High-temperature resistivity and the superconductivity of potassium and rubidium at high pressures

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The growth of the high-temperature resistivity of K and Rb upon compression is attributed to electron rearrangement manifested by a change of the relative positions of the *s* and *d* bands in the conduction band, which leads to an increase in localization of the conduction electrons. The resistivity ρ is calculated on the basis of data on the wave functions and structure of electron bands obtained in the Wigner–Seitz spherical approximation. The calculated dependence of ρ on the degree of compression possesses a maximum which coincides with the beginning of the filling of the *d* bands. The K and Rb superconductivity parameters as functions of compression are calculated in the same approximation. The superconducting transition of K (at $P \sim 150$ kbar) and Rb (at $P \sim 100$ kbar) is predicted quantitatively. The main cause of appearance of superconductivity in these metals is the restructuring of the electron spectrum upon compression.

1. INTRODUCTION

There is a whole series of metals at the present time which undergo a rearrangement of their electron structure under pressure, which has a significant effect on their properties. In particular, such metals include the heavy alkali metals K, Rb and Cs, according to the band calculations of various authors.[1,2] These metals have the property of "pretransition," which is expressed in the presence in their electron spectrum of relatively low-lying d-bands, which are not filled at normal pressure. Thus, there is a partially filled 4s-band in K at normal pressure and an unfilled 3d-band is located rather close to it, above the Fermi level. There is a similar situation in Rb, but the bands mentioned are replaced by the 5s- and 4d-bands, respectively. At a relative compression $\delta = 2.5$ ($\delta = \rho/\rho_0$, ρ_0 is the density of the normal material and ρ that of the compressed material) in K and $\delta = 2$ in Rb, is an electron rearrangement takes place, consisting of the filling of the previously empty d-band. This materially changes the character of the electron-phonon interaction, leads to the appearance of additional channels of electron-phonon scattering and to a sharp increase in the matrix element of electron-phonon interaction, which can in turn lead on the one hand to a strong increase in the resistivity and on the other to the appearance of superconductivity.

The growth of the resistivity of K and Rb upon compression has actually been observed experimentally by Drikhamer[3,4] and the size of the effect is very great (for a compression to 0.5 Mbar, the resistivity increases by almost two orders of magnitude). There are as yet no experimental data on the superconductivity of K and Rb.

The present paper is devoted to a quantitative account of the effect of the described electron rearrangement under pressure on the electrical conductivity, and the possibility of appearance of superconductivity (the possibility of explanation of pressure-induced superconductivity was suggested in[5]).

It should be noted that account of an unfilled d-band close to an s-band and the consequent hybridization of the s- and d-bands in heavy alkali metals is important even at normal density. This is confirmed, for example, by the results of the research of Bortolani and Calandra,[6] where account of the unfilled d-band, considered as a resonant d-level, in the calculation of the electric resistivity of I, Rb and Cs, led to a significant improvement in the agreement between the calculated results and experiment.

The model chosen by us for calculations of the resistivity is discussed in Sec. 2, along with the superconducting parameters of K and Rb under pressure. Section 3 contains the results of calculations of the high-temperature lattice resistivity. Section 3 also gives the equations that generalize the equations of superconductivity theory to the case of a multiband metal, from the solution of which a BCS-type formula is obtained for the critical temperature of such a metal. Quantities which characterize the superconducting properties are then expressed in terms of the band-structure parameters in the spherical model, after which the results of calculations of the superconducting parameters in K and R are given for various pressures, as well as their analysis.

2. CHOICE OF MODEL

For the calculation of the quantities of interest to us, we use the self-consistent field approximation. This corresponds to the Hartree self-consistent field method for the electron spectrum and wave functions, and also the random phase approximation for the effective electron-electron and Coulomb interactions. For specific calculations of the compression dependence of the resistivity and, in what follows, also the superconductivity parameters, we shall use the wave functions and energy eigenvalues in the spherical approximation.[7] This means that the solution of the Hartree self-consistent set of equations is sought in the region of the unit cell of the crystal, which is replaced by an equivalent sphere with center at the lattice site, with boundary conditions replaced by Bloch conditions on the sphere.

