EUROPHYSICS LETTERS 15 September 2006

Europhys. Lett., **75** (6), pp. 943–949 (2006)
DOI: 10.1209/epl/i2006-10218-2

Enhancing superconductivity: Magnetic impurities and their quenching by magnetic fields

T.-C. Wei, D. Pekker, A. Rogachev, A. Bezryadin and P. M. Goldbart
Department of Physics, University of Illinois at Urbana-Champaign
1110 West Green Street, Urbana, IL 61801-3080, USA

received 10 April 2006; accepted in final form 1 August 2006
published online 23 August 2006

PACS. 74.25.Sv – Critical currents.
PACS. 74.62.-c – Transition temperature variations.
PACS. 74.78.-w – Superconducting films and low-dimensional structures.

Abstract. – Magnetic fields and magnetic impurities are each known to suppress superconductivity. However, as the field quenches (*i.e.* polarizes) the impurities, rich consequences, including field-enhanced superconductivity, can emerge when both effects are present. In superconducting wires and thin films this field-spin interplay is investigated via the Eilenberger-Usadel scheme. Non-monotonic dependence of the critical current on the field (and therefore field-enhanced superconductivity) is found to be possible, even in parameter regimes for which the superconducting critical temperature decreases monotonically with increasing field. The present work complements that of Kharitonov and Feigel'man (*JETP Lett.*, **82** (2005) 421), which predicts regimes of non-monotonic behavior of the critical temperature.

Introduction. – In their classic work, Abrikosov and Gor'kov [1] predicted that unpolarized, uncorrelated magnetic impurities suppress superconductivity, due to the de-pairing effects associated with the spin-exchange scattering of electrons by magnetic impurities. Among their results is the reduction, with increasing magnetic impurity concentration, of the critical temperature T_{C}, along with the possibility of "gapless" superconductivity in an intermediate regime of the impurity concentrations. It was soon recognized that other de-pairing mechanisms, such as those involving the coupling of the orbital and spin degrees of freedom of the electrons to a magnetic field, can lead to equivalent suppressions of superconductivity [2–5].

Conventional wisdom holds that magnetic fields and magnetic moments each tend to suppress superconductivity [6]. Therefore, it seems natural to suspect that any increase in a magnetic field, applied to a superconductor containing magnetic impurities, would lead to additional suppression of the superconductivity. However, very recently, Kharitonov and Feigel'man [7] have predicted the existence of a regime in which, by contrast, an increase in the field applied to a superconductor containing magnetic impurities leads to a critical temperature that first increases with magnetic field but eventually behaves more conventionally, decreasing with the field and, ultimately, vanishing at a critical value of the field. More strikingly, they have predicted that, over a certain range of concentrations of magnetic impurities, a magnetic field can actually induce the normal state to become superconducting.

The Kharitonov-Feigel'man treatment focuses on determining the critical temperature via the linear instability of the normal state. The purpose of the present letter is to address properties of the superconducting state itself, most notably the critical current and its dependence on temperature and externally applied magnetic field. The approach that we shall take is to derive the (transport-like) Eilenberger-Usadel equations [8, 9], starting from the Gor'kov equations. We account for the following effects: potential and spin-orbit scattering of electrons from non-magnetic impurities, and spin-exchange scattering from magnetic impurities, along with the orbital and Zeeman effects of the magnetic field. In addition to obtaining the critical current, we shall recover the Kharitonov-Feigel'man prediction for the critical temperature, as well as the dependence of the order parameter on temperature and magnetic field. In particular, we shall show that not only are there reasonable parameter regimes in which both the critical current and the transition temperature vary non-monotonically with increasing magnetic field, but also there are reasonable parameter regimes in which only the low-temperature critical current is non-monotonic, even though the critical temperature varies monotonically with field. We believe the present theory is applicable to explaining certain recent experiments on superconducting wires [10].

