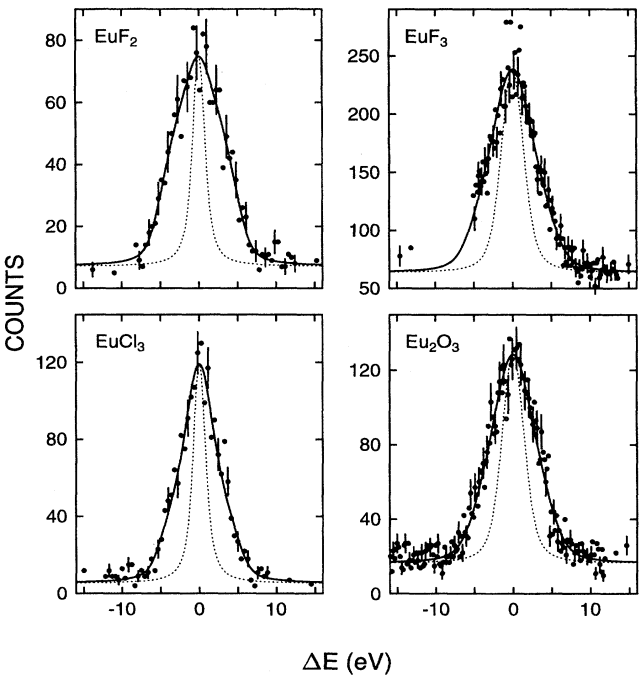

FIG. 1. Experimental (filled circles) and simulated (solid line) γ-ray line shapes for the 842-keV transition in ^{152}Sm measured in different target materials. The instrumental response function is also shown (dashed line). The simulations were performed with the recoil charge equal to the charge of the Eu atom in each lattice.

tysonite, and uranium trichloride for EuF_2, EuF_3, and $EuCl_3$, respectively. For Eu_2O_3 the bixbyite structure (cubic C form) was adopted because the phase corresponding to the experimental temperature has monoclinic structure which is computationally more time consuming to implement in the simulation code than the cubic one.

The computational cells in the simulations comprised 1500 ($5\times5\times5$ unit cells), 1792 ($7\times4\times4$), 1536 ($4\times3\times8$), and 2160 ($3\times3\times3$) atoms for EuF_2, EuF_3, $EuCl_3$, and Eu_2O_3, respectively. Some test runs were performed for EuF_2 using a computational cell of $8\times8\times8$ unit cells containing 6144 atoms. The obtained results were practically identical to those simulated with the 1500-atom cell.

In the beginning of a recoil event, an appropriate isotropically distributed recoil velocity was generated for the recoiling ion. The event was simulated for 300 fs using a time step of 0.5 fs. In order to calculate the Doppler-broadened γ-ray line shape from the time-dependent velocity distribution of the recoiling ion,[13] on the average 200 recoil events were simulated for each target material. Due to low recoil energies, short simulation times, and large computational cells there was no need to perform temperature scaling during the simulations.

Because of the ionic character of the compounds and the low recoil energy, the Coulomb potential affects the slowing down considerably. The potential energy of the long-range Coulomb interaction was calculated using the Ewald summation method.[15] The ionic charge of lattice Eu ions was (in units of the unit charge e) 2+ in EuF_2 and 3+ in EuF_3, $EuCl_3$, and Eu_2O_3. The charges of the anions were assigned so that charge neutrality of the unit cell was achieved. Polarization of the ions was not included in the simulations. The short-range interactions were described by the Buckingham function[16]

$$V_{sr}(r)=A\exp(-r/\rho)-C/r^6. \quad (1)$$

For the O-O interaction the values of the parameters A, ρ, and C were taken from Ref. 17. First estimates for the rest of the parameters were obtained from the model of Ziegler, Biersack, and Littmark.[18] Then the parameters (mostly the cation-anion interaction) were adjusted to obtain approximately right cohesion energies (available for EuF_3, $EuCl_3$, and Eu_2O_3 in the literature[19,20]) and stable structures[14] for the compounds. The results were found to be insensitive to the Eu-Eu and Sm-Eu interactions. The elastic constants calculated[21] with the obtained parameters gave reasonable values for EuF_2 when compared with the experimental ones of Ref. 19. The parameters are given in Table I.

