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Hidden Order in URu2Si2

Studied by High-Pressure Experiments

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Pressure-Induced Magnetic Phase Transition of the 5f Electron System URu_2Si_2

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A puzzling phase transition at $T_o \sim 17.5$ K of the heavy-electron 5f system URu_2Si_2 is characterized by development of an unusually small staggered moment μ_o concomitant with large bulk anomalies and sharp magnon excitations. Through elastic and inelastic neutron scattering, we have studied the influence of pressure P up to ~ 2.8 GPa on this phase transition. For pressures less than 1.3 GPa, μ_o at 1.4 K is quasi-linearly enhanced from $\sim 0.017(3)\ \mu_B$ to $\sim 0.25(2)\ \mu_B$, while its onset temperature increases only ten present. We have also observed a sharp phase transition at $P_c \sim 1.5$ GPa, above which a 3D-Ising type of antiferromagnetic phase ($\mu_o \sim 0.4\ \mu_B$) appears. Sharp magnon excitations at $Q = (1,0,0)$ and $(1,0.4,0)$ collapse in this high-P phase. The lattice parameter a decreases discontinuously at P_c even for $T > T_o$, indicating that pressure induces a significant change in electronic states in a higher characteristic energy scale. We show that the observed magnetic instability could qualitatively well be explained by competition between quadrupolar and dipolar interactions in a non-trivial Γ_5 doublet.

KEYWORDS: URu_2Si_2, tiny magnetic moment, neutron scattering, high pressure

§1. Introduction

The ternary intermetallic compound URu_2Si_2 is a well-known heavy-electron system in that 5f electrons clearly undergo two successive phase transitions at ~ 17.5 K and ~ 1 K.[1-4] It crystallizes in the body-centered tetragonal $ThCr_2Si_2$ structure with space group of I4/mmm. The 1K transition is known as the onset of unconventional superconductivity due to heavy quasi-particles, while the origin of the 17.5 K transition is still in controversy in spite of intensive studies for the past fifteen years.

Microscopic studies of neutron scattering[5-7] and X-ray magnetic scattering[8] have revealed a type-I antiferromagnetic modulation with 5f magnetic dipoles polarized along the c axis developing below about 17.5 K. The magnitude of the staggered moment μ_o is estimated to be ~ 0.02–$0.04\ \mu_B$/U, which is extremely small compared with the effective moment ($\sim 3\ \mu_B$) estimated from the high-temperature magnetic susceptibility.[1,2] The similar tiny-moment state has been reported for UPt_3[9] and $CeRh_2Si_2$ (under high pressure),[10] but URu_2Si_2 is unique in the following two respects.

First, the development of μ_o is accompanied by disproportionally large bulk anomalies.[1-4] For example, the specific-heat jump ($\Delta C/T \sim 300$mJ/K^{2}mol) at 17.5 K involving a large reduction of the 5f entropy ($S \sim 0.2R\ln 2$) cannot simply be reproduced by the one-present condensation of the 5f moments.[11] Secondly, an energy gap occurs in the density of states in the ordered state. This is indicated by a term, $\exp(-\Delta/T)$, appearing in various macroscopic quantities such as resistivity[12] and specific heat,[3] and also manifested in measurements of neutron scattering,[13] far-infrared reflectance,[14] and tunneling spectroscopy.[15] The evaluated gap averages ~ 100 K.

In the neutron scattering, the excitations are observed as spin waves which are polarized along the c-axis with a large dipolar matrix element of $\mu_z^{ex} \equiv g\mu_B|\langle 0|J_z|1\rangle| \sim 1.2$–$1.8\ \mu_B$.[5,6] The inelastic reflections are particularly strong at $Q = (1,0,0)$ and $(1,0.4,0)$, where the dispersion curve takes minima with gap energies of $\Delta_{(1,0,0)} \sim 2.0$ meV and $\Delta_{(1,0.4,0)} \sim 4.1$ meV. In the paramagnetic state, on the other hand, the system shows a broad continuous spectrum of antiferromagnetically correlated spin fluctuations. The fluctuating moment is large, comparable to μ_z^{ex}, and the corresponding staggered susceptibility diverges as the system approaches the phase transition.[16] However, the quasi-elastic peak seen just above 17.5 K does not grow into an adequate Bragg peak, but turns into the above magnon spectrum.

To explain the unusual features of the phase transition, various scenarios have been proposed, which can be classified into two groups: (i) the transition is uniquely caused by magnetic dipoles with highly reduced g-values;[17-19] and (ii) there is hidden order of a non-dipolar degree of freedom.[20-27] Staggered quadrupolar

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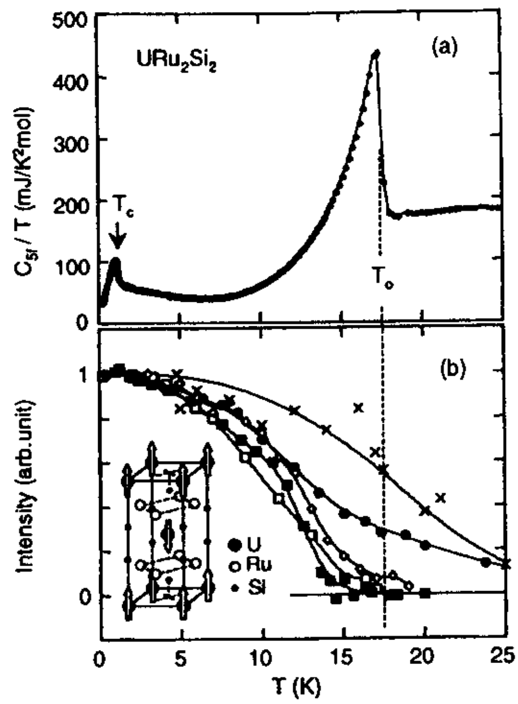

Fig. 1. (a) The 5f-electronic specific heat C_{5f}/T of a polycrystal of URu_2Si_2 annealed at 800 °C for one month. Non-5f contribution was subtracted by using the data of $ThRu_2Si_2$. Presence of two phase transitions are clearly indicated by jumps in the C_{5f}/T curve. Transition temperatures are evaluated as $T_c \sim 1.1$ K and $T_o \sim 17.5$ K, if they are defined by the midpoints of the jumps. It is known from previous macroscopic measurements that T_o is hardly affected by sample quality and by annealing conditions. The lowest value of T_o is reported to be ~ 17 K[2] for a polycrystalline sample. (b) Temperature variations of integrated intensity of the type-I antiferromagnetic Bragg peak for a high-purity single crystal (□) from Mason *et al.*,[4] and for single crystals annealed at 950–1000°C for 7–8 days (●,◇,■) from Broholm *et al.*,[5] Pâk *et al.*,[24] and Honma *et al.*,[48] respectively. Temperature variations of the μSR relaxation rate (×) for a polycrysatalline sample[49] are also shown. The lines are guides to the eye.

order is believed to be one of the most promising scenarios for the hidden order, which was first suggested by Miyako and his colleagues on the basis of non-linear susceptibility measurements.[20] After their definition, we use a notation T_o to express the transition temperature determined from macroscopic measurements. Three-spin order,[21] singlet orderings,[25] uranium-pair distortions[26] and d-type spin density waves[27] have also been argued. The models of the second group ascribe the observed tiny moment to side effects, such as secondary order, dynamical fluctuations and coincidental order of a parasitic phase. Each of the dipolar states may have its own energy scale, and to take account of this possibility we define T_m as the onset temperature of μ_o, distinguishing it from T_o.

To solve the problem, it is necessary to see how μ_o relates to the macroscopic anomalies. In contrast to the mean-field-like anomaly of the bulk properties at T_o, reported T variations of μ_o differ largely from each other, as typically illustrated in Fig. 1. A recent comparison of T_o and T_m for the same sample has revealed that T_m becomes lower than T_o in the absence of annealing.[28] In addition, careful high-field studies[16,29–31] have suggested that T_o and μ_o are not scaled by a unique function of field. Moreover, ^{29}Si-NMR measurements have never detected the internal fields caused by the development of μ_o.[32,33] These experimental results seem to support the hidden-order hypotheses.

To further investigate the relation between the tiny moments and the macroscopic anomalies, we have performed neutron scattering experiments under high pressure up to 2.8 GPa. Previous measurements of resistivity and specific heat in P up to 8 GPa have shown that the ordered phase is slightly stabilized by pressure, with a rate of $dT_o/dP \sim 1.3$ K/GPa.[34–39] We now show that pressure dramatically increases μ_o and causes a new phase transition which is accompanied by collapse of the magnetic excitations. We have previously made a brief report on the elastic scattering.[40] In this paper, we give the detailed description on the experiments and analyses, including the results of inelastic scattering.

§2. Sample and Experimental Technique

The measurements were made on a 8-mm-long approximately cylindrical single crystal with a diameter of 5mm, grown by the Czochralski technique in a tri-arc furnace in Osaka University. The crystal was annealed at 1000 °C for one week in a vacuum sealed quartz tube. Pressure was applied by means of a barrel-shaped piston cylinder device[41] at room temperature, which was then cooled in a ^{4}He cryostat for temperatures between 1.4 K and 300 K. A solution of Fluorinert 70 and 77 (Sumitomo 3M Co. Ltd., Tokyo) of equal ratio served as the quasihydrostatic pressure transmitting medium. The pressure was monitored by measuring the lattice constant of NaCl, which was encapsuled together with the sample.

The present neutron-scattering experiments were performed on the triple-axis spectrometer TAS-1 at the JRR-3M reactor of Japan Atomic Energy Research Institute. Vertically bent pyrolytic graphite PG(002) crystals were used as a monochromator and an analyzer. Elastic scans were made by using neutrons with a wavelength of $\lambda = 2.3551$ Å, and a 40'-80'-40'-80' horizontal collimation. Higher-order contamination was suppressed with double 4-cm-thick pyrolytic graphite (PG) filters as well as a 4-cm-thick Al_2O_3 filter. The scans were performed in the ($hk0$) scattering plane, particularly on the antiferromagnetic Bragg reflections (100) and (210), and on the nuclear ones (200), (020) and (110). For inelastic scattering measurements, we made constant-Q scans at Q = (1,0,0) and (1,0.4,0), using neutrons with a fixed incident energy of $E_0 = 14.7$ meV and a horizontal collimation of (open)-80'-40'-80'. The energy resolution, determined from the vanadium incoherent scattering, was ~ 0.95 meV (FWHM).

The lattice constant a of our sample at 1.4 K at ambient pressure is 4.13(1) Å, which is in good agreement with previous reports.[5,16] Radial scans at (200) and (110) show a crystal mosaic spread of $\sim 20'$ at $P = 0$, which increases to $\sim$ 40' by applying pressure.

§3. Experimental Results

In Fig. 2, we plot the pressure variations of elastic scans at 1.4 K along the $(1+\zeta,0,0)$ and $(1,\zeta,0)$ directions around the (100) antiferromagnetic Bragg peak. The instrumental background and the higher-order contributions of nuclear reflections were determined by scans at 35 K and subtracted from the data. The (100) reflection develops rapidly as pressure increases. No other peaks were found in a survey along the principle axes of the first Brillouin zone: $(\zeta,\zeta,0)$ and $(\zeta,1-\zeta,0)$ for $0.5 \le \zeta \le 1$ and $(1,\zeta,0)$ for $0 \le \zeta \le 1$. In addition, the intensities of (100) and (210) reflections follow the $|Q|$ dependence expected from the U^{4+} magnetic form factor[42] by taking the polarization factor unity. These ensure that the type-I antiferromagnetic structure at $P = 0$ is unchanged by the application of pressure.

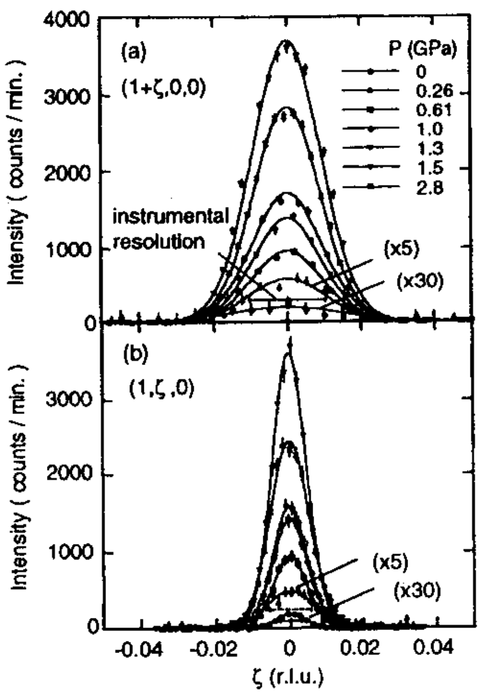

Fig. 2. Pressure variations of line shape at 1.4 K of the (100) antiferromagnetic response in URu_2Si_2 shown in scans along the $(1+\zeta,0,0)$ (a) and $(1,\zeta,0)$ (b) directions. Widths (FWHM) of the scans taken without filters are shown by horizontal bars as a measure of the instrumental resolution. The resolution is unchanged by applying pressure in the longitudinal scans, while broaden in the vertical scans because of an increase of the crystal mosaic (given by the broken line for $P > 0$).

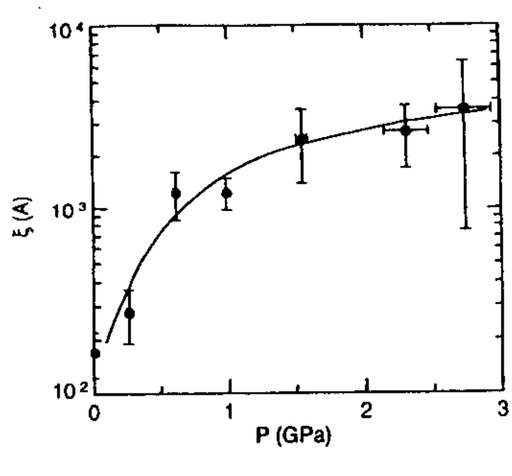

Fig. 3. Pressure dependence of the correlation length along the a^* direction obtained by fitting the (100) peaks with a Lorentzian function convoluted with a Gaussian resolution function.

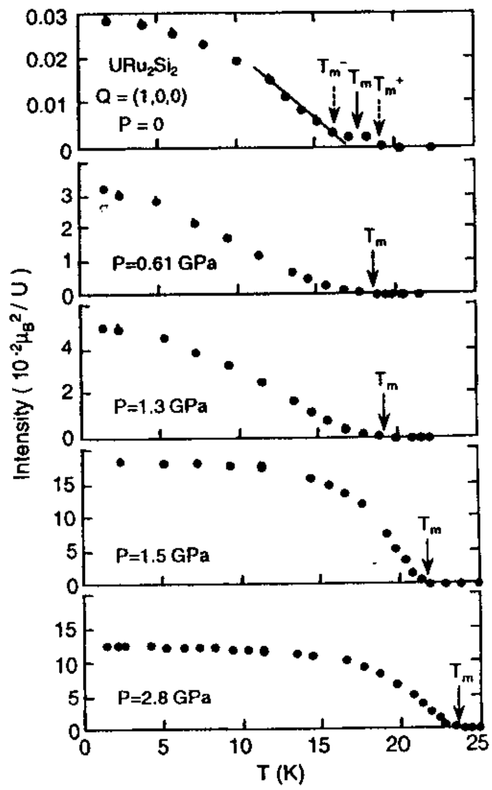

Fig. 4. Temperature dependence of integrated intensity of the (100) Bragg reflection of URu_2Si_2 for several pressures.

The widths (FWHM) of the (100) peaks for $P = 0$ and 0.26 GPa are significantly larger than the instrumental resolution ($\sim$ 0.021(1) reciprocal-lattice units for the a^* direction), which was measured by $\lambda/2$ scattering from the nuclear (200) Bragg reflection. From the best fit to the data by a Lorentzian function convoluted with the Gaussian resolution function, the correlation length ξ along the a^* direction is estimated to be about 180 Å at $P = 0$ and 280 Å at 0.26 GPa. For the higher pressures $P \ge 0.61$ GPa, the simple fits give $\xi > 10^3$ Å, indicating that the line shapes are resolution limited (Fig. 3).

The temperature dependence of the integrated intensity $I(T)$ at (100) varies significantly as P traverses 1.5 GPa ($\equiv P_c$) (Fig. 4). For $P < P_c$, the onset of $I(T)$ is

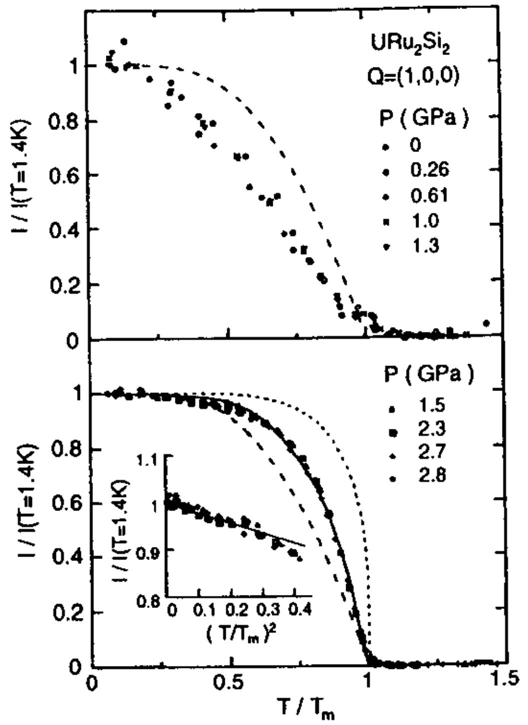

Fig. 5. Normalised intensities $I/I(1.4\mathrm{K})$ plotted as a function of T/T_m for $P < P_c$ (top) and $P > P_c$ (bottom). Theoretical calculations based on 2D,[50] 3D[43] and mean-field Ising models are also given by dotted, solid and broken lines. The inset plots $I/I(1.4\mathrm{K})$ versus $(T/T_m)^2$ at low temperatures. The thin line is a guide to the eye.

