Polaronic band narrowing and the magnetic susceptibility of vanadium compounds with the A-15 structure

A. S. Aleksandrov, V. F. Elesin, and V. V. Kabanov

Engineering Physics Institute, Moscow
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It is shown that at certain values of the corresponding coupling constant g^2, the strong electron-phonon interaction in narrow-band compounds with the A-15 structure lifts the overlap of the d subbands of different symmetry and leads to a substantial increase in the height of the peaks in the density of states. The band structure and density of states of V_3Si and V_3Ga are calculated with allowance for the polaron effect. It is shown that the strong electron-phonon interaction in V_3Si is responsible for its large magnetic susceptibility $\chi(T)$, which depends on g^2 and on the phonon spectrum. Comparison with the experimental results on $\chi(T)$ indicates that the polaron model of the electron spectrum[4] is valid for A-15 vanadium compounds.

The familiar explanation of the anomalous properties of high-temperature superconductors consisting of intermetallic compounds with the A-15 structure is based on the one-electron Hartee-Fock approximation, which neglects the electron-phonon interaction and Coulomb correlations. As was pointed out by Gor'kov,[1] the necessary, but scarcely justified, neglect of interaction effects makes it impossible to fix the choice of band shape in the one-electron approximation by other than phenomenological means.

Based on the results of the adiabatic theory of the electron-phonon interaction, the view has been expressed[2,3] that the strong electron-phonon interaction can smear out the fine structure of the one-electron density of states.

As we have earlier pointed out,[4] however, in A-15 intermetallides the electron-phonon interaction can be so strong, and the initial d subbands so narrow, that the usual treatment of this interaction on the basis of a small parameter $(m/M)^{1/4}$ might not apply. Here, if the existence condition for a small polaron is met,[4] the electron-phonon interaction does not smear out the fine structure but leads to a localization of the electrons, i.e., to a narrowing of the initial d band by a factor of $\exp(g^2)$ and, accordingly, to a heightening of the peaks in the density of states. Thus, owing to the polaron effect the strong electron-phonon interaction can alone lead to the existence of a fine structure in the density of states with an energy scale of several hundred kelvins. We have noted[4] that quantitative comparison of the polaron model with experiment requires calculation of the energy dependence of the polaronic density of states $N_p(\varepsilon)$.

The electronic density of states of A-15 compounds in the one-electron approximation have been calculated in a number of papers[5] by the pseudopotential and tight-binding methods. At the present time the energy resolution in such calculations is 10^{-3} Ry, which, in principle, permits resolution of the narrow peaks in $N(\varepsilon)$ due to the flat parts of the Fermi surface. However, the fraction of states in such peaks (i.e., the height of the peaks) is insufficient to explain the experimentally observed values of χ, λ, and several other parameters. This was demonstrated in Ref. 6, which implies that the discrepancy between the experimental values of λ and the theoretical values obtained from standard band calculations and the McMillan formula reaches 300% in vanadium compounds, while the accuracy of the band calculations is no worse than 20%.

In the present paper it is shown that band calculations give a value for the spin susceptibility of V_3Si which is lower than the experimental value by a factor of six or seven, and that allowance for the polaron effect eliminates this discrepancy. The polaron model, which takes into account the strong electron-phonon interaction in A-15 compounds, can thus serve as a realistic basis for explaining the anomalous properties of the intermetallides and, possibly, of other high-temperature superconductors.

1. ELECTRON SPECTRUM AND DENSITY OF STATES OF A-15 COMPOUNDS WITH ALLOWANCE FOR THE POLARON EFFECT

For calculating $N_p(\varepsilon)$ we used a simplified version of the tight-binding approximation of Weger and Goldberg.[2] The simplification lies in the neglect of hybridization both between d states and s, p states and between the various d orbitals forming the irreducible representations of the symmetry group A-15. This is possible because the necessary width of the peaks is achieved by taking the polaron factor into account (by proper choice of g^2), eliminating the need for a high degree of accuracy in the numerical calculations, as improvement here would only lead to insignificant changes in the hyperfine structure of $N_p(\varepsilon)$ within the narrow peak.

