Impurity Effect on Flux-Flow Resistivity in Gapless Superconductors*

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The flux-flow resistivity ρ_f has been calculated for gapless superconductors with arbitrary amounts of magnetic and nonmagnetic scattering. The most striking result occurs in the limit of low concentrations of magnetic impurities, where the initial slope of ρ_f with respect to the applied magnetic field may range from zero to infinity depending on the amount of nonmagnetic scattering.

Recently, we have calculated the flux-flow resistivity ρ_f of dirty, gapless superconductors containing a high concentration of magnetic impurities, in both the high-[1] and low-field[3,4] limits. The dependence of ρ_f on the average magnetic field $\langle B\rangle$, if measured in units of normal-state resistivity ρ_n and upper critical field H_{c2}, respectively, was found to have an initial slope 0.38 and a final slope 5.2, with the whole curve falling somewhat below the simple relation $\rho_f/\rho_n = \langle B\rangle/H_{c2}$ envisioned by Kim and co-workers[5] for the low-temperature limit. Our low-field result was also obtained by Gor'kov and Kopnin[6] (except for their error of neglecting the losses directly due to the electric field). They also applied their method to dirty gapless superconductors with a low concentration of magnetic impurities and found a very small initial slope for ρ_f near $\langle B\rangle = 0$. This result prompted us to extend our previous investigation to a complete dependence of ρ_f in the gapless regime on the amounts of magnetic and nonmagnetic scattering. Our result indicates that such an empirical relation as a universal dependence of ρ_f/ρ_n on $\langle B\rangle/H_{c2}$ and $T/$(transition temperature T_c), independent of alloy composition as is proposed in Ref. 5, does not apply to gapless superconductors. In fact, such a relation is found to be violated to the maximum possible degree in the limit of low magnetic impurity concentrations where the initial slope of ρ_f is found to range from zero to infinity depending on the ratio τ_s/τ_1, where τ_s and τ_1 are the spin-flip and total scattering lifetimes, respectively.

Throughout this paper we shall assume the gapless condition $\tau_s\Delta_0 \ll 1$ which is roughly $T_c(T_c - T) \ll (3.5\tau_s)^{-2} \approx (T_{c0} - T_c)^2/12$, where $\Delta_0(T)$ is the equilibrium value of the order parameter in the absence of fields, and T_{c0} is the transition temperature in the absence of magnetic impurities. A complete set of time-dependent Ginzburg-Landau equations may then be derived along the line of Gor'kov and Éliashberg[7] and Éliashberg[8] to include arbitrary amounts of magnetic and nonmagnetic scattering:

$$C_1\gamma\left(\frac{\partial}{\partial t}+u_1\right) f - \nabla^2 f + 4e^2Q^2 f + \xi^{-2}(f^3 - f) = 0, \tag{1}$$

$$C_1\gamma f^2\left(\frac{\partial}{\partial t}\chi + u_2\right) + \nabla\cdot(f^2\vec{Q}) = 0 \quad [\vec{Q} \equiv \vec{A} - \nabla\chi], \tag{2}$$

$$\left(\gamma\frac{\partial}{\partial t} - \nabla^2\right) u_1 = C_1\xi^{-2}\frac{\partial}{\partial t} f^2, \tag{3}$$

$$u_2 = \zeta_1^{\,2}\int_{-\infty}^{\infty} d\epsilon\, \zeta_\epsilon^{\,-2}\psi_\epsilon, \tag{4}$$

$$\left(\gamma\frac{\partial}{\partial t} - \nabla^2 + \zeta_\epsilon^{\,-2} f^2\right)\psi_\epsilon = \frac{1}{4T}\cosh^{-2}\frac{\epsilon}{2T}\left[-\zeta_\epsilon^{\,-2} f^2\frac{\partial}{\partial t}\chi + \gamma\frac{\partial}{\partial t}\varphi + \frac{\partial}{\partial t}\nabla\cdot\vec{A}\right], \tag{5}$$

$$\vec{j} = \sigma\left[-\nabla\psi - \frac{\partial}{\partial t}\vec{A}\right] - f^2\vec{Q}/4\pi\lambda^2, \tag{6}$$

$$\rho = (\psi - \varphi)/4\pi\lambda_{TF}^{\,2}, \quad \psi = \int_{-\infty}^{\infty} d\epsilon\, \psi_\epsilon. \tag{7}$$

