

AC-Calorimetry at High Pressure and Low Temperature

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Abstract. Recent developments of the AC-calorimetric technique adapted for the needs of high pressure experiments are discussed. A semi-quantitative measurement of the specific heat with a Bridgman-type of pressure cell as well as a diamond anvil cell is possible in the temperature range $0.1 \text{ K} < T < 10 \text{ K}$. The pressure transmitting medium used to ensure good pressure conditions determines to a great extent via its thermal conductivity the operating frequency and thus the accessible temperature range. Investigations with different pressure transmitting media for $T > 1.5 \text{ K}$ reveal for solid He a cut-off frequency which is considerably higher than for steatite. Experiments below 1 K and pressures above 10 GPa clearly show that the pressure dependence of the linear temperature coefficient of the specific heat can be measured. It is in qualitative agreement to a related quantity obtained quasi-simultaneously by electrical resistivity measurements on the same sample.

The specific heat (C) is an important thermodynamic quantity. Its temperature dependence can deliver hints about microscopic energy scales and provides a powerful tool to identify phase transitions. In this respect temperature (T) dependent measurements are an indispensable means not only for experimentalists. This has triggered the development of different and very sophisticated technical realizations to obtain $C(T)$ from the millikelvin range up to very high temperature. The available methods can be divided in two categories. Adiabatic techniques are considered as the most accurate way to estimate the absolute value of $C(T)$. They require sample masses of several grams and the subtraction of the addenda, i. e., the specific heat of sample holder and thermometer. Among the non-adiabatic (or dynamic) methods, AC-calorimetry is a suitable technique for samples with masses well below one milligram. The specific heat can be measured with very high sensitivity, despite the small masses. However, the absolute accuracy which can be achieved is less than for the adiabatic methods.

Adiabatic techniques are used to detect pressure-induced phase transitions or to investigate the evolution of electronic properties as the unit cell volume is reduced. The sample masses needed demand large volume pressure cells, such as a piston-cylinder cell. With this technique the accessible pressure range is, however, limited to about 3.5 GPa. Very often it would be desirable for the pressure range to be extended. In this case an anvil-type of pressure cell is the only alternative. Such a high pressure tool demands a

much smaller sample volume which makes an adiabatic measurement a hopeless venture. Thus, AC-calorimetry is an ideal method to be used for pressures beyond the limit of piston-cylinder cells.

1 AC-calorimetry adapted for high pressure

The general set-up of the AC-calorimetric technique for measuring the specific heat is sketched as a simplified model in Fig. 1(a). The sample is thermally excited by an oscillating heating power $P = P_0[1 + \cos(\omega t)]$, e.g. generated by a current of frequency $\omega/2$ through a resistance heater. The temperature oscillations at frequency ω are detected with a thermometer attached to the sample. Sullivan and Seidel [1] obtained a relation among the amplitude T_{ac} of the temperature oscillations and the specific heat C of the sample:

$$T_{ac} = \frac{P_0}{\omega C} \left\{ 1 + \frac{1}{\omega^2 \tau_1^2} + \omega^2 \tau_2^2 \right\}^{-1/2}. \quad (1)$$

This equation contains the time constants $\tau_1 = C/\kappa$ and τ_2 , with κ the thermal conductivity of the thermal link between sample and temperature bath (see Fig. 1(a)). It was derived in the ideal case, when the heat capacity of thermometer, heater, and heat link between sample and temperature bath are negligible and assuming a perfect coupling between heater, sample, and thermometer. The measured value of T_{ac} depends on the measuring frequency ω (Fig. 1(b)): At low frequency ($\omega \ll \omega_1 = \kappa/C$) the mean sample temperature is above the bath temperature T_0 by $T_{ac} \propto P_0/\kappa$. The recorded temperature oscillation T_{ac} yields the specific heat of the sample if the frequencies are in the range $\omega_1 \ll \omega \ll \omega_2 = 1/\tau_2$. The possibility of tuning both the amplitude and the frequency of the excitation is the main advantage of this method; as long as κ can be made small enough, the sensitivity of the measurement does not depend on the mass of the sample.

This technique was employed by several groups [2,3,4,5] to investigate the pressure dependence of the specific heat. The conditions for the AC-technique in a pressure cell are far away from being ideal. In particular the thermal properties of the pressure transmitting medium have to be taken into account. This was done by Baloga and Garland [3] for the case of high gas densities and low sample thermal conductivities. In their accessible temperature range (245 K $< T <$ 300 K) the general relation between T_{ac} and C for the AC-calorimetric expression (1) can be recovered if the product of specific heat and thermal conductivity of the pressure transmitting medium is negligible with respect to that of the sample. Then the heat wave does not propagate too far into the pressure transmitting medium and its specific heat does not contribute too much to T_{ac} . Typical frequencies are of the order of 1 Hz. Eichler and Gey [4] were the first to use the AC-technique for metallic samples in a piston-cylinder cell ($p_{max} \approx 3.5$ GPa) at low temperature (1.3 K $< T <$ 7 K). Here, the sample was embedded in diamond powder. It acts as pressure