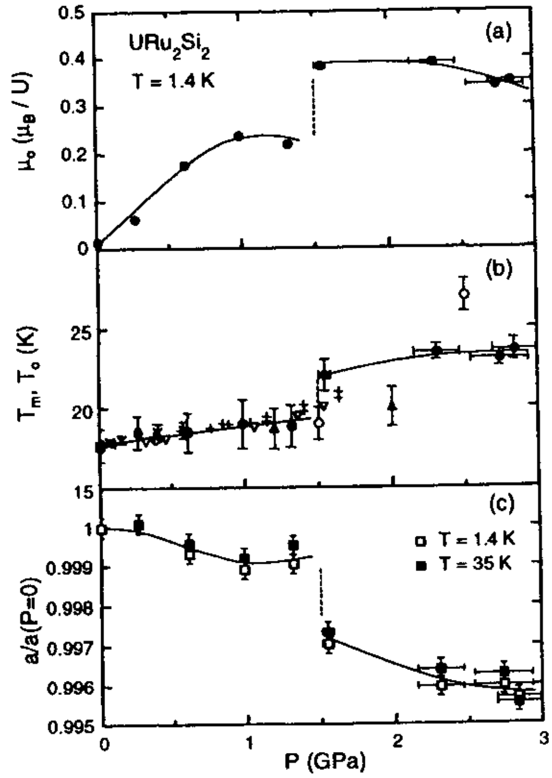

Fig. 6. Pressure variations of (a) staggered magnetic moment μ_o at 1.4 K; (b) the onset temperature T_m of the moment determined from this work (•) and the transition temperature T_o determined from resistivity (◇,[34] ▽,[35] △,[37] +,[38] o[39]) and specific heat (×[36]); and (c) the relative lattice constant $a(P)/a(0)$ at 1.4 K and 35 K. T_m is defined by $(T_m^+ + T_m^-)/2$ (see the text), and the range $\delta T_m (\equiv T_m^+ - T_m^-)$ is shown by using error bars. The lines are guides to the eye.

not sharp: $I(T)$ gradually develops at a temperature T_m^+, which is higher than T_o, and shows a T-linear behavior below a lower temperature T_m^-. Here we empirically define the "antiferromagnetic transition" temperature T_m by the midpoint of T_m^+ and T_m^-. The range of the rounding, $\delta T_m \equiv T_m^+ - T_m^-$, is estimated to be 2–3 K, which is too wide to be usual critical scattering. Above P_c, on the other hand, the transition becomes sharper ($\delta T_m <$ 2 K), accompanied by an abrupt increase in T_m at P_c.

If $I(T)$ is normalized to its value at 1.4 K, it scales with T/T_m for various pressures on each side of P_c (Fig. 5). This indicates that two homogeneously ordered phases are separated by a (probably first-order) phase transition at P_c. The growth of $I(T)$ for $P < P_c$ is much weaker than that expected for the mean-field Ising model, showing an unusually slow saturation of the staggered moment. On the other hand, the overall feature of $I(T)$ for $P > P_c$ is approximately described by a 3D-Ising model.[43] In the low temperature range $T/T_m < 0.5$, however, $I(T)$ rather follows a T^2 function (the inset of Fig. 5), indicating a presence of gapless collective excitations.[44]

In Fig. 6, we plot the pressure dependence of μ_o, T_m and the lattice constant a. The magnitude of μ_o at 1.4 K is obtained through the normalization of the integrated intensity at (100) with respect to the weak nuclear Bragg peak at (110). The variation of the (110) intensity with pressure is small ($< 5\%$) and independent of the crystal mosaic, so that the influence of extinction on this reference peak is negligible. μ_o at $P = 0$ is estimated to be about 0.017(3) μ_B, which is slightly smaller than the values ($\sim$ 0.02–0.04 μ_B) of previous studies,[5,6,8,28] probably because of a difference in the selection of reference peaks. As pressure is applied, μ_o increases linearly at a rate $\sim$ 0.25 μ_B/GPa, and shows a tendency to saturate at $P \sim$ 1.3 GPa. Around P_c, μ_o abruptly increases from 0.23 μ_B to 0.40 μ_B, and then slightly decreases.

In contrast to the strong variation of μ_o, T_m shows a slight increase from 17.7 K to 18.9 K, as P is increased from 0 to 1.3 GPa. A simple linear fit of T_m in this range yields a rate $\sim$ 1.0 K/GPa, which roughly follows the reported P-variations of T_o. Upon further compression, T_m jumps to 22 K at P_c, showing a spring of $\sim$ 2.8 K from a value ($\sim$ 19.2 K) extrapolated with the above fit. For $P > P_c$, T_m again gradually increases and reaches $\sim$ 23.5 K around 2.8 GPa. The pressure dependence of T_o in this range is less clear, and the few available data points deviate from the behavior of T_m, see Fig. 6(b).

The lattice constant a, which is determined from the scans at (200), decreases slightly under pressure

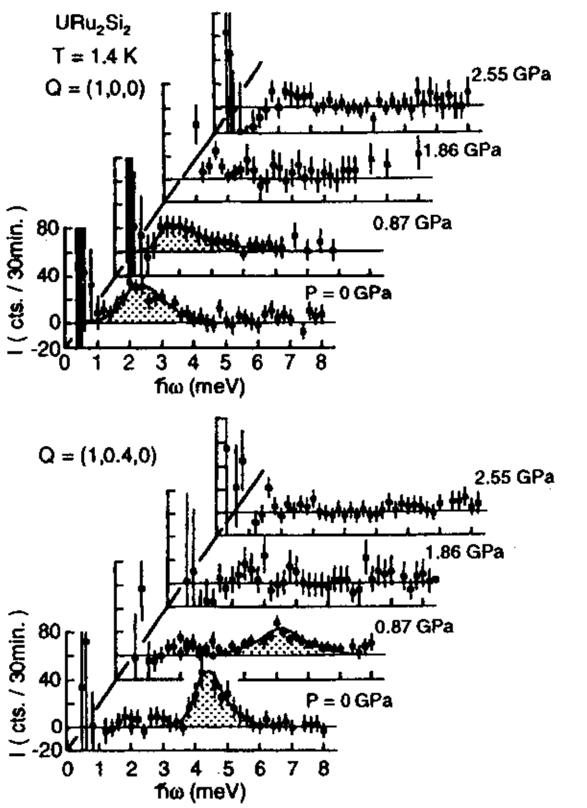

Fig. 7. Pressure variations of constant-Q scans at 1.4 K for $Q = (1,0,0)$ and $(1,0.4,0)$ of URu_2Si_2. Incoherent scattering and the instrumental background are subtracted. The lines and shades are guides to the eye.

(Fig. 6(c)). From a linear fit of a at 1.4 K for $P < P_c$, we derive $-\partial \ln a/\partial P \sim 6.7 \times 10^{-4}$ GPa^{-1}. If the compression is isotropic, this yields an isothermal compressibility κ_T of 2×10^{-3} GPa^{-1}, which is about 4 times smaller than what was previously estimated from the compressibilities of the constituent elements.[4)]

Around P_c, the lattice shrinks with a discontinuous change of $-\Delta a/a \sim 0.2$ %. Assuming again the isotropic compression, we evaluate $\Delta \ln V/\Delta\mu_o \sim -0.04\mu_B^{-1}$ and $\Delta \ln T_m/\Delta \ln V \sim -27$ associated with this transition. Note that a similar lattice anomaly at P_c is observed at 35 K, much higher than T_o. This implies that the system has another energy scale characteristic of the volume shrinkage in the paramagnetic region.

To check the possibility of a structural phase transition at P_c, we have carefully investigated the widths of the nuclear reflections in longitudinal and radial scans at two equivalent reciprocal points of (2,0,0) and (0,2,0). No significant broadening of the peaks is detected at P_c, allowing us to conclude that there is no lattice distortion lowering the fourfold symmetry within the detectability limit of $|a-b|/a \sim 0.05$ % and $\cos^{-1}(\widehat{a}\cdot\widehat{b}) \sim 2'$. The c axis is perpendicular to the scattering plane and cannot be measured in the present experimental configuration. Precise X-ray measurements under high pressure in an extended T-range are now in progress.

In Fig. 7, we plot the pressure variations of the energy scans at 1.4 K at (1,0,0) and (1,0.4,0), after subtracting the instrumental background including incoherent scattering from the pressure cell and the sample. The background was measured by scans at the corresponding $|Q|$-invariant positions (0.702,0.702,0) and (0.762,0.762,0), where the system is known to have neither magnon nor phonon excitations.[5)] At ambient pressure, the observed spectra show clear peak anomalies centering around $\Delta_{(1,0,0)} \sim 2.3$ meV and $\Delta_{(1,0,0)} \sim 4.3$ meV, which are in good agreement with the previous results.[5,16)] For $P \sim 0.9$ GPa, the peak intensity of the excitations is reduced by around half, but they still exist with roughly the same transfer energies. Above $P_c \sim 1.5$ GPa, on the other hand, we could observe no significant anomalies at least within the energy range of $\hbar\omega \leq 8$ meV. Large scattering amplitude at $\hbar\omega \sim 0$ for (1,0,0) arises mainly from the magnetic Bragg scattering and the $\lambda/2$ nuclear reflections. For (1,0.4,0), on the other hand, it is mainly due to the statistical error in the subtraction.

§4. Discussion

4.1 *A phenomenological analysis based on Landau's theory on the low-pressure ordered phase*

The remarkable contrast between μ_o and T_m below P_c offers a test to the various theoretical scenarios for the 17.5 K transition. Let us first examine the possibility of a single transition at T_m ($= T_o$) due to magnetic dipoles. In general, Landau theory with a magnetic free energy

$$F_m = -a(T_m - T)m^2 + bm^4 - g\mu_B mh, \qquad (1)$$

gives

$$m^2 = \frac{a}{2b}(T_m - T) \qquad (2)$$

and

$$\Delta C/T_m \sim (Nk_B/T_m)(m_o/m_{para})^2, \qquad (3)$$

where m and h denote the staggered dipole and field. Since T_m and ΔC are independent of g, their weak variations by pressure will be compatible with the ten-times enlargement of μ_o ($\equiv g\mu_B m_o$), *if only g is sensitive to pressure*. The existing theories along this line explain the reduction of g by assuming crystalline-electric-field (CEF) effects with low-lying singlets,[17)] and further by combining such with quantum spin fluctuations.[18,19)] For small g-values, the theories predict the relations, $g \sim \exp(-\Delta/2k_BT_m)$ and $g \sim T_m/T^*$, where Δ and T^* denote the gap energy between two singlets and a characteristic energy of a quasi particle band. The observed behavior will thus be reproduced, if Δ and T^* is strongly reduced by applying pressure. Previous macroscopic studies however suggest opposite tendencies: the resistivity maximum at around 73 K ($P = 0$), which is usually regarded as T^*, shifts to higher temperatures[35,37,39)] under pressure, and the low-T susceptibility, $\chi(0) \sim \Delta^{-1}$, decreases as P increases.[34)] The simple application of those models is thus unlikely to explain the behavior of μ_o with pressure.

On the other hand, the models that predict a hidden order due to a non-dipolar degree of freedom ψ could be consistent with the present observation. This category is further divided into two branches according to whether ψ is odd or even under time reversal, which we call (A) and (B) after the classification given by Shah et al.[45] Three-spin order[21] and d-type spin density waves[27] belong to the former, and quadrupolar order[20,22-24] and structural distortions[25,26] belong to the latter. We should notice that the polarized neutron scattering has confirmed that the reflections arise purely from magnetic dipoles.[7] For each branch, therefore, it has been proposed that the observed magnetic dipoles are induced by the order of ψ as secondary order.

The Landau free energy for type (A) theories is given as

$$F^{(A)} = -\alpha(T_o - T)\psi^2 + \beta\psi^4 + Am^2 - \eta m\psi, \qquad (4)$$

where α, β and A are positive, and the dimensionless order parameters m and ψ vary in the range $0 \leq m, \psi \leq 1$.[45] Minimization of $F^{(A)}$ with respect to m gives $m = -\delta\psi$, where $\delta \equiv \eta/2A$, showing that the staggered dipole moment appears in proportion to the primary order parameter. The stability condition for ψ then yields a mean-field solution, $\psi^2 \sim \frac{\alpha}{2\beta}(T'_o - T)$, where T'_o denotes the renomalized transition temperature given as $T'_o \sim T_o(1 + \kappa\delta^2)$. The coefficient κ is given as $A/\alpha T_o$, where A is originally written as $A = -a(T_m - T)$. Since $T_m \leq T_o$ and $\alpha \sim a \sim k_B$, κ is less than unity. Therefore, the shift of T_o due to the coupling between ψ and m is the order of δ^2. If the fluctuating dipole moment $\mu_{para} \sim 1.2\ \mu_B$ seen above T_o[16] corresponds to the maximum induced value of the secondary order parameter ($m \sim 1$), then the observed increase in μ_o gives the variation of δ ($\sim m_o \sim \mu_o/\mu_{para}$) from 0.014 ($P = 0$) to 0.21 ($P \sim P_c$). This results in $\mathrm{d}T_o/\mathrm{d}P \sim T_o \mathrm{d}m_o^2/\mathrm{d}P \sim 0.8$ K/GPa, which is in good agreement with the previous bulk measurements (~ 1.3 K/GPa).[34-39]

In type (B), on the other hand, the simplest free energy invariant under time-reversal[45,46] must take the form

$$\begin{aligned} F^{(B)} =& -\alpha(T_o - T)\psi^2 + \beta\psi^4 \\ &+ a(T_m - T)m^2 + bm^4 - \zeta m^2\psi^2. \end{aligned} \qquad (5)$$

Minimization of $F^{(B)}$ with respect to ψ gives $\psi^2 = \frac{\alpha}{2\beta}(T'_o - T)$, where $T'_o = T_o + \frac{\zeta}{\alpha}m^2$. Therefore, T_o is unchanged, so far as m is continuously induced by the primary order ($m = 0$ at $T_m \leq T_o$). The secondary order of this type influences the specific heat at T_m as $\Delta C/T_m \sim Nk_B m_o^2/T_m$. By the same arguments as in type (A), we obtain $\mathrm{d}(\Delta C/T_m)/\mathrm{d}P \sim 20$ mJ/K^{2}molGPa, when $T_m \sim T_o$. This cancels out with the P-increase in T_o, resulting in a roughly P-independent jump in $C(T)$. This is consistent with previous $C(T)$ studies up to 0.6 GPa,[36] in which $\Delta C_m/T_o$ is nearly constant, if entropy balance is considered. Note that in type (B) T_m can in principle differ from T_o, which could also be consistent with the annealing effects.[28]

4.2 *Quadrupolar and dipolar instabilities of the Γ_5 non-Kramers doublet*

The phenomenalogical consideration shown above supports the hidden order hypotheses for the ordered phase below P_c. We shall now argue more concretely that the staggered quadrupolar order due to the non-Kramers doublet Γ_5 fairly well explain the observed magnetic properties including the magnetic instability at P_c.

In our diluting experiments for $Th_{1-x}U_xRu_2Si_2$ ($x \leq 0.07$), we have observed a strong uniaxial magnetic anisotropy with the c-axis susceptibility diverging down to ~ 0.1 K.[23] This evidences a magnetically degenerate lowest state, and we have proposed the Γ_5 doublet to be the most promising CEF state to account for the observed anisotropy. The doublet is usually expressed as the form,

$$|\Gamma_5^{\pm}\rangle = \alpha|\pm 3\rangle + \beta|\mp 1\rangle, \qquad (6)$$

where $|\pm$ integer $\rangle$ denote the eigenfunctions for the z component of the total angular momentum of $J = 4$. $|\Gamma_5^{\pm}\rangle$ diagonalizes J_z with eigenvalues of $\pm(3\alpha^2 - \beta^2)$, but has no matrix elements for J_x and J_y. Therefore, Γ_5 always behaves as an Ising spin state for magnetic fields. Interestingly, Γ_5 may also behave as a quadrupolar doublet in response to strain fields of $\epsilon_{xx} - \epsilon_{yy}$ and ϵ_{xy}. One can see this by transforming the wave function (6) into

$$|\phi_1^{\pm}\rangle = \frac{1}{\sqrt{2}}(|\Gamma_5^{+}\rangle \pm |\Gamma_5^{-}\rangle) \qquad (7)$$

and

$$|\phi_2^{\pm}\rangle = \frac{1}{\sqrt{2}}(|\Gamma_5^{+}\rangle \pm i|\Gamma_5^{-}\rangle), \qquad (8)$$

which then diagonalize the quadrupolar operators $O_2^2 = J_x^2 - J_y^2$ and $P_{xy} = \frac{1}{2}(J_xJ_y + J_yJ_x)$ with eigenvalues of $\pm(6\sqrt{7}\alpha\beta + 10\beta^2)$ and $\pm(3\sqrt{7}\alpha\beta - 5\beta^2)$, respectively. Therefore, both the quadrupolar and dipolar orderings may occur in the system that has the Γ_5 lowest state. Note that these two instabilities compete with each other, since neither O_2^2 nor P_{xy} commutes with J_z. If the quadrupolar order takes place, then $|\phi_1^{\pm}\rangle$ or $|\phi_2^{\pm}\rangle$ splits into two quadrupolar singlets, shifting the dipolar matrix elements to off-diagonal. This is consistent with the feature of URu_2Si_2 that the phase transition at T_o is accompanied by the diverging staggered susceptibility and the subsequent development of longitudinal magnon excitations.[16] The presence of the quadrupolar instability has also been suggested by recent measurements of elastic constants.[47]

The relationship between the quadrupolar and dipolar degrees of freedom in Γ_5 can be described in terms of a pseudo-spin $S = \frac{1}{2}$, if its components S_x, S_y and S_z are mapped onto O_2^2, P_{xy} and J_z, respectively. The quadrupolar order may occur when the spin-spin interactions are of XY-type. Ohkawa and Shimizu have recently pointed out this equivalence, and applied an extended s-d model to the Γ_5 local state. They have found that dynamical spin susceptibility along the c axis shows a peak anomaly in the vicinity of $\omega = 0$ and $Q = (1,0,0)$ as a result of the staggered quadrupolar order.[24] From the calculations they attribute the tiny dipole moments to the dynamical spin fluctuations with a life time in between the time scales of static and neutron scattering measurements. This is supported by recent NMR measurements under pressure, in which the dipolar order has not been observed even for $P \sim 0.4$ GPa.[51]

Interestingly, they have further proposed the phase transition at P_c to be a switching in the order parameter between quadrupole and dipole in Γ_5. Opposite to the above situation, the dipolar order splits $|\Gamma_5^\pm\rangle$ into two singlets, which are coupled with each other only by O_2^2 or P_{xy}. Therefore, the magnon excitations should disappear in the dipolar ordered state of Γ_5. This is consistent with the present results of inelastic neutron scattering, at least, in the survey ranging up to 8 meV (Fig. 7). Note that electrical resistivity below T_o follows a gap-type function even for $P > P_c$,[37,39] evidencing the presence of excitations that can be detected by thermal probes.

