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Magnetic Properties Of Intermetalic Uranium Compounds

*Dr. N.A.Khalefa**

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Abstract

Magnetization density, neutron diffraction and a spin polarized have been made on uranium compounds, U_{pt3}, UB_{el3}. The compounds containing Pt, crystallizes in the hexagonal $N_{13}S_n$ structure the space group $P_6\bar{3}c$/mmc. The Heavy Fermion Compound UBe13 crystallizes in the cubic $NaZn_{13}$ structure with the space group $Fm\bar{3}c$.

In addition to the investigation of the nuclear structures of two compounds the influence of the magnetic degrees of freedom on the physical properties has been investigated. For U_{pt3} the conduction electron play an important role. It is to be expected that band structure effects significantly determine the properties of Up_{t3} at low temperature. In contrast for UBe13 the conduction electron density of states is found to be significantly smaller that in Up_{t3}. As the density of states at the Fermi level is predominately of 5f electron character in UB_{el3}, this Heavy Fermion system is easier to model magnetically compared to Up_{t3}.

الخلاصة

الكثافة المغناطيسية وحيود النيوترون والبرم المستقطب اجريت على مركبات اليورانيوم الحاوية على فلز البلاتين والبريليوم.. المركب الحاوي على فلز البلاتين ذو تركيب بلوري سداسي نوع $N_{13}Sn$ تركيبه الفراغي نوع $P6_3/mmc$ اما المركب الاحادي على البريليوم ذو تركيب بلوري مكعب نوع $NaZn_{13}$ والمجموعة الفراغية له هي Fm_{3c}. من الاختبارات الاضافية هي تحديد التركيب النووي لهذين المركبين وتأثير حرية المغناطيسية على خصائصها الفيزيائية.. تبين بأن لألكترون التوصيل تأثير هام على خصائص المركب Up_{t3} وتأثير الترابط التركيبي حدد ماهية هذه الخصائص للمركب في درجات الحرارة المنخفضة كما هو متوقع. التباين للمركب UB_{el3} هو ان لحالات كثافة الكترونات التوصيل اهمية اقل مما هو عليه لمركب Up_{t3}.. كثافة الحالات لمستوي فيرمي يحدد تأثير الكترون نوع 5f على خصائص UB_{el3} وان هذا المركب اسهل تعديل للتمغنط مقارنة مع نوع Up_{t3}.

* College of Education , Dept. of physics, University of Al-Anbar

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2412-0758/University of Technology-Iraq, Baghdad, Iraq
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1- Introduction

The discovery of Heavy Fermion Super conductor such as Up_{t3} and UB_{e13}, indicated the superconductivity in Heavy Fermion compounds is by nomeans unusual. Rather the large number of different Heavy Fermion system found up till now show that the large electron mass enhancement is compatible with various different ground states such as superconducting ground state in Up_{t3} and UB_{e13} or antiferromagmetically ordered ground state which is found in UCd_{11} and U_2Z_{n17} or a ground state which does show neither the tendency towards magnetic order nor a tendency towards superconductivity observed in $CeAl_3$ and C_eC_{u6}. compounds Up_{t3}, UB_{e13} show transitions to superconducting ground state at temperature below 1 kelvin. The Heavy Fermion compound UB_{e13} has been identified as a Heavy Fermion superconductor by Ott et al [1]. This group established the existence of a superconductor ground state in UB_{e13} with a transition temperature of Tc=0.8 [K]. The electronic contribution to the specific heat is found to enhanced with a γ UB_{e13} value of 1100 [mj/mol.K^2]. The high temperature magnetic susceptibility shows a Curie Weiss behaviour for temperature above 100[K]. The effective magnetic moment obtained in these measurements is determined to values around 3 μ_B per uranium atom. T Curie Weiss temperature θ is negative with value between –50[K] and –70[K] reported in the literature [2]- below 100[K] the magnetic susceptibility deviates from a simple Curie Weiss law rendering the measured values at low temperature to be smaller than expected on the basis of the high temperature Curie Weiss susceptibility. Up_{t3} is a superconducting Heavy Fermion system with a specific heat γ is found to be enhanced in Up_{t3} with a value of λ=422[mj/mol.K^2]. The superconducting transition temperature is determined to be Tc=-5[k]. specific heat measurements carried out by Frings et al [3] at low temperatures and in normal state were interpreted as indicating strong spin fluctuations in Up_{t3}. At higher temperatures the magnetic susceptibility is described by a Curie Weiss law with an effective magnetic moment of 2.6± 2 /μ_B U.atom [2]. Measurement on single crystals

