

SUPERCONDUCTIVITY, SPIN FLUCTUATIONS AND MAGNETIC ORDER IN URANIUM COMPOUNDS UNDER HIGH PRESSURE

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Invited paper

The systematics in the occurrence of superconductivity and magnetic order in the actinide compounds is usually represented by a critical value for the nearest neighbour distance between the actinide ions. On the basis of these systematics a suppression of magnetism and an enhancement of superconductivity with pressure is expected.

High-pressure results are reviewed for a superconducting system (U₂Co), a spin-fluctuating compound (UPt₃) and for a series of (nearly) ferromagnetic compounds (UX₂ with X = Fe, Co, Ni, Pt, Ga and Ge).

1. Introduction

The electronic and magnetic properties of the actinide metals and their compounds are largely determined by the partially filled 5f shell [1-4]. The 5f electrons in the actinides are less localized than the 4f electrons in the corresponding rare earth series. In the light actinide elements Th to Pu, no magnetic order has been observed. Instead, superconductivity occurs in Th, Pa and U, whereas spin-fluctuation effects are found in Np and Pu. Magnetic order has been reported for Cm and Bk; Am is supposed to be in a non-magnetic ground state. The overlap between the 5f wave functions of neighbouring atoms decreases going from the light to the heavy actinide metals. This overlap can also be varied in a series of suitably chosen uranium compounds. In such a series the gradual change from itinerant electron states (superconductivity) to localized states (complex magnetic structures with large magnetic anisotropies) via the intermediate spin-fluctuation state can be studied. The systematics in the occurrence of superconductivity and magnetic order in the actinide compounds is usually represented by a critical value for the distance between nearest neighbour actinide atoms. This value is known as the Hill limit and amounts to 3.4-3.6 Å [1]. Below this value no spontaneous magnetic order of the actinide sublattice should occur, whereas beyond

this value no superconductivity is expected to be found. The Hill limit provides the restrictions under which magnetic order or superconductivity occurs in the actinide systems. It does not predict these phenomena. In most of the intermetallic compounds between uranium and the d-transition metals, for instance, no magnetic order is observed, although in many cases the distance between the uranium atoms is well above the Hill limit.

Recently, it became clear that the systematics of the Hill plot is not followed by some exciting materials like UBe₁₃ [5] and UPt₃ [6]. In both compounds the nearest neighbour U-U distances are too large to expect superconductivity. From an analysis of the temperature dependence of the critical field for superconductivity, large effective masses (about 200m_e) have been derived for the electrons that participate in the superconductivity, indicating that uranium 5f electrons are involved [5, 7]. Other exceptions to the systematics of the Hill plot are the ferromagnetic compounds UFe₂ and UNi₂. In both compounds the nearest neighbour U-U distances are smaller than the critical value. The magnetic properties of UFe₂ are mainly due to the iron sublattice [8]. In case of UNi₂, however, magnetization studies on single crystalline samples reveal extremely large magnetic anisotropies, a characteristic property of f-electron magnetism [9].

In our review of high-pressure effects in

uranium compounds, some of these exceptions will be discussed in more detail. Subsequently, we report on high-pressure studies on the superconducting transition in U_6Co , on the spin-fluctuation phenomena in UPt_3 and on the magnetic properties of a series of UX_2 compounds.

2. Superconductivity in U_6Co

Superconductivity in the U_6X ($X = Mn, Fe, Co, Ni$) is known for more than 15 years [10]. Recent specific heat measurements on U_6Fe [11] and U_6Co [12] reveal a relatively large value for the coefficient of the electronic term, indicating a density of states per atom at the Fermi level that is at least two times larger than in metallic uranium. For that reason these compounds are considered to belong to the class of the heavy-Fermion superconductors, although the effective electron masses are not so extremely large in these compounds as, for instance, in $CeCu_2Si_2$ [13], UBe_{13} [5] and UPt_3 [7] where values of the order of $200m_e$ have been reported. Typically large values for the temperature dependence of the upper critical field are found in the heavy-Fermion superconductors. A collection of values for the superconducting parameters is given in table I. The values for the temperature dependence of the upper critical field of the U_6X compounds follow in good approximation the "dirty-limit" expression [14]:

