Resonant Phonon Scattering by Calcium Colloids in Electron-Irradiated Calcium Fluoride

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1. Introduction

High frequency phonons with wavelengths in the nm-range should be scattered by mesoscopic defects in single crystals due to excitation of elastic resonances. The technique of thermal conductivity has been applied to investigate such an interaction in the case of Ag-colloids in NaCl [1] and γ-irradiated LiF [2]. Much better spectral resolution ($\sim$ 1 GHz) is possible by phonon spectroscopy with superconducting tunnelling junctions [3] whence more detailed information on the interaction with these defects is to be expected. Here we report on such an investigation of Ca-colloids produced by e^--irradiation in CaF_2. We find a good correlation of the observed phonon scattering (and its variation with thermal treatment) with a resonance peak in the optical Mie scattering [4].

2. Experimental results

Figure 1 shows the phonon transmission spectra above 285 GHz, the threshold of the Sn-junction, for several differently treated CaF_2 samples. Common for all samples is a scattering dip (3) near 750 GHz of unknown origin. After e^--irradiation at 120 K two additional dips appear at 398 GHz and 628 GHz which can be annealed by thermal treatment at 720 K. No additional structures were found in the frequency range between $\sim$ 100 GHz and 285 GHz. The appearance of dip (1) as well as its reduction with various annealing steps [Fig. 2] is correlated with an optical absorption line at 550 nm due to Mie scattering at Ca-colloids. A colloid radius of 6 nm is inferred from the position of this line [4]. A small shift of the minimum frequency with annealing temperature as visible in Fig. 2, however, has no observable correspondence in the optical scattering. The annealing behaviour of dip (2) is somewhat different: It deepens at 570 K to 670 K and vanishes also at 720 K. Only a small feature (4) at 377 GHz appears after low temperature e^--irradiation at 4.2 K. Additional Mie scattering is not observed in this case. γ-irradiation had no effect on the phonon spectra.

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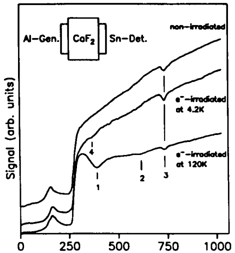

Fig. 1: Phonon transmission of electron irradiated CaF_2.

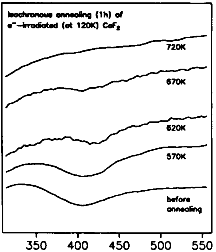

Fig. 2: Reduction of dip (1) at 398 GHz after isochronous (1 h) annealing.

3. Discussion

The correspondence between the annealing behaviour of dip (1) at 398 GHz and the optical absorption line due to Mie scattering at 550 nm led us to assume that dip (1) is due to phonon scattering at Ca-colloids. Evaluating the relative concentration of scatterers from the area below the optical absorption line and from the depth of dip (1) for every annealing temperature both resulting values agree within the error limits only if inelastic phonon scattering is supposed. To estimate the frequency dependence we applied an isotropic continuum model [5] for the total scattering cross section of a sphere for excitation of low lying resonances by transverse acoustic phonons. The resulting frequency pattern consisting of several nearly equidistant sharp peaks is smoothened by convolution with the typical size distribution [4] of the Ca colloids centered at 6 nm. A smooth and broad resonance peak near 400 GHz as experimentally observed is obtained only if a Q–factor as low as 5 is assumed for the individual resonances.

4. References

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