of UPt_3 indicate the magnetic susceptibility to be strong unisotropic with C-axis being the hard magnetic direction[4]. The properties of UPt_3 have been reviewed by de visser et al [5,6]. Determination of the magnetization density in UPt_3 will reveal that the electrons responsible for the magnetism in this compound are located on both the uranium as well as the platinum atoms. Consequently the magnetic properties are therefore comples.

2- Experimental

As single crystal structure determination has been carried out using the diffractometer D15 at the ILL in General. The crystal had cylindrical shape. About 10 mm in height 1.5 mm in diameter. Approximately 400 Bragg reflections were measured at temperature of T=5[K] and T=23[K]. This enabled the determination of about 100 crystallographically independent Bragg reflection for each temperature. The atoms are located in two hexagonal planes in the unit cell, given by X,Y,Z=1/4 and X,Y,Z=3/4. Both the origin of the unit cell and point (1/2,1/2,1/2) are centers of inversion symmetry for this structure. These points are not occupied by any atom in UPt_3. The inversion symmetry of (1/2,1/2,1/2) allows the upper plane of the unit cell to be obtained from the lower one by point inversion (1/2,1/2,1/2). An additional consequence of the inversion symmetry of this structure is the reality of the nuclear factor. The lattice parameters of UPt_3 determined to be a=b=5.763 A° and c= 4.894 A°. The primitive cell contains two formula of UPt_3 with the atoms being located at positions as indicated in Tab.(1). Fig(1). Using the parameters drived from the refinement and the lattice constant (7) the nearest neighbour U-U distance was determined to be 4.12 [A°]. The nearst neighbour distance between Pt atoms is 2.81 [A°]. The various interatomic distance are indicated in fig(2) and fig(3). The structure determination confirms the $p6_3/mmc$ space group for UPt_3 as reported in the literature (pearson's crystallographic Data [8].

The temperature factor of the thermal vibrations where found to be small and no anomalies could be detected. Details of the structural parameter are give in Tab(2) with a knowledge of the lattice parameter, the distances

between the atoms can be calculated. A measurement of the static induced magnetization density in Up_{t3} was carried out on the diffractometer D3. The same single crystal was used as for the unclear structure determination. The crystal was oriented such that the (210) direction of the crystal (this direction corresponds to any easy axis of magnetization) was paraller to the external field. Susceptibility and magnetization measurement [5,6] indicate that the crystallographic c-axis is the hard magnetic direction. Measurements were carried out at a temperature of 5[k] and in a field of 4.65 Tesla. An induced magnetization experiment had been interpreted in terms of an uranium 5f electror magnetization only. Using an effective moment of .105 MB/U atom is deduced for a field of 4.65 Tesla at temperature of T=4.2 [k]. This corresponding to magnetic structure factor of .21 MB(a) there are two uranium atoms per unit cell for the X=0 reflection. Restricting the uranium atom form factor to the dipole approximation. [9] an initial refinement was carried out. The aligned magnetic moment on the U and the factor C_2 were taken as the parameters to be determined. The best agreement to the observed magnetic structure factors was obtained for an U atom moment Mu of 0.55 μB and C_2=1.96. The observed form factor and the resultant refinement obtained with the above parameters is shown in Fig(4). Within the model the magnetic structure factor and the U form factor are related by the nuclear uranium sublattic structure factor. For some reflections a finite fliping ratio is observed despite a zero U structure factor. The present model can not account of these observations and this reflections are not shown in fig (4). The total magnetic moment to approximately half the moment observed in the induced magnetization experiment. Further, the observed points are scattered around the curve representing the best fit to the data. The value of C_2=1.96 indicate an F occupation between configuration F^3 and F^4 the C_2 value do not significantly improve the fit compared to that obtained using C_2=1.75 which corresponds to a U5f configuration fig. (5).

