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LETTER TO THE EDITOR

Pressure dependence at low temperatures of thermodynamic properties of the heavy fermion compound CeAl_3

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Abstract. Values of $(\partial\beta/\partial P)_T$ derived thermodynamically from the zero pressure ultrasonic data of Niksch and co-workers indicate that the absolute magnitude of the volumetric thermal expansion coefficient β should be initially enhanced by pressure both at very low temperatures, where β is negative, and at higher temperatures when β becomes positive. Values of $[\partial^2(C_P/T)/\partial P^2]_T$, similarly derived, are in qualitative agreement with measurements of C_P under pressure above 0.5 K, but at lower temperatures exacerbate an apparent inconsistency with thermal expansion data noted earlier by Brodale and co-workers; it is unlikely that this anomaly is due solely to sample variation.

CeAl_3 has been named as a prototype heavy fermion material (Ott 1987). At high temperatures it behaves like a Curie-Weiss antiferromagnet with a single f-electron localized on each Ce^{3+} ion. No phase transitions occur as the temperature is lowered, and at low temperatures highly enhanced heat capacity and magnetic susceptibility indicate that the f-electrons are now highly correlated and hybridized with the conduction electrons, forming a very narrow band in the density of states. However, no quantitative theory has been established (Grewe and Steglich 1991).

So far it has been possible to prepare only small (\sim mm-sized) single crystals of CeAl_3 (Jaccard *et al* 1987), and the data discussed in this letter all come from measurements on polycrystalline samples. CeAl_3 is difficult to obtain pure, and samples usually contain appreciable concentrations (a few per cent) of CeAl_2 and $\text{Ce}_3\text{Al}_{11}$ (Ribault *et al* 1979). In addition, the texture of the polycrystals may not be macroscopically uniform: for example, Jaccard *et al* found that CeAl_2 formed chiefly at the surface of their annealed material, and that oriented regions of CeAl_3 extended over distances of some mm, giving rise to macroscopic anisotropy. It is therefore not surprising that properties can be strongly sample-dependent (Ribault *et al* 1979, Brodale *et al* 1986; see also figure 1 below). Furthermore, differences between heat capacity measurements by Andres *et al* (1975) and by Bredl *et al* (1984) on samples 'cut from the same button' suggest that properties may change during sample storage, although the differences could also be due to inhomogeneity of the original button. It follows that the quantities derived in the present letter from the results of measurements on different samples cannot be precise. However, CeAl_3 exhibits such extreme behaviour that even semi-quantitative or qualitative results are significant.

At low temperatures the thermal expansion is very large and changes sign with temperature (figure 1). This behaviour is related to the pressure dependence of the

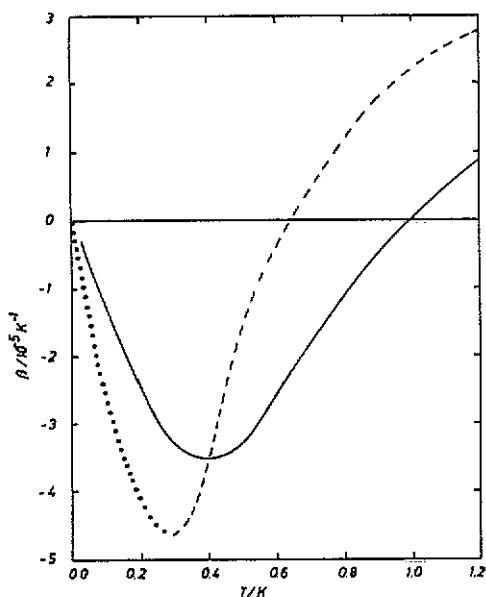


Figure 1. Coefficient of volumetric expansion β for polycrystalline CeAl_3 . —, Ribault *et al* (1979); - - -, Andres *et al* (1975) (....., extrapolation).

heat capacity by the thermodynamic relation

$$[\partial(C_P/T)/\partial P]_T = -[\partial(\beta V)/\partial T]_P \quad (1)$$

where β is the coefficient of volumetric expansion. The heat capacity below 1 K has been measured by Brodale *et al* (1986) at a number of pressures (figure 2). The results correlate well with those for β above 1 K, in that they indicate a change of sign in $[\partial(C_P/T)/\partial P]_T$ at about 2 K when β passes through a maximum. There is however no indication of another change of sign in $[\partial(C_P/T)/\partial P]_T$ at lower temperatures, which we should expect to see when β passes through its minimum somewhere below 0.5 K. Brodale *et al* believed that the most probable explanation is that the change of sign does indeed occur, but only at pressures lower than 0.4 kbar.

The present letter examines additional information, which is available from the temperature dependence of the compressibility χ .