$$\frac{\partial B_{c2}}{\partial T} = -4480\gamma\rho \quad (T/K), \quad (1)$$

where γ , the coefficient of the linear term in the

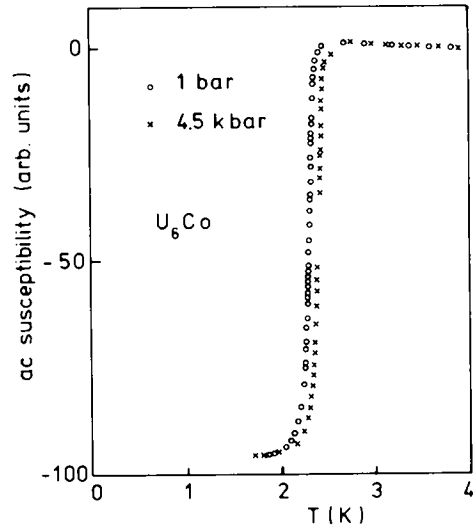


Fig. 1. The superconducting transition in polycrystalline U_6Co as a function of pressure.

specific heat, is expressed in J/m^3K^2 and ρ , the resistivity in the normal state at the superconducting transition, in $10^{-8} \Omega m$.

The susceptibility of U_6Co has been measured at 4.2 K in fields up to 20 T and is found to be not particularly large: $\chi = (34 \pm 5) \times 10^{-9} m^3/mol.f.u.$ Defining a Stoner factor S as the ratio of the observed values for the susceptibility and the coefficient of the electronic term in the specific heat: $S = 5.80 \times 10^6 \chi/\gamma$, a value for S of 1.6 is derived. Larger values for S must be expected if electron-phonon enhancement effects are considered. The effect of pressure on the superconducting transition temperature of U_6Co has been studied by performing ac-susceptibility measurements at zero pressure and under solid helium pressures of 4.5 kbar, see fig. 1. The

Table I
Superconducting parameters of some "heavy-Fermion superconductors"

| Compound | T_c (K) | γ (mJ/K ² mol.f.u.) | $\partial B_{c2}/\partial T$ (T/K) | $\partial T_c/\partial p$ (K/kbar) | Ref. |
|--------------|--------------|--|---------------------------------------|---------------------------------------|------|
| $CeCu_2Si_2$ | 0.6 | 1006 | -5.8 | | 13 |
| UBe_{13} | 0.85 | 1100 | -25.7 | | 5 |
| UPt_3 | 0.48 | 422 | -4.4 | | 7 |
| U_nFe | 3.8 | 155 | -3.4 | | 11 |
| U_6Co | 2.3 | 126 | -3.7 | 0.022 | 12 |

resulting value for $\Delta T_s/\Delta p$ is (0.022 ± 0.003) K/kbar. This pressure effect is undoubtedly positive and fits to the general concept that superconductivity is promoted in the uranium compounds by decreasing the interatomic distances. In a more quantitative interpretation of this positive pressure effect we have to consider the pressure dependence of the phonon frequencies and that of a series of many-body parameters arising from electron-phonon interaction (λ_{eph}) spinfluctuations (λ_s) and a Coulomb term (λ_c).

For two other compounds of the superconducting series of U_6X compounds, U_6Fe and U_6Mn , no significant change in T_s with pressure has been observed, although it was not excluded that a small effect in these experiments was masked by the width of the transition [15]. As a first approximation we make use of the expression for T_s , recently applied to the A15 superconductors by Orlando and Beasley [16]:

$$T_s = 0.375 \theta_D e^{-\alpha}, \quad (2)$$

with θ_D the Debye temperature and α a combination of many-body parameters: $\alpha = (1 + \lambda_{\text{eph}} + \lambda_s)(\lambda_{\text{eph}} - \lambda_s - \lambda_c)^{-1}$. For the relative pressure dependence of T_s we write

$$\frac{\partial \ln T_s}{\partial p} = \frac{\partial \ln \theta_D}{\partial p} - \frac{\partial \alpha}{\partial p}. \quad (3)$$

