Evidence for triplet superconductivity in Josephson junctions with ferromagnetic 
\text{Cu}_2\text{MnAl}-\text{Heusler barriers}

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We have studied Josephson junctions with barriers prepared from the Heusler compound \text{Cu}_2\text{MnAl}. In the as-prepared state the \text{Cu}_2\text{MnAl} layers are non ferromagnetic and the critical Josephson current density \(j_c\) decreases exponentially with the thickness of the Heusler layers \(d_F\). On annealing the junctions at 240°C the Heusler layers develop ferromagnetic order and we observe a dependence \(j_c(d_F)\) with \(j_c\) strongly enhanced and weakly thickness dependent in the thickness range 7.0 nm < \(d_F\) < 10.6 nm. We attribute this feature to a triplet component in the superconducting pairing function generated by the specific magnetization profile inside thin \text{Cu}_2\text{MnAl} layers.

Superconducting pairing functions with a symmetry different from conventional s-wave singlet pairing are in the focus of interest since the advent of BCS theory \cite{BCS}. However, unconventional pairing functions are rare in nature and experimental realizations had to wait until detecting systems like heavy fermions- \cite{HF}, high-\(T_c\) \textit{s} \(-\text{i} \text{r} \text{f} \) and the \text{Sr}_2\text{RuO}_4\-superconductors \cite{SrRuO4}. Unconventional pairing states might also be induced by the superconducting proximity effect at superconducting/ferromagnetic (S/F) interfaces (see \cite{Kohler, Zabel} for recent reviews). The exchange field in the ferromagnetic layer favors triplet pairing i.e. a superconducting condensate function with parallel spins. The penetration depth of superconducting triplet pairs into a ferromagnet in the limit of dirty metals with weak pair breaking scattering is given by \(\xi = \sqrt{\hbar D_F/2\pi k_B T}\) whereas the penetration depth of singlet pairs is limited by the ferromagnetic exchange energy \(E_{ex}\) via \(\xi_F = \sqrt{\hbar D_F/E_{ex}}\) \cite{Kohler}. Thus at low temperatures triplet pairs can penetrate deeply into a ferromagnetic metal.

In early experiments \cite{Russek} the resistance change of a ferromagnetic nanowire crossing a superconducting nanowire below the superconducting transition temperature was analyzed and the results were interpreted in favor of triplet superconductivity. The best experimental verification of long range triplet superconductivity would be the direct measurement of the decay length of the superconducting pairing function, as can be done by measuring the critical current \(j_c(d_F)\) in Josephson junctions with ferromagnetic barrier layers of variable thickness \(d_F\). However, up to now the experimental studies of \(j_c(d_F)\) did not indicate long range triplet superconductivity \cite{Kohler}. Singlet pairing with a transition from 0- to \(\pi\)-coupling (0- and \(\pi\) denoting the phase shift of the pair wave function across the barrier) could reasonably well explain all data.

Recent theoretical work suggested an interesting possibility to enhance the amplitude of the triplet component at the S/F interface drastically. Instead of a homogeneous magnetization profile as in Ref.\cite{Kohler} one should better use a ferromagnetic layer system with some intrinsic spin canting e.g. an in-plane \text{Neel} wall at the interface \cite{Sprungmann}, an S/F/S multilayer with non parallel orientation of the F-layers \cite{Zabel}, or a trilayer system with canted spins on both sides facing the S-layers \cite{Sprungmann}. The triplet component in the condensate function is enhanced by conversion of singlet Cooper pairs into triplet pairs by spin active interfaces \cite{Kohler}. The dominating superconducting wavefunction is the so called odd-frequency triplet pairing, i.e. a superconducting wave function even in space, even in spin but odd in time (or odd in the Matsubara frequencies). This exotic type of pairing state has been proposed originally for the pairing in the \text{He}^3 superfluid \cite{Kohler}.

Transport measurements on lateral Josephson junctions with half metallic \text{CrO}_2 \cite{Zabel} and the Rare Earth metal Ho \cite{Zabel} as the barrier material gave first indications of weak long range triplet contributions to the supercurrent. In the present Letter we will show that the unique properties of thin films of the Heusler alloy \text{Cu}_2\text{MnAl} can create a magnetization profile inside the \text{Cu}_2\text{MnAl} layer, which effectively generates triplet superconductivity.

