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Neutron scattering studies on uranium compounds in high magnetic fields

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Abstract

Uranium intermetallics exhibit various interesting phenomena as a function of magnetic field, temperature and/or pressure. Bulk measurements usually provide first information regarding their physical properties. Nevertheless, neutron scattering provides important information that is otherwise not achievable. In this contribution we discuss issues of neutron scattering technique in magnetic fields. A few experimental results obtained on UNiGe, UNiAl and U_2Pd_2In obtained in fields up to 17 T are used as examples of the state of art of this technique at present. We also briefly report on a project to build a dedicated high-field facility for neutron research with the aim to achieve fields up to 40 T.

1. Introduction

Uranium intermetallics exhibit a rich variety of interesting phenomena (e.g. long-range order, superconductivity, Kondo, heavy-fermion and non-Fermi-liquid states etc). Change of thermodynamic parameters like temperature, magnetic field and pressure can further alter these phenomena. In this contribution we focus on antiferromagnetic (AF) U compounds, in which magnetic moments usually reside only on uranium ions. In sufficiently high fields AF interactions are overcome and a field-induced metamagnetic state is formed. In the limit of very high field a field-forced ferromagnetic (FIF) state is established. Most of the magnetic studies are performed by means of magnetization measurements, which sense only the total magnetic response of the material and are not able to reveal individual contributions. For that, neutron diffraction is an indispensable tool.

2. Experimental details

Conditions of a certain orientation of the crystallographic axes, neutron beam and magnet coil impose serious geometrical limitations on the scattering experiment. Two basic experimental

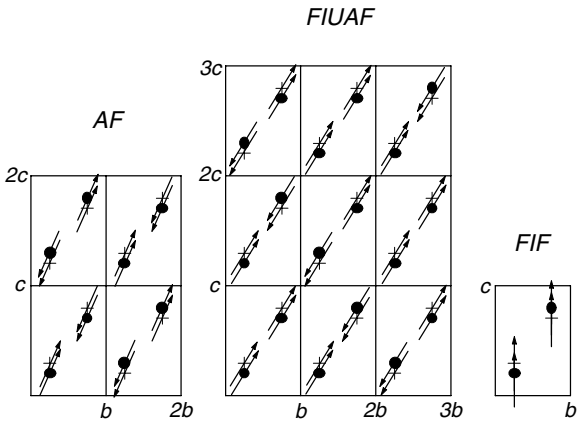

Figure 1. Schematic representation of the magnetic structures of UNiGe (AF—ground state antiferromagnetic phase, FIUAF—field-induced uncompensated AF phase, FIF—field-induced ferromagnetic phase). Dots and crosses denote components parallel/antiparallel to the *a* axis.

geometries exist, with the horizontal or vertical field with respect to the incident beam. In the former the direction of the magnetic field lies in the scattering plane. This can be achieved either with a simple solenoid in a small-angle scattering experiment or with a magnet that has the coil split into two or more coils having wedges in the scattering plane, limiting the access for neutrons. In the vertical geometry one has to use a split pair coil design having almost no wedges but smaller maximum field.

To demonstrate both geometries we describe here three experiments on uranium compounds, namely UNiGe, UNiAl and U_2Pd_2In. All systems were investigated in a single-crystalline form. Neutron data were obtained using diffractometers installed either at HMI Berlin (E4) or at LLB Saclay (6T2) in magnetic fields up to 17 T.

3. Results

In the orthorhombic UNiGe the nearest-neighbour uranium atoms form zig-zag chains along the *a* axis [1]. Bulk measurements revealed an easy-plane magnetocrystalline anisotropy with the hard-magnetization direction along the *a* axis [2]. UNiGe exhibits incommensurate (INC) AF order between $T_N = 50$ K and $T_M = 42.5$ K at which an order–order transition to the ground AF state appears. The INC phase has a propagation vector $\boldsymbol{q}_{INC} = (0, \delta_1, \delta_2)$, where $\delta_{1,2}$ are close to 0.36. The ground state has a $\boldsymbol{q} = (0, 1/2, 1/2)$ [3].

While a magnetic field as high as 38 T applied along the *a* axis does not affect the AF state of UNiGe, for the other two principal directions an almost saturated magnetization $M_s = 1.45\ \mu_B$ is attained above two subsequent metamagnetic transitions (MTs) that occur at 17 and 25 T for $B \parallel b$ and 3 and 10 T for $B \parallel c$, respectively. The intermediate field-induced uncompensated antiferromagnetic (FIUAF) phases are characterized by propagation vectors $\boldsymbol{q} = (0, 1/3, 1/3)$. Above the second MT a FIF state is established (see figure 1).

