Properties of superconductors with a strong electron-phonon interaction

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An electron and phonon system with a strong coupling, $\lambda \gg 1$, is considered. The temperature dependence of the upper critical field H_{c2} has a positive curvature. A relationship is established among the following quantities: the electron state density $N(0)$, the abrupt change in the specific heat ΔC, the upper and lower H_{c1} critical fields, and the residual resistivity ρ.

The superconducting properties of several compounds are found to be markedly different from the standard behavior of the BCS model. These compounds include, for example, the organic superconductors β_H-(BEDT-TTF)$_2$I$_3$ with $T_c \simeq 8$ K and β-(BEDT-TTF)AuI$_2$ with $T_c \simeq 5$ K. In the first compound the curvature on the $H_{c2}(T)$ curve is positive for the field direction perpendicular to the layers, and the reduced field, $h_{c2}(T) = H_{c2}(T)/(-dH_{c2}/dT)_{T_c}$, reaches a value of 1.4 as $T \to 0$, which is twice as large as the corresponding BCS value.[1] In the second compound the H_{c2} curve is linear down to the lowest temperatures and at $T \ll T_c$ the ratio $\Delta(T)/T_c$ reaches a value[2] of 7, whereas in the BCS model it is equal to 1.76. Anomalies of approximately the same order of magnitude in the behavior of H_{c2} were detected in the system[3] Ba(Pb$_{1-x}$Bi$_x$)O$_3$. Differences from the BCS model of this magnitude cannot be explained in terms of the intermediate-coupling model, $\lambda \simeq 1$, although theoretical calculations based on Eliashberg's equations show that $h_{c2}(0)$ and $\Delta(0)/T_c$ have a tendency to increase with increasing λ (Refs. 4 and 5).

Let us consider the properties of the Eliashberg model with a single phonon mode [the spectral density of the electron-phonon interaction is $\alpha^2(\omega)F(\omega) = \lambda\Omega\delta(\omega - \Omega)$] as a function of λ for large values of λ. In this model $T_c = 0.18\Omega\sqrt{\lambda}$ when $\lambda \gg 1$ and the ratio $\Delta(0)/T_c$ increases monotonically with increasing λ, reaching 4.75 when $\lambda = 28$, according to the results of numerical calculations.[5,6]

The model with large values of λ which we are considering can be encountered in crystals near the structural instability. In general, the coupling constant and the phonon spectrum are described by the equation

$$\lambda = N(0)\langle I^2\rangle\left\langle \frac{1}{M\omega^2}\right\rangle, \qquad \langle\omega^2\rangle = \langle\Omega_0^2\rangle - \frac{4}{5}n_0 M^{-1}N(0)\langle I^2\rangle, \tag{1}$$

where $\langle I^2\rangle$ is the mean value of the squared matrix element of the electron-phonon interaction on the Fermi surface, M is the ion mass, n_0 is the number of electrons per atom, $\langle\omega^2\rangle$ is the mean value of the phonon frequencies squared, and Ω_0 is a pseudo-

atomic frequency (the seed phonon frequency in the Fröhlich model) (see Refs. 7–9). As the structural instability is approached, one of the modes softens and the coupling constant, which is determined primarily by the interaction of electrons with this mode, may increase without bound. The results of the model we are considering here can be used to describe a system even with a broader phonon spectrum. This can be done by replacing Ω^n by the corresponding moments of the function $\alpha^2(\omega)F(\omega)$ in the final expressions. As the structural instability point is approached, an increase in λ causes T_c to increase and $T_c \sim \Omega\sqrt{\lambda}$ to increase but, according to (1), the limiting value $T_{c\max} = 0.18\,[N(0)\langle I^2\rangle M^{-1}]^{1/2}$, whose upper limit is a value on the order of $0.2\Omega_0$, is reached as $\lambda \to \infty$.