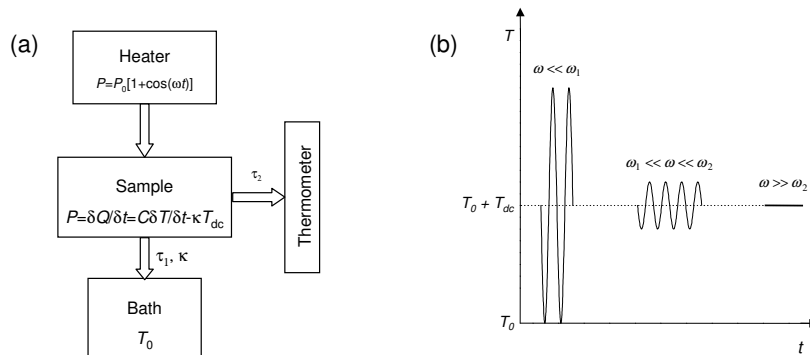


Fig. 1. (a) Sketch of a general AC-calorimetric assembly. The sample, thermal bath, thermometer, and heater are in contact by a thermal link with thermal conductivity κ . τ_1 is a measure of the thermal relaxation between sample and bath; τ_2 comprises the relaxation of thermometer, heater, and sample. (b) Sample temperature $T(t)$ for different frequency domains: For $\omega \ll \omega_1$, $T_{ac} \equiv T_{dc} \propto P_0/\kappa$ is not frequency dependent and is a measure of the thermal conductivity κ . In the range $\omega_1 \ll \omega \ll \omega_2$, the amplitude of the AC-part $T_{ac} \propto (\omega C)^{-1}$ depends on the measuring frequency and yields the specific heat of the sample. At $\omega \gg \omega_2$, T_{ac} is strongly reduced. Independent of the frequency, the mean sample temperature is $T = T_0 + T_{dc}$.

transmitting medium and provided the thermal resistance between the sample and the pressure cell. The measuring frequency was 120 Hz.

Pressures well above 3.5 GPa can only be achieved with opposed anvils, i.e., with a clamped Bridgman anvil technique or a diamond anvil cell (DAC). In Bridgman cells, the anvils are often made out of tungsten carbide (WC) or synthetic diamond and the pressure chamber consists of pyrophyllite (a sheet silicate, $\text{Al}_2\text{Si}_4\text{O}_{10}(\text{OH})_2$). The sample is in between two disks of e.g., the soft mineral steatite ($3\text{MgO} \cdot 4\text{SiO}_2 \cdot \text{H}_2\text{O}$) which acts as pressure transmitting medium. In a DAC a metallic gasket contains the sample and the pressure transmitting medium. Compared to a Bridgman cell a DAC comprises several advantages. First of all the pressure range can be extended easily to 50 GPa. Furthermore, the transparent anvils give optical access to the sample and the pressure can be determined with the ruby fluorescence method. Finally, the most important point is the possibility using He as pressure transmitting medium. With respect to hydrostatic pressure conditions, solidified He is an ideal medium since it is highly plastic and inert. However, these desirable features might mislead in underestimating the efforts in the elaborate assembly of the AC-calorimetric circuit in a DAC.

The feasibility of the AC-technique at pressures well above the limit of the piston-cylinder cells has to be tested, regardless of the type of high pressure cell. From the general principle of the AC-calorimetry (Fig. 1) it is evident that the main challenge are the unknown thermal properties of the pressure transmitting medium. To shed some light on this, the thermal conductivity

