THE MAGNETIC PROPERTIES OF SUPERCONDUCTORS WITH $l \neq 0$ PAIRING. THEORY OF THE KNIGHT SHIFT

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A theory of the Knight shift in superconductors is developed in which formation of pairs with nonzero angular momentum is taken into account. It is found that for pairing in the triplet state the Knight shift at T = 0 does not vanish. The theoretical predictions are in good agreement with the experimental data.

IN the theory of superconductivity [1,2] it is assumed that the Cooper pairs are in S-states. It should be noted that higher harmonics in the interaction between electrons, which arise as a result of exchange with phonons, have not been investigated. Evidently, these harmonics exist and can have an attraction sign. In this case, the formation of pairs with nonzero orbital momentum is possible [3-6] in superconductors as well as in superfluid He^3.

The author has shown [7] that the interaction between the conduction electrons of an antiferromagnet, which is brought about by the spin waves, leads to the formation of Cooper pairs in the triplet state. It is possible that this mechanism is responsible for the superconductivity of ruthenium and osmium, in which no isotope effect is observed. [8,9] The electrons in an antiferromagnet interact only if the projections of their spins on the axis of easy magnetization are oppositely directed. Hence the electrons in an anitferromagnet form pairs with $s_z = 0$.

There has been no experimental evidence up to this time that the spin of Cooper pairs is zero. Moreover, those experiments that can give information about the spin of the pairs can be explained by assuming that the Cooper pairs are formed in the triplet state. We have in mind here the measurements of the Knight shift of the frequency of nuclear magnetic resonance, [10,12] which at absolute zero has the same order of magnitude as in the normal state, in contradiction to the conclusions of the BCS theory. [1] According to this theory, the Knight shift, which is proportional to the spin paramagnetic susceptibility of the conduction electrons, should tend to zero as the temperature is lowered, [13,14] since the ordering of the spins of the electrons, which form Cooper pairs in the singlet state, is energetically unfavorable.

If the Cooper pairs form in the triplet state, then the ordering of the spins of the electron pairs in the magnetic field is energetically favorable, i.e., at T = 0 the paramagnetic susceptibility, and consequently the Knight shift, is different from zero. It will be shown below that the calculation of the paramagnetic susceptibility of a superconductor with pairing in the triplet state leads to good agreement with the experimental data. [1)]

The Hamiltonian of the interaction between the electrons taking into account the higher harmonics is of the form

$$\mathcal{H}_{int} = \frac{1}{2} \sum_{\mathbf{p}, \mathbf{p}', \sigma, \sigma', \mathbf{q}} V_{\sigma\sigma'}(\mathbf{p}, \mathbf{p}')\, a^{+}_{\mathbf{p}\sigma} a^{+}_{\mathbf{q}-\mathbf{p}, \sigma'} a_{\mathbf{p}'\sigma} a_{\mathbf{q}-\mathbf{p}', \sigma'}. \quad (1)$$

The coefficients $V_{l\sigma\sigma'}$ in the expansion of the "potential" of the interaction in Legendre polynomials

$$V_{\sigma\sigma'}(\mathbf{p}, \mathbf{p}') = \sum_{l} (2l+1)\, V_{l\sigma\sigma'} P_l(\cos \widehat{\mathbf{p}, \mathbf{p}'}),$$

characterize the intensity of the interaction of the electrons that form a pair with momentum l and a projection of spin $\sigma+\sigma'$, and a positive sign on $V_{l\sigma\sigma'}$ signifies attraction. For even l only the off-diagonal elements of the matrix $V_{l\sigma\sigma'}$ are different from zero, because of the Pauli principle.

