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Superconducting scanning tunneling microscopy tips in a magnetic field: Geometry-controlled order of the phase transition

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The properties of geometrically confined superconductors significantly differ from their bulk counterparts. Here, we demonstrate the geometrical impact for superconducting scanning tunneling microscopy (STM) tips, where the confinement ranges from the atomic to the mesoscopic scale. To this end, we compare the experimentally determined magnetic field dependence for several vanadium tips to microscopic calculations based on the Usadel equation. For our theoretical model of a superconducting cone, we find a direct correlation between the geometry and the order of the superconducting phase transition. Increasing the opening angle of the cone changes the phase transition from first to second order. Comparing our experimental findings to the theory reveals first and second order quantum phase transitions in the vanadium STM tips. In addition, the theory also explains experimentally observed broadening effects by the specific tip geometry.

When geometrically confined to dimensions smaller than the London penetration depth, superconductors in a magnetic field exhibit properties that can significantly differ from their bulk counterparts. For example, the critical magnetic fields, at which superconducting thin films become normal conducting, are considerably enhanced compared to the bulk due to geometrical confinement.1–4 At the critical field, the order of the superconducting phase transition also depends on geometrical factors such as the film thickness.3,4 In addition to thin films, various other geometries have been studied experimentally and theoretically such as disks, rings, or spheres.5–10 A cone presents a particularly interesting and challenging geometry covering length scales from the atomic scale apex to the macroscopic base.11 At mesoscopic length scales where superconductivity has to be described, cones are a highly suitable approximation for superconducting tips in scanning tunneling microscopy (STM). Superconducting STM tips can be employed for enhancing the energy resolution (e. g. Refs. 12 and 13), for accessing parameters of a superconductor,14 as probes for absolute spin polarization,15 or for designing Josephson junctions.16–18 In a conical geometry, it is a priori not clear if the superconducting properties are affected due to the mesoscopic confinement or if quantum size effects have to be considered as in zero-dimensional (0D) superconductors.19–22 In this context, the magnetic field dependence of the superconducting gap is of fundamental interest. The order of the superconducting phase transition also remains an open question. Therefore, it is essential to understand the impact of the confinement on tunneling experiments employing superconducting STM tips.

Here, we investigate the influence of the geometry on the superconducting phase transition of STM tips in magnetic fields. To this end, we measure the magnetic field dependence of several vanadium tips by STM and use the Usadel equation for modeling the tips as sharp superconducting cones in magnetic fields. Comparing our experimental results with the calculations, we characterize the order of the superconducting phase transitions in the V tips. We find that we can tune the order of the phase transition by changing the geometry of the tip apex, i.e., first order transitions for sharp tips and second order transitions for blunt tips. Furthermore, our approach allows to correlate experimentally observed broadening effects to the geometric confinement of the superconductor.

Fig. 1(a) shows differential conductance (dI/dV) tunneling spectra of a superconducting V tip on single crystal V(100) as a function of magnetic field B and at a temperature of 15 mK.23,29 Since the critical field for bulk V \( B_{c,\text{bulk}} < 0.5 \text{T} \), the sample is normal conducting for all measurements shown. The dI/dV-spectra show the superconducting quasi-particle density of states (DOS) of the STM tip up to its critical field \( B_c \) of about 4.2 T, which is higher than in the bulk due to the dimensional confinement near the apex.15 The lifted spin degeneracy results in the characteristic four-peak-structure of the superconducting coherence peaks.24

To characterize our data, we start with the simplest 0D model of a small superconductor in a magnetic field. In this model analyzed by Maki,25,26 the DOS is given by

\[
\rho_{\parallel}(E) = \frac{(\rho_0/2) \text{sgn}(E) \Re \left\{ u_+ \sqrt{u_+^2 - 1} \right\}},
\]

where \( u_+ \) and \( u_- \) are implicitly defined by

\[ u_+ = \sqrt{1 + \frac{\rho_0^2}{\rho_0^2 - 1}} \]

and

\[ u_- = \sqrt{\left| \frac{\rho_0^2}{\rho_0^2 - 1} \right|} \]