Here γ^{-1} is the diffusion constant, e is the electronic charge, $fe^{i2e\chi}$ is the order parameter divided by Δ_0, ξ and λ are the temperature-dependent coherence length and penetration depth, respectively. $\sigma = \rho_n^{-1}$, and λ_{TF} is the Thomas-Fermi screening length. The scalar and vector potentials φ and $\vec{A}$ are coupled to the charge and current densities ρ and $\vec{j}$ by the Maxwell equations. Four dimensionless con-

stants appear in Eqs. (1)–(7), besides the Ginzburg-Landau parameter $\kappa = \lambda/\xi$:

$$C_1 = \psi^{(1)}(\tfrac{1}{2}+\rho_s)\left(\sum_{n=0}^{\infty}\frac{\rho_1}{(n+\frac{1}{2}+\rho_s)^2(n+\frac{1}{2}+\rho_1)}\right)^{-1}, \tag{8}$$

$$C_2 = -\frac{\psi^{(1)}(\frac{1}{2}+\rho_s)+\rho_s\psi^{(2)}(\frac{1}{2}+\rho_s)}{\rho_s C_1[\psi^{(2)}(\frac{1}{2}+\rho_s)+\frac{1}{3}\rho_s\psi^{(3)}(\frac{1}{2}+\rho_s)]}, \tag{9}$$

$$\xi^2/\zeta_\epsilon{}^2 = C_3/[1+(\tau_s\epsilon)^2], \tag{10}$$

where

$$C_3 = 4C_2\psi^{(1)}(\tfrac{1}{2}+\rho_s)/[\psi^{(1)}(\tfrac{1}{2}+\rho_s)+\rho_s\psi^{(2)}(\tfrac{1}{2}+\rho_s)], \tag{11}$$

and we have defined $\zeta_1{}^2 = 4\pi\lambda^2\sigma/C_1\gamma$ such that

$$C_4 \equiv \xi^2/\zeta_1{}^2 = \rho_s\psi^{(1)}(\tfrac{1}{2}+\rho_s)C_3. \tag{12}$$

In these expressions $\rho_s = 1/(2\pi\tau_s T)$ and $\rho_1 = 1/(4\pi\tau_1 T)$, and $\psi^{(n)}$ are the polygamma functions.

In Eq. (5) the individual energy component $e\psi_\epsilon$ of the electrochemical potential $e\psi$ is seen to be governed by an energy-dependent screening length ζ_ϵ. The important energy range is $|\epsilon| \lesssim T$ because of the hyperbolic cosine factor in Eq. (5). In the limit when $\tau_s T \ll 1$, corresponding to high concentrations of magnetic impurities, we have $\zeta_\epsilon = \zeta_1$ ($\equiv\zeta$) and the set of equations reduces to that studied in Ref. 1. If further $\tau_1 \ll \tau_s$ (the dirty limit), then $\zeta^2 = \xi^2/12$ as stated in Ref. 1, but in the opposite limit when $\tau_1 = \tau_s$ we have $\zeta^2 = \xi^2/4.64$, so that the screening length ζ defined in Refs. 1–4 does have a limited range of variation.

