# Sound wave anomalies in superconducting compounds

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**Abstract.** Temperature dependence of the elastic constants in the superconducting state is described on the basis of the Ginsburg-Landau theory. This approach shows very general character and allows to describe quantitative renormalization of the elastic constants in the superconducting state for different materials including non-conventional superconductors. Various examples (A-15 compounds, Chevrel compounds, CeRu<sub>2</sub>, Ba<sub>0.63</sub>K<sub>0.37</sub>BiO<sub>3</sub>, Yb<sub>3</sub>Rh<sub>4</sub>Sn<sub>13</sub>, Ca<sub>3</sub>Rh<sub>4</sub>Sn<sub>13</sub>, HfV<sub>2</sub>, some heavy fermion compounds and Sr<sub>2</sub>RuO<sub>4</sub>) are given.

PACS. 74.25.Ld Mechanical and acoustical properties, elasticity, and ultrasonic attenuation

# **1** Introduction

Sound wave effects in superconducting elements like Sn, Pb, Nb etc are well documented. In superconducting compounds like A-15 material, Chevrel phase structures, stannides, high temperature superconductors or heavy fermion systems there is a rich variety of ultrasonic effects (see e.g. Ref. [1]). In most cases they have not been quantitatively described. Here we want to give a simple phenomenological description of these effects using the Ginzburg-Landau theory of superconductivity. In most cases of ultrasonic experiments in these compounds we are in the limit  $ql_e \ll 1$  (q wave number of sound wave,  $l_e$  electronic mean free path). Therefore we notice pronounced sound velocity effects and only in very rare cases typical sound attenuation effects due to contribution of conduction electrons.

In the next section we derive the necessary formulas for longitudinal and transverse sound velocities or elastic constants for the model in question. Our approach is similar to the one, first given by Testardi [2]. In the following chapter we apply derived formulas for fits to the various experimental results. In most cases these fits give a satisfactory agreement with the experimental data. We treat only the field free case B = 0 in this paper.

# 2 Calculation of the elastic constants: Ginzburg - Landau model

Following the Landau theory of the second order phase transitions we start with the order parameter expansion of the free energy density:

$$\Delta F = (a/2)|\eta|^2 + (b/4)|\eta|^4 + \dots$$
(1)

 $\Delta F = F - F_o$  is a change of the free energy in the superconducting state. Here we have to use for the complex superconducting order parameter (OP) the absolute value  $|\eta|$ . In the Ginsburg-Landau theory  $|\eta|^2$  is proportional to the density of Cooper pairs. In the case of unconventional superconductors the OP can have two or more components.  $dF/d|\eta| = 0$  gives the equilibrium value

$$\eta|^2 = -a/b = a_o(T_c - T)/b, \tag{2}$$

where we have used the Landau Ansatz  $a = a_o(T - T_c)$ . Here T is temperature and  $T_c$  is the temperature of the superconducting phase transition,  $a_0 > 0$  is a constant. Substituting the OP back into equation (1) gives for the free energy density

$$\Delta F = -a_o^2/4b(T - T_c)^2 = -\Phi_o(1 - T/T_c)^2.$$
(3)

This is valid for temperatures  $T < T_c$ . Here we have introduced  $\Phi_o = a_o^2 T_c^2/4b$  which we can interpret as the T = 0condensation energy density. For the following we assume that  $\Phi_o(\varepsilon)$  and  $T_c(\varepsilon)$  are strain dependent. Then we can calculate the elastic constants, using the symmetry strain  $\varepsilon_{\Gamma}$ , with  $c_{\Gamma} = d^2 F/d\varepsilon_{\Gamma}^2$ :

$$c_{\Gamma} = c_{\Gamma}^{o} - 2(\partial T_{c}/\partial \varepsilon_{\Gamma})^{2} \varPhi_{o} T/T_{c}^{3}(-2+3T/T_{c}) - 2(\partial^{2}T_{c}/\partial \varepsilon_{\Gamma}^{2}) \varPhi_{o} T/T_{c}^{2}(1-T/T_{c}) - 4(\partial T_{c}/\partial \varepsilon_{\Gamma})(\partial \varPhi_{o}/\partial \varepsilon_{\Gamma})T/T_{c}^{2}(1-T/T_{c}) - \partial^{2} \varPhi_{o}/\partial \varepsilon_{\Gamma}^{2}(1-T/T_{c})^{2}.$$
(4)

Here  $c_{\Gamma}^{o}$  is the background elastic constant,  $\Gamma$  is the symmetry label for the elastic constant and the corresponding strain. The four terms involving the different strain derivatives we discuss in the following.

