Magnetic-Field Enhancement of Superconductivity in Ultranarrow Wires

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We study the effect of an applied magnetic field on sub-10-nm wide MoGe and Nb superconducting wires. We find that magnetic fields can enhance the critical supercurrent at low temperatures, and do so more strongly for narrower wires. We conjecture that magnetic moments are present, but their pair-breaking effect, active at lower magnetic fields, is suppressed by higher fields. The corresponding microscopic theory, which we have developed, quantitatively explains all experimental observations, and suggests that magnetic moments have formed on the wire surfaces.

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Magnetic fields have long been known to suppress superconductivity through two main effects: first, by aligning the electron spins (i.e., the Zeeman effect) and second, by raising the kinetic energy of electrons via Meissner screening currents (i.e., the orbital effect) [1]. However, some exceptions to this general convention have been observed in nanoscale systems, where the field can penetrate, essentially unattenuated, throughout the sample. For instance, a small applied magnetic field has been observed to cause negative magnetoresistance (i.e., a decrease of resistance with increasing field) in narrow superconducting strips [2]. More strikingly, an applied magnetic field has been shown to strongly enhance superconductivity in nanowires: the so called antiproximity effect [3]. We know of no commonly accepted theoretical explanations for these effects in nanoscale systems.

Several (non-mutually-exclusive) theoretical pictures have been proposed for how magnetism or magnetic fields may enhance superconductivity. First, the applied field may reduce the charge-imbalance relaxation time associated with phase-slip centers, thus resulting in negative magnetoresistance at high currents and near T_c [4]. Second, the field may enhance dissipative fluctuations, thus localizing the phase of the superconductor and thereby stabilizing superconductivity [5]; this is thought to be relevant to the antiproximity effect [3]. Third, in disordered superconductors having grain boundaries, negative magnetoresistance may arise from interference between normal and π junctions [6]. Finally, the pair-breaking effect of magnetic moments may be quenched by either an applied field [7] or an exchange field [8], the latter being relevant to magnetic superconductors.

In this Letter, we present results from experiments on ultranarrow, sub-10-nm wide MoGe and Nb homogeneous superconducting wires that are nominally free of magnetic-impurity atoms. We have found that at low temperatures magnetic fields can enhance their critical currents by up to 30%, reaching a maximum at fields of 2–4 T. To explain this behavior we conjecture that magnetic moments (due, e.g., to the surface oxide [9]) are present in the nanowires. Correspondingly, we have developed a microscopic theory [10], which shows that polarization of such local moments by a magnetic field can quench their exchange coupling with the electrons in Cooper pairs [11] and enhance the superconducting critical current I_c. Our theory is consistent with all experimental observations, and also suggests that in the present experiments the magnetic moments are located on the surfaces of the wires.

To fabricate sub-10-nm wide wires we have used the molecular templating technique [12,13]. Our nanowires were made from the superconducting amorphous alloy $Mo_{0.79}Ge_{0.21}$ or Nb, deposited onto a freestanding fluorinated carbon nanotube suspended over a trench in a multilayered Si/SiO2/SiN substrate. The combined fraction of Fe, Co, and Ni was less than 10^{-4} at. % in the MoGe sputtering target and less than 10^{-2} at. % in the Nb target. We exposed the MoGe wires to the ambient atmosphere, which led to the oxidation of their surfaces. This process reduced the width of the conducting core by about 5 nm [14]. A transmission electron microscopy study of MoGe wires fabricated under the same conditions indicated that the wires are homogeneous [14]. Our Nb nanowires were covered with a protective Si layer [15]. The parameters of the wires are given in Table I.