3- Magnetic correlation in UB_{el3}:-

The magnetic correlation in UB_{el3} have been investigated in paramagnetic scattering experiment constant K and constant energy scans have been performed on the diffractometer D5 ILL in General at avariety of different temperature. The scans which were carried out as a function of the scattering vector and at constant energy transfer revealed the presence of antiferromagnetic correlation in UB_{el3} for temperature up to 100[k] fig[6] to fig[9] show the results of neutron scattering experiments carried out T=100[mk], 10[k], 100[k] and 300[k]. The magnetic scattering cross section was studied experimentally as a function of energy transfer for scattering fector of 1.06[A°], 1.56[A°] and 2.56[A°] and temperature of 10[k] and 100[k] fig[10], fig[11], and for k=1.06 A^{-1} at T=300[k] Fig [12]. The analysis of inelastic neutron scattering experiments has been presented and discussed by Holland. Moriz et al [10]. Similar presentation are also given by Holland. Moriz [11] and severing [12].

4- Discussion

The experimental results of this investigation as reported and compared to those results which are reported by Stasis et al [13] for a similler investigation. The results of the induced magnetization density determination have yield valuable information concerning the role of conduction electrons in these Heavy Fermion Compounds.

The physical picture which emerges from the investigation of the induced magnetization density in UB_{el3} is significantly different from the one obtained for Up_{t3}. while for UB_{el3} the dipole approximation and the uranium only f-electron model works well it completely fails to account for the observation in Up_{t3}. Both Heavy Fermion systems have in common a small uranium 5f electron magnetic moment which is aligned parallel to the external magnetic field direction. Therefore. However this similarity between Up_{t3} and UB_{el3} seems to end. While the magnetization density is observed to be of fairly simple character in UB_{el3} the magnetization density in Up_{t3} is found to be complex. The pure uranium 5f-electron magnetization density is of the same order of (10 me v) and spherically symmetric in bothe compounds. But Up_{t3} and UB_{el3} differ significantly when the

non uranium 5f electron contribution are considered. In Up_{t3} these non-5f electrons account for approximately half of the total magnetic moment aligned in the external magnetic field. In contrast the magnetization in UB_{el3} is found to have no appreciable conduction electron contribution. Also no magnetic contribution is found which is centered on the sites of the Be atom. The complexity of the magnetization distribution in Up_{t3} is reflected in the static magnetization measurement at low temperature Fobserved a complex behaviour as a function of field in Up_{t3}. The magnetization is observed to be highly anisotropic with the C-axis being the magnetically hard direction within the basal plane the magnetic susceptibility is larger indicating an easier alignment of the magnetic moments along the direction of the external field. As pointed out above this interpretation is consistent with the results of the paramagnetic scattering experiment carried out in a powder sample of UB_{e13}. There is an energy scale of the order of 100[mev] was indentified. The energy scale is equivalent to a temperature of the order of 100[k] and it corresponds to the temperature at which the deviation of the magnetic susceptibility from the Curie-Weiss behaviour is observed. At this temperature and below antifferomagnetic short range order correlation being to appear in UB_{el3}.

The experimental findings of the induced magnetization density in Up_{t3} and UB_{el3} also indicate. That both compounds differ substantially in the conduction electron density of state at the Fermi level. Thus the determination of the induced magnetization density in a neutron scattering experiment will enable via the identification of the various sources of magnetization density to asses the various electronic wave functions which contribute to the density of sates at the Fermi leve. However, the argument is only of qualitative character, because the filling of electron eigenstates is determined in momentum space where as the magnetization density is shown in position space. There is no straight forward and simple connection between the one and the other. A part from the information that a partially filled band must cut the Fermi level some where.

5- Conclusions

In this work magnetization density spin polarized neutron scattering has been used for the investigation of two members of Heavy Fermion Systems UB_{el3} and Up_{t3}. The nuclear structures of compounds the influence of magnetic moments on the physical properties have been investigated.

