Point-contact spectroscopy of the phononic mechanism of superconductivity in YB$_6$

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Abstract
Lortz et al (2006 Phys. Rev. B 73 024512) have utilized specific heat and resistivity measurements as ‘thermal spectroscopies’ to deconvolve the spectrum of the electron–phonon interaction in YB$_6$, assuming a major role of the low frequency phonon mode in mediating superconductivity. Here, we present direct point-contact spectroscopy studies of the superconducting interaction in this system. As a result, the normalized superconducting gap reveals a strong coupling with $2\Delta/k_B T_c = 4$ and, moreover, the spectra contain nonlinearities typical of the electron–phonon interaction at energies around 8 meV. The measurements in a magnetic field evidence that the phonon features found in the second derivative of the current–voltage characteristics are due to the energy dependence of the superconducting energy gap as their energy position shrinks equally as the gap is closed. This provides direct proof that the superconducting coupling in the system is due to the low energy Einstein-like phonon mode associated with the yttrium ion vibrations, in perfect agreement with determinations from bulk measurements.

1. Introduction
The surprising discovery of superconductivity in MgB$_2$ at almost 40 K [1] has re-attracted attention to the superconductors with an electron–phonon interaction mechanism. Due to their eight phonon branches and a high density of scatterers in the form of boron atoms the metallic and non-magnetic hexaborides were regarded once as possible candidates for high-temperature superconductors [2]. These expectations were not met as YB$_6$ has the highest transition temperature, $T_c = 8.4$ K, among them [3] and isostuctural LaB$_6$ with a very similar electronic structure is not superconducting at all [4]. It was believed for a long time that a dominant contribution to the electron–phonon interaction (EPI) in hexaborides leading eventually to superconductivity came from the boron sublattice with a lot of phonon branches stretched up to 160 meV [5]. However, later, Mandrus et al [6] noticed that due to the large spaces of the metal atoms among the boron octahedral cages the metal atoms can develop large unharmonic vibrational amplitudes with strong EPIs. Then, LaB$_6$ or YB$_6$ can be modeled as a Debye solid with the rigid boron framework and the metal ions can be treated as independent harmonic oscillators (Einstein oscillators). The local vibrational modes of the La ions are the most important for the explanation of the low-temperature resistivity and heat capacity in LaB$_6$, as was also suggested by the neutron experiments of Smith et al [7] showing rather rapid flattening of the acoustic modes near 13 meV due to non-interacting vibrations of La ions. Some of us [8] have measured the EPI function of LaB$_6$ directly by point-contact spectroscopy (PCS), detecting the whole set of eight modes starting from the prominent peak at 13–15 meV up to 160 meV with a resulting electron–phonon coupling constant of $\lambda \approx 0.15$, showing very weak electron–phonon coupling in the system. In the case of YB$_6$ the rattling motion of the Y ion is now supposed to be dominant in the electron–phonon coupling. Based on this assumption Lortz et al [9] have exploited specific heat
and resistivity measurements as ‘thermal spectroscopies’ to deconvolve the phonon density of states and the spectrum of the electron–phonon interaction in $YB_6$. Their results suggest that the superconductivity is mainly driven by a low lying phonon mode (at $\approx 8$ meV), which is associated with the yttrium ions in oversized boron cages. The lower vibration frequency of lighter Y in $YB_6$ than the frequency of La vibrations in $LaB_6$ has been attributed to the weaker bond of Y due to its smaller radius in the same boron cage. The electronic specific heat reveals that $YB_6$ is a strong superconductor with a reduced energy gap of $2\Delta/k_BT_c \approx 4.1$ and a coupling constant of $\lambda \approx 1$. Three spectroscopy experiments performed on $YB_6$ have been published to the best of our knowledge so far. Kunii et al [10] determined from GaAs point contacts on $YB_6$ the superconducting gap $\Delta = 1.22$ meV, which with the bulk $T_c = 7.1$ K (the local $T_c$ was not measured) yields $2\Delta/k_BT_c \approx 3.8$. An EPI peak at 11 meV was also observed with the gap energy subtracted. The photo-emission spectroscopy results of Souma et al [11] indicate similar conclusions. Schneider et al [12] prepared a tunneling sandwich on $YB_6$ film naturally oxidized with an In top electrode and obtained $2\Delta/k_BT_c \approx 4$, with an intense EPI peak at 8 meV in discrepancy with Kunii and $\lambda \approx 0.9$. No significant contribution to the EPI was detected above 16 meV.