§5. Conclusion

We have found URu_2Si_2 to undergo a new phase transition under high pressure, at $P_c \sim 1.5$ GPa. Below P_c, the sublattice magnetization associated with the 17.5 K transition is strongly enhanced, where the unusual T-linear behavior is conserved. In contrast to the ten-times increase in the saturation moment, however, the transition temperature rises only ten percent with pressure. These results for $P < P_c$ could indicate that the tiny dipole moments are intrinsically induced by the 17.5 K transition, but that they are not the primary order parameter. A simple analysis based on the Landau's theory supports the hidden (non-dipolar) order scenarios. Above P_c, on the other hand, the T variations of the staggered moment are well described by a 3D-Ising model, where sharp magnon excitations below P_c disappear. We have shown that the magnetic instabilities at P_c as well as the exotic phase below P_c could be ascribed to the competitions between the quadrupolar and dipolar degrees of freedom in the Γ_5 non-Kramers doublet. In this context the low-pressure phase will be described as a staggered quadrupolar ordered state concomitant with dynamical spin fluctuations.[24] To obtain a conclusive proof, we are making the resonant X-ray scattering and the NMR measurements in a wide P-range.

Acknowledgements

One of us (H.A.) is indebted to F.J. Ohkawa for helpful discussions. This work was partly supported by the JAERI-JRR3M Collaborative Research Program, and by Grant-in-Aid for Scientific Research from Ministry of Education, Science, Sports and Culture of Japan.

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8

July 28, 2000 Trieste

Hidden Order in URu_2Si_2 Studied by High-Pressure Experiments

Hokkaido University , Sapporo, JAPAN
Hiroshi Amitsuka

Backgrounds

5f Systems

The "17.5K Transition" of URu_2Si_2

Neutron Scat. Exper. under High-P

Ohkawa Model

Advances and Prospects

Collaborators

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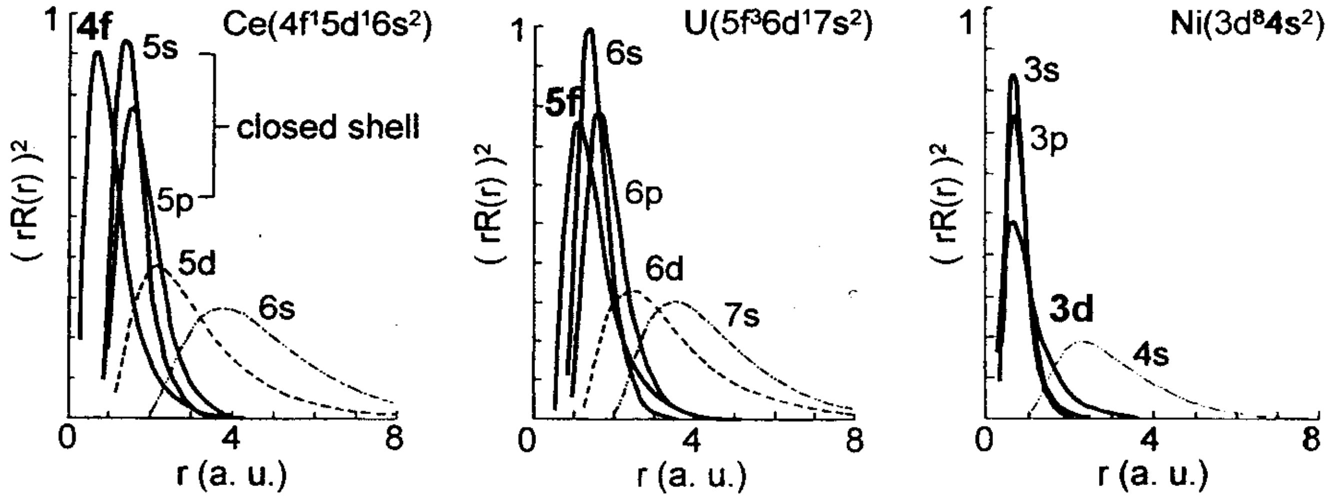
Tata Institute of Fundamental Research, Bombay, India

J.A. Mydosh
Kamarlingh Onnes Laboratory, Leiden Univ. The Netherlands

5f States

Ionic : $\langle r \rangle_{4f} < \langle r \rangle_{5f} < \langle r \rangle_{3d}$



Periodic

4f

Intra-Atomic
Many-Body States
+
c-f Hybridization

5f

?

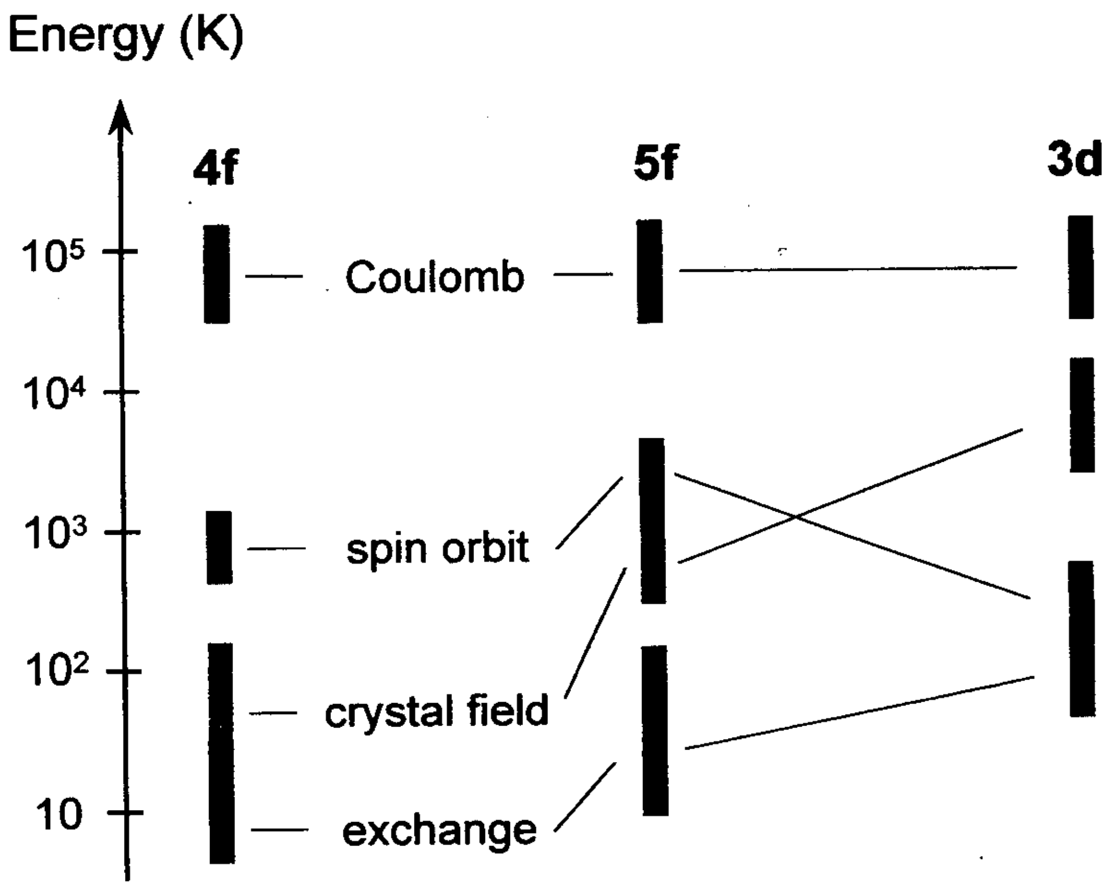
3d

Bands
+
Spin Fluctuations

T. Kasuya JPSJ (Japanese ed.)42(1987)722

5f States

Standard Energy Scale



Handbook on the Physics and Chemistry of the Actinides,
edited by A.J. Freeman and G.H. Lander,
Elsevier Sci. Pub., Chapter 2, 1985, p. 34

Topics in the Strongly Correlated 5f Electron Systems

Unconventional Superconductivity

UPt_3, UPd_2Al_3, URu_2Si_2, UBe_{13}

Tiny or Small Moment Magnetism

UPt_3, URu_2Si_2, $\longrightarrow$ μ_{ord} $\sim 10^{-2}\ \mu_B$

UPd_2Al_3, UNi_2Al_3, UIr_2Si_2 $\longrightarrow$ $\sim 10^{-1}\ \mu_B$

Non-Dipolar Order?

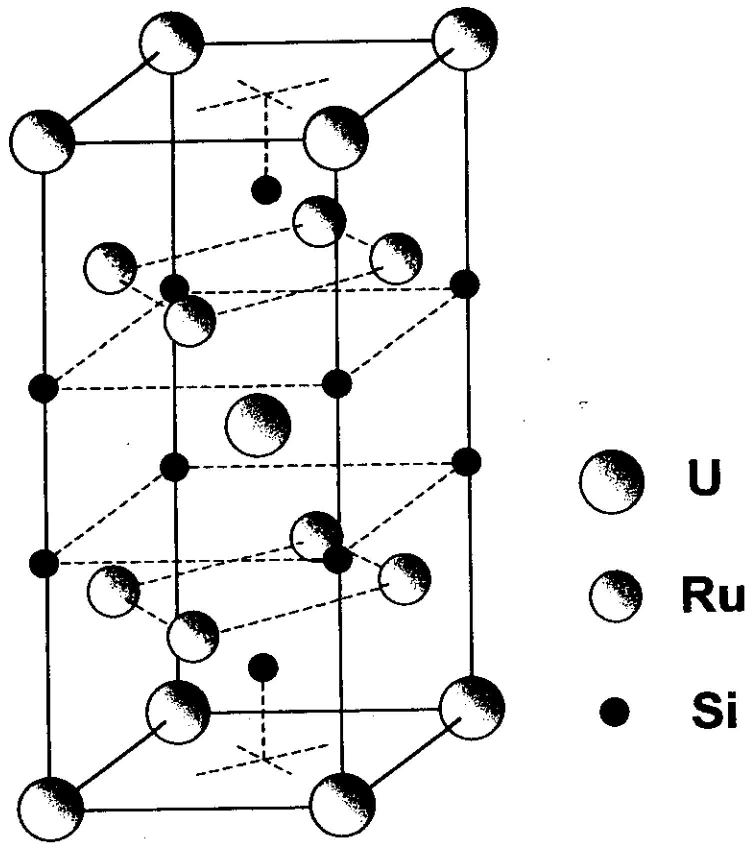
UPd_3, URu_2Si_2, UCu_2Sn Quadrupoles ?

Quadrupolar Kondo Effects and NFL?

(Y, U)Pd_3, (Th, U)Be_{13}, (Th, U)Ru_2Si_2

an Exotic Local c-f Interaction ?

URu_2Si_2

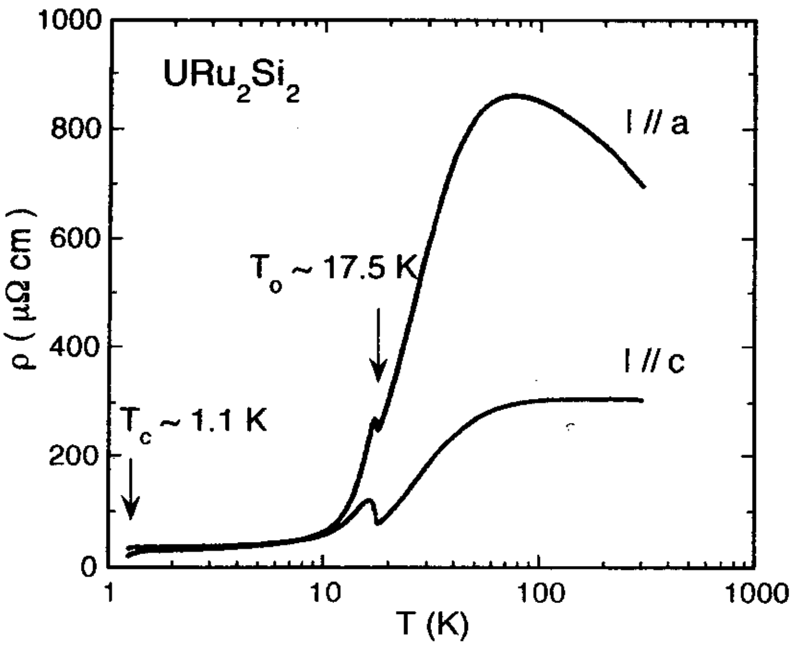


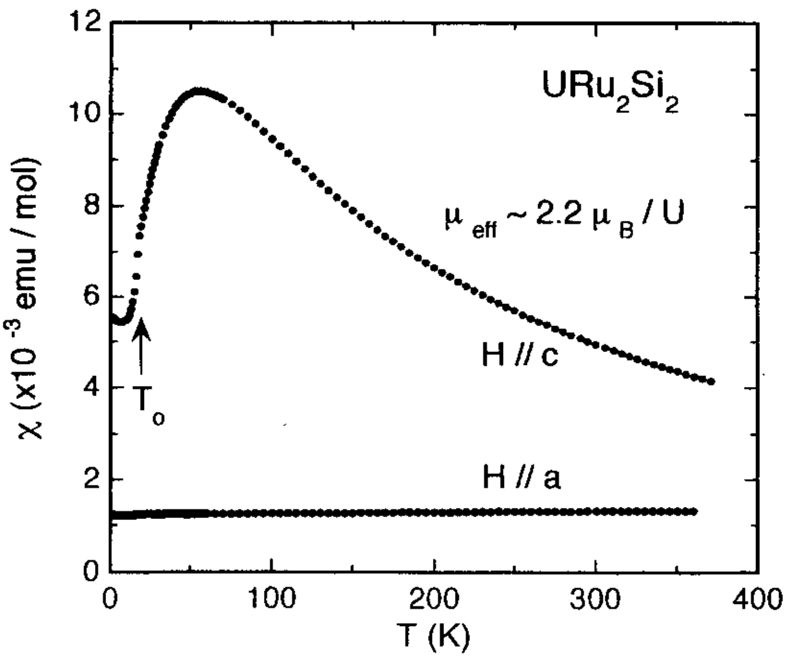
$ThCr_2Si_2$ structure (I4/mmm)

	a (Å)	c (Å)	V (Å^3)
URu_2Si_2	4.127	9.570	163

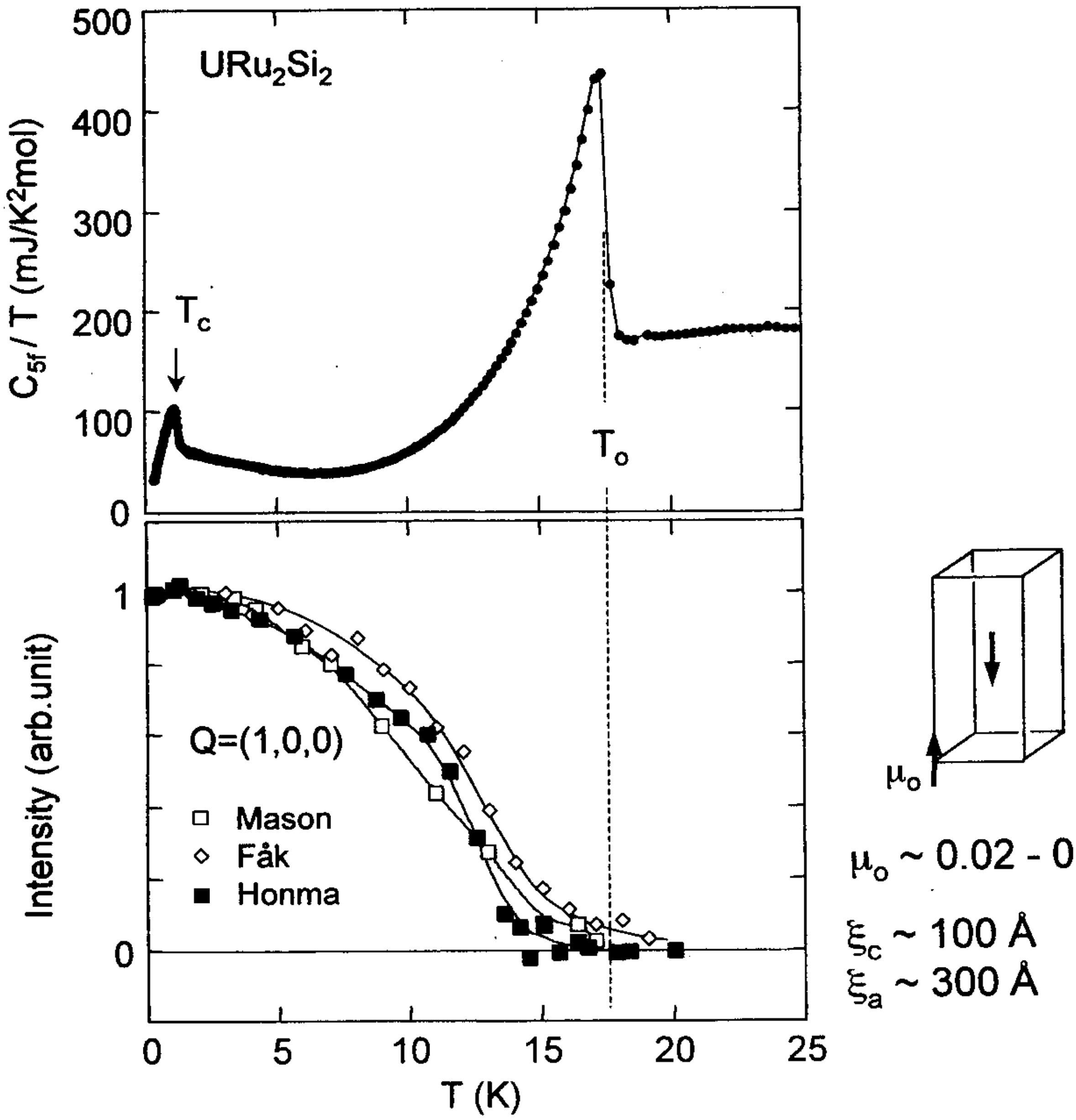
ρ & χ

T.T.M. Palstra et al., PRL55(1985)2727
W. Schlabitz et al., Z. Phys. B62(1986)171
M.B. Maple et al., PRL56(1986)185