The experimental results indicate that substances show a varied behaviour. Despite the similarities in their bulk properties as observed in measurement which probe macroscopic characteristics for example, the onset of superconductivity is abserved for compounds at temperature below T=1[k]. Their properties on the atomic scale are strikingly different. The lack of similarity is vividly illustrated by the variation in the magnetization densities of Up_{t3} and UB_{el3}. These measurement illustrate the different level of importance of conduction electron in Up_{t3} and UB_{el3}. while the nuclear structure determination experiments indicate a normal behaver of the nuclear degrees of freedom in the uranium based Heavy Fermion systems. In this compounds direct and indirect evidence point towards the importance of an energy scale which is significantly larger than 10[k]. This larger energy scale is felt responsible for the low value of the aligned magnetic moment at low temperature observed in the induce magnetization measurement reported above. In UB_{el3} it is the fluctuation of the magnetic moment which is determined by this energy scale. In this case these fluctuation are not deriven by temperature. Therefore, to be considered as constituting quantum or zero point fluctuation.

In order to assess the important of the quantum fluctuations more detailed experiment are needed. These involve a detailed investigation of phonons in this compounds. For UB_{el3}, magnetic scattering experiment on single crystals will have to be carried out in order to elucidate the role of the antifferomagnetic fluctuation.

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Atom	Wychoff symbol	coordinates		
U	2c	$\frac{1}{3}, \frac{2}{3}, \frac{1}{4}$	$\frac{2}{3}, \frac{1}{3}, \frac{3}{4}$	
Pt	6h	$x, \bar{x}, \frac{1}{4}$	$2\bar{x}, \bar{x}, \frac{1}{4}$	$x, \bar{x}, \frac{1}{4}$
		$\bar{x}, 2\bar{x}, \frac{3}{4}$	$2x, x, \frac{3}{4}$	$\bar{x}, x, \frac{3}{4}$

Tab. 1. Atomic position of U and Pt atoms in Upt_3 for the space group $P6_3$ / mmc.

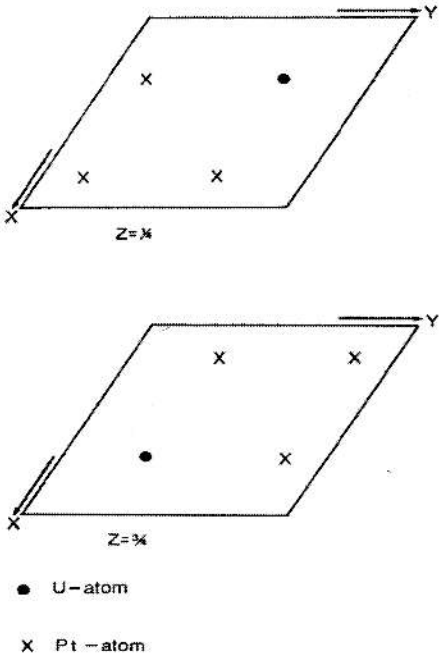


Fig.1

Position of the 2 uranium atoms and 6 platinum atoms in the unit cell of UPt_3. The atoms are located in two planes given by x, y, z = $\frac{1}{4}$ (a) and x, y, z = $\frac{3}{4}$ (b) with x = $\frac{1}{2}$, y = $\frac{1}{2}$, z = $\frac{1}{2}$ as a point of inversion.

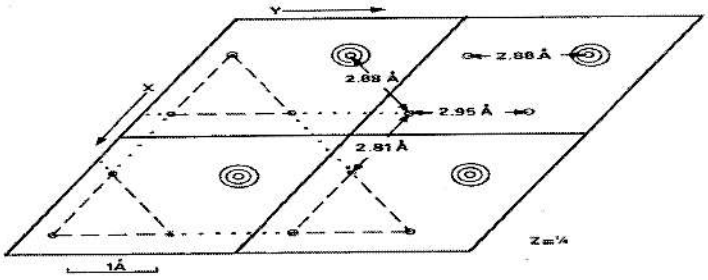


Fig.2
Position of the atoms in the x,y,z =1/4 plane for four unit cell . The distances between atoms are indicated. The platinum atoms are arranged in two different equilateral triangles with side length 2.81 [Å] (dotted triangle) and 2.95 [Å] (this triangle is indicated by the broken line). The uranium atom is surrounded by 6 platinum atoms in this plane with a 3-fold rotational axis perpendicular to the plane x,y,z=1/4.