Taking into account only the orbital effect of the field and ignoring the Coulomb repulsion in the Matsubara technique, we can write the equations for H_{c2} in the form (see Ref. 4)

$$\Delta(i\omega_n) = \pi T \sum_m \lambda(\omega_n - \omega_m)[\chi^{-1}(|\tilde{\omega}_m|) - (2\tau)^{-1}]\Delta(i\omega_m),$$
$$\tilde{\omega}_n = \omega_n + \pi T \sum_m \lambda(\omega_n - \omega_m)\operatorname{sign}\omega_m + (2\tau)^{-1}\operatorname{sign}\omega_n, \qquad (2)$$
$$\chi(x) = \frac{2}{\sqrt{\alpha}} \int_0^\infty dq e^{-q^2} \arctan[q\sqrt{\alpha}/x], \quad \alpha = \pi H_{c2} v_F^2/\Phi_0,$$
$$\lambda(\omega_n - \omega_m) = \lambda\Omega^2/[\Omega^2 + (\omega_n - \omega_m)^2], \qquad \omega_n = \pi T(2n-1),$$

where τ is the time it takes the electrons to be scattered by nonmagnetic impurities, and Φ_0 is a fluxoid. We will now consider a dirty superconductor, $\lambda\Omega\tau \ll 1$. For $\lambda \gg 1$ we find

$$H_{c2}(0) = 0.54\frac{\Phi_0 T_c}{D}\sqrt{\lambda}, \qquad D = \frac{1}{3}v_F^2\tau,$$
$$H_{c2}(T) = 1.20\frac{\Phi_0 T_c t}{D}, \qquad t = \frac{T_c - T}{T_c}, \qquad t \ll 1. \qquad (3)$$

From (3) we find the reduced field $h_{c2}(0) = 0.45\sqrt{\lambda}$ for $\lambda \gg 1$. For the intermediate values of λ the numerical calculation yields the curves $h_{c2}(T/T_c)$ shown in Fig. 1. For $\lambda \gg 1$ the field h_{c2} is much greater than the value (0.7) found in the BCS model. For $\lambda \gtrsim 4$ the curvature of the $h_{c2}(T/T_c)$ curve is positive.

The Gorkov relation for an arbitrary value of λ is

$$-H'_{c2} \equiv -(dH_{c2}/dT)_{T_c} = 8r(\lambda)ec\rho N(0)/\pi, \qquad (4)$$

where $r(\lambda)$ varies from 1.3 when $\lambda = 1$ to 2.86 in the limit $\lambda \to \infty$; $r = 1$ in the BCS model. For $\lambda \gg 1$ the electron specific heat $C_e(T)$ is not yet known near $T_c \gtrsim \Omega$. As $T \to 0$, we have $C_e = \gamma T$, where the large coefficient is $\gamma = (2\pi^2/3)k_B^2 N(0)(1+\lambda)$.

Expressing $H_{c2}(T)$ in terms of the correlation length $\xi(T)$ as $H_{c2} \sim \Phi_0/\xi^2$, we find near T_c the standard expression in the BCS theory, $\xi(T) \sim (\xi_0 l)^{1/2} t^{-1/2}$, where

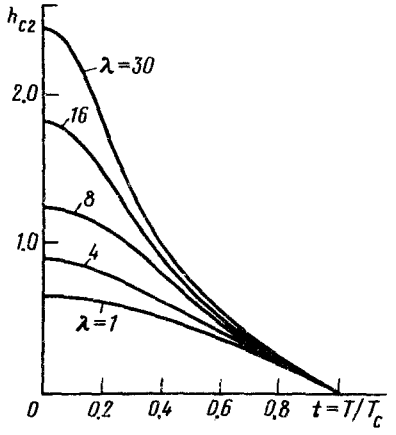

FIG. 1.

$l = v_f\tau$ and $\xi_0 \approx v_F/T_c$, but for $T \to 0$ and $\lambda \gg 1$ the length ξ_0 decreases, $\xi_0 \approx v_F/T_c\sqrt{\lambda}$. The crossover occurs at $T \approx \Omega$. The thermal phonons Ω are cold-trapped below this temperature. The presence of these phonons diminishes the contribution of the electron-phonon interaction to the Cooper pairs by a factor of $l/n_{\rm ph}(T)$, where $n_{\rm ph}(T)$ is the number of thermal phonons. At $T_c \gg \Omega$ from the relation $T_c \sim \lambda\Omega n_{\rm ph}^{-1}(T_c) \approx \lambda\Omega(\Omega/T_c)$ we finally obtain $T_c \sim \Omega\sqrt{\lambda}$, instead of $\lambda\Omega$, as would be the case in the coupling-constant (λ) model. At $T \ll \Omega$, however, the contribution of the electron-phonon interaction to the pairing increases and we can assume that the gap $\Delta(0) \sim \lambda\Omega$. Numerical calculations of Carbotte *et al.*[6] also suggest such a relation[6] [$\Delta(0) \approx 0.16\,\lambda\Omega$], although these authors have assumed on the basis of nonrigorous considerations that, like T_c, $\Delta(0) \sim \Omega\sqrt{\lambda}$. The results for H_{c2} obtained by us for a dirty superconductor suggest that $h_{c2} \sim \lambda$ in pure superconductors and that the positive curvature on the $H_{c2}(T)$ curve begins with $\lambda \approx 2$.

