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Abstract

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PHYSICS

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ON THE QUESTION OF MECHANISMS OF SUPERCONDUCTIVITY

(Presented by Academician Ya. B. Zel'dovich, 22 IV 1965)

In Little's work ⁽¹⁾ the question of superconductivity in polymer chains is considered. Serious difficulties arise here in connection with the one-dimensional character of the model proposed by him. B. T. Geilikman ⁽²⁾ considered a possibility apparently realized in alloys, i.e., in the usual three-dimensional case. The Cooper pairing in his work is due to the interaction of the metal *s*-electrons not with phonons, but with the electrons of unfilled *d*- and *f*-shells (transition metals), or with the electronic system of an isolated atom. As a result an effective attraction arises between the *s*-electrons, corresponding to large values of the critical temperature. V. L. Ginzburg ⁽³⁾ proposed a mechanism for the establishment of surface superconductivity, caused by the interaction of the electrons of the surface layer with a dielectric deposited on the surface (see also ⁽⁴⁾).

In the present work one more possibility for the establishment of a superconducting state is considered, analogous in a number of respects to the usual Fröhlich mechanism, but allowing interaction of electrons in a broader energy region near the Fermi surface.

The role of the system responsible for the interelectronic attraction may be played by molecules embedded in the crystal lattice. In conducting molecular crystals, the molecules of which it is composed are considered. Molecules are characterized by vibrational energy levels ΔE , the separation of which is usually much larger than θ (θ is the Debye temperature; for example, for HCl $\Delta E \simeq 4 \cdot 10^3^\circ$, for NO $\Delta E \simeq 2.5 \cdot 10^3^\circ$, for C₆H₆ $\Delta E \simeq 9 \cdot 10^2^\circ$). The effective interelectronic interaction that arises in this case may be represented as follows: an electron, by its field, causes polarization of the molecule, accompanied by its transition to an excited vibrational level. Polarization of the molecule causes changes in the electric field created by it, which affects the state of another electron. The energy of the latter changes owing to the return of the molecule to its ground state. The interaction between the electrons may be regarded as an exchange of "virtual" quanta describing the excitation of the vibrational molecular levels. The mechanism described is completely analogous to the usual one, with the only difference that, in the case considered, the role of the vibrational

energy levels of the lattice is played by the vibrational levels of the molecule. It is essential that $\Delta E \gg k\theta$.

The effective interaction is then described by the expression:

$$\Gamma = g^2 D(\varepsilon_2 - \varepsilon_1, \Delta E) = g^2 \frac{\Delta E}{(\varepsilon_2 - \varepsilon_1)^2 - (\Delta E)^2},$$

where $\varepsilon_2 - \varepsilon_1$ is the change in electronic energy; ΔE is the energy of the vibrational molecular quantum (attraction arises, as usual, for $\varepsilon_2 - \varepsilon_1 < \Delta E$); D is the Green's function describing the propagation of the vibrational quantum, g is the matrix element, equal to

$$g = \int \psi_1^* \psi_1^* u \nabla V \psi_2 \psi_{II} d\tau, \quad (2)$$

where ψ_1, ψ_2 are electron wave functions that are eigenfunctions of the unperturbed molecular-oscillation Hamiltonian; ψ_I and ψ_{II} are products of functions describing linear harmonic oscillators corresponding to molecular normal vibrations; u is the displacement in the molecule; $u \nabla V$ is the potential of interaction of an electron with molecular vibrations, quite analogous to the potential of electron-phonon interaction ⁽⁵⁾.

The Hamiltonian of the four-fermion interaction under consideration can be written in the form

$$H = \sum_{k_2, k_1, n} g^2 \frac{(\Delta E)_n}{(\varepsilon_{k_1} - \varepsilon_{k'})^2 - (\Delta E)_n^2} a_{k_1} a_{k_2} a_{k'_1}^+ a_{k'_2}^+ \simeq g_{\text{eff}} \sum_{k_i, k'_i} a_{k_1} a_{k_2} a_{k'_1}^+ a_{k'_2}^+, \quad (3)$$

Further investigating Hamiltonian (3) by the methods of the usual theory of superconductivity ^(6, 7), we obtain the expression for the critical temperature

$$T_k \simeq \Delta E \exp[-1/N(0)g_{\text{eff}}] \quad (4)$$

($N(0)$ is the density of states near the Fermi surface).

The quantity g_{eff} , equal, as is seen from (1) and (2), to $g_{\text{eff}} \simeq g_1 \theta^2 / (\Delta E)^2 \gamma$ ($\gamma = M_1/M$; M is the mass of a lattice ion; M_1 is the effective mass of the harmonic oscillator, $g_1 = C^2/MN u_0^2$; C is the mean kinetic energy of the electron due to its interaction with the incorporated molecules), can vary depending on the values of the parameters $\theta/\Delta E$, γ , and g_1 . Thus, for example, when molecules possessing considerable dipole moments (for example, C_6H_5Cl , HCl , etc.) are incorporated in sufficient concentration, the value of g_1 becomes comparable with the magnitude of the ordinary electron-phonon interaction constant g_n .

Substituting into the expression for g_{eff} the possible values $\theta \simeq 10^2$, $\Delta E \simeq 10^3$, $\gamma \simeq 10^{-2}$, we obtain $g_{\text{eff}} \simeq g_n$. In this case the relation $\Delta E \gg \theta$ makes possible, according to (4), a state with a value of the critical temperature larger than usual.

The indicated value of γ is obtained when molecules consisting mainly of light atoms are incorporated into a lattice containing heavy ions (for example, into Hg; mercury is, moreover, of interest because of its low melting point, which facilitates incorporation of molecules, usually carried out into a molten substance with subsequent cooling).

For setting up the experiment it is necessary to have a metal or semiconductor with molecules incorporated in sufficiently high concentration. Such incorporation is analogous to obtaining inclusion compounds⁽⁸⁾ or molecular crystals. In molecular spectroscopy, where these substances are used, small concentrations of included molecules are required. Incorporation with a higher concentration is, however, quite possible. Evidently, the inclusion required for establishing the superconducting state is easier to achieve in metallic or semiconductor films. A method for obtaining the required inclusions may also be co-condensation of vapors containing molecules of the host and incorporated substances.* It is possible to obtain a crystal with an ordered arrangement of incorporated molecules.

In the work of V. L. Ginzburg and D. A. Kirzhnits⁽⁹⁾, as is known, the question of surface superconductivity was first considered. It is possible that this effect may be observed owing to ordinary adsorption of molecules on a surface, leading, by means of the mechanism considered above, to Cooper pairing of electrons localized at surface levels.

At present molecular crystals with high electrical conductivity are known (see, for example, ⁽¹⁰⁾), due to overlap of the orbitals

* The existence of this method was communicated to the author by Ya. B. Zel'dovich.

π -electrons. In this case superconductivity associated with the interaction of conduction electrons with the vibrational degrees of freedom of the molecules forming the crystal is possible. The estimates given above, of course, remain valid in this case as well.

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Note: Figure translations are in progress. See original paper for figures.

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