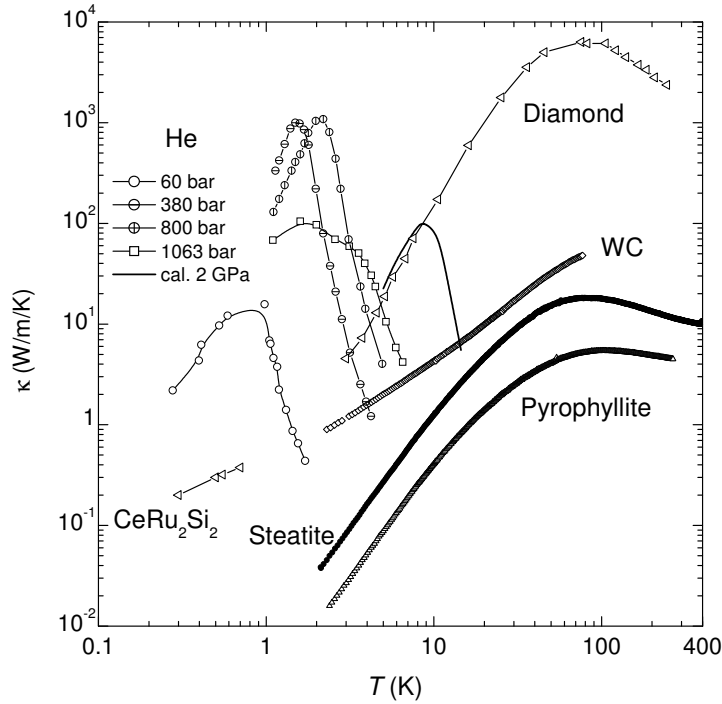


Fig. 2. Thermal conductivities $\kappa(T)$ of different materials used in high pressure devices with opposed anvils. WC and diamond are often used as anvils. Data for diamond (type Ib) are taken from [6]. Pyrophyllite and steatite serve as gasket and pressure transmitting medium, respectively. Due to its plasticity solid He permits homogeneous pressure conditions. The $\kappa(T)$ data for He are taken from [7] (60 bar) and [8] (380 bar and 800 bar). Unpurified He (1063 bar [8]) has a significantly different $\kappa(T)$. At the indicated pressures the crystals were grown. The solid line represents a calculated $\kappa(T)$ of He at 2 GPa (see text). $\kappa(T)$ of CeRu_2Si_2 [9] stands in for the thermal conductivity of heavy Fermion compounds at low temperature.

of the two preferred media, steatite and He will be discussed qualitatively in the following.

A priori it is not evident if the pressure media could satisfy the assumed requirements in the deduction of (1) because little is known about their thermal conductivity under pressure. To get an overview of $\kappa(T)$ of the materials used in a Bridgman device, the thermal conductivity of WC, pyrophyllite, and steatite at ambient pressure have been measured (Fig. 2). Literature data of diamond [6] and solid He at different pressure [7,8] are also depicted in Fig. 2. At low temperatures $\kappa(T)$ of steatite can be a factor 10^4 smaller than that of solid He at about 0.1 GPa. Moreover, the purity of He significantly affects the shape and size of $\kappa(T)$. For very pure He [10] the maximum thermal conductivity can be one order of magnitude higher than for unpu-

rified He at almost the same pressure [8]. Fortunately, it is very likely that the solidified He in the pressure chamber of a DAC is polycrystalline and contains impurities, brought in during the filling procedure. Therefore, $\kappa(T)$ might not reach the high values expected for pure He at the same pressure. As was pointed out in [8] the maximum value of $\kappa(T)$ occurs roughly at $\Theta_D/50$ (Θ_D : Debye temperature). Since He is highly compressible, Θ_D and thus, the maximum of $\kappa(T)$, rapidly increases with pressure. As a result the value of $\kappa(T)$ at, e.g. 1 K, could decrease considerably. In order to get a rough estimate of $\kappa(T)$ of He at several GPa, the following assumptions are made: (i) The low temperature slope remains unchanged at high pressure. (ii) The maximum value of $\kappa(T)$ at T_{max} does not increase with pressure. (iii) $T_{max} \approx \Theta_D/50$ can be estimated using $\Theta_D(V) = \Theta_D(V_0) \left[\frac{V}{V_0} \right]^\gamma$ with $\gamma = 2.4$ [11]. (iv) The maximum value of $\kappa(T)$ for He in a DAC might be comparable to that of impure He at about 0.1 GPa (Fig. 2). Based on the equation of state for solid He [12] the density for low temperature and 2 GPa is inferred to be about $V \approx 6 \text{ cm}^3/\text{mole}$. This density together with $\Theta_D(V_0) \approx 90 \text{ K}$ at $V_0 = 11.77 \text{ cm}^3/\text{mole}$ [11] yields $T_{max} \approx 9 \text{ K}$. Then $\kappa(T)$ at 2 GPa (line in Fig. 2) can be estimated with the assumptions specified above.

$\kappa(T)$ of steatite and He shown in Fig. 2 clarifies the differences in an AC-calorimetric experiment with these pressure media. The cut-off frequency in the case of steatite changes continuously since $\kappa(T)$ is a monotonic function below 10 K. Moreover, it is very likely that its shape will not be effected strongly by pressure. Thus, ω_1 at a given temperature should slightly vary with pressure. For steatite $\kappa(T) = aT^{2.3}$, with $a = 6.7 \times 10^{-3} \text{ W/m/K}^{3.3}$, is a good approximation of the data below 8 K. Together with heat capacity of a typical heavy Fermion compound like CeRu₂Ge₂ [13] or CePd₂Ge₂ [14] a cut-off frequency $\omega_1/(2\pi) \approx 100 \text{ Hz}$ at ambient pressure is calculated. $\kappa(T)$ of pressurized He, however, varies drastically with temperature and pressure. Comparing $\kappa(T)$ of He at 2 GPa and 4.2 K with that of steatite at ambient pressure shows that ω_1 will be roughly a factor 10 larger and of the order of several kHz. At these frequencies severe constraints are put on the homogeneity of the temperature in the sample. A homogeneous temperature distribution in the sample is given if the thermal wavelength $\lambda_{th} \propto \sqrt{\kappa/(C\omega)}$ is of the order of the sample thickness (typically about 30 μm). This condition is already fulfilled at about $\omega/(2\pi) \approx 1 \text{ kHz}$ for the compounds mentioned above. Nevertheless, these frequencies are well below ω_2 since metallic samples and thermometer ensure high thermal conductivity.