In an isotropic system, when the interaction is independent of the direction of the total spin of the pair, all components of the matrix $V_{l\sigma\sigma'}$ with odd l are equal to each other: $V_{l\sigma\sigma'} = V_l$. In phonon superconductors the energy of the pair can depend on the direction of the spin because of the presence

[1)] Abrikosov and Gor'kov [15] have suggested that the source of the non-zero Knight shift is spin-orbit interaction on the boundaries of the crystallites. At the same time, the experiments of Androes and Knight [11] did not disclose any significant dependence of the Knight shift on the dimensions of the sample, although this is not a reliable conclusion, as Abrikosov and Gor'kov [15] have remarked.

of intracrystalline fields. In this case $V_{l\sigma\sigma} \neq V_{l\sigma,-\sigma}$. In a uniaxial antiferromagnet only those spins interact which have antiparallel spin projections, i.e., the diagonal elements $V_{l\sigma\sigma}$ are zero.

Unfortunately, at the present time there is no agreement about the symmetry properties of the state of thermodynamic equilibrium of a Fermi-system with pairing to nonzero momentum. Anderson and Morel [3] consider that in pairing with $l \neq 0$ the energy gap in the spectrum of elementary excitations of a system with the Hamiltonian (1) is anisotropic.[2)] Gor'kov and Galitskiĭ [4] have found another possible structure for the ground state of a system with Hamiltonian (1) by the Green's function method. The energy of the ground state assumed by Gor'kov and Galitskiĭ is less than in the scheme of Anderson and Morel, the gap in the elementary excitation spectrum is isotropic. Recently Balian and Werthamer [16] generalized the scheme of Anderson and Morel and, assuming that the pairs form in p-states and the interaction is independent of the spin direction, found by the variational method a solution with an isotropic gap that was not equivalent to the solution of Gor'kov and Galitskiĭ, although the values of the energy gap and the energy of the ground state calculated in [4] and [16] are the same. We shall calculate the spin paramagnetic susceptibility below by both methods.

In order to determine the susceptibility, it is necessary to find the average of the magnetization operator:

$$\mathbf{M} = -2\mu_0 (\psi^+(\mathbf{x})\, \mathbf{s}\psi(\mathbf{x})). \tag{2}$$

Here μ_0 is the Bohr magneton, $\psi^+(\mathbf{x})$ and $\psi(\mathbf{x})$ are second-quantization operators in the coordinate representation, and $\mathbf{s}$ is the electron spin operator. In the Hamiltonian of the interaction of the conduction electrons with an external magnetic field $\mathbf{H}$ we will take into account only the "paramagnetic" term:

$$\mathcal{H}_1 = 2\mu_0 \int d^3\mathbf{x}\psi^+(\mathbf{x})\, \mathbf{H}\mathbf{s}\psi(\mathbf{x}) \equiv \int d^3\mathbf{x}\mathcal{H}_1(\mathbf{x}). \tag{3}$$

We shall assume that the dimensions of the sample are such that the magnetic field within it is practically homogeneous, but we shall not take into account the alteration in the correlation functions caused by the fact that these dimensions are small. This alteration is not important in our problem if the spin-orbit interaction is neglected. [14]

For calculating the averages we make use of the "scattering matrix" of the thermodynamic theory of perturbations [17]:

$$S(\beta) = \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \int \cdots \int d^4x_1 \ldots d^4x_n T\mathcal{H}_1(x_1) \ldots \mathcal{H}_1(x_n). \tag{4}$$

Calculating the magnetization to first order in the magnetic field $\mathbf{H}$, it is not difficult to obtain for the components of the tensor χ_{ik} the following general relations:

$$\chi_{ik} = \mu_0^2 \sum_{\alpha,\beta,\gamma,\delta} (\sigma_i)_{\alpha\beta} (\sigma_k)_{\gamma\delta} \int d^4x' \langle \psi_\gamma^+(x')\, \psi_\delta(x')\, \psi_\alpha^+(\mathbf{x})\, \psi_\beta(\mathbf{x}) \rangle, \tag{5}$$

where $\hat{\sigma}_i$ are the Pauli matrices.