Our SIFS-Josephson junctions with a lateral size of 10 \(\times\) 50 \(\mu\)m\(^2\) or 10 \(\times\) 200 \(\mu\)m\(^2\) were fabricated by de-sputtering and micro-structured by optical lithography and ion beam etching, as described in detail in Ref. \cite{Sprungmann}. The barrier between the two Nb-electrodes is composed of an about 1 nm thick \text{AlO}_x\-layer prepared by thermal oxidation of \text{Al} and a \text{Cu}_2\text{MnAl}\-layer with increasing thickness along the wafer axis. The thin \text{AlO}_x\ layer determines the normal resistance \(R_n\) and allows a precise analysis of the critical current without changing the physics discussed above. In a single preparation run we prepared several hundred junctions covering the thickness range from typically 5 nm to 15 nm for the \text{Heusler} alloy while keeping the other layer thicknesses constant. The I-V \textit{-characteristics} of the junctions were measured in a flow cryostat in the temperature range between 1.8 K and 6 K using home made, fully automatized electronics. The critical current was defined using a voltage drop criterion of \(\sim 0.5\ \mu\text{V}\). Transport measurements were done on Josephson junctions in the as-prepared state as well as after annealing at 240°C for 24 h in an UHV oven.
During this annealing process, which does not degrade the junction, the Heusler layers become ferromagnetic. The magnetic properties of the thin Heusler layers were studied by a SQUID magnetometer on similar but not micro-structured layer stacks.

The magnetization measurements of our samples with the Cu$_2$MnAl-layers in the as-prepared state reveal that they are non-ferromagnetic (see Fig.1). When prepared at room temperature Cu$_2$MnAl grows in the A2-structure i.e. with a random distribution of all atoms on a bcc lattice [17]. Nearest neighbor Mn-atoms are coupled by strong antiferromagnetic exchange interactions whereas Mn-next nearest neighbors with an Al-atom in-between are coupled ferromagnetically via a RKKY type interaction [18]. The competition of these two interactions and the randomness of the atomic distribution in the A2 structure gives rise to a spin glass type magnetic order as seen in Fig.1. Upon annealing at 240°C the unit cell symmetry transforms to the ordered L2$_1$-type Heusler structure which is combined of four interpenetrating fcc-sublattices occupied by Mn, Cu and Al, exclusively [17]. With this symmetry in Cu$_2$MnAl there are no Mn-Mn nearest neighbors and the ferromagnetic Mn-Al-Mn exchange interactions lead to ferromagnetic order with a magnetic moment of about 3.2 $\mu_B$ per Mn-atom and a ferromagnetic Curie temperature of 603 K [17].

The I-V-characteristics of a Josephson junction with a Heusler barrier thickness $d_F = 6.7$ nm before and after annealing is shown in Fig.2. At $T = 4.2$ K we observe the characteristics of an underdamped junction up to a thickness range of $d_F = 7.5$ nm in the as-prepared state and 9.5 nm after annealing. Beyond this thickness range the Josephson dynamics is overdamped, as typical for junctions with thicker ferromagnetic barriers due to the high subgap currents [22].
The thickness dependence of the critical current density in the as-prepared state is plotted in Fig. 3. One observes an exponentially damped curve with a decay length $\xi = 0.8$ nm, as typical for a system with strong pair breaking scattering. Strong inelastic pair breaking scattering in the Heusler layers must be expected, since in the spin glass state there is a high density of randomly oriented Mn magnetic moments. The decay length of a dirty metal ($\xi_m \ll \xi_F$) with inelastic pair breaking scattering ($E_{\text{inc}} \gg k_B T$) is given by $\xi_F = \sqrt{\hbar D_F/E_{\text{inc}}}$ with the diffusion constant $D_F$ and the scattering energy $E_{\text{inc}} = h/\tau_{\text{inc}}$ (scattering time $\tau_{\text{inc}}$). Single Cu$_2$MnAl layers in the as-prepared state have a residual resistivity of $\rho_m \approx 275 \ \mu\Omega\text{cm}$ and with the Fermi velocity $v_F$ taken from the literature we estimate $D_F$ and get $E_{\text{inc}} = 45$ meV for the inelastic scattering energy.

In Fig. 2 we show characteristic examples of Fraunhofer patterns for two junctions before and after annealing. The sample with $d_F = 6.7$ nm has a slight decrease of the critical current after annealing, as typical for the whole thickness range below $d_F = 7$ nm (see Fig. 3). This decrease is due to a slight increase of the AlO$_x$ barrier resistance $R_n$. From the observed change of the residual electrical resistivity upon annealing we find that $D_F$ increases by about a factor of $\sim 3$.

