Phonon-Induced Spin Flip in Extremely Thin Superconducting Al Tunneling Junctions in High Magnetic Fields

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Superconducting Al tunneling junctions of film thicknesses of about 30 Å were used as phonon generators and detectors in parallel magnetic fields up to 5 T. These extremely thin tunneling junctions show Zeeman splitting of the energy gap according to the splitting of states with parallel and antiparallel spin. From the dependence of the detection signal on the magnetic field in phonon spectroscopy and phonon pulse measurements it is concluded that quasiparticle-phonon transitions can be accompanied by a spin flip. [S0031-9007(96)01204-5]

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In this Letter we report the first observation of a phonon-induced quasiparticle spin flip in superconducting tunneling junctions (STJs). STJs can be used for spectroscopic investigations with acoustic phonons in the frequency range from 80 GHz to several THz [1,2]. Because of the sharp energy gap of the superconducting material (here Al) the phonon emission spectrum of a STJ under a bias voltage shows a sharp voltage determined high frequency edge which by modulation (differentiation) techniques experimentally corresponds to a quasimonochromatic line. By varying the applied bias voltage the position of the quasimonochromatic line can be moved over a wide frequency or energy range. This tunable phonon source together with a second STJ for phonon detection allows high resolution phonon absorption spectroscopy. Another possibility for frequency tuning arises from applying an external parallel magnetic field to the superconducting films. In this case the narrow line of recombination phonons with the gap energy of 2Δ can be shifted [3]. However, this method has the disadvantage of broadening the phonon line very strongly with increasing magnetic field, thus severely reducing frequency resolution. This is caused by a smearing of the energy gap edge due to the influence of the field on the quasiparticle orbits.

As demonstrated by Meservey *et al.* [4,5], the influence of the magnetic field on the quasiparticle orbits can be strongly suppressed by using very thin superconducting films in a parallel field or strongly disturbed materials with a very short electron mean free path. In these cases the influence of the magnetic field on the quasiparticle spins becomes dominant causing a Zeeman splitting of the energy gap of $2\mu_B B$ according to the splitting of the states parallel and antiparallel to the applied field B. In addition, the critical field B_c can be considerably enhanced (Al over 5 T); the Zeeman splitting of the energy gap can be observed in the current-voltage characteristics of the STJ.

The idea of the present work now was to use very thin STJs for phonon detection experiments in magnetic fields in order to study the phonon emission and detection mechanism of STJs in the presence of Zeeman splitting. A special aspect was to investigate the possibility of an improved frequency tuning by the external magnetic field which is illustrated in Fig. 1 for the phonon detection process: In zero field only phonons with an energy that exceeds the energy gap 2Δ of the detector junction can be absorbed. In the presence of Zeeman splitting the effective energy gap for a transition under quasiparticle spin flip is reduced; however, for transitions without quasiparticle spin flip nothing changes. As the magnitude of the splitting $2\mu_B B$ is adjustable by the external magnetic field the detector threshold can be tuned. Similar considerations apply to the generator junction. Since there is no spin flip of quasiparticles during oxide barrier tunneling [6], the existence of spin flip accompanied quasiparticle-phonon transitions would be a necessary condition for a magnetic field frequency tuning of phonons.

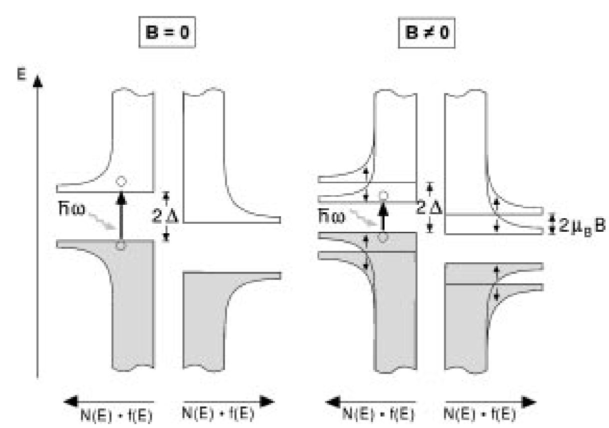

FIG. 1. Phonon detection using a superconducting tunneling junction with and without external magnetic field.

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The experiments we performed can be divided into two classes: phonon spectroscopy measurements to obtain energy resolved information of the phonon emission and absorption processes and phonon pulse measurements to differentiate between the interaction of the longitudinal and the transverse mode. The mode separation is possible because of the different sound velocities of the modes in using short pulses (120 ns) and a sufficiently long propagation path, here 8 mm (thickness of crystal).