C/T & Tiny-Moment AF State

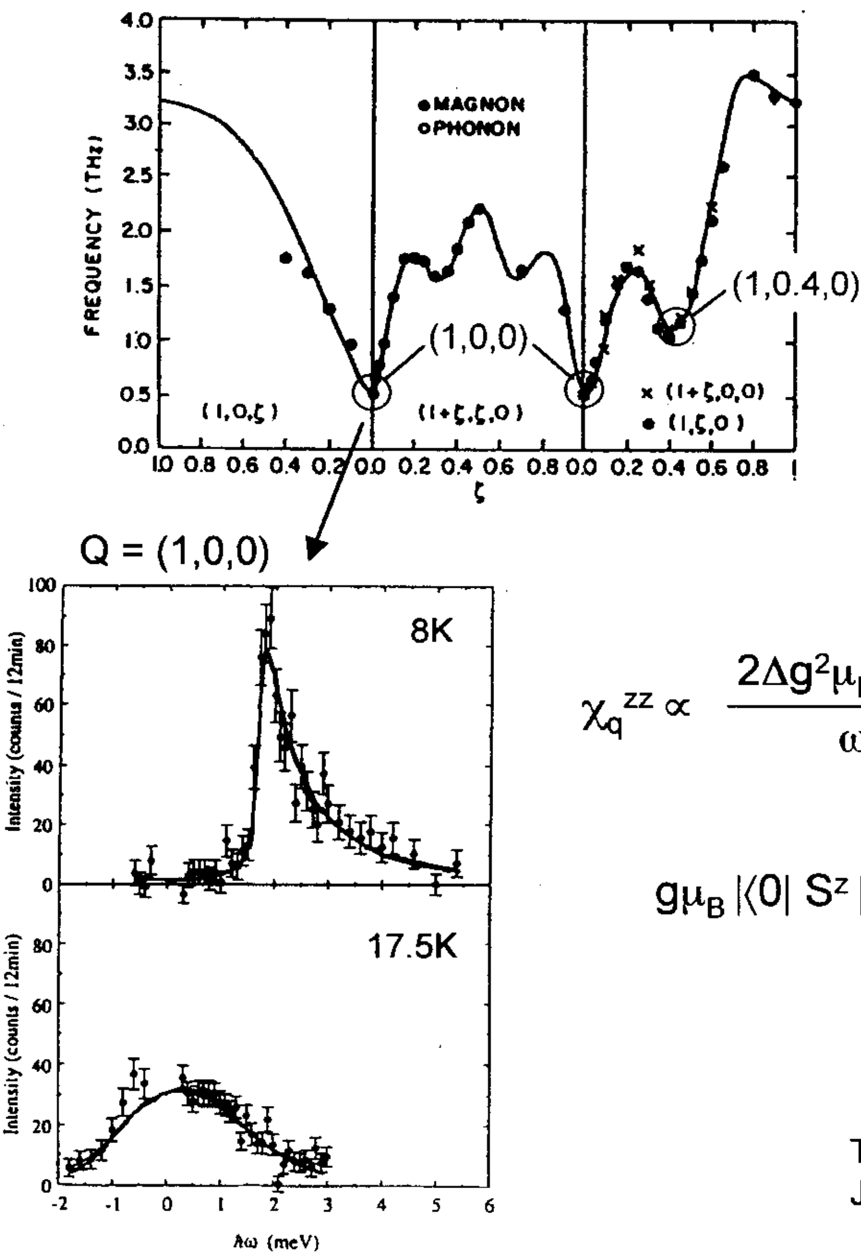


$\mu_o \sim 0.02 - 0.04\ \mu_B$

$\xi_c \sim 100$ Å

$\xi_a \sim 300$ Å

Magnon excitations



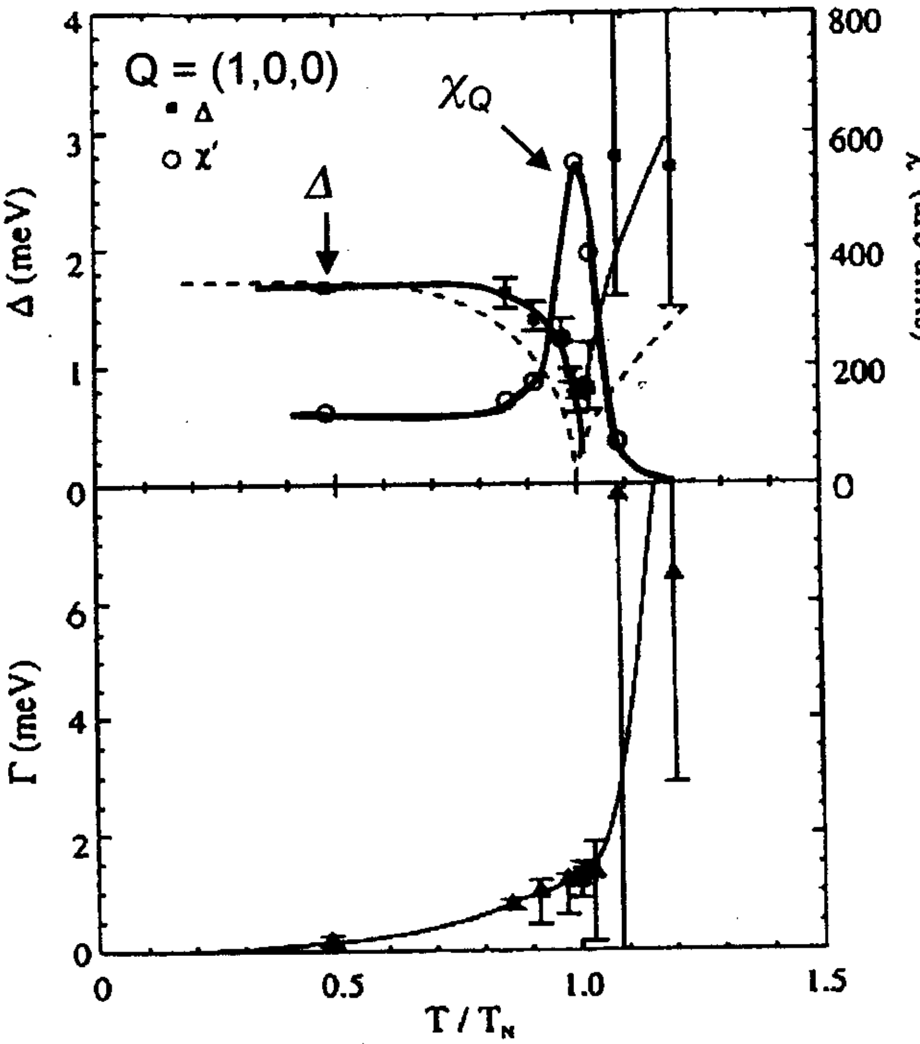
C. Broholm et al.
PRB43(1991)12809

$$\chi_q^{zz} \propto \frac{2\Delta g^2 \mu_B^2 |\langle 0| S^z |1\rangle|^2}{\omega_q^2 - \omega^2}$$

$$g\mu_B |\langle 0| S^z |1\rangle| \sim 1.2\ \mu_B$$

T.E. Mason et al.
J. Phys. 7(1995)5089

χ_Q



T.E. Mason et al.
J. Phys. 7(1995)5089

If f electrons are well localized ·····

$$\frac{\Delta C}{T_N} \sim \frac{Nk_B}{T_N}\left(\frac{\mu_o}{\mu_{para}}\right)^2$$

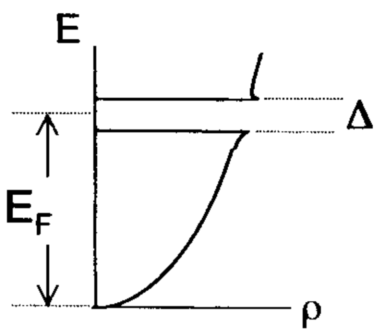
$$\leftarrow \begin{cases} \mu_o \sim 0.03\ \mu_B \\ \mu_{para} \sim 1.2\ \mu_B \end{cases}$$

$\sim$ 0.3 mJ/K²mol

by experiments ··· $\updownarrow$

300 mJ/K²mol

If they are itinerant ·····



$$\mu_o \sim \mu_B \times \left(\frac{\Delta}{E_F}\right) \sim 0.03\ \mu_B$$

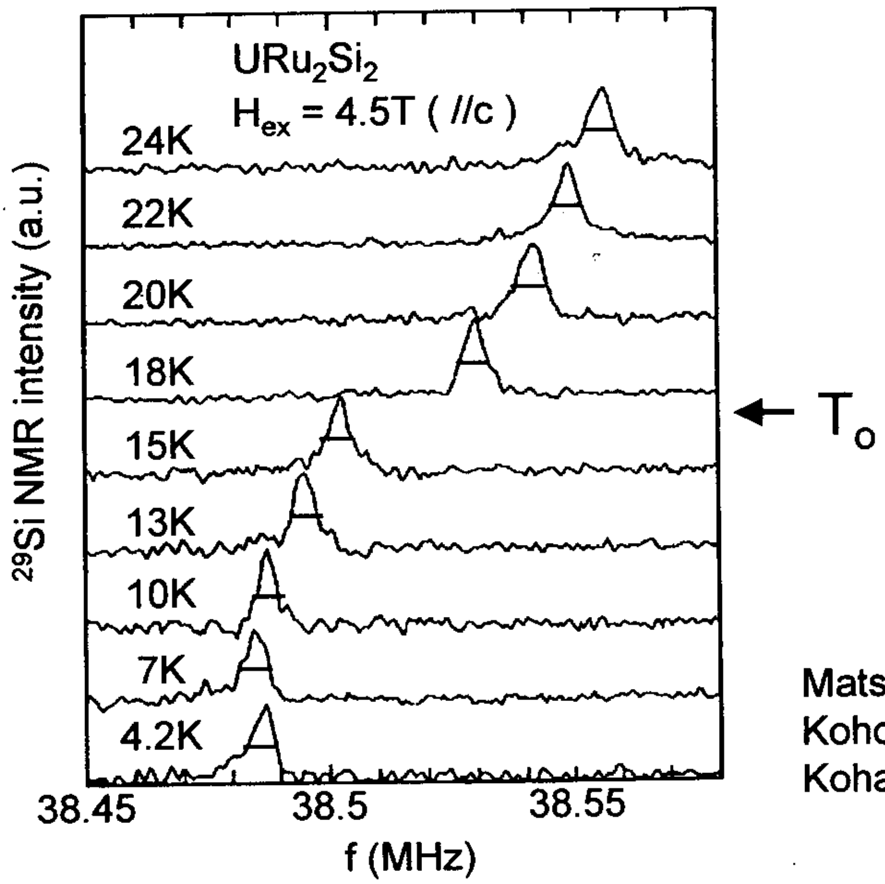
$$S_{mag} \sim R\ \ln 2 \times \left(\frac{\Delta}{E_F}\right) \sim 0.03R\ \ln 2$$

by experiments ··· $\updownarrow$

$\sim$ 0.2R ln2

^{29}Si NMR

Absence of internal fields below T_o



Matsuda,
Kohori,
Kohara

Are the tiny moments quasi-static fluctuations?

$$10^{-5} < \tau(s) < 10^{-10}$$

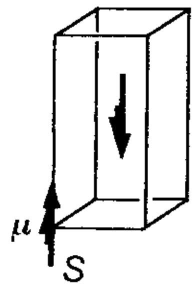
Proposed Scenarios for the Order Parameter

Magnetic Dipoles ?

Small g-factor

Crystal Fields?

Quantum Spin Fluctuations



Niewenhuys '8?
Sikkema '96
Okuno, Miyake
Yamagami '99

Intrinsic (static)

Hidden OP ?

Quadrupole	*Miyako '91* *Santini, Amoretti '94* *Amitsuka '94* *Ohkawa '99* *Tsuruta '00*
U-pair	*Kasuya '97*
d-wave SDW	*Ikeda, Ohashi '98*
Valece+Str.	*Barzykin, Gor'kov*

tiny moment

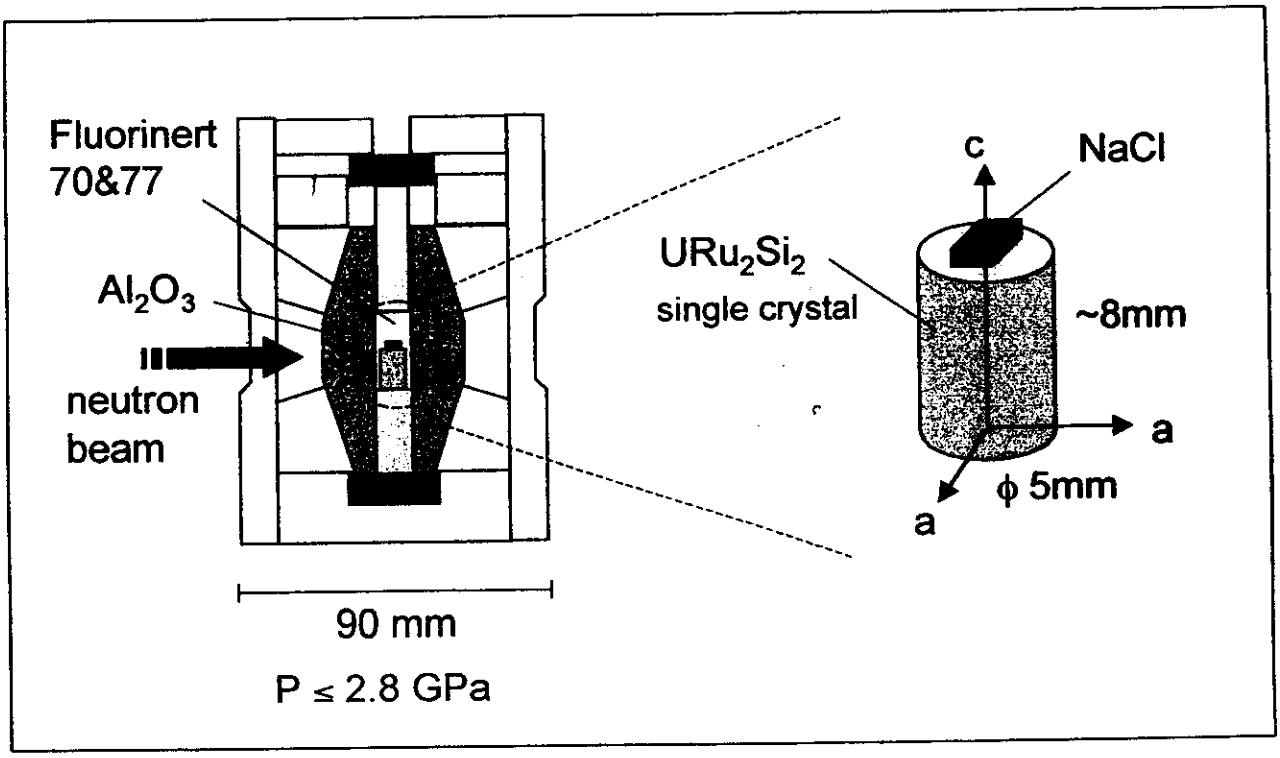
μ_o

Side effects

fluctuations?

a parasitic phase?

Neutron Scattering Experiments under High Pressure

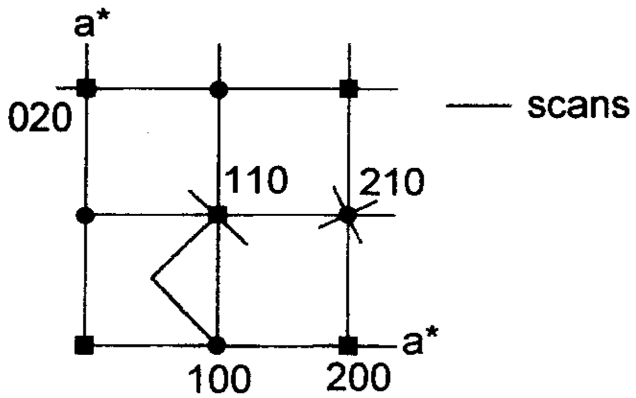


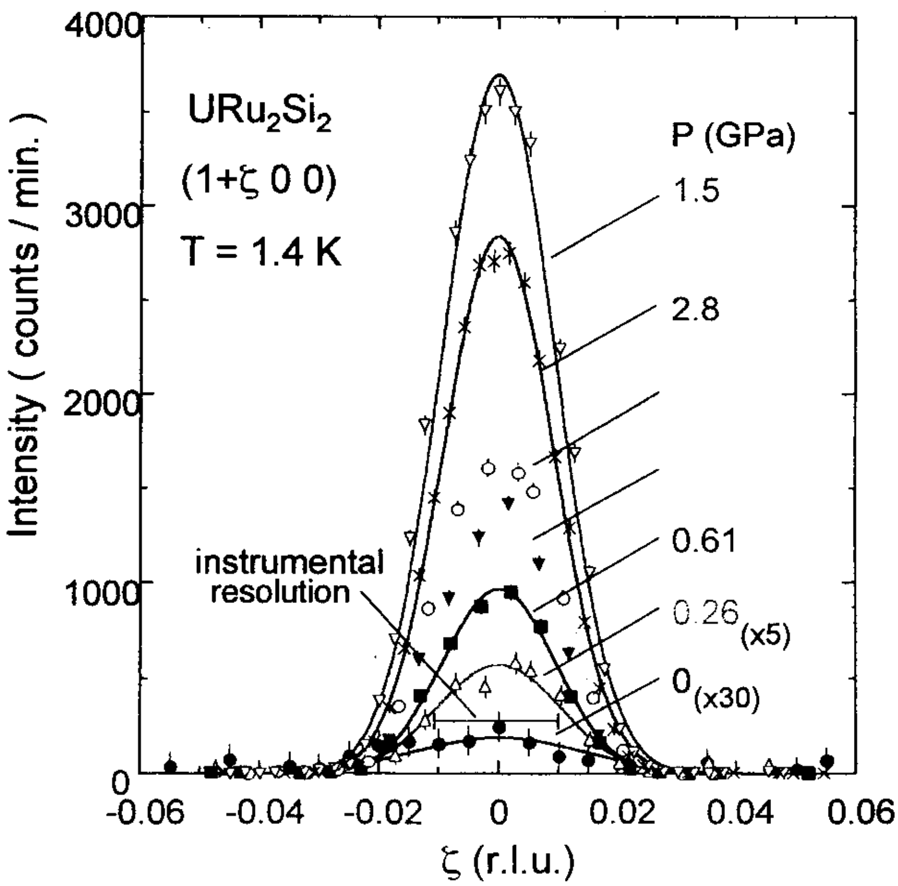
TAS-1 (JRR-3M, JAERI)

Elastic: $k_i = k_f = 2.6679\ A^{-1}$
40' – 80' – 40' – 80' (Al_2O_3 & PG × 2 filters)