We present a detailed experimental study on $YB_6$ single crystal with a $T_c$ of 7.5 K via point-contact (PC) spectroscopy. A single s-wave superconducting energy gap with a reduced value of $2\Delta/k_BT_c$ close to 4 together with its classical temperature and magnetic field dependence have been found. Moreover, an electron–phonon interaction peak has been directly observed in the second derivative of the current–voltage characteristics in the superconducting state. Upon application of magnetic field the energy position of this peak shifts to lower energies. From data analysis the magnetic field dependence of the superconducting energy gap has been inferred. Importantly, the energy position of the EPI feature shifts with increasing magnetic field to lower energy in exactly the same way as the superconducting gap is closing. This provides direct proof that the low energy phonon mode near 7.6 meV is mediating superconducting pairing, in agreement with the conclusions of Lortz et al.

2. Point-contact spectroscopy of strong coupling superconductors

A micro-constriction between two metals with a contact diameter $d$ much smaller than the mean free path of electrons $l$ can serve as a device for quasi-particle spectroscopy since the applied voltage $V$ is directly related to the quasi-particle energy $\Delta E = eV$, where $e$ is the electron charge.

In the case of two normal metals forming the junction the PC current $I$ comprises besides the major term $V/R_S$, the $Pb$ contribution $I_{Ps}(V)$ of order of $\approx d/l_{in}$, where $l_{in}$ is the electronic inelastic mean free path, yielding nonlinearities in the $I$–$V$ curve at characteristic EPI energies/voltages. The small nonlinearities are better pronounced as peaks in the second derivative $d^2V/dI^2(V)$, which is directly related to the point-contact form of the electron–phonon interaction function $g_{PC}(\omega)F(\omega)$ [13, 14]. Here, the matrix element $g_{PC}(\omega)$ describes the strength of the electron–phonon interaction in the PC geometry and $F(\omega)$ is the phonon density of states.

When one of the PC-forming electrodes is a superconductor, below $T_c$ a phase coherent state of Cooper pairs is formed in it. For a bias energy $|eV| < \Delta$, direct transfer of the quasi-particles is not possible due to the existence of the energy gap $\Delta$ in the spectrum of the superconductor. The transport of the charge carriers is realized through Andreev reflection with an excess current $I_{exc}(V)$, which makes the PC current inside the gap voltage two times larger than in the normal state. For biases larger than the gap voltage the excess current becomes constant and equals $I_{exc} \propto \Delta/R_N$.

The PC conductance $\sigma = dI/dV$ shows a double increase below the gap voltage $|V| < \Delta/e$ compared to the normal state or to what is observed at very large bias where the coupling via the gap is inefficient. If a barrier is formed at the point-contact junction a Giaever-like tunneling component contributes to the charge transfer as well. The evolution of the $dI/dV$ versus $V$ curves for different interfaces characterized by arbitrary transmission probability $T$ has been modeled by Blonder, Klapwijk and Tinkham (BTK) theory [15]. In case of a PC interface with an intermediate transmission probability $0 < T < 1$ a minimum appears at zero bias, $eV = 0$, but two peaks are also visible at $eV \sim \pm \Delta/e$. The experimentally measured PC conductance data can be compared with this model using as input parameters the energy gap $\Delta$, the parameter $Z$ (measure of the interface barrier strength with transmission coefficient $T = 1/(1 + Z^2)$), and a parameter $\Gamma$ for the quasi-particle lifetime broadening of the spectrum [16]. The fitting procedure is described, for example, in [17].