In the scheme of Gor'kov and Galitskiĭ [4] the averages of the four Fermi operators are expressed via Green functions for two operators:

$$\langle T\psi_\alpha(x)\, \psi_\beta^+(x')\, \psi_\gamma(y)\, \psi_\delta^+(y') \rangle = g_{\alpha\beta}(x - x')\, g_{\gamma\delta}(y - y') - g_{\alpha\delta}(x - y')\, g_{\gamma\beta}(y - x') - \sum_m F_{\alpha\gamma}^m(x - y)\, F_{\beta\delta}^{+(m)}(x' - y'). \tag{6}$$

The Green's functions $\hat{g}$, $\hat{F}^{(m)}$, and $\hat{F}^{+(m)}$ are determined as follows:

$$g_{\alpha\beta}(x - x') = -\langle T\psi_\alpha(x)\, \psi_\beta^+(x') \rangle, \tag{7}$$

$$F_{\alpha\beta}^{(m)}(x - x') = \langle N | T\psi_\alpha(x)\, \psi_\beta(x') | N + 2, l, m \rangle, \tag{8}$$

$$F_{\alpha\beta}^{+(m)}(x - x') = \langle N + 2, l, m | T\psi_\alpha^+(x)\, \psi_\beta^+(x') | N \rangle, \tag{9}$$

where the state $|N+2, l, m\rangle$ differs from the state $|N\rangle$ of the system with average number of particles N by the addition of pairs with momentum l and projection m.

Considering Eq. (6) and transforming to Fourier components of the Green's functions, we obtain

$$\chi_{ik} = \frac{\mu_0^2}{(2\pi)^3 \beta} \sum_n \sum_{\alpha,\beta,\gamma,\delta} (\sigma_i)_{\alpha\beta} (\sigma_k)_{\gamma\delta} \int d^3\mathbf{p} \left\{ -g_{\delta\alpha}(\mathbf{p}, \omega_n)\, g_{\beta\gamma}(\mathbf{p}, \omega_n) + \sum_m F_{\delta\beta}^{(m)}(\mathbf{p}, \omega_n)\, F_{\alpha\gamma}^{+(m)}(\mathbf{p}, \omega_n) \right\}. \tag{10}$$

In [4] the Fourier components of the Green functions depending on real time were calculated. A similar calculation for the Matsubara Green functions leads to the following formulas:

$$g_{\alpha\beta}(p, \omega_n) = -\frac{i\omega_n + \zeta_p}{\omega_n^2 + \varepsilon_p^2} \delta_{\alpha\beta}, \tag{11}$$

$$F_{\alpha\beta}^{(m)}(\mathbf{p}, \omega_n) = F(p, \omega_n)\, Y_{lm}(\mathbf{n})\, I_{\alpha\beta}, \tag{12}$$

$$F_{\alpha\beta}^{+(m)}(\mathbf{p}, \omega_n) = F(p, \omega_n)\, Y_{lm}^*(\mathbf{n})\, I_{\beta\alpha}^*, \tag{13}$$

$$F(p, \omega_n) = \sqrt{\frac{4\pi}{2l+1}} \frac{\Delta}{\omega_n^2 + \varepsilon_p^2}, \tag{14}$$

where $\epsilon_p = (\zeta_p^2 + \Delta^2)^{1/2}$ is the energy of the ele-

[2)] This means that in real superconductors the symmetry of the gap $\Delta(\mathbf{p})$ is different from the symmetry of the reciprocal lattice.

mentary excitations. The matrix $I_{\alpha\beta}$ is antisymmetric for pairing in the singlet state ($I^{(s)}_{\alpha\beta} = -I^{(s)}_{\beta\alpha}$) and symmetric for the triplet state ($I^{(t)}_{\alpha\beta} = I^{(t)}_{\beta\alpha}$). In both cases the matrix $\hat{I}$ is unitary: $\hat{I}\hat{I}^{+} = \hat{1}$. The matrix $\hat{I}^{(s)}$ (to within a phase factor $e^{i\alpha}$) equals

$$\hat{I}^{(s)} = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix} = i\hat{\sigma}_y. \tag{15}$$

If the interaction between the electrons forming a pair in the triplet state depends on the orientation of the total spin, then we can choose the coordinate axes in such a way that the maximum positive coefficient $V_{l\sigma\sigma'}$ will be non-diagonal. In the scheme of Gor'kov and Galitskiĭ this means that the only allowable value of s_z is zero, i.e.,

$$\hat{I}^{(t)} = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} = \hat{\sigma}_x. \tag{16}$$

If the interaction is isotropic, then for $s = 1$ the projection of the total spin of the pair takes values $s_z = -1, 0, 1$. In this case, in the absence of a magnetic field the choice of matrix $\hat{I}$ is not unique and is limited only by the conditions of unitarity and symmetry. This non-uniqueness reflects the presence of degeneracy, which is caused by the fact that in the equilibrium state the number of pairs with $s_z = 0$ can be arbitrary and is removed in an external magnetic field.