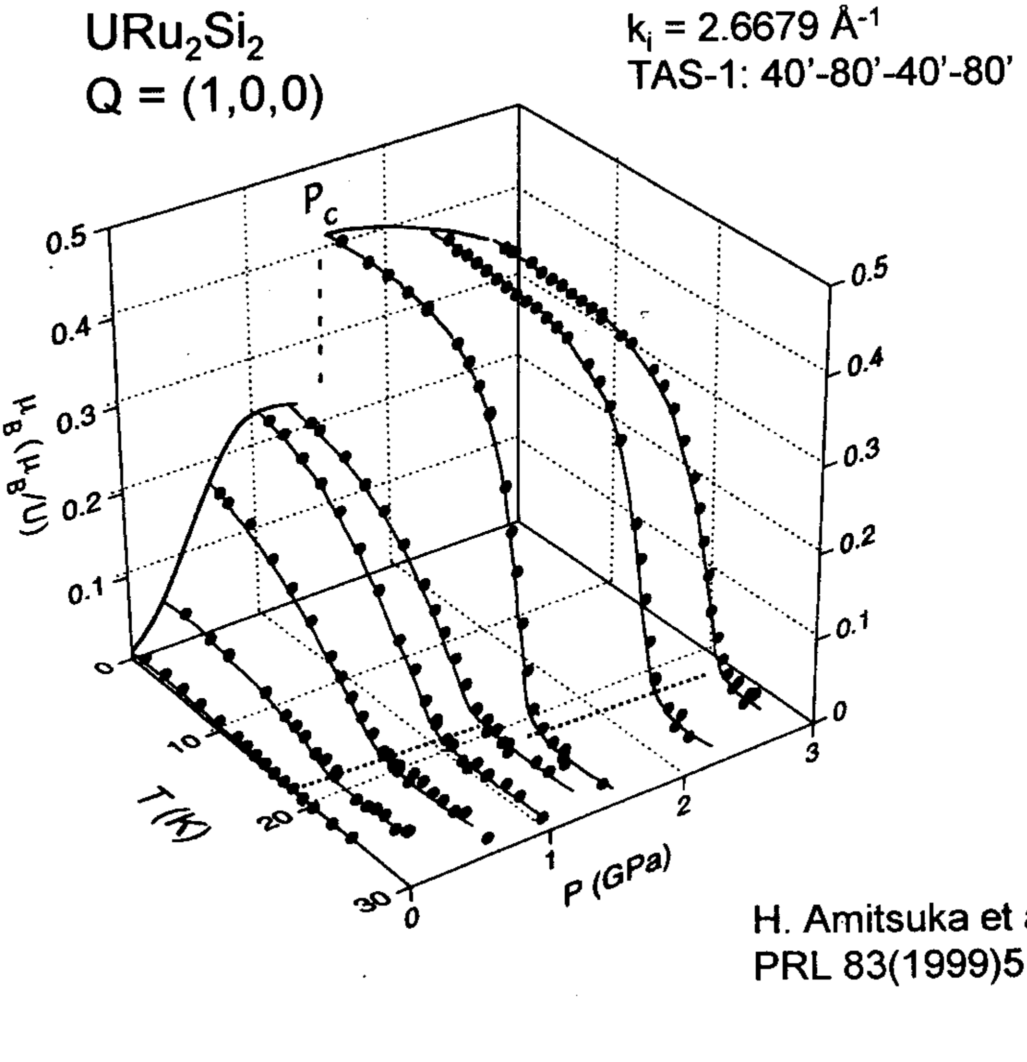
Inelastic: $k_i = 2.6635\ A^{-1}$ fixed
B – 80' – 40' – 80' (Al_2O_3 & PG × 1 filters)

Elastic scattering



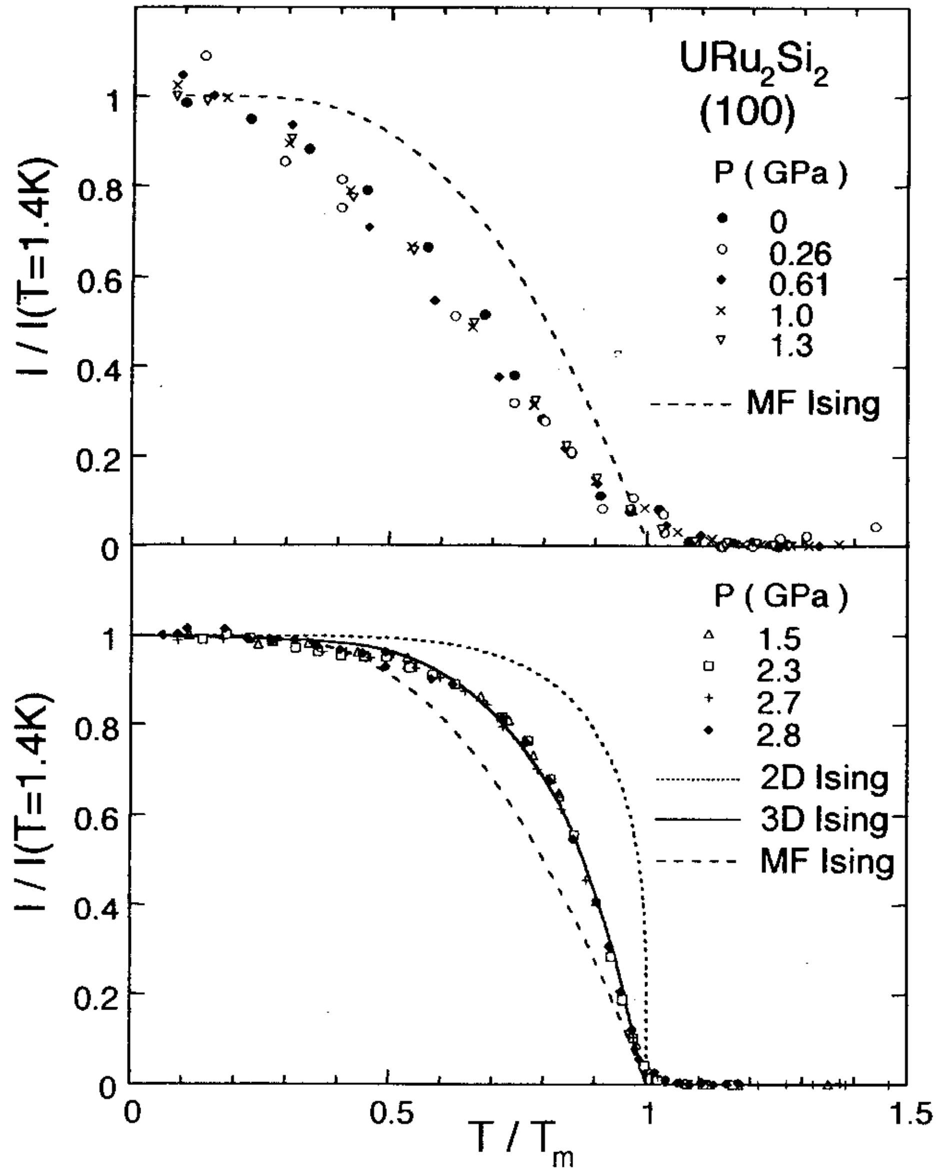


μ_o (P, T)



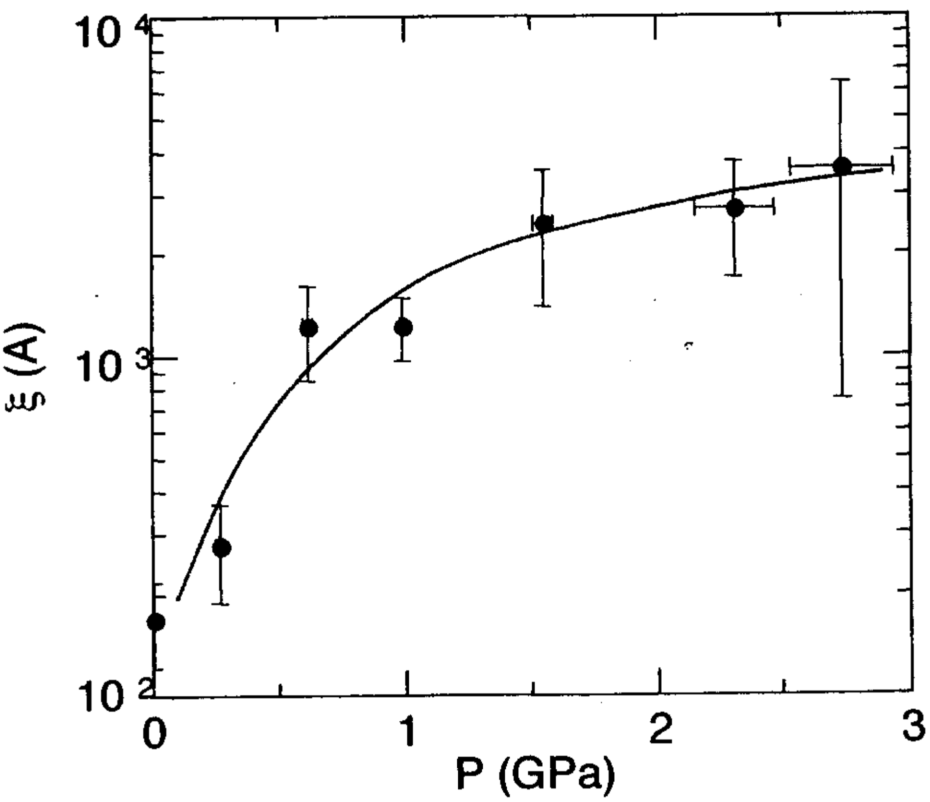
H. Amitsuka et al.
PRL 83(1999)5114

I / I_o vs T / T_m

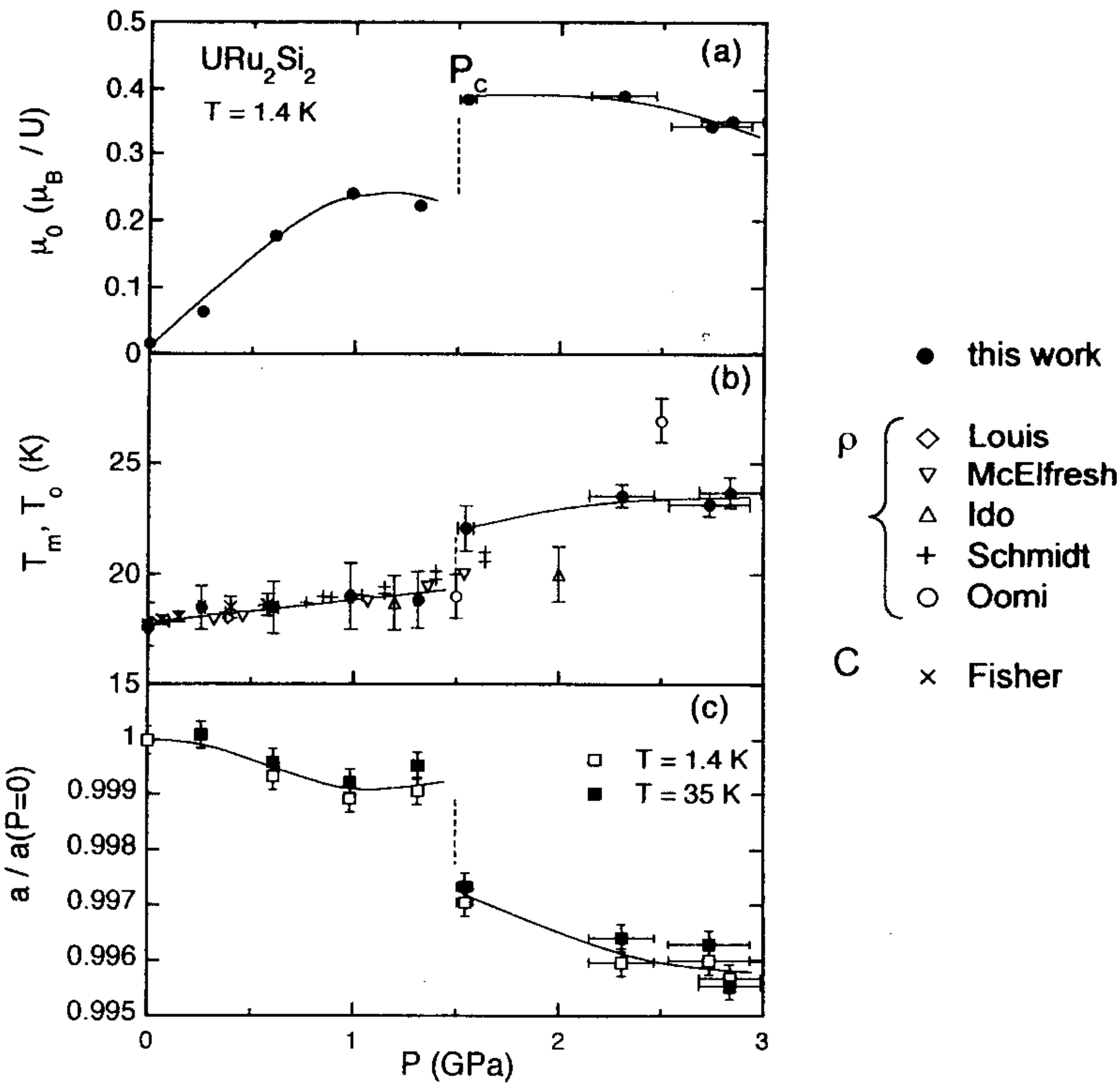


URu2Si2
(100)
P (GPa)
0
0.26
0.61
1.0
1.3
MF Ising
P (GPa)
1.5
2.3
2.7
2.8
2D Ising
3D Ising
MF Ising
I / I(T=1.4K)
I / I(T=1.4K)
T / Tm
0
0.5
1
1.5
0
0.2
0.4
0.6
0.8
1

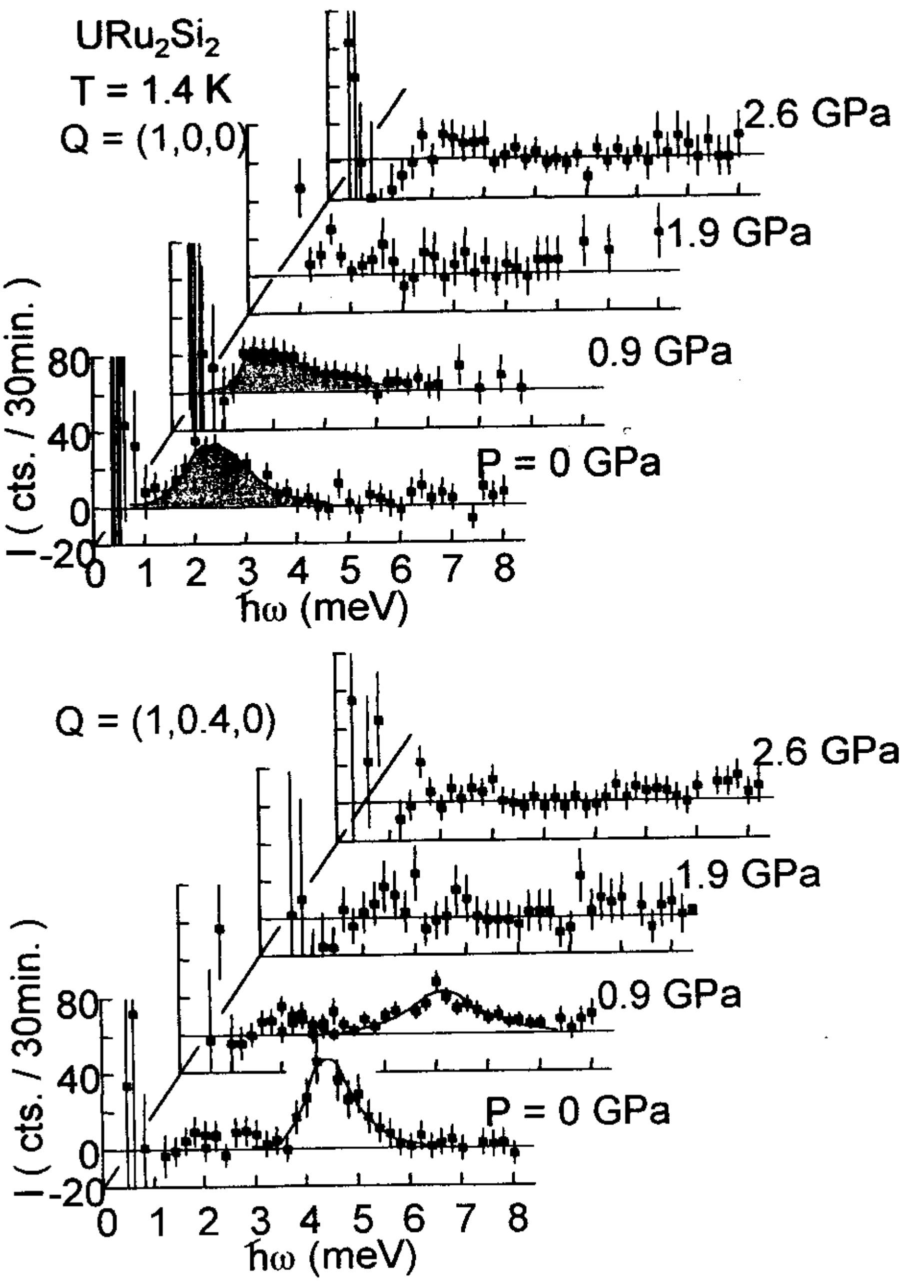
Coherence length



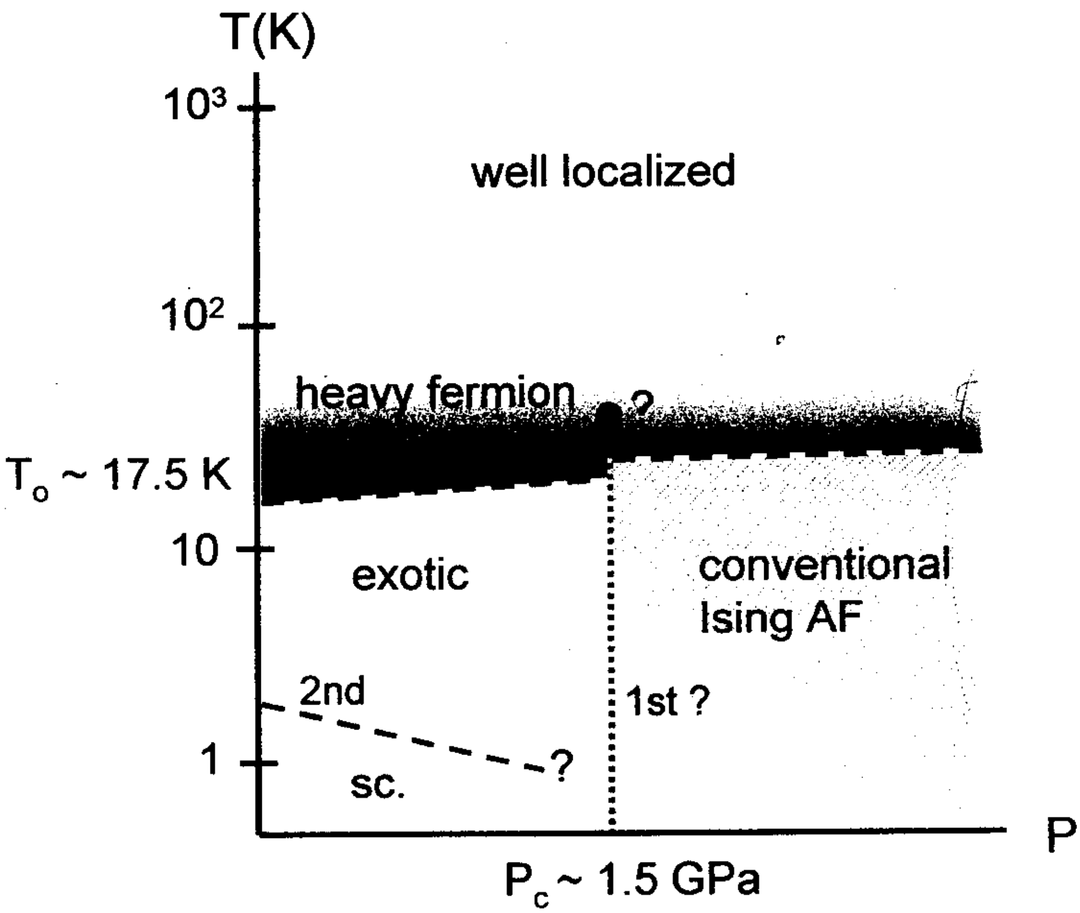
P variations of μ_o, T_m, T_o, and the lattice parameter *a*