For phonon spectroscopy experiments we evaporated almost identical superconducting Al-Al_2O_3-Al tunneling junctions with film thicknesses of 30 to 120 Å on the opposite sides of polished Ge or Si crystals. The crystals were oxygen doped in order to use the resonant phonon absorptions of the oxygen atoms as reference frequencies [7,8].

By applying a bias voltage $U \geq 2\Delta_{\rm gen}/e$ to the generator junction, excited quasiparticles are injected and decay via relaxation and recombination processes under emission of phonons. Part of these phonons reach the detector junction on the opposite side of the crystal. In the detector junction phonons with an energy $\Omega \geq 2\Delta_{\rm det}$ are absorbed by splitting Cooper pairs thus increasing the tunneling current in the "thermal regime." Energy-sensitive detection is obtained by recording the derivative of the detector signal with respect to the generator voltage using an appropriate modulation technique [9]. This differential generator-detector signal furthermore is simply referred to as "detection signal."

For our phonon pulse experiments we replaced the generator junction by a metallic film (Cr or Au). Heating this film with short current pulses yields phonon pulses with a broadband thermal emission spectrum. Phonon detection occurred in the same way as in the phonon spectroscopy experiments.

The STJs were produced by thermal evaporation of pure Al onto liquid-nitrogen-cooled crystals under ultrahigh vacuum conditions. The experiments were performed at temperatures below 0.5 K in a ^{3}He cryostat with a built-in superconducting magnet coil able to produce homogeneous magnetic fields up to 5 T. The parallelism of the field in the region of the sample was better then 10 sec of arc.

First we recorded the current-voltage characteristics in order to check whether the thinnest STJs showed the expected Zeeman splitting in an external parallel magnetic field. In Fig. 2 the magnified lower part of *I*-*V* characteristics of a 30 Å STJ is displayed in comparison to the characteristics of a 120 Å STJ: In contrast to the characteristics of the 120 Å STJ which showed strong smearing of the energy gap with increasing magnetic field the *I*-*V* characteristics of the 30 Å STJs showed an excellent sharpness of the energy gap up to the highest applied fields of 5 T. From the *I*-*V* characteristics of the 30 Å STJ we found in addition that with increasing magnetic field a small signal step moves from the main onset corresponding to the energy gap to lower voltages. This small step arises from so-called spin-mixed states

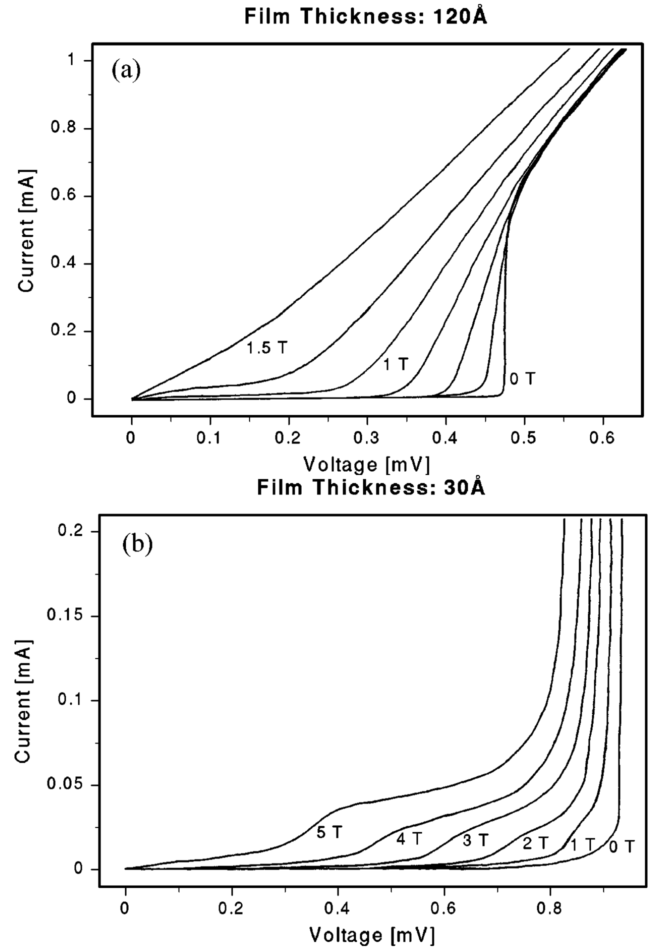

FIG. 2. Current-voltage characteristics of a 120 Å junction (above) and a 30 Å junction (below, magnified lower part of characteristics) in a parallel magnetic field.

due to spin-orbit coupling. The energy distance between the small step and the main onset agrees very well with the Zeeman splitting energy. Furthermore in comparison to the 120 Å characteristics an enhancement of the energy gap as well as of the critical temperature was observed.