These considerations show that an AC-calorimetric measurement of metallic samples enclosed in solid He is more difficult for $1 \text{ K} < T < 10 \text{ K}$ as for the same sample embedded in steatite. Outside this temperature interval $\kappa(T)$ of He can be as small as that of steatite at ambient pressure and it might expected that T_{ac} is dominated by the specific heat of the sample. If the assumptions made above hold for even higher pressure, the cut-off frequency

could be reduced significantly and the AC-calorimetry in a DAC would become feasible in a larger temperature range.

2 AC-calorimetry in different pressure environments

The previous section illuminated the general aspects of the AC-calorimetry and contemplated the frequency domain in which experiments could be conducted. Two independent experiments [15,16,17] using the same compound but in different pressure devices and pressure transmitting media provide experimental information about the cut-off frequency. Both investigations explored pressures up to 8 GPa and temperatures in the range $1.5 \text{ K} < T < 10 \text{ K}$.

Figure 3 shows the pressure chamber of the Bridgman cell before closing the device. The typical thickness of the sample, thermocouple and heating wires are 20, 12, and $3 \mu\text{m}$, respectively. Two different ways of supplying the heat to the samples were tested. For sample A a thin electrical insulation ($4\text{--}5 \mu\text{m}$ of an epoxy/ Al_2O_3 mixture) prevents electrical contact with the heater but still established a good thermal contact. Sample B is set apart on a metallic (Pb) foil, electrically (and thus thermally) linked to the heater through a gold wire. No heating current passes through this sample. In the course of the experiment it turned out that the configuration A provided a homogeneous temperature distribution whereas the configuration B was ensuring hydrostatic pressure conditions.

The heating power was chosen in such a way that the temperature oscillations were in the range $2 \text{ mK} < T_{ac} < 20 \text{ mK}$. They were measured with a AuFe/Au thermocouple ($\text{Au} + 0.07 \text{ at\% Fe}$). The thermovoltage V_{ac} arises from the temperature difference between the sample (at $T_0 + \Delta T$) and the edge of the sample chamber (at T_0) [18]. The thermovoltage was amplified at room temperature in two stages and read by means of a lock-in detection referred to the frequency of the heating current. However, two potential drawbacks should not be concealed: (i) the temperature of the samples is measured with a thermocouple, under the assumption that the ambient pressure calibration holds at high pressure. (ii) The total amount of heat supplied to the samples is not known, despite the resistive heating. This prevents so far the acquisition of absolute values for the specific heat.

CeRu_2Ge_2 exhibits two magnetic phase transitions at ambient pressure leading to large features in the specific heat. Together with the well known influence of pressure on these transitions [13] this compound is a good candidate for testing AC-calorimetry at high pressure. Figure 4(a) shows the result of the AC-measurements at 0.7 GPa in comparison to the specific heat obtained by a relaxation method at ambient pressure. Pressure slightly shifted the transition temperatures as expected from the (T, p) phase diagram [13]. The height of the specific heat jump at the second order transition ($T_N \approx 9 \text{ K}$) represents 47% of the total signal compared to 51% for the ambient pressure curve. This indicates that T_{ac} is dominated by the heat capacity of the sam-