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**Table 1.** s.c. transition temp.  $T_c$ , Sommerfeld parameter  $\gamma$  in (mJ/mol)K<sup>-2</sup>, mass density  $\rho$ , elastic constants  $c_{ij}$  at T = 200 K in  $10^{11}$  erg/cm<sup>3</sup>, coupling constants  $A_1$ ,  $A_2$ .

Material	$T_c$	$\gamma$	Density	$c_{11}$	$(c_{11} - c_{12})$	$c_{44}$	$A_1$	$A_2$	$ (\Delta c/c_0)_{OP} $	Acoustic
	Κ		$ m g/cm^3$		2		$\times 10^{-3}$	$\times 10^{-3}$		mode
$V_3Ge$	6.0	31	6.87	29.3	9.2	7.03	1	0.46		$c_{11} - c_{12}$
$V_3Si$	17.0	59	5.86	28.7	8.34	8.09				
$Nb_3Sn$	18.2	63	8.80	24.5	6.8	7.66				
PhMocSe	13.9	105	63	$0.20^{b}$	$10.75^{b}$		_	-6.5		T
1 0100008	10.2	100	0.0	0.20	10.75		_	-5.5		L
Euo «Sno «Mo«S»Bro 1	78		11.5	$2.18^{b}$	$3 \ 13^{b}$		_	-2.55		T
Ed0.8510.4100858E10.1	1.0		11.0	2.10	0.10			-4.5		Ĺ
CeBua	6.1	50	10.6	13.85	2.29	0.57	4	1.8		$C_{11} = C_{12}$
001042	0.1	00	1010	10.00		0.01	0.08	-0.015		C11 C12
Ba.63 K.37 BiO3	31						2.1	-0.075		$c_{11}$
$Yb_3Rh_4Sn_{13}$	6.5		8.9	12.62	2.93	2.05				
$Ca_3Rh_4Sn_{13}$	7.1		8.3	13.91	3.29	1.86	0.5	-4.5		$c_L$
$HfV_2$	9.0	58.1	9.3	12.8	1.3	1.7			$8.4 \times 10^{-5}$	CL
2	0.0								0.2	-1
UPt <sub>3</sub>	0.54	450	19.40	32.10	9.30	3.90			$5.6 \times 10^{-5}$	$c_{11}$
$URu_2Si_2$	1.40	120	10.01	25.51	10.40	13.34			$12 \times 10^{-5}$	$c_{11}$
$CeCu_2Si_2$	$0.50^{a}$	600	6.40	17.10	0.95	4.70			$5 \times 10^{-5}$	$c_{11}$
$\mathrm{Sr}_{2}\mathrm{RuO}_{4}$	1.42	38					0.55	0.25	$1.3 \times 10^{-4}$	$c_{11}$
							0.118	0.062	$8 \times 10^{-6}$	$c_{66}$
							0.0726	0.033	$5.5 \times 10^{-6}$	$c_{44}$
							1.54	0.7	$1.8 \times 10^{-4}$	$c_{11} - c_{12}$

<sup>a</sup>- $T_c$  without A-phase.

<sup>b</sup>-  $c_L$ ,  $c_T$ 

In order to illustrate the significance of the various terms in equation (4) one can expand the Landau free energy density [see Eq. (3)] further with the strain terms included. By expanding  $T_c(\varepsilon)$  and  $\Phi_o(\varepsilon)$ , in addition to the Landau free energy  $\Delta F_L$  of equation (1) the additional terms are obtained:

$$F_{sp} = g_{\Gamma} \varepsilon_{\Gamma} |\eta|_{\Gamma}^{2} + g_{\Gamma}' \varepsilon_{\Gamma} |\eta|_{\Gamma}^{4} + h_{\Gamma} \varepsilon_{\Gamma}^{2} |\eta|_{\Gamma}^{2} + h_{\Gamma}' \varepsilon_{\Gamma}^{2} |\eta|_{\Gamma}^{4} + \dots$$
(5)

Here we assume that these terms, linear and quadratic in the strain  $\varepsilon_{\Gamma}$ , exist for the symmetry  $\Gamma$ . The coupling constants g, g', h and h' can be related to the strain derivatives of  $T_c$  and  $\Phi_o$ . These relations are described in the Appendix.