The change in free energy F in a magnetic field, calculated from the perturbation theory, depends on the choice of the matrix $\hat{I}$ of the zeroth approximation. The minimum free energy is attained if the quantity $\Sigma_\alpha(|I_{\alpha\alpha}|^2 - |I_{\alpha,-\alpha}|^2)$ is a maximum (here α is the projection of the electron spin on the direction of the magnetic field). Because of this requirement the matrix $\hat{I}^{(t)}$ for an isotropic system equals

$$\hat{I}^{(t)} = \begin{pmatrix} 1 & 0 \\ 0 & \pm 1 \end{pmatrix} \tag{17}$$

and only electrons with the same spin projections associate in pairs. For this choice of $\hat{I}$, the value of the susceptibility χ, which needs to be calculated from the formula for χ_{zz}, turns out to be greatest.

It is clear that in the isotropic case, i.e., for pairing in the singlet state, or when the interaction in the triplet state is independent of the orientation of the total spin of the pair, the susceptibility is also isotropic: $\chi_{ik} = \chi\delta_{ik}$. Below we will introduce the symbols $\chi^{(s)}$ and $\chi^{(t)}$ for the susceptibility of an isotropic superconductor for pairing in the singlet state and triplet state, respectively.

The susceptibility is anisotropic when the interaction of the electrons that form a pair with $s = 1$ depends on the orientation of the total spin relative to a chosen axis (z axis). This axis we choose in such a way that the only allowed (in the scheme of Gor'kov and Galitskiĭ) value of the total spin of the pair s_z equals zero. The values of the paramagnetic susceptibility of the current carriers in a field parallel to the chosen axis $\chi_{\|}$ and in a field perpendicular to this axis $\chi_\perp$ are different: $\chi_{\|} \neq \chi_\perp$. The Knight shift, therefore, is anisotropic.

Accomplishing the summation over the spin indices in Eqs. (14) and (15), and making use of the spherical harmonic addition theorem, we obtain:

$$\chi^{(s)} = \chi_{\|} = -\frac{2\mu_0^2}{(2\pi)^3\beta}\sum_n \int d^3\mathbf{p}\left\{g^2(\mathbf{p}, \omega_n) + \frac{2l+1}{4\pi}F^2(\mathbf{p}, \omega_n)\right\}, \tag{18}$$

$$\chi^{(t)} = \chi_\perp = -\frac{2\mu_0^2}{(2\pi)^3\beta}\sum_n \int d^3\mathbf{p}\left\{g^2(\mathbf{p}, \omega_n) - \frac{2l+1}{4\pi}F^2(\mathbf{p}, \omega_n)\right\}. \tag{19}$$

It is convenient to transform from a summation over n to an integration by means of the substitution

$$\sum_n \ldots \to \frac{i\beta}{4\pi}\int_{(C)} d\omega \operatorname{tg}\frac{\beta\omega}{2}\ldots, \tag{20}$$

where (C) is a contour consisting of two straight, parallel real axes. Closing the contour by semicircles and summing the residues of the sub-integral functions in the points $\omega = \pm i\epsilon_p$, we find that

$$\chi^{(s)} = \chi_{\|} = \frac{\mu_0^2}{(2\pi)^3}\frac{\beta}{2}\int \frac{d^3\mathbf{p}}{\operatorname{ch}^2(\beta\varepsilon_p/2)}, \tag{21}$$

$$\chi^{(t)} = \chi_\perp = \frac{\mu_0^2}{(2\pi)^3}\int d^3\mathbf{p}\left\{\frac{\beta\zeta_p^2}{2\varepsilon_p^2}\operatorname{ch}^{-2}\frac{\beta\varepsilon_p}{2} + \frac{\Delta^2}{\varepsilon_p^3}\operatorname{th}\frac{\beta\varepsilon_p}{2}\right\}. \tag{22}$$