Taking a value of 2 for the phonon Grüneisen parameter $-\partial \ln \theta_D/\partial \ln V$ and, by lack of information on the compressibility κ of U_6Co , a value for κ of 1.0×10^{-11} m²/N, equal to that of metallic uranium, we calculate

$$\frac{\partial \alpha}{\partial p} = -7.6 \times 10^{-11} \text{ m}^2/\text{N}.$$

Using a value of 11 / K for θ_D as derived from the specific heat results for U_6Co [12] we obtain for α a value of 2.93 and for its relative volume derivative $\partial \ln \alpha/\partial \ln V$ a value of 2.6. The positive volume dependence of α most likely points to a reduction of spin fluctuation effects

and/or effective Coulomb interactions at decreasing interatomic distances.

3. Spin-fluctuation phenomena in UPt_3

Actinide metals and intermetallic compounds with large density of states at the Fermi level often exhibit spin-fluctuation effects. In general, these spin fluctuations manifest themselves by a $T^3 \ln T$ term in the specific heat, a T^2 -term in the low-temperature susceptibility with either a positive or negative sign and by a T^2 -term in the resistivity at low temperatures in combination with a large increase of the resistivity (of the order of 100 $\mu\Omega\text{cm}$) going up to room temperature. According to these criteria UAl_2 and UPt_3 have been classified as spin-fluctuation systems among other uranium compounds. Both compounds have been studied under pressure: Fournier et al. performed susceptibility measurements on UAl_2 [17], De Visser et al. [18] measured the resistivity of single-crystalline UPt_3 (hexagonal $MgCd_3$ -type of structure). The susceptibility [19] and the resistivity [18] of UPt_3 are strongly anisotropic, indicating that large orbital moments of the uranium 5f electrons are involved. Most remarkable is the superconductivity in UPt_3 that has been observed below 0.5 K [6]. Extended studies of the Meissner effect [20] reveal that superconductivity must be considered as a bulk property in this compound. Regarding spin-fluctuation phenomena as the precursor of magnetic moment formation, we expect a suppression of spin-fluctuation effects at the application of hydrostatic pressures. This suppression has been observed indeed in resistivity measurements under pressure, see fig. 2. These high-pressure data can be summarized as follows:

at the lowest temperature $\rho(T)$ approaches the expression:

$$\rho(T) = \rho_0 + AT^2, \quad (4)$$

where the residual resistivity, of the order of a few $\mu\Omega\text{cm}$, is insensitive to pressure; the coefficient A is strongly anisotropic and amounts to 1.6 and 0.7 $\mu\Omega\text{cm}/\text{K}^2$ for the basal plane and the hexagonal axis, respectively; a relative decrease in A