When a point-contact micro-constriction is formed between a normal metal and a strongly coupled superconductor, due to the significant energy dependence of the superconducting gap $\Delta(eV)$ a small negative correction to the elastic excess current $\delta I_{exc}(V)$ caused by the Andreev reflection will have a measurable effect [14]. Generally, the PC current will read as

$$I(V) = \frac{V}{R_S} + \delta I_{ph}(V) + I_{exc}(V) + \delta I_{exc}(V),$$

where the first three terms are described above and the last term is given as $\delta I_{exc}(V) \equiv (\Delta(V)/\hbar\omega)^2$ [18], where $\omega$ is the characteristic frequency of phonons mediating the superconducting pairing. This term exceeds the inelastic component of the PC current and the electron–phonon interaction modes mediating superconductivity can be seen in the second derivative $d^2V/dI^2(V)$ as peaks not exactly at characteristic EPI energies $eV$, but, importantly, shifted to higher energies by the value of the superconducting energy gap $\Delta$, in the same way as in the tunneling spectroscopy. When the gap is closed, for example, by increasing the temperature or in an applied magnetic field, the EPI peaks should move to lower energies following the gap and decrease their intensity. This would be a smoking gun for the coupling mode which mediates the superconductivity in the system.
3. Experiment

The measurements presented in this paper were performed on high-quality single-crystalline YB$_6$ samples prepared by the traveling solvent floating zone method [19]. All measurements were performed on crystals from the same batch. The crystals had a cubic form with an edge of about 0.5 mm. The values of the critical temperature $T_c$ were determined by point-contact spectroscopy, resistivity and ac-calorimetry specific-heat measurements [20]. The first two techniques give $T_c = 7.4$–7.5 K and the specific heat measurements sensitive to the bulk of the sample yield from the entropy balance construction around the anomaly $T_c = 7.32$ K, a reasonably close value.

The point-contact micro-constrictions were formed in situ by pressing a Pt tip onto the YB$_6$ surface using a differential screw mechanism allowing for a positioning of the tip on different spots on the sample. The PC tips were cut off from 50 µm Pt wires. Spectra with the best resolution were obtained on the shiny blue YB$_6$ surfaces prepared by cleaving before cooling down in the cryostat. PC spectroscopy measurements (the first and second derivatives of the $I$–$V$ characteristics) were realized by the standard lock-in modulation technique [8].

4. Results and discussion

The PC resistances were typically in the range $R_N \equiv 3$–30 Ω. As shown in our previous short communication [21], in some point contacts with the largest PC resistances a small gap below the BCS limit was observed. In those junctions also a non-BCS temperature dependence of the gap had been obtained typical of the proximity effect from the bulk to the degraded surface layer. Here, we have collected a large number of point contacts showing no signature of proximity effect and with a very little scattered gap size. Representative examples of the point-contact spectra $dI/dV$ obtained at $T = 4.2$ K are shown in figure 1. The lines display the experimental data after normalization to the normal-state PC conductance, measured above the transition temperature $T_c$ or above the upper critical field value $H_{c2}$. The open symbols represent the fits by the BTK model. Although our experiments were performed on freshly cleaved YB$_6$ surfaces, a finite $Z$ parameter was always found. Its size scattered as $Z \approx 0.3$–0.8. This means that there is always an effective interface barrier between the Pt tip and the sample. In some cases we were able to form point contacts with a high spectral resolution, as witnessed by a small value of the smearing parameter $\Gamma$ obtained from the fits. For the three spectra shown in figure 1 from the top downwards the values of $\Gamma$ achieved 5%, 13% and 37% of the gap values, respectively. The values of the superconducting gap obtained at 4.2 K on many junctions were scattered between 1.18 and 1.22 meV.