The unit cell of V_3Si (V_3Ga) contains two atoms of silicon (Ga) and six atoms of vanadium. The symmetry of the crystalline field in which the V atom is located shows that the states π_1 and π_2 (magnetic quantum number $m = \pm 1$) form the basis of a two-dimensional irreducible representation. The remaining states $\sigma(m = 0)$ and δ_1 and δ_2 $(m = \pm 2)$ belong to different one-dimensional representations and are nondegenerate.

The wave function in the tight-binding approximation is constructed, following Weger and Goldberg,[2] as a superposition of bonding and antibonding states:

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$$\sigma_x(\mathbf{k})=\sum_{\mathbf{R}}\exp(i\mathbf{kR})\frac{1}{\sqrt{2}}[\sigma_x(\mathbf{r}-\mathbf{R}+\tau_x)+\sigma_x(\mathbf{r}-\mathbf{R}-\tau_x)] \quad (1)$$

for the bonding orbital, and

$$\sigma_x^*(\mathbf{k})=i\sum_{\mathbf{R}}\exp(i\mathbf{kR})\frac{1}{\sqrt{2}}[\sigma_x(\mathbf{r}-\mathbf{R}+\tau_x)-\sigma_x(\mathbf{r}-\mathbf{R}-\tau_x)] \quad (2)$$

for the antibonding orbital, where $\mathbf{R}=a(l\mathbf{i}+m\mathbf{j}+(n+\frac{1}{2})\mathbf{k})$, a is the lattice parameter, $\tau_x=\frac{1}{4}a\mathbf{i}$, and (l,m,n) is a set of whole numbers. The spectrum is determined with allowance for the two-center and three-center overlap integrals by diagonalizing the 30×30 energy matrix. Because all the matrix elements coupling orbitals of different symmetry have been dropped, and the 30×30 matrix decomposes into three 6×6 matrices, which determine the spectrum of the σ, δ_1, and δ_2 subbands, and one 12×12 matrix, for the π subband.

In the numerical calculation we used for the effective overlap integrals the values which Weger and Goldberg obtained for V_3Ga and V_3Si by interpolation from the direct calculations of Mattheiss.[7] According to Ref. 4, the existence condition for small-radius polarons is satisfied in A-15 compounds, and consequently the overlap integrals J are renormalized by the strong electron-phonon interaction:

$$\mathcal{J}=J\exp(-g^2), \quad (3)$$

where

$$g^2=\frac{1}{2N}\sum_{\mathbf{q},\lambda}\frac{\mathrm{cth}(\omega_{\mathbf{q}\lambda}/2T)}{2\omega_{\mathbf{q}\lambda}^2}\{|\Gamma_\alpha^\lambda(\mathbf{q})|^2+|\Gamma_{\alpha'}^\lambda(\mathbf{q})|^2-2\,\mathrm{Re}\,\Gamma_\alpha^\lambda(\mathbf{q})\Gamma_{\alpha'}^\lambda(\mathbf{q})^*\exp[i\mathbf{q}(\mathbf{m}-\mathbf{m}')]\},$$

$\omega_{\mathbf{q}\lambda}$ is the frequency of vibrational mode $(\mathbf{q},\lambda)$, and $\Gamma_\alpha^\lambda(\mathbf{q})$ is the matrix element for the interaction of a d electron with atom α of unit cell $\mathbf{m}$.

To simplify the calculations the factor g^2 was chosen the same for all the overlap integrals governing the spectrum of the given subband. The density of polaron states

$$N_p(\varepsilon)=\frac{1}{(2\pi)^3}\int d^3k\,\delta(\varepsilon-\varepsilon(\mathbf{k})) \quad (4)$$

was calculated by the Monte Carlo method; the width of the band was divided into 100 equal parts and the density-of-states integral was calculated over each of the parts:

$$N_p(\varepsilon)\approx\frac{1}{\Delta\varepsilon}\int_{\varepsilon}^{\varepsilon+\Delta\varepsilon}N_p(\xi)\,d\xi=\frac{1}{\Delta\varepsilon}\int_{\varepsilon(\mathbf{k})}\frac{d^3k}{(2\pi)^3},\quad \varepsilon<\varepsilon(\mathbf{k})<\varepsilon+\Delta\varepsilon. \quad (5)$$

The accuracy of our simplified method is illustrated by Fig. 1a, which shows the density of states for the δ_2 band of V_3Ga without allowance for the polaronic narrowing ($g^2=0$) in comparison with the Weger–Goldberg result.