For high symmetry directions shear waves do not couple in lowest order because  $\partial T_c/\partial \varepsilon_{sh} = \partial T_c/\partial (-\varepsilon_{sh}) = 0$ . This was noted by Pippard [3]. This no longer holds true in cases of a multidimensional order parameters as occurs for e.g. in heavy fermion compounds. Higher order terms involving  $\partial^2 T_c/\partial \varepsilon^2$  and other derivatives of  $\Phi_o$  in equation (4) are also possible for shear waves.

## 3 Fit-procedure of experimental results

We want to fit equation (4) to very different superconductors. They include A-15 compounds, Chevrel phase materials, stannides, CeRu<sub>2</sub>, HfV<sub>2</sub>, Ba<sub>1-x</sub>K<sub>x</sub>BiO<sub>3</sub>, Sr<sub>2</sub>RuO<sub>4</sub> and heavy fermions. The experimental results are taken from the literature. In Table 1 we collect relevant data for the different superconductors investigated. We write equation (4) in the form

$$\Delta c/c_o = -\left[2((\partial T_c/\partial\varepsilon)/T_c)^2 (\Phi_o/c_o)F_o(T) + A_1F_1(T) + A_2F_2(T)\right]$$
(6)

with  $F_o = (T/T_c)(-2 + 3T/T_c)$   $F_1 = T/T_c(1 - T/T_c)$  $F_2 = (1 - T/T_c)^2$ .

The first term in equation (6) is the order parameter coupling discussed in the Appendix with  $g = (a_o/2)\partial T_c/\partial \varepsilon$ . For the following two terms we get for the coefficients  $A_1$ ,  $A_2$  the expressions

$$c_o A_1 = 2\partial^2 T_c / \partial \varepsilon^2 \Phi_o / T_c + 4 / T_c \partial T_c / \partial \varepsilon \partial \Phi_o / \partial \varepsilon$$
$$A_2 = (\partial^2 \Phi_o / \partial \varepsilon^2) / c_o). \quad (7)$$

Note that  $A_1$  and  $A_2$  and also  $F_i$  are dimensionless. If the first term (with  $g_{\Gamma}$ ) of the strain-order parameter coupling expressions of equation (5) is negligible, the first term in equation (6) does not contribute. The term strain-order parameter coupling follows directly from the Ginzburg Landau treatment equation (5). It does not involve only  $dT_c/d\varepsilon$  but also higher terms  $(g'_{\Gamma} \text{ and } h'_{\Gamma})$ . Since  $A_1$  involves two terms and  $A_2$  also derivatives of  $\Phi_o$  it is best



**Fig. 1.**  $V_3$ Ge : Temperature dependence of the  $c_{11} - c_{12}$  mode near  $T_c$  [4]. Empty circles present measurements at B = 0, filled circles – measurements at B = 2.3 T. In the last case the sample stays in the normal state. Dashed line is a fit with equation (6) (see text for details).

to quote the  $A_{1,2}$  values. Since with this survey we would like to stimulate microscopic theories of these effects, we just quote the A-values and the experimental  $\Delta c/c_o$  values together with other physical quantities for the different superconductors in Table 1.

#### 3.1 A-15 compounds

These superconductors were analysed already by Testardi (see Ref. [2]). In this review a relation between a structural instability and superconductivity was elaborated, and corresponding behaviour of elastic constant in the superconducting state was studied. A-15 compounds have a cubic structure and had the highest critical temperature before the copper oxide high temperature superconductors were discovered. A characteristic feature of these compounds is a tetragonal structural instability. Relatively high superconducting transition temperature in these compounds has been related to the lattice anharmonicity because of this structural instability.

In the following we will discuss V<sub>3</sub>Ge for which elastic constant data exist for  $T < T_c$  [4]. In Figure 1 elastic constant data are shown for the  $(c_{11} - c_{12})/2$  mode. Clearly this mode exhibits a temperature dependence which can be interpreted with equations (4, 6). The fit gives the following values for  $A_1 = 10^{-3}$ ,  $A_2 = 0.46 \times 10^{-3}$ . In this case the experimental data exists only for a limited temperature range below  $T_c$ , from 4.5 K to 6 K. Therefore one cannot say whether the values of fit parameters are preserved at low temperatures.