Due to low recoil energies the finite temperature of the target material affects the recoil process through the thermal displacement of atoms and the thermal velocity itself. The temperature was taken into account by simulating the system before initiating the recoil event undisturbed for 2 ps during which the temperature was scaled at intervals of 25 fs. The target temperatures were determined from the broadening of the γ-ray line shape deexciting the long-lived [$\tau=2.1$ ns (Ref. 22)] state at 122 keV in ^{152}Sm. The distribution of the thermal velocities of the Sm ions in the lattice can be described by Maxwell-Boltzmann distribution. This in turn yields a Gaussian-shaped Doppler-broadened γ-ray line-

TABLE I. Parameters for the short-range potential (1) used in the simulations. For Sm, same values as for Eu were used.

Interaction	A (eV)	ρ (Å)	C (eVÅ^6)
Eu–Eu	1715.0	0.317	0.0
Eu–F	3429.1	0.280	14.0
F–F	369.1	0.280	12.5
Eu–Cl	3886.0	0.349	169.6
Cl–Cl	7911.5	0.383	2026.8
Eu–O	5045.4	0.290	34.0
O–O	22764.3	0.149	27.9

shape. From the experimental widths of the line shapes the temperatures of 1040 ± 310 K for EuF_2 and $EuCl_3$, and 1610 ± 340 K for EuF_3 and Eu_2O_3 were determined. Previous γ-ray-induced Doppler-broadening measurements performed at the ILL indicate that the average temperature of the targets is at least 620 K.[9] Simulations were also performed using this temperature. Lifetime results obtained were on the average 0.9 fs shorter than the ones (Table II) obtained using the higher temperatures. This uncertainty caused by the inaccuracy in the target temperature determination is included in the lifetime results.

The electron shell rearrangement process caused by the vacancy in the K shell of the Sm ion created in the electron capture of the Eu ion affects the recoil process in two ways. First, the most energetic Auger electrons and x rays emitted during the rearrangement process affect the slowing down by imparting an extra recoil velocity to the Sm ion. In the simulations, the recoil velocities from the *K-LL*, *K-LM*, *K-LN*, *K-MM*, *K-MN*, *L-MM*, and *L-MN* Auger transitions and *KL* and *KM* x-ray transitions were included. The calculation of the recoil velocities and their relative intensities were based on semiempirical Auger-electron energies,[23] experimental x-ray energies,[24,25] transition probabilities,[26,27] and fluorescence yields.[28] Based on the transition probabilities,[26,27] it was assumed that these Auger electrons and x-ray photons were emitted immediately after the electron capture decay of the Eu ion, i.e., in the very beginning of the recoil event.

Second, the rearrangement process causes the ion to eventually obtain a high charge state[29] and this changes the Cou-

TABLE II. Lifetime of the 963-keV state in ^{152}Sm obtained from NID measurements in different target materials using the equilibrium charges of Eu (2+ for EuF_2 and 3+ for the others) for the Sm ion in simulations. Lifetimes are weighted averages of the values for the 842- and 963-keV transitions. In addition to the statistical uncertainty, the quoted errors include uncertainties of the target temperature (0.9 fs), simulation statistics (2.0 fs), and instrumental response function (1.2 fs for EuF_3 and Eu_2O_3).

Target material	Lifetime (fs)
EuF_2	24.2 ± 2.7
EuF_3	22.4 ± 2.9
$EuCl_3$	36.8 ± 2.8
Eu_2O_3	27.8 ± 2.8

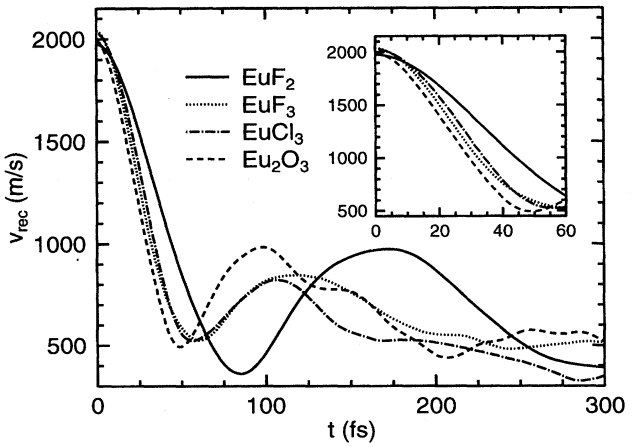

FIG. 2. Average magnitude of the velocity vector of the recoiling Sm ion as a function of time after the nuclear decay of ^{152}Eu as obtained from the MD simulations for different target materials.