Electrical transport measurements were performed on the wires in a ^{3}He cryostat equipped with carefully filtered leads. The zero-bias resistance R of the wires is shown as a function of temperature T in Fig. 1. For each $R(T)$ curve, the higher-temperature transition corresponds to the superconducting transition in the film electrodes, which are connected in series with the wires. The resistance measured immediately below the film transition is taken as the normal-state resistance R_N of the wire. Each curve also shows a lower-temperature transition, corresponding to the appearance of superconductivity in the wire itself. To fit resistance data we have used a phenomenological formula, $R = R_N \exp[-\Delta F/k_B T]$ [16], that accounts for thermally activated phase slips (TAPS) below critical temperature T_c, where ΔF is the free-energy barrier for phase slips [17]. The fitting parameters that determine ΔF are T_c and the

TABLE I. Summary of nanowire parameters (all lengths are in nm). The symbols (⊥) and (∥) indicate orientations of the magnetic field. Wire sample parameters: t is the nominal thickness of deposited MoGe or Nb; w, width measured via scanning electron microscopy (actual width and thickness of the conducting core of wires are reduced, compared to t and w, due to oxidation); L, length; R_N, normal-state resistance; d_R, diameter, calculated from R_N, L and the resistivity of MoGe (180 $\mu\Omega$ cm) and Nb (30 $\mu\Omega$ cm)[15], assuming circular cross section; $I_c(0)$, zero-field critical current at 0.3 K. Parameters produced by the fitting of R vs T curves at $B = 0$ T using TAPS theory: T_c is the critical temperature of the wire; $\xi(0)$, dirty-limit coherence length. Parameters used to fit our theory to $I_c(B)$ data (Fig. 2): τ_B is the exchange-scattering time due to local magnetic moments; $d_{\rm fit}$, effective diameter of the wire; T_{c0}, critical temperature of the wire without local moments; $I_c(0)/I_{\rm dp}(0)$, rescaling factor.

Sample	t	w	L	R_N (kΩ)	d_R	$I_c(0)$ (nA)	T_c (K)	$\xi(0)$	τ_B (ps)	$d_{\rm fit}$	T_{c0} (K)	$I_c(0)/I_{\rm dp}(0)$
MG1a (⊥)	10	21	106	2.14	10.6	1930	3.8	18	3.6	8.9	5.0	1.01
MG1b (∥)				2.26	10.3	1760	3.7	19	3.5	8.9	5.0	0.92
MG2 (⊥)	8	17	128	3.24	9.5	1010	3.6	17	2.4	8.7	5.6	0.69
MG3a ⊥)	7	17.5	156	3.86	9.6	880	2.9	17	3.4	8.5	4.4	0.75
MG3b (∥)				3.86	9.6	800	2.9	17	3.1	8.3	4.4	0.82
MG4 (∥)	8	12.5	104	4.84	7.0	63	1.9	39	1.9	9.1	4.6	0.22
Nb1 (⊥)	7	18	120	0.70	8	7170	5.7	8.1	5.9	6.4	6.5	0.89
Nb2 (⊥)	4	11	110	4.25	3.1	109	1.5	28.5	4.9	3.1	2.5	0.72

zero-temperature coherence length $\xi(0)$ (see Table I). The anomalously large $\xi(0)$ (and small I_c; see below) detected in the sample MG4 is not yet understood, but might reflect the proximity of this sample to the quantum critical point of the superconductor-insulator transition that for MoGe wires of length about 100 nm occurs at $R_N \approx 6.5$ kΩ [12]. The $R(T)$ dependence of our samples does not show any evidence of quantum phase slips [18].

For samples MG1-MG3, increasing the magnetic field B shifts the resistive transition of the wires to progressively lower temperatures, in agreement with previously observed behavior [19]. However, for the MoGe sample with the lowest T_C (i.e., MG4), the $R(T)$ curve displays a more complex response to the magnetic field: whereas at the highest fields ($B \approx$ 5–9 T) the aforementioned suppression of superconductivity is observed, there is a regime of lower fields ($B \approx$ 0–3 T) for which the resistive transition of the wire shifts oppositely, i.e., to higher temperatures with increasing B, as shown in the inset to Fig. 1. This constitutes negative magnetoresistance, which has previously been observed in Pb wires [2].