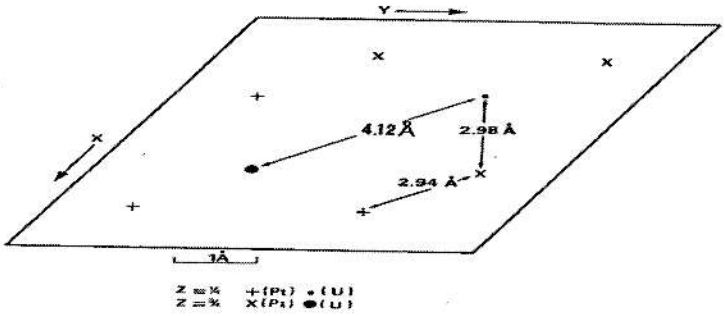


Fig.3
Position of the atoms in the unit cell as projected down onto the x,y plane the various distances between atoms in different z-planes of the unit cell are indicated. Every uranium atom is surrounded by 3 platinum atoms from above and 3 other ones from below. The shortest distance between uranium atoms is 4.12 [Å] as shown in the plot.

Atom	parameter	Temperature T = 5 [K]	T = 23 [K]
U	B11	.03 (8)	.06 (8)
	B33	.10 (9)	.10 (10)
Pt	x	.8374 (2)	.8375 (2)
	B11	-.038 (47)	.001 (45)
	B22	-.113 (73)	-.081 (70)
	B33	-.030 (50)	-.032 (44)

The structural parameters of UPt_3 and their estimated standard deviations obtained in the final refinement are shown below.

Tab.2

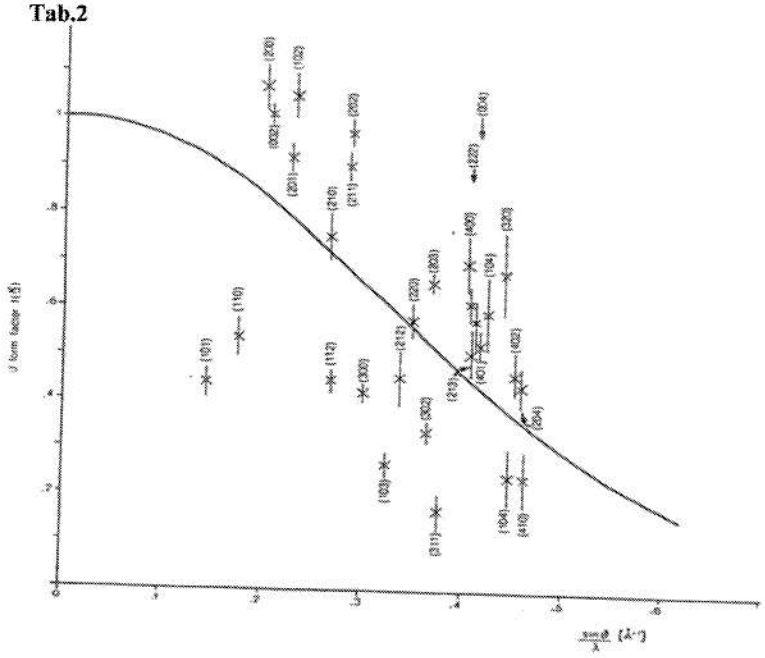


Fig. 4 Uranium form factor fit (continuous line) for UPt_3 and observations (crosses with error bars for the best two parameter fit using an uranium form factor in dipole approximation. The calculation was carried out taking only the uranium 5f electron magnetization into account.

