Zero to first sound transition in copper

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Abstract. Recent inelastic neutron scattering data in Cu (Larose and Brockhouse 1976) show an unexplained positive dispersion effect around the reduced wavevector $\xi = 0{\cdot}2$ in the T_1A branch with a sound velocity proportional to $(c_{11} - c_{12})^{1/2}$. In this letter this effect is explained in terms of a zero to first sound transition. The zero and first sound elastic constants, as well as the mean inverse lifetimes of the thermal phonons are derived for five temperatures from room temperature up to the melting point. Neutron scattering data in Ag and Au are also considered under these aspects.

In anharmonic crystals the elastic constants depend on the applied frequency. At low frequencies one measures first sound elastic constants, while in the high-frequency region zero sound elastic constants are observed (Cowley 1967). The transition from zero to first sound is expected at frequencies $\Omega \approx \bar{\Gamma}$, where $\bar{\Gamma}$ is an averaged inverse lifetime of the thermal phonons. The difference between zero and first sound increases with increasing temperature and, in addition, because of the temperature dependence of $\bar{\Gamma}$ itself, the transition will be shifted towards higher frequencies at higher temperatures. Frequency dependent elastic constants were derived by Niklasson (1972) and by Loidl *et al* (1976). We obtain for cubic crystals

$$c_{\alpha\beta\gamma\delta}(T, \Omega\boldsymbol{Q}) = c^{\mathrm{is}}_{\alpha\beta\gamma\delta}(T) + c^{6}_{\alpha\beta\gamma\delta}(T, \Omega) + c^{7}_{\alpha\beta\gamma\delta}(T, \Omega\boldsymbol{Q}) \tag{1}$$

with

$$c^{6}_{\alpha\beta\gamma\delta}(T, \Omega) = \frac{\hbar^2}{k_B T}\frac{1}{N v_a}\frac{\left\{\sum_{\boldsymbol{q}j}\gamma(\boldsymbol{q}_j)n(\boldsymbol{q}_j)[n(\boldsymbol{q}_j)+1]\omega^2(\boldsymbol{q}_j)\right\}^2}{\sum_{\boldsymbol{q}j} n(\boldsymbol{q}_j)[n(\boldsymbol{q}_j)+1]\omega^2(\boldsymbol{q}_j)} \times \left(1 - \frac{\Omega}{\Omega + 2\mathrm{i}\bar{\Gamma}}\right)\delta_{\alpha\beta}\delta_{\gamma\delta} \tag{2}$$

and

$$c^{7}_{\alpha\beta\gamma\delta}(T, \boldsymbol{Q}\Omega) = \frac{\hbar^2}{k_B T}\cdot\frac{1}{2N v_a}\sum_{\boldsymbol{q}j}\gamma^{jj}_{\alpha\beta}(\boldsymbol{q})\gamma^{jj}_{\gamma\delta}(\boldsymbol{q})n(\boldsymbol{q}_j)[n(\boldsymbol{q}_j)+1]$$
$$\times\ \omega^2(\boldsymbol{q}_j)\left(\frac{\Omega}{\Omega - \boldsymbol{Q}\cdot\nabla_q\omega(\boldsymbol{q}_j) + 2\mathrm{i}\bar{\Gamma}} + \frac{\Omega}{\Omega + \boldsymbol{Q}\cdot\nabla_q\omega(\boldsymbol{q}_j) + 2\mathrm{i}\bar{\Gamma}}\right) \tag{3}$$

$c^{\mathrm{is}}_{\alpha\beta\gamma\delta}(T)$ are the isothermal elastic constants. $c^{6}_{\alpha\beta\gamma\delta}(T, \Omega)$ denote the difference between isothermal and adiabatic elastic constants; the Kronecker deltas restrict its contribution

to c_{11} and c_{12}. $\gamma(\mathbf{q}_j)$ are the mode Grüneisen parameters and $\gamma^{ij}_{\alpha\beta}(\boldsymbol{q})$ are the elements of the Grüneisen tensor. The other symbols are self explanatory and used in the same notation as in the references above.

In copper we want to calculate the frequency dependence of the elastic constant $c'' = (c_{11} - c_{12})/2$, which determines the gradient of the dispersion curve in the $[0\xi\xi]T_1A$ branch. As transverse phonons do not couple to the local temperature we can neglect the contribution $c^6_{\alpha\beta\gamma\delta}(T, \Omega)$.