Let us pause to give a physical picture of the relevant de-pairing mechanisms. First, magnetic impurities cause spin-exchange scattering of the electrons (including both spin-flip and non-spin-flip terms, relative to an arbitrary spin quantization axis), and therefore lead to the breaking of Cooper pairs [1]. Next, consider the effects of magnetic fields. The associated vector potential scrambles the relative quantum phases of the partners of a Cooper pair, as they move diffusively in the presence of impurity scattering (viz., the orbital effect), which suppresses superconductivity [2, 3]. On the other hand, the magnetic field polarizes the magnetic impurity spins, which decreases the rate of exchange scattering, thus diminishing this contribution to de-pairing [7]. In addition, the Zeeman effect associated with the effective field (coming from the magnetic field and the impurity spins) splits the energy of the up and down spins in the Cooper pair, thus tending to suppress superconductivity [6]. But strong spin-orbit scattering tends to weaken the de-pairing by the Zeeman effect [5]. Thus, we see that the magnetic field produces competing tendencies: it causes de-pairing via the orbital and Zeeman effects but it mollifies the de-pairing caused by magnetic impurities. This competition can manifest itself through the non-monotonic behavior of observables such as the critical temperature and critical current. In order for these manifestations to be observable, the magnetic field needs to be present throughout the samples, a scenario readily accessible in wires and thin films.

Model. – The full Hamiltonian is $H = H_0 + H_{\mathrm{int}} + H_{\mathrm{Z}}$. We take the impurity-free part of the Hamiltonian to be of the BCS form [6, 11]

$$H_0 = \int \mathrm{d}r\, \frac{-1}{2m} \psi^\dagger_\alpha \Big(\nabla - \frac{ie}{c}A\Big)^2 \psi_\alpha + \frac{V_0}{2}\int \mathrm{d}r\, \Big(\langle \psi^\dagger_\alpha \psi^\dagger_\beta\rangle \psi_\beta\psi_\alpha + \psi^\dagger_\alpha\psi^\dagger_\beta \langle\psi_\beta\psi_\alpha\rangle\Big) - \mu \int \mathrm{d}r\, \psi^\dagger_\alpha\psi_\alpha, \tag{1}$$

where $\psi^\dagger_\alpha(r)$ creates an electron having mass m, charge e, position r and spin projection α, A is the vector potential, c is the speed of light, μ is the chemical potential, V_0 is the BCS pairing interaction, and $\langle \cdots \rangle$ denotes the appropriate thermal average. Throughout this letter we shall put $\hbar = 1$ and $k_B = 1$. Assuming the superconducting pairing is spin-singlet, we introduce the complex order parameter Δ via

$$-V_0\langle \psi_\alpha\psi_\beta\rangle = i\sigma^y_{\alpha\beta}\Delta, \qquad V_0\langle \psi^\dagger_\alpha\psi^\dagger_\beta\rangle = i\sigma^y_{\alpha\beta}\Delta^*, \tag{2}$$

where $\sigma^{x,y,z}_{\alpha\beta}$ are the Pauli matrices. We assume that the electrons undergo potential (u_1) scattering from impurities, located at a set of random positions $\{x_i\}$, spin-exchange (u_2)

scattering from magnetic impurities, located at positions $\{x_j\}$, and spin-orbit scattering ($v_{\rm so}$) from a third set of impurities (or defects) located at positions $\{z_k\}$, as well as being Zeeman-coupled to the applied magnetic field: these effects are included via $H_{\rm int} = \int {\rm d}r\, \psi^\dagger_\alpha V_{\alpha\beta}\psi_\beta$, with $V_{\alpha\beta}$ being given by

$$V_{\alpha\beta} = \sum\nolimits_i u_1(r-x_i)\delta_{\alpha\beta} + \sum\nolimits_j u_2(r-y_j)\vec{S}_j\cdot\vec{\sigma}_{\alpha\beta} + \sum\nolimits_k \vec{\nabla}v_{\rm so}(r-z_k)\cdot\left(\vec{\sigma}_{\alpha\beta}\times\vec{p}\right) + \mu_B B\,\sigma^z_{\alpha\beta}, \quad (3)$$

where $\vec{S}_j$ is the spin of the j-th magnetic impurity. We note that cross terms, *i.e.*, those involving distinct interactions, can be ignored when evaluating self-energy [5, 7]. Furthermore, we shall assume that the Kondo temperature is much lower than the temperatures of interest to us.