We observed a much stronger enhancement of superconductivity in the low-temperature critical current of our nanowires. In Fig. 2(a) we show the normalized critical currents for several MoGe wires, measured in a parallel magnetic field at $T = 0.3$ K. Experimentally, I_c is taken to be the current at which the wire switches to the resistive state [see the inset in Fig. 2(c)]. For all MoGe samples, I_c displays remarkable behavior, initially growing with increasing magnetic field before reaching a maximum at $B \sim$ 2–4 T. The relative magnitude of the enhancement of I_c grows with the reduction of the cross-sectional area of the wire. Nanowires made of Nb display the same tendency [see Fig. 2(b)].

To assess whether the effect is nonlocal in origin (e.g., is associated with patterns of supercurrent) we measured two wires (MG1 and MG3) both in parallel and perpendicular magnetic fields. Between measurements, each sample was removed from the cryostat and rotated on the chip. After this procedure, each wire changed slightly, due to additional oxidation. We use labels MG1a and MG3a for the wires before the rotation and MG1b and MG3b for the corresponding wires after the rotation. The critical current for sample MG3a and Mg3b, normalized by its value at zero field, is shown in Fig. 2(c). We found, at the qualitative level, that the curves $I_c(B)$ are very similar for both field orientations. This suggests that the enhancement of I_c is local in origin.

Assuming that local physics is at the core of the problem and is associated with localized spins, we have developed a theoretical model (see Ref. [10]) that yields the dependence of I_c on B and T. We have included the following ingredients: (i) local magnetic moments, which cause ex-

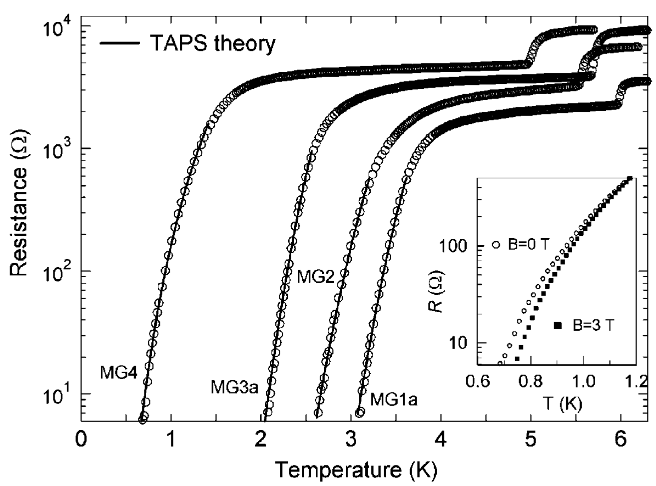

FIG. 1. Temperature dependence of the resistance of MoGe nanowires. For each sample, the solid line indicates a fit to the TAPS theory [16]. Inset: R vs T dependence of wire MG4 at $B = 0$ and 3 T.

