Nuclear inelastic scattering with ^{161}Dy

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We have extended the technique of nuclear inelastic scattering to the 25.651 keV nuclear transition of ^{161}Dy. Energy spectra of nuclear inelastic absorption of synchrotron radiation in ^{161}Dy metal foil were measured at room temperature and at $T = 15$ K with 1.0 meV resolution. From low temperature data a density of phonon states and several thermodynamic parameters were derived.

Nuclear inelastic scattering made a fast progress during the last years. Discovered in 1995,[1] it has progressed from first demonstrational experiments[2,3] to an established technique.[4] This rapid development became possible due to several distinct advantages: the technique is fast, precise, requires only a small amount of material and can be applied to samples in various aggregate states.

However, nuclear inelastic scattering also has a serious limitation which follows from the resonant nature of nuclear interaction: the method can only be applied to the so called ''Mössbauer isotopes.'' On the other hand, this has the advantage that it allows one to determine element specific dynamics. Up to now, the main progress has been achieved with the ^{57}Fe isotope.[1–4] In addition to that, several successful experiments with ^{119}Sn were performed[5–7] and first results with ^{151}Eu were reported.[8] Thus, the list of elements explored with nuclear inelastic scattering is far from being complete, and the further extension of the technique to other suitable isotopes is an obvious demand.

This paper demonstrates the feasibility of nuclear inelastic scattering with ^{161}Dy. We have measured the energy spectra of phonon-accompanied nuclear inelastic absorption of synchrotron radiation in a dysprosium metal foil at room temperature and at 15 K. The low-temperature data allowed us to determine the density of phonon states and to calculate several thermodynamic parameters.

The first excited nuclear state of ^{161}Dy has an energy of 25.651 keV and a lifetime of 40.7 ns. The energy is higher than those of the previously explored ^{151}Eu (21.541 keV) and ^{119}Sn (23.880 keV) isotopes. This makes crystal optics design harder. However, the longer lifetime compared to Sn and Eu allows an easier separation of the delayed nuclear scattering from the instantaneous electronic scattering. This considerably simplifies the measurements of nuclear inelastic scattering in case of ^{161}Dy.

The first excitation of the 25.651 keV nuclear level of ^{161}Dy by synchrotron radiation was observed in an incoher-

ent scattering geometry with a $\sim$50 meV bandpass monochromator.[9] More recently, nuclear forward scattering of synchrotron radiation by dysprosium foil has been studied using an exact back-scattering monochromator with a 6.8 meV bandpass.[10] With the same monochromator nuclear inelastic scattering measurements have been attempted as well.[10] However, this energy resolution is far from being enough in the case of dysprosium metal, which has a Debye energy of about 15 meV.

For the present studies, we have elaborated a high energy-resolution monochromator according to the conventional ''nested'' scheme[11,12] with an energy bandpass of 1 meV. For the outer channel-cut crystal an asymmetric Si (4 4 4) reflection with an asymmetry factor of 0.10 and for the inner channel-cut crystal a symmetric Si (18 12 6) reflection were chosen.

The experiment was performed at the nuclear resonance end-station ID22N at the European Synchrotron Radiation Facility during a 16 bunch mode of storage ring operation. The description of the standard beamline setup for nuclear resonant scattering may be found in Ref. 13. Details of the nuclear inelastic scattering technique are described in Ref. 4. Similar to our previous experiments,[14] we used a collimating compound refractive lens in order to improve the throughput of the monochromator. The 1.0 meV bandpass of the monochromator was evaluated from a fit of the instrumental function with a Gaussian distribution. Within this bandwidth, the monochromator provided 5.7×10^6 photons per second at 90 mA current of the storage ring.

The sample, a 2×5 mm^2 large and 50 μm thick foil of dysprosium metal, was enriched to 95.7% in the resonant ^{161}Dy isotope. For low-temperature measurements the sample was mounted inside a closed cycle cryostat.

Similar to the case of the ^{57}Fe isotope, the deexcitation of a ^{161}Dy nucleus via the internal conversion channel results in the emission of atomic fluorescence radiation with relatively low energies L_α (6.5 keV), L_β (7.2 and 7.6 keV), and L_γ (8.4
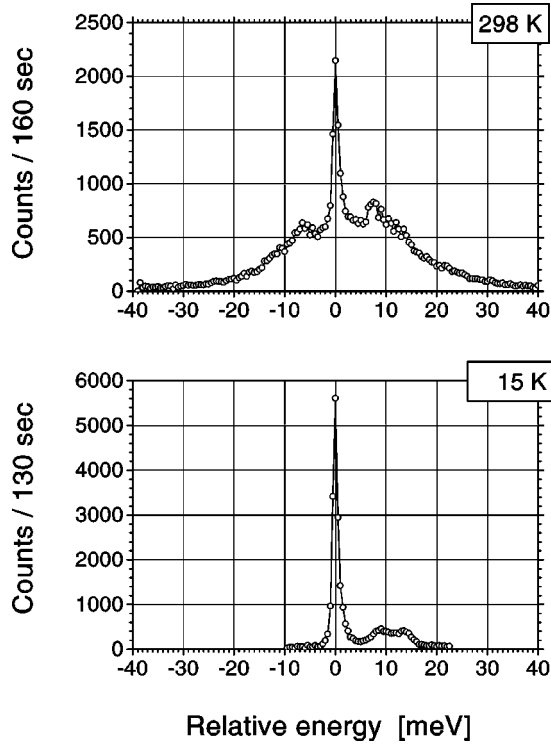
FIG. 1. Energy spectra of nuclear inelastic absorption of synchrotron radiation in a ^{161}Dy metal foil at room temperature and at 15 K.