Calculating the integrals in Eqs. (21) and (22), we obtain

$$\chi^{(s)}/\chi_0 = \chi_{\|}/\chi_0 = N_n(\beta)/N, \tag{23}$$

$$\chi^{(t)}/\chi_0 = \chi_\perp/\chi_0 = 1, \tag{24}$$

where $\chi_0 = \mu_0^2 p_0^2/\pi^2 v_0$ is the Pauli paramagnetic susceptibility of free electrons (p_0 and v_0 are the momentum and velocity on the Fermi surface), and $N_n(\beta)/N$ is the ratio of the number of "normal" electrons to the total number of conduction electrons:

$$\frac{N_n(\beta)}{N} = \int_0^\infty \operatorname{ch}^{-2}\sqrt{z^2 + \left(\frac{\beta\Delta}{2}\right)^2}\,dz. \tag{25}$$

The quantities $\chi^{(s)}$ and $\chi_{\|}$ tend rapidly toward zero at low temperatures.

The paramagnetic susceptibility of polycrystalline anisotropic superconductors equals

*tg = tan.

†ch = cosh, th = tanh.

$$\chi^{(t)}_{\text{polycr}} = \frac{1}{3}\chi_{\parallel} + \frac{2}{3}\chi_{\perp} = \chi_0\left(\frac{2}{3} + N_n(\beta)/3N\right). \quad (26)$$

The Knight shift in mercury[10] and tin[11] at absolute zero equals approximately $\frac{2}{3}$ of the shift in the normal state, and in vanadium[12] the Knight shift apparently does not depend on temperature and does not change in the superconducting transition. Thus, a nonzero Knight shift can be explained if one assumes that the Cooper pairs form in the triplet state.

Until now we have been using the method of Gor'kov and Galitskiĭ to calculate susceptibilities. In the scheme of Anderson-Morel-Balian-Werthamer[3,16] the uncoupling of two-particle systems is the same as in the work of Gor'kov[18] for pairing in S-states:

$$\langle T\psi_\alpha(x)\,\psi^+_\beta(x')\,\psi_\gamma(y)\,\psi^+_\delta(y')\rangle = g_{\alpha\beta}(x-x')\,g_{\gamma\delta}(y-y') - g_{\alpha\delta}(x-y')\,g_{\gamma\beta}(y-x') - F_{\alpha\gamma}(x-y)\,F^+_{\beta\delta}(x'-y'). \quad (27)$$

It is easy to show that for pairing in the singlet state, or in the case when in the interaction $V_{\sigma\sigma'}(\mathbf{p},\mathbf{p}')$ only the off-diagonal components $V_{\sigma,-\sigma}(\mathbf{p},\mathbf{p}')$ are nonzero, i.e., in the triplet state, the only allowed value of the projection of the total spin of the pair s_z is zero,[3)] the method of [3,16] leads to Eqs. (23), (24), and (26), obtained by the method of Gor'kov and Galitskiĭ. It is necessary to remember, however, that if the gap is anisotropic, then the ratio of the number of normal electrons to the total number of conduction electrons $N_n(\beta)/N$ does not have the form (25), although, of course, it tends toward zero as the temperature is lowered.

We will now assume that the interaction is isotropic: $V_{\sigma\sigma'}(\mathbf{p},\mathbf{p}') = V(\mathbf{p},\mathbf{p}')$. For pairing in the triplet state all components of the matrices $\hat{F}(\mathbf{x}-\mathbf{x}')$ and $\hat{F}^+(\mathbf{x}-\mathbf{x}')$ are different from zero:

$$\hat{F}(\mathbf{p},\omega_n) = \hat{\Delta}(\mathbf{p})/(\omega_n^2 + \varepsilon_p^2), \quad (28)$$

where $\hat{\Delta}(\mathbf{p})$ satisfies the equation

$$\hat{\Delta}(\mathbf{p}) = \frac{1}{(2\pi)^3\beta}\sum_n \int d^3\mathbf{p}'\,V(\mathbf{p},\mathbf{p}')\,\frac{\hat{\Delta}(\mathbf{p}')}{\omega_n^2 + \varepsilon_{\mathbf{p}'}^2} \quad (29)$$

and the normalization condition

$$\hat{\Delta}(\mathbf{p})\,\hat{\Delta}^+(\mathbf{p}) = \Delta^2(\mathbf{p})\,\hat{1}. \quad (30)$$