Inelastic scattering

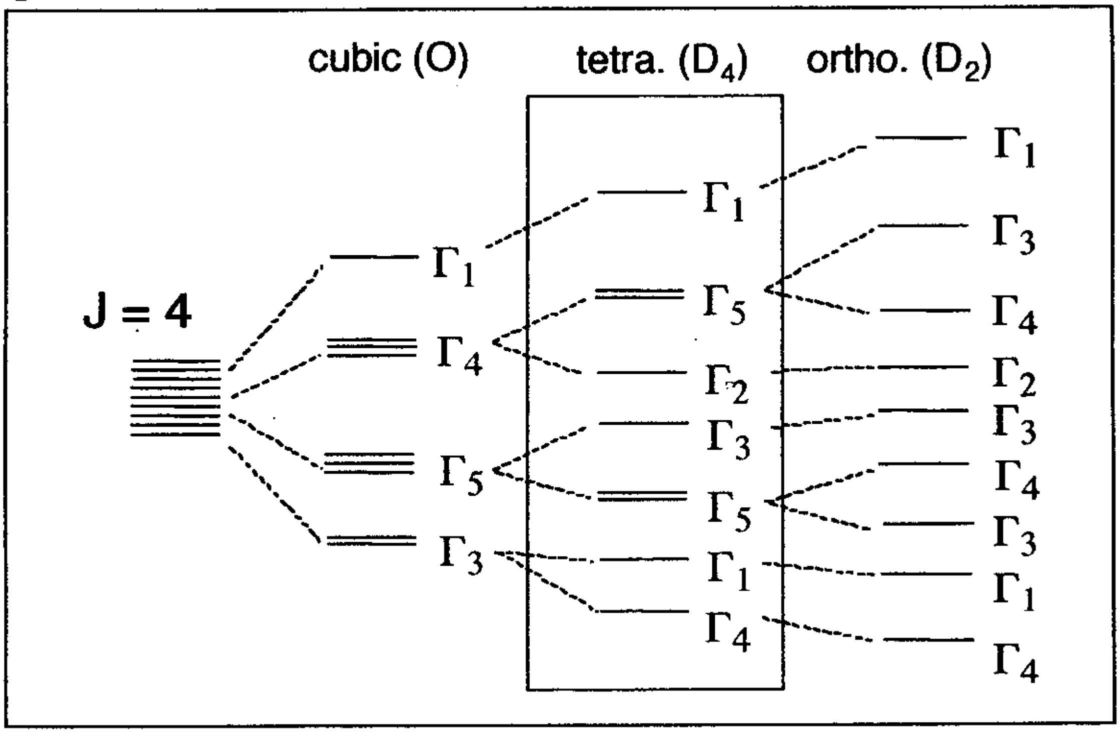


P-T phase diagram



CEF branching in $f^2(U^{4+})$ configuration

In general....

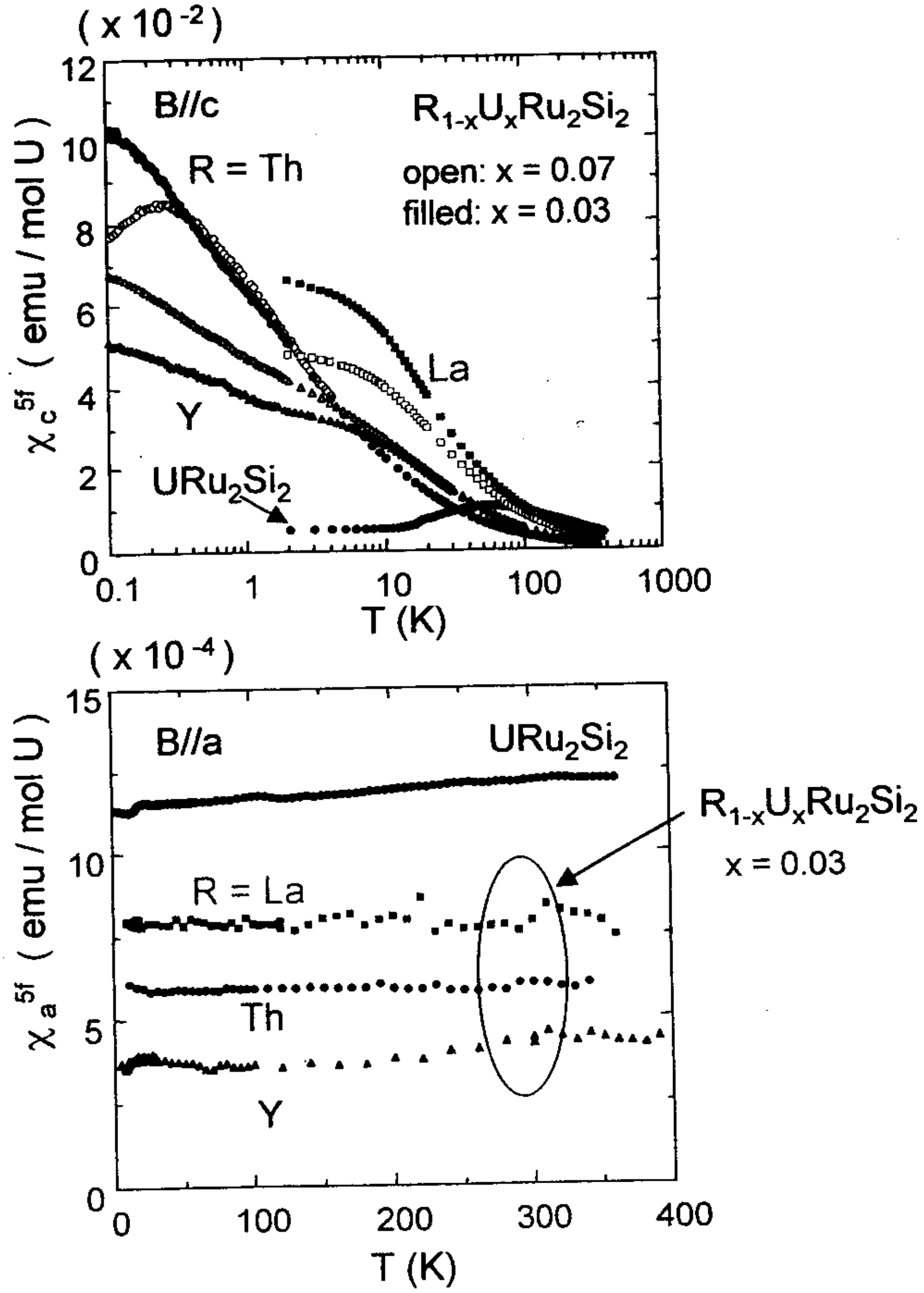


Is the CEF lowest state of URu_2Si_2 ...

, or ?

singlet	doublet
Γ_1, Γ_2, etc.	Γ_5

$\chi(T)$ in the dilute U limit of URu_2Si_2



Γ_5 non-Kramers doublet

Heavy Electron State at Low-T

$\chi_c \to \infty$ (T $\to$ 0) in the dilute U limit

Strong Uniaxial Magnetic Ansiotropy

$$|\Gamma_5 \pm\rangle = \cos\alpha|\pm 3\rangle + \sin\alpha|\mp 1\rangle = \begin{cases} |\uparrow\rangle \\ |\downarrow\rangle \end{cases}$$

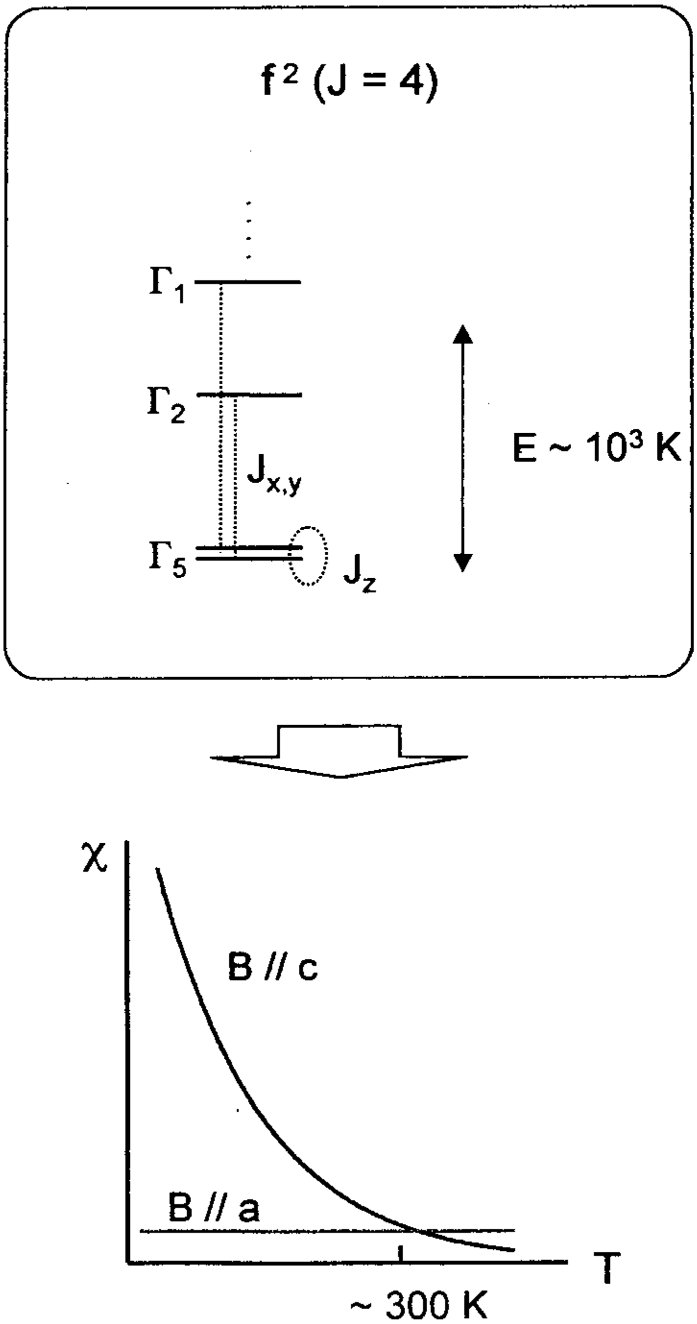
$$J_z \longrightarrow \begin{array}{cc} |\uparrow\rangle & |\downarrow\rangle \end{array}$$
$$\begin{pmatrix} +g_z & \\ & -g_z \end{pmatrix}$$

$$g_z = g_J\,(3\cos^2\alpha + \sin^2\alpha)$$

$$J_{x,y} \longrightarrow \mathbb{O}$$

$$g_x = g_y = 0$$

A promising CEF scheme with the Γ_5 lowest state



Ohkawa Model

(**An extended periodic s-d model with the Γ_5 local state**)

F.J. Ohkawa and H. Shimizu
J. Phys. 11(1999)L519

$$\begin{array}{c} \\ \\ \end{array}\quad \begin{array}{cc} |\uparrow\rangle & |\downarrow\rangle \end{array}$$

$$O_z = J_z \longrightarrow \begin{pmatrix} g_z & \\ & -g_z \end{pmatrix} \propto S_z$$

$$O_{x2\text{-}y2} = \frac{1}{2}(J_+^2 + J_-^2) \rightarrow \begin{pmatrix} & q_1 \\ q_1 & \end{pmatrix} \propto S_x$$

$$O_{xy} = \frac{-i}{2}(J_+^2 - J_-^2) \rightarrow \begin{pmatrix} & -iq_2 \\ iq_2 & \end{pmatrix} \propto S_y$$

$S = \frac{1}{2}$ **pseudo spin**

$g_x = g_y = 0$
$g_z \neq 0$

$$H_{\text{multi}} = \frac{1}{2}\sum_{\Gamma}\sum_{i,j} A_{\Gamma;\,ij}\, O_{\Gamma;\,i} O_{\Gamma;\,j}$$

$$\Downarrow$$

$$H_{\text{spin}} = \frac{1}{2}\sum_{\lambda=x,y,z}\sum_{i,j} J_{\lambda;\,ij}\, S_{\lambda;\,i} S_{\lambda;\,j}$$

$$H_{\text{eff}} = H_c + H_{sd} + H_{\text{spin}} \; (+ H_{\text{Zeeman}})$$

Pressure Induced Magnetic Transition

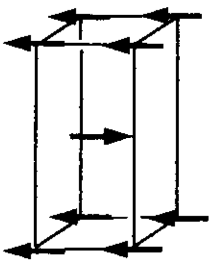
$P < P_c$

XY

$(|J_x| = |J_y| > |J_z|)$

$\langle S_z \rangle = 0$

$\sqrt{\langle S_x \rangle^2 + \langle S_y \rangle^2} \neq 0$



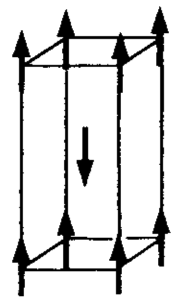
$P > P_c$

Ising

$(|J_x| = |J_y| < |J_z|)$

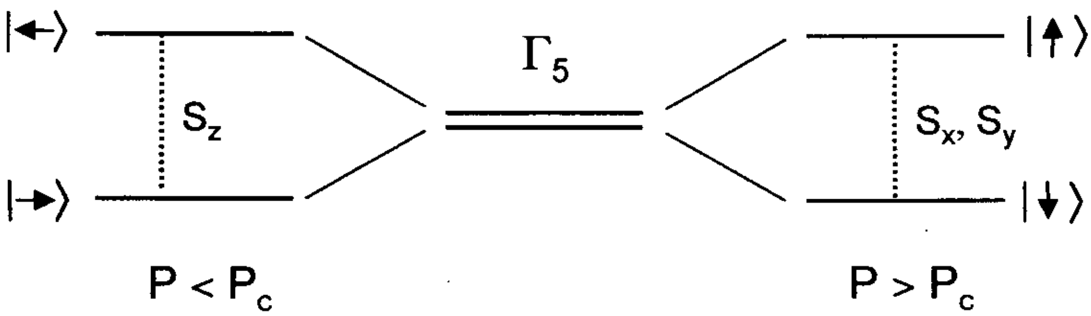
$\langle S_z \rangle \neq 0$

$\sqrt{\langle S_x \rangle^2 + \langle S_y \rangle^2} = 0$



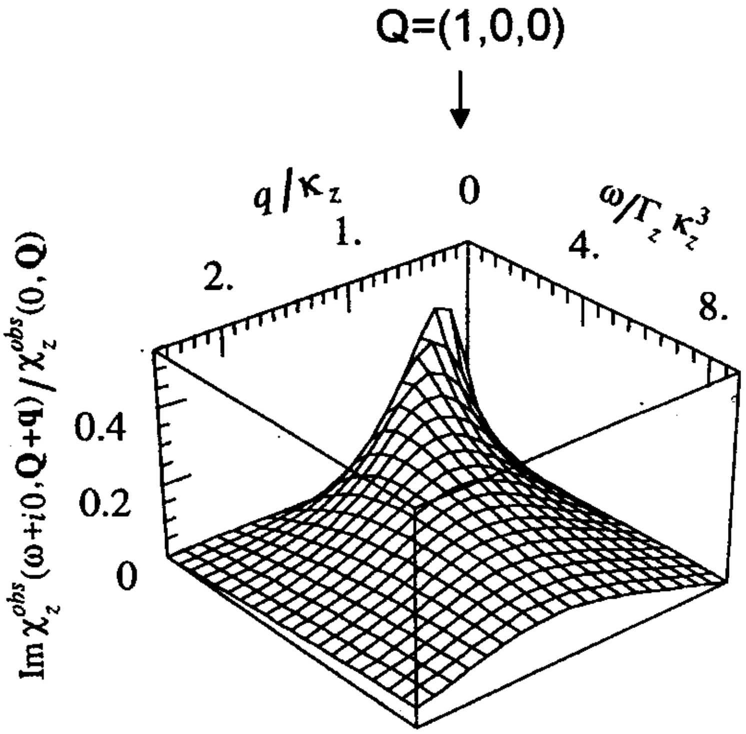
If quadrupolar interactions are dominant .....

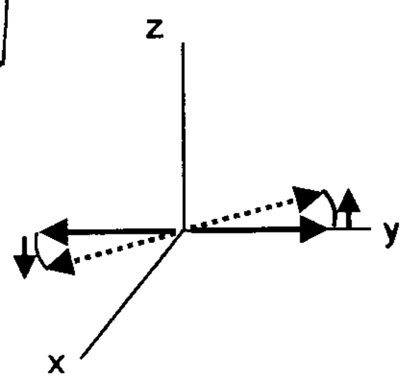
If dipolar interactions are dominant .....