for the spectral density of states at the Fermi level. The DOS is then given by

\[ \rho_{\parallel}(E) = \frac{(\rho_0/2) \text{sgn}(E) \Re \left\{ u_+ \sqrt{u_+^2 - 1} \right\}}{u_+^2}. \]
broadening parameter in Maki's model (EMM) with an additional phenomenological parameter. Its appearance is the price to pay for a good fitting of experimental data using the EMM with experimental data by an oversimplified 0D Maki's model. Therefore, we employ an extended Maki model. Its dose not completely go to zero as predicted by the original Maki’s model. Therefore, we employ an extended Maki model (EMM). The analysis reveals large variations of the critical fields (2 T ≤ Bc ≤ 4.5 T) as well as of the superconducting gaps at zero field (260 μeV ≤ Δ0 ≤ 580 μeV). The observed Zeeman splitting follows the theoretical prediction of a system with spin s = 1/2 and g = 2 for all investigated V tips [gray line in Fig. 1(c)].

Further, the behavior of the superconducting gap Δ in the magnetic field depends on the specific tip (cf. Fig. 1(c)). While tip 1 shows a discontinuous transition to the normal state at high fields, other tips, such as tip 5, show a more continuous phase transition at lower fields. This behavior becomes more obvious when comparing the measured superconducting gaps to ellipses drawn as a guide to the eye in Fig. 1(c). We further find different initial superconducting gaps Δ0 at zero field, all of which are smaller than the superconducting bulk gap of V (Δ0, bulk = 820 μeV at T = 0 K). This reduction might be explained by the influence of vanadium oxide at the tip surface, changes in the phonon dispersion, and correspondingly the electron-phonon interaction, or grain size effects within the material.

For a quantitative description of the non-uniform superconducting state in the V tips, we employ a quasi-classical approach based on the Usadel equation. This approach is suitable for the polycrystalline V tips in the dirty limit, where the electron mean free path l is smaller than the coherence length. We model the STM tips as superconducting cones with the opening angle χ in an external magnetic field B applied along the tip axis (z-direction) [see insets Figs. 2(a) and 2(b)]. For sharp tips (χ ≪ 1), one can use the adiabatic approximation neglecting variations perpendicular to the cone axis. The resulting one-dimensional (1D) Usadel equation is written in terms of the spectral angle θ, which is correlated with the specific geometry of each tip.

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\[
\frac{\partial \Psi}{\partial \tau} = \left( -\frac{\epsilon}{\hbar} - \frac{g^2\cos^2\theta}{2} \right) \Psi + \frac{\hbar^2}{2m}\frac{\partial^2 \Psi}{\partial x^2}.
\]

where \(\epsilon\) is the imaginary Matsubara energy, \(\tau\) is the dimensionless \(z\)-coordinate defined by \(\tau = z\sqrt{\pi B_0/2\Phi_0}\), \(\Phi_0 = h/2e\) is the superconducting flux quantum, and \(\pm\) refers to the spin orientation. The critical angle \(\chi_c\) (described in more detail below) is defined as

\[
\chi_c = (2\sqrt{2}/3)(c \mu_B/\epsilon D) \approx 2\sqrt{2}(m_e/m)/(k_F l) \ll 1,
\]

which is in good agreement with the EMM requires larger \(\Gamma\) values than in Fig. 1(a). Repeating these experiments for several V tips (made from five different pieces of V wire), we extract the superconducting parameters by the fitting routine based on the EMM. Fig. 1(c) shows the superconducting gap Δ (solid symbols) and the Zeeman energy (open symbols) as a function of the external magnetic field B. The analysis reveals that the filling of the superconducting gap where the depairing parameter \(\Gamma\) does not completely go to zero as predicted by the original Maki’s model. Therefore, we employ an extended Maki model (EMM). The analysis reveals large variations of the critical fields (2 T ≤ Bc ≤ 4.5 T) as well as of the superconducting gaps at zero field (260 μeV ≤ Δ0 ≤ 580 μeV). The observed Zeeman splitting follows the theoretical prediction of a system with spin \(s = 1/2\) and \(g = 2\) for all investigated V tips [gray line in Fig. 1(c)].