The existence of two components in propagation vectors of INC, AF and FIUAF phases and the necessity to apply a magnetic field either along the *b* or *c* axis imply that on constant wavelength neutron sources one has to use split coil horizontal cryomagnets having for principal technical reasons a rather small maximum magnetic field (limited in our case to 6 T) and a restricted angular range leading to very restricted set of reflections. To study the FIF ($H \parallel c$)

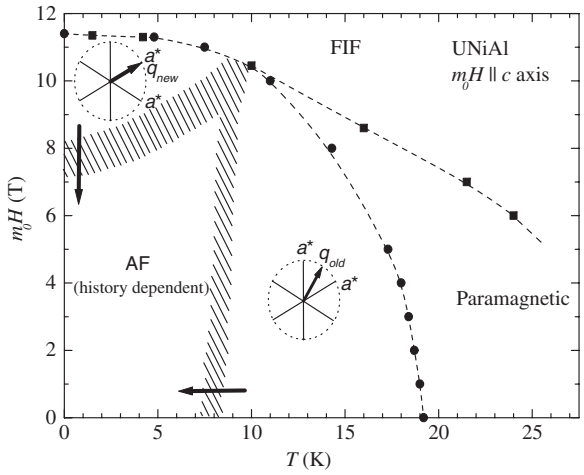

Figure 2. Magnetic phase diagram of UNiAl. Shaded areas denote region where both old and and new AF structures exist.

state a vertical superconducting magnet that is capable of reaching fields as high as 14.5 T was used. In this case, because of the very small opening of $\pm 2^\circ$, no pure magnetic reflections are observable and only FIF components can be measured. Nevertheless, even such constrained conditions revealed unambiguously that in all the phases (even in the FIF state) a significant AF a axis component exists. Magnetization measurements which show a slow increase above the second MT were analysed in terms of simple ferromagnetic order with field-induced U/Ni moments. Neutron experiments were able to identify this magnetization increase with a gradual alignment of U moments with increasing magnetic field. This result, in turn, provides a clear indication for the presence of anisotropic interactions in UNiGe [4].

The hexagonal UNiAl orders AF below $T_N = 19.3$ K [1] and exhibits strong uniaxial magnetic anisotropy in with the c axis as the easy-magnetization direction. Upon cooling in zero field the AF structure in UNiAl is characterized by $\boldsymbol{q}_{\rm old} = (0.1, 0.1, 0.5)$. The magnetic structure refinement points to two U moments in the UNiAl unit cell coupled together ferromagnetically and the third one, which is smaller, uncoupled. Moments are sine-wave modulated in the basal plane along [110] and coupled antiparallel to those in the next plane. When a magnetic field $H \parallel c$ is applied, UNiAl undergoes an MT at 11.35 T [1]. Neutron diffraction experiments in magnetic fields have confirmed the FIF phase to be ferromagnetic. To perform such an experiment one has to use a split-pair vertical magnet which has a large opening and lifting counter-detector. Fortunately, the 12 T cryomagnet used at ILL Grenoble and LLB Saclay has an opening of $-5^\circ + 12^\circ$. Combination with short wavelength made it possible to observe reflections with index $l = 0.5$.

Recently, magnetic-history-dependent phenomena have been observed in UNiAl [5]. When the magnetic field increases above 7 T, original magnetic reflections get broader and above 8 T new magnetic reflections appear which can be described by propagation vector $\boldsymbol{q}_{\rm new} = (0.173, 0.0, 0.5)$. Between 7 and 8 T no well defined AF state exists. Instead, a ring of enhanced intensity around the origin of reciprocal space appears. This is interpreted in terms of coexistence of $\boldsymbol{q}_{\rm old}$ and $\boldsymbol{q}_{\rm new}$ that have equal length.

New magnetic reflections are then stable down to zero field and disappear above 7–8 K, where the old reflections re-appear, upon heating (figure 2). Analysis of the data set belonging to the new AF structure lead to essentially very similar AF structure

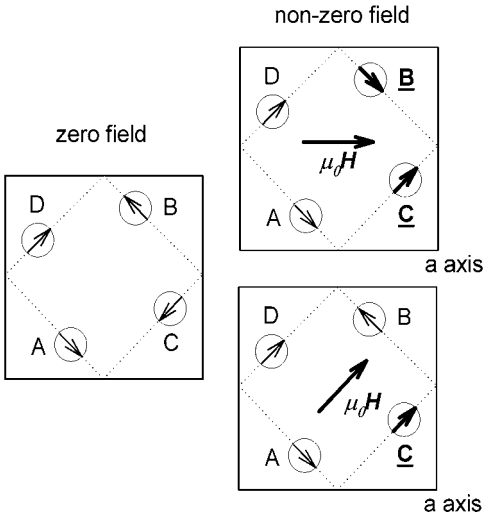

Figure 3. Schematic representation of the field-induced non-collinear magnetic structures of U_2Pd_2In established upon application of field applied along [100] (top right) and [110] directions.