The key thermodynamic relation is

$$[\partial\beta/\partial P]_T = \partial^2 \ln V / \partial P \partial T = -[\partial\chi_T/\partial T]_P. \quad (2)$$

Differentiation with respect to T gives

$$\partial^2 \beta / \partial P \partial T = -[\partial^2 \chi_T / \partial T^2]_P \quad (3)$$

and so, by equation (1), after removal of terms negligible for CeAl_3 at low temperatures,

$$[\partial^2(C_P/T)/\partial P^2]_T \simeq V[\partial^2 \chi_T / \partial T^2]_P. \quad (4)$$

χ_T is related to the adiabatic compressibility by

$$\chi_T / \chi_S = 1 + \beta \gamma T \quad (5)$$

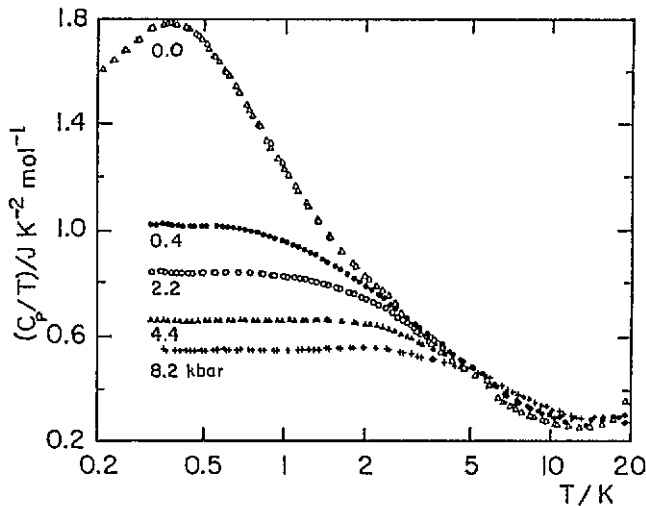


Figure 2. C_P/T for polycrystalline CeAl_3 at various pressures (Brodale *et al* 1986).

where γ is the thermodynamic Grüneisen function (see e.g. Barron *et al* 1980) defined by

$$\gamma \equiv [\partial(PV)/\partial U]_V = \beta V/\chi_S C_P. \quad (6)$$

If needed, equations (1) to (4) can be generalized for anisotropic materials; e.g. (2) becomes

$$[\partial\alpha_\lambda/\partial t_\mu]_{T,t'} = [\partial s_{\lambda\mu}^T/\partial T]_t \quad (7)$$

where (Barron *et al* 1980) α_λ is a thermal expansion component, t_μ a stress component and $s_{\lambda\mu}^T$ an isothermal elastic compliance.

We make the usual assumption that ultrasound in heavy fermion materials is transmitted adiabatically (Thalmeier and Lüthi 1991)†. Ultrasonic velocities have been measured between 50 mK and 175 K by Nicksch *et al* (1980), in a polycrystalline sample from the same batch as the sample used by Andres *et al* (1975). Values of the compressibility below 1.2 K (derived from figure 45 of Thalmeier and Lüthi (1991)) are plotted as χ_S in figure 3. The variation with temperature is due predominantly to that of the longitudinal sound velocity. At higher temperatures χ_S continues to decrease with temperature, but at a slower rate.

Because β is so large, the difference between χ_S and χ_T cannot be neglected even at very low temperatures (Lüthi and Yoshizawa 1987). In applying equations (5) and (6) we have a choice of experimental values for β and C_P . Smoothed results obtained with two different data sets are shown in figure 3. Set A, with β from Ribault *et al* (1979) and C_P from Brodalen *et al* (1986), is preferred for consistency with the direct pressure measurements of Brodalen *et al*; their zero pressure heat

† The theory of Becker and Fulde (1987) suggests that for sufficiently low ultrasonic frequencies this might cause χ_S to be underestimated by an amount of comparable order to $\chi_T - \chi_S$; but no quantitative estimates are available.

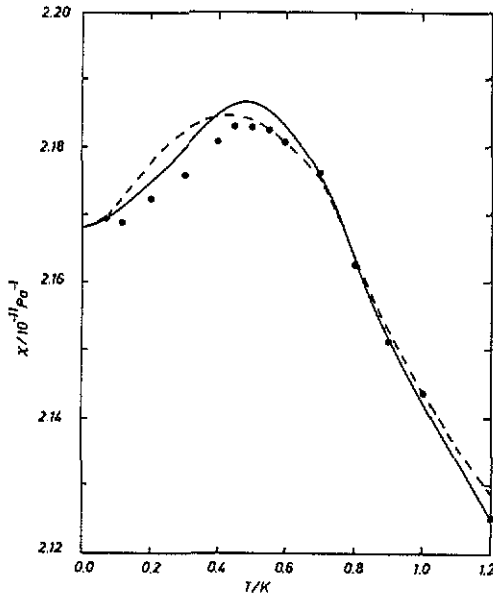


Figure 3. Compressibility of polycrystalline CeAl_3 . \bullet , χ_S (Niksch *et al* 1980); —, χ_T (data set A); ---, χ_T (data set B).