The wave functions $|i\mathbf{p}\rangle$ inside the Wigner-Seitz sphere are of the form

$$|i\mathbf{p}\rangle = \sum_{l \geqslant |\mu|} A_{il}(p) f_l(\xi_i(p), r) Y_{l\mu}^{\mathbf{p}_0}(\mathbf{n}), \qquad (1)$$

where $\mathbf{u} = \mathbf{r}/|\mathbf{r}|$, the index $\mathbf{p}_0$ for the spherical harmonics Y_{lm} indicates that their arguments are defined in the set of coordinates with axis $z \parallel \mathbf{p}$. The numbers

μ_i classify the bands with given values of the projection of the orbital angular momentum of the electron in the direction of the quasimomentum p, $\xi_i(p)$ is the unrenormalized electron-phonon interaction energy of the electron Fermi excitations, reckoned from the chemical potential.

Such a choice is based on the supposition, confirmed by the results of the calculations given below, that the basic role both in the anomalous growth of the resistivity and in the development of superconductivity is played by the matrix element of the effective electron-phonon interaction, due to the rearrangement of the electron structure under pressure and the large change in the radial functions f_l associated with it; all the other factors (phonon spectrum, anisotropy of the collision times and the superconducting gap, etc.) are less essential.

For the same reason, we chose the spherized Debye spectrum with one longitudinal and two transverse modes as the model for the phonon spectrum. The expressions for the longitudinal and transverse sound velocities

$$c_l = c_{BS}\left[p_F m \Big/ \sum_i p_i m_i - 9r_s/35p_F^2\right]^{1/2},$$
$$c_t = c_{BS}[9r_s/70p_F^2]^{1/2}$$

(c_{BS} is the velocity from the Bohm-Staver formula, m is the mass of the free electron, p_F is the Fermi momentum of a homogeneous electron gas with density equal to the density of the conduction electrons in the metal; $r_S = r_0/a_0$, r_0 is the radius of the Wigner-Seitz sphere, and a_0 is the Bohr radius) were obtained by generalization of the well-known Bohm-Staver formula[8] for the longitudinal sound velocity in the jellium model with account of multi-bandedness and the microscopic inhomogeneity of the metal.

3. ELECTRIC RESISTIVITY

The natural generalization of the usual approximation[9] in the calculation of high-temperature electrical conductivity ($T \gg \Theta_D$) to the case of several overlapping bands is the following set of equations:

$$\sum_j (R_{ij}\tau_i - S_{ij}\tau_j) = 1; \tag{2}$$

$$R_{ij} = \frac{v_0 T}{2\pi M} p_i m_j \int d(\cos\theta_{p'}) \sum_s \frac{|\langle j\mathbf{p}'|\varphi_{s\mathbf{k}}|i\mathbf{p}\rangle|^2}{\omega_s^2(\mathbf{k})}, \tag{3}$$

$$S_{ij} = \frac{v_0 T}{2\pi M} p_i m_j \frac{v_j}{v_i} \int d(\cos\theta_{p'})\cos\theta_{p'} \sum_s \frac{|\langle j\mathbf{p}'|\varphi_{s\mathbf{k}}|i\mathbf{p}\rangle|^2}{\omega_s^2(\mathbf{k})}. \tag{4}$$

(The expression (4) takes on a similar form after averaging over the orientations of the crystal lattice.) The following notation is used here: p_i and v_i are the Fermi momentum and velocity in the i-th band, $m_i = p_i/v_i$ is the effective mass, v_0 is the volume of the unit cell, M is the mass of the ion, and φ_{sk} is the effective potential of electron-phonon interaction in the dielectric screening approximation.