as the old one. However, now all the U magnetic moments are from symmetry arguments decoupled and modulated along a different direction. The microscopic difference between both structures is that while in the old AF structure there are two kinds of chain (one built by U atoms only and the other by the sequence Ni–U–Ni–U), in the new one there is only one type (Ni–U–U–U–Ni). The originally decoupled U moment in the old AF structure increases with field faster than the other two and unbalances the original magnetic symmetry expressed by $\boldsymbol{q}_{\rm old}$. At a certain critical field it appears equivalent to the other two moments and exchange interaction along the Ni–U–U–U–Ni chain becomes more important.

The tetragonal U_2Pd_2In orders AF below $T_N = 38$ K [1]. A complicated non-collinear ground state arrangement of U moments exists in this compound (figure 3). At 4.2 K, magnetization measurements show MTs at 29 T, and 26 T with increasing field applied along the [100] and [110] directions (figure 3), respectively, while in the c axis no transition is observed up to 57 T although is this the easy-magnetization direction. A maximum field of 17 T has been set up for use at HMI with the help of Dy pole tips that focus the magnetic field produced by a 14.5 T cryomagnet onto the sample. However, this field is not sufficient to study field-induced magnetic structures in U_2Pd_2In at low temperatures and elevated temperatures are necessary. It appears that above 33 K one can at 17 T achieve modifications of the ground state AF structure. Unfortunately, the original steplike character of the MTs is lost and an S-shape is observed on magnetization curves at this temperature.

Specific geometrical requirements and constraints lead to the necessity to perform three independent experiments in which we collected ($hk0$), ($h0l$) and ($h-hl$) reflections for $B \parallel [001]$, $B \parallel [010]$ and $B \parallel [110]$, respectively.

Analysis of all the data sets scaled together revealed field-induced noncollinear magnetic structures (figure 3) [5] that agree with *ab initio* theoretical calculations for low temperatures [6]. These results clearly demonstrate the importance of hybridization-induced magnetocrystalline anisotropy in U_2Pd_2In that locks U moments in the [110] type mirror planes even at elevated temperatures.

4. Discussion and conclusions

Uranium compounds are known to exhibit a huge magnetocrystalline anisotropy due to strong hybridization of 5f states with other states in the compound [1]. The anisotropy energy is much larger than the perturbation caused by external magnetic field that is eventually able to change the type of magnetic order but not to tilt magnetic moments out of specific directions. Therefore, one has to use rather high magnetic fields and apply them along particular directions. Presented examples show on a microscopic scale that even field as high as 14.5–17 T (magnetization data suggest that the limit is much higher) are not able to align U moments parallel to each other (in UNiGe the hard-axis AF component exists in the FIF phase; in the case of U_2Pd_2In U moments are locked in the [110] type of mirror plane). On the other hand, the example of UNiAl shows that there might be several magnetic structures that compete with each other and a field of several Tesla can stabilize one or the other even if the direction of the moments remains.

In general, application of magnetic field and its combination with neutron scattering is of prime importance and interest in solid state physics. While in some cases the field intensity is sufficiently large to achieve desired magnetic phases, there are important cases where much stronger magnetic fields are needed. This is because of the energy scale involved in magnetism. The energy of the magnetic exchange J competes with perturbation caused by the external field and thermal fluctuations. For a given J it is quite easy to affect the magnetic state by changing the temperature. In the case of magnetic field we are restricted to fields smaller than 22–24 T (superconducting magnets), 30–45 T (resistive and hybrid magnets) and 80–100 T (long-pulse magnets; we do not consider destructive ones). For neutron research the current limit is at 17 T.

Recently, HMI submitted a proposal to the national authorities to built a dedicated high-field installation devoted to neutron research. Two resistive magnets, one horizontal (tapered solenoid able to achieve fields as high as 40 T) and one vertical (split pair coil having maximum field of up to 30 T) are foreseen to cover requests for experiments ranging from small-angle scattering through classical magnetic structure studies to experiments up to now unthinkable. A 40 MW power station will be necessary to energize the magnets. To achieve optimized performance, a time of flight spectrometer equipped with large-area detectors standing at the end of a ballistic neutron guide that is able to deliver thermal as well as cold neutrons over a distance of more than 70 m will be built. The facility is planned to be in operation three years after it is approved. No costs for proposal-oriented academic research are foreseen. There is no doubt that such a facility will open entirely new research possibilities.

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