Other A-15 compounds cannot easily be interpreted in this way because strong attenuation effects prevent measurements of symmetry modes for these materials. An example is Nb<sub>3</sub>Sn [5] where the assumption of a constant bulk modulus had to be invoked and no anomalies were seen at the superconducting phase transition for  $T < T_c$  presumably because of strong domain-wall stress effects due to the structural transition at  $T_a = 45$  K.



Fig. 2.  $c_L(T)$ ,  $c_T(T)$  and  $\rho(T)$  for PbMo<sub>6</sub>S<sub>8</sub> (a);  $c_L(T)$  and  $c_T(T)$  for Eu<sub>0.6</sub>Sn<sub>0.4</sub>Mo<sub>6</sub>S<sub>8</sub>Br<sub>0.1</sub> (b). Dashed lines give a fit with equation (6) with  $F_1 = 0$  for  $T \leq T_c$ . Experimental results from reference [8].

#### 3.2 Chevrel compounds

These are ternary molybdenum sulphide compounds with the general formula  $MMoS_8$  where M stands for a large number of metals. The superconducting properties are mainly determined by the band structure of the  $Mo_6S_8$ cluster. Single crystals of these materials are hardly available. Therefore most of the work was performed on sintered substances. Reviews on these compounds can be found in references [6,7].

In Figure 2a we show elastic constant data (longitudinal and transverse) for PbMo<sub>6</sub>S<sub>8</sub> [8]. The electrical resistivity is also given in the figure ( $T_c = 13.2$  K). A fit to the measured elastic constants with only the function  $F_2(T)$ reproduces the temperature dependence very well. In Figure 2b we give likewise the results for the other Chevrel compound Eu<sub>0.6</sub>Sn<sub>0.4</sub>Mo<sub>6</sub>S<sub>8</sub>Br<sub>0.1</sub> with  $T_c = 7.8$  K. Again the fit involving only  $F_2$  describes the experimental results (longitudinal and transverse) very well. The corresponding fit parameters are giving in Table 1.

Chevrel compounds exhibit in addition other pronounced acoustic effects: a softening of the elastic modes



**Fig. 3.**  $c_{11}$  versus T (a) and  $(c_{11} - c_{12})/2$  versus T (b) for CeRu<sub>2</sub>. Dashed lines are fits with equation (6) for  $T \leq T_c$ . Experimental results from reference [10].

for  $T > T_c$  and in magnetic fields for  $T < T_c$  anomalies in velocity and attenuation which can be interpreted quantitatively with a vortices-strain coupling [8].

# 3.3 CeRu<sub>2</sub>

This is a Laves phase compound with intriguing properties. It has a superconducting transition at  $T_c = 6.3$  K. It was investigated with acoustic measurements by different groups [9–11]. The normal state elastic properties exhibit a pronounced softening for the  $(c_{11}-c_{12})/2$  tetragonal mode. This is a precursor mode for a possible cubictetragonal phase transition for which the coupling constant is not quite large enough, however [10].

In Figure 3 we show data for the  $c_{11}$  and  $(c_{11} - c_{12})/2$ modes in the low temperature region  $T \leq 10$  K. For the longitudinal mode  $c_{11}$  we obtain a good fit with  $F_1$  and  $F_2$ . We can also interpret the temperature behaviour of  $c_{11} - c_{12}$  with our model. We get an equally good fit with  $F_1$ ,  $F_2$  (see dashed lines in Fig. 3).



**Fig. 4.**  $c_{11}(T)$  for Ba<sub>0.63</sub> K<sub>0.37</sub> BiO<sub>3</sub> [12]. Dashed line is a fit to equation (6) for  $T \leq T_c$ .

The magnetic field dependent behaviour of the elastic constants gives evidence for a pronounced peak effect. It was discussed with the so-called TAFF-model (thermally assisted flux flow) [10,11].

#### 3.4 Ba<sub>0.63</sub>K<sub>0.37</sub>BiO<sub>3</sub>

This high temperature copper-free superconductor has a superconducting transition at  $T_c \approx 32$  K. The potassium concentration 0.37 is close to the phase boundary of cubic – orthogonal phases. The unique properties of this material depend strongly on the dynamics of the BiO<sub>6</sub> octahedra.

The acoustical properties were investigated by Zherlitsyn et al. [12]. Due to some inhomogeneous distribution of potassium in the crystal the superconducting phase transition might have some finite width. For our sample an onset of superconductivity was at 31.7 K, and the width of the transition was approximately 6 K. We found a strong softening of more than 10% for the  $c_{11}$  and  $c_{44}$  modes in the normal state. Some hysteretic behaviour of these modes around  $T \sim 100$  K was interpreted with the anharmonic dynamics of the BiO<sub>6</sub> octahedra.

