Observation of Localized Modes in TbO\textsubscript{x} Using the Mößbauer Effect

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Received December 5, 1978

The resonant absorption of the 58 keV Mößbauer line under excitation of one phonon states in TbO\textsubscript{x} was investigated. Using an ultra centrifuge, the γ energy was shifted by ±25 meV. Narrow absorption lines at 14.1 meV, 16.0 meV and presumably at 12.3 meV are interpreted as being caused by localized modes of the terbium ion.

Introduction

If an excited nucleus is bound in a crystal, the recoil of a deexciting quantum may lead to the excitation of lattice vibrations, whereas with a probability \( f_i \) phonons are created and/or absorbed. Thus, the shape of the γ spectrum is determined by the dynamics of the lattice. The analogous consideration is valid for the resonant absorption of the γ radiation. Soon after the discovery of the Mößbauer effect, Vissher [1] proposed to use the narrow Mößbauer line \((i=0)\) of a γ source to investigate the phonon spectrum of a resonant absorber. Since \( f_i \) is related approximately to the Debye Waller factor (DWF) \( f_0 \) by \( f_i = f_0 \left( -\ln f_0 \right)^i \), for this investigation only γ sources are suitable having large values of \( f_0 \) so that contributions of \( i \geq 2 \) are negligible.

Previous experiments failed due to the small cross sections for resonant absorption. Recently [2], a successful experiment was performed using the 58 keV transition in \(^{159}\text{Tb}\) for the investigation of the phonon spectrum of terbium metal. The main advantage of this method is the high energy resolution feasible which in principle is limited by the natural line width of \( F_{58 \text{ keV}} = 7.9 \mu\text{eV} \). Therefore the study of localized modes seems to be a typical application.

In the present investigation, the resonant absorption of the 58 keV Mößbauer line in TbO\textsubscript{x} \((x=1.5 \ldots 2)\) was studied. The γ energy was varied by moving the source in an ultra centrifuge. TbO\textsubscript{x} was chosen, since in infrared spectroscopic studies [3] the rare earth oxides of the type RE\textsubscript{2}O\textsubscript{3} exhibited narrow absorbing states at 10.9 meV, 15.9 meV and 16.6 meV which were interpreted as localized modes, where the RE ion oscillates in a potential formed by the rigid lattice of the surrounding atoms. The crystal structure of TbO\textsubscript{x}\footnote{supplied by E. Merck, Darmstadt} is uncertain. Although x ray analysis by the Debye Scherrer method showed the similarity to the RE\textsubscript{2}O\textsubscript{3} structure, the infrared spectra [3] yielded, besides of electronic excitation states, only one distinct absorption line at 12.3 meV.

Experiments

Figure 1 shows the experimental arrangement. The ultra centrifuge was driven by an electro motor. The revolutions per second were stabilized to be ±1 rps. Measurements were performed between −200 rps and +400 rps corresponding to source velocities between −75 m/s and 150 m/s.

At the circumference of the rotor, four sources\footnote{We thank the KFA Karlsruhe for several irradiations in the FR 2-reactor} of \(^{159}\text{Dy}_2\text{O}_3\) were attached each having a strength of about 10 mCi. The rotor was surrounded by a copper shield which was cooled by liquid nitrogen. 1 Torr of helium was used as contact gas. By this means, the temperature of the sources amounted to 85 K independent of the velocity. The fraction of the recoilfree emitted 58 keV quanta was \( f_0 = 22 \% \) [4].
The terbium absorber consisted of 300 mg/cm$^2$ TbO$_x$ and was kept in a helium cryostat at a temperature of 20 K. The transmission of the $\gamma$ radiation through the absorber was determined using a germanium detector. A laser beam reflected by a mirror which was attached to the rotor axis indicated the position of the sources. $\gamma$ radiation was registered only when a source passed the interval indicated in Fig. 1. In order to accomplish a good velocity resolution, the multiscaling technique was applied and during each passage of a source through the region, 16 subsequent spectra were taken. By this, the region was subdivided into subregions each corresponding to an angle of 3.75°, yielding an energy resolution of 100 $\mu$eV at an energy shift of 10 meV.

Figure 2 shows the $\gamma$ spectrum after traversing the TbO$_x$ absorber. In order to avoid uncertainties due to velocity dependent distortions of the rotor axis and delays in the registration, only the intensity ratio, $I_{58}/I_\beta$, of the 58 keV radiation to the $K_\beta$ radiation was considered in the analysis. Control measurements using a Gd$_2$O$_3$ absorber showed that this ratio is velocity independent, when no resonance absorption takes place.

In Fig. 3, $I_{58}/I_\beta$ averaged over the 16 subspectra is plotted versus the maximum source velocity. Resonance absorption is indicated by a decrease of $I_{58}/I_\beta$.

Fig. 1. Experimental arrangement. The four $^{159}$Dy sources are cooled to 85 K. The source velocity was varied between -75 m/s and 200 m/s. $\gamma$ quanta were registered only, when a source was in the indicated region.

Fig. 2. $\gamma$ spectrum of a $^{159}$Dy-source after traversing the TbO$_x$ absorber. In the analysis, the ratio $I_{58}/I_\beta$ was determined.