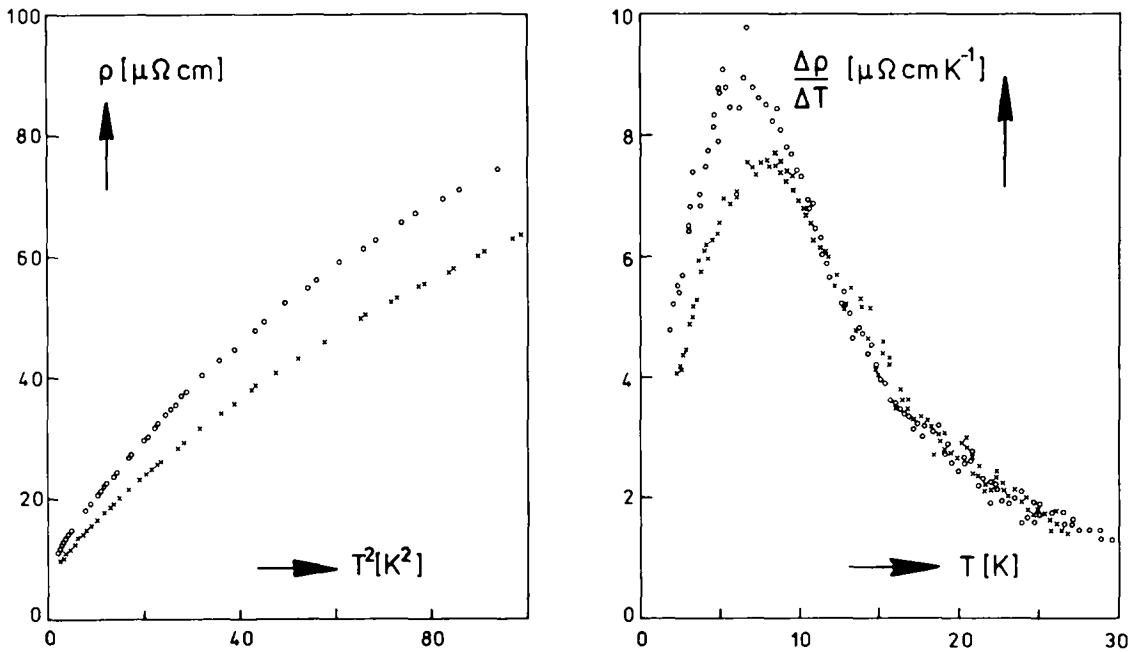


Fig. 2. Electrical resistivity of single-crystalline UPt_3 versus T^2 at zero pressure (○) and at a pressure of 4.2 kbar (×) and its temperature derivative, measured along the b -axis.

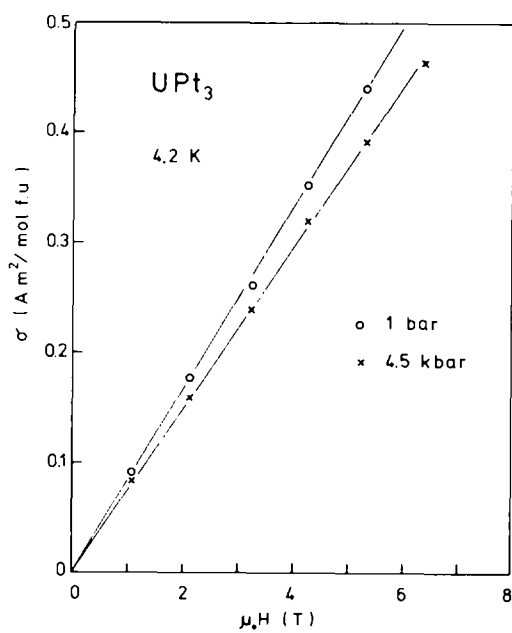


Fig. 3. Magnetisation curve of polycrystalline UPt_3 at 4.2 K at different pressures.

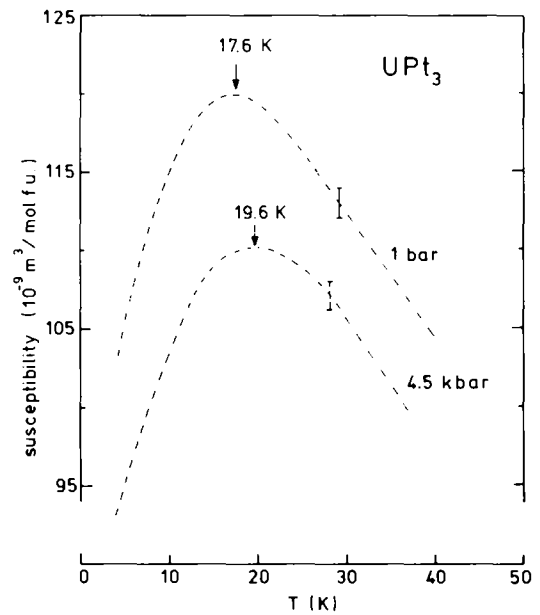


Fig. 4. Dc susceptibility of polycrystalline UPt_3 as a function of temperature at different pressures; the applied field is 5.5 T.