In Figure 4 we show the temperature dependence of the  $c_{11}$  mode in the vicinity of the superconducting phase transition. Below the superconducting transition we can fit the results adequately to equation (6) with the  $F_1$  and  $F_2$  terms of equation (6).

#### 3.5 Yb<sub>3</sub>Rh<sub>4</sub>Sn<sub>13</sub> and Ca<sub>3</sub>Rh<sub>4</sub>Sn<sub>13</sub>

These two ternary stannide compounds have transition temperatures of 6.5 K and 7.1 K respectively. They form a primitive cubic phase with two inequivalent sites for the Sn atoms:  $Sn(1)Yb_3Rh_4Sn(2)_{12}$  with Sn(1) forming a A-15 type sublattice. The superconducting properties of these materials have striking similarities with CeRu<sub>2</sub>.



**Fig. 5.**  $c_L$  mode for Yb<sub>3</sub>Rh<sub>4</sub>Sn<sub>13</sub> and Ca<sub>3</sub>Rh<sub>4</sub>Sn<sub>13</sub> [13], dashed lines are fits with equation (6) using  $F_1$  and  $F_2$  (see text for detailes).

This is not the case for the acoustical properties because the much smaller electron - lattice coupling renders the lattice effects much smaller than for CeRu<sub>2</sub>. This holds also for the ultrasonic properties in the normal state for the two classes of materials.

These two compounds were investigated acoustically by Haas et al. [13]. In the normal state the  $c_{44}$  mode shows softening of several percent from room temperature down to  $T_c$  for both compounds. The other modes,  $c_L$  and  $(c_{11} - c_{12})/2$ , exhibit much smaller temperature anomalies below 50 K. The various elastic modes, both longitudinal and transverse, exhibit pronounced anomalies at and below  $T_c$ . In Figure 5 we show the longitudinal elastic mode propagating in [110]  $c_L = (c_{11} + c_{12} + 2c_{44})/2$ for Ca<sub>3</sub>Rh<sub>4</sub>Sn<sub>13</sub> [13]. Note that in this case we present data down to 50 mK. We found a logarithmic temperature dependence for the longitudinal modes in the low temperature range T < 1 K (not shown in Fig. 5). Apart from this logarithmic temperature dependence the  $c_L$  mode for  $T < T_c$  can be well accounted for by  $F_1$  and  $F_2$ .

## 3.6 HfV<sub>2</sub>

This Laves phase compound, together with ZrV<sub>2</sub>, is an interesting superconductor because of the high electronic density of states at the Fermi energy, leading to a large specific heat with a  $T^3$  law at low temperatures. This classifies HfV<sub>2</sub> as a strong coupling superconductor with a high value of  $\Delta C/\gamma T_c \sim 1.9$ , compared to the BCS value of 1.43.

Elastic constant measurements on single crystals and polycrystals of HfV<sub>2</sub> showed a strong anomaly in the  $(c_{11} - c_{12})/2$  mode at the structural transition of  $T_a = 118$  K [14]. Additional X-ray investigation showed the transition being from cubic to orthorhombic.

At the superconducting transition  $T_c$  one observes a clear step function anomaly for the longitudinal mode  $c_L$  and no anomaly for the transverse mode  $(c_{11} - c_{12})/2$ .

This indicates an order parameter – strain coupling of the first term in equations (4, 6). No indications of the other higher order terms is noticeable for  $T < T_c$ . This behaviour is close to the behaviour of elemental superconductors or heavy fermion superconductors. Since for HfV<sub>2</sub> we are in the region  $ql \ll 1$  we cannot observe any sound attenuation due to conduction electrons, neither for  $T > T_c$  nor for  $T < T_c$ . In Figure 6a we show the stepfunction behaviour of the  $c_L$  mode at  $T_c$  together with the smooth temperature dependence of the  $(c_{11} - c_{12})/2$ mode.

#### 3.7 Heavy fermion compounds

In the heavy fermion compounds one has clear indications of strain – order parameter coupling. This is observed for both  $UPt_3$  and  $URu_2Si_2$  in the superconducting state [15,16] (see Fig. 6b, c).