The free-energy functional for the Éliashberg model was found by Rainer and Bergmann[10] for the homogeneous order parameter $\Delta(i\omega_n)$,

$$(\mathcal{F}_n - \mathcal{F}_s)/N(0) = \pi T \sum_n \omega_n (\sin\varphi_n - \operatorname{sign}\omega_n)$$
$$+ \pi^2 T^2 \sum_{n,m} [\cos(\varphi_n - \varphi_m) - \operatorname{sign}(\omega_n \omega_m)]\lambda(\omega_n - \omega_m),$$
$$\Delta(i\omega_n) = \pi T \sum_m \lambda(\omega_n - \omega_m)\cos\varphi_m, \quad \varphi_n \equiv \varphi(i\omega_n). \tag{5}$$

Augmenting (5) with the gradient terms, which can easily be reconstructed near T_c from the known values of $H_{c2}(T)$, and restricting the analysis in (5) to the terms $\varphi = \varphi(\pm\pi T)$ (the accuracy of such an approximation for T_c is about 15%), we find the Ginzburg-Landau functional for the order parameter $\psi = \pi/2 - \varphi$ for $\lambda \gg 1$

$$\mathcal{F}_s - \mathcal{F}_n = N(0)(2\pi T_c)^2\left[\frac{1}{2}\xi^2 |(\vec{\nabla} - i\frac{2e}{c}\mathbf{A})\psi|^2 - t\psi^2 + \frac{1}{4}\psi^4\right], \quad (6)$$

where $\xi^2 = 0.27\, D/T_c$.

The Ginzburg-Landau functional implies the relations

$$N(0) = \frac{H'_{c1}H'_{c2}}{\ln(\kappa + 0.497)4\pi f(\lambda)} = \frac{\Delta C}{T_c f(\lambda)}; \quad \frac{\kappa^2}{\ln\kappa + 0{,}497} = \frac{H'_{c2}}{2H'_{c1}}. \quad (7)$$

For $\lambda \gg 1$ we have $f \approx 8\pi^2$; $f = 9.4$ in the BCS model. For $\lambda \gg 1$ the jump in the specific heat is 8.4 times higher than the corresponding value in the BCS model, but its relative value is small, because at $T_c \gtrsim \Omega$ the phonon and normal-electron components in the specific heat are large. The value $h_{c2}(0)$ and relations (4) and (7) can be used to estimate the value of λ from the experimental data.

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[1]F. Creuzet, G. Creuzet, D. Jerome, D. Schweitzer, and H. J. Keller, Phys. Lett. **46**, L1079 (1985).
[2]M. E. Hawley, K. E. Gray, B. D. Terris, H. H. Wang, K. D. Carlson, and J. M. Williams, Phys. Rev. Lett. **57**, 629 (1986).
[3]A. M. Gabovich and D. P. Moiseev, Usp. Fiz. Nauk **150**, 599 (1986) [Sov. Phys. Usp. **29**, 1135 (1986)].
[4]M. Schossmann and E. Schachinger, Phys. Rev. B **33**, 6123 (1986).
[5]B. T. Geilikman, V. Z. Kresin, and N. F. Masharov, J. Low Temp. Phys. **18**, 241 (1975).
[6]J. P. Carbotte, F. Marsiglio, and B. Mitrovic, Phys. Rev. B **33**, 6135 (1986).
[7]I. R. Gomersall and B. L. Gyorffy, Phys. Rev. Lett. **33**, 1286 (1974).
[8]P. B. Allen, in Dynamical Properties for Solids 3, ed. G. K. Horton and A. A. Maradudin, North Holland, 1980.
[9]O. V. Dolgov and E. G. Maksimov, Usp. Fiz. Nauk **138**, 95 (1982) [Sov. Phys. Usp. **25**, 688 (1982)].
[10]D. Rainer and G. Bergmann, J. Low Temp. Phys. **14**, 501 (1974).

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