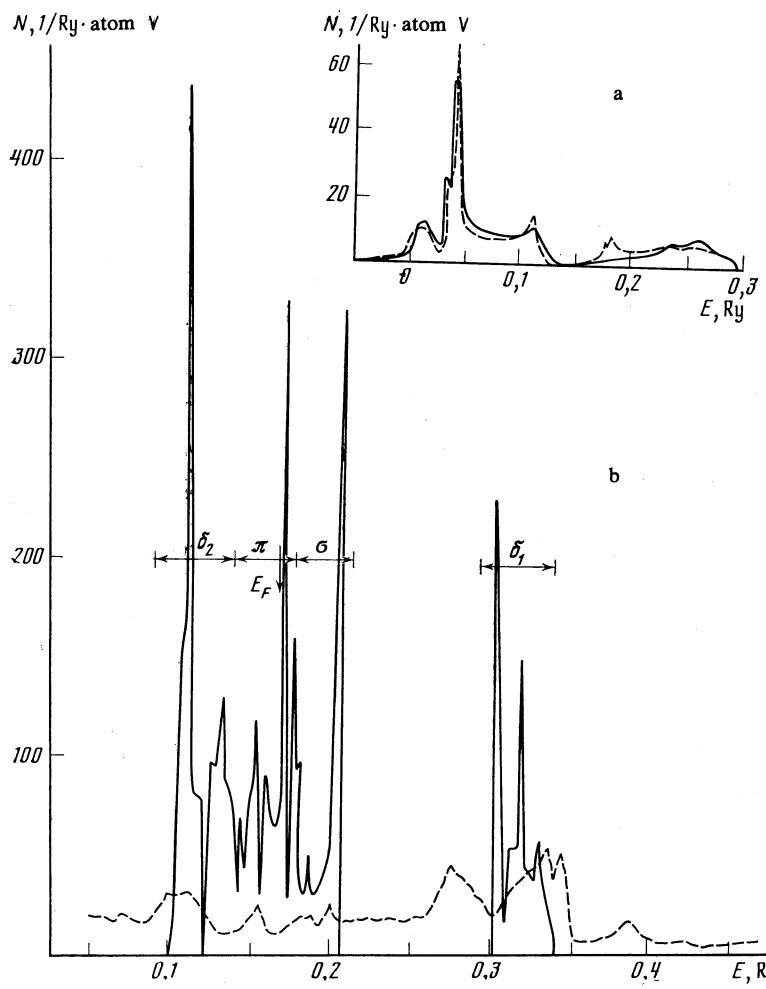

FIG. 1. a) Density of states of the δ_2 subband of V_3Ga in comparison with the calculation of Weger and Goldberg (dashed curve). b) Density of states of the d band of V_3Ga (the dashed curve was calculated without allowance for the polaron effect,[2] while the solid curve was calculated with the polaron effect taken into account for $g^2=2.1$). The arrow indicates the position of the Fermi level.

Our calculation shows that the polaron effect leads to a substantial renormalization of the density of states of the entire d band: to a sharp decrease in the width of the individual subbands and to an increase in the density of states at the peaks. Figure 1b shows the polaronic density of states of V_3Ga in comparison with the Weger–Goldberg result.[2] Because the one-center integrals (the crystalline field), which determine the position of the band center, are not renormalized by the polaron effect, incorporation of the electron-phonon interaction eliminates the overlap of the d subbands even for $g^2 \gtrsim 2$.

