Study of Atomic Motion in Oriented EuO Single Crystals Using Neutrino Induced Doppler Broadening

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The motion of atoms caused by neutrino emission following electron capture is observed in EuO crystals by measuring the Doppler shift of subsequently emitted gamma rays. The slowing down of the recoiling atom, which depends on the forces exerted by other atoms in the crystal, is calculated by molecular dynamics simulations using different interatomic potentials. The comparison between measured and simulated gamma-ray line shapes opens the way to a precise determination of the interatomic potential and of the lifetime of the nuclear state. [S0031-9007(97)02810-X]

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We report the first ultrahigh resolution study of Doppler shifts produced by nuclear recoil after neutrino emission in an oriented single crystal. The reaction used, electron capture by the atomic nucleus, leads to the emission of a neutrino which induces a well defined recoil. This is very hard to achieve with gamma rays emitted after thermal neutron capture in medium–heavy nuclei due to gamma cascades [1]. The motion following isotropically distributed recoils causes a Doppler broadening of subsequently emitted γ radiation. By measuring the broadening using the GAMS4 two-axis flat-crystal spectrometer at the Institute Laue Langevin (ILL) one obtains information on the atomic motion. The latter is governed by the interaction of the recoiling atom with the surrounding atoms. The use of an oriented single crystal yields the maximum of information that can be extracted from such a measurement. Various descriptions and theories for interatomic potentials in solids have been proposed and investigated in the literature [2]. They range from *ab initio* total energy calculations [2] up to empirical potentials determined from experimental crystal properties [3] (such as elastic constants, phonon dispersion, lattice constants, etc.). However, due to the lack of appropriate methods, very few theoretical potentials have been tested and compared to experimental data in the energy range between 0.5 and 10 eV. Now, the Doppler broadening technique [4] combined with computational techniques like molecular dynamics (MD) simulations permits one to observe and test the reliability of such potentials in the mentioned energy range.

The movement of the atom depends on its thermal motion and on the extra kinetic energy given by emission of a neutrino after electron capture [5]. After the nuclear reaction, the atomic nucleus resides in an excited state with a certain lifetime τ and gamma rays will depopulate this nuclear level. The lifetime defines a characteristic time interval (typically 4τ) in which most gamma rays are emitted. Therefore, the atomic velocities within this time interval are responsible for the Doppler broadening. The motion of the atom consists first in a slowing down and subsequently in a thermal movement. The nuclear lifetime decides which kind of motion dominates the Doppler broadening: while the slowing down takes a certain time the thermal motion is of infinite duration. For instance, a long lifetime makes the fraction of gamma rays emitted during the slowing down, a very fast event, negligible compared to those emitted after thermalization.

In this work, line shapes from neutrino induced Doppler broadening (NID) experiments obtained with an oriented EuO single crystal are compared to the line shapes calculated on the basis of MD simulations of the recoiling atoms. The purpose was to test pair potentials for Eu-Eu, Eu-O, and O-O. In this context the use of single crystals promises to have a better sensitivity than former experiments realized with powder targets [5,6]. With oriented EuO crystals, two advantages appear. On the one hand no averaging over all directions of observation with respect to the crystal orientation needs to be done and on the other hand all the atoms sit in similar surroundings, such that only one position has to be simulated, because the fcc structure of the crystal [7] leads to unique interatomic distances.

The experiment was performed using three single-crystal targets with a total mass of 672 mg embedded in a graphite matrix and placed with the $\langle 100 \rangle$ orientation in the direction of the spectrometer. The crystals were placed at the in-pile target position of GAMS4 under a neutron flux of $5 \times 10^{14}\ \mathrm{cm}^{-2}\,\mathrm{s}^{-1}$ [8]. After neutron capture, 36% of the formed ^{152}Eu atomic nuclei end up in the 0^- state at 45.6 keV [9]. This isomeric 0^- state decays with a half-life of 9.3 h with 28% via electron capture forming ^{152}Sm. After electron capture a 1^- level at 963.4 keV in ^{152}Sm is populated (see Fig. 1). Some other states are also created in this process but they play a minor role in this experiment, since the feeding of the 963.4 keV level by electron capture represents 99.97% of the total feeding [5]. The 1^- level decays with a lifetime of 29(4) fs [9] via emission of either a 963.4 keV gamma ray to the ground state, or a 841.6 keV gamma ray to a