To calculate ρ_f in the low-field limit, we repeat the procedure developed in Ref. 2. An energy-balance relation is first derived using Eqs. (1)–(7):

$$(\partial/\partial t)F + \nabla\cdot\vec{j}^E = -W, \tag{13}$$

$$W = \sigma\left(-\nabla\psi - \frac{\partial}{\partial t}\vec{A}\right)^2 + \frac{\sigma}{\zeta_1{}^2}\left[\left(\frac{\partial\chi}{\partial t}+\psi\right)\left(\frac{\partial\chi}{\partial t}+u_2\right)f^2 + \frac{1}{4e^2}\frac{\partial f}{\partial t}\left(\frac{\partial f}{\partial t}+u_1 f\right)\right], \tag{14}$$

where the free energy density F and the energy current $\vec{j}^E$ are the same as those defined in Ref. 2. The average dissipation rate $\langle W\rangle$ is equal to the transport current $\vec{j}_t$ times the average electric field $\langle\vec{\mathcal{E}}\rangle$, so that as before[2-4] we can use it to find the dependence of $\vec{j}_t$ on the flux-flow velocity $\vec{v}$, from which we obtain the flux-flow resistivity $\rho_f = \langle\mathcal{E}\rangle/j_t = v\langle B\rangle/j_t$. After using Eqs. (1)–(7) to transform W as in Ref. 2, we obtain

$$\frac{H_{c2}}{\rho_n}\frac{d\rho_f}{d\langle B\rangle}\bigg|_{\langle B\rangle=0} = \left\{\tfrac{1}{2}C_4\left(C_\gamma + \frac{1}{\pi v^2}\int\frac{\partial f}{\partial t}u_1 f\frac{dV}{L}\right) + \frac{\mathcal{E}(0)}{vH_{c2}}\right\}^{-1}, \tag{15}$$

where we have assumed $\kappa \gg 1$. The constant $C_\gamma = 0.2791$.[3] Equation (3) may be solved to first order in $\vec{v}$[6]:

$$u_1 = C_2\xi^{-2}(\vec{v}\cdot\hat{r})r^{-1}\int_0^r (f^2-1)r'\,dr', \tag{16}$$

which gives, after use is made of Eq. (39) of Ref. 3,

$$(\pi v^2)^{-1}\int(\partial f/\partial t)u_1 f(dV/L) = \tfrac{1}{2}C_2. \tag{17}$$

The electric field at the origin of an isolated moving vortex $\mathcal{E}(0)$ may be obtained to first order in $\vec{v}$ by solving Eq. (5) and then using $\vec{\mathcal{E}} = \nabla\psi + (\vec{v}\cdot\nabla)\vec{A}$. We have done this only approximately by replacing f^2 by $r^2/(r^2+\delta\xi^2)$ in Eq. (5). This approximation was used in Ref. 2 with $\delta = 1$ which gave rather good results. Here, we take $\delta = 2.83$ to reproduce the exact result for the limit of high concentration of magnetic impurities calculated in Ref. 3. In the opposite limit, when $\tau_s T_c \gg 1$, $\zeta_\epsilon \ll \xi$, so that f^2 may be replaced by its behavior near the origin,[3] $f^2/2.94\xi^2$, our approximation is seen to be still very good. The result is

$$\frac{H_{c2}}{\rho_n}\frac{d\rho_f}{d\langle B\rangle}\bigg|_{\langle B\rangle=0} = \left\{\tfrac{1}{4}C_4(0.5582+C_2) + \frac{\pi\rho_s}{2\delta}\int_0^\infty \frac{K_0(x_0(1+x^2)^{-1/2})x_0\cosh^{-2}\pi\rho_s x}{K_1(x_0(1+x^2)^{-1/2})(1+x^2)^{1/2}}\,dx\right\}^{-1}, \tag{18}$$

where $x_0{}^2 = \delta C_3$, and K_n is the modified Bessel function of order n. This dependence of the normalized

initial slope of ρ_f on impurity concentrations is shown in Fig. 1, where we have chosen as independent variables the ratio $\tau_s/\tau_1 = 2\rho_1/\rho_s$ and

$$t \equiv \exp[\psi(\tfrac{1}{2}) - \psi(\tfrac{1}{2}+\rho_s)]. \quad (19)$$

In view of the gapless condition $\tau_s\Delta_0 \ll 1$, one may ignore the difference between T and T_c in the definition of ρ_s and identify t as the reduced transition temperature T_c/T_{c0}.