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where $h$ and $k$ are mass and the effective mass $m_e$. The order parameter $\Delta(\tilde{z})$ is determined from the self-consistency equation at $T = 0$

$$\Delta(\tilde{z}) = \lambda \Re \int_0^{\hbar \omega_D} \sin \theta_c(\tilde{z}) d\epsilon, \quad (5)$$

where $\lambda$ is Cooper-channel interaction constant, and $\hbar \omega_D$ is the Debye energy. The DOS is obtained by analytic continuation

$$\rho_{11}(E, \tilde{z}) = \left( \rho_0 / 2 \right) \text{sgn}(E) \Re \cos \theta_c(\tilde{z}), \quad (6)$$

where $\theta_c(\tilde{z})$ refers to the spin orientation in Eq. (3). With $u_{\pm} = -i \coth \theta_c$. Eq. (6) generalizes Eq. (1) to a non-uniform case.

Calculating the free energy of such a system reveals that the nature of the quantum phase transition at the critical field is determined by the ratio $\alpha / \alpha_c$. While for small opening angles $\alpha < \alpha_c$ (sharp tips), a first order phase transition with abrupt disappearance of $\Delta$ at $B = B_c$ is expected, larger opening angles $\alpha > \alpha_c$ (blunter tips) exhibit a second order phase transition, with $\Delta$ continuously vanishing at $B = B_c$. 29

In Figs. 2(a) and 2(b), the superconducting gap in a tip with $\alpha / \alpha_c = 0.4$ and 3.2 is displayed, respectively, as a function of the dimensionless coordinate $\tilde{z}$. The figures show that only the apex of the cone remains superconducting in an external magnetic field $B > B_{c_{\text{bulk}}}$. Increasing the field shrinks the superconducting region, which becomes more confined to the apex. At a critical field $B_c$ of 4.35 T in (a) and 1.27 T in (b), the superconducting gap vanishes and the whole cone is normal conducting. This demonstrates a strong influence of the confined geometry, i.e., the opening angle $\alpha$.

The geometrical confinement also affects the quasi-particle DOS measured in tunnel experiments. In Figs. 2(c) and 2(d), the calculated quasi-particle DOS at the apex of a superconducting cone is displayed for $\alpha / \alpha_c = 0.4$ and 3.2, respectively, for different external magnetic fields. The spectral features are well-defined and the increasing Zeeman splitting is clearly observable in (c), while in (d), the wider opening angle results in broadened features with the spin-up and spin-down contributions completely smeared out.

Due to the high computational cost related with the self-consistency equation Eq. (5), the Usadel approach is unsuitable as fitting routine for the experimental $dI/dV$-spectra. To establish the relation between the microscopic theory and the phenomenological EMM, we employ the latter (now with $b = \zeta = 0$) to fit the calculated spectra [dashed lines in Figs. 2(c) and 2(d)]. The superconducting gaps obtained from the EMM fits match the results obtained from the Usadel equation.

In Fig. 3(a), the magnetic field dependence of the superconducting gap $\Delta$ is presented for several superconducting cones with varying opening angles $0.2 \leq \alpha / \alpha_c \leq 4$. Increasing $\alpha / \alpha_c$ clearly decreases the critical field $B_c$ of the cone. More importantly, at the critical field, the ratio $\alpha / \alpha_c$ determines the order of the superconducting phase transition. Sharp cones with $\alpha / \alpha_c < 1$ exhibit a first order phase transition to the normal state. For $\alpha / \alpha_c < 1$, $\Delta$ only decreases slowly up to the critical field where it abruptly vanishes and the cone becomes normal. Blunter tips with $\alpha / \alpha_c > 1$ (but still $\alpha \ll 1$) undergo a second order phase transition, in which the superconducting gap continuously decreases to zero. For a quantitative analysis of the spectral broadening, Fig. 3(b) shows the reduced broadening parameter $\Gamma = \Gamma / \Delta$ of the calculated quasi-particle DOS fitted by the EMM as a function of the magnetic field. For all cones with opening angles $0.2 \leq \alpha / \alpha_c \leq 4$, the spectral broadening is well-described by the phenomenological parameter $\Gamma$. More importantly, the rate of change $d\Gamma / dB$ in the magnetic field is also a function of the opening angle. When increasing $\alpha / \alpha_c$, the spectral broadening becomes more sensitive to the external field, and therefore, the observation of features such as coherence peaks split by the Zeeman energy is more difficult.