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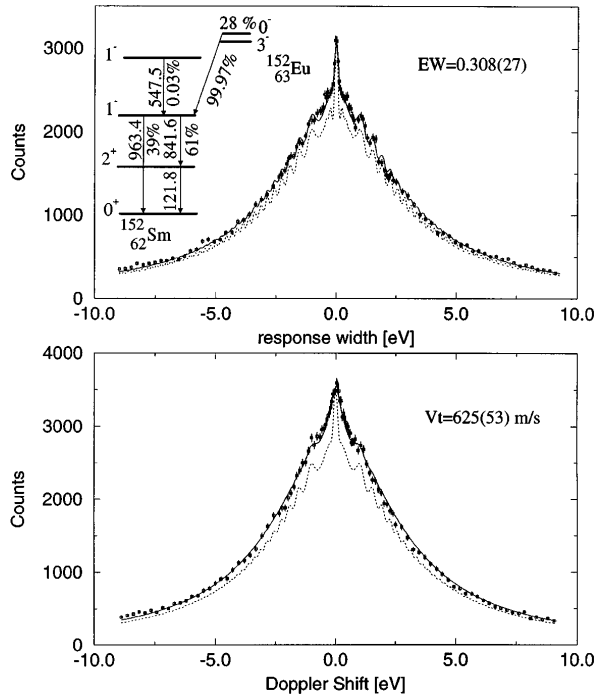

FIG. 1. The gamma-ray line shape of the 121.8 keV transition in first order of reflection obtained from a sum of 10 nondispersive scans (top) and from a sum of 12 dispersive scans (bottom). In the upper part, the dotted line represents the diffraction theory and the full lines correspond to the diffraction curve folded with a fitted excess width. In the lower part, the dotted line corresponds to the instrumental response as determined in the upper part and the full line is the fit which includes the thermal broadening. A sum of several measured scans is represented, since it is preferable to perform many short scans to avoid effects due to slow drifts of the spectrometer. The inset shows the partial level scheme for the nuclear reaction.

2^+ long lived ($\tau = 1.44$ ns) state. The last gamma ray is followed by a 121.8 keV gamma ray to the ground state. The Doppler broadening of the line shape originates from the recoil given to the atom by emission of a neutrino at the moment of the electron capture (β^+ decay). The initial energy of the recoiling atom E_r is given by

$$E_r = \frac{E_\nu^2}{2Mc^2} = \frac{Q^2}{2Mc^2}, \qquad (1)$$

with Q the energy liberated in the reaction, M the atomic rest mass, c the velocity of light, and E_ν the neutrino energy. Formula (1) gives a kinetic recoil energy $E_r = 3$ eV for the ^{152}Sm atom [5]. Another process affects the atomic recoil: After the reaction, an electron is missing, in most cases a K electron [10], and the rearrangement process induces either an x ray (in 90% of the cases) or an Auger electron (10%) [11]. Both of these processes impart also a small recoil to the atom. X rays and Auger electrons are emitted immediately after the electron capture decay, therefore at the beginning of the recoil.

The total line shape is described by three contributions: (i) the instrumental response, (ii) Doppler broadening due to the thermal motion of the atoms, and (iii) the recoil induced Doppler broadening. The instrumental response can be determined by nondispersive scans of the transition in question and is given by dynamical diffraction theory folded with a small Gaussian extra broadening (excess width). Thermal broadening can be measured in dispersive mode [3] using transitions emitted from nuclear states having a long lifetime (here the 121.8 and the 221.2 keV, a gamma ray emitted after neutron capture) where the nuclei had sufficient time to thermalize. The nondispersive and dispersive line shapes of the 121.8 keV line in first order of reflection are given in Fig. 1. With a total of 25 scans, the 121.8 and 221.2 keV lines result in a thermal velocity of 598(47) m/s for the Eu atom. This thermal velocity corresponds to an effective crystal temperature of 2173(344) K. This temperature is above the calculated value ($\sim$1600 K) for a EuO crystal with the specific irradiation conditions at the target position. In the present analysis, the measured value was adopted. The sensitivity of lifetime to temperature was checked by starting the molecular dynamic simulations with different crystal temperatures and affects the lifetime value by less than 5.0%.