Another important circumstance is the substantial temperature dependence of the polaron spectrum and of $N_p(\varepsilon)$, on a scale of the order of the characteristic vibrational frequency [see Eq. (3)]. This circumstance, together with the migration of the chemical potential within the narrow degenerate polaronic band upon changes in temperature, can explain the marked spatial redistribution of electrons that has been observed[8] in V_3Si in the temperature range 300–15 K.[1)]

The position of the Fermi level is easily determined for $g^2 \gtrsim 2$, in which case there is no overlap of the individual subbands. A vanadium atom has five valence electrons. According to Ref. 8, there is a transfer of charge from the silicon atoms to the vanadium atoms (1.8–2.4 electrons per Si atom). Thus, each vanadium atom has 5.6–5.8 electrons. Since two of the electrons of each vanadium atom occupy the lowest subband δ_2, one expects that the Fermi level lies in the immediate vicinity of the upper boundary of the π subband, in agreement with Ref. 9.

2. MAGNETIC SUSCEPTIBILITY, COMPARISON WITH EXPERIMENT

In the adiabatic approximation[10] the electron-phonon interaction leads to a small correction of order $(m/M)^{1/2}$ (m is the electron mass, M is the mass of the atom) to the magnetic susceptibility $\chi(T)$. Under conditions of polaronic narrowing of the bands the situation is directly opposite: the electron-phonon interaction leads to a strong localization of the spins and, consequently, to a substantial increase in $\chi(T)$. Thus, as we have already pointed out,[4] allowance for the polaron effect can eliminate the discrepancy between the theoretical and experimental values of χ and λ.

We assume that the polaron-polaron correlations in A-15 are rather small ($T_c \ll \tilde{J}$). In this case one can calculate $\chi(T)$ using the one-polaron approximation, in which the polaronic spin susceptibility is of the form

$$\chi_p(T) = 6\frac{\mu_B^2}{T}\int d\varepsilon N_p(\varepsilon)\exp\left(\frac{\varepsilon-\mu}{T}\right)\left[\exp\left(\frac{\varepsilon-\mu}{T}\right)+1\right]^{-2}, \tag{6}$$

where the change with temperature of the chemical potential $\mu(T)$ is described by the equation

$$n_e = \int d\varepsilon N_p(\varepsilon)\left[\exp\left(\frac{\varepsilon-\mu}{T}\right)+1\right]^{-1}. \tag{7}$$

Here μ_B is the Bohr magneton, and n_e is the number of electrons per vanadium atom.

Because the height of the peaks increases by a factor of $\exp(g^2)$, $\chi_p(T)$ should be noticeably larger than the electronic spin susceptibility $\chi_e(T)$. The shape of $\chi_p(T)$ depends on the position of the Fermi level $\mu(0)$, the value of g^2, and the phonon spectrum (3). Assuming that the main contribution to g^2 is due to optical modes (corresponding to oscillations of the silicon atoms with respect to the vanadium) at a frequency $\omega_0 \approx 800$ K (determined from the experimental[11] phonon density spectrum), we obtain

$$g^2(T) = g_0^2 \operatorname{cth}(\omega_0/2T), \tag{8}$$

where g_0^2 is independent of temperature.

The position of $\mu(0)$ can be found from the experimental value of the charge transfer[8] from Si to V with the aid of expression (7). The functions $\chi_p(T)$ obtained from (6) with allowance for (7) and (8) is shown in Fig. 2 for various values of g_0^2. For certain values of g_0^2 and sufficiently small ω_0 the function $\chi_p(T)$ can be nonmonotonic (curve 1) on account of the temperature dependence of the width W of the polaronic band [see Eqs. (3) and (8)]. In this case the growth of the density of states at low temperatures as a result of the decrease of W leads at first to an increase in $\chi_p(T)$ with increasing temperature. In the high-temperature region, $T \gtrsim W$, the function $\chi_p(T)$ is described by the Curie law,

$$\chi_p(T) \propto 1/T, \tag{9}$$

on account of electron localization due to the polaron effect.

