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Elastic Properties of the Heavy Fermion Superconductor URu₂Si₂

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Measurements of the elastic constants in the normal and superconducting state on a high quality URu₂Si₂ single crystal exhibit large anomalies in the c_{11} mode versus temperature around 30 K and T_c . No such anomalies are visible in the c_{33} mode and in the shear elastic constants. We describe this behaviour with the anisotropic Grüneisen parameter coupling. Using the c_{11} anomaly a B-T phase diagram is determined which clearly shows the single phase bulk properties of superconducting URu₂Si₂. The magnetic field dependences of the shear waves in the superconducting and normal state can be interpreted quantitatively with a Lorentz force mechanism.

1. INTRODUCTION

In literature URu₂Si₂ is described as a heavy fermion superconductor with a $T_c \sim 1.2$ K and an antiferromagnetically ordered state with a $T_N \sim 17.5$ K.¹ The coefficient of the specific heat $\gamma(T=0$ K) has a value of 65 mJ/mol K^{2,2} which places URu₂Si₂ into the list of moderately heavy fermion compounds. This medium value of γ implies a rather high fluctuation temperature $T_f \sim 70$ K. The sublattice magnetic moment $\mu = 0.03 \pm 0.01 \mu_B$ is very small.³ Up until now there are no convincing measurements which point to a coupling between the superconducting order parameter and the sublattice magnetization. If such a coupling exists it would not lead to a splitting of T_c ,⁴ because of the spatial orientation of the spin density wave.

In previous work⁵ we have given results on elastic constants in the paramagnetic, antiferromagnetic and superconducting state both as a function of temperature and magnetic field. The interpretation of these results was difficult because of different superconducting phases present in the crystal (sample B) as discussed previously in the literature.⁶ But we were able to determine a B-T phase diagram which indicated two superconducting phases by elastic constant anomalies and ac-susceptibility. Ultrasound measurements are a powerful tool to determine phase transitions. They possess the great advantage of being insensitive to surface phases which is helpful in the case of URu₂Si₂.

In the present paper we give new results of elastic constant measurements in the temperature range from 0.06 K and magnetic fields up to 12 Tesla for a crystal of high quality which exhibits a sharp superconducting transition at 1.2 K (sample A). The sample is well characterised by resistivity-, ac-susceptibility and specific heat measurements. It is possible to combine the elastic constant which is a thermodynamic property with the thermal expansion and the specific heat to obtain the Grüneisen parameter. With the Grüneisen parameter coupling we are able to interpret the temperature and field dependence of the elastic modes in the paramagnetic and in the superconducting state.

2. NORMAL STATE PROPERTIES

We shall interpret our data with the Grüneisen parameter coupling. For heavy fermion systems this coupling is due to the heavy electrons and for the normal state it can be characterised by $\Omega_i = -\delta \ln T_f / \delta \ln \varepsilon_i$ (see e.g.^{7,8}). We give first the necessary formulae in the uniaxial case appropriate for the anisotropic heavy fermion system URu₂Si₂.

The Grüneisen parameter can be determined from the corresponding pressure dependence of the characteristic temperature T_f , T_N and T_c using Eq. 1 or from specific heat and thermal expansion with Eq. 2.⁷

$$\begin{aligned}\Omega_a &= -\frac{\delta \ln X}{\delta \varepsilon_a} = \left((c_{11} + c_{12}) \frac{\delta X}{\delta p_a} + c_{13} \frac{\delta X}{\delta p_c} \right) \left(\frac{1}{X} \right) \\ \Omega_c &= -\frac{\delta \ln X}{\delta \varepsilon_c} = \left(2c_{13} \frac{\delta X}{\delta p_a} + c_{33} \frac{\delta X}{\delta p_c} \right) \left(\frac{1}{X} \right)\end{aligned}\tag{1}$$

Here X denotes T_f , T_N or T_c respectively. ε_a and ε_c stand for the strains in a- and c-direction. Likewise we obtain⁷ with the molar volume V_m , the specific heat C_p and the linear thermal expansion coefficient α_i :

$$\begin{aligned}\Omega_a &= \frac{V_m}{C_P} ((c_{11} + c_{12}) \alpha_a + c_{13} \alpha_c) \\ \Omega_c &= \frac{V_m}{C_P} (2c_{13} \alpha_a + c_{33} \alpha_c)\end{aligned}\quad (2)$$