**UPt<sub>3</sub>:** Especially for UPt<sub>3</sub> with the complicated phase diagrams including 3 phases one clearly observes for the longitudinal elastic constants step function like behaviour as a function of temperature and magnetic field corresponding to the first term of equations (4, 6) as shown in Figure 6b. But for UPt<sub>3</sub> we have a multi-component order parameter. Therefore the strain-order parameter interaction and the elastic constant expressions are more complicated than discussed above. One does not find any sign of additional contributions involving  $F_1$ ,  $F_2$ . The reason seems to be the large electronic Grüneisen parameter coupling observed in this case for both normal and superconducting states. Shear waves do not show any anomalies however.

**URu<sub>2</sub>Si<sub>2</sub>:** For URu<sub>2</sub>Si<sub>2</sub> one also observes for longitudinal waves only strain – order parameter coupling involving  $g_{\Gamma}$  as discussed above for equations (4, 6) (see Fig. 6c). In this case the *B*-*T* phase diagram consists only of one superconducting phase. Again a strong electronic Grüneisen parameter coupling seems to explain the effects adequately, especially for the  $c_{11}$  mode [17].

**CeCu<sub>2</sub>Si<sub>2</sub>:** This compound exhibits also distinct lattice properties. Usually, in stoichiometric compounds, the superconducting phase is enclosed by the so-called A phase and the strain – order parameter coupling [see Eq. (6)] can account for the A-phase expulsion and superconducting phase description [18]. For non-stoichiometric CeCu<sub>2</sub>Si<sub>2</sub> where no A phase is present one observes an ordinary step function behaviour indicating again strainorder parameter coupling of equation (6) [19] as shown in Figure 6d. In fact the temperature dependence of the  $c_{11}$  mode below  $T_c$  can be fitted quantitatively with the function  $F_o$  of equation (6). For magnetic fields  $B > B_{c2} \approx 1.7$  T the step function is suppressed.

## 3.8 Sr<sub>2</sub>RuO<sub>4</sub>

The ruthenate  $Sr_2RuO_4$  has the same layered perovskite structure of the  $K_2NiF_4$ -type as the cuprate superconductor  $La_{2-x}Sr_xCuO_4$ . This latter one is a *d*-wave superconductor. Like wise one argues that  $Sr_2RuO_4$  is also



**Fig. 6.** a)  $c_L$  and  $(c_{11} - c_{12})/2$  modes in the vicinity of  $T_c$  for HfV<sub>2</sub> (Ref. [14]); b) Temperature dependence of  $c_{11}$  and  $c_{33}$  modes for UPt<sub>3</sub> (Ref. [16]); c) Temperature dependence of  $c_{11}$  mode for URu<sub>2</sub>Si<sub>2</sub> (Ref. [17]); d)  $c_{11}(T)$  in CeCu<sub>2</sub>Si<sub>2</sub> for crystal without A-phase [19]. Full line is a guide to the eye.

an unconventional superconductor with a *p*-wave pairing symmetry and a transition temperature  $T_c = 1.42$  K [20].

Ultrasonic measurements in the superconducting state have been performed by several groups [21,22]. In addition the elastic constant tensor was determined with resonant ultrasonic spectroscopy RUS [23]. The temperature dependence of some elastic modes  $c_{11}$ ,  $(c_{11} - c_{12})/2$ ,  $c_{44}$ and  $c_{66}$  are shown in Figure 7 taken from the references listed above. In this case an order parameter-strain coupling was observed for the longitudinal mode  $c_{11}$  as for the heavy fermions and perhaps also for  $(c_{11} - c_{12})/2$ . In addition the other terms of equation (6) with  $F_1$  and  $F_2$ contribute strongly for all measured modes. The analysis of references [21,22] was made along the same lines as in this paper.

## 4 Discussion and conclusions

The results, presented in Figures 1–7 give a good description of the phenomenological expressions of equation (6). As fit parameters we used  $c_o$ , the background elastic con-