The impurity spins interact with the magnetic field through their own Zeeman term: $H_{\rm Z} = -\omega_s S^z$, where $\omega_s \equiv g_s\mu_B B$, g_s is the impurity-spin g-factor, and μ_B is the Bohr magneton. Thus, the impurity spins are not treated as static, but rather have their own dynamics, induced by the applied magnetic field. We shall approximate the dynamics of the impurity spins as if it were governed solely by the magnetic field, ignoring any influence on them coming from the electrons. Then, as the impurity spins are in thermal equilibrium, we may take the Matsubara correlators for a single spin to be

$$\langle T_\tau S^\pm(\tau_1)S^\mp(\tau_2)\rangle = T\sum\nolimits_{\omega'} D^{\pm\mp}_{\omega'} e^{-i\omega'(\tau_1-\tau_2)}, \qquad d^z \equiv \langle T_\tau S^z(\tau_1)S^z(\tau_2)\rangle = \overline{(S^z)^2}, \quad (4)$$

$$D^{+-}_{\omega'} \equiv 2\overline{S^z}/(-i\omega' + \omega_s), \qquad D^{-+}_{\omega'} \equiv 2\overline{S^z}/(+i\omega' + \omega_s), \quad (5)$$

where ω' ($\equiv 2\pi nT$) is a bosonic Matsubara frequency and $\overline{\cdots}$ denotes a thermal average. We ignore correlations between distinct impurity spins, as their effects are of the second order in the impurity concentration.

To facilitate the forthcoming analysis, we define the Nambu-Gor'kov four-component spinor (see, *e.g.*, refs. [5, 12]) via $\Psi^\dagger(x) \equiv \left(\psi^\dagger_\uparrow(r,\tau), \psi^\dagger_\downarrow(r,\tau), \psi_\uparrow(r,\tau), \psi_\downarrow(r,\tau)\right)$. Then the electron-sector Green functions are defined in the standard way via

$$\mathcal{G}_{ij}(1:2) \equiv -\langle T_\tau \Psi_i(1)\Psi^\dagger_j(2)\rangle \equiv \begin{pmatrix} \hat{G}(1:2) & \hat{F}(1:2) \\ \hat{F}^\dagger(1:2) & \hat{G}^\dagger(1:2)\end{pmatrix}, \quad (6)$$

where $\hat{G}$, $\hat{G}^\dagger$, $\hat{F}$, and $\hat{F}^\dagger$ are each two-by-two matrices (as indicated by the ˆ symbol), *i.e.*, the normal and anomalous Green functions, respectively. As the pairing is assumed to be singlet, $\hat{F}$ is off-diagonal whereas $\hat{G}$ is diagonal.

Eilenberger-Usadel equations. – The critical temperature and critical current are two of the most readily observable quantities. The procedure is first to derive Eilenberger equations [8] and then, assuming the dirty limit, to obtain the Usadel equations. The self-consistency condition between the anomalous Green function and the order parameter leads, in the small order-parameter limit, to an equation determining the critical temperature. Moreover, solving the resulting transport-like equations, together with the self-consistency equation, gives the transport current, and this, when maximized over superfluid velocity, yields the critical current.