The volume Grüneisen parameter Ω_V is obtained from the uniaxial ones by:

$$\Omega_V = c_B [2\Omega_a(s_{11} + s_{12} + s_{13}) + \Omega_c(s_{33} + 2s_{13})] \quad (3)$$

Here $c_B = (2c_{11} + 2c_{12} + 4c_{13} + c_{33})/9$ is the bulk modulus and the s_{ij} are elements of the elastic compliance matrix. We use these formulae for the interpretation of our data.

Table 1 shows the elastic constants of a multiphase sample (B) and singlephase crystal (A) of URu₂Si₂ extrapolated to $T = 0$ K. The absolute value of the elastic constants in sample A are higher especially for the shear modes compared with the multiphase crystal B⁵ (see Table 1). We measured the c_{11} , c_{33} , c_{44} and c_{66} modes directly and the $1/2(c_{11} - c_{12})$ mode using a wedge arrangement. The c_{13} elastic constant was estimated by comparison with the isostructural compounds CeCu₂Si₂ and CeRu₂Si₂. The temperature dependence of the elastic constants deviates only minimally between sample A and B for the normal state properties. An overview of the temperature dependence of the elastic constants of crystal A is given in Fig. 1. In Fig. 1 there is a minimum in the longitudinal c_{11} mode clearly visible around 30 K while it is absent in the longitudinal c_{33} mode. This indicates the anisotropy of the physical properties in URu₂Si₂. Due to the Grüneisen parameter coupling no anomalous behaviour is visible in the volume conserving transverse modes c_{44} and c_{66} .

The anomalies near T_N are perceptible as kinks in the different $c_{ii}(T)$ -curves. They are of the same magnitude in relative units of $\Delta c/c_0$ compared to the ones of sample B in reference.⁵ Remarkable is that the changes of the elastic constants near T_N are roughly of the same order of magnitude as in UPd₂Al₃ which possesses a magnetic moment of 0.85 μ_B in the same

TABLE I

Elastic Constants for Different URu₂Si₂ Samples Extrapolated for $T = 0$ K ($\rho = 10.01$ g/cm³)

	c_{11} * 10 ¹¹ [erg/cm ³]	c_{33}	c_{44}	c_{66}	c_{12}	c_{13}
sample A	25.5	31.3	13.3	18.8	4.8	(8.6)
sample B	24.7	30.2	10.1	14.2

Fig. 1. Temperature dependence of the elastic constants c_{11} , c_{33} , c_{44} and c_{66} of sample A between 4 K and 250 K. [$\times 10^{11}$ erg/cm³].

temperature region. This points to a special kind of magnetic order in URu₂Si₂ as discussed in reference 9.

From the elastic constants, thermal expansion and pressure dependence of the characteristic temperatures T_f , T_N and T_c we can determine the corresponding Grüneisen parameters in the normal and superconducting state Ω^n , Ω^s . Because of the strong anisotropy of the tetragonal compound URu₂Si₂, the uniaxial Grüneisen parameters have to be used. With the values of the thermal expansion, specific heat and absolute elastic constants we obtain uniaxial Grüneisen parameters for sample A in the normal state for 1.5 K of $\Omega_a^n = +57.4$ and $\Omega_c^n = -15.1$. Clearly visible is the anisotropy between Ω_a^n and Ω_c^n . Using equation 3 one obtains for $\Omega_v^n = +41.1$ a value which is close to $\Omega_v^n = +43.2$ determined out of c_B , α_v and C_p at the same temperature. With the values of the pressure dependence of T_N $\delta T_N/\delta p_a = 126$ mK/kbar and $\delta T_N/\delta p_c = -41$ mK/kbar,¹¹ and the absolute elastic constants from table 1 we obtain magnetic Grüneisen parameters $\Omega_a^m = +19.3$ and $\Omega_c^m = +5.0$. From the temperature dependences of the thermal expansion and specific heat we get for $T = 18$ K ($T > T_N$) $\Omega_a^n = +25.5$ and $\Omega_c^n = +4.9$. These values are nearly the same as the magnetic Grüneisen parameters $\Omega_{a,c}^m$. The analysis above shows that these parameters exhibit considerable anisotropy in the temperature region 1 K-18 K. The low temperature Grüneisen parameters are listed in table 2. It should be stressed again that we deal here with electronic Grüneisen parameters, the lattice Grüneisen parameters being of order 1 are negligible.