Since the 1^- state of ^{152}Sm reached after the electron capture is short lived, the Doppler broadening of the 841.6 keV transition comes essentially from the recoil after the neutrino emission. The gamma-ray line shape is calculated by simulating the motion of the atom using a constant energy and volume molecular dynamics method. At the beginning, the 1000 atoms of the MD cells are placed in a fcc array and periodic boundary conditions are applied in all three dimensions. Before giving the initial kick simulating the recoil after neutrino emission ($v_r/c = 6.54 \times 10^{-6}$) to the most central atom, all atoms in the cell are thermalized to the desired temperature of 2173 K by scaling the velocity of a few percent of the atoms each 20 fs. The thermalization is then simulated for a 15 ps period with a time step of 0.5 fs. After 3 ps the cell has a constant temperature and the velocity distribution of the atoms can be described by a Maxwellian. Once thermalization is achieved, snapshots of positions and velocities were recorded every 100 fs. They are taken as initial conditions for the second MD program which includes the kick induced by the neutrino emission to the central atom in the cell. As mentioned before, orbital electron capture produces x rays or Auger electrons by the immediate refilling of the vacated orbit. These reactions give a small recoil (around 5%–15% of the neutrino induced recoil) to the atom and are included in the MD program for the most probable x rays and Auger transitions. The probabilities and energies of these transitions are taken from Ref. [12]. In the second program, the motion of the recoiling atom is followed during 125 fs, which corresponds to 4 times the maximum expected lifetime τ of the nuclear state. The contribution to the line shape of the gamma rays emitted after 4τ is negligible, since, following the exponential decay law, the number of gamma rays emitted after 4τ

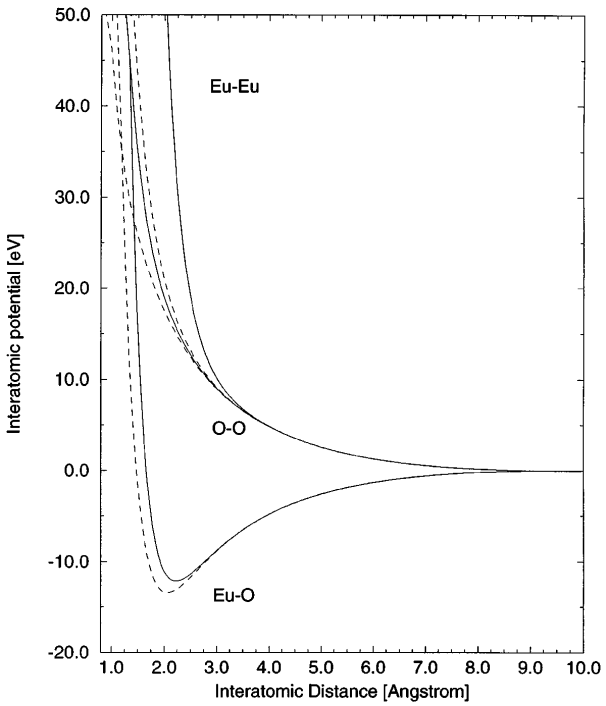

FIG. 2. Theoretical MD potentials for Eu-Eu, Eu-O, and O-O interactions. The solid line represents the Born-Mayer and the dotted line the Buckingham potential.