with 40% has been observed at applying a pressure of about 4 kbar near 4.2 K: the resistivity strongly increases with temperature below 100 K and reaches values of 238 and 132 $\mu\Omega\text{ cm}$ at room temperature for the basal plane and the hexagonal axis, respectively; the resistivity slightly decreases with pressure in the high-temperature region: in the basal plane a temperature-independent reduction of 1.4 $\mu\Omega\text{ cm/kbar}$ is observed above 45 K. The relatively small and negative pressure effect at higher temperatures points to a slight dehybridization of the *s* and *f* electron states by which the spin-fluctuation scattering is reduced. Ascribing the residual resistivity to impurity effects (atomic disordering, lattice imperfections, impurity atoms), we expect no pressure dependence for ρ_0 . The temperature derivative of the resistivity of UPt_3 along the *b*-axis shows a maximum at zero pressure around 6 K. The temperature at which this maximum occurs shifts to 8 K under a pressure of 4.5 kbar. The main result of our high-pressure resistivity studies is the enormously large pressure dependence of the coefficient *A* in eq. (4) in the low temperature region.

The magnetisation curve at 4.2 K has been studied on a polycrystalline sample under pressures of 4.5 kbar in magnetic fields up to 6.5 T, see figs. 3 and 4. The dc susceptibility decreases under pressure resulting in a value for $\partial \ln \chi / \partial p$ of -24 Mbar^{-1} . In addition, we measured the pressure dependence of the magnetisation in a field of 5.5 T between 4.2 K and 40 K. From these experiments we conclude that the temperature at which the maximum in the susceptibility occurs shifts from 17.6 K at zero pressure to 19.6 K at a pressure of 4.5 kbar. The value for $\partial \ln T_{\text{max}} / \partial p$ is: 25 Mbar^{-1} . From these numbers for the relative pressure derivatives we conclude that the product χT_{max} is independent of pressure, indicating that the susceptibility is inverse proportional to T_{max} .

4. Magnetic order

High-pressure studies on the intermetallic uranium compounds contribute to the under-

standing of the different types of magnetic order present in the actinide metals. Transition metal systems with low values for the magnetic ordering temperature (e.g. ZrZn_2 , Ni_2Al) often show large pressure effects on the spontaneous magnetization near zero temperature as well as on the transition temperature itself. For these materials the relative pressure dependence of these parameters are both negative and of the same order of magnitude. The absolute values for this relative pressure effect increase in one series (e.g. the non-stoichiometric $\text{Ni}_{1-x}\text{Al}_{1+x}$ alloys) at decreasing values of the spontaneous magnetization or the magnetic ordering temperature. These results can be understood in the Stoner-Wohlfarth model for weak itinerant ferromagnetism [21].

Following the results of our high-pressure studies on the intermetallic compounds $\text{U}(\text{Fe}_{1-x}\text{Co}_x)_2$ with *x*-values between 0 and 0.3, we can characterize the ferromagnetism in this series as itinerant. The compound UNi_2 with a value for T_c of 27 K reveals a much smaller value for the relative pressure dependence of the magnetic parameters than $x = 0.3$ of the $\text{U}(\text{Fe}_{1-x}\text{Co}_x)_2$ series which has nearly the same value for T_c . This observation provided one of the arguments to develop a different type of description for the ferromagnetic order in UNi_2 : magnetic order in this compound is due to the interaction between a small number of local moments by means of the susceptible UNi -matrix [21, 23]. These local moments were thought to be connected with defects in the atomic structure, possibly nickel antistructure atoms. Another strong argument for this type of description is the weak temperature dependence of the susceptibility between 100 K and 1000 K:

$$\chi(100 \text{ K}) = 14 \times 10^{-5} \text{ m}^3/\text{mol.f.u.},$$

$$\chi(1000 \text{ K}) = 11 \times 10^{-5} \text{ m}^3/\text{mol.f.u.}$$

Since that time single crystals of UNi_2 became available. The magnetization curves for this hexagonal Laves phase compound are strongly anisotropic indicating that uranium moments with large orbital contributions are involved [9].