Let us now discuss the anomaly in c_{11} near 30 K. In figure 2 we plot the temperature dependence of this mode for the region 4 K-80 K. From the estimates of $T_f \sim 70$ K given in the introduction we conclude that this anomaly is due to the Grüneisen parameter coupling. In a system with large Ω_v it is necessary to distinguish between isothermal and adiabatic sound propagation. For an adiabatic elastic constant the temperature dependence of a longitudinal mode is given by:⁷

$$c_s = c_0 + \Omega_{a,c}^2 * U \quad (4)$$

TABLE II

Low Temperature Grüneisen Parameter in the Normal and Superconducting State for Sample A

	$\Omega_a^{n,s}$	$\Omega_c^{n,s}$	$\Omega_v^{n,s}$
normal state ($T = 1.50$ K)	+57.4	-15.1	+41.3
superconducting state ($T = 1.07$ K)	-79.0	+16.9	-28.4

Fig. 2. c_{11} -mode at 140 MHz in the temperature range between 4-80 K together with calculated fits c_S (dotted line) and c_T (full line), (see text)

Here c_0 is the $T=0$ elastic constant, $\Omega_{a,c}$ the uniaxial Grüneisen parameter and U the internal energy. The dotted line in figure 2 shows the change in the $\Delta c/c_0$ -curve for adiabatic sound propagation with data for $\Omega_a(T)$ determined by equation 2 with thermal expansion from ref.¹³ and specific heat from ref.¹⁴. The internal energy was calculated by integrating the specific heat. It is seen that the experimental results for c_{11} are not describable with the adiabatic elastic constant as for Ce-compounds.⁷ Therefore we turn to the isothermal elastic constant c_T . It is given by

$$c_T = c_S - \Omega_{a,c}^2 * T * C \quad (5)$$

In figure 2 c_T is shown by the full curve. We use the temperature dependent uniaxial Grüneisen parameter Ω_a again. It is seen that the experimental points are closest to the c_T mode.

Now one has to make sure that the soundwave propagates in the isothermal regime. It is important to distinguish between the electronic thermal conductivity and contributions of non-electronic carriers especially phonons which exhibit a large value of κ_{ph} at low temperatures in URu₂Si₂.¹² The gaskinetic expression for the thermal conductivity of phonons is used:

$$\kappa_{ph} = \frac{1}{3} * v^2 * C_{ph} * \tau_{ph} \quad (6)$$

Here v denotes the sound velocity, C_{ph} the specific heat and τ_{ph} the relaxation time of the phonons. Together with our sound velocities in the tetragonal plane of 4040 m/s, the specific heat $C_{ph} = 26.02 \text{ J/m}^3 \text{ K}^{15}$ and the thermal conductivity $\kappa_{ph} = 0.78 \text{ W/K} * \text{m}^{12}$ (all values at 1.5 K) we obtain with equation 6 a value of $5.5 * 10^{-9} \text{ s}$ for τ_{ph} . The condition for the isothermal sound propagation is given by:¹⁶

$$\omega * \tau_{ph} \geq 1 \quad (7)$$

which is valid for the above τ_{ph} at all frequencies higher than 30 MHz. This indicates that the sound propagates isothermally in URu₂Si₂ for the frequency range of 30-150 MHz. Measurements at 10 MHz indicate a beginning of an isothermal-adiabatic crossover.

On the other hand, taking only the electronic contribution to the thermal conductivity one has to compare the sound velocity and the Fermi velocity.¹⁶ The condition for isothermal sound propagation is in this case:

$$\omega * \tau_c > \left(\frac{v}{v_F} \right)^2 \quad (8)$$

Taking appropriate values for the sound velocity v , the Fermi velocity v_F , the ultrasound frequency and the electronic relaxation time τ_c one does not

obtain any isothermal region for URu_2Si_2 . The conclusion from the first estimate is that sound propagates isothermally at least in the temperature region 1 K-10 K. The fit in figure 2 shows that this region extends up to 30 K.