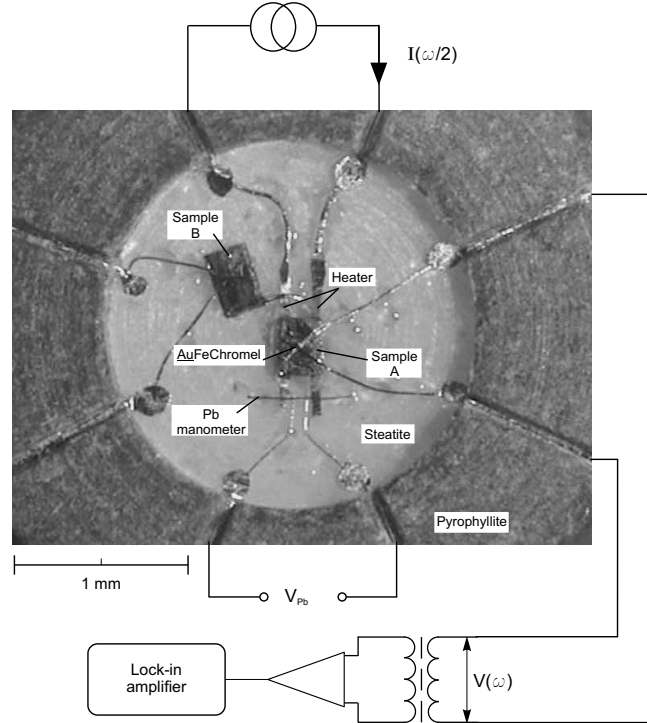


Fig. 3. Top view of the inner part of a Bridgman-type of pressure cell before closing. Two samples of CeRu_2Ge_2 are arranged for an AC-calorimetric experiment. Sample A is placed on top of the heater wires but is insulated from them. Sample B is in contact with a metallic foil and thermally linked to the heater through a Au-wire. The Chromel-AuFe thermocouples measure the sample temperature. The Pb-wire serves as pressure gauge. The entire assembly is mounted on a disk of steatite.

ple. An additional support for this statement is given by a frequency test. According to (1) the relation $T_{ac} \propto 1/\omega$ for $\omega \gg \omega_1$ should hold, which is indeed observed (inset Fig. 4(a)). A fit of a low pass filter to the data yields $\omega_1/(2\pi) = 450$ Hz. Frequency tests at various temperatures and pressures are a necessary task to determine ω_1 and to ensure the validity of the relation between T_{ac} and the specific heat of the sample. The height of the first order transition ($T_C \approx 7$ K) is very sensitive to any distribution of T_C and should not be compared to the peak in $C_p(T)$ at ambient pressure. Moreover, AC-calorimetry is not the proper tool to measure a latent heat [19] since it only detects the reversible part at frequency ω on a temperature scale T_{ac} . Nevertheless, the position of a first order transition can be detected by an AC-calorimetric measurement.

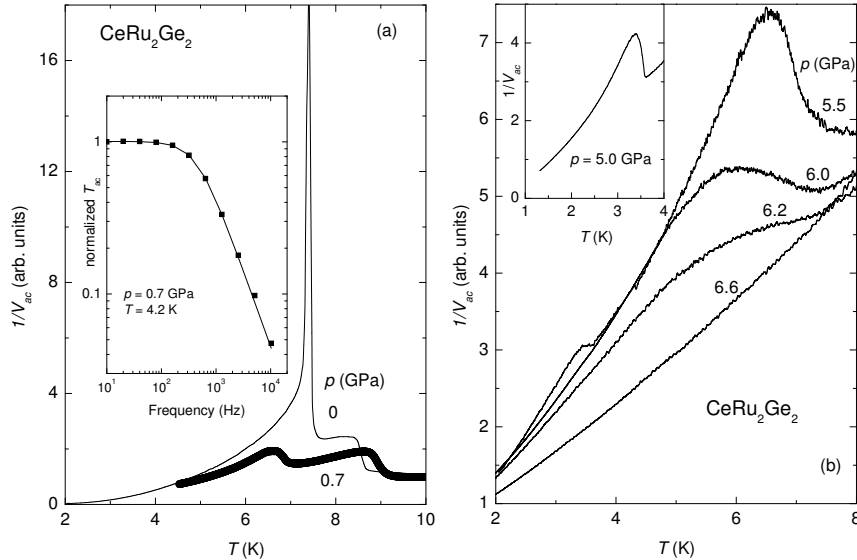


Fig. 4. (a) Comparison of the inverse of the lock-in signal, $1/V_{ac} \propto C$, of CeRu_2Ge_2 enclosed in steatite at 0.7 GPa and the specific heat C/T measured with a relaxation method at ambient pressure [13]. The data sets are normalized at 10 K. A frequency test at 4.2 K is depicted in the inset ($\omega_1 \approx 450$ Hz). (b) Temperature dependence of $1/V_{ac} \propto C$ of CeRu_2Ge_2 above 5 GPa. The pronounced feature related to the antiferromagnetic transition is suppressed by pressure. The inset shows the specific heat at 5.0 GPa with an anomaly at low temperature. The feature can still be seen near 3.5 K at 5.5 GPa (main figure).

The AC-calorimetry data of CeRu_2Ge_2 above 5 GPa shown in Fig. 4(b) demonstrate the potential of this method. The influence of pressure on the antiferromagnetic transition is visible and the deduced $T_N(p)$ data agree with the (T, p) phase diagram extracted from transport measurements [13]. A critical pressure $p_c \approx 7$ GPa is necessary to suppress the long-range magnetic order. The broadening of the antiferromagnetic transition is very likely related to intrinsic effects although a small pressure inhomogeneity could be partly responsible for it. In addition to this transition an anomaly at lower temperature was resolved (inset of Fig. 4(b)). These measurements were the first to show that this anomaly seen so far only by transport measurements [13], has thermodynamic origin and is a bulk property.