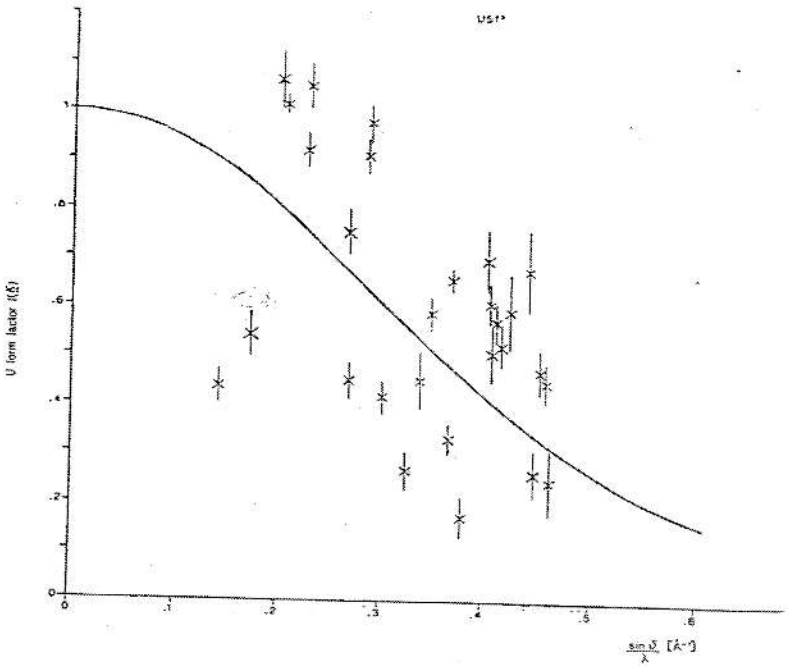


Fig.5

Uranium form factor fit (continuous line) and observations (crosses) for the uranium atom with a f^3 configuration. The fit was performed within the dipole approximation, fixing the value of C_2 to 1.75. In the model calculation it was assumed that only the uranium 5f electrons contribute to the magnetization in UPt_3.

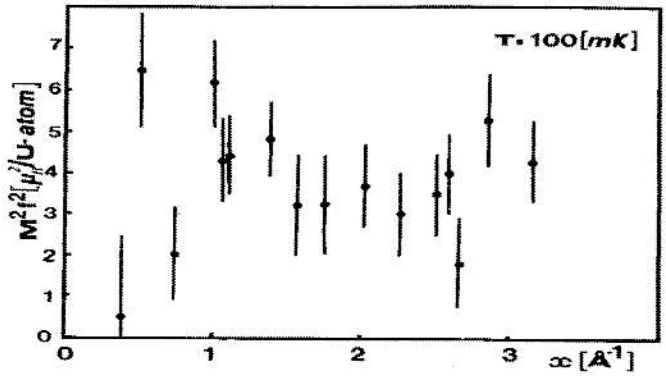


Fig.6

Constant energy scan in UBe_{13} with the spectrometer set in the elastic position. The temperature at which the measurement was carried out, T = 100 [mK], is below the superconducting transition in UBe_{13}. The enhancement observed around certain scattering vectors indicates the presence of antiferromagnetic correlations down into the superconducting state.

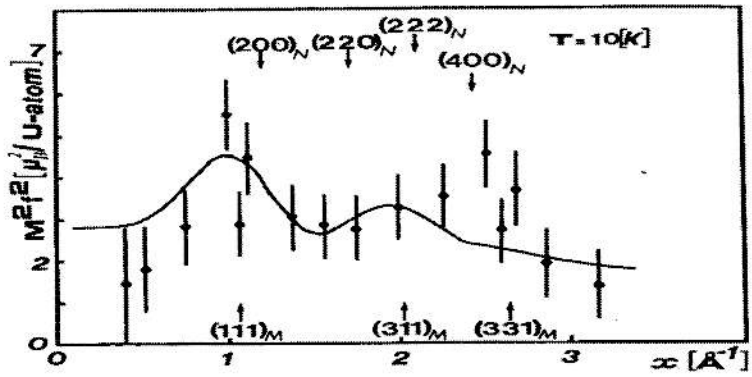


Fig.7

Elastic energy scan in UBe_{13} at a temperature of T = 10 [K]. The position of the nuclear Bragg reflections (indexed by hkl) is indicated by $(hkl)_N$ in the figure. The position of the antiferromagnetic Bragg reflections for the structure characterized by $\underline{k} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ is indicated by $(hkl)_M$. The continuous line corresponds to a fit using a model as described in the text.