Figure 2 also shows the spin susceptibility $\chi_e(T)$ calculated without allowance for the polaron effect (curve 5). To rule out inaccuracies due to the insufficient energy resolution in the Weger–Goldberg method, in calculating $\chi_e(T)$ we took $N(\varepsilon)$ and $\mu(0)$ from the recent numerical calculations of Jarlborg *et al.*,[12] in which the energy resolution is no worse than 10^{-3} Ry. As is seen from comparison of the curves in Fig. 2, the polaronic spin susceptibility is several times larger than the electronic spin susceptibility at low temperatures, while at high temperatures it is smaller.

Figure 3 shows a comparison with experiment for V_3Si. Correct determination of the temperature-independent component χ_0 of the total susceptibility $\chi(T) = \chi_0 + \chi_p(T)$ requires measurements over a wide temperature interval $\Delta T \sim 1000$ K. Such measurements have recently been

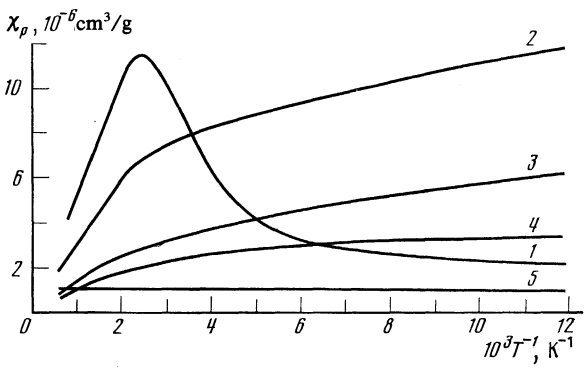

FIG. 2. Polaronic spin susceptibility of V_3Si: 1) $g_0^2 = 2$, $\mu(0) = 17$ mRy, $\omega_0 = 400$ K; 2) $g_0^2 = 3.2$, $\mu(0) = 8.2$ mRy, $\omega_0 = 840$ K; 3) $g_0^2 = 2.1$, $\mu(0) = 27$ mRy, $\omega_0 = 840$ K; 4) $g_0^2 = 1.43$, $\mu(0) = 49$ mRy, $\omega_0 = 840$ K; 5) calculation without allowance for the polaron effect.

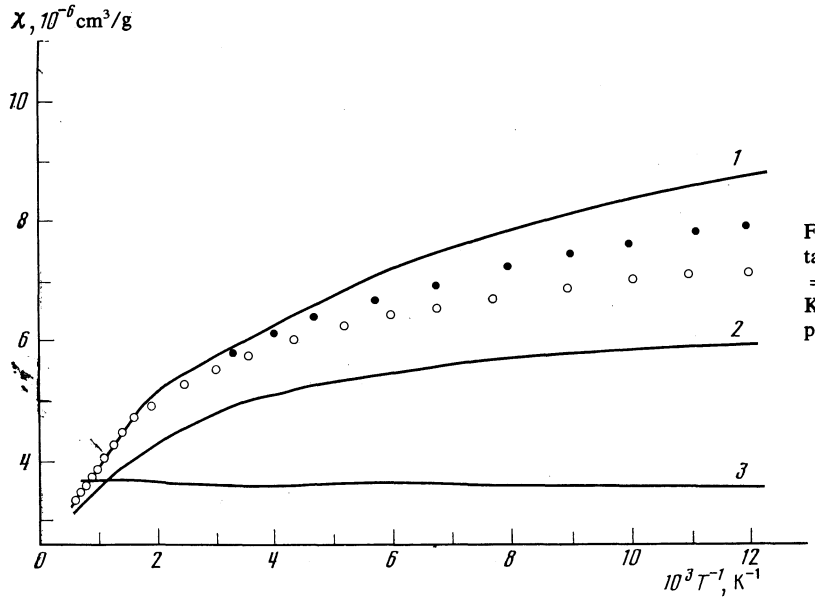

FIG. 3. Total magnetic susceptibility of V_3Si: ○) experimental data of Ref. 13, ●) experimental data of Ref. 14, 1) $g_0^2 = 2.1$, $\omega_0 = 840$ K, $\mu(0) = 27$ mRy; 2) $g_0^2 = 1.43$, $\omega_0 = 840$ K, $\mu(0) = 49$ mRy; 3) calculation without allowance for the polaron effect.