Table II
Relative pressure derivatives of the magnetic parameters of UX_2 compounds

| Compound | σ_0 (μ_B/cell) | $\chi(4.2\text{ K})$ ($10^{-9}\text{m}^3/\text{mol.f.u.}$) | T_c (K) | $\frac{\partial \ln \sigma_0}{\partial p}$ | $\frac{\partial \ln \chi}{\partial p}$ | $\frac{\partial \ln T_c}{\partial p}$ |
|-------------------------|---------------------------------------|---|--------------|--|--|---------------------------------------|
| | | | | (Mbar^{-1}) | (Mbar^{-1}) | (Mbar^{-1}) |
| UFe_2 | 1.1 | | 160 | -4.5 | | -3.4 |
| $U(Fe_{0.9}Co_{0.1})_2$ | 0.84 | | 129 | -6.0 | | -4.8 |
| $U(Fe_{0.8}Co_{0.2})_2$ | 0.54 | | 87 | -8.9 | | -9 |
| $U(Fe_{0.7}Co_{0.3})_2$ | 0.14 | | 24 | -44 | | -37 |
| $U(Fe_{0.6}Co_{0.4})_2$ | | 59 | | | -27 | |
| UCO_2 | | 17 | | | -4.5 | |
| UNi_2 polycr. | 0.05 | 16 | 27 | -10 | -11 | -6.7 |
| (a,b) plane | 0.08 | 12 | 27 | | | |
| c-axis | <0.004 | $\ll 12$ | | | | |
| UPt_2 | | 51 | | | -10 | |
| UAl_2 | | 57 | | | -14* | |
| UGa_2 | 2.4 | | 120 | -16 | | 1.9 |
| UGe_2 polycr. | 0.7 | | 52 | -6.8 | | -24 |
| a, b-axis | <0.09 | | | | | |
| c-axis | 1.4 | | 52 | -2.4 | | |

* From ref. 17 at 4.2 K.

The susceptibility along the hexagonal axis is an order of magnitude less than that along any direction in the hexagonal plane. As a consequence, the internal field, connected with the small fraction of local magnetic moments, produces a much larger spontaneous magnetic moment in the hexagonal plane, compared to that along the hexagonal axis.

In a more systematic study of pressure effects on the magnetic properties of uranium compounds we investigated a number of UX_2 compounds, see table II.

For the non-cubic compounds, i.e. hexagonal UNi_2 and UGa_2 and orthorhombic UGe_2 , one has to be careful in interpreting the pressure effects on the spontaneous magnetization near zero temperature, measured on polycrystalline samples. Due to the large magnetic anisotropy, saturation can not be reached, even not in fields up to 10 T. For these materials the pressure effect on the magnetization curves could equally point to a change in magnetic anisotropy as to a variation of the spontaneous magnetic moment. For that reason we prepared single crystals of UGe_2 and UNi_2 and measured so far the pressure

effects on the magnetization curves for polycrystalline (UGe_2 , UNi_2) and single-crystalline (UGe_2) samples. The difference between the single-crystalline and polycrystalline data for UGe_2 in table II illustrate the above presented discussion.

Magnetization data around T_c are shown for

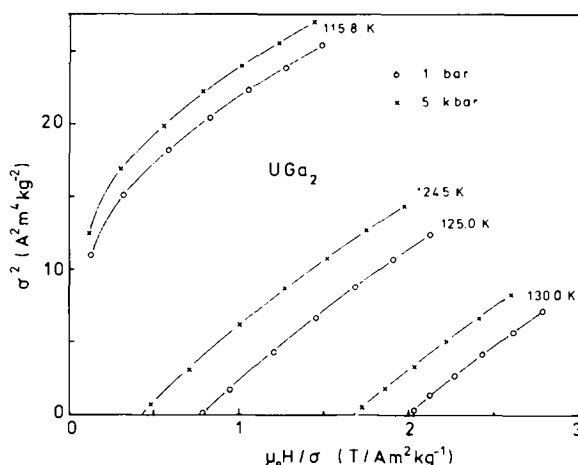


Fig. 5. Magnetisation data for polycrystalline UGa_2 in a plot of σ^2 versus $\mu_0 H/\sigma$ at different temperatures and pressures.