Point-contact spectroscopy explores the superconductivity in an area with dimensions of the order of the superconducting coherence length. In some cases the superconducting transition temperature $T_c$ at the surface can differ from the bulk value. That is why it is important to determine the local transition temperature $T_c$ of the junction before making any conclusions on such an important parameter as the superconducting coupling strength $2\Delta/k_BT_c$. In the published spectroscopy measurements on YB$_6$ the local critical temperature $T_c$ values have not always been determined, which could affect the calculation of the coupling strength. A correct determination of this ratio requires experimental measurement of $T_c$ and $\Delta$ in the same experiment.

A representative temperature dependence of the PC spectra is plotted in figure 2 (lines). This is a high resolution spectrum without any smearing parameter ($\Gamma = 0$). The open circles are obtained from the fits to the BTK model. During the fitting procedure we first determined the parameters $\Delta$ and $Z$ at the lowest temperature, here equal to 1.6 K. Later, for higher temperatures only $\Delta$ was used as a fit parameter, while $Z$ was kept constant. The inset of figure 2 shows the temperature dependence of the superconducting energy gap determined from the fit. The energy gap $\Delta(0) = 1.3$ meV closes at the critical temperature $T_c = 7.4$ K leading to a coupling strength of $2\Delta/k_BT_c = 4.07$. Note that from a number of measurements we always obtained $T_c$ close to 7.4–7.5 K. The transition temperature was determined either by noting when the PC conductance no longer displayed energy gap features or by extrapolating the temperature dependence of the gap to its zero value. The zero-temperature energy gap value was obtained either by extrapolating the temperature dependence of $\Delta$ to zero temperature or by measurements below 2 K where no extrapolation was necessary. As a result of many measurements we obtained a superconducting gap of $\Delta(0) = 1.30 \pm 0.03$ meV and $2\Delta/k_BT_c$ values between 3.9 and 4.1. We can conclude that in YB$_6$ the point-contact spectroscopy has revealed a single $s$-wave-gap superconductivity with intermediate strength of coupling. The temperature dependence of the superconducting gap follows the BCS prediction well, as documented by the full line in the inset of figure 2.

In the following we examine the effect of an applied magnetic field on the PC spectra and particularly on the
superconducting energy gap. The magnetic field was applied perpendicularly to the PC junction area and parallel with a Pt tip. The estimated PC diameters were in the range of hundreds of nanometers while the coherence length in YB$_6$ is about 30 nm. Then, above the lower critical magnetic field the junction was in a mixed state with many Abrikosov vortices penetrating the junction. The vortex cores form a normal-state part $N^A$ of the junction with an area $A$. Their fraction of the whole junction written as $n = N^A/A$ is in a first approximation proportional to the applied field divided by the upper critical one ($H/H_{c2}$). $n$ increases linearly with magnetic field and at the upper critical magnetic field where the whole junction is in the normal state $n = 1$. The normalized point-contact conductance in the mixed state will be the sum $\sigma/\sigma_0 = n + (1 - n) \sigma_c$, where $n$ represents the normal-state channel and $(1 - n)\sigma_c$ is the superconducting-channel contribution.

This simple empirical model (successfully applied for the study of two-gap superconductivity in MgB$_2$ [22, 23]) has been used for the fit of the normalized-conductance curves measured in the presence of magnetic field. Figure 3 shows the evolution of one PC spectrum with magnetic field (lines) and fitting curves (symbols). The fit parameters $Z$, $\Gamma$ and $\Delta$ have been determined from the zero-field PC spectrum (solid lines) measured at $T = 4.5$ K. Fits by BTK curves in the mixed state model (open symbols) yield the parameters $Z = 0.64$, $\Gamma = 0.23$ meV and $\Delta(4.5$ K) = 1.15 meV. The inset shows the temperature dependence of $H_{c2}$ from our specific heat (squares) and point-contact measurements (stars). The line is the WHH model.