stant,  $(\partial T_c/\partial \varepsilon)^2 \Phi_o$  the coupling constant for the strain – order parameter coupling and the parameters  $A_1$  and  $A_2$ . In Table 1 we have listed these parameters  $A_1$  and  $A_2$ which are dimensionless. As a corresponding dimensionless quantity we also list  $(\Delta c/c_0)_{OP}$  which is the dimensionless step-function due to the order parameter-strain coupling, proportional to  $g_{\Gamma}^2$ , as discussed with equations (A1, A2) in the Appendix. In this way we can compare the different types of superconductors. The background elastic constant  $c_o$  is in most cases temperature independent for  $T < T_c$  since  $T_c$  is small compared to the Debye temperature. However in several investigated cases, the elastic constant  $c_{\Gamma}$ , shows pronounced softening for  $T > T_c$ . In these cases we assume that the softening is arrested at  $T_c$ . This can be investigated by applying magnetic fields  $B > B_{c2}$ . Such behaviour we found for the Chevrel compounds (Fig. 2a, b), for CeRu<sub>2</sub> (Fig. 3) and for  $Ca_3Rh_4Sn_{13}$  (Fig. 5). For the other compounds one could extrapolate into the  $T < T_c$  region, although for some cases the situation with the background elastic constants, in particular for T < 10 K, should be investigated by additional experiments.



Fig. 7. Temperature dependence of different acoustic modes  $c_{11}$  (a),  $c_{44}$  (b),  $c_{66}$  (c),  $c_{11} - c_{12}$  (d) in the vicinity of the superconducting transition in Sr<sub>2</sub>RuO<sub>4</sub> [21,22]. Dashed lines show a fit with equation (6) for  $T \leq T_c$  with parameters given in Table 1.

We see from the Figures 1-7 that only  $HfV_2$ , the heavy fermion compounds UPt3, URu2Si2, CeCu2Si2 and the layer compound Sr<sub>2</sub>RuO<sub>4</sub> exhibit strain-order parameter coupling effects involving the  $g_{\Gamma}$  term of equation (5), and only for longitudinal modes. The step-function at  $T_c$  for  $\Delta c/c$  varies between  $5 \times 10^{-5}$  and  $12 \times 10^{-5}$  (see Fig. 6). This can be related for the heavy fermions with the large density of states at the Fermi energy  $E_F$  for these materials. Taking as a measure the Sommerfeld  $\gamma$ -values given in Table 1, it is evident that these materials have an observable strain-order parameter coupling. For  $HfV_2$  the  $\gamma$  value is not so large, but it is considered a strong coupling superconductor. But also  $Sr_2RuO_4$  has a relatively small  $\gamma$  value. Apart from the step function behaviour the strain – order parameter coupling gives for  $T < T_c$  a temperature dependence expressed by  $F_o$  of equation (6), as observed for CeCu<sub>2</sub>Si<sub>2</sub> in Figure 6. It is interesting to note that shear waves may exhibit a strain - order parameter coupling [see Eq. (5)] also for multidimensional order parameters. This has been observed in Sr<sub>2</sub>RuO<sub>4</sub> for the transverse modes, especially for the  $(c_{11} - c_{12})$ mode [22, 24] (see also Fig. 7 and Tab. 1).

Higher order terms, involving  $\partial^2 T_c / \partial \varepsilon^2$  or the corresponding parameters  $A_1$ ,  $A_2$  are necessary for the fits in all discussed compounds except HfV<sub>2</sub> and the heavy fermions. From Table 1 we notice that  $A_2$  is practically the same (within a factor of 2-3) for all superconductors with the exception of Ba<sub>.63</sub>K<sub>.37</sub>BiO<sub>3</sub> and Sr<sub>2</sub>RuO<sub>4</sub>, whereas  $A_1$  is 0 for Chevrel compounds and very large for CeRu<sub>2</sub> and Ba<sub>0.63</sub> K<sub>0.37</sub> BiO<sub>3</sub>.  $A_1$  and  $A_2$  involve strain derivatives of both  $T_c$  and  $\Phi_o$  [see Eq. (7)]. The fits to the experimental results in Figures 1–7 shows that the strain derivatives of  $\Phi_o$  are also needed (see Tab. 1).

We have treated only the field free B = 0 case. For  $0 < B \leq B_{c2}$  pronounced dispersive effects are observed for both attenuation and velocity changes. This is especially pronounced for the Chevrel compounds and CeRu<sub>2</sub>. These effects can be interpreted quantitatively with the so-called TAFF model. For a discussion of this model see reference [25].

Furthermore we have treated only elastic constant effects, neglecting attenuation effects. For all superconducting compounds discussed above we are in the limit  $ql_e \ll 1$ 

with q the wave vector and  $l_e$  the electron mean free path. Therefore attenuation effects due to conduction electrons are completely negligible, in contrast to the elemental superconductors, with the exception of the  $(c_{11} - c_{12})$ -mode in Sr<sub>2</sub>RuO<sub>4</sub> [21,22]. Attenuation effects in the other compounds are only observable in the presence of magnetic fields as discussed before.