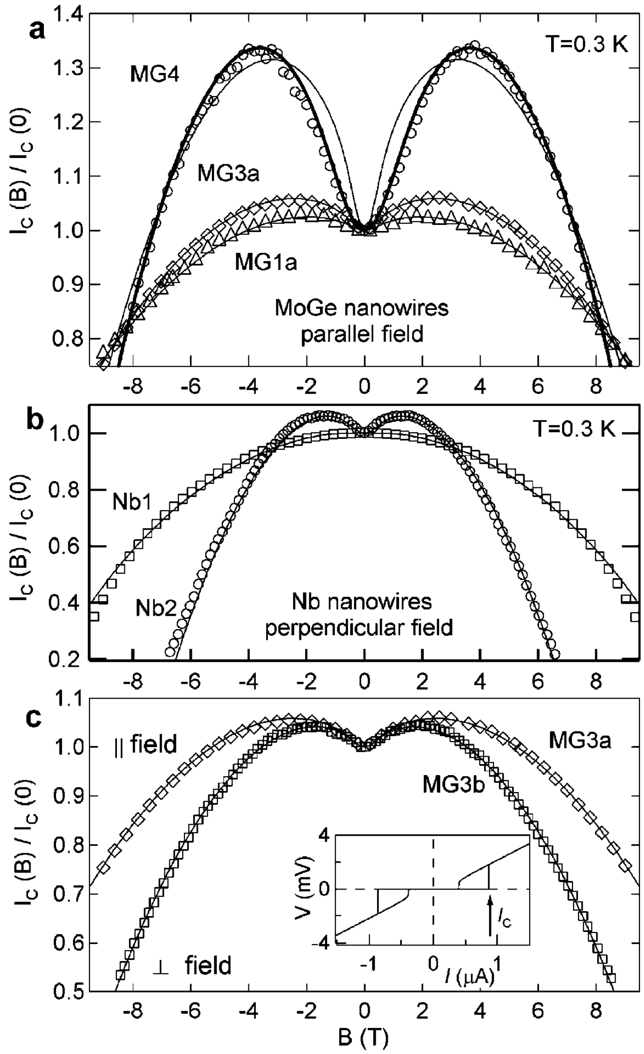

FIG. 2. (a) Normalized critical current vs magnetic field at $T = 0.3$ K for MoGe wires (parallel field). The thin solid lines are fits to our microscopic theory (for the parameters see Table I). The thick solid line corresponds to a fit in which we allow variation of additional parameters: The gyromagnetic ratio g of local magnetic moments ($g = 0.9$) and the average of the exchange coupling between electrons and local magnetic moments divided by Fermi energy $\langle \tilde{J} \rangle / E_F = -0.3$. For other fits, $g = 2$ and $\langle \tilde{J} \rangle / E_F = 0$. (b) Normalized critical current of Nb nanowires in a perpendicular magnetic field. (c) Normalized critical current of nanowires MG3a and MG3b at $T = 0.3$ K, measured in parallel and perpendicular magnetic fields. Inset: A typical hysteretic voltage vs current curve.

change scattering of electrons and thus lead to the breaking of Cooper pairs (the zero-field exchange-scattering time is given by $\tau_B = E_F / 2\pi \langle \tilde{J}^2 \rangle x_m$, where x_m is the fractional concentration of local moments, $\tilde{J}$ is the exchange coupling, and E_F is the Fermi energy); (ii) the vector potential (associated with the applied magnetic field), which scrambles the relative phases of the partners in a Cooper pair as they move diffusively in the presence of impurity scattering (viz., the orbital effect), which also suppresses superconductivity; (iii) the polarizing effect of the magnetic field on localized spins, which decreases the rate of exchange scattering, and consequently enhances superconductivity; and (iv) the Zeeman effect, associated with the applied field, which splits the energy of the up and down spins of the Cooper pair and thus tends to suppress superconductivity. (Note that strong spin-orbit scattering tends to weaken depairing due to the Zeeman effect.) These ingredients, which were also employed by Kharitonov and Feigel'man in their work on critical temperatures [7], embody the competing tendencies produced by the magnetic field: depairing via the orbital and Zeeman effects, but also the mollification of the depairing caused by local magnetic moments.

To obtain the critical current we first derive the semiclassical Eilenberger-Usadel equations [20] for the anomalous Green function, taking into account terms that describe spin-orbit scattering (with scattering time τ_{SO}), local magnetic moments, and the magnetic field [10]. Then we seek the current-carrying solution that maximizes the current, and identify it as the depairing current $I_{dp}(B)$. Because of thermal fluctuations, the switching from superconducting to the resistive state occurs at an experimentally measured value $I_c(B)$ that is less than $I_{dp}(B)$. To account for this effect of premature switching, we introduce the ratio $I_c(B)/I_{dp}(B)$ as a fitting parameter, and assume that this ratio does not depend on B [21].