To implement this procedure [8, 9, 13–15], one first derives the equations of motion for $\mathcal{G}$ (viz., the Gor'kov equations). By suitably subtracting these equations from one another one arrives at a form amenable to a semiclassical analysis, for which the rapidly and slowly varying parts in the Green function (corresponding to the dependence on the relative and center-of-mass coordinates of a Cooper pair, respectively) can be separated. Next, one treats the interaction Hamiltonian as an insertion in the self-energy, which leads to a new set of semiclassical Gor'kov equations. These equations are still too complicated to use effectively, but they can be simplified to the so-called Eilenberger equations [8] (at the expense of losing detailed information about excitations) by introducing the energy-integrated Green functions:

$$\hat{g}(\omega,k,R) \equiv \frac{i}{\pi}\int \mathrm{d}\xi_k\, \hat{G}(\omega,k,R), \qquad \hat{f}(\omega,k,R) \equiv \frac{1}{\pi}\int \mathrm{d}\xi_k\, \hat{F}(\omega,k,R), \tag{7}$$

and similarly for $\hat{g}^\dagger(\omega,k,R)$ and $\hat{f}^\dagger(\omega,k,R)$. Here, ω is the fermionic Matsubara frequency Fourier-conjugate to the relative time, k is the relative momentum conjugate to the relative coordinate, and R is the center-of-mass coordinate. However, the resulting equations do not determine g's and f's uniquely, and need to be supplemented by the normalization conditions [8, 9, 13–15] as well as the self-consistency condition

$$\hat{g}^2 + \hat{f}\hat{f}^\dagger = \hat{g}^{\dagger 2} + \hat{f}^\dagger \hat{f} = \hat{1}, \qquad \Delta = |g| \sum\nolimits_\omega f_{12}(\omega). \tag{8}$$

In the dirty limit (*i.e.*, $\omega\tau_{\mathrm{tr}} \ll G$ and $\Delta\tau_{\mathrm{tr}} \ll F$), where τ_{tr} is the transport relaxation time (which we shall not distinguish from the elastic mean-free time), the Eilenberger equations can be simplified further, because, in this limit, the energy-integrated Green functions are almost isotropic in k. This allows one to retain only the two lowest spherical harmonics ($l = 0, 1$), and to regard the $l = 1$ term as a small correction (*i.e.*, $|\check{k}\cdot\vec{F}| \ll |F|$), so that we may write

$$g(\omega,\check{k},R) = G(\omega,R) + \check{k}\cdot\vec{G}(\omega,R), \qquad f(\omega,\check{k},R) = F(\omega,R) + \check{k}\cdot\vec{F}(\omega,R), \tag{9}$$

where $\check{k}$ is the unit vector along k. In this limit the normalization conditions simplify to

$$G_{11}^2 = 1 - F_{12}F_{21}^\dagger, \qquad G_{22}^2 = 1 - F_{21}F_{12}^\dagger, \tag{10}$$

and the Eilenberger equations reduce to the Usadel equations [9] for $F_{12}(\omega,R)$, $F_{21}(\omega,R)$, $F_{12}^\dagger(\omega,R)$, and $F_{21}^\dagger(\omega,R)$.

Application to thin wires and films. – Let us consider a wire (or film) thinner than the coherence length. In this regime, we may assume that the order parameter has the form $\Delta(R) = \tilde{\Delta}e^{iuR_x}$, where R_x is the coordinate measured along the direction of the current (*e.g.*, along the length of the wire) and u parametrizes the superfluid velocity via $v_s = \hbar u/2m$. Similarly, we may assume that the semiclassical anomalous Green functions have the form

$$F_{\alpha\beta}(\omega,R) = \tilde{F}_{\alpha\beta}(\omega)e^{iuR_x}, \qquad F_{\alpha\beta}^\dagger(\omega,R) = \tilde{F}_{\alpha\beta}^\dagger(\omega)e^{-iuR_x}. \tag{11}$$