The bare potential φ^0_{sk} is of the form

$$\varphi_{s\mathbf{k}}^0(\mathbf{r}) = (\mathbf{e}_s\mathbf{k})\nabla_r \sum_\alpha v(\mathbf{r} - \mathbf{R}_\alpha)\exp\{i\mathbf{k}\mathbf{R}_\alpha\}, \tag{5}$$

where $v(r - R_\alpha)$ is the potential of the electron-phonon interaction, e_S is the polarization vector, and R_α is the crystal lattice vector.

With allowance for the relaxation times τ_i that have been introduced, we get the following elementary relation for the resistivity[1)]

$$\rho = \frac{3\pi^2}{e^2}\left(\sum_i \frac{p_i^3\tau_i}{m_i}\right)^{-1}. \tag{6}$$

The expression for the matrix element of the bare electron-phonon interaction (5) in the spherical approximation has the form

$$\langle j\mathbf{p}'|\varphi_{s\mathbf{k}}^0|i\mathbf{p}\rangle = \sum_{l_1 l_2} A_{jl_2}(p_j) A_{il_1}(p_i) N^s_{l_2 l_1}(k) Q^{ij,s}_{l_2 l_1}(\mathbf{p}', \mathbf{p}),$$
$$N^s_{l_2 l_1}(k) = J_{l_1 l_2}\delta_{l_1} + I^s_{l_2 l_1}(k),$$

and the radial integral

$$J_{l_1 l_2} = \int_0^{r_0} dr\, r^2 f_{l_1}(r) f_{l_2}(r) \frac{dv_0(r)}{dr}$$

corresponds to scattering by a central ion, while $I^S_{l_2 l_1}(k)$ is the contribution from scattering by the remaining ions of the lattice; $Q^{ij,s}_{l_2 l_1}$ is the integral over the solid angle of the product of three spherical harmonics. In the region of Umklapp processes, the expression

$$|\langle j\mathbf{p}'|\varphi_{s\mathbf{k}}|i\mathbf{p}\rangle|^2/\omega_{sk}^2,$$

which enters into (3) and (4), was averaged over the directions of the reciprocal lattice vector in a manner similar to that used by Jones.[9]

The resistivities of K and Rb were calculated with a computer for different degrees of compression, within the framework of the scheme set forth above. In the expansion of the wave functions (1), we can limit ourselves, with excellent accuracy, to terms up to $l = 3$, inclusively, Figure 1 shows the theoretical dependence of the resistivity of K and Rb on the degree of compression δ. Both in K and in Rb there is a sharp increase in the resistivity, with a maximum at the emergence of the d-band to the Fermi surface. This takes place in K at $\delta = 2.5$ and in Rb at $\delta = 2$ (see Figs. 2 and 3, where the band structures of K and Rb are shown in the Wigner-Seitz spherical model for several degrees of compression).

Direct comparison of our data with the results of Stager and Drikhamer[4] is difficult, since they made no corrections for the changes in the geometry of the sample under compression and in the resistance of the contact. Allowance for these changes when the resistivity is calculated from the total resistance can lead to the appearance of the maximum that we obtained in our calculations on the experimental curve. With account of this circumstance, the agreement of our data with the experimental results is quite good.

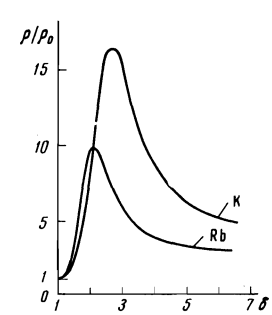

FIG. 1. Dependence of the resistivity of K and Rb on the degree of compression δ.