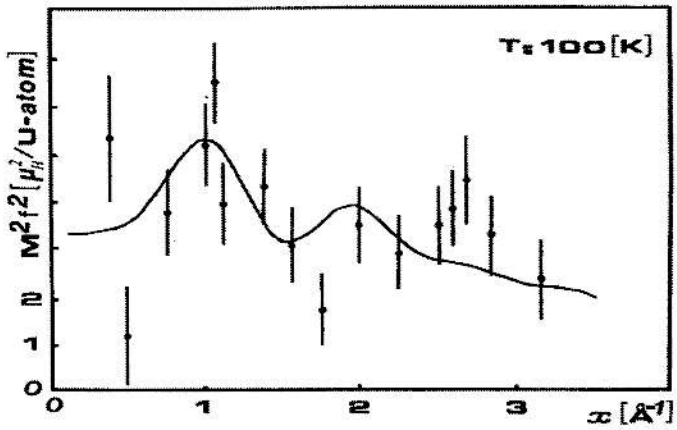


Fig.8

A scan carried out at constant energy transfer and at the elastic position of the spectrometer. The sample temperature was T = 100 [K]. The continuous line corresponds to a model fit as described in the text.

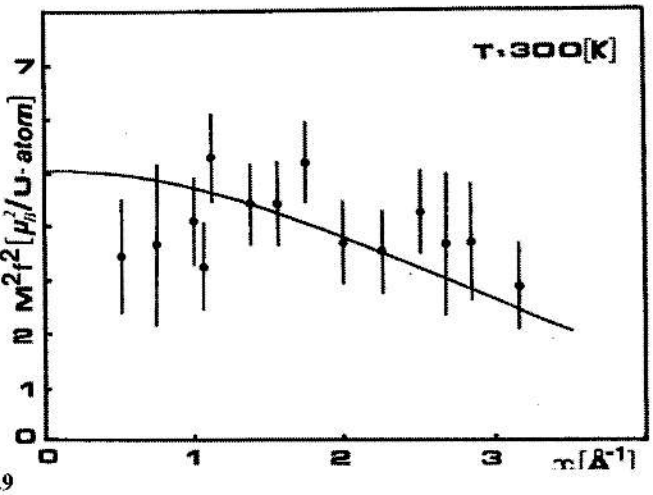


Fig.9

Constant energy scan in UBe_{13} carried out at zero energy transfer and at room temperature. The continuous line corresponds to a model fit using a purely paramagnetic description of the magnetic moment which is located on the uranium atom. The falloff at larger values of $|\kappa|$ is due to the magnetic form factor.

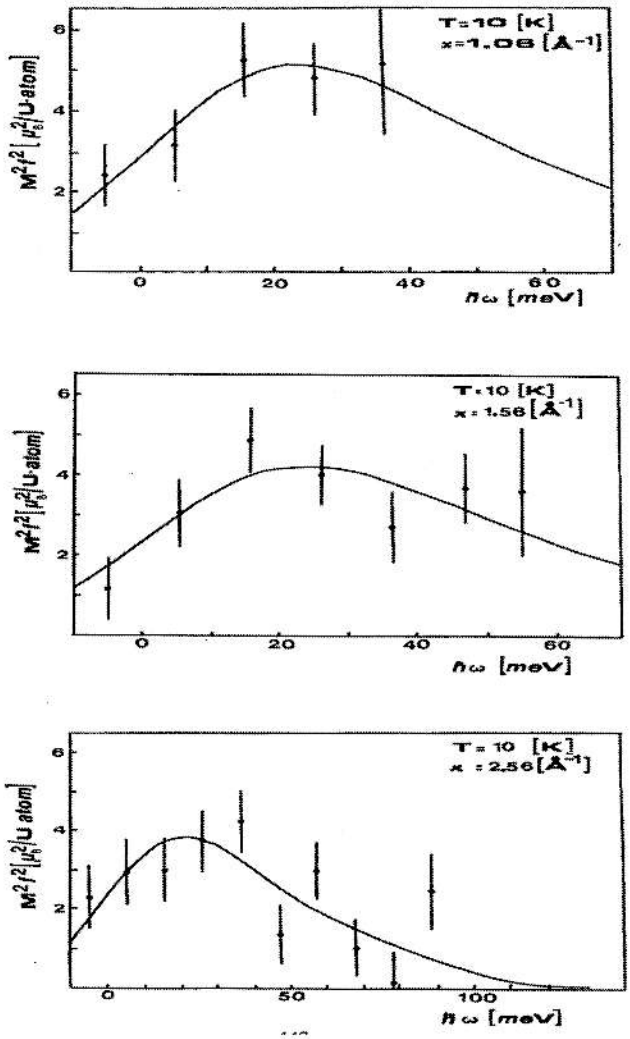


Fig.10

Inelastic measurements of UBe_{13} at a temperature of T = 10 [K] and at constant scattering vectors of 1.06 [$Å^{-1}$], 1.56 [$Å^{-1}$] and 2.56 [$Å^{-1}$].