For the second sample with the thickness $d_F = 9.3$ nm in Fig. 3 the critical current in the annealed state is a factor of 14 larger than in the as-prepared state! This is very surprising, since the increasing ferromagnetic exchange field should suppress rather than enhance the critical current carried by singlet Cooper pairs. As plotted in Fig. 3 this enhancement is observed for all samples in the thickness range between 7.5 nm and 10.5 nm, the enhancement factor reaches a maximum value of nearly 100 close to $d_F = 10.5$ nm. Above 10.5 nm there is a sharp drop-off and the critical current approaches the value for the as-prepared state again. In order to verify our data we prepared two sample series in this thickness range with equal AlO$_x$ barriers. Both of them are included in Fig. 3 and follow the same curve, including the sharp drop-off at $d_F = 10.5$ nm.

The theory for singlet pairing of junctions with a ferromagnetic barrier of variable thickness $d_F$ in the dirty limit predicts the functional dependence

$$j_c(d_F) = j_0 |\cos(d_F/\xi_{F2})| e^{-d_F/\xi_{F1}}$$

with the decay length $\xi_{F1}$ and the oscillation length $\xi_{F2}$ [2]. Without pair breaking scattering both lengths are given by the penetration depth $\xi_F$ [2]. The first term in equation describes the Josephson phase transition between 0 and $\pi$ and leads to a deviation of the current density from an exponentially damped curve towards smaller values when approaching the transition range, in sharp contrast to what we have observed for the annealed state in Fig. 3. For the decay length in the plateau region in Fig. 3 we derive a value of $\xi = 5.7$ nm, which in the framework of singlet superconductivity cannot be interpreted in any reasonable way. For singlet superconductivity the critical current density should decay following the initial slope in Fig. 3, i.e., with a decay length $\xi_{F1} \leq 1$ nm. We thus are led to the conclusion that in the plateau region of Fig. 3 an additional component of the supercurrent appears, which compensates the exponentially damped singlet supercurrent. The theoretical work [3, 4] strongly suggests that
this additional component should be identified as an odd-frequency triplet supercurrent. The superposition of a short range singlet and a long range triplet component \( (j_s(d_F)) \) becomes more apparent if we assume a model function for the singlet supercurrent \( j_s(d_F) \) corresponding to Eq. 1 and subtract it from the total supercurrent. For the dotted line in Fig 8 we assumed an exchange and a scattering energy of \( E_{ex} = E_{sc} \approx 60 \text{ meV} \), which seems reasonable, because in many alloys the exchange and the inelastic scattering energy have the same magnitude [8].

As a further piece of evidence for the occurrence of unconventional superconducting pairing in the plateau region of Fig 8 we show the temperature dependence of the critical current in Fig 4. For \( d_F < 7 \text{ nm} \) \( j_c \) increases with decreasing \( T \), this is the conventional behavior for a singlet supercurrent. Within the plateau region we observe a broad maximum in \( j_c(T) \) at about 4.5 K with \( j_c \) decreasing for lower temperatures. As shown in the theoretical work [22] this anomalous temperature dependence results from the competition of different contributions to the supercurrent in junctions with unconventional pairing. For instance, it can be caused by the superposition of a triplet and a singlet supercurrent with opposite Josephson coupling.

Our assumption of the appearance of triplet supercurrent being responsible for the \( j_c \)-plateau in Fig 8 implies that inside the Heusler layers there is a magnetization profile which very effectively generates triplet superconductivity. Referring to Fig 4 one finds that the \( j_c \)-plateau coincides with a narrow thickness range just above the onset of ferromagnetic order. For slightly larger thicknesses, but still rather small values of the saturation magnetization, the extra critical current becomes rapidly suppressed again. This indicates that the microscopic origin for the conversion of the singlet into triplet pairs depends sensitively on the magnetization profile inside the Heusler layers. In the thickness range just above the onset of ferromagnetism, ferromagnetic order already exists in the core of the Heusler layers, whereas at the interfaces spin glass order still prevails. The coupling between the two types of magnetic order will induce a small ferromagnetic magnetization close to the interfaces, however, with some canting of the local magnetization direction because of the coupling to the coexisting random spin glass type of order. These canted interface moments might be identified as the “spin active zone” needed for the conversion of singlet pairs into triplet pairs [12]. Thus a picture emerges which qualitatively resembles the three layer ferromagnetic system in the theoretical work of Ref. [11], where triplet pairing is also found in a narrow thickness range of the spin active interfaces only.

In summary, we have shown that making use of the unique magnetic properties of Cu2MnAl-Heusler layers we can create a magnetization profile inside the Heusler layer which can very effectively generate triplet superconductivity. Aside from the intriguing problem of the detailed microscopic origin of this process, from a practical point of view we want to stress that the preparation of Heusler films is perfectly compatible with established preparation routes for Josephson junctions, unlike other possible barrier materials like CrO2 or Ho. Thus Heusler layers open a new route to routinely prepare Josephson junctions with nearly pure triplet supercurrents for applications and basic research issues.

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