made[13] (see Fig. 3). These measurements confirm the behavior (9) at $T \gtrsim 600$ K and give a value

$$\chi_0 \doteq 2.6 \cdot 10^{-6} \text{ cm}^3/\text{g} \qquad (10)$$

which is appreciably lower than earlier estimates of χ_0 obtained by extrapolation of the $\chi(T)$ curve for $T < 600$ K. As is seen in Fig. 3, the discrepancy with experiment of the one-electron band calculations is almost 300% at low temperatures, while allowance for the polaronic narrowing of the d band permits description of the experimental curve. The best agreement is obtained at the following values of the parameters:

$$g_0^2 = 2.1, \quad \omega_0 = 840 \text{ K}, \quad \mu(0) = 0.027 \text{ Ry}. \qquad (11)$$

The frequency value $\omega_0 = 840$ K supports the correctness of the estimate[11] of the frequency of the optical vibrations of the silicon ions relative to the vanadium and agrees with the spectral density extracted from tunneling experiments.

The value of the chemical potential $\mu(0)$ given in (11) corresponds to an almost filled, doubly degenerate π band (see Fig. 1b). Such a position of $\mu(0)$ agrees with the results of NMR experiments[9] in V_3Si, where it was shown that the symmetry of the d-electron wave functions at the Fermi level corresponds to π states. In addition, as was noted above, this agrees with the stoichiometric composition of V_3Si and is consistent with the experimental data on the charge transfer.[8]

It should be noted that in the low-temperature region ($T < 100$ K) the theoretical curve gives too large a value of $\chi(T)$. On the one hand, this may be due to the influence of polaron-polaron correlations, which are especially pronounced as one approaches the transition to the superconducting state. In superconducting A-15 compounds the polaron-polaron attraction leads to a virtual (for $T > T_c$) pairing of opposite spins and a decrease in χ. On the other hand, as can be seen from Fig. 3, which shows the experimental data for various samples,[14] the value of $\chi(T)$ in this temperature region depends on the residual resistance of the sample. It has been noted[15] that defects in the A-15 structure smear the peaks of $N(\varepsilon)$ and decrease χ. Thus, the nonideality of the investigated V_3Si crystals is also responsible for some discrepancy in the low-temperature region. Furthermore, for $T < 100$ K the hyperfine structure of $N_p(\varepsilon)$ due to hybridization of the various orbitals (this was not considered in our calculation) and the residual polaron-polaron interaction (see below) can be important.

We might also note that the fine structure of the density of states in vanadium compounds and other A-15 structures can be studied by irradiation,[15] investigation of the influence on χ of the stoichiometric composition,[16] and other experiments.

CONCLUSION

We have shown in this paper that the strong electron-phonon interaction lifts the overlap of the d subbands of different symmetry and sharply increases the height of the peaks in the density of states and the spin magnetic susceptibility. The Coulomb repulsion also leads to an increase in χ.[17] As was pointed out by Gor'kov,[1] the two renormalization effects are independent, and so the effective state densities determined from the specific heat C and the susceptibility do not have to coincide (if χ is renormalized by the Coulomb repulsion). At the same time, the renormalized state densities determined experimentally from χ and C are quite similar in spite of the large renormalization. This indicates that the nature of the renormalization of χ and C is the same, as is predicted by the polaron model.[4] In addition, as we noted above, the interaction between carriers in A-15 compounds at attractive, not repulsive, and can therefore only decrease χ.

