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Satellite Phonon Absorption Lines above the 875 GHz Resonance of Interstitial Oxygen in Silicon

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Acoustic phonon spectroscopy with superconducting tunneling junctions as phonon generator and detector revealed a large number of sharp absorption lines between 875 GHz (oxygen resonance) and 1.35 THz for silicon doped with interstitial oxygen (Si:O_i). The strength of these lines scales with the square of the oxygen concentration ranging from 10^{17} to 10^{18} cm^{-3}. Under mechanical stress the lines show a frequency shift almost identical to the main oxygen resonance at 875 GHz as well as to the also observable isotope resonance. These satellite absorption lines are therefore discussed as O_i-O_i neighbour interaction. This is supported by the influence of annealing.

1. Introduction

Acoustic phonon spectroscopy with superconducting thin film Al-I-Al tunnel junctions as phonon generator and Sn-I-Sn junctions as detector covers a frequency range from 280 GHz up to 3 THz with a maximum resolution of 2 GHz /1/. One of the many examples demonstrating the high frequency resolution and sensitivity of this spectroscopy is the phonon absorption /2/ of the 875 GHz (3.62 meV) line of Si:O_i (interstitial oxygen in Silicon single crystals) known from far infrared measurements /3/. This line corresponds to the transition between the lowest "rotational" states of the O_i between two Si atoms. By increasing the O_i concentration to 10^{18} cm^{-3} it was possible to observe also the $^{18}O_i$ isotope line /4/ at the natural concentration of $2\cdot10^{15}$ cm^{-3} indicating a phonon spectroscopic sensitivity at least two orders of magnitude higher than obtained with far infrared techniques. In these measurements also weak additional satellite lines /4/ predominantly in the frequency range from 875 GHz to 1.25 THz were observed as shown in Fig. 1, a more recent experimental result. Since these lines showed up in all samples of sufficiently high O_i concentration, neighbour-neighbour interactions appeared possible.

2. Satellite Phonon Absorption Lines in Si:O_i

Fig. 1 shows a phonon absorption measurement for a Silicon crystal of 1.3 mm length with a $^{16}O_i$ concentration of $1.3\cdot10^{18}$ cm^{-3}. The signal was obtained by a modulation technique /1/ revealing the line shape for the ^{18}O isotope as well as for the "satellites" marked by corresponding arrows. The ^{16}O absorption instead is experimentally saturated due to the much higher concentration resulting in a wide, almost rectangular frequency stopband. By the specific phonon spectrum emitted from the Al-generator /1/ this wide stopband causes the step-like signal decrease in the regime of the ^{16}O line. The natural line width of the ^{18}O isotope from our results amounts to $\Delta\nu = 4$ GHz. This line width is also observed for the ^{16}O isotope at sufficiently low concentration. Increased experimental resolution together with careful computer assisted noise reduction and data analysis revealed more details (see Fig. 2) of the satellite phonon absorption spectrum. The position and the relative strength of the satellite lines are independent of the O_i concentration. It should be noted that overlap is expected to be responsible for those lines which appear wider than the ^{18}O line.

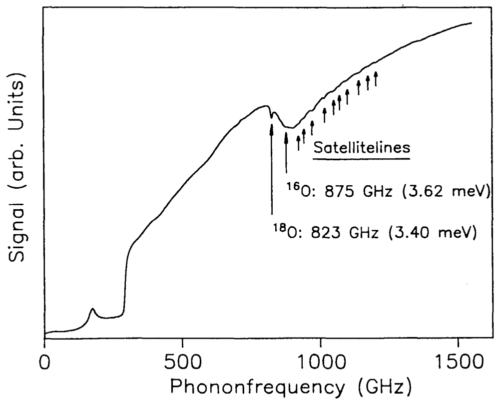

Fig. 1. Phonon absorption spectrum of Si:$^{16}O_i$ with resolved ^{18}O isotope line and satellite lines above 875 GHz.

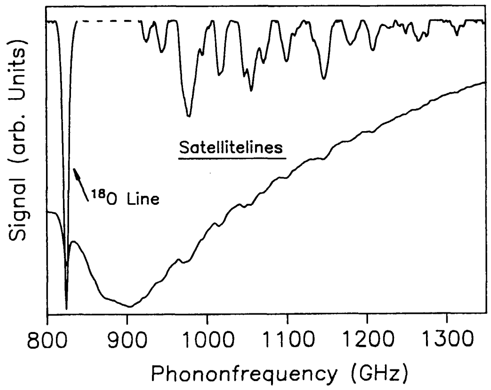

Fig. 2. Highly resolved satellite lines in acoustic phonon absorption above 875 GHz by computer analysis of the lower curve corresponding to Fig.1.

From the strength of the $^{18}O_i$ dip in Fig. 1 compared to the signal amplitude step of the $^{16}O_i$ isotope, we estimate a phonon absorption cross section of $\sigma_0 = 6\cdot10^{-15}$ cm^2. The directly determined natural line width is about 4 GHz for ^{18}O as well as for ^{16}O as checked by line width measurements for ^{16}O at lower concentrations.