It should be emphasized that possible crystal field effects on elastic constants can be excluded because the CEF strain susceptibility affects all modes giving especially a large value for c_{33} . However a pronounced softening is not visible in the measurements. Furthermore, a fit for the c_{11} mode does not agree with the measured value because the position of the minimum appears at 20 K instead of 27 K. This is in contrast to statements in the literature¹⁷ but agrees nicely with the postulated large fluctuation temperature $T_f \sim 70$ K, prohibiting the observation of CEF-effects.⁷

3. PROPERTIES IN THE SUPERCONDUCTING STATE

Crystal A exhibits in some points different results in the superconducting state than crystal B discussed previously.⁵ This has to do with the multi-phase properties in the latter crystal compared to the single phase material of the former. In Fig. 3 we show elastic constants as a function of temperature through the superconducting transition for various elastic modes. It is seen that c_{11} is the only mode which exhibits a steplike anomaly at T_c of the order of $\Delta c/c_0 \sim 10^{-4}$. All the other modes like c_{33} and the various shear modes show very small or no anomalies at all. This is in direct correspondence to the normal state properties discussed above where also c_{11} was the only mode to exhibit a temperature anomaly. So in URu_2Si_2 there exists a intimate connection between normal and superconducting Grüneisen parameters, a relation already observed in UPt_3 .⁵ Accordingly the electron-phonon coupling is the same in the normal and superconducting states. This coupling mechanism we shall now analyse.

Taking the uniaxial pressure dependence of T_c ,¹¹ the elastic constants and equation 1 we get for the uniaxial superconducting Grüneisen parameters $\Omega_a^s = -79.0$ and $\Omega_c^s = +16.9$. Similar values can be obtained from the step function of c_{11} with $\Delta c_{11} = \Omega_a^s \Delta C^s$ ($|\Omega_a^s| = 43.3$, $|\Omega_c^s| = 22.1$). The value of $|\Omega_c^s|$ was determined from the change in the sound velocity at B_{c2} . The superconducting Grüneisen parameters are listed in table 2. The strong anisotropy of the tetragonal system is obvious. Remarkable are the different signs between the superconducting and the normal state-Grüneisen parameter $\Omega_{a,c}^s$ and $\Omega_{a,c}^n$. This can be understood if one takes a BCS-expression for T_c in which the Debye temperature is exchanged by the characteristic temperature T_f :⁵

$$T_c = T_f * \exp^{-(1/NV)}$$

Fig. 3. Temperature dependence of the elastic constants c_{11} , c_{33} , c_{44} and c_{66} of sample A in the superconducting state. T_c is marked by the large anomaly in the c_{11} mode.

If the pressure dependence of the electron coupling potential can be neglected one obtains different signs of Ω^s and Ω^n for $T_c \ll T_f$.⁵

Figure 4 shows a plot of the *ac*-susceptibility together with the c_{11} anomaly versus temperature of sample A for different magnetic fields. χ_{ac} -measurements and the c_{11} mode exhibit the onset of superconductivity at nearly the same temperature. But there are differences in the width of the transitions. The elastic constants which probe the bulk superconductivity

Fig. 4. Anomalies in c_{11} and *ac*-susceptibility of sample A in the superconducting state as a function of magnetic field and temperature.

give a T_c of 1.07 K. T_c is defined by an equal area construction with the help of a step function.⁵ The same construction was done for the specific heat.¹⁰ From the shift of the curves with the magnetic field we can determine the B - T phase diagram which is given in Fig. 5 for $B//a$. Here we show the data of sample A together with sample B which is of lower quality.