F.J. Ohkawa and H. Shimizu
J. Phys. 11(1999)L519

The Tiny-Moment AF State





Quantum Spin Fluctuations
Developing under the Pseudo-Spin
Ordered State in the XY Plane

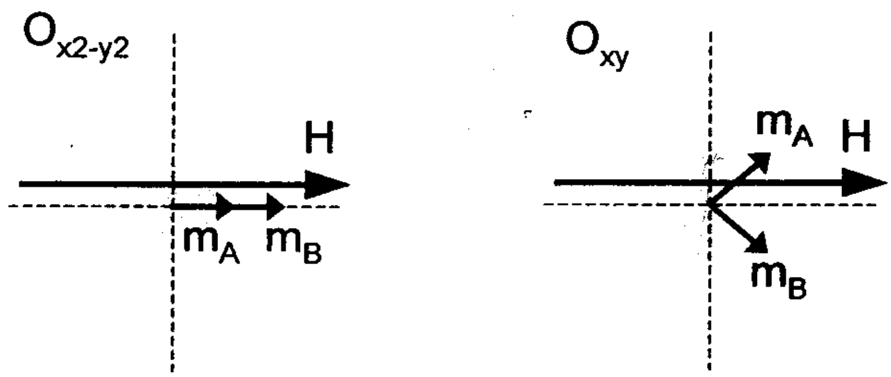
F.J. Ohkawa and H. Shimizu
J. Phys. 11(1999)L519

Advances & Prospects

Observation of the Quadrupole Order

Direct: Resonant X-Ray Scat. (M_{III} : 5d → 5f)

Indirect: NMR & NSE in Transverse Fields



Time Scale of the AF fluctuation

μSR

NMR

under High P ?

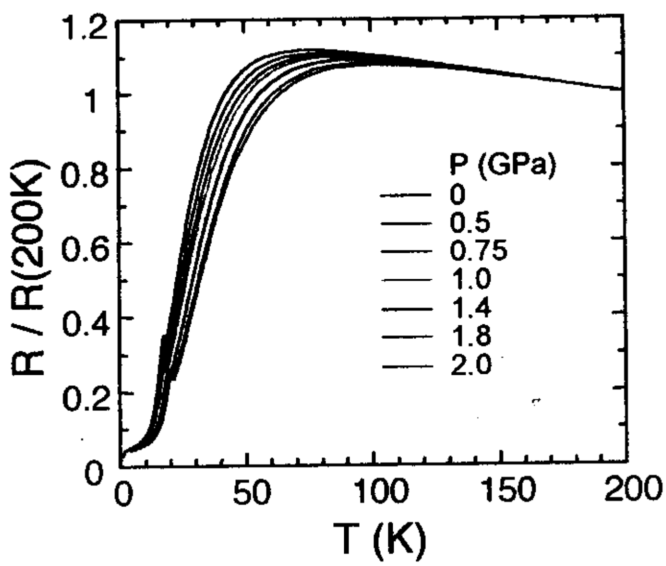
P-T Phase Diagrdam

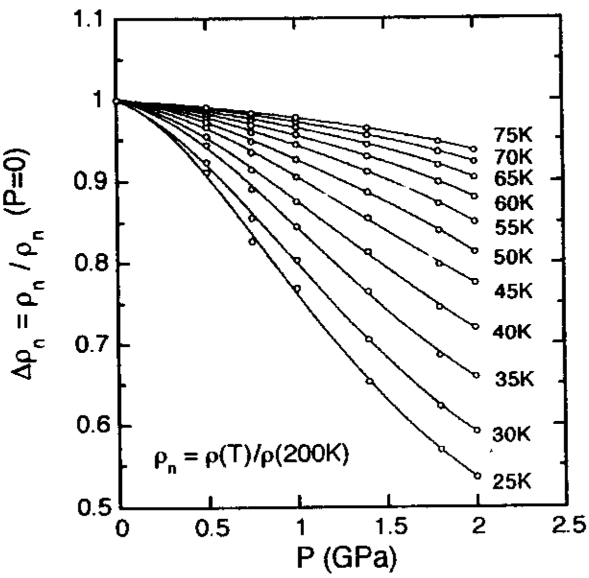
Bulk Measurements,

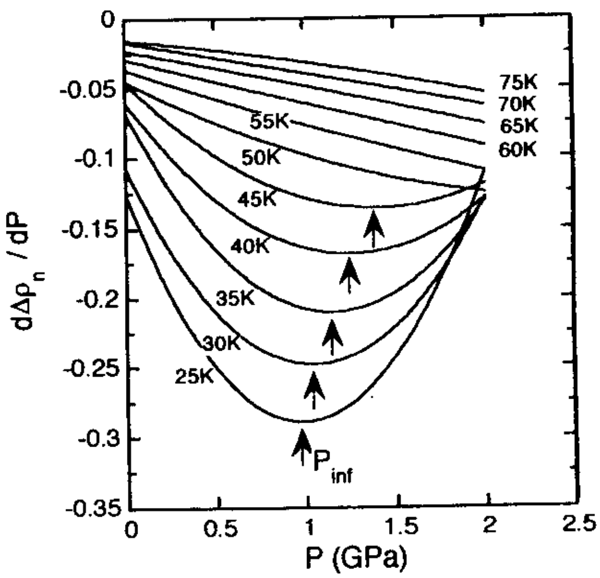
X-ray Diffraction

ρ (T) under high P

C. Sekine
MIT in Japan







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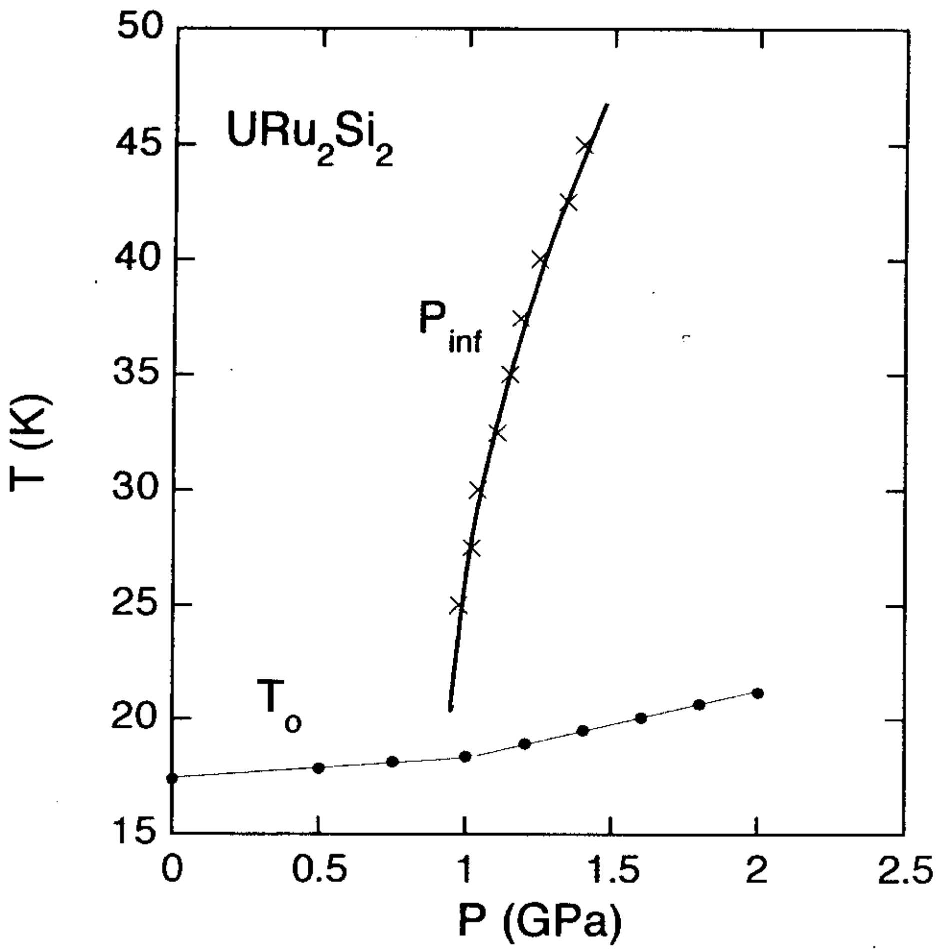
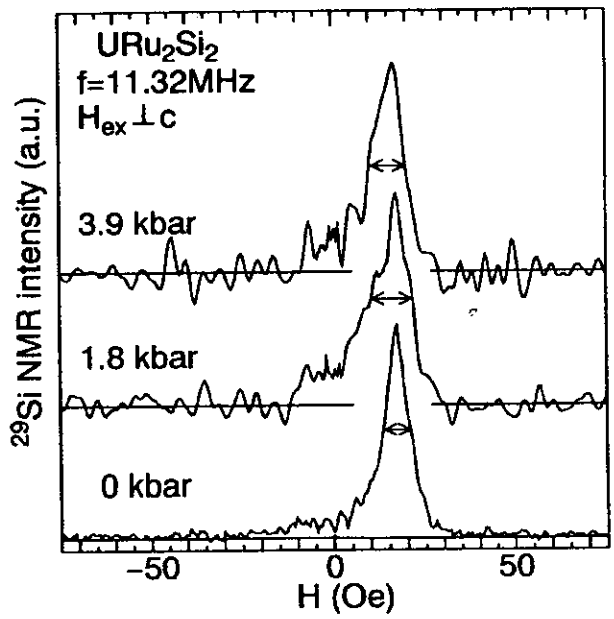
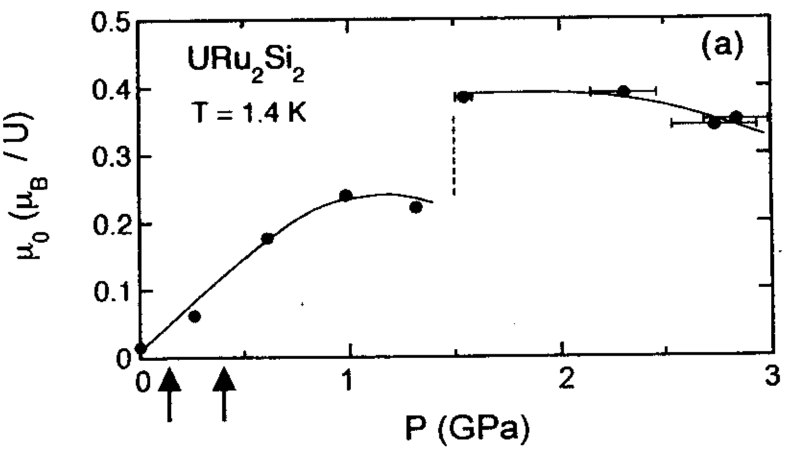
ρ (T) under high P



^{29}Si NMR under high P

K. Matsuda
Y. Kohori
T. Kohara





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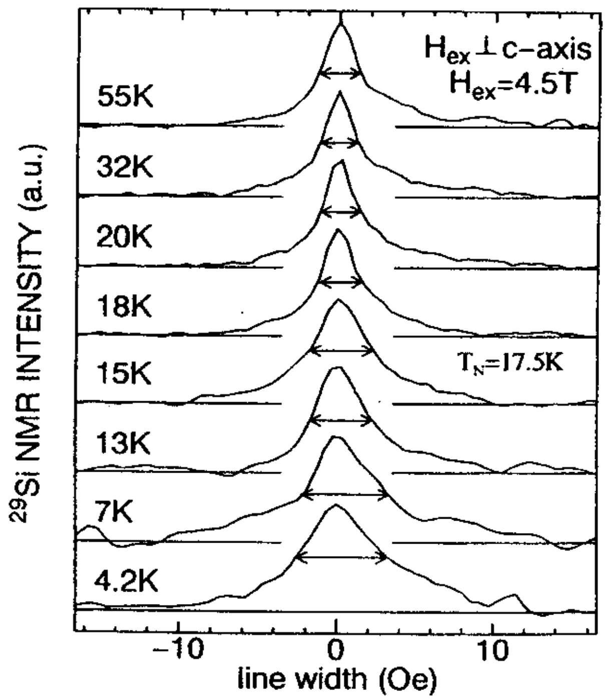
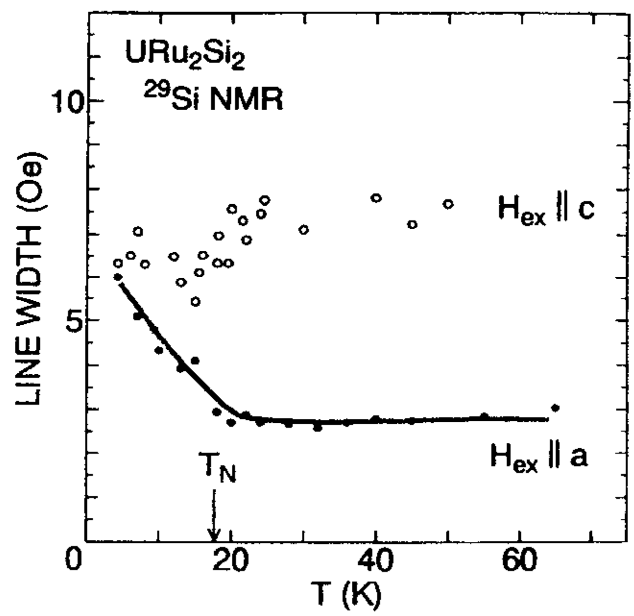
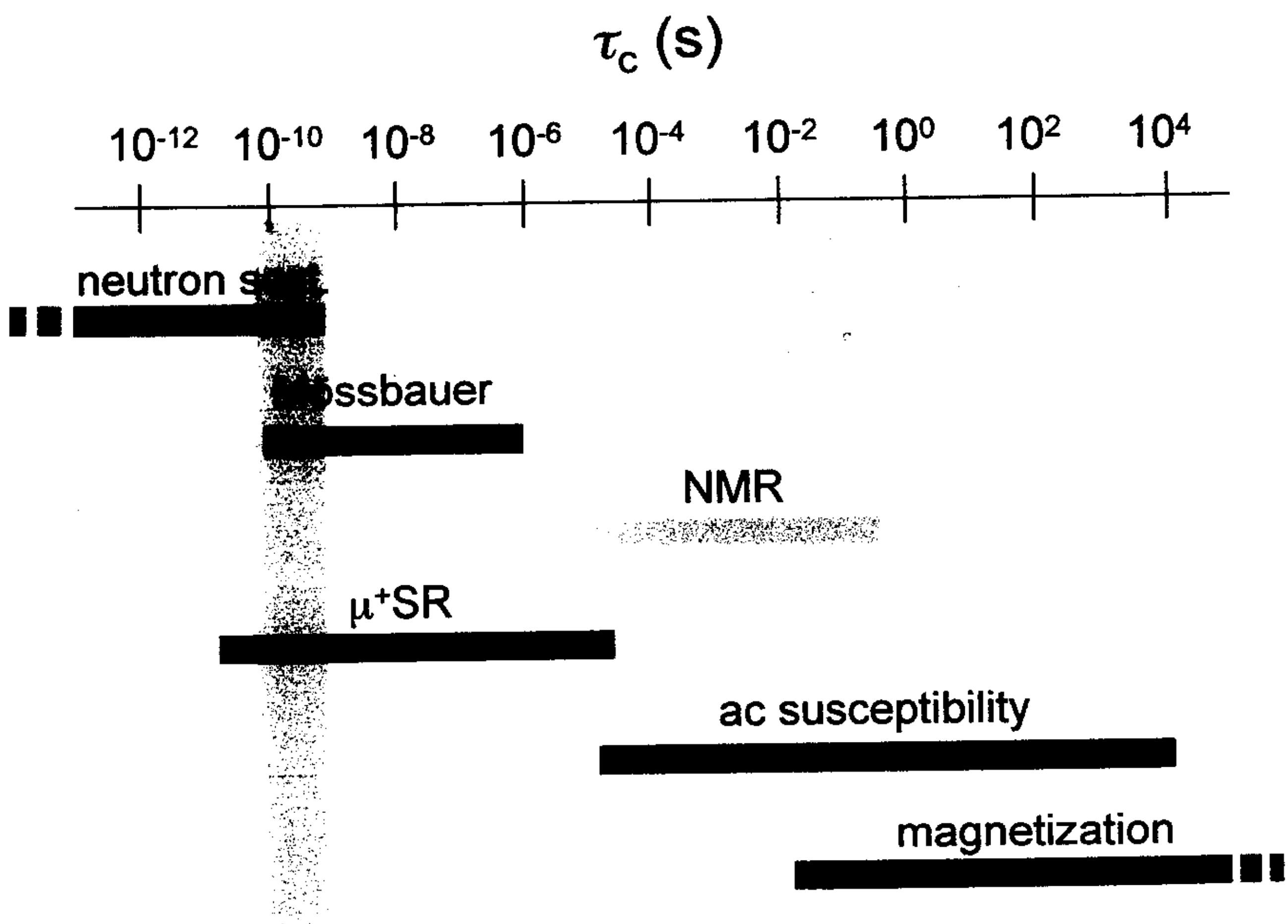
^{29}Si NMR in fields $\perp$ c-axis

K. Matsuda
Y. Kohori
T. Kohara



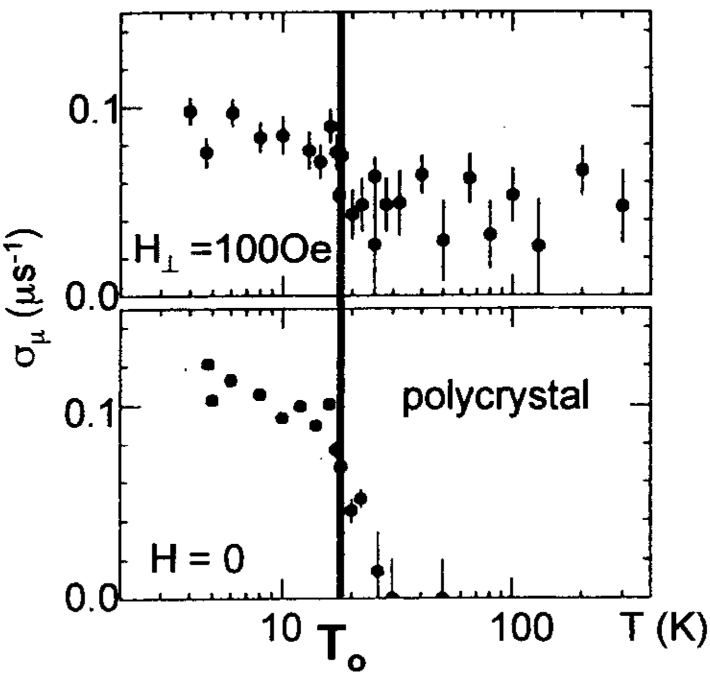


Standard time scales of observations for various methods



URu_2Si_2 ?

