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LETTER TO THE EDITOR

Phonon echoes in an amorphous superconductor in the vortex state

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Abstract. Phonon echoes are observed in the amorphous superconductor $CuZr_2$ ($T_c = 2.4$ K) in the vortex state at 10 mK. The phase-memory time T_2 of the tunnelling states and their relaxation time T_1 are measured at 10 mK. The latter is also obtained from a saturation–recovery experiment. T_1 and T_2 vary exponentially as a function of magnetic field between 0 and 5 kOe. This effect is explained by considering an interaction between the tunnelling states and the vortices that arise from the normal-electron density distribution near the vortex core.

Phonon echoes (the acoustic analogue of spin or photon echoes) are a powerful tool to study the relaxation rates of the tunnelling states (TS) in amorphous matter [1]. They have been observed in fused silica [2, 3] and in magnetic rare-earth glasses [4, 5]. In normal conducting metallic glasses, the strong coupling between the electrons and the TS leads to very short relaxation times and the phonon echoes cannot be observed [6, 7]. In superconductors, the appearance of an energy gap is predicted to inhibit the fast-electron-TS process [8]. This prediction has been confirmed recently by a phonon echo experiment in superconducting amorphous $Pd_{30}Zr_{70}$ [9]. The authors of [9] have measured the TS relaxation times T_1 and T_2 well below T_c in zero magnetic field and have shown there is no evidence for an electronic contribution.

Amorphous superconductors are of the second kind with large κ. So they exist in the vortex state in a large magnetic-field range [10]. In this Letter we report on a phonon-echo experiment in superconducting amorphous $CuZr_2$ ($T_c = 2.4$ K) in the vortex state (i.e. in a magnetic field H such that $H_{c1} \ll H < H_{c2}$). The relaxation times T_1 and T_2 vary exponentially as a function of magnetic field. We show that this behaviour is evidence for the existence of an interaction between the TS and the vortices. This interaction, which to our knowledge has never previously been considered, arises from the normal-electron density distribution near the vortices, which varies exponentially when moving away from the vortex core.

The sample was obtained from a sputtered $CuZr_2$ film of thickness 40 μm (T_c = 2.4 K). It was formed from many 2 × 6 mm^2 platelets cut from the $CuZr_2$ film and pressed together in a copper–beryllium clamp (without any glue). The end faces of the platelets were polished flat and parallel. Then, Y-cut quartz transducers were bonded to these faces. The magnetic field was normal to the platelets.

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We have performed sound velocity measurements to determine the parameters of the TS. These measurements have been made with a phase comparison method at 660 MHz and for acoustic shear waves. In zero magnetic field, the sound velocity varies as $\ln T$ in the temperature range 50–500 mK, as usual [11]. From this temperature dependence the coupling parameter $nM^2/\rho V^2$ can be determined (n is the TS density, M is the coupling strength between the TS and the acoustic wave, ρ is the specific mass and V is the sound velocity). Thus, we have obtained $nM_t^2 = 4 \times 10^7\,\mathrm{erg\,cm^{-3}}$ ($\rho = 6.95\,\mathrm{g\,cm^{-3}}$ and $V_t = 1.94 \times 10^5\,\mathrm{cm\,s^{-1}}$). Above 500 mK, the velocity increases still slightly, reaches a maximum around 1 K and then passes through a shallow minimum around 2.4 K (the transition temperature). In high magnetic field (50 kOe, value higher than $H_{c2} = 35$ kOe), there is the same $\ln T$ dependence above 50 mK but with a slope reduced by a factor 2. This behaviour has been already observed in disordered NbZr [12]. In the vortex state (i.e. for $H_{c1} \ll H < H_{c2}$) the slope of the logarithmic temperature dependence of the velocity decreases with increasing the magnetic field (which is itself proportional to the vortex number). Its value is 3% smaller than that of the zero magnetic field at 3 kOe and 16% smaller at 20 kOe.

We have observed phonon echoes at 10 mK, 660 MHz and in magnetic field up to 5 kOe. In order to avoid the problems related to the vortex pinning, the magnetic field was set up above T_c, and then the sample was cooled down to 10 mK. The phase-memory time T_2 was measured from the decay of the spontaneous phonon echo as a function of the time separation τ_{12} between the two pulses. The TS–lattice relaxtion time T_1 was measured from the decay of the stimulated phonon echo as a function of the time delay τ_{13} of a third pulse (1). Figure 1 shows the variation of T_2 as a function of magnetic field. T_2 varies as $\exp(-H/H_0)$ from 5 μs to 0.43 μs between 0 and 5 kOe. The experimental value of H_0 is 1.9 kOe. Above 5 kOe, the phase-memory time is too short to be measured. Figure 2 shows the variation of T_1 as a function of magnetic field. Again, the variation is exponential according to $\exp(-H/H_1)$ with $H_1 = 1.1$ kOe. In figure 2, we have extrapolated the straight line up to 5 kOe. Although we have not measured T_1 at this field, we have seen that T_1 was certainly shorter than 2 μs.

