Characteristics of the magnetic order in CeCu$_2$Si$_2$ revealed by neutron spin-echo measurements

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Abstract. In antiferromagnetically ordered $A$-type CeCu$_2$Si$_2$ the linewidth of the magnetic Bragg peaks, measured by elastic neutron scattering, is larger than the resolution limit for $T < T_{c,\rho}$, with $T_{c,\rho}$ identified by resistivity measurements as the onset of a very broad transition to the superconducting state. In contrast to that, the linewidth in $A/S$-type CeCu$_2$Si$_2$, where bulk superconductivity (SC) expels magnetic order, is resolution-limited. This suggests that filamentary SC limits the size of magnetically ordered regions in $A$-type crystals. We present neutron spin-echo measurements which yield significantly faster exponential decay rates of the normalized intermediate scattering function $S(Q_{AF}, t)/S(Q_{AF}, 0)$ in $A$-type CeCu$_2$Si$_2$ at $T < T_{c,\rho}$ than at $T_{c,\rho} < T < T_N$ as well as in $A/S$-type CeCu$_3$Si$_2$ at $T < T_c$. We conclude that filamentary SC also causes reduced lifetimes and enhanced fluctuation rates of the magnetic order in $A$-type CeCu$_2$Si$_2$.

1. Introduction

The interplay of antiferromagnetism (AF) and superconductivity (SC) is especially complex in CeCu$_2$Si$_2$, the first heavy-fermion superconductor to be discovered [1]. The present study focuses on the magnetic order in the system by addressing the question whether its nature is static or rather dynamic.

Due to the closeness of the hybridization of 4f and conduction electrons to the critical value, where a quantum phase transition occurs, the ground state properties of CeCu$_2$Si$_2$, which crystallizes in the tetragonal ThCr$_2$Si$_2$ structure, depend very sensitively on the exact stoichiometry within the homogeneity range and on the growing conditions. While a slight Cu excess yields SC samples (S-type), Si rich ones show AF order (A-type). Samples with a composition very close to the nominal stoichiometry exhibit both properties (A/S-type) [2].

After large single crystals with well-defined ground states became available, neutron scattering studies in particular have contributed new insights. Thus the nature of the A-phase was found to be AF order of incommensurate spin-density-wave type [3]. In A-type CeCu$_2$Si$_2$ the ordered magnetic moment is $\mu_{ord} \approx 0.1 \mu_B$ at temperatures well below $T_N \approx 0.8$ K. The propagation vector of the spin-density wave $\mathbf{\tau} = (0.215 0.215 0.53)$ is determined by the nesting properties of the Fermi surface. In neutron rocking scans performed on a three-axis spectrometer at $B = 0$
in A-type CeCu$_2$Si$_2$, the width of the magnetic Bragg peak at $\vec{Q}_{\text{AF}} = (0.22 0.22 1.45)$ turns out to be considerably broader than the resolution of the spectrometer for $T < 0.5$ K, while it is resolution-limited at $T > 0.5$ K before broadening again around $T_N$ ([4], figure 1). Line-broadening beyond the resolution indicates a limitation of the correlation length. At $T = 0.1$ K, the width of the magnetic Bragg reflection corresponds to a correlation length of the magnetic order of $\xi \approx 130$ Å. This behaviour coincides with a broad transition into the SC state starting at $T_{c,\rho} = 0.45$ K and extending down to 0.15 K, which was observed in measurements of the electrical resistivity [4]. The SC appears to be of percolative nature, causing a path of infinite conductance. From these results it can be concluded that filamentary SC limits the size of magnetically ordered regions in A-type CeCu$_2$Si$_2$, resulting in a reduced correlation length.

A/S-type CeCu$_2$Si$_2$ exhibits a transition into the AF ordered phase at $T_N \approx 0.7$ K, the propagation vector being approximately the same as in pure A-type samples, and a SC transition at $T_c \approx 0.5$ K. Neutron scattering and $\mu$SR measurements provide evidence that the SC volume fraction grows at the expense of the magnetic one on decreasing $T$ [6, 7]. This means that SC regions supersede magnetically ordered regions, and there is no microscopic coexistence of both phenomena. In contrast to the A-type crystal, the width of the magnetic Bragg peak at $\vec{Q}_{\text{AF}} = (0.21 0.21 1.46)$ at $B = 0$ is resolution-limited in the temperature range $0.4$ K $< T < 0.7$ K, thus also below $T_c$, before the magnetic intensity disappears completely for $T < 0.4$ K ([5], figure 2).