3. Phonon Absorption by the Satellite Lines as Function of the O_i Concentration

Since the satellite lines are only observed at high O_i concentration of the order of 10^{18} cm^{-3} neighbour-neighbour interactions can be their possible origin. Assuming a statistical distribution of O_i atoms, the density of pairs must scale quadratically with the O_i concentration. The corresponding experimental result is presented in Fig. 3. For different O_i concentration ranging from $4 \cdot 10^{17}$ to $1.4 \cdot 10^{18}$cm^{-3} the phonon scattering (absorption) for the strong satellite line at 975 GHz has been quantitatively determined again using the $^{16}O_i$ absorption structure for calibration. The O_i concentration was measured by infrared absorption at 9 µm wave length. Fig. 3 shows the reciprocal mean free path obtained from the experimental absorption as function of the pair density parameter $(C_{O_i}/C_{Si})^2 \times C_{Si}$ indicating the statistical number of pairs per Si atom with C_{O_i} and C_{Si} the number of O_i and Si atoms respectively per volume. The scale on the right side of the diagram indicates the concentration $C_{O_{ii}}$ of pair scatterers at 975 GHz as obtained from the experimental phonon mean free path assuming the same scattering cross section as for O_i. The experimental result clearly agrees with a quadratic concentration dependence of the absorption (full line) within the measuring accuracy. Also the absolute absorption strength falls in the range of the estimated pair concentration with about 10^{14} pairs/cm^3. We can read from the right scale of Fig. 3 that each statistical pair at 975 GHz has a tenfold occupancy. This allows the conclusion that the 975 GHz satellite can be attributed to second or third nearest neighbour pairs depending on the number of equivalent neighbour sites with the same crystal anisotropy influence or the same symmetry of the positions.

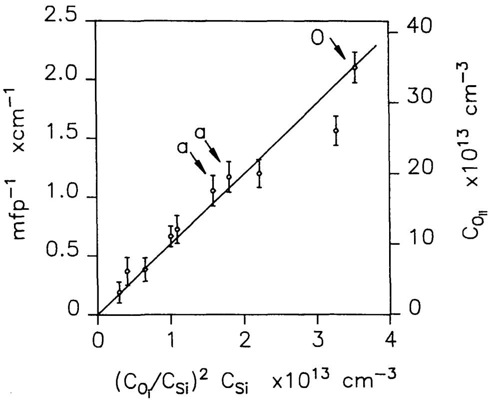

Fig. 3. Reciprocal phonon mean free path and O_{ii} pair concentration (right scale) for the 975 GHz satellite versus the square of O_i concentration. "a" indicates results for annealed samples with original values "o".

4. Influence of Uniaxial Stress

As shown in Fig. 4 all satellite lines are shifted by roughly 10 GHz by application of pressure in the /110/ direction. This shift is also observed for the ^{18}O isotope line. The higher resolution of the isotope line in addition reveals an unshifted contribution as to be expected for this stress direction. It is to be noted that the "dominant line shift" is negative with increased pressure.

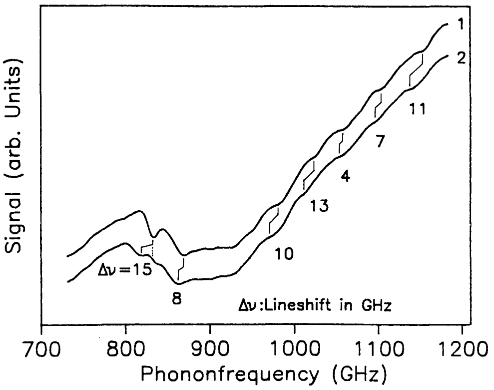

Fig. 4. Shift of the satellite lines by a static stress in /110/ direction.
1. zero stress, 2. p = 700 bar

5. Influence of annealing and of C-impurities

The results of Fig. 3 contain two experiments marked "a" in which the O_i concentration has been changed by annealing two samples of the original highest O_i concentration material corresponding to the measurement marked by "o". The annealed samples exhibit the same satellite line distribution with respect to frequency and relative absorption. Only The absolute absorption of the satellite lines is reduced in proportion to the square of the $^{16}O_i$ concentration. Apparently the statistics of the O_i distribution are not changed by annealing.

In order to check the influence of other impurities, measurements with high additional concentrations of carbon with $6 \cdot 10^{17}$ cm^{-3} and $2 \cdot 10^{15}$ cm^{-3} in the presence of $1.0 \cdot 10^{18}$ O_i have been performed. In both cases the carbon atoms had no influence on the oxygen lines as well as on the satellite lines.

6. Conclusion

The observation of O_i neighbour interactions possibly is of relevance for investigations of the growth dynamics of oxygen agglomerates in annealing procedures. The present results are an example of the high resolution and sensitivity of acoustic phonon spectroscopy.

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