At present, only the limit $\rho_1 \gg \rho_s$ seems to be observable in practice. Assuming this and also $\rho_s \ll 1$, one finds that C_2 dominates in Eq. (18), so that

$$\frac{H_{c2}}{\rho_n}\frac{d\rho_f}{d\langle B\rangle}\bigg|_{\langle B\rangle=0} = 2\rho_s\left[\frac{28\zeta(3)C_1}{\pi^3}\right]^2.$$

If one further assumes that $\rho_1 \gg 1$ for all ρ_s, then $C_1 = 1$ and the result is that found by Gor'kov and Kapnin.[6] But if the ratio ρ_s/ρ_1 is held constant as ρ_s is reduced, as would be the case if only one type of magnetic impurity and no other scattering centers are present, then the normalized initial slope will go through a minimum near $\rho_1 \sim 1$, $t \sim \exp[-\pi^2\tau_1/\tau_s]$, and then diverge upward like $(8/\pi^2)(\rho_s/\rho_1{}^2)$.

The flux-flow resistivity for field $\langle B\rangle$ near H_{c2} may be calculated simply by applying the method of Caroli and Maki[9] or by solving Eqs. (1)–(7) linearized with respect to f and v.[1] (The anomalous contributions to the currents which were investigated by Thompson[10] are not important under the gapless condition $\tau_s\Delta_0 \ll 1$, because the main pair breaker is then the magnetic impurities, not the field.) The result is a simple generalization of that in Ref. 1 with ζ replaced by ζ_1:

$$\frac{H_{c2}}{\rho_n}\frac{d\rho_f}{d\langle B\rangle}\bigg|_{\langle B\rangle=H_{c_2}} = \frac{\xi^2}{2\zeta_1{}^2}\frac{2\kappa^2}{(2\kappa^2-2)1.16+1}. \quad (20)$$

This result in the limit $\kappa \gg 1$ is plotted in Fig. 2 with respect to t for the same set of values for τ_s/τ_1 as in Fig. 1. The two figures together give an overall field dependence of ρ_f for various impurity concentrations.

We conclude by noting that the lowest possible value for τ_s/τ_1 is, in principle, unity, corresponding to only spin-flip scattering being present. To approach this limit, one must dope very

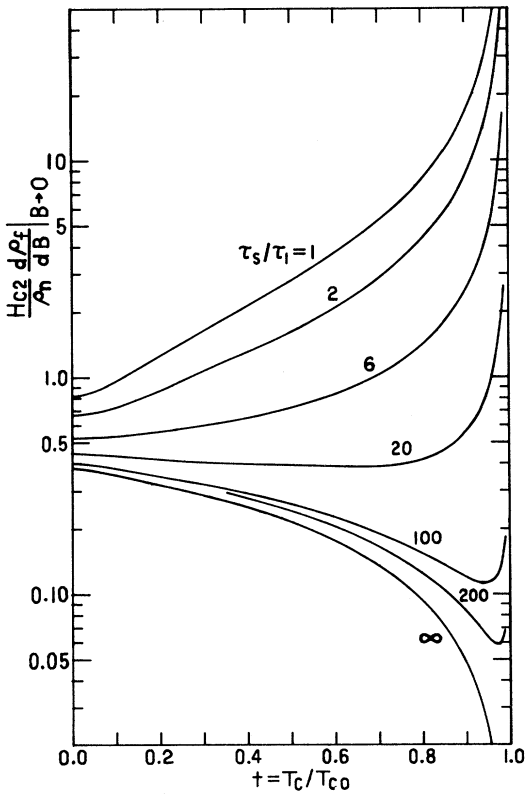

FIG. 1. The normalized initial slope of the flux-flow resistivity ρ_f with respect to the average magnetic induction B at $B=0$ plotted against the reduced transition temperature $t = T_c/T_{c0}$ for various values of τ_s/τ_1, the ratio of spin-flip to total scattering lifetime of the conduction electrons.