Working with a DAC allowed Demuer and coworkers [17] to use a different way to supply the oscillating heat power to the sample. They attached an optical fiber to the DAC and heated the sample with the light of an Ar-ion laser. It was chopped mechanically at frequencies up to 3 kHz. The temperature oscillations of a Au-Chromel thermocouple bonded directly on the sample by spark welding were measured with a lock-in amplifier. In this

experiment CeRu_2Ge_2 was enclosed with solidified He. The cut-off frequency was estimated to 4 kHz at 0.5 GPa and 7 K. This value supports the estimated order of magnitude for ω_1 in the case of pressurized He given in Sec. 1. The high thermal conductivity of He limits the application of the AC-method at low pressures. Nevertheless, the magnetic phase transitions could be observed although only a part of the signal at the fixed measuring frequency of 1.5 kHz was due to the specific heat of the sample. In this investigation an increased width of the transition was also established. Intrinsic effects seem to be responsible for this if the good hydrostatic pressure conditions in the experiment are kept in mind. In addition, a similar broadening in specific heat experiments at ambient and low pressure have been reported [20,21] when T_N is pushed to zero temperature either by doping or pressure.

The analysis of the thermal conductivity data in Sec. 1 suggests that He could be used as a pressure transmitting medium at low temperature even at pressures of a few GPa. This presumption is corroborated by the results of Holmes and coworkers [22]. With a combined measurement of electrical resistivity and AC-calorimetry the heavy Fermion superconductor CeCu_2Si_2 was investigated down to 0.1 K for pressures up to 7 GPa. The jump in the AC-signal caused by the entrance into the superconducting state provided a semi-quantitative measure of the sample specific heat. The onset of the specific heat occurred when the resistive transition was completed and affirms the bulk property of the superconducting state.

3 AC-calorimetry below 1 K and beyond 10 GPa

A demonstration of the feasibility of the AC-technique below 1 K and pressures well above 10 GPa is the experiment on $\text{CePd}_{2.02}\text{Ge}_{1.98}$ in a Bridgman-type of pressure cell [14]. The set-up of the experiment was chosen in such a way that electrical resistivity and AC-calorimetry could be performed on the same crystal. This makes it possible to check whether an anomaly in T_{ac} is related to the sample or not with an independent electrical resistivity measurement. Figure 5 shows the arrangement in the pressure chamber. It contains two different samples of the solid-solution $\text{CePd}_{2+x}\text{Ge}_{2-x}$, but only one of them ($x = 0.02$) was connected for the AC-experiment. The sample was heated with a current supplied through Au-wires attached to the sample. This reduces the components in the pressure chamber and avoids a pressure gradient due to the heat wires. With this arrangement it is also possible to calibrate the AuFe/Au thermocouple up to very high pressure and over a wide temperature range [14]. It was observed that the absolute thermopower $S(T)$ of AuFe at 4.2 K and 1.0 K at 12 GPa is about 20% smaller than the values at ambient pressure. These rather small changes show that the results are not affected qualitatively if the ambient pressure values of $S(T)$ are used. Thus, the drawback of a missing temperature calibration for the thermocouple mentioned in Sec. 2 could be in principle eliminated. The reliability of

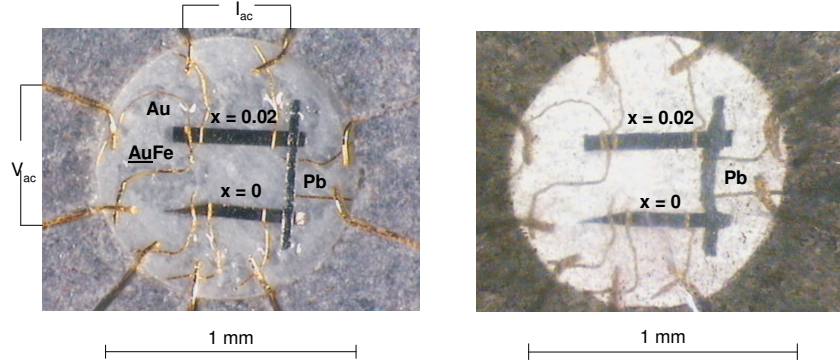


Fig. 5. Left: Pressure cell before closing with two samples of $\text{CePd}_{2+x}\text{Ge}_{2-x}$ ($x = 0$ and 0.02) and the pressure gauge (Pb-foil). The AC-calorimetric circuit is mounted on one sample ($x = 0.02$). The temperature oscillations are read with an additional AuFe wire when an AC-heating current is applied. Right: The pressure cell after pressure release from 22 GPa. The almost unchanged configuration shows the reliability of the pressure device.

the pressure cell is obvious as can be seen in Fig. 5. After the pressure was released from 22 GPa the overall shape of the pressure cell as well as its initial diameter were almost unchanged and the distance between the voltage leads increased by less than 5%.