By invoking the symmetry amongst the $\tilde{F}$'s (*i.e.*, $\tilde{F}_{\alpha\beta}^* = -\tilde{F}_{\alpha\beta}^\dagger$ and $\tilde{F}_{\alpha\beta}/\tilde{\Delta} = -(\tilde{F}_{\beta\alpha}/\tilde{\Delta})^*$) we may reduce the four Usadel equations (for $\tilde{F}_{12}$, $\tilde{F}_{21}$, $\tilde{F}_{12}^\dagger$, and $\tilde{F}_{21}^\dagger$) to a single one:

$$\left[\omega + i\delta_B + \frac{T}{2\tau_{\mathrm{B}}}\sum_{\omega'}\left(D_{\omega'}^{-+}G_{22}(\omega-\omega')\right) + \left(\frac{d^z}{\tau_{\mathrm{B}}} + \frac{\tilde{D}}{2}\right)G_{11}(\omega) + \frac{1}{3\tau_{\mathrm{so}}}G_{22}(\omega)\right]\frac{\tilde{F}_{12}(\omega)}{\tilde{\Delta}} -$$
$$- G_{11}(\omega) = -G_{11}(\omega)\frac{T}{2\tau_{\mathrm{B}}}\sum_{\omega'}\left(D_{\omega'}^{-+}\frac{\tilde{F}_{12}^*(\omega-\omega')}{\tilde{\Delta}^*}\right) + \frac{1}{3\tau_{\mathrm{so}}}G_{11}(\omega)\frac{\tilde{F}_{12}^*(\omega)}{\tilde{\Delta}^*}, \tag{12}$$

in which we have used eqs. (4) and (5), $\delta_B \equiv \mu_B B + n_i u_2(0)\overline{S^z}$, $\tilde{D} \equiv D\langle\langle(u - 2eA/c)^2\rangle\rangle$, where the London gauge has been adopted and $\langle\langle\cdots\rangle\rangle$ defines a spatial average over the sample thickness, and $D \equiv v_F^2 \tau_{\rm tr}/3$ is the diffusion constant. The spin-exchange and spin-orbit scattering times, $\tau_{\rm B}$ and $\tau_{\rm so}$, are defined via Fermi surface averages:

$$\frac{1}{2\tau_{\rm B}} \equiv N_0 n_i \pi \int \frac{{\rm d}^2\check{k}'}{4\pi} |u_2|^2, \qquad \frac{1}{2\tau_{\rm so}} \equiv N_0 n_{\rm so} \pi \int \frac{{\rm d}^2\check{k}'}{4\pi} |v_{\rm so}|^2 p_F^2 |\check{k} \times \check{k}'|^2. \tag{13}$$

Here, N_0 is the (single-spin) density of electronic states at the Fermi surface, n_i is the concentration of magnetic impurities, $n_{\rm so}$ is the concentration of spin-orbit scatterers, $p_F = m v_F$ is the Fermi momentum, u_2 and v_{so} are potentials introduced in eq. (3). The normalization condition then becomes

$$\tilde{G}_{11}(\omega) = {\rm sgn}(\omega)[1 - \tilde{F}_{12}^2(\omega)]^{1/2}, \qquad \tilde{G}_{22}(\omega) = {\rm sgn}(\omega)[1 - \tilde{F}_{12}^{*2}(\omega)]^{1/2} = \tilde{G}_{11}^*(\omega). \tag{14}$$

Furthermore, the self-consistency condition (8) becomes

$$\ln \frac{T_{C0}}{T} = \pi T \sum_\omega \left(\frac{1}{|\omega|} - \frac{\tilde{F}_{12}(\omega)}{\tilde{\Delta}} \right), \tag{15}$$

in which we have exchanged the coupling constant g for T_{C0}, *i.e.*, the critical temperature of the superconductor in the absence of magnetic impurities and the magnetic fields.