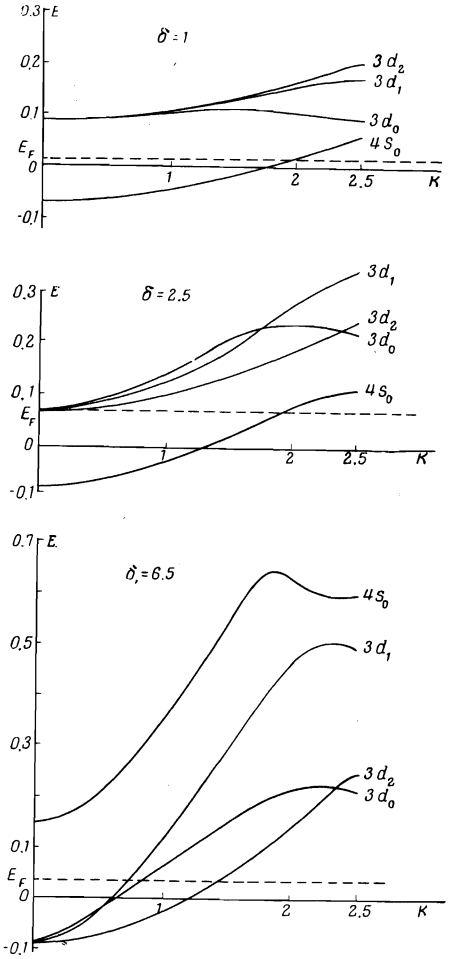

FIG. 2. Energy bands in K for different degrees of compression.

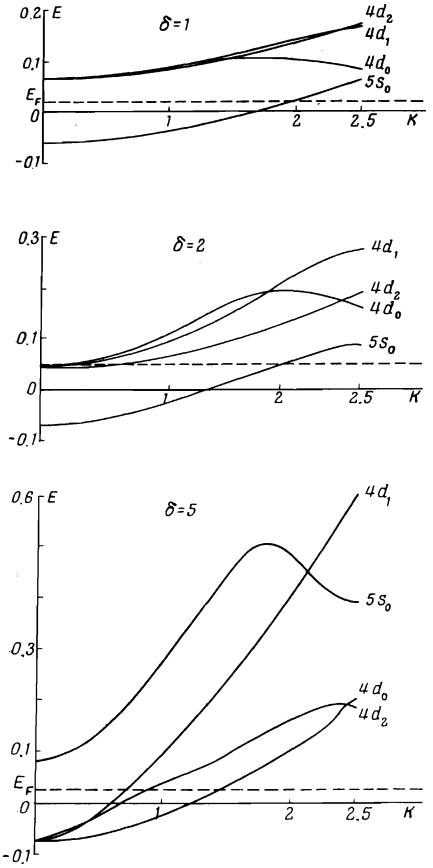

FIG. 3. Energy bands in Rb for different degrees of compression.

As has already been pointed out above, the growth of the resistivity is connected fundamentally with the strong change in the radial functions $f_l(\xi, r)$ with pressure, which reflects the increase in the localization of the conduction electrons. This leads to an increase in the radial integrals $N^S_{l_2 l l_1}$ due to a sharp growth of the terms $J_{l_1 l_2}$ corresponding to scattering by the central ion. This growth masks the decrease in the resistivity which would have taken place as a consequence of the increase in the Debye frequency of the carriers with compression, assuming no change in the character of the wave functions. As the d-bands begin to fill up, the growth of the integrals J slows down, so that the already mentioned factors that contribute to the decrease in the resistivity become dominant. Thus a curve with a maximum is obtained and is evidently characteristic of metals in which electronic rearrangement under pressure takes place.

It should be noted that, within the framework of the representations that have been developed, the weaker increase in the resistivity of Rb is connected with the fact that the electrons of the 4d-band in it are less localized than the 3d electrons in K.

The change in the coefficients A_l for the s-band with compression, up to pressures corresponding to the moment of emergence of the d-band to the Fermi surface, is given in Table I for K and Rb. These data make it possible to assess the change in the filling of the s-band with compression. The dimensionless values of the integrals $J_{l_1 l_2}$ are also given in the table in units of $4Z^{5/3}(e^2/a_0^2) \times 10^4$.