By carrying out this procedure for the case of spin-1/2 magnetic impurities we have obtained numerical solutions for a wide range of material parameters, temperatures and magnetic fields, and have thus found three distinct regimes: a naturally expected one, in which both I_c and T_c simply decrease with B; and two anomalous variants. The first gives nonmonotonic behavior for both I_c and T_c, both first rising and then falling with B. The second is even more striking: although T_c simply decreases with B, at low temperatures I_c first rises and then falls. Most of our wires exhibit behavior in this last regime. To make a quantitative comparison between our experiments and our theory, we have performed fits to our data, allowing variations in the wire diameter and the exchange-scattering time. For the remaining parameters we have used following values: the g factor $g = 2$, the spin-orbit scattering times $\tau_{so} = 5.0 \times 10^{-14}$ s for MoGe and 2.3×10^{-13} s for Nb [19], and the diffusion constant for MoGe $D = 1$ cm^2/s [22]. Our theoretical model provides excellent fits to the data (Fig. 2).

An important consequence of the theory is that the behavior of I_c at small fields is dominated by scattering from magnetic impurities, and thus should not depend on the relative orientation of the field and the wire. At larger fields the orbital effect becomes important, and is larger for the perpendicular field orientation. Figure 2(c) shows that our experimental data for samples MG3a and MG3b exhibit these properties. Further evidence in favor of our theoretical picture comes from the fact that the fits to

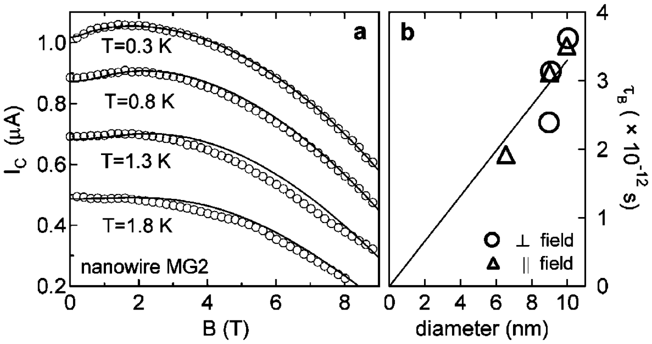

FIG. 3. (a) Critical current vs magnetic field for various temperatures. Solid lines are fits to the microscopic theory. Only the $T = 0.3$ K curve was fitted. The same microscopic parameters were used to generate curves at higher temperatures. The rescaling ratio $I_c(0)/I_{dp}(0)$ was adjusted at each temperature. (b) Exchange-scattering time vs wire diameter for MoGe nanowires. The straight line is the linear fit.

perpendicular- and parallel-field data return very close values for the τ_B, which is proportional to the impurity concentration (Table I).

At high temperatures thermal fluctuations in the moment-orientations make the quenching by the applied field less effective and, hence, higher fields are required to quench the local moments. Thus, the anomaly is expected to diminish. This is indeed what we observe experimentally [Fig. 3(a)]: at our lowest temperature (0.3 K) the anomaly is clearly observed; but at temperatures higher than roughly 1.8 K the anomaly is completely washed out. This loss of the anomaly at higher temperatures is consistent with the absence of any observed negative magnetoresistance for samples MG1-MG3.

Finally, in Fig. 3(b) we display τ_B as a function of the wire diameter d. Assuming that the magnitude of the exchange integral $|\tilde{J}| \approx 0.2$ eV (Ref. [23] p. 264), we find the magnetic-impurity fraction x_m to be of order of 0.2 at. %. The content of magnetic atoms in fluorinated carbon nanotubes (1.4 wt. %) corresponds to an effective x_m of about 0.003 at. % and is too small to account for the anomaly in $I_c(B)$. If the moments were distributed homogeneously throughout the MoGe then x_m, and therefore τ_B, would not depend on the wire diameter. Instead, our data suggest that τ_B depends linearly on d, consistent with the magnetic moments being distributed over the surface of the wires. This is also supported by the fact that our thick-film Nb and MoGe [14] samples do not reveal any change in T_c, compared to the bulk [22]. The observation of anomalous behavior both in MoGe and Nb nanowires suggests that such behavior is likely to occur for nanodevices made from other superconducting materials, unless a suitable treatment is applied to avoid the formation of local moments.