In our phonon spectroscopy experiments we observed that some of the detection signal structures were drastically changed when applying an external magnetic field. Figure 3 shows the detection signal of a 30 Å STJ as a function of the energy of the relaxation phonons with and without magnetic field. Structures A and B refer to the onset of recombination and relaxation phonons, respectively; structure C results from resonant phonon absorption caused by interstitial oxygen in the germanium crystal. It can be seen that as a consequence of the sharp energy gap the zero field signal structures can be clearly recognized in all applied fields. However, apart from a slightly increasing slope of the signal with growing field strength there appear additional structures close to A and B, especially visible above 2 T. The influence of the magnetic field is seen more clearly in Fig. 4 showing the difference between the detection signal and its smoothed

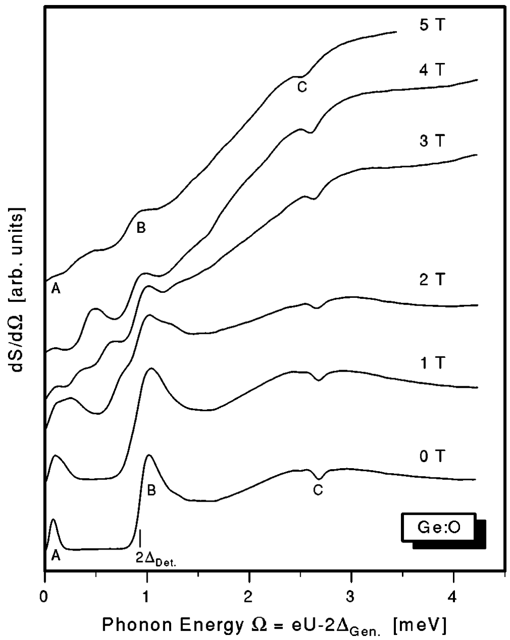

FIG. 3. Detection signal of a 30 Å junction on Ge:O in a magnetic field. A: Recombination phonons. B: Relaxation phonons. C: Resonant phonon absorption due to interstitial oxygen. Measurement curves have been displaced vertically for clarity. *X* axis is scaled in units of phonon energy of the relaxation phonons.

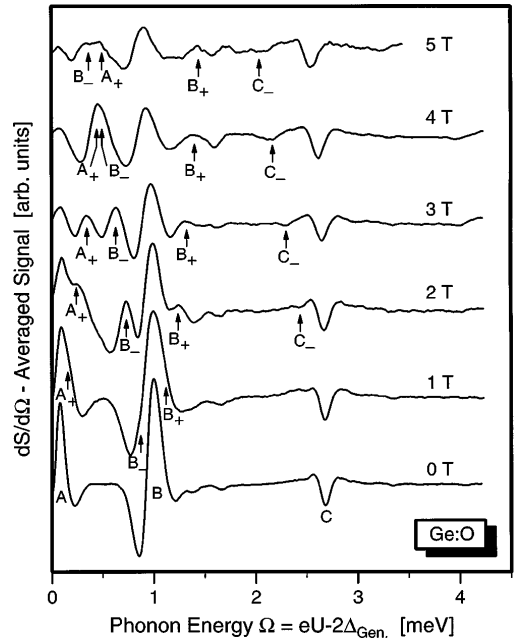

FIG. 4. "High pass filtered" detection signal of a 30 Å junction on Ge:O in a magnetic field. A: Recombination phonons. B: Relaxation phonons. C: Resonant phonon absorption due to interstitial oxygen. Measurement curves have been displaced vertically for clarity. *X* axis is scaled in units of phonon energy of the relaxation phonons.

function with respect to the phonon energy within a bandwidth of 0.4 meV. This corresponds to a "high pass filtering." From regarding Fig. 4 it becomes obvious that the additional structures arise from a splitting of the structures A and B. The energy distance from the split-up structures A_+, B_-, B_+ to their "parent structures" A and B, respectively, agrees very well with the Zeeman splitting energy $2\mu_B B$. From a more detailed analysis of the detection signal it can be seen that structure C at 2.66 meV also shows Zeeman splitting, although the relative magnitude of the splitting structure C_- is found to be significantly smaller than those of the others. In the additional measurement in Si:O with an absorption line at 3.63 meV we found an even smaller relative magnitude of the corresponding high frequency splitting structure. In summary, it turned out that all investigated splitting structures show a decreasing relative magnitude with increasing phonon energy.

