Phonon Absorption-Spectroscopy in the Presence of Strong Elastic Phonon Scattering

J. Mebert¹, O. Koblinger², S. Döttinger¹, and W. Eisenmenger¹

¹Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57,
D-7000 Stuttgart 80, Fed. Rep. of Germany

²IBM-Deutschland, GmbH, Schönaicherstr. 220,
D-7030 Böblingen, Fed. Rep. of Germany

1. Introduction

Rare Earth doped crystals are known to show very sharp absorption lines in IR-absorption measurements due to 4f-intra-shell transitions of the Rare Earth dopants. The line sharpness is caused by screening the 4f-orbit by electrons occupying the spatially extended 5s and 5p shells. Absorption lines have been found at 21.2 cm^{-1} and 32.8 cm^{-1} in Er^{3+} doped CaF_2 [1] and at 52 cm^{-1} in Er^{3+} doped LaF_3 [2].

In this energy range phonon spectroscopy is a good and useful method in absorption measurements. The predominant advantages are the very high frequency resolution of about 10 GHz (in IR typical 30 GHz) and the existence of other coupling mechanisms for the 4f-intra shell transitions. In IR-spectroscopy these transitions are characterized by a small oscillator strength because they are forbidden in first order by the parity selection rule. Phonon transitions are induced by orbit-lattice coupling allowing transitions without parity change.

In pulse measurements a strong elastic phonon scattering can be observed. Such elastic phonon-scattering was also published by different other authors [3/4] in recent times. In this work we show that in the presence of a strong phonon scattering background absorption structures can only be well resolved by reducing sample thickness to the phonon mean free path. This mean free path can be determined by analyzing the pulse shape of 285 GHz phonons. By reducing sample thickness to the appropriate value of 0.3 mm the 21.2 cm^{-1} crystalline field transition in $\text{CaF}_2:\text{Er}^{3+}$ could be evaluated with the very high resolution of 5 GHz. In experiments performed on a 1mm thick $\text{LaF}_3:\text{Er}^{3+}$ sample we observed an absorption line at 14.2 cm^{-1} not visible in FIR absorption measurements.

2. Phonon spectroscopy measurements at $\text{CaF}_2:\text{Er}^{3+}$

Phonon spectroscopy measurements with an Al and a Sn tunneling junction as voltage tunable phonon generator and detector respectively were applied to Er^{3+} doped CaF_2

using the well known modulation technique. The basic intention was to evaluate the crystalline field absorption lines reported above. Fig. 1 shows the corresponding phonon absorption spectrum. The sample thickness in this case was $d = 2\text{ mm}$. At 636 GHz (21.2 cm^{-1}) a weak, hardly resolved absorption line can be recognized. This structure can be ascribed to the 21.2 cm^{-1} absorption line known from FIR measurements.

Fig. 1 Phonon absorption spectrum of $\text{CaF}_2:\text{Er}^{3+}$
 $d = 2\text{ mm}$

3. Pulse measurements with superconductive Sn-tunnel junctions

In this technique superconductive Sn-tunnel junctions are used as well as phonon generator and phonon detector. The advantage of this arrangement is the possibility of investigating nonequilibrium phonon propagation at the fixed frequency of 285 GHz coupled with a high S/N ratio. Fig 2a, b, and c show the Sn/Sn pulse-measurements of $\text{SrF}_2:\text{Eu}^{2+}$, $\text{LaF}_3:\text{Er}^{3+}$ and $\text{CaF}_2:\text{Er}^{3+}$ respectively.

The generator pulse width was 0.2 μs in case a, 0.4 μs in case b and 0.3 μs in c. The detector signal on the other hand extends over a range of about 10 μs . This large extension of the detector signal indicates a strong elastic scattering of the 285 GHz phonons increasing both the pulse propagation time and the pulse width. Such nonequilibrium phonon propagation determined by strong elastic scattering processes is called "diffusive" phonon propagation. The essential scattering mechanisms leading to diffusive phonon propagation are mass defect scattering and scattering processes at lattice imperfections [5].

4. The influence of strong elastic phonon scattering on phonon absorption measurements

In phonon absorption spectroscopy absorption structures are indicated by absorption and reemission of resonant phonons i.e. elastic scattering of the resonant phonons at the absorption centers. In the presence of a strong elastic scattering background the resonant scattering at absorption centers is a weak additional effect, hardly detectable. By reducing the sample thickness to the elastic scattering mean free path of the phonons the elastic scattering background can be reduced and the resonant phonon scattering at absorption centers becomes resolvable. In a first order approximation the mean free phonon path in the frequency range from 200 GHz to 1 THz can be estimated by the mean free path of 285 GHz phonons. This value can be determined by fitting the pulse measurements (Fig. 2) to an analytic expression for the detector signal based on the diffusive equations well

Fig. 2 Sn/Sn pulse measurements of a) $\text{SrF}_2:\text{Eu}^{2+}$
b) $\text{LaF}_3:\text{Er}^{3+}$
c) $\text{CaF}_2:\text{Er}^{3+}$

known from kinetic theory. The adaption values are shown in Fig. 2. The values obtained for λ (285 GHz) are

$$\begin{aligned} \lambda &= 1.8 \pm 0.6 \text{ mm} && \text{in SrF}_2:\text{Eu}^{2+} ; && \lambda = 0.7 - 1 \text{ mm} && \text{in LaF}_3:\text{Er}^{3+} \\ \lambda &= 0.2 - 0.4 \text{ mm} && \text{in CaF}_2:\text{Er}^{3+} \end{aligned}$$

5. Phonon spectroscopy measurements at thin samples ($d=\lambda$ (285 GHz))

By reducing the sample thickness to the value of mean free path of 285 GHz phonons the resonance structure at 21.2 cm^{-1} hardly detectable at the thick $\text{CaF}_2:\text{Er}^{3+}$ sample (see 2.) could now be well resolved (Fig.3 a). Fig.3 b shows the high resolution phonon absorption spectrum of this resonance ($\Delta\nu = 5.2 \text{ GHz}$). An additional signal modification can be seen at 30 cm^{-1} (b) with less intensity. This absorption can be ascribed to respective Er^{3+} crystalline field transitions already known from FIR-measurements (see 1.).

Fig.3 Phonon absorption spectrum of $\text{CaF}_2:\text{Er}^{3+}$; $d=300 \mu\text{m}$

Fig. 4
Phonon absorption spectrum of $\text{LaF}_3:\text{Er}^{3+}$
 $d = 1 \text{ mm}$

Phonon absorption measurements were also applied to appropriate thin $\text{LaF}_3:\text{Er}^{3+}$ samples. Fig. 4 shows the related spectrum to a $d=1 \text{ mm}$ thick sample.

At 14.2 cm^{-1} a deep absorption line was found not visible in FIR-measurements (a). This line so far could not be identified. For frequencies higher than 52 cm^{-1} a signal reduction is observed (b). This can be explained by the frequency limit of the TA-branches in the phonon dispersion relation of LaF_3 . Phonons with frequencies higher than this limit cannot propagate in the sample.

[1]	Ward/Clayman	J. Phys. C Solid State Phys. Vol.8, p 872 (1975)
[2]	Hadni/Strimer	Phys. Rev. B; 5,11, 4609 (1972)
[3]	Bron, Keilmann	Phys. Rev. B 12, 2: 2496 (1975)
[4]	Eisfeld, Renk	Int. Conf. in Phonon Scattering, Brown University
Providence 1979
[5]	Klemens	Proc. Phys. Soc. 68, 12A, 73 (1955)