4. SUPERCONDUCTIVITY

To study the problem of the appearance of superconductivity in a metal which undergoes the above-discussed rearrangement of the electronic structure under pressure, it is necessary to modify the usual gap equation. Instead of it, as can be shown, we have the following set of integral equations

$$\Delta_i(\mathbf{p}_0) = \ln\frac{\omega_0\gamma}{\pi T_c}\sum_j \int \frac{dS_{\mathbf{p}_0'}}{v_j} g_{ij}(\mathbf{p}_0, \mathbf{p}_0')\Delta_j(\mathbf{p}_0'), \tag{7}$$

$$g_{ij}(\mathbf{p}_0, \mathbf{p}_0') = [\lambda_{ij}(\mathbf{p}_0, \mathbf{p}_0') - \mu_{ij}^*] \Big/ \Big[1 + \sum_l \int \frac{dS_{\mathbf{p}_0''}}{v_l}\lambda_{il}(\mathbf{p}_0, \mathbf{p}_0'')\Big], \tag{8}$$

$$\mu_{ij}^* = \Big[\Big(\hat{1} + \hat{\mu}\ln\frac{\omega_p}{\omega_0}\Big)^{-1}\hat{\mu}\Big]_{ij}, \tag{9}$$

where λ_{ij} and μ_{ij} are the electron-phonon and Coulomb matrices, which correspond to the constants λ and μ in the single-band case;[10] i and j are the band indices; $\mathbf{p}_0 = \mathbf{p}/|\mathbf{p}|$; $\int dS_{p_0'}$ denotes integration over the Fermi surface of the j-th band;

$$v_j = \int \frac{d\mathbf{p}'}{(2\pi)^3}\delta(\xi_j(\mathbf{p}'))$$

is the density of states on the Fermi surface; ω_p is the plasma frequency of the electrons, ω_0 a quantity of the order of the Debye frequency. Neglecting the dependence of the gaps and of the interaction on the absolute value of the quasimomentum is obviously equivalent to a generalization of the properties of the isotropic model

TABLE I. Data for the 4s-bands and the values of the integrals $J_{l_1 l_2}$ in K and Rb

	K				Rb		
	$\delta = 1$	1.5	2	2.5	$\delta = 1$	1.5	2
A_0	−0.635	−0.631	−0.629	−0.630	−0.666	−0.667	−0.671
A_1	0.650	0.630	0.594	0.547	0.598	0.538	0.475
A_2	0.383	0.427	0.470	0.512	0.418	0.473	0.515
A_3	0.048	0.136	0.157	0.180	−0.137	−0.184	−0.217
J_{01}	0.10	1.54	4.45	6.98	0.04	0.62	1.46
J_{12}	−1.00	−4.38	−8.14	−13.0	−0.63	−1.90	−3.30
J_{23}	4.40	7.03	10.2	15.8	−1.38	−2.22	−3.13

established by Éliashberg[11] and Migdal[12] to include a real metal.

In the derivation of (7), effects of retardation in the phonon-induced electron-electron interaction were neglected by replacing them with a static expression in the energy range $\sim\omega_D$ near the Fermi level. In a similar way, the usually employed[10] approximation of a piecewise constant energy-transfer function was used for the Coulomb interaction.

An equation similar to (7) has been studied in the single-band case by Pokrovskiĭ.[13] It follows from Eq. (7) that the temperature of the superconducting transition is determined by a formula of the type given by the BCS theory:

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$$T_c = \tilde{\omega} e^{-1/\eta}; \tag{10}$$

Here, in accord with Dynes,[14] we have set $\tilde{\omega} = \omega_D/1.45$ and η is the largest eigenvalue of the set of equations

$$\eta\Delta_i(\mathbf{p}_0) = \sum_j \int \frac{dS_{\mathbf{p}_0'}}{v_j} g_{ij}(\mathbf{p}_0, \mathbf{p}_0')\Delta_j(\mathbf{p}_0'). \tag{11}$$