keV) lines, with L_α being the most probable channel. Due to the higher efficiency of avalanche photodiode detectors[15] for this radiation compared to the scattered 25.651 keV quanta and due to high probability of internal conversion, we measured mostly nuclear *absorption*, with only a negligible contribution of nuclear *scattering*.

The energy spectra of nuclear absorption are shown in Fig. 1. Here the intensity of atomic fluorescence is plotted versus the difference between the energy of the incident radiation and the exact energy of the nuclear resonance. The count rate in the inelastic peak at ~9 meV was about 5 counts per second (cps) at room and about 4 cps at low temperature. The spectra give the probability of nuclear inelastic absorption as a function of energy transfer due to phonon annihilation ($E<0$) and phonon creation ($E>0$) in the lattice, respectively. According to the law of detailed balance the annihilation part vanishes at low temperature.

At room temperature, the energy spectrum of nuclear inelastic absorption is dominated by multiphonon contributions. The statistical accuracy of these data is insufficient to extract directly the single-phonon term and to derive the density of phonon states. In contrast to that, the energy spectrum measured at 15 K consists mostly of single-phonon absorption with only a small contribution of multiphonon components. We have derived the density of phonon states from these data according to the conventional procedure described, e.g., in Refs. 2,4. A double Fourier transformation technique[16,17] was used in order to eliminate multiphonon contributions. Due to some uncertainty in the instrumental function, subtraction of elastic contribution of scattering (i.e.,
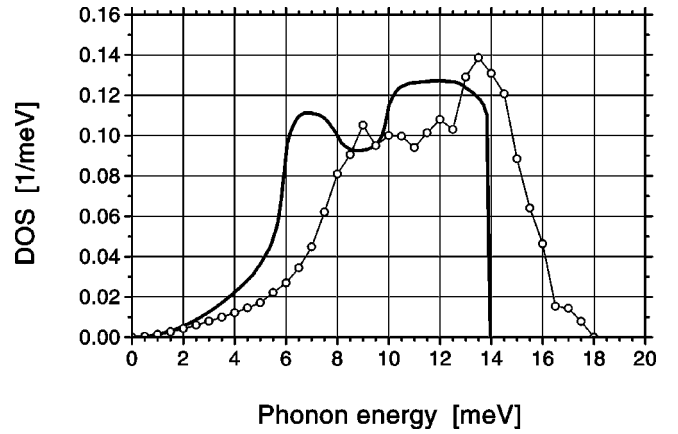

FIG. 2. Density of phonon states for Dy metal: open circles and thin line (to guide the eyes) show the density of phonon states as derived from the energy spectrum of nuclear inelastic absorption measured at 15 K. The solid line shows the results of the theoretical calculations of Ref. 20, where the lattice dynamics of dysprosium was evaluated using an interpolation between terbium and holmium elastic constants at room temperature.

the central peak) was not reliable within the energy range of ±5 meV. For the density of states in this energy region we used the Debye approximation. The derived density of phonon states is shown in Fig. 2.

In general, an experiment on nuclear inelastic absorption does not necessarily provide an exact density of states $g(E)$, which is

$$g(E) = \frac{1}{3n_c} V_0 \sum_j \int \frac{d\mathbf{q}}{(2\pi)^3} \delta(E - \hbar\omega_j(\mathbf{q})), \qquad (1)$$

but rather a partial density of phonon states for atoms of the resonant isotope, which is projected on the direction of the incident x-ray beam[16,17]

$$\widetilde{g}(E,\mathbf{s}) = \frac{1}{n_r} V_0 \sum_{j,a} \int \frac{d\mathbf{q}}{(2\pi)^3} \delta(E - \hbar\omega_j(\mathbf{q})) |\mathbf{s} \cdot \mathbf{e}_{j,a}(\mathbf{q})|^2. \qquad (2)$$

Here and above n_c and n_r are the total number of atoms and the number of resonant atoms in the unit cell, respectively; V_0 is the volume of the unit cell; the indices a and j are used, respectively, for resonant atoms in the unit cell and vibrational modes; E and $\mathbf{q}$ are phonon energy and wave vector; $\mathbf{s}$ is the unit vector along the direction of the incident x-ray beam; $\mathbf{e}_{j,a}(\mathbf{q})$ is the polarization vector of vibrations of the ath resonant atom and the jth vibrational mode.