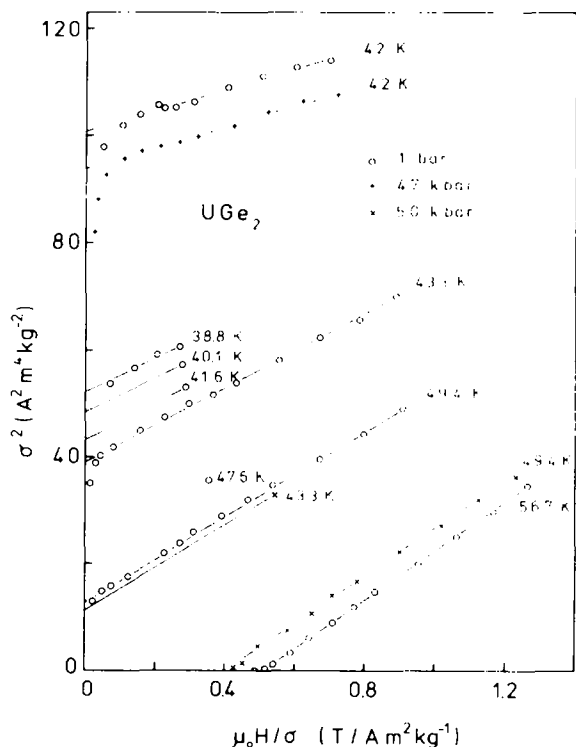


Fig. 6. Magnetisation data for polycrystalline UGe₂ in a plot of σ^2 versus $\mu_0 H/\sigma$ at different temperatures and pressures.

UGa₂ and UGe₂ in a plot of σ^2 versus H/σ in figs. 5 and 6. The curves for UGa₂ reflect critical phenomena. An extended study of these critical phenomena as a function of pressure has been performed for UFe₂ [21, 23, 7]. Results of this study have been used for estimating the reduction in the absolute value of the magnetic moment for an itinerant ferromagnet between zero tem-

perature and T_c . The pressure effect on T_c is extremely large for UGe₂, contrary to the variation with pressure of the spontaneous magnetization. The strong anisotropy and the different values for the relative pressure dependences of the spontaneous magnetization and the Curie temperature are considered as a proof for the localized nature of magnetism in this compound. Nevertheless, we observe a constant non-zero value for the differential susceptibility of UGe₂ along the easy axis of magnetization in fields up to 35 T [7].

Pressure effects on the Curie temperature of UGa₂ are undoubtedly positive, whereas the magnetization curve for a polycrystalline sample at 4.2 K exhibits negative pressure effects. Positive pressure effects on the magnetic ordering temperature have been observed, for instance, in nickel and nickel-rich alloys with Cu, Pt etc. These pressure effects can be understood with the well-known expression that follows from the Stoner-Edwards-Wohlfarth theory [24, 25]:

$$\frac{\partial \ln T_c}{\partial p} = \frac{\alpha}{T_c^2} + \frac{5}{3} \kappa, \tag{5}$$

where the factor $\frac{5}{3}$ represents the relative volume derivative of the effective degeneracy temperature and with α a parameter that can be expressed in the band parameters and the effective Coulomb interaction. For magnetism due to 5f electrons the factor $\frac{5}{3}$ should be replaced by $\frac{7}{3}$. Assuming a value for the compressibility of UGa₂ of 1 Mbar⁻¹ we find that $\partial \ln T_c / \partial p$ is close to the result calculated with eq. (5) accepting that

Table III

Values for the magnetic parameters of a series of paramagnetic UX₂ compounds; $S = 5.80 \times 10^6 \chi/\gamma$. $\gamma = \lim_{T \rightarrow 0} c/T$. χ is defined as the initial susceptibility for UCo₂, UPt₂ and UAl₂ and as the differential susceptibility in high magnetic fields for UNi₂; data from refs. 21 and 23

| Compound | γ (10 ⁻³ JK ⁻¹ /mol.f.u.) | χ (10 ⁻⁶ m ³ /mol.f.u.) | S | $\frac{\partial \ln \chi}{\partial p}$ (Mbar ⁻¹) | κ (Mbar ⁻¹) |
|------------------|---|---|-----|---|-----------------------------------|
| UCo ₂ | 35 | 17 | 2.8 | 4.5 | |
| UNi ₂ | 65 | 14 | 1.2 | 11 | |
| UPt ₂ | 77 | 51 | 3.8 | 10 | |
| UAl ₂ | 133 | 57 | 2.5 | 14* | 1.65* |

*From ref. 17 at 4.2 K.

the first term in the right hand side of eq. (5) is negligible.