With neglect of the anisotropies of the energy gaps, the system (7) reduces to the set of linear algebraic equations

$$\Delta_i = \ln\frac{\omega_0\gamma}{\pi T_c}\sum_j g_{ij}\Delta_j, \tag{12}$$

$$g_{ij} = (\lambda_{ij} - \mu_{ij}^*) \Big/ \Big(1 + \sum_l \lambda_{il}\Big) \tag{13}$$

and η in Eq. (10) is now the largest root of the equation

$$\|\eta\delta_{ij} - g_{ij}\| = 0. \tag{14}$$

It must be noted that the set of equations (12) was obtained by Geĭlikman, Zaĭtsev and Kresin[15] without a detailed analysis of the structure of the matrix g_{ij}. In the case of a single-band metal, Eq. (13) goes over into the well-known McMillan formula.[16]

Thus, the problem of the computation of the critical temperature reduces essentially to finding the matrices μ_{ij} and λ_{ij} and then, with the help of Eqs. (13), and (14), the eigenvalue η. So far as the electron-phonon constants λ_{ij} are concerned, they are uniquely determined by the relation

$$\lambda_{ij} = R_{ij}/2\pi T, \tag{15}$$

where the R_{ij} are given by Eq. (2).

The matric μ_{ij}, with account of the approximation of the dielectric constant $\epsilon(k)$ for the Coulomb interaction, can be rewritten in the form

$$\mu_{ij} = \nu_j \int \frac{dO_{\mathbf{p}'}}{4\pi} \sum_{\mathbf{g}} \frac{4\pi e^2 |\langle j\mathbf{p}'|\exp\{i(\mathbf{p}+\mathbf{q})\mathbf{r}\}|i\mathbf{p}\rangle|^2}{|\mathbf{q}+\mathbf{g}|^2 \varepsilon(q+g)}. \tag{16}$$

For calculation of the sum with $\mathbf{g} \neq 0$ ($\mathbf{g}$ are the lattice vectors) in (16), we used the following approximation: we replaced summation over reciprocal lattice vectors of identical length but different direction by averaging over the directions with subsequent multiplication by the number of vectors belonging to the corresponding coordination sphere. Then

$$\sum_{\mathbf{g}\neq 0} = \frac{1}{4\pi}\sum_n c_n \int dO_{\mathbf{g}_n}, \qquad \mu_{ij} = \mu_{ij}^0 + \sum_{n\neq 0} c_n \mu_{ij}^n, \tag{17}$$

where c_n is the number of vectors on the n-th coordination sphere of the reciprocal lattice.

Calculations of the matrices λ_{ij}, μ_{ij}, μ_{ij}^*, g_{ij} and also of the eigenvalues η and the transition temperature have been made on the basis of the formulas presented above for K and Rb at various compressions. Some results of these calculations are shown in the tables.

Table II contains the results of calculations for those compressions in K and Rb for which no electronic rearrangement occurred and the metal remained single-band in character. The compression scale was recalculated into a pressure scale on the basis of data on the equation of state of the metals.[1] There is some error here (in particular in Rb), connected basically with inexact account of the contribution of exchange processes to the pressure. The calculation for $\delta = 1$ was in some sense a test of the validity of the chosen approximation, confirming the absence of superconductivity in K and Rb at normal pressure. The values of μ^* at $\delta = 1$ are close to those computed earlier in the homogeneous electron gas model (0.11).

It is seen from Table II that a considerable increase takes place in λ for K at $\delta = 2$, leading to the possible appearance of superconductivity. This growth, as analysis of intermediate results shows, is due to growth of the matrix element of electron-phonon interaction, owing to the increase in the partial contribution of the d-electrons (the terms with $l = 2$ in the sum (1)). In Rb, as has already been mentioned above, the rearrangement takes place at lower values of the compression and the increase in the matrix element is smaller there than for K. In the language of our model, this is explained by the fact that the radial function f_2 in Rb corresponds to the state 4d and has a single zero, in contrast with K, where the corresponding state is 3d and f_2 has no zeroes at all.

