# PHONON LINEWIDTHS OF NaF AT HIGH TEMPERATURES

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The linewidths of transverse acoustic modes in NaF are calculated for temperatures up to the melting point. We show that the widths decrease in a certain long wavelength region with increasing temperatures. The results are compared with recent neutron experiments up to 700 K.

The attenuation of an acoustic phonon with frequency $\Omega$ due to its interaction with thermal phonons is strongly dependent on the averaged inverse lifetime $\bar{\Gamma}$ of the thermal phonons in the crystal. In the region $\Omega \ll \bar{\Gamma}$ we have first sound and here Akhiezer's $\omega^2$-law is expected to hold. In the opposite limit $\Omega \gg \bar{\Gamma}$ the energy of the long wavelength phonon $\hbar\Omega$ is supposed to be large compared with the energy width $\hbar\bar{\Gamma}$ of the thermal modes. In this case energy and momentum conservation select the possible interactions between the sound wave and the thermal phonons in the crystal. This region is known as the zero sound or Landau–Rumer regime. The temperature dependence of the phonon widths has been found to be quite different in both regimes. At temperatures which are sufficiently high compared with the Debye temperature, the first sound damping is independent of temperature, while zero sound damping exhibits a linear temperature dependence [1,2].

In ref. [3] we have evaluated the elastic constants in the whole frequency region. Real and imaginary part were given by

$$C_{\alpha\beta\gamma\delta}(T,\Omega\boldsymbol{Q}) = C^{\mathrm{is}}_{\alpha\beta\gamma\delta}(T) + \frac{\hbar^2}{k_{\mathrm{B}}T}\frac{1}{2Nv_{\mathrm{a}}}$$

$$\sum_{\boldsymbol{q}j} \gamma^{jj}_{\alpha\beta}(\boldsymbol{q})\,\gamma^{jj}_{\gamma\delta}(\boldsymbol{q})\,\omega^2\begin{pmatrix}\boldsymbol{q}\\ j\end{pmatrix}\; n\begin{pmatrix}\boldsymbol{q}\\ j\end{pmatrix}\left[n\begin{pmatrix}\boldsymbol{q}\\ j\end{pmatrix}+1\right]$$

$$\times\left[\frac{\Omega}{\Omega-\boldsymbol{Q}\nabla_q\,\omega\binom{\boldsymbol{q}}{j}+2\mathrm{i}\bar{\Gamma}}+\frac{\Omega}{\Omega+\boldsymbol{Q}\nabla_q\,\omega\binom{\boldsymbol{q}}{j}+2\mathrm{i}\bar{\Gamma}}\right].$$

We have introduced the Grüneisentensor $\gamma^{jj}_{\alpha\beta}(\boldsymbol{q})$ of a phonon with wavevector $\boldsymbol{q}$ belonging to the branch $j$. The other symbols are selfexplaining; we use the same notation as in our previous publication. The above expression is exact for transverse branches; in the longitudinal branches, however, the difference between adiabatic and isothermal elastic constants is neglected. We now introduce the selfenergy matrix $\Pi_{\alpha\gamma}$ in the long wavelength region

$$\Pi_{\alpha\gamma}(T,\Omega\boldsymbol{Q}) = \frac{1}{\rho}\sum_{\beta\delta} C_{\alpha\beta\gamma\delta}(T,\Omega\boldsymbol{Q})Q_\alpha Q_\delta\ .$$

The inverse lifetime of the long wavelength phonons is then determined using the eigenvectors $\boldsymbol{e}\binom{\boldsymbol{Q}}{j}$

$$\Gamma\left(T,\Omega\begin{matrix}\boldsymbol{Q}\\ j\end{matrix}\right) = \frac{1}{2\Omega}\sum_{\alpha\gamma} e^*_\alpha\begin{pmatrix}\boldsymbol{Q}\\ j\end{pmatrix}\mathrm{Im}\left[\Pi_{\alpha\beta}(T,\Omega\boldsymbol{Q})\right]e_\gamma\begin{pmatrix}\boldsymbol{Q}\\ j\end{pmatrix}$$

The calculations for NaF are based on a breathing shell model for the phonon dispersion and on realistic microscopic Grüneisen parameter $\gamma^{jj}_{\alpha\beta}(\boldsymbol{q})$ for the third order coupling [3]. The averaged inverse lifetimes $\bar{\Gamma}$ were taken from our previous analysis [4] of the real part of the elastic constants up to 700 K, where we had been able to interpret the data from our inelastic neutron scattering experiments in the transition region between zero and first sound and determined averaged inverse lifetimes and isothermal elastic constants $C^{\mathrm{is}}(T)$

Table 1
Averaged inverse lifetimes $\bar{\Gamma}(T)$ of NaF as a function of temperature.

| temperature $T$(K) | 295 | 500 | 600 | 700 | 1000 | 1200 |
|---|---|---|---|---|---|---|
| inverse lifetime $\bar{\Gamma}(T)$ in rad THz | 1 | 3 | 5 | 7 | 13* | 17* |

* Linearly extrapolated.