A finite magnetic correlation length is usually accompanied by a finite lifetime of the magnetic order, as indicated by $\mu$SR and NQR measurements on CeCu$_2$Si$_2$ powder samples [8, 9].
However, no broadening of the magnetic peak was observed in energy scans on a three-axis spectrometer [4]. Therefore, in order to corroborate the assumption of enhanced fluctuations of the AF order in A-type CeCu$_2$Si$_2$ below $T_{c,\rho}$, neutron spin-echo experiments were performed. The spin-echo technique yields a far better energy resolution than a three-axis spectrometer, with the loss of neutron intensity still being acceptable, and probes dynamical processes at time scales larger than $10^{-10}$ s.

2. Experiments and results

The CeCu$_2$Si$_2$ A-type ($m \approx 1.3$ g) and A/S-type ($m \approx 0.7$ g) single crystals studied were characterized by heat capacity measurements using the quasi-adiabatic heat-pulse method (figure 3). A-type CeCu$_2$Si$_2$ shows a transition into the AF ordered state, while no traces of a SC transition can be observed, supporting the supposition that there is only filamentary instead of bulk SC. A/S-type CeCu$_2$Si$_2$ exhibits a second order transition into the AF ordered state, similar to the A-type crystal, and a broadened first-order transition into the SC state, where magnetism is expelled.

Neutron spin-echo measurements were performed on the spectrometer IN11 at the Institut Laue-Langevin in Grenoble, France. Both crystals were mounted with the [1 1 0][0 0 1] plane being the scattering plane. The incident wavelength was $\lambda = 6.1$ Å. For both samples the same instrumental set-up was used: As the magnetic moments turned out to be aligned in the scattering plane, the neutron spin had to be rotated out of the plane. Four echo periods with eight points per period were recorded for different Fourier times at the position of the magnetic Bragg peak, $\vec{Q}_{AF} \approx (0.22 0.22 1.47)$. Measurements were performed in A-type CeCu$_2$Si$_2$ at $T = 72$ mK, this means at $T < T_{c,\rho}$, where the magnetic peak is broadened, and at $T = 600$ mK, this means at $T_{c,\rho} < T < T_N$, where the magnetic peak is resolution-limited, as well as in A/S-type CeCu$_2$Si$_2$ at $T = 450$ mK. This temperature was chosen as a compromise: The magnetic peak has still half the maximum intensity as found at $T_{c,\rho}$, although the sample is already superconducting. Nonetheless, due to the lower magnetic moment and the smaller mass of the crystal, the spin-echo intensity in A/S-type CeCu$_2$Si$_2$ is about 10 times smaller than in the A-type sample.

Spin echoes can be fitted by a sine function, with the echo amplitude corresponding to the intermediate scattering function $S(\vec{Q}_{AF}, t)$. Figure 4 displays the intermediate scattering function $S(\vec{Q}_{AF}, t)$ normalized to $S(\vec{Q}_{AF}, t = 0)$ for different Fourier times. With increasing Fourier time $t$ the intermediate scattering function decreases much faster in A-type CeCu$_2$Si$_2$ at $T = 72$ mK, where filamentary SC has developed, than at 600 mK or in the A/S-type crystal,
where there is either no or bulk SC. In each case $S(Q_{AF}, t)/S(Q_{AF}, 0)$ can be fitted by a single exponential decay, yielding lifetimes of $\tau \approx 7.5$ ns (A, 72 mK), 20 ns (A, 600 mK) and 22 ns (A/S, 450 mK). This corresponds to fluctuation rates of the AF order of 133 MHz, 50 MHz and 45 MHz, respectively.

In order to account for effects caused by inhomogeneities of the magnetic guide fields and the depolarization of the neutron beam at high Fourier times, $S(Q_{AF}, t)/S(Q_{AF}, 0)$ is usually normalized to the intermediate scattering function measured at the same $\vec{Q}$ position on the standard sample TiZr, the scattering of which is purely elastic. Unfortunately, the anomalous set-up with the neutron polarization rotated out of the scattering plane prevented us from observing spin echoes in TiZr. This is why this normalization cannot be performed, and the fitted lifetimes do not represent actual values, but lower limits to the real lifetimes. The qualitative conclusion, however, is not influenced by the missing normalization: The magnetic order is more dynamic in A-type CeCu$_2$Si$_2$ at low $T$ than at higher $T$ or in the A/S-type sample.

3. Conclusions
Neutron spin-echo measurements on A-type and A/S-type CeCu$_2$Si$_2$ indicate that, while there is quasi-static long-range AF order in the A/S-type as well as in the A-type crystal at $T_{c,\rho} < T < T_N$, the filamentary SC in A-type CeCu$_2$Si$_2$ at $T < T_{c,\rho}$ does not only limit the size of magnetically ordered regions, but also lead to a finite lifetime and an enhanced fluctuation rate of the antiferromagnetism.

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