There are two essential features visible in this phase diagram. One important point is that the anomalies in the c_{11} -mode occur for higher magnetic fields at the same temperature in sample A in comparison with crystal B. The B_{c2} -curve of sample A determined by the c_{11} anomaly (open triangles) and the one determined from the susceptibility measurements (closed triangles) are identical for magnetic fields greater than 1.5 Tesla. This could be interpreted as a sign for a single crystal with only one superconducting phase. Whereas sample B is of lower quality with at least two superconducting phases shown by the different B_{c2} -curves in Fig. 5 (open and closed circles). The second point is the difference between the magnetic field dependence of the elastic constant and the susceptibility in the temperature region near T_c . Here it is obvious that the curve determined by ac -susceptibility has a smaller initial slope ($\sim 4 \text{ T/K}$) compared to the one defined by of the c_{11} -anomaly which has a initial slope of $\sim 16 \text{ T/K}$ at T_c . This large value is an intrinsic property of the bulk superconductivity because measurements of the specific heat^{10,18} reveal the same results with

Fig. 5. B - T phasediagram for $B//a$ determined by the c_{11} anomaly and a susceptibility for two samples of different quality. Sample A (triangles) has only a vestige of a second superconducting phase at the surface. Sample B (dots) is a multiphase crystal.

Fig. 6. Elastic constant change and ultrasonic attenuation for the c_{11} -mode of sample A in the superconducting state. The attenuation exhibits a maximum at T_c .

a large slope of the B_{c2} -curve near T_c for $B//a$. For $B//c$ specific heat¹⁰ yields a slope of $\sim 4\text{--}5\text{ T/K}$ similar to the ac -susceptibility indicating the strong anisotropy of the tetragonal system.

In sample A there still remains a difference between the superconducting transition temperature T_c determined by χ_{ac} and ultrasound of $\sim 100\text{ mK}$ (Fig. 5). A similar difference was observed between resistivity and specific heat investigations by Hasselbach on another high quality sample.¹⁸ Because of the smaller initial slope and higher T_c obtained by χ_{ac} -measurements we suggest that a second superconducting phase exists at the surface of our sample which could be caused by the ageing of our crystal.

The positive curvature, i.e., the tail in the B - T phase diagram which is measured with ac -susceptibility and resistivity for $B//a$ (Fig. 5), is also indicated in specific heat and elastic constants but not so pronounced, because the width of the superconducting phase transition significantly broadens in a magnetic field. So together with the steeper slope of the B_{c2} -curve obtained by the elastic constants and specific heat experiments it is difficult to verify a positive curvature via these bulk measurements.

Fig. 6 gives data on ultrasonic attenuation together with the corresponding elastic constant anomaly for the c_{11} -mode. It is seen that the pronounced peak in the attenuation coincides with the middle of the elastic constant step indicating that the maximum of α occurs at T_c . Furthermore there is a rapid decrease of the attenuation below 1 K which is much stronger than in UPt_3 .^{19,20} The limited frequency range does not warrant however a detailed analysis of BCS attenuation versus power law attenuation. The transition in the superconducting state is connected with large anomalies in the thermal expansion²² but it is not clear whether they are responsible for the small minimum in α around 0.9 K or if there is a connection to the anomaly detected by μ_{SR}^+ at the same temperature for sample A.²¹

Shear waves do not give any effects as a function of temperature which is obvious because of the validity of the Grüneisen parameter coupling. However as a function of magnetic field clearcut anomalies are observed. For crystal B we interpreted⁵ the quadratic field dependence by the Alpher-Rubin effect.²³ Sample A exhibits similar results as crystal B. They are shown in fig. 7a for the c_{66} - and in Fig. 7b for the c_{44} -mode. In the superconducting state at temperatures $\leq 100\text{ mK}$ the $\Delta c/c_0$ exhibits a quadratic field dependence up to 7 Tesla. Then the elastic constant decreases until the critical field for this temperature is reached at ~ 11 Tesla. For higher temperatures this maximum is shifted to lower fields. So it is also possible to determine the B - T phase diagram shown in Fig. 5 out of such measurements. But here we will analyse our data with the Alpher-Rubin effect.

Fig. 7. Magnetic field dependence of the c_{66} -mode (9a) and c_{44} -mode (4b) in the superconducting state for different temperatures. Full lines are from calculations (see text).