High temperature cuprate superconductors have not been dealt with in this paper. The reason is either the apparent lack of pronounced sound wave effects for the B = 0temperature dependence or for the La-Sr-CuO<sub>4</sub> the interference with a structural transition as can be seen from the magnetic field dependence (Ref. [26]). For a theoretical discussion of this class of superconductors, similar to the one given here, see reference [27].

## Appendix A

Here we give the relations between the terms of equation (4) and equation (5). For illustration we consider only  $T_c(\varepsilon) = T_c^0 + (\partial T_c/\partial \varepsilon)\varepsilon + (1/2) (\partial^2 T_c/\partial \varepsilon^2) \varepsilon^2$  and neglect the corresponding  $\partial \Phi/\partial \varepsilon$ ,  $\partial^2 \Phi/\partial \varepsilon^2$  terms.

From equation (5) with  $\Delta c_{\Gamma} = c_{\Gamma} - c_{\Gamma}^{0} = (d^{2}F_{sp}/d\varepsilon^{2})$ and with  $(d|\eta|/d\varepsilon) = -2g\eta/(a+3b|\eta|^{2})$  we obtain

$$\Delta c_{\Gamma} = -2g_{\Gamma}^2/b + 2h_{\Gamma}|\eta|^2 + 2h'_{\Gamma}|\eta^4 + 4g'g|\eta|^2/b \quad (A.1)$$

or introducing the specific heat  $\Delta C = T a_o^2/2b = 2\Phi_o T/T_c^2$ we obtain

$$\Delta c_{\Gamma} = -4g_{\Gamma}^2 \Delta C/(a_o^2 T) + 4h_{\Gamma} \Delta C(T_c - T)/(a_o T)$$
  
+higher order terms. (A.2)

The first term in equation (A2) is proportional to  $g_{\Gamma}^2$ and  $\Delta C/T$ . The first term of equation (A1) gives a step function for  $T \leq T_c$  of  $\Delta c_{\Gamma} = -2g_{\Gamma}^2/b$ . This we can identify with the first term of equation (4) at  $T_c$ :  $\Delta c_{\Gamma} = -2(\partial T_c/\partial \varepsilon)^2 \Phi_o/T_c^2$ . Inserting  $\Phi_0$  gives  $2g_{\Gamma} = a_o \partial T_c / \partial \varepsilon$ . Equations (A1, A2) predict a stepfunction (first term) and equation (4) gives a stepfunction at  $T_c$  with a temperature dependent term below  $T_c$ . This latter term arises from the h'-term quadratic in  $\varepsilon_{\Gamma}$  of equation (5). With this comparison we can check whether we have an OP coupling of the type of equation (A1). The second term of equations (A1, A2) gives  $\Delta c_{\Gamma} = 0$  at  $T_c$  and a temperature dependent term in T for  $T < T_c$ . Comparing with the second term of equation (4) we get  $h = a_o T_c [4(\partial T_c/\partial \varepsilon)^2/T_c^2]$  $- (\partial^2 T_c / \partial \varepsilon^2) / T_c ]/4$  and corresponding expressions for the constants g' and h'. In the following equation (A3) we

collect these expressions, where we have only considered  $T_c(\varepsilon)$ :

$$g_{\Gamma} = -(a_0/2)\partial T_c/\partial \varepsilon_{\Gamma} \qquad g'_{\Gamma} = (b/2T_c)\partial T_c/\partial \varepsilon_{\Gamma}$$
$$h_{\Gamma} = (a_o T_c/4)[4(\partial T_c/\partial \varepsilon_{\Gamma})^2/T_c^2 - \partial^2 T_c/\partial \varepsilon_{\Gamma}^2/T_c]$$
$$h'_{\Gamma} = (b/4)[-3(\partial T_c/\partial \varepsilon_{\Gamma})^2/T_c^2 + (\partial^2 T_c/\partial \varepsilon_{\Gamma}^2)/T_c]. \quad (A.3)$$

Similar expressions can be gained for the other terms involving derivatives of  $\Phi_o$ . We see that we can interpret the terms involving the strain derivatives of  $T_c$  and  $\Phi_o$  with this Landau strain expansion (Eq. (5)) quantitatively. A similar analysis was made using the two-fluid model of superconductivity [21,22].

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