The value we have found for the electron-phonon coupling constant in V_3Si, $g^2 \approx 2$, corresponds to a polaron shift $E_p \sim g^2\omega_0 \approx 15$ mRy, which is sufficient to satisfy the existence condition $2E_p \gtrsim J$ for the small polaron in the δ_1 and δ_2

subbands. However, the overlap integrals for nearest neighbors in the σ and π subbands turns out to be larger than $2E_p$. Therefore, these subbands most likely form a polaron of intermediate radius, with an effective mass smaller than the mass of a small polaron[18] and with a corresponding renormalized band which is wider on account of the residual electron-phonon interaction, which is not taken into account in the small-polaron model. Allowance for this interaction clearly leads to a decrease in the theoretical value of χ, which would require larger values of g^2 for comparison with experiment. Thus the value $g^2 \approx 2$ should be regarded as a lower estimate. The behavior of χ as a function of temperature and the large value of $\chi(T)$ obtained with allowance for the polaron effect (see Fig. 2) are also characteristic of other classes of high-temperature superconductors (e.g., the Laves and Chevrel phases), indicating that the polaron effect may play a substantial role in them as well.

The polaronic density of states averaged over an energy interval of the order of the width of the polaron band $2W \sim 2z\tilde{J}$ turns out to be very high in polaronic superconductors:

$$\bar{N}_p \approx 1/2W \geqslant 10 \text{ states eV·atom·spin}, \quad (12)$$

where z is the number of neighboring transition-metal ions to which hops take place ($2 < z < 10$). How can this circumstance can be reconciled with the relatively low value of T_c, and what is the mechanism which limits T_c? As we mentioned earlier,[4] for a polaronic superconductor in the case of weak coupling of the polarons, T_c is determined by the ratio of the sum of the intercenter zv_1 and contact v_0 interactions to the width $2W$ [see Eq. (19) of Ref. 4]:

$$T_c < W \exp(-1/\lambda^*), \quad \lambda^* = -(zv_1+v_0)/2W. \quad (13)$$

The contact interaction consists of the positive Hubbard repulsion U and the negative polaron shift $-E_p$, while the intercenter interaction v_1 is approximately equal to $-E_p$ (the corresponding estimate is given in Ref. 19). The upper boundary of T_c is thus determined by the ratio

$$\lambda^* \approx [(z+1)E_p - U]/2W. \quad (14)$$

According to the estimate obtained for E_p in the present paper, the first term in the numerator of (14) is of the order of 1 eV. As was recently pointed out,[20] the ionization potential in vacuum for ions of the transition metals which form superconducting compounds is extremely low: ≈ 6 eV for V and Nb. According to Hubbard,[21] screening effects in a crystal decrease this potential by several times; thus we have $U \sim 1$ eV in formula (14) and, hence, $\lambda^* \approx 1$–5 in A–15 compounds.

An increase in the electron-phonon coupling constant g^2 leads to an increase in the value of the polaron shift and a simultaneous sharp decrease in the width of the polaronic band. According to (13) and (14), this would seemingly cause T_c to increase all the way to $T_c \sim W$. In the limit $\lambda^* \gg 1$, however, as was established in Refs. 19, 22, and 23, small polarons form spatially separated small-radius bipolarons having a superconductivity similar to the superfluidity of a Bose liquid. In this limit T_c falls off with increasing g^2 as a result of the growth of the effective mass $m^* \sim \exp(2g^2)$ of the small bipolaron[23] (for an ideal Bose gas the condensation temperature is $T_c \sim 1/m^*$).

It can be supposed that compounds with record high values of T_c occupy an intermediate position between polaronic superconductors with a BCS-type superconductivity and bipolaronic superconductors with the superconductivity of a charged Bose gas.[22,23] Certain A-15 intermetallides can be found on the "bipolaron" part of the $T_c(\lambda)$ curve (see Fig. 5 of Ref. 19) and can display the specific features of a bipolaronic superconductor, among these being the existence of bound pairs (small bipolarons) at temperatures above T_c. In this regard we mention the feature observed[24] on the voltage-current characteristics of V_3Si tunneling contacts at sufficiently high bias voltages (~ 0.04 V). This feature persisted at $T > T_c$ and was interpreted as the breakup of a small bipolaron by the external electric field. Meanwhile, the quantitative calculation of T_c for intermediate electron-phonon and polaron-polaron coupling remains an unsolved problem.

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[1)]We are indebted to L. P. Gor'kov for calling our attention to the experiment of Ref. 8.

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