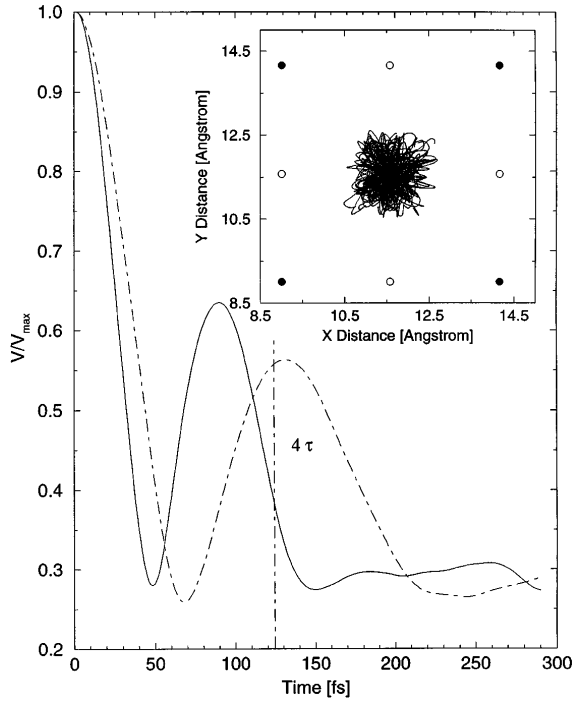

FIG. 3. Average velocities as a function of time for 250 recoiling Sm atoms in a EuO single crystal obtained with MD simulations. The full line represents the Born-Mayer and the dotted line the Buckingham potential. The vertical line shows the 4τ simulation limit. The inset shows the MD trajectories of 250 recoiling Sm atoms projected in the $\langle 100 \rangle$ plane using the Buckingham potential. The open circles denote the O and the full circles the Eu atoms equilibrium positions.

equals 1.8%. Both MD programs solve numerically the Newtonian equations of motion for each atom using a Verlet algorithmn[13]. The total number of events (one event corresponds to the following of a recoiling atom during 4τ) is equal to 1500. The two different potentials used in the simulations are shown in Fig. 2 and are of the form

$$V_{1,2}(r) = \frac{q_1 q_2}{4\pi\varepsilon_0} \cdot \frac{1}{r} + A \exp\left(-\frac{r}{\rho}\right) - \frac{C}{r^6}, \quad (2)$$

with the different parameters listed in Table I. Both potentials contain a Coulomb term for the ionic interaction. To $V_1(r)$ only a Born-Mayer term for the repulsive interaction due to the overlap of the electron clouds ($C = 0$) [14,15] is added, whereas $V_2(r)$ takes into account a Buckingham dispersive term calculated using crystal parameters given in Ref. [6]. In both potentials q_1 and q_2 represent the ionic charges and are equal to $+2$ for Eu and -2 for O atoms. The long-range force is treated using a cutoff radius method. The more general Ewald method used to treat the long-range part of the potential was also tested in this work. However, since it does not significantly change the final result, it was neglected to reduce the simulation time. Although after the nuclear reaction the recoiling atom is no longer a europium atom, no distinction is made for the calculation of the potential for Eu or Sm, since Sm and Eu are chemically closed elements ($4f$, rare earth). Figure 3 gives a comparison between the average velocity as a function of time for 250 recoiling Sm atoms obtained with both potentials. The inset shows the projected trajectories for 250 recoiling atoms given by the MD simulation using the Buckingham potential. It is clear from Fig. 3 that the slowing-down process is faster for the Born-Mayer simulations and therefore thermalization is achieved more rapidly.

Once the instrumental response is measured by nondispersive scans of the line, one can calculate the gamma-ray line shapes for different lifetime values from the MD

TABLE I. Parameters of the different theoretical MD potentials for the EuO crystal.