capacity curve at low temperatures is fairly close both to that of Bredl *et al* (1984) for a sample from the same batch as that used for the elastic measurements, and to that of Benoit *et al* (1981) for the sample used by Ribault *et al* to measure β . Set B uses the markedly different earlier values for both β and C_P of Andres *et al* (1975), so as to give some indication of the dependence of our conclusions on the choice of data. The curves have been extrapolated to zero slope at $T = 0$ in accordance with the third law.

By equation (2), figure 3 shows that β should decrease under pressure below about 0.45 K and thereafter increase with pressure. The magnitude of $[\partial\beta/\partial P]_T$ over parts of the low temperature range is large (table 1). It is interesting that β and $[\partial\beta/\partial P]_T$ both change sign, so that the absolute magnitude of the thermal expansion increases with pressure both below ~ 0.45 K, when β is negative, and above ~ 1.0 K, when β is positive. Elastic measurements under moderate pressure should determine whether this is because the material becomes more compressible or because the temperature variation of the Grüneisen function becomes steeper.

Table 1. Magnitudes of thermodynamic properties of polycrystalline CeAl_3 below 1 K.

T (K)	Data set	$(\partial\beta/\partial P)_T$ ($10^{-5}\text{K}^{-1}\text{ kbar}^{-1}$)	$[\partial(C_P/T)/\partial P]_T$ ($\text{J mol}^{-1}\text{K}^{-2}\text{ kbar}^{-1}$)	$[\partial^2(C_P/T)/\partial P^2]_T$ ($\text{J mol}^{-1}\text{K}^{-2}\text{ kbar}^{-2}$)
0.35	A	-5-	2	0
	B	-3	-5	-1
0.5	A	0	-4	-3
	B	1	-9	-2
0.8	A	12	-3	0
	B	11	-4	0
1.0	A	9	-3	1
	B	9	-2	1

Despite the greater uncertainty caused by taking second derivatives, there is little doubt that $[\partial^2 \chi_T / \partial T^2]_P$ and consequently $[\partial^2(C_P/T) / \partial P^2]_T$ are positive at sufficiently low temperatures, negative between ~ 0.3 and ~ 0.8 K, and thereafter positive until much higher temperatures. This means that the increase of C_P with pressure at very low temperatures and its decrease with pressure between the minimum in β and ~ 0.8 K both initially become steeper with increasing pressure; it is only above about 0.8 K that the decrease in C_P with pressure becomes initially less steep as the pressure is increased.

To obtain numerical estimates from equations (1) and (4) we take the molar volume as $51.5 \text{ cm}^3 \text{ mol}^{-1}$. The data shown in figures 1 and 3 then lead to the rough estimates of pressure derivatives shown in table 1. Consistency with the direct pressure measurements shown in figure 2 is reasonable at higher temperatures. For example, at 0.50 K the negative initial slope of C_P/T against P is not large enough by itself to account for a drop of $0.7 \text{ J mol}^{-1} \text{ K}^{-2}$ in an interval of 0.4 kbar, even if we take the larger of the two experimental values, but $[\partial^2(C_P/T) / \partial P^2]_T$ is also strongly negative. Around 0.35 K, however, where Brodale *et al* found a similar drop in C_P/T over 0.4 kbar, not only is the initial slope positive for our preferred set A (for set B it is still negative but smaller in magnitude), but also $[\partial^2(C_P/T) / \partial P^2]_T$ is now approximately zero (still negative but smaller for set B). One effect of the present analysis is thus to exacerbate the anomaly in this region first pointed out by Brodale *et al*.

To sum up, the negative value of $(\partial\beta/\partial P)_T$ at the lowest temperatures gives further confirmation that the extreme heavy fermion properties of CeAl_3 are initially enhanced by the application of pressure. First and second pressure derivatives of C_P/T are in qualitative agreement with direct pressure measurements at higher temperatures, but around 0.35 K the data are hard to reconcile with each other. The effect of the application of moderate pressures on a variety of properties should thus be a rewarding field of investigation, particularly if samples are carefully prepared and measurements of different properties (including texture) are made on the same sample. Measurements carried out shortly after sample preparation and then repeated after suitable intervals should reveal any effects of aging.

I thank Professor M Springford for comments on the first draft of this letter, and the referee for stressing the importance of texture and for drawing my attention to the paper of Jaccard *et al*.

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