The spin-mixed states in the density of states distribution contribute less than 2% of the total density of states. This follows from *I-V* characteristics like Fig. 2 which are typical for the generator as well as for the detector tunneling junction. Therefore the phonon splitting structures of Fig. 4 with up to 50% of the zero field structures cannot be explained by a contribution of spin-mixed states. However, the observed splitting structures can be well explained by permitting quasiparticle spin flips during quasiparticle-phonon transitions. Because generation as well as absorption of phonons could be influenced by a quasiparticle spin flip the phonon spectroscopy experiments alone do not allow conclusions whether these spin flips accompany only emission or only absorption processes or both.

The decrease of the relative magnitude of the splitting structures with increasing phonon energy indicates a decreasing probability for quasiparticle spin flip. It is suggested that this is due to a shortening of the quasiparticle relaxation time with increasing energy. In the high energy region the quasiparticle relaxation time then could drop under the required time for a spin flip of $h/2\mu_B B$ thus reducing the spin-flip probability.

In phonon pulse measurements we observed that the detection signal of the transverse modes increased significantly stronger with the applied magnetic field than the detection signal of the longitudinal mode. An increased detection signal can be explained by a reduced detector threshold $2\Delta_{\mathrm{det}} - 2\mu_B B$ for the absorption of phonons under magnetic field due to phonon-induced quasiparticle spin flip during the absorption process (see also Fig. 1). Therefore we conclude that the absorption of transverse phonons accompanied by a quasiparticle spin-flip scattering is stronger than the same mechanism with longitudinal phonons.

Additionally, we observed a stronger increase of the mean quasiparticle lifetime of the transverse detection signal (calculated from the pulse form) on the magnetic field compared to the longitudinal signal. This is attributed to an enhanced recombination quasiparticle lifetime for quasiparticles recombining across the reduced energy gap in the magnetic field because these quasiparticles can recombine only under emission of transverse phonons accompanied by a quasiparticle spin flip. In addition, the recombination lifetime increases with decreasing phonon energy by the corresponding reduction of the phonon density of states which determines the electron-phonon interaction rate. A simple classical electromagnetic approach for transverse phonon-electron interaction has been used by Pippard [10].

The results of a theoretical investigation concerning the quasiparticle-spin-phonon interaction mechanism are presented in the following Letter [11].

With regard to future applications the presented results show that, apart from the STJ-generator bias voltage, an external magnetic field also is suitable for phonon frequency tuning, in case that it can be applied parallel to very thin superconducting tunneling junctions. Thus under these conditions phonon spectroscopy with superconducting tunneling junctions is also possible in high magnetic fields.

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[1] W. Eisenmenger and A. H. Dayem, Phys. Rev. Lett. **18**, 125 (1967).
[2] W. Eisenmenger, Phys. Acoust. **XII**, 79 (1976).
[3] V. Narayanamurti and R. C. Dynes, Phys. Rev. Lett. **27**, 410 (1971).
[4] R. Meservey, P. M. Tedrow, and P. Fulde, Phys. Rev. Lett. **25**, 1270 (1970).
[5] P. Fulde, Adv. Phys. **22**, 668 (1973).
[6] P. M. Tedrow and R. Meservey, Phys. Rev. Lett. **27**, 919 (1971).
[7] E. Dittrich, W. Scheitler, and W. Eisenmenger, Jpn. J. Appl. Phys. **26**, 873 (1987).
[8] M. Gienger, M. Glaser, and K. Lassmann, Solid State Commun. **86**, 285 (1993).
[9] W. Forkel, M. Welte, and W. Eisenmenger, Phys. Rev. Lett. **31**, 215 (1973).
[10] A. B. Pippard, Proc. R. Soc. London A **257**, 165 (1960); cf. also [2].
[11] C. Grimaldi and P. Fulde, following Letter, Phys. Rev. Lett. **77**, 2550 (1996).