Potential interaction	Eu-Eu	O-O	Eu-O
Born-Mayer	$A = 42\,020$ eV	$A = 2143.4$ eV	$A = 10\,048.38$ eV
$V_1(r)$	$\rho = 0.285\,08$ Å	$\rho = 0.263\,96$ Å	$\rho = 0.274\,49$ Å
Buckingham	$A = 1715.0$ eV	$A = 22\,764.3$ eV	$A = 5045.4$ eV
$V_2(r)$	$\rho = 0.317$ Å	$\rho = 0.149$ Å	$\rho = 0.290$ Å
	$C = 0.0$ eV Å^6	$C = 27.9$ eV Å^6	$C = 34.0$ eV Å^6

simulations of the slowing down. Using the least-squares fitting routine GRIDDLE [16] such line shapes are fitted to the experimental data, with the lifetime and experimentally related numbers (background, intensity, etc.) as free parameters. As a result of this procedure a lifetime value τ and a χ^2 per degree of freedom are obtained. Both permit us to quantify the agreement/disagreement between theory (MD simulations) and experiment (NID data). The result of the fit gives for the Born-Mayer potential a lifetime of 17.2(7) fs with a χ^2 of 1.149. This result is much lower than the reference value of 29(4) fs [9] and is due to the repulsive part of the Born-Mayer potential, which is too steep at short distances (see Fig. 2). The atoms are in this case more bound in the lattice and the slowing-down process is much faster which decreases the fitted lifetime of the nuclear state under study. The other potential gives a lifetime of 28(1) fs with a χ^2 equal to 1.148. This value agrees very well with the lifetime mentioned in Ref. [9]. The fit of 28 scans of the 841.6 keV line obtained with the Buckingham potential is shown in Fig. 4.

In conclusion, we can state that for EuO and the given energy range the Buckingham potential is much more suited since it reproduces the nuclear lifetime. The Born-Mayer potential is not a good candidate since the disagreement between the reference and the fitted lifetime is quite pronounced. For our sets of measurement we conclude that a repulsive interaction originating only from the overlap of the electron clouds is not sufficient and that a dispersive term is needed in the 1–100 eV range.

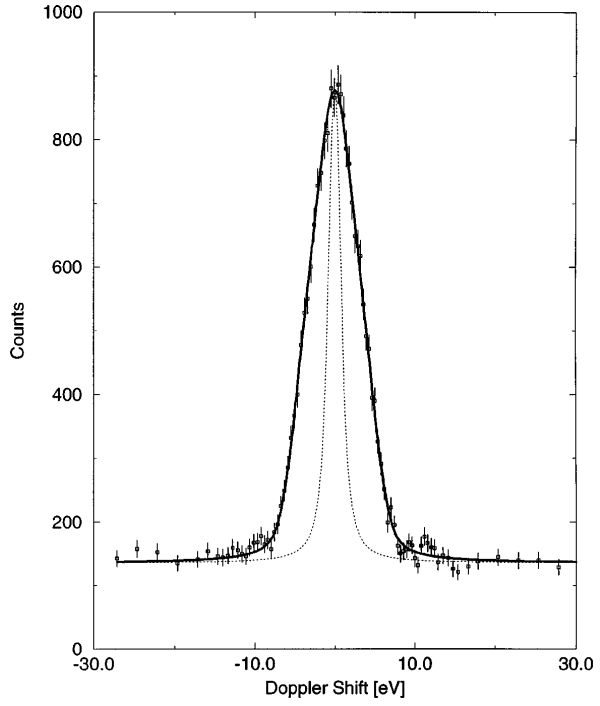

FIG. 4. Experimental (squares and error bars) and simulated (solid line) gamma-ray line shape of 28 scans for the 841.6 keV transition. The Buckingham potential is used in the simulations and a lifetime of 28(1) fs is fitted. The dotted line represents the instrumental response function of the GAMS4 spectrometer obtained from nondispersive scans.

The NID method permits the testing of the reliability of different potentials at low energy. It can determine which potential describes the best the interaction between atoms in a solid. The method used here allows also the determination of a much more precise nuclear lifetime. The error is 4 times smaller than the one given in the literature [9]. The line shape of the broadened gamma-ray line should vary with different crystal orientations because of the crystalline structure symmetries. The use of single crystals is therefore preferable over powder targets, because it allows one to check line shapes from different orientations [17]. For this purpose other orientations of single crystalline EuO will be tested at the GAMS4 spectrometer in the future and more detailed MD simulations will be performed to investigate the influence of (1) other types of potentials and slowing-down theories, (2) the effects of crystal orientation on the line shape, and (3) the temperature of the crystal.

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