It can be shown that this solution is asymptotically accurate.

The solution of Balian and Werthamer,[16] who considered pairing in p-states, has the form

$$\hat{\Delta}(\mathbf{p}) = \Delta\hat{\mathbf{n}}\hat{\sigma}\hat{\sigma}_l, \quad (31)$$

where $\mathbf{n}$ is unit vector in the direction of the momentum $\mathbf{p}$, and the energy gap Δ satisfies the same equation as in the model of Gor'kov and Galitskiĭ[4]:

$$1 = \frac{V_1}{2(2\pi)^3}\int d^3\mathbf{p}\,\frac{1}{\varepsilon_p}\,\text{th}\,\frac{\beta\varepsilon_p}{2}. \quad (32)$$

Balian and Werthamer, by assuming that $\Delta(\mathbf{p}) = \hat{\Delta}\hat{\mathbf{n}}\hat{\sigma}\hat{\sigma}_y$, found that for pairing in a p-state the susceptibility equals $\chi^{(t)} = \chi_0(\frac{2}{3} + N_n(\beta)/3N)$. Thus, independently of model, one can successfully explain the nonzero Knight shift in superconductors at $T = 0$, by assuming that the pairs are in a triplet state.

Measurements of the anisotropy of the Knight shift in monocrystalline samples can give not only information about the spin of the Cooper pairs but also throw light on the properties of the interaction $V_{\sigma\sigma'}(\mathbf{p},\mathbf{p}')$ between the electrons.

In conclusion, without giving the details of the calculation, we write the relation between the Fourier components of the current $\mathbf{j}(\mathbf{x})$ and the transverse component of the vector potential $\mathbf{A}(\mathbf{x})$ obtained from the model of Gor'kov and Galitskiĭ[4][4)]:

$$\mathbf{j}(\mathbf{k}) = -(Ne^2/m)\,Q(k)\,\mathbf{A}(\mathbf{k}); \quad (33)$$

$$Q(k) = 1 + \frac{3}{4\beta}\sum_n \int_0^\pi \sin^3\theta\,d\theta \times \int_{-\infty}^{\infty} d\zeta\,\frac{(i\omega_n+\zeta_+)(i\omega_n+\zeta_-) + \Delta_+\Delta_- P_l(\mathbf{n}_+\mathbf{n}_-)}{(\omega_n^2+\varepsilon_+^2)(\omega_n^2+\varepsilon_-^2)}. \quad (34)$$

We have introduced the abbreviated symbols $\mathbf{p}_\pm = \mathbf{p} \pm \mathbf{k}/2$ and chosen the polar axis in p-space along the vector $\mathbf{k}$.

It is easy to show that for characteristic values of $\mathbf{k}$ that are of order of the inverse skin depth δ, the nucleus of $Q(\mathbf{k})$ does not depend on l to the accuracy of terms of order $(la/\delta)^2$, where a is the lattice constant.

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[3)] In this case, when all differences between the maximum coefficient $(V_{l\sigma,-\sigma})_{\max}$ and the other coefficients $V_{l'\sigma\sigma'}$ have the same order of magnitude as $(V_{l\sigma,-\sigma})_{\max}$, the number of pairs with $s = 1$ and $s_z \neq 0$ calculated by the method of Anderson and Morel[3] and Balian and Werthamer[16] are found to be negligibly small, i.e., these pairs are of no consequence in our problem.

[4)] Such a relation with $l = 1$ can be obtained by the method of Balian and Werthamer[16] in the case of isotropic pairing in a p-state.

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