In general, the partial projected density of states $\widetilde{g}(E,\mathbf{s})$ differs from the exact density of states $g(E)$, because contributions of various vibrational modes are weighted by the squared projections of the polarization vectors of the resonant atoms on the direction of the incident beam. In some cases, however, nuclear inelastic absorption gives the exact density of phonon states. One well-known case, for instance, is the cubic Bravais lattice.[18] Another case, which takes place here, corresponds to a polycrystalline sample with an

arbitrary monoatomic lattice. At room temperature, dysprosium has a hcp ($P6_3/mmc$) lattice. Therefore, in the case of a single crystal, the projected density of states has a polar anisotropy relative to c axis,[19] and differs from the exact density of states. In the polycrystalline case, however, one has to average Eq. (2) over all directions with respect to the incident x-ray beam. We introduce the tensor

$$T^{\mu\nu}(E) = \frac{1}{n_r} V_0 \sum_{j,a} \int \frac{d\mathbf{q}}{(2\pi)^3} \delta(E - \hbar\omega_j(\mathbf{q}))(e_{j,a})_\mu(e_{j,a})_\nu^*.$$
(3)

Here the indices $\mu,\nu = x,y,z$ number vector coordinates. Using this tensor for a polycrystalline sample without a texture the averaging over all directions of the incident beam can be expressed as

$$\hat{g}(E,\mathbf{s}) = \frac{1}{4\pi} \int d\mathbf{s} \sum_{\mu,\nu} s_\mu T^{\mu\nu}(E) s_\nu$$

$$= \sum_{\mu,\nu} T^{\mu\nu}(E) \frac{1}{4\pi} \int d\mathbf{s}\, s_\mu s_\nu.$$
(4)

One can directly check that the integral over all directions is zero for all nondiagonal components of the tensor, whereas it is 1/3 for all diagonal components. Thus, one arrives at

$$\hat{g}(E,\mathbf{s}) = \frac{1}{3} \sum_\mu T^{\mu\mu}(E)$$

$$= \frac{1}{3n_r} V_0 \sum_j \int \frac{d\mathbf{q}}{(2\pi)^3} \delta(E - \hbar\omega_j(\mathbf{q}))$$

$$\times \left\{ \sum_a |\mathbf{e}_{ja}(\mathbf{q})|^2 \right\}.$$
(5)

For a monoatomic lattice, the sum inside the figure brackets includes all atoms of the unit cell. By definition, it equals unity, so that Eq. (5) becomes identical to Eq. (1). Thus, for a polycrystalline sample with a monoatomic lattice nuclear inelastic absorption provides the exact density of phonon states.

In Fig. 2 we compare the density of phonon states obtained in our measurements with that from Ref. 20, where the dynamics of dysprosium lattice was evaluated using an interpolation between the elastic constants of terbium and holmium. One should note that the experimental data are smoothed by the finite energy resolution of 1.0 meV, whereas the theoretical curve is calculated for an ideal instru-

mental function. Our results show that, in first approximation, an interpolation of the terbium and holmium data gives an acceptable description of the lattice dynamics in dysprosium. At closer examination, however, one notes that the experimental spectrum is shifted to higher energies. One should, however, keep in mind that the experimental density of states corresponds to a temperature of 15 K, whereas in the theoretical estimations room temperature data were used. Therefore, the difference might be explained by damping of lattice vibrations at room temperature.

In addition to the density of phonon states from the low-temperature ($T = 15$ K) experimental data we have derived the average Lamb-Mössbauer factor $f_{LM} = 0.78 \pm 0.01$ and the average mean-square displacement of the dysprosium atoms $\langle x^2 \rangle = 14.7 \pm 0.7$ pm^2. In both cases, the experimental uncertainties were estimated from deviations of the values obtained directly from the measured energy spectra and those calculated from the density of phonon states.

In summary, we have demonstrated the feasibility of nuclear inelastic scattering for the 25.651 keV nuclear transition of ^{161}Dy. To our knowledge, this is the highest energy at which nuclear inelastic absorption was applied. Energy spectra of nuclear absorption by a ^{161}Dy foil were measured at room temperature and at 15 K. The density of phonon states in dysprosium metal was derived from the experimental data taken at 15 K. Lamb-Mössbauer factor and atomic mean-square displacement were calculated. Our confidence in these results was aided by work at SPring-8, where measurements with a 0.5 meV monochromator has given similar results.[21]

In the future, several important applications of nuclear inelastic spectroscopy with ^{161}Dy may be foreseen. For instance, dysprosium enters several high-temperature superconducting materials. Nuclear inelastic absorption studies with these compounds may give insight into their lattice dynamics and thermodynamics. Several dysprosium compounds have strong magnetic activity, and this technique can help clarifying, for instance, the influence of magneto-acoustic interactions on lattice dynamics. Finally, dysprosium forms stable complexes with fullerenes, which are model objects for studies of rotational dynamics. This challenges an application of nuclear inelastic absorption to the new field of soft excitations.

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