Results for the critical temperature. – These can be obtained in the standard way, *i.e.*, by i) setting $u = 0$ and expanding eqs. (12) to linear order in $\tilde{F}$ (at fixed $\tilde{\Delta}$), and ii) setting $\tilde{\Delta} \to 0$ and applying the self-consistency condition. In the limit of strong spin-orbit scattering (*i.e.*, $\tau_{\rm so} \ll 1/\omega$ and $\tau_{\rm B}$), step i) yields

$$\left[|\omega| + \tilde{\Gamma}_\omega + \frac{D}{2} \left\langle\!\!\left\langle \left(\frac{2eA}{c} \right)^2 \right\rangle\!\!\right\rangle + \frac{3\tau_{\rm so}}{2} \delta_B'(\omega)^2 \right] {\rm Re}\, C(\omega) \approx 1 - \frac{T}{\tau_{\rm B}} \sum_{\omega'} \frac{\omega_s \overline{S^z}}{\omega'^2 + \omega_s^2} {\rm Re}\, C(\omega - \omega'), \tag{16a}$$

where $C \equiv \tilde{F}_{12}/\tilde{\Delta}$ and

$$\delta_B'(\omega) \equiv \delta_B - \frac{T}{\tau_{\rm B}} \sum_{\omega_c > |\omega'| > |\omega|} \frac{2|\omega'|\overline{S^z}}{\omega'^2 + \omega_s^2}, \qquad \tilde{\Gamma}_\omega \equiv \frac{d^z}{\tau_{\rm B}} + \frac{T}{\tau_{\rm B}} \sum_{|\omega'| < |\omega|} \frac{\omega_s \overline{S^z}}{\omega'^2 + \omega_s^2}, \tag{16b}$$

in which a cutoff ω_c has been imposed on ω' in δ_B'. This is essentially the Cooperon equation in the strong spin-orbit scattering limit, first derived by Kharitonov and Feigel'man [7], up to an inconsequential renormalization of δ_B. Step ii) involves solving the self-consistency equation (15) to obtain the critical temperature T_C.

Figure 1 shows the dependence of the critical temperature of wires or thin films on the (parallel) magnetic field for several values of magnetic impurity concentration. Note the qualitative features first obtained by Kharitonov and Feigel'man [7]: starting at low concentrations of magnetic impurities, the critical temperature decreases monotonically with the applied magnetic field. For larger concentrations, a marked non-monotonicity develops, and for yet larger concentrations, a regime is found in which the magnetic field first induces superconductivity but ultimately destroys it.

The non-monotonicity arises due to the two competing roles the magnetic field plays, mentioned earlier. To see this, let us consider the strong spin-orbit scattering limit, where the fourth term on the l.h.s. of eq. (16a) can be simply ignored. The third term (*i.e.*, the orbital contribution to de-pairing) scales as B^2 and is frequency indepdent, whereas the second term (*i.e.*, the de-pairing effect from the exchange scattering) develops a dip at low frequencies as the field B increases before it saturates, as shown in the inset of fig. 2. Therefore, for certain range of the impurity concentration it is possible to have non-monotonic behavior of the critical temperature (and similarly of the critical current, see below).