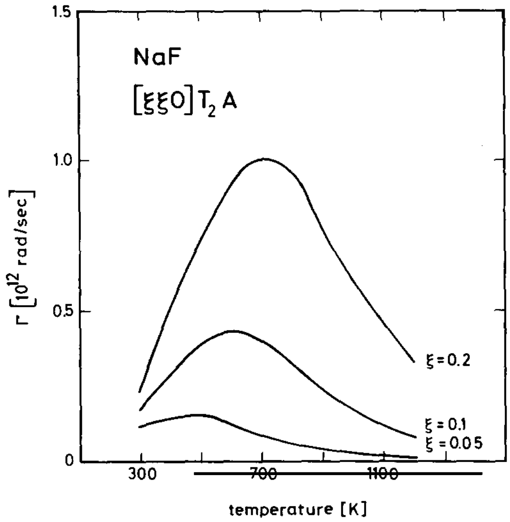

Fig. 1. Width Γ of $T_2A$ phonons in the [110] direction of the Brillouin zone in NaF.

for each temperature. Above 700 K $\bar{\Gamma}$ is assumed to have a linear temperature dependence. In table 1 the $\bar{\Gamma}$-values for the present calculation are listed.

The surprising features of the calculations become evident in fig. 1 presenting the temperature dependence of the widths with the reduced wave vector coordinate ξ as parameter. The theory predicts long wavelength phonons to exhibit increasing widths with rising temperature as long as they are zero sound phonons; however, in the region of first sound the situation is reversed. The phonon widths become smaller again even though the temperature approximates the melting point. This seems to be an important result which makes neutron scattering a tool for investigating (first sound) phonons in a temperature region near the melting point, where 'normal' phonons are almost damped out.

We are now in the position to interpret recent neutron scattering experiments in NaF [5] from applying our theory. In fig. 2 we show the change of the half widths at half maximum of $T_2A$ phonons with temperature. The theoretical calculations without any free parameter fit the experimental data satisfactorily well. Only at 500 K a larger deviation is observed which may be attribute to the errors in the difference of two nearly equal quantities. The advantage of such a plot is to get rid of the experimental resolution. Finally

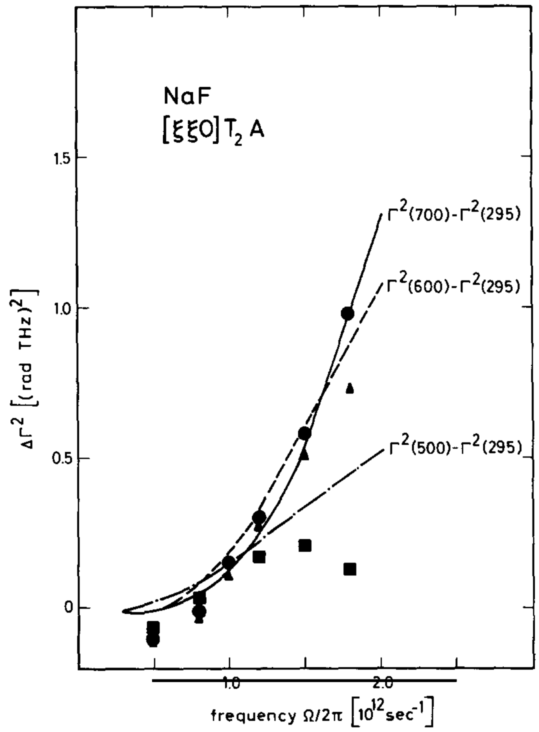

Fig. 2. Change of the half-widths at half-maximum of $T_2A$ phonons for different temperatures. The experimental data [5] refer to the following temperatures: ● 700 K–295 K, ▲ 600K–295K, ■ 500K–295K.

we may conclude that measurements in the first sound region could help to elucidate the following questions:

a) The phonon lifetimes in the first sound region are directly governed by the averaged inverse lifetimes. Its determination could clarify the question if $\bar{\Gamma}$ as determined from the damping of first sound noticeably differs from $\bar{\Gamma}$ that contributes to the heat flow [6], because different phonons are involved in the scattering process.

b) Although melting is usually considered as first order phase transition without critical phenomena, premelting effects e.g. an increased concentration of lattice defects are discussed [7]. It is at the moment a controversal point, whether the damping changes from Akhiezer's $\omega^2$-dependence due to pure first sound to a $\omega^{7/4}$-power law due to point defects [8,9].

c) For alkali halide crystals a direct connection between a soft shear mode [ξξo] $T_2A$ and the melting point has been proposed [10]. We have shown the possibility of observing well defined phonons even at very high temperatures which includes the possibility to study the softening of this branch as the melting point is approached.

Also we would like to comment on the very recent inelastic neutron scattering data of copper reaching from room temperature to the melting point of the crystal [11]. The authors reported an unexplained positive dispersion effect in their $|\gamma/\xi|$-data especially in the [ξξ0] $T_2A$ branch which we interpret as a first to zero sound transition. They also mention that no anomaly in the phonon widths was observed. Unfortunately, however, the reported widths do not cover the long wavelength region of first sound phonons which would be important to proof the predictions pointed out in this letter.

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