As has been discussed above, point-contact spectroscopy is capable of exploring the electron–phonon interaction in superconductors either due to inelastic quasi-particle scattering on phonons or due to an energy dependent superconducting energy gap present in the elastic component of the point-contact spectra. In both mechanisms the second derivative $d^2V/dI^2$ is proportional to the EPI function. The first mechanism requires clean ballistic contacts and reveals approximately the same spectrum in the superconducting as well as in the normal state. The latter mechanism prevails in strongly coupled superconductors, where the intensity of the peaks in $d^2V/dI^2$ related to the phonons (bosons) mediating the superconducting coupling is significantly increased in the superconducting state as compared with the normal state and, moreover, the peak positions are shifted in energy by the value of the superconducting energy gap $\Delta$. With a relatively strong superconducting coupling of $2\Delta/k_BT_c = 4 \pm 0.1$ in our YB$_6$ samples we expected that strong coupling features could also appear in the elastic

\[\sigma_n(T) = \sigma(0) \left(1 - \frac{T}{T_c}\right)^{-\frac{1}{2}}.\]

\[\Delta(T) = \Delta(0) \left(1 - \frac{T}{T_c}\right)\]

\[H_{c2}(T) = H_{c2}(0) \left(1 - \frac{T}{T_c}\right)^{\frac{1}{2}}.\]

\[z(T) = z(0) \left(1 - \frac{T}{T_c}\right)^{-\frac{1}{2}}.\]

\[C(T) = C(0) \left(1 - \frac{T}{T_c}\right)^{-\frac{3}{2}}.\]

\[\rho(T) = \rho(0) \left(1 - \frac{T}{T_c}\right)^{-\frac{3}{2}}.\]

\[\chi(T) = \chi(0) \left(1 - \frac{T}{T_c}\right)^{\frac{1}{2}}.\]
component at the characteristic phonon energies. By trial and error we looked for dV/dI spectra showing a ‘smooth’ linearly increasing background, a typical feature of good quality metallic point contacts with more direct than tunneling conductance. Figure 4 plots a set of second derivatives, d²V/d²(V), of the point-contact spectra obtained at T = 4.2 K in the superconducting as well as the normal state of a Pt–YB₆ hetero-contact. The curves taken at H = 0 and 0.1 T show the spectra measured in the superconducting state and mixed state, respectively, while the spectrum taken at H = 0.4 T, above the value of the upper critical magnetic field H_c2(4.2 K) = 0.18 T (see inset of figure 3), represents the normal-state behavior. All spectra shown here reveal well defined nonlinearities in the 30 mV window. In the zero-field spectrum a sharp peak is visible at the bias (energy) \( \omega_1/e \approx 8.6 \) mV, an intense peak is also situated around \( \omega_2/e \approx 13 \) mV, followed by a hump at about 18 mV and a peak at around \( \sim 23 \) mV. The structure is superimposed on a typical point-contact background which is related to the nonequilibrium phonon generation near the point-contact orifice [8].

Point-contact spectra measured through a junction between two different metals are proportional to the EPI functions of both electrodes. For comparison, we show in figure 4 also the point-contact spectrum of the electron–phonon interaction obtained on the Pt homo-contact; the curve is reproduced from [14]. In this spectrum the PC background has been already subtracted. Two dominant Pt phonon modes are visible with peaks at \( \sim 13 \) and 23 mV and also a hump at about 18 mV can be recognized. Comparing the Pt–EPI spectrum with the spectrum of Pt one can see that the maxima observed above 10 mV are positioned at the characteristic phonon energies of Pt without any energy shift. When the superconductivity in our YB₆ sample is suppressed in increasing magnetic field, the intensities and positions of these maxima are practically unchanged. This is strongly suggestive that in our PC spectra the inelastic scattering of electrons on the Pt phonons dominates at energies above 10 meV. On the other hand, the low-energy sharp peak observed in the superconducting state at \( \omega_1/e \approx 8.6 \pm 0.1 \) mV changes clearly in an applied magnetic field. The intensity of this peak is reduced in an increased field and a gradual shift of its position is visible from \( \sim 8.6 \) mV to \( \sim 7.6 \) mV in the normal state, where the peak transforms to a shoulder. Such a shift of the energy position between the spectra in the superconducting and normal states indicates that this is the mode mediating superconducting coupling. A detailed comparison of this shift with a progressive reduction of the superconducting energy gap in a magnetic field is shown in figure 5. Clearly, the phonon-peak position (solid symbols) follows the suppression of the energy gap \( \Delta(H) \) (open symbols) in increasing magnetic field. At higher fields above \( H_c2 \) the phonon contribution in the spectrum due to the energy dependent superconducting gap is absent and the small nonlinearity at about 7.6 mV which is still present as displayed in figure 4 is due to an inelastic quasi-particle scattering on this phonon mode. It is visible together with the phonon modes of the Pt tip. In contrast to the reports in [10, 11], no structure was observed at \( \sim 11 \) meV in any of the several junctions revealing detectable nonlinearities in the d²V/d²(V) spectra on our Pt–YB₆ hetero-contacts. No structures apart of the smoothly increasing background were observed at higher voltages above 30 mV.