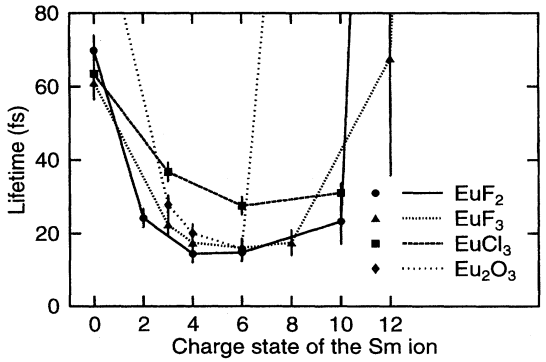

FIG. 3. Dependence of the fitted lifetime of the 963-keV level in ^{152}Sm on the charge state of recoiling ion in different target materials.

lomb potential the ion experiences. Because of the incomplete information available in the literature for the transition probabilities involved in the rearrangement process and the time scale of the neutralization of the highly charged ion in solid material, the charge state of the recoil is not exactly known during the slowing-down process. The effect of the ion charge state was investigated by performing the simulations with different charge states.

Examples of the simulated NID line shapes along with the measured ones are shown in Fig. 1.

The magnitude of the recoil velocity vector of the Sm ion obtained as an average from about 200 MD simulation events for different target materials is shown in Fig. 2. The attenuating oscillatory motion of the recoiling ion is clearly demonstrated. Together with the trajectory plots for a few recoils events, this shows that the ion does not leave its lattice site — as expected in this recoil energy range.

Assuming that the electron shell rearrangement process and the neutralization of the highly charged ion take place in a time scale much shorter than the decay of the excited nuclear state under study, the lifetime of the nuclear state is obtained from the simulations performed using the equilibrium charges of the Eu ion for the recoiling Sm ion. These results are given in Table II. Lifetime values obtained with different target materials are in a fair agreement. Only the chloride value differs substantially from the others. This difference can be attributed to uncertainties in the interatomic potentials because the simulation results were observed to be rather sensitive to the potential parameters. In order to obtain more accurate potentials more experimental data in addition to the cohesion energy and the structure are needed. Moreover, the fact that equilibrium properties of the materials are used to deduce potentials that are used to model interactions in nonequilibrium phenomena might bring some uncertainty to the interatomic potentials.

Figure 3 shows how the charge state of the recoiling ion affects the lifetime to be obtained in the fitting. In all materials the fitted lifetime increases when the charge state goes below $5+$. This can be expected since the Coulomb interaction between the recoiling ion and lattice ions decreases, which in turn lengthens the slowing-down time. However, at large charges ($>+6$) there are differences between target materials. In the case of oxide, the fast increase in lifetime stems from the fact that the crystal structure around the ion is destroyed by the Coulomb potential and the potential energy of the ion is converted to kinetic energy implying a longer slowing-down time. In the case of fluoride and chloride targets the increase in the fitted lifetime at large charges is not as strong as in the case of oxide. In these compounds anions have smaller charges than the O ions in oxide and larger recoil ion charges are required to create disorder in their crystal structures.

The literature values for the lifetime of the 963-keV level are 41 ± 3 fs (Ref. 22) and 29 ± 4 fs.[30] The latter one is in good agreement with the present result 28 ± 6 fs, calculated as an average value of the results for different target materials. It should, however, be kept in mind that the uncertainties given for the literature values do not include the possible systematic errors arising from the analysis of the experimental data. In the result of the present work the inaccuracies in the modeling of the slowing down — originating mainly from the uncertainties in the interatomic potentials — show up as the variation of the lifetime results for different target materials.

In conclusion, we have studied the slowing-down process of a recoiling 3-eV ^{152}Sm ion in ionic target materials, EuF_2, EuF_3, $EuCl_3$, and Eu_2O_3, by simulating the experimental Doppler-broadened line shapes of γ rays emitted by the recoiling nucleus. In the simulations the molecular-dynamics method and rigid-ion pair potentials were employed.

The results show that the slowing down of the recoiling ion is sensitive to the potential well it experiences in the host lattice, i.e., on the interaction potential between the ion and the atoms of the target material. Also, a strong dependence of the slowing down of the ion on the charge state was observed.

This work was supported by the Academy of Finland. The work has been made possible by computer resources from the Center of Scientific Computing, Espoo, Finland. Dr. J. Gale is acknowledged for providing the computer code GULP for the authors' use. J.J. acknowledges support from the Swiss National Science Foundation.

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