Previous Reports

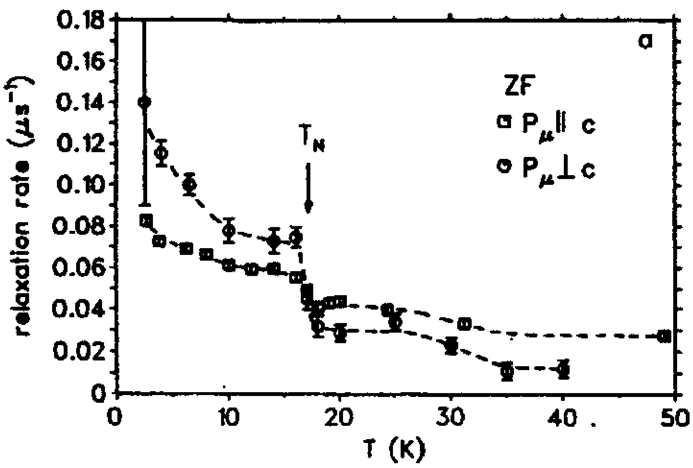


$\sqrt{\langle H_\mu{}^2 \rangle} \sim$ 1Oe

$\updownarrow$

$\mu_o \sim 0.001\ \mu_B$

MacLaughlin et al.
PRB37(1988)3153



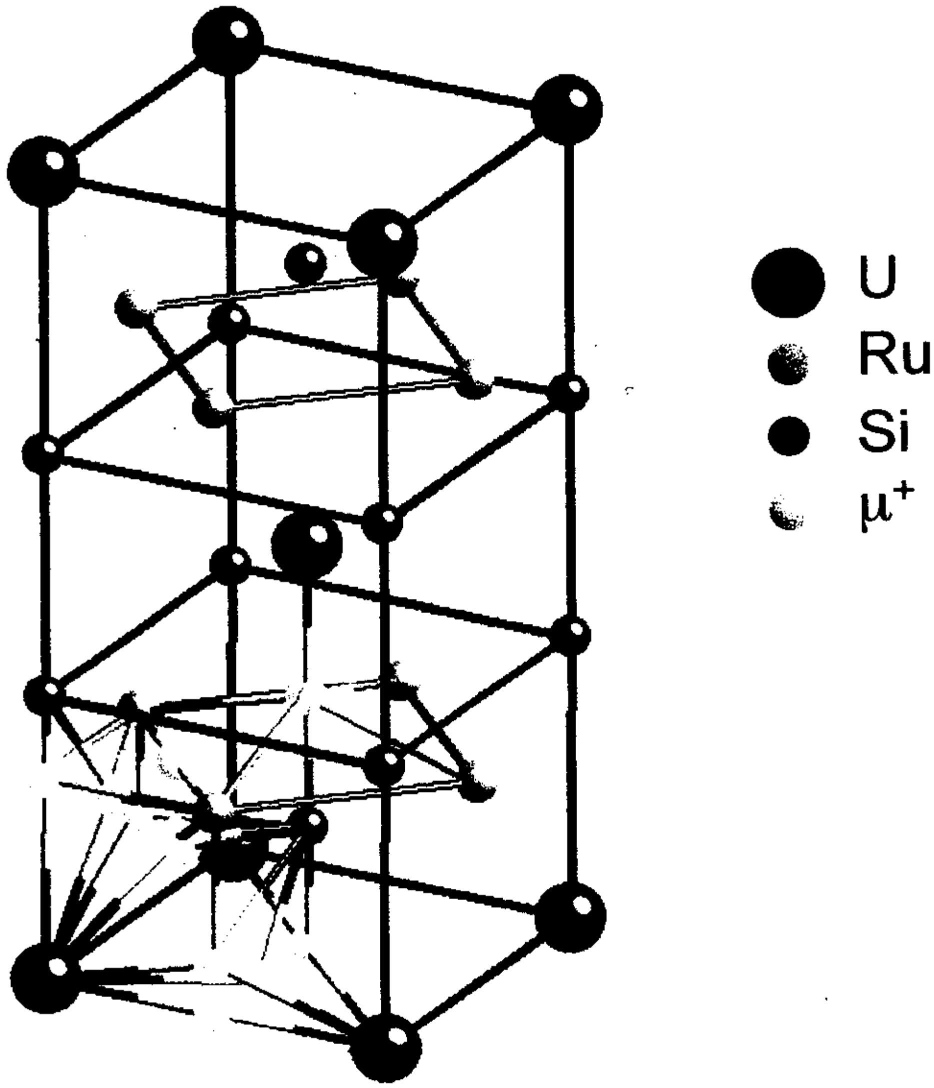
$\sqrt{\langle H_\mu{}^2 \rangle} \sim$ 0.2Oe

$\updownarrow$

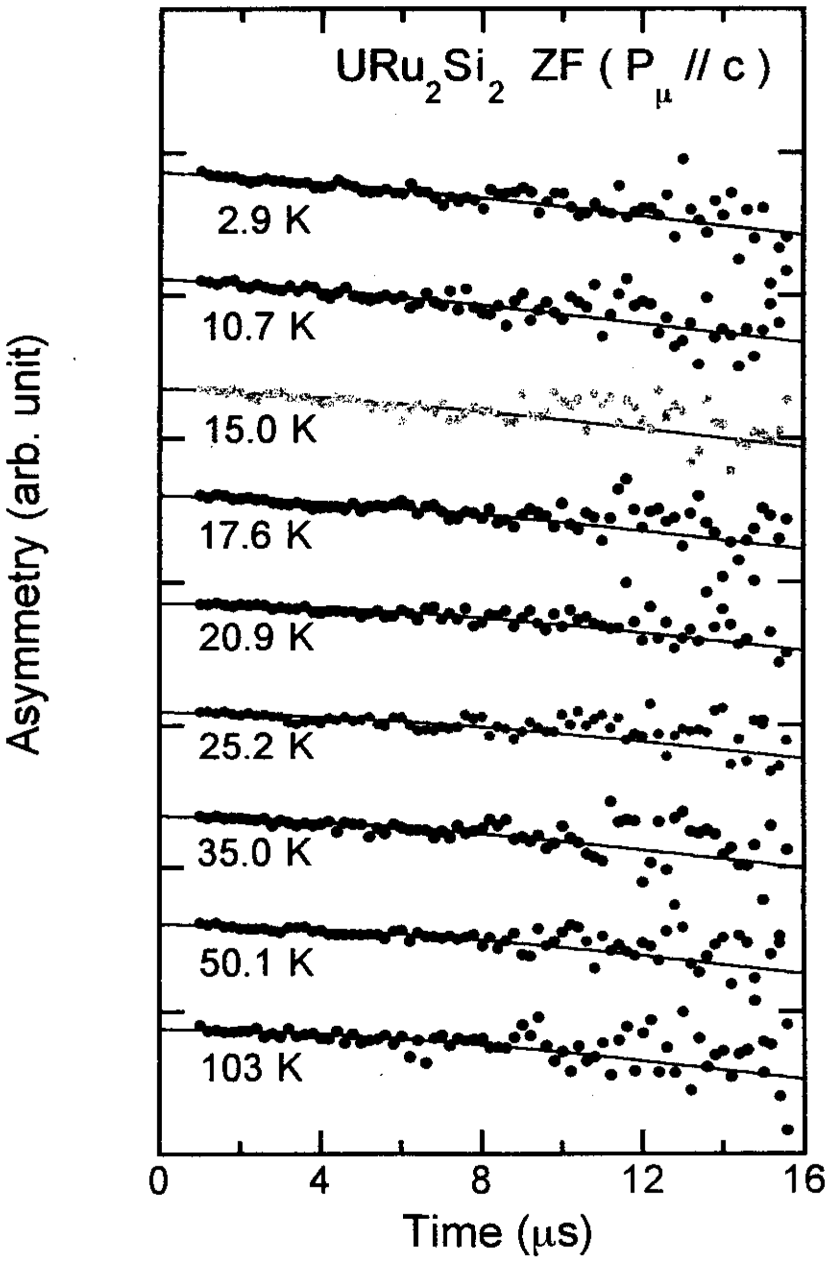
$\mu_o \sim 0.0002\ \mu_B$

E.A.Knetch et al.
Physica B(1993)300

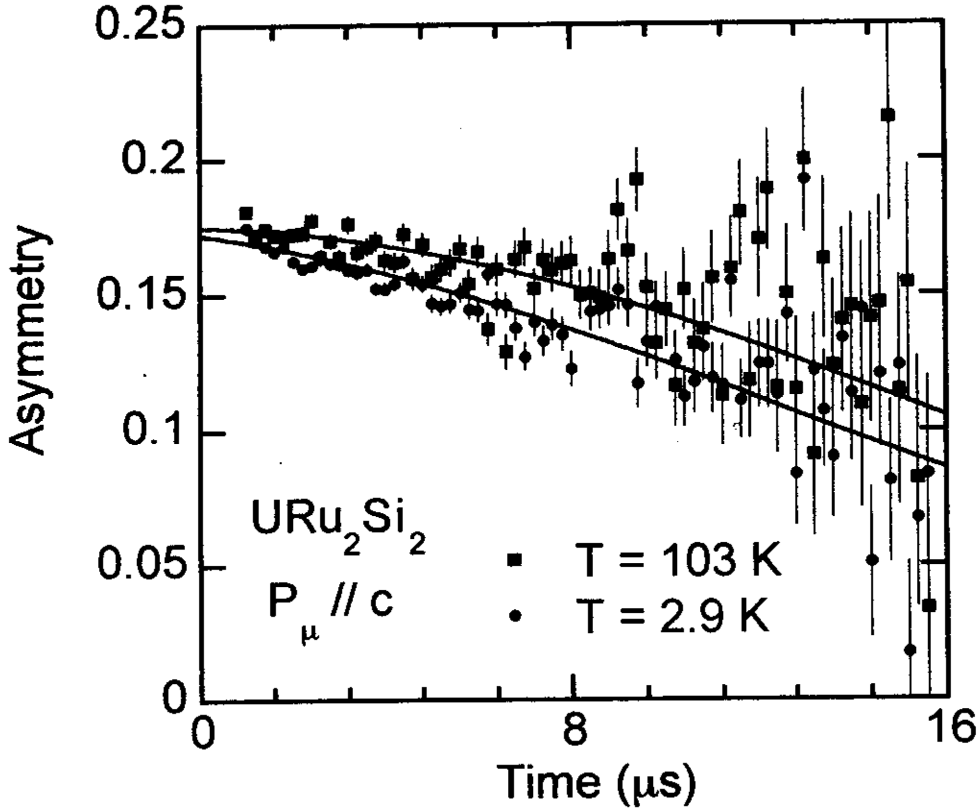
μ^+ stopping sites in URu_2Si_2



T-variations of zero field pulse μSR spectra of URu_2Si_2



Zero-field pulse μSR spectra of URu_2Si_2 at 2.9K and 103K



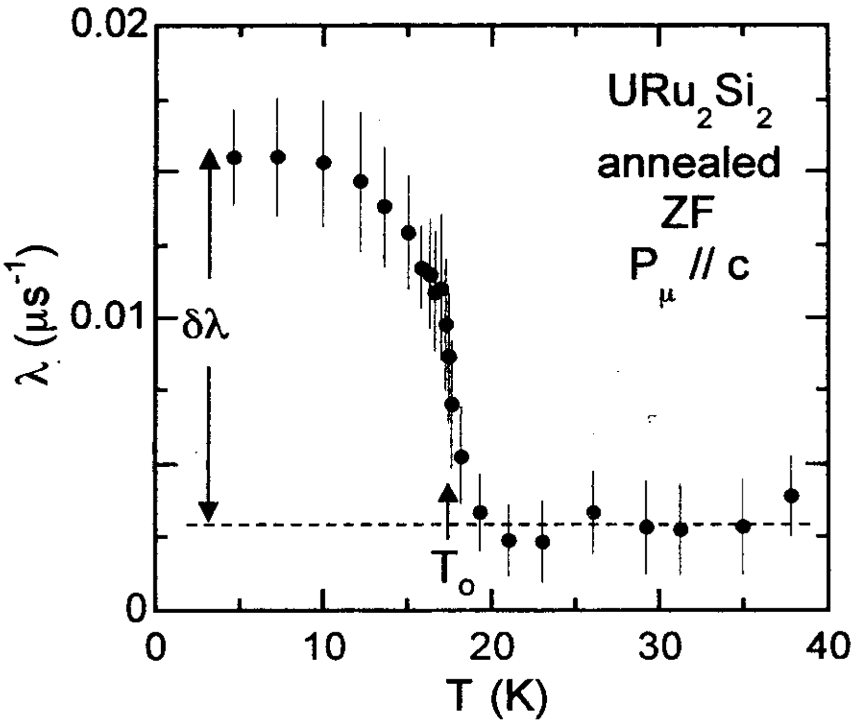
$$G_z(t) = \left\{\frac{1}{3} + \frac{2}{3}\left(1 - \sigma_{KT}^2 t^2\right) \exp\left(-\frac{1}{2}\sigma_{KT}^2 t^2\right)\right\} \exp(-\lambda t)$$

{ Kubo-Toyabe function (nuclear static moments) } × { single exponetial (electronic contribution) }

$\sigma_{KT} \sim 0.042\ \mu s^{-1}$

$\sqrt{\langle H_\mu^2 \rangle} \sim 0.5\text{Oe}$

T-variations of the relaxation rate λ

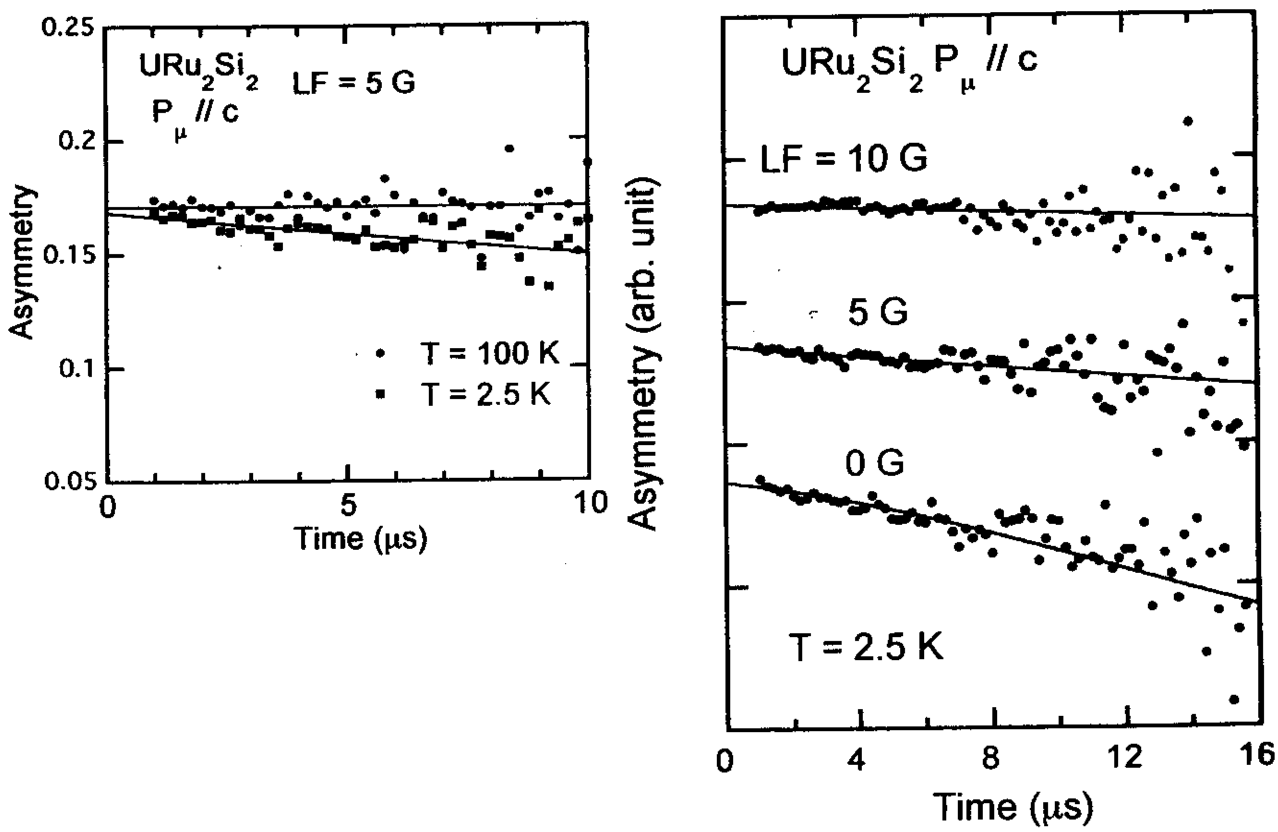


$$\delta\lambda \sim 0.013\ \mu s^{-1}$$

If the μ^+ relaxation is caused by random static fields,

$$\langle H_\mu \rangle = \frac{\delta\lambda}{\gamma_\mu} \sim 0.15\ \text{Oe}.$$

μSR spectra in longitudinal fields (Tests of "decoupling")



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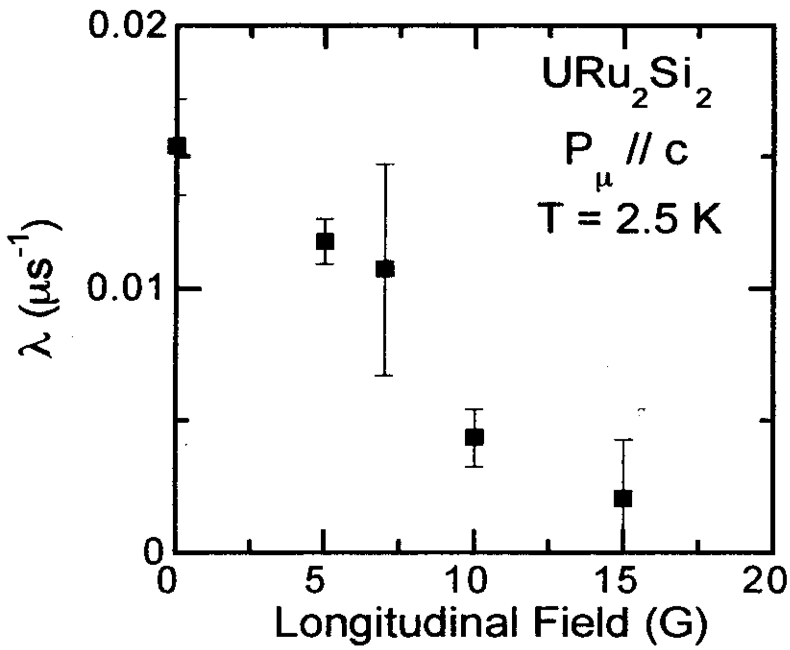
μSR spectra in longitudinal fields (Tests of "decoupling")



The μ^+ relaxation is caused by dynamical fields.

For fast fluctuations.......

$$\lambda = 2\Delta^2 \tau_c$$

$$\Delta = \gamma_\mu \sqrt{\langle H_\mu^2 \rangle} \sim 100\ (\mu s^{-1}\ \mu_B^{-1})$$

$$\mu_o = 0.02 \sim 0.04\ \mu_B \quad \text{(NSE)}$$

$$\tau_c \sim 0.1 - 2 \times 10^{-9}\ \text{sec.}$$