The formula to describe the change of the sound velocity in a normal metal via this Lorentz force effect can be written:²⁴

$$\Delta v_t = \left(\frac{B^2 * \sin^2 \Theta}{8\pi * \rho * v_t^2} \right) * \left(\frac{1}{1 + \beta^2} \right) \quad (9)$$

$$\beta = \frac{c^2 * \omega}{4\pi * \sigma * v_t^2} = 1/2 * \lambda^2 * k^2$$

Here λ denotes a skin penetration depth. Because β depends upon the conductivity σ of the sample it is important to know the residual resistivity ρ_0 and the coefficients of the magnetoresistance $a_{a,c}$ to describe the change in the sound velocity. In a type II superconductor the magnetic field has an influence on the elastic constant via a Lorentz force effect which couples to the free electrons in the vortex core.

In the superconducting state it is possible to describe the motion of a vortex by the flux resistivity ρ_F ,²⁵ where ρ_F is proportional to the normal state resistivity.

$$\rho_F = \rho_n \frac{B}{B_{c2}(T)} * h(T, B) \quad (10)$$

$h(T, B)$ is an empirically determined function containing information about the thermal activation of the vortex lattice and crystallographic defects of the sample. $h(T, B) = 1$ for $T = 0$ K and less than one for $T < T_c$ and $B < B_{c2}$.

All heavy fermion systems possess an extremely large magnetoresistance at low temperatures so that one must include it in the formula for ρ_F . Accordingly ρ_F in a heavy fermion superconductor is given by:²⁶

$$\rho_F = \rho_n * \frac{B}{B_{c2}(T)} (1 + a_{a,c} * B^2) * h(T, B) \quad (11)$$

Knowing the residual resistivity ($11.5 \mu\Omega\text{cm}$), the magnetoresistivity and the absolute elastic constant we can describe the change of the transversal modes versus magnetic field for the normal state with Eq. 9 and for the superconducting state with Eq. 11. The coefficients of the magnetoresistance are determined by measurements of a small bar like piece of our single crystal. The magnetoresistance has a temperature dependence which is quite similar for samples of different quality. With the resistivity data we calculate the change of the sound velocity. The curves are also shown in Fig. 7a and 7b. We use $h(T, B) = 1$ for the calculations because we are not able to determine the fluxresistivity ρ_F .

From the β in Eq. 9 it is possible to determine the effective skin depth λ in the normal- and superconducting states.⁵ The temperature dependence of the effective skin depth in the superconducting state agrees qualitatively with the penetration depth determined by *ac*-susceptibility.¹⁰

4. CONCLUSION

This investigation of elastic properties of a single phase URu₂Si₂ crystal yields the following important results:

- The absence of anomalies for shear waves indicates a prevalent electronic Grüneisen parameter coupling in the normal state. However, with a Ω_V^n of 41.1 at 1.5 K URu₂Si₂ exhibits a Grüneisen parameter which is not so large as that of other heavy fermion system.⁷
- Of the longitudinal modes c_{11} exhibits much stronger anomalies than the c_{33} mode. So the strong anisotropy of the system is also visible in the uniaxial Grüneisen parameters. The large anomaly in c_{11} can be interpreted in terms of an isothermal-adiabatic crossover phenomenon.
- The anomalies in the elastic constants at T_N are very strong considering the small sublattice magnetic moment. This points to a new type of magnetic state, a spin nematic transition, as discussed in detail in reference.⁹
- In the superconducting state we observe clearcut strain-order parameter effects both as a function of temperature and magnetic field. The normal and superconducting uniaxial Grüneisen parameters are strongly correlated inasmuch as the c_{11} mode exhibits also in the superconducting state much stronger effects than the c_{33} mode.
- The B-T phase diagram exhibits a large initial slope for $B//a$. Due to the possible ageing of the sample there seems to be a vestige of a second superconducting phase at the surface of our sample visible in the *ac*-susceptibility.
- The magnetic field dependences of the shear waves in the superconducting and normal phases can be interpreted quantitatively with the Alpher-Rubin effect which is due to a Lorentz force mechanism.

Summarizing all the points listed above ultrasound measurements are a powerful tool to characterise the intrinsic properties of heavy fermion superconductors. Besides UPt₃, URu₂Si₂ represents a second example where the strain order parameter coupling has been studied in detail.

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