We have also performed a saturation–recovery experiment which is another way to obtain information on the TS–lattice relaxation times. It consists in saturating the TS system with a first pulse and afterwards measuring the variation of the attenuation of a probing pulse as a function of the delay time between the two pulses [1]. In principle, the recovery time is a direct measurement of T_1. Experimentally, there is a distribution of relaxation times and the decay is not exponential. We have chosen for T_1 the characteristic time of the initial decay (which corresponds to the shortest T_1). Our results are reported in figure 2. As usual, the values obtained from saturation recovery are larger than those obtained from stimulated echoes [1]. Here again, the variation of T_1 as a function of magnetic field is exponential up to 5 kOe according to $\exp(-H/H_1)$ with $H_1 = 0.9$ kOe.

Our results in zero magnetic field are similar to those of [9]. T_2 is the same as in PdZr and T_1 is five times larger. Although these values are smaller than the corresponding ones in fused silica [1, 5], the authors of [9] have shown that they can be explained by taking into account the one-phonon process alone and using a relaxation rate [13]

$$T_1^{-1}(\mathrm{ph}) = (1/2\pi\rho\hbar^4)(M_l^2/V_l^5 + 2M_t^2/V_t^5)\,E^3 \coth(E/2k_B T) \qquad (1)$$

where V_l and V_t are the longitudinal and transverse sound velocities, respectively. In normal conducting metallic glasses there is a very high relaxation rate due to interaction with conduction electrons and which is given by [6]

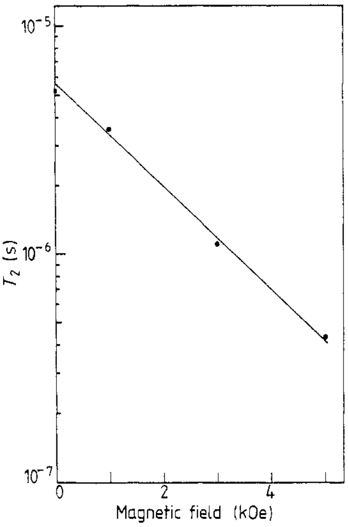

Figure 1. Phase-memory time T_2 obtained from spontaneous phonon echoes as a function of magnetic field at 10 mK and with acoustic shear waves of 660 MHz.

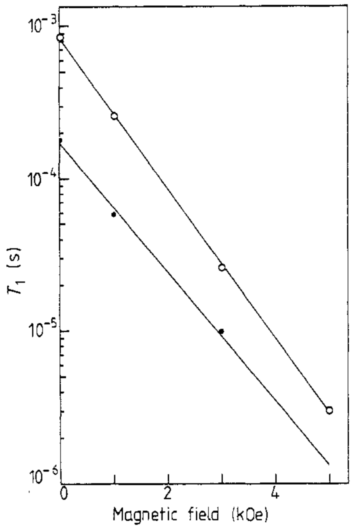

Figure 2. Relaxation time T_1 obtained from stimulated phonon echoes (full circles) and from saturation recovery (open circles) as a function of magnetic field at 10 mK and with acoustic shear waves of 660 MHz.

$$T_1^{-1}(e) - (\pi/4\hbar)(Nv_\perp)^2\, E \coth(E/2k_BT) \tag{2}$$

where N is the electronic density of states at the Fermi level and $v_\perp$ is the off-diagonal scattering matrix element. In the superconducting state it has been predicted that the electronic contribution varies below T_c according to [8]

$$T_1^{-1}(e) = (\pi/\hbar)(Nv_\perp)^2 k_BT[\exp(\Delta/k_BT) + 1]^{-1} \tag{3}$$

where Δ is the temperature-dependent superconducting energy gap. In the present case $T_c = 2.4$ K and at 10 mK this contribution is completely frozen out. Such a behaviour has been observed recently in PdZr [9].

Before looking at the behaviour of the TS in magnetic field, we have to determine the parameters of $CuZr_2$ in the superconducting state. The critical temperature is $T_c = 2.4$ K and the upper critical magnetic field is $H_{c2} = 35$ kOe [14]. Taking for the density of states at the Fermi level $N = 1.8$ states eV^{-1}/atom [15], we get for the thermodynamic critical field $H_c = 280$ Oe. In the framework of the Ginzburg–Landau theory, the coherence length ξ, the penetration depth λ and the parameter κ can be determined [10]. We have obtained $\kappa = 90$, $\xi = 100$ Å and $\lambda = 0.9\,\mu$m. The penetration depth is indeed much larger than the coherence length. With these parameters the first penetration field is $H_{c1} = 10$ Oe. Thus, the magnetic field range 1–5 kOe of our experiments satisfies the condition for the existence of the mixed state $H_{c1} \ll H < H_{c2}$. In the mixed state the vortices form a triangular array. The distance d between neighbouring vortex lines is given by [10]

$$B = \Phi_0 n_L = (2/\sqrt{3})\Phi_0/d^2 \tag{4}$$

where Φ_0 is the flux quantum and n_L is the vortex density. At 3 kOe (in the middle of our magnetic-field range), equation (4) gives $n_L = 1.45 \times 10^{10}$ vortices cm^{-2} and $d =$