A direct comparison between the values for $\partial \ln \sigma_0 / \partial p$ and $\partial \ln T_c / \partial p$ of polycrystalline UGa₂ is less relevant because of the large magnetic anisotropy at low temperature. Although this large anisotropy points to localized magnetism, for this material too, suggestions have been made that part of the magnetic moment arises from conduction electron polarization [26]. The remaining paramagnetic compounds, the cubic Laves phases UAl₂ and UCo₂ and the orthorhombic compound UPt₂ all show a decrease of the susceptibility at 4.2 K with increasing pressures. Following Brommer [27] we write for the relative pressure derivative of the susceptibility:

$$\frac{\partial \ln \chi_0}{\partial p} = -\kappa \lambda I' (S - 1) - \kappa I', \quad (6)$$

with $I' = \partial \ln N(E_F) / \partial \ln V$, $\lambda I' = \partial \ln IN(E_F) / \partial \ln V$, S the Stoner enhancement factor and I the effective electron-electron interaction. For d metals we use $I' = \frac{5}{3}$, for f metals $I' = \frac{7}{3}$.

Values for the relevant parameters are collected in table III. The Stoner factor is calculated from the expression: $S = 5.80 \times 10^6 \chi / \gamma$ with χ in m³/mol.f.u. and γ in JK⁻²/mol.f.u. In this expression the different mechanisms that lead to enhanced γ -values are not considered just as the diamagnetic and orbital contributions to the susceptibility are not. Only in case of UAl₂ we dispose over a complete set of data to be able to derive a value for the many-body parameter λ . In order to get physically meaningful results for λ (i.e. between 0 and 1), the Stoner factor should be larger than 3.7 for UAl₂. Additional information on the electron-phonon and spin-fluctuation enhancement as well as on the different contributions to the susceptibility is required before presenting definite values for λ . The same information is needed for UCo₂, UNi₂ and UPt₂ for which compounds, in addition, the compressibility has to be determined.

5. Concluding remarks

The high-pressure technique is a useful tool for studying many-body phenomena in the uranium

intermetallics. Pressure effects on the susceptibility of the UX₂ compounds can, in principle, be translated in values for the many-body parameter that reflects the reduction of the bare intraatomic Coulomb interaction by correlation effects. High-pressure studies on superconducting materials inform us on the volume derivative of a combination of many-body parameters.

In a qualitative analysis we have shown by following the relative pressure dependence of the spontaneous magnetization at 4.2 K as a function of composition, that ferromagnetism in the U(Fe_{1-x}Co_x)₂ series is of an itinerant nature. Values for the relative pressure derivatives of σ_0 and T_c are of the same order of magnitude and of the same sign for this series. According to the strong anisotropy and the high-pressure results for σ_0 and T_c , ferromagnetism on UGe₂ and UGa₂ is more of a localized type.

Pressure effects on the magnetic parameters of UNi₂ ($T_c = 27$ K) are less pronounced than those of U(Fe_{0.7}Co_{0.3})₂ ($T_c = 24$ K). The magnetic moment is very small (0.08 μ_B /f.u.), the anisotropy extremely large, the susceptibility nearly temperature independent between 100 K and 1000 K. Two models have been proposed so far in the literature: a homogeneous type of magnetism due to a splitting of the 5f-electron band [28] and a second model in which defects in the structure play an important role [21, 23]. Also in this latter case the magnetic moments are due to the uranium 5f electrons. They are induced, however, by a small fraction of local magnetic moments associated with defects or partial disordering of the atomic structure.

The spin-fluctuation compound UPt₃ shows rather large pressure effects in the resistivity and in the dc susceptibility. Spin fluctuations are suppressed under pressure in this compound. Finally, we observe positive pressure effects on the superconducting transition temperature in U₆Co.

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