It is noteworthy that our measurements have revealed only the strongest peak of the complete EPI spectrum of YB₆, while the other branches due to the quasi-particle interactions with the optical phonon modes of the boron octahedra are not pronounced. That they have been observed in our PCS studies.

Figure 4. The second derivative, d²V/d²(V), spectrum measured at T = 4.2 K on a Pt–YB₆ point-contact in the superconducting (at 0 and 0.1 T) and normal (at 0.4 T) states. The down-most curve plots the EPI function of a Pt–Pt homo-contact [14].

Figure 5. The magnetic-field dependence of the superconducting energy gap of YB₆ (left ordinate) resulting from the measurements shown in figure 3—open circles. The solid line is a square-root field dependence. The full circles indicate a position development of the phonon mode of YB₆ in a magnetic field from the spectrum in figure 4 (right ordinate).
on non-superconducting LaB₆ [8] can be explained by the extremely high quality of the LaB₆ crystal with the residual resistivity ratio RRR ≳ 200, while in the case of YB₆ RRR ≳ 4. In the case of our point-contact measurements on the LuB₁₂ single crystals with RRR = 70 we have observed a dominant EPI peak at about 14 meV coming from Lu vibrations and two small maxima at 24 and 30 meV from optical boron branches [26].

Our findings strongly suggest the importance of the low-energy yttrium phonon mode with an energy of 7.6 meV in the superconducting coupling of YB₆, while other phonon modes coming from the boron octahedra vibrations are not significantly coupled to the electronic quasi-particles. This result is in full agreement with the conclusions of the ‘thermal spectroscopy’ of Lortz et al [9] based on heat capacity and resistivity measurements.

5. Conclusions

Point-contact spectroscopy measurements have been performed to study the superconducting coupling in YB₆. Values of the superconducting energy gap of Δ = 1.30 ± 0.03 meV and of the strength of the superconductivity of 2Δ/k_B T_c = 4.0 ± 0.1 have been determined. We have shown that, while the maxima of the d²V/d²(V) spectra observed on the Pt–YB₆ point contacts at energies above 10 meV are related to the inelastic scattering of electrons on the phonons of the Pt tip, the low-energy mode, observed in the superconducting state at 8.6 meV, is shifted to lower energies in the same way as the superconducting energy gap of YB₆ is gradually suppressed by the magnetic field and shows up as a small feature at 7.6 meV when YB₆ is driven to the normal state. This finding provides direct experimental evidence of the phonon-mediated superconductivity in YB₆ by the low-energy yttrium phonon mode near 7.6 meV. Our measurements confirm the results of the deconvolution of the electron–phonon interaction from the specific-heat and resistivity measurements confirm the results of the deconvolution of the electron–phonon interaction from the specific-heat and resistivity measurements by Lortz et al, showing that YB₆ is a superconductor with an Einstein lattice of Y ions rattling in the spacious cage of boron octahedra.

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