900 Å. Assuming that each vortex line is equivalent to a normal region of radius ξ, the ratio of the normal to the superconducting volume is found to be 0.05 at 3 kOe. This ratio is consistent with the small decrease of the slope of the logarithmic dependence of the sound velocity reported above.

Since we are in the experimental conditions where $\xi < d < \lambda$, each TS is at a distance to the nearest vortex of the order of ξ and much shorter than λ. Then, one can expect for an exponential effect a connection with the coherence length rather than with the penetration depth. To be more explicit in this statement we can evaluate the mean distance of the resonant TS from the nearest vortex. The TS density in $CuZr_2$ is $n = 5 \times 10^{32}\,erg^{-1}\,cm^{-3}$ [14]. In zero magnetic field we have found $T_2 = 5\,\mu s$. Hence, the density of the resonant TS is about $n\hbar/T_2 = 5 \times 10^{11}\,cm^{-3}$. Assuming a cubic TS lattice, the mean distance of the resonant TS between each other is then 1.2 μm, which is much larger than the distance between vortices in our magnetic-field range. In a plane perpendicular to the vortex lines, these form a triangular network and there are many fewer resonant TS than triangular cells. Assuming that the TS are located randomly in this plane, one finds easily that their mean distance $\langle r \rangle$ from the nearest vortex is $d/3$ where d is the distance between vortices. At 3 kOe this gives a value $\langle r \rangle/\xi = 3$. This small value implies that we cannot treat the vortex line as a simple normal conducting cylinder but we have to consider the spatial variation of the order parameter $\psi(r)$ where r is the distance from the centre of the vortex [10]. A reasonable approximation to $\psi(r)$ is $\tanh(r/\xi)$ [16]. Since the number of electrons in the superconducting state is proportional to $|\psi(r)|^2$, we can write for the density n_N of normal electrons as a function of the distance from the vortex centre

$$n_N = N(1 - \tanh^2(r/\xi)) \tag{5}$$

where N is the electron density at the Fermi level. At 3 kOe we have seen that $\langle r \rangle/\xi = 3$. For this value (5) gives $n_N = 10^{-2}\,N$. This electronic density of states is non-negligible on account of the strong effect of the normal electrons on the TS relaxation. Using (2) but now replacing N by n_N, we can obtain the electronic contribution to the relaxation rate. Taking $Nv_\perp = 0.85$ (as for PdZr [6]), we get $T_1 = 4\,\mu s$ at 660 MHz and 3 kOe. As compared with our experimental value of 10 μs obtained from the stimulated-echo experiment, it is a satisfactory order of magnitude. Trying to go further with (5) can lead for $r > \xi$ to an exponential dependence for T_1 but taking then for r the mean value $\langle r \rangle = d/3$ is less satisfactory and does not give the experimental magnetic-field dependence. It is not surprising on account of the roughness of the approximations. Replacing the distribution of the distances of the TS from the vortices by a mean distance is oversimplifying. Moreover, our experiments are in a magnetic-field range where the vortices form a rather dense lattice and the interactions extend to distant neighbours. Nevertheless, the present evaluation shows clearly that the electronic contribution near the vortices is adequate to account for the T_1 measured.

Let us consider now our results on T_2. The phase-memory time also varies exponentially with H. This can be explained by taking into account spectral diffusion [17]. This process assumes that due to elastic dipolar interaction the splitting energy of the resonant TS is modulated when the neighbouring non-resonant TS change their state with a rate T_1^{-1}. These latter TS which are thermally active ($E < 2k_BT$) are more numerous than the resonant ones. They undergo the electronic contribution of the vortices and their T_1 are the shorter as they are near the vortex cores. Thus, it is not surprising to find again the same magnetic-field dependence as for T_1.

In conclusion, phonon-echo experiments in superconductors in the vortex state are a powerful tool to study the vortices and the TS. We have shown there is a strong interaction of the TS with the vortices which arises from the normal-electron density distribution near the vortex lines. Although the physical system is complex, it is in return beautiful. One cannot approximate as often the vortex lines as normal conducting cylinders but one must consider the spatial variation of the order parameter. By acting on the coherence length one can hope to obtain more information on the microscopic nature of the TS, in particular their spatial extension.

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