For a first evaluation we write equation (1) in the high-temperature limit as

$$c''(T, \Omega) = c''_{is}(T) + k_B T \delta^2 \frac{1}{v_a} \frac{\Omega}{\Omega + 2i\bar{\Gamma}}. \tag{4}$$

Here we introduced an averaged anharmonic coupling constant δ, which is related to the macroscopic Grüneisen parameter and ignored the term $\boldsymbol{Q} \cdot \nabla_q \omega(\mathbf{q}_j)$, assuming that the dominant contributions to the sum over the Brillouin zone come from short wavelength phonons. Expressions of similar forms were used by Niklasson and Sjölander (1974) and recently by Damien and Deprez (1976) analysing Brillouin scattering data in adamantane.

The frequency dependence of the real part of the elastic constant c'' is then determined by

$$\mathrm{Re}[c''(T, \Omega)] = c''_{is}(T) + k_B T \delta^2 \frac{1}{v_a} \frac{\Omega^2}{\Omega^2 + 4\bar{\Gamma}^2}. \tag{5}$$

The halfwidth at half maximum of phonons in the T_1A branch is obtained from

$$\Gamma(T, \Omega\boldsymbol{Q}) = \frac{1}{2\Omega} \mathrm{Im}[\pi(T, \Omega\boldsymbol{Q})] \tag{6}$$

with the self-energy

$$\pi(T, \Omega\boldsymbol{Q}) = \frac{1}{\rho} c''(T, \Omega) Q^2. \tag{7}$$

Using equation (5) we evaluated the wavevector dependence of the ratio $|\nu/\xi|$ for the five temperatures of the measurements in Cu (22, 400, 700, 920, 1063°C). From equation (6) and (7) we obtained the linewidths of the phonons in the T_1A branch at 22, 920 and 1063°C.

In this calculation we treated $c''_{is}(T)$, $\bar{\Gamma}(T)$ and δ as fit parameters. For the model to be consistent obviously c''_{is} should coincide with ultrasonic data, $\bar{\Gamma}$ should increase with increasing temperature and δ should remain constant over the whole temperature range.

Figure 1 shows the experimental data of the phase velocity compared with our theoretical results. The agreement between experiment and theory is satisfactory.

For comparison table 1 presents the values of the ultrasonic data (Chang and Himmel 1966) and the parameters used in our calculation: isothermal elastic constants, averaged inverse lifetimes and averaged anharmonic coupling constants. In the last column the calculated zero sound elastic constants in the high-frequency limit $\Omega \gg \bar{\Gamma}$ are listed. There is good agreement between the ultrasonic data and the isothermal elastic constants. The averaged inverse lifetimes of the thermal phonons increase with increasing temperature and also the anharmonic coupling

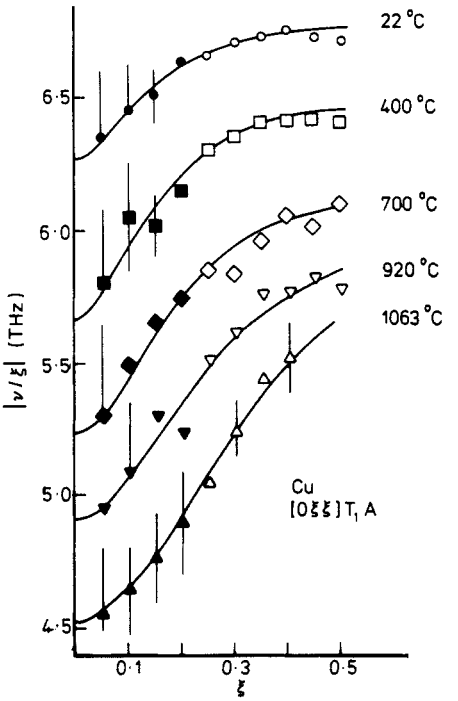

Figure 1. Temperature and wavevector dependence of the phonon phase velocity $|\nu/\xi|$ in Cu. The full symbols are mean values from energy loss and energy gain measurements, the open symbols represent neutron energy gain data only (Larose and Brockhouse 1976). Full curve: theory of this work.

remains almost constant. With the same set of parameters we calculated the halfwidths of the phonons in the T_1A branch. The results are presented in figure 2. The room temperature data as well as the experimental points at 920°C are well described, however, the theory overestimates the linewidths at 1063°C.

Starting from equation (3) we also made some refined calculations with a more precise resonance denominator, approximating

$$\boldsymbol{Q} \cdot \nabla_q \omega(\boldsymbol{q}_j) \approx \tilde{v} Q \cos(\boldsymbol{Q}\boldsymbol{q})$$

where the sum over the Brillouin zone was evaluated using an isotropic model with a mean sound velocity $\tilde{v}$. As a result, the values of the zero and first sound

Table 1. Ultrasonic data c''_{us} (Chang and Himmel 1966; † values are linearly extrapolated), parameters of the numerical calculations: isothermal elastic constants c''_{is}, averaged inverse lifetimes $\bar{\Gamma}$ and anharmonic coupling constants δ. The last column shows the calculated zero sound elastic constants c''_{zero}.