$\text{CePd}_{2.02}\text{Ge}_{1.98}$ was chosen because in its stoichiometric form it is the Ge-doped counterpart of the antiferromagnetically ordered heavy Fermion compound CePd_2Si_2 ($T_N = 10$ K). The latter system enters a superconducting ground state when the magnetic order is suppressed ($p_c = 2.7$ GPa) [23]. Applying pressure to $\text{CePd}_{2.02}\text{Ge}_{1.98}$ ($T_N = 5.16$ K [14]) should increase T_N to a maximum and then it should approach zero temperature. The aim of the AC-calorimetric measurement was to extract the electronic contribution to the specific heat. Figure 6 shows the inverse of the registered lock-in signal V_{ac} below 10 K at various pressures. The pronounced anomaly in $1/V_{ac}(T)$ for pressures between 6.0 GPa and 10 GPa is caused by the entrance into the antiferromagnetically ordered phase. The height of the anomaly decreases and it becomes a very broad feature as the system approaches $p_c = 11.0$ GPa [14]. A similar broadening upon approaching the critical pressure was reported for CePd_2Si_2 , despite the lower pressure and the use of He as pressure transmitting medium [24]. Recalling the increased transition width reported in Sec. 2 it is very likely that this is an intrinsic phenomenon. Two frequency tests at $p = 8.9$ GPa are shown in the inset of Fig. 6. A fit of a low pass filter to the data yield cut-off frequencies of $\omega_1/(2\pi) = 350$ Hz and 1060 Hz for $T = 4.2$ K and 9 K, respectively. Assuming the validity of $\omega_1 = \kappa/C$, these values and the $1/V_{ac}$ data at the corresponding temperatures result in $\kappa(4.2 \text{ K})/\kappa(9 \text{ K}) \approx 0.2$, almost the same ratio as at ambient pressure (see

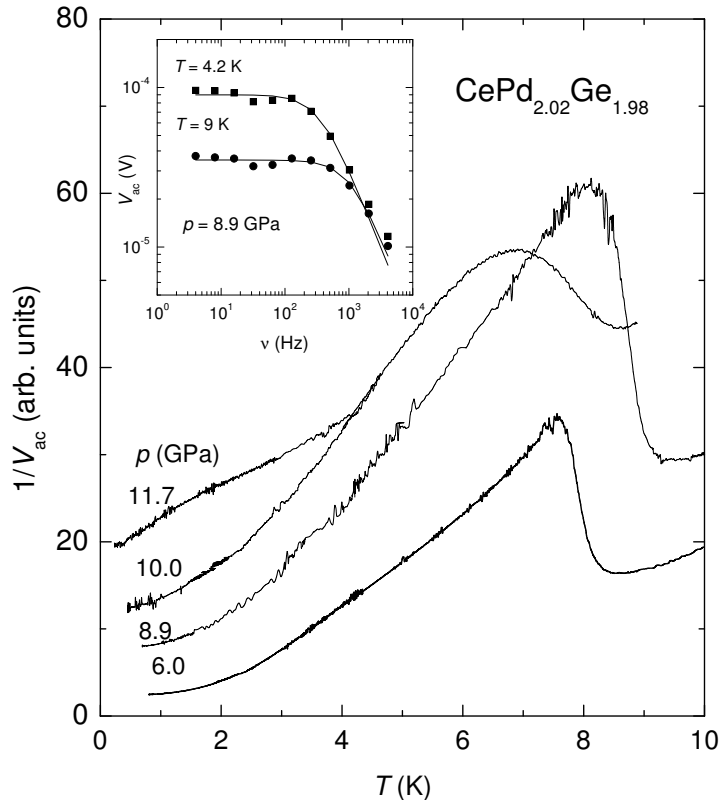


Fig. 6. Temperature dependence of the inverse lock-in voltage V_{ac} of $\text{CePd}_{2.02}\text{Ge}_{1.98}$. The entrance into the antiferromagnetically ordered state is clearly visible. Inset: Frequency test at $p = 8.9$ GPa for different temperatures. The solid lines represent a fit of a low pass filter to the data with cut-off frequencies $\omega_1/(2\pi) = 350$ Hz and 1060 Hz for 4.2 K and 9 K, respectively.

Fig. 2). Hence, pressures up to 9 GPa seem to have a weak effect on $\kappa(T)$ of stearite below 10 K.