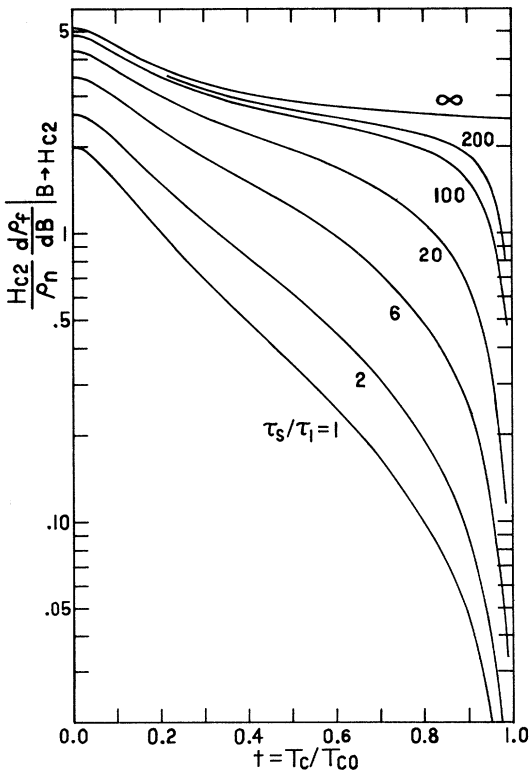

FIG. 2. Normalized final slope of ρ_f with respect to B at $B = H_{c2}$ plotted as in Fig. 1.

pure samples with a controlled amount of magnetic impurities of a type chemically very similar to the host atoms, preferentially also giving resonant spin-flip scattering to the conduction electrons. The condition $\kappa \gg 1$ assumed in this calculation is not a serious obstacle, because our previous work has indicated that the κ dependence of ρ_f is weak both in the high-field [see Eq. (20)] and in the low-field[2] limits. Furthermore, by using very thin films, even type-I materials can exhibit flux-flow behavior with an effective $\kappa \gg 1$.[4] Because of the wide variation of ρ_f predicted here, experimental work under the gapless conditions studied here is desirable.

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Two-Photon Photogeneration in Amorphous Selenium

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Two-photon photogeneration has been observed in amorphous selenium. The field dependence of this uniform bulk photogeneration process saturates at low fields and fits a Poole-Frenkel model down to 300 V/cm. The previously reported strong field dependence of the generation efficiency at low fields for strongly absorbed light is attributed to surface recombination.

Time-of-flight and charge-integration measurements have been invaluable tools for studying intrinsic photogeneration and charge transport in insulators. This is primarily because they enable one to determine the bulk current and effective transit time for either carrier (depending on the polarity of the bias voltage). Knowledge of both the current and the transit time then allows the rate of supply of carriers to the bulk to be unambiguously determined. For various experimental reasons these measurements have generally been restricted to the use of strongly absorbed light. Under these circumstances, however, since all of the carriers are produced very near the surface, surface recombination, trapping, and exciton dissociation, as well as bulk recombination in the generation region, can strongly influence the results. This is particularly serious for those materials where the onset of strong optical absorption occurs lower in energy than the onset of efficient photogeneration of free charge. In amorphous selenium, for example, it has not been possible to separate unambiguously the electric field dependence of intrinsic photogeneration from surface recombination and trapping effects[1-3] (all of which affect the rate of carrier supply), since only very strongly absorbed light generates carriers efficiently. Similar problems occur in molecular materials ranging from orthorhombic sulfur to anthracene.

We suggest that two-photon processes provide a powerful tool for the study of intrinsic photogeneration in these materials, since they allow carrier pairs to be created in the bulk with energies equal to pairs created by very strongly absorbed light near the surface. We report here the observation of two-photon absorption in amorphous selenium and the results of unambiguous photogeneration efficiency measurements using the two-photon process to generate bulk carriers *uniformly*. The observed efficiency of this bulk generation process is consistent with a Poole-Frenkel mechanism from 3×10^2 to 10^5 V/cm. This indicates that the linear field depen-