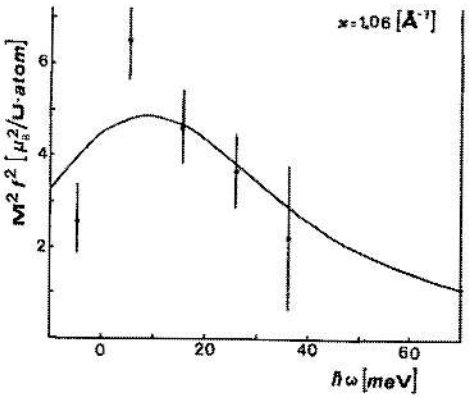


Fig.11

Inelastic measurement and fit at a temperature of T = 100 [K]. The scans were carried out at constant scattering vectors of 1.06 [Å^{-1}], 1.56 [Å^{-1}] and 2.56 [Å^{-1}].

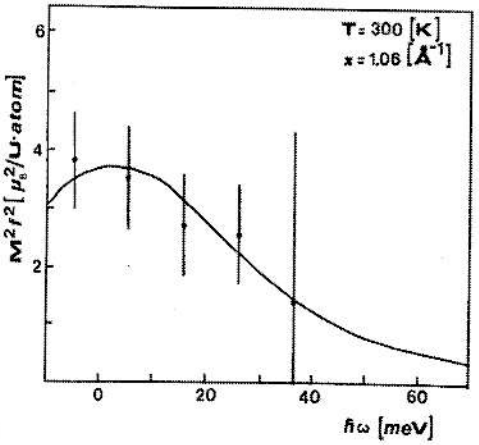
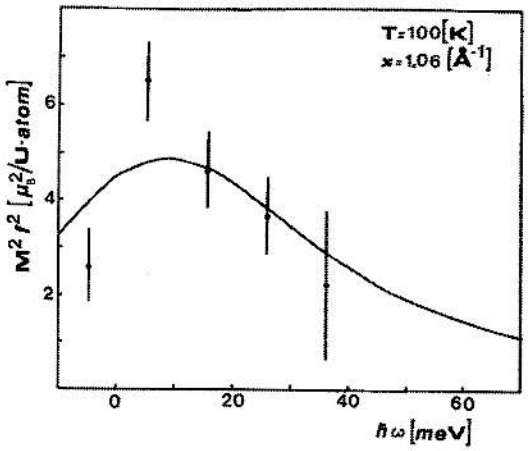
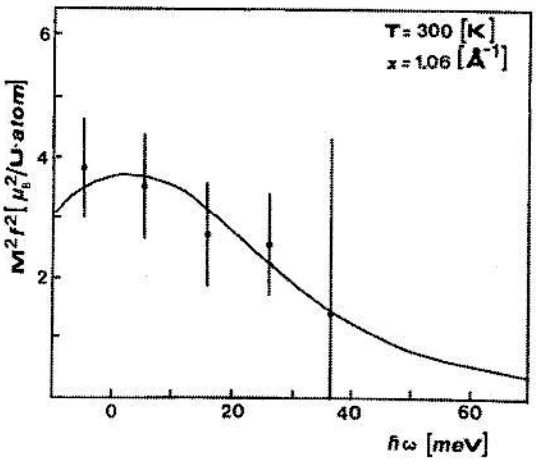


Fig.12

Inelastic measurements of UBe_{13} at room temperature (T = 300 [K]) and at a constant scattering vector of 1.06 [Å^{-1}].



Inelastic measurement and fit at a temperature of T = 100 [K]. The scans were carried out at constant scattering vectors of 1.06 [Å^{-1}], 1.56 [Å^{-1}] and 2.56 [Å^{-1}].



Inelastic measurements of UBe_{13} at room temperature (T = 300 [K]) and at a constant scattering vector of 1.06 [Å^{-1}].