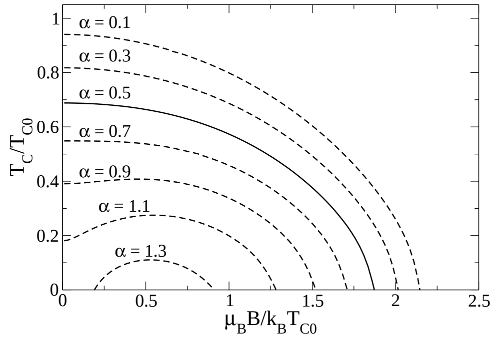

Fig. 1

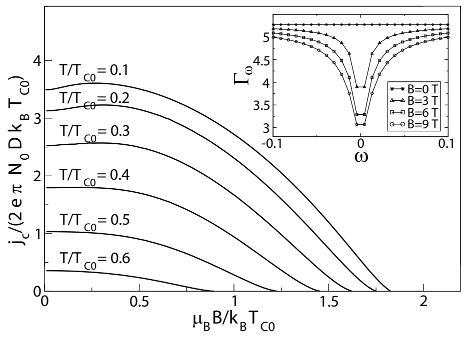

Fig. 2

Fig. 1 – Critical temperature *vs.* (parallel) magnetic field for a range of exchange-scattering strengths characterized by the dimensionless parameter $\alpha \equiv \hbar/(k_B T_{C0}\tau_{\rm B})$. The strength for potential scattering is characterized by the parameter $\hbar/(k_B T_{C0}\tau_{\rm tr}) = 10^4$, and that for spin-orbit scattering by $\hbar/(k_B T_{C0}\tau_{\rm so}) = 10^3$; the sample thickness is $d = 90.0\,\hbar/p_F$, where p_F is the Fermi momentum; the impurity gyromagnetic ratio is chosen to be $g_s = 2.0$; and the typical scale of the exchange energy u_2 in eq. (3) is taken to be $E_F/7.5$, where E_F is the Fermi energy.

Fig. 2 – Critical current *vs.* (parallel) magnetic field at several values of the temperature, with the strength of the exchange scattering set to be $\alpha = 0.5$ (corresponding to the solid line in fig. 1), and all other parameters being the same as those used in fig. 1. The inset shows an example of the rate $\tilde{\Gamma}_\omega$ in eq. (16b) for $B = 0, 3, 6, 9$ T (from top to bottom).

Results for the critical current density. – To obtain the critical current density j_c we first determine the current density (averaged over the sample thickness) from the solution of the Usadel equation, via

$$j(u) = 2eN_0\pi DT \sum\nolimits_\omega \mathrm{Re}\left(\tilde{F}_{12}^2(\omega) \left[u - \frac{2e}{c} \langle\!\langle A \rangle\!\rangle \right] \right), \tag{17}$$

and then maximize $j(u)$ with respect to u. In the previous section, we saw that, over a certain range of magnetic impurity concentrations, T_C displays an up-turn with field at small fields, but eventually decreases. Not surprisingly, our calculations show that such non-monotonic behavior is also reflected in the critical current.

Perhaps more interestingly, however, we have also found that for small concentrations of magnetic impurities, although the critical temperature displays *no* non-monotonicity with the field, the critical current *does* exhibit non-monotonicity, at least for lower temperatures. This phenomenon, which is exemplified in fig. 2, sets magnetic impurities apart from other depairing mechanisms. The reason why the critical current shows non-monotonicity more readily than the critical temperature does is that the former can be measured at lower temperatures, for which the impurities are more strongly polarized by the field.

Conclusion and outlook. – We have addressed the issue of superconductivity, allowing for the simultaneous effects of magnetic fields and magnetic impurity scattering, as well as spin-orbit impurity scattering. Although sufficiently strong magnetic fields inevitably destroy superconductivity, their quenching effects on the magnetic impurities can, at lower field strengths, lead to the enhancement of superconductivity, as first predicted by Kharitonov and Feigel'man via their analysis of the superconducting transition temperature. In the present letter, we adopt the Eilenberger-Usadel semiclassical approach to address the critical current of the superconducting state and, moreover, we also recover the behavior of critical temperature obtained by Kharitonov and Feigel'man. We have found that any non-monotonicity in the field dependence of the critical temperature is always accompanied by non-monotonicity in the field dependence of the critical current. However, we have also found that for a wide range of physically reasonable values of the parameters the critical current exhibits non-monotonic behavior with field at lower temperatures, even though at such parameter values there is no such behavior in the critical temperature.

Especially for small samples, for which thermal fluctuations can smear the transition to the superconducting state over a rather broad range of temperatures, the critical current is expected to provide a more robust signature of the enhancement of superconductivity, as it can be measured at arbitrarily low temperatures. In addition, critical currents can be measured over a range of temperatures, and thus provide rather stringent tests of any theoretical models. Recent experiments measuring the critical temperatures and critical currents of superconducting MoGe and Nb nanowires show a behavior consistent with the predictions of the present letter, inasmuch as they display monotonically varying critical temperatures but non-monotonically varying critical currents [10].

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We acknowledge useful discussions with A. J. Leggett and M. Yu. Kharitonov. This work was supported via NSF EIA01-21568 and CAREER award DMR01-34770, via DOE DEFG02-91ER45439, and via the A. P. Sloan Foundation.

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