For comparing the results obtained from the Usadel equation to our experimental findings in Fig. 4(a), we normalize

**FIG. 2.** Calculated superconductivity of cones with opening angle $\alpha$ in an external magnetic field. (a) The apex of a sharp cone ($\alpha / \alpha_c = 0.4$) remains superconducting for magnetic fields up to 4.35 T. (b) For a blunt cone ($\alpha / \alpha_c = 3.2$), the superconducting part is more confined to the apex and the critical field is smaller ($B_c = 1.27$ T). (c) The DOS of the sharp tip ($\alpha / \alpha_c = 0.4$) exhibits clear spectral features, and the lifted spin degeneracy is clearly visible due to the Zeeman energy. (d) The DOS of the blunt tip ($\alpha / \alpha_c = 3.2$) appears more broadened.

**FIG. 3.** Superconducting parameters extracted by the EMM fit of the Usadel spectra. (a) The magnetic field dependence of the superconducting gap $\Delta$ is determined by the opening angle $\alpha$. For $\alpha / \alpha_c < 1$, the phase transition is of first order, and for $\alpha / \alpha_c > 1$ a second order phase transition is observed. (b) The spectral broadening is described by the parameter $\Gamma = \Gamma / \Delta$, which increases with $\alpha$. 
the measurements and calculations to the zero-field gaps $\Delta_0$ and critical fields $B_c$. The black line for $\alpha/\varepsilon_c = 1$ marks the separation between phase transitions of first and second order. The superconducting gaps of tip 1 lie in the region above the separation line and, therefore, tip 1 undergoes a phase transition of first order. For tip 2, the classification of the phase transition is ambiguous, since it is too close to the line $\alpha/\varepsilon_c = 1$. Tips 3–5 exhibit a second order phase transition as already indicated by the continuously vanishing gaps [Fig. 1(e)]. In Fig. 4(b), the rate of change of the normalized broadening parameter $\tilde{\Gamma}$ in the normalized magnetic field $\tilde{B}$ allows for an additional classification of the superconducting phase transition in the V tips.

In conclusion, we have investigated the quantum phase transitions of superconducting V STM tips of various geometries in magnetic fields. Solving an effective 1D Usadel equation, we have demonstrated the direct correlation of the cone geometry and the order of the superconducting phase transition: first order for very sharp tips ($\alpha < \varepsilon_c$) and second order for blunter tips ($\alpha > \varepsilon_c$). Our microscopic approach provides a physical interpretation for the experimentally observed broadening of the $dI/dV$ spectra and sheds light on the origin of the phenomenological parameter $\Gamma$ introduced to fit the data by the very simple Maki model. This parameter is not related to any pair-breaking but is a formal way to remedy the inapplicability of the 0D Maki model to systems with nonuniform superconductivity. For experimental applications, a detailed understanding of the superconductivity in the cone geometry is essential. Our study facilitates the application of superconducting STM tips in presence of an external magnetic field as additional tuning parameter, which enables techniques such as Meservey-Tedrow-Fulde STM (MTF-STM) or Josephson STM. Both techniques greatly benefit from clearly distinguishable spectral $dl/dV$ features, e.g., for resolving the Zeeman splitting and probing absolute spin polarization in MTF-STM. The combination of Josephson STM with external magnetic fields enables a wide range of additional experiments, such as single electron spin resonance measurements. Our findings suggest that both techniques benefit from superconducting tips with small opening angles ($\alpha \ll 1$) resulting in small broadening ($\Gamma \ll \Delta$) and first order phase transitions at high critical fields ($\tilde{B}_c \gg \tilde{B}_{c,\text{bulk}}$).