The most important observation in this experiment is the pressure dependence of the value of $1/V_{ac}$ taken at about 0.3 K. The inverse of the lock-in voltage, $1/V_{ac}$, strongly increases, reaches a maximum in the vicinity of p_c and levels off at high pressure (Fig. 7). The critical pressure was inferred from the $\tilde{A}(p)$ -anomaly in the temperature dependence of the electrical resistivity $\rho(T) = \rho_0 + \tilde{A}T^n$, with ρ_0 the residual resistivity, and the fitting parameters \tilde{A} and n [14]. Below 1 K, $1/V_{ac}$ is proportional to C/T , since the temperature dependence of the absolute thermopower, $S(T) \propto T$, is a fairly good assumption. Above this temperature the $S(T)$ dependence is certainly different and $1/V_{ac}$ has to be interpreted with caution. Thus, $1/V_{ac}(T)$ at low tempera-

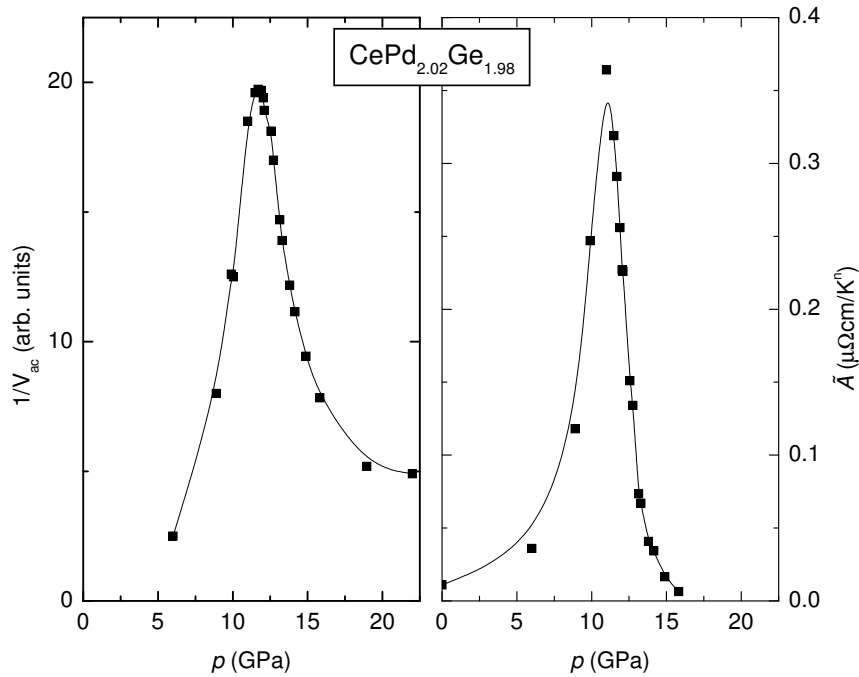


Fig. 7. Left: Pressure dependence of the inverse lock-in voltage V_{ac} of CePd_{2.02}Ge_{1.98} obtained at the lowest temperatures reached in each run. The maximum is attained at a pressure very close to the critical pressure where the magnetic ordering temperature is pushed to zero. Right: The temperature coefficient \tilde{A} of the electrical resistivity. It shows an anomaly at the magnetic/non-magnetic phase transition.

ture can be regarded as a direct measure of the electronic correlations. The pronounced pressure dependence of $1/V_{ac}$ shows that the electronic correlations are considerably enhanced as pressure approaches p_c and that the signal originates mainly from the sample. However, above 15 GPa, the pressure dependence is not strong enough to follow the $\tilde{A}(p)$ -dependence according to the empirical Kadowaki-Woods relation [25]. A possible reason for this deviation might be that at these pressures V_{ac} does not represent entirely the heat capacity of the sample. A step towards a quantitative measure of the specific heat at these conditions would be to achieve a control of the supplied heating power and the thermal contact between sample and pressure transmitting medium. Nevertheless, the strong pressure dependence of $1/V_{ac}$ at low temperature is reminiscent to $\tilde{A}(p)$ and is a motivation for further studies.

4 Conclusions

The AC-calorimetric technique adapted for high pressure experiments at low temperature ($T < 10$ K) was discussed. The oscillating sample temperature provides the specific heat of the sample if the measuring frequency is above the cut-off frequency $\omega_1 = \kappa/C$. It is determined by the thermal conductivity of the pressure transmitting medium and the specific heat of the sample. A qualitative estimate of $\kappa(T)$ for steatite and solid He, the two preferred pressure media was made. The cut-off frequency for steatite is less than 1 kHz whereas several kHz was inferred for solid He (at ≈ 2 GPa and 4.2 K). An experimental confirmation of the order of magnitude for these values was found for pressures up to 7 GPa and temperatures in the range of $1.5 \text{ K} < T < 10 \text{ K}$. The large values in the case of He put a temperature limit for the use of a DAC whereas a Bridgman-type of high pressure cell can be used below 10 K and pressures well above 10 GPa. Due to the strong pressure dependence of $\kappa(T)$ of He, the maximum in $\kappa(T)$ shifts towards higher temperature. This might open the low temperature region for the AC-calorimetric method also for a DAC. These promising results build up the hope of a quantitative understanding of the AC-calorimetry and interesting specific heat data under extreme conditions might be expected.

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