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[1] M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1996), 2nd ed.
[2] P. Xiong, A. V. Herzog, and R. C. Dynes, Phys. Rev. Lett. **78**, 927 (1997).
[3] M. Tian *et al.*, Phys. Rev. Lett. **95**, 076802 (2005).
[4] J. Clarke, in *Nonequilibrium Superconductivity, Phonons, and Kapitza Boundaries*, edited by K. E. Gray (Plenum, New York, 1981).
[5] H. C. Fu, A. Sidel, J. Clarke, and D.-H. Lee, Phys. Rev. Lett. **96**, 157005 (2006).
[6] S. A. Kivelson and B. Z. Spivak, Phys. Rev. B **45**, 10 490 (1992).
[7] M. Yu. Kharitonov and M. V. Feigel'man, JETP Lett. **82**, 421 (2005).
[8] P. C. Canfield, P. L. Gammel, and D. J. Bishop, Phys. Today **51**, No. 10, 40 (1998).
[9] F. Pierre and N. O. Birge, Phys. Rev. Lett. **89**, 206804 (2002); M. Xiao, I. Martin, E. Yablonovich, and H. W. Jiang, Nature (London) **430**, 435 (2004).
[10] T.-C. Wei, D. Pekker, A. Rogachev, A. Bezryadin, and P. M. Goldbart, Europhys. Lett. **75**, 943 (2006).
[11] M. G. Vavilov and L. I. Glazman, Phys. Rev. B **67**, 115310 (2003).
[12] A. Bezryadin, C. N. Lau, and M. Tinkham, Nature (London) **404**, 971 (2000).
[13] Y. Zhang and H. Dai, Appl. Phys. Lett. **77**, 3015 (2000); D. S. Hopkins, D. Pekker, P. M. Goldbart, and A. Bezryadin, Science **308**, 1762 (2005).
[14] A. T. Bollinger, A. Rogachev, M. Remeika, and A. Bezryadin, Phys. Rev. B **69**, 180503(R) (2004).
[15] A. Rogachev and A. Bezryadin, Appl. Phys. Lett. **83**, 512 (2003).
[16] S. L. Chu, A. T. Bollinger, and A. Bezryadin, Phys. Rev. B **70**, 214506 (2004).
[17] J. S. Langer and V. Ambegaokar, Phys. Rev. **164**, 498 (1967).
[18] A. D. Zaikin, D. S. Golubev, A. van Otterlo, and G. T. Zimanyi, Phys. Rev. Lett. **78**, 1552 (1997).
[19] A. Rogachev, A. T. Bollinger, and A. Bezryadin, Phys. Rev. Lett. **94**, 017004 (2005).
[20] G. Eilenberger, Z. Phys. **214**, 195 (1968); K. D. Usadel, Phys. Rev. Lett. **25**, 507 (1970).
[21] By using the width of the switching currents distribution (δI_c) measured at each value of the magnetic field and the equations for Josephson junction (Tinkham, Ref. [1] p. 208–209), we roughly estimated that for each wire the ratio $I_c(B)/I_{dp}(B) \sim I_c(B)/[I_c(B) + 23\delta I_c(B)]$ changes by no more than 10% with magnetic field.
[22] J. M. Graybeal, Ph.D. thesis, Stanford University, 1985.
[23] P. G. de Gennes, *Superconductivity of Metals and Alloys* (Benjamin, New York, 1966).