Table III gives the results on the dependence of the transition temperature of K and Rb on pressure for multi-band variants. In particular, as is seen from Table III, superconductivity sets in for Rb at $\delta = 2$.

TABLE II

δ	P, kbar	λ	μ^*	η	T_c, °K
		Potassium			
1	10	0.071	0.114	−0.043	—
1.5	55	0.084	0.109	−0.025	—
2	140	0.270	0.107	0.129	0.1
		Rubidium			
1	8	0.064	0.103	−0.037	—
1.5	50	0.153	0.099	0.047	10^{-12}

TABLE III

δ	P, kbar	η	T_c, °K
	Potassium		
2.5	275	0.280	4.6
3	325	0.352	10.8
5	450	0.358	16
	Rubidium		
2	90	0.229	1.1
2.4	140	0.285	2.9
3	150	0.302	4.1
4	425	0.324	6.1

TABLE IV

δ	λ_{ss}	$\lambda_{d_2d_2}$	μ^*_{ss}	$\mu^*_{d_2d_2}$
	Potassium			
2.5	0.514	0.025	0.106	0.024
3	0.452	0.130	0.098	0.052
5	0.033	0.274	0.014	0.070
	Rubidium			
2	0.364	0.051	0.092	0.034
2.4	0.316	0.122	0.089	0.051
3	0.188	0.175	0.085	0.057
4	0.019	0.217	0.007	0.058

The effect of electron rearrangement on the superconductivity parameters can be traced in the example, shown in Table IV, of the compression dependence of several matrix elements λ_{ij} and μ^*_{ij}, the change in which seems to us to be most characteristic. We note that after overlapping of the bands, λ_{ij} and μ^*_{ij} are 6×6 matrices.

The increase in the element λ_{ss} up to overlapping of the bands is connected basically with the increase in the weight of the d-states in the wave function of the s-band. The decrease in the density of states of the electrons of the s-band with compression, taking place after overlap, and the simultaneous increase of it in the d-band, lead to a decrease in λ_{ss} and an increase in $\lambda_{d_2d_2}$. The character of the change in the elements of the matrix μ^*_{ij} is explained by basically the same reasons.

Direct comparison of the calculated data with experiment is impossible because of the absence of experimental data. However, the presence of a connection between superconductivity and the electrical resistance, given by Eq. (15), and the good agreement of the results of the preceding section with experiment, indicates the reasonableness of the approximations used.

Of course, because of the strong exponential dependence of the coupling constant, the transition temperature is much more sensitive to the choice of model than the resistivity, and therefore it is difficult to expect very close agreement of the calculated absolute values of T_c with the experimental values. However, the choice of the model used in our calculations seems to be justified to a sufficient degree for the assessment of the possibility of the superconducting transition and the determination of the qualitative dependence of T_c on the pressure.

5. CONCLUSION

All the foregoing analysis has been based on the supposition of the decisive role of rearrangement of the electron structure in K and Rb under pressure in the phenomena investigated by us. The analysis of the results completely confirm this supposition. The compression dependence of the resistivity calculated by us agrees well with experiment. On the basis of calculations of the superconducting parameters, the appearance of superconductivity has been predicted for K at $P \sim 150$ kbar and for Rb at $P \sim 100$ bar. This gives us hope for accomplishments of the next step proposed by us in the investigation of the appearance of superconductivity in Cs, Y and Ba, where this phenomenon has been discovered experimentally, and it is evidently due to the same cause.

In conclusion, we express our gratitude to A. I. Voropinov and V. G. Podval'nyĭ for preparing the programs of calculation of the equation of state and the results of the calculation of the exchange pressure in K. The authors also sincerely thank Professor B. T. Geĭlikman, Academician L. F. Vereshchagin, and E. S. Itskevich for useful discussions of the results of the present research.

[1)]Here and below, we set $\hbar = k = 1$ (k = Boltzmann's constant) and consider a crystal of unit volume.

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