T (°C)	c''_{us} (10^{10}dyn cm^{-2})	c''_{is}	$\bar{\Gamma}$ (THz)	δ	c''_{zero} (10^{10}dyn cm^{-2})
22	23·6	23·0	0·4	3·3	26·8
400	19·7	18·5	0·45	2·8	24·5
700	16·7†	15·7	0·52	2·4	22·3
900	14·6†	13·8	0·65	2·4	21·3
1063	13·2†	11·8	0·8	2·6	21·8

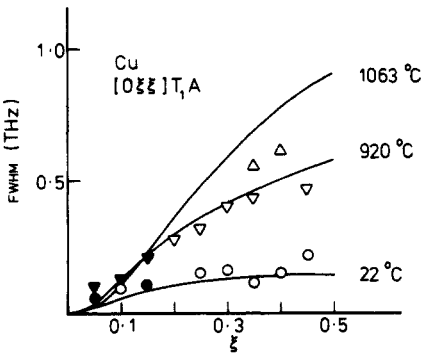

Figure 2. Temperature and wavevector dependence of the full width of phonons in Cu. Experimental data: Larose and Brockhouse (1976); the full symbols represent mean values from neutron energy loss and energy gain data, the open symbols are energy gain measurements only. Full curve: theory of this work.

elastic constants kept almost the same, but the values of the averaged inverse lifetimes $\bar{\Gamma}$ increased now from 0·6 THz at room temperature to 1·8 THz at 1063°C. This indicates, that the transition frequencies are shifted due to the phonon dispersion of the crystal. We conclude that realistic $\bar{\Gamma}$ values will be in between the results of the two pictures we used in our calculation.

It seems important to point out, that a pronounced transition effect between zero and first sound is only to be observed in the T_1A branch. In cubic crystals with FCC structure, at least demonstrated for some alkali halides (Cowley 1967, Loidl *et al* 1976), there is generally speaking a small renormalization effect of the order of 1% in c_{44}; the contributions to c_{11} and c_{12} are larger and positive in c_{11} while negative in c_{12}. Regarding the noble metals, where c_{12} is not much less than c_{11} we expect a very large percentage effect in the elastic constant $c'' = (c_{11} - c_{12})/2$. I recall, that the T_1A branch is rather soft compared with the frequencies of the other branches. As the averaged inverse lifetime of the thermal phonons is related to an averaged frequency, in this branch the transition region between zero and first sound is shifted far into the Brillouin zone.

The difference between the zero and first sound elastic constants c'' in Cu is of the order of 17% at room temperature. With the same anharmonic coupling and the above considerations the effects in c_{11} and $c' = (c_{11} + c_{12} + 2c_{44})/2$ turned out to be about 2%, which is far inside the error bars of the inelastic neutron scattering data. At 400°C, the highest temperature where both ultrasonic and neutron data are available, the difference between the zero and first sound elastic constants c_{11} should be of the order of 4%, neglecting the difference between adiabatic and isothermal elastic constants, which would reduce this value. For comparison, c_{11} from

Table 2. Elastic constants in the noble metals at room temperature derived from ultrasonic measurements (Chang and Himmel 1966). The units are 10^{10} dyn cm^{-2}.

	c_{11}	c_{44}	$(c_{11}+c_{12}+2c_{44})/2$	$(c_{11}-c_{12})/2$
Cu	170·0	75·8	222·0	23·55
Ag	124·0	46·5	155·5	15·25
Au	192·5	42·4	220·0	14·85

neutron scattering data is quoted with uncertainties of the order of $\pm 4\%$, while from ultrasonic data the relative uncertainties are $\pm 0{\cdot}7\%$.

Considering the other noble metals, the conditions mentioned above are nearly the same for Ag and even more pronounced for Au. This becomes evident from a comparison of the ultrasonic elastic constants (Chang and Himmel 1966) listed in table 2. Therefore the zero to first sound transition must be indicated in the neutron scattering data from the T_1A branches even at room temperature. In the case of Ag (Kamitakahara and Brockhouse 1969) this branch was not observed at a sufficiently fine mesh of wavevectors, nonetheless, already the measured points indicate a positive dispersion. In the case of Au (Lynn *et al* 1973) with a high density of experimental points, this branch clearly shows a positive dispersion effect at room temperature, which is even more pronounced than that found in Cu.

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