Alteration of the superconducting properties of vanadium by introduction of tantalum impurity atoms

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The superconducting transition temperature T_c, the temperature dependence of the specific heat in the 1.5—40°K range in the absence of a field or in a field ~18 kOe, and the electrical conductivity between 5 and 1000°K are measured for dilute solutions of Ta in V. The experimental data are compared with calculations based on the MacMillan[3,4] and Maksimov[10] theory. It is shown that decrease of T_c is primarily due to weakening of electron-phonon interaction with increasing Ta content, despite the fact that the growth is accomplished by a softening of the crystal phonon spectrum in the presence of the impurity, due to the appearance of quasilocal oscillations. The variation of T_c can also be explained qualitatively on the basis of data pertaining to variation of the phonon spectrum of an impurity-containing crystal and variation of the amplitude for scattering of electrons by ions. A correlation is observed between variation of the electric resistance of alloys at high temperatures $T \geq 2\Theta$ and variation of electron-phonon interaction.

1. INTRODUCTION

The question of the influence exerted on the superconducting transition temperature T_c of a metal by atoms of a nonmagnetic impurity with mass m′, which differs greatly from the mass m of the atoms of the host metal, when local ($m' \ll m$) or quasilocal ($m' \gg m$) phonon states appear in the phonon spectrum, is one of the interesting but little investigated problems of superconductivity physics. Since in the general case introduction of impurity atoms with $m' \neq m$ into a metal alters not only the phonon spectrum but also the density of the electronic states on the Fermi surface and the Coulomb repulsion potential, an experimental study of the influence of heavy or light impurity atoms on T_c calls for the use of methods that make it possible to determine the variation of both the phonon and the electron spectra. Such methods have been made possible by the development of tunnel spectroscopy with superconductors as well as of inelastic scattering of neutrons in conjunction with measurements of the temperature dependence of the specific heat and of the electric conductivity.

The influence of local and quasilocal oscillations in the phonon spectrum on T_c of metals with impurities was considered theoretically by a number of workers[1,2]. In practice, however, the analysis of the connection between the properties of the metal in the normal state and T_c is based, both for pure metals and for metals with impurities or alloys, on the MacMillan equation[3] or its modification[4]. The theoretical papers[1-4] are based on approximate solutions of the integral equations obtained by Eliashberg[5] for the energy-gap function, and are rigorously valid for regular crystals. In crystals containing many impurities and in disordered compounds, the phonon and electron systems can undergo appreciable changes in comparison with a regular crystal. Obviously, the theoretical concepts applied to such disordered systems should take into consideration these changes and take into account the dynamic character of the defects. Until recently, only static defects were considered in the theory of superconductivity, as for example in the paper by Markowitz and Kadanoff[6].

A new approach to the determination of the critical temperature, applicable for a non-ideal crystal, can be developed by using the Van-Hove correlation-function formalism to describe the interaction in an electron-ion system[7]. It is known that such an approach is fruitful in the investigation of the thermodynamic and kinetic properties of non-ideal crystals[8,9]. On the basis of such an approach, Maksimov[10] redetermined the electron-phonon interaction constant λ expressed it in terms of the correlation function of the crystal, and showed that an isotopic impurity does not influence the value of λ of an isotropic superconductor, although it does deform the phonon spectrum, and that the change of T_c can be approximately expressed in terms of the change of the high-temperature resistance of the metal with impurity in comparison with the resistance of the initial metal.

The present paper is devoted to a study of the superconducting properties of dilute disordered solutions of Ta in V and the concentration region Ta < 6 at.%, and to the correlation of the concentration dependence of T_c of these solutions as a function of the phonon spectrum and of the electric conductivity. The investigated range of concentrations of Ta corresponds to the region of noninteracting impurities. The changes in the phonon spectrum of V following introduction of Ta is quantitatively determined from data on inelastic neutron scattering and on the temperature dependence of the specific heat of V and of its alloys with Ta at low temperatures[11,12].

The concentration dependence of T_c in the V – Ta system at a Ta content ≥ 10 at.% was investigated in[13]. It was established that the electronic specific heat coefficient γ and the characteristic temperature Θ decrease monotonically with increasing Ta concentration, and that T_c has a minimum at ~ 50 at.% Ta. Such variations in the transition temperature could not be due to changes in the density of the electronic states and in the phonon spectrum. The explanation offered by the authors was tantamount to a general statement that in the case of V – Ta alloys the changes in the interaction responsible for the superconductivity lead to changes of T_c. At low concentrations of Ta in V, when the phonon spectrum of V is deformed by quasi-local vibrations of the impurity Ta atoms and the neighboring V atoms perturbed by the impurity[12], the concentration dependence of T_c was not investigated. The use of interpolated data on T_c, Θ, and γ is not justified in this concentration region.

2. EXPERIMENTAL PART

The specific heat of pure V and of its alloys with Ta ($V_{99.18}Ta_{0.82}$, $V_{96.45}Ta_{3.55}$, $V_{94.32}Ta_{5.68}$) were measured in an adiabatic calorimetric setup in which measurements could be made in the range 1.2—40°K and in magnetic fields up to 18 kOe. The magnetic field was produced with the aid of a superconducting solenoid. The homogeneity of the field along the sample was $\sim 10^{-4}$. The use of germanium and platinum resistance thermometers, of a null method based on the R-308 potentiometer, and of a reliably operating adiabatic calorimeter has resulted in sufficiently high accuracy in the measurements of the specific heat. The error in the determination of the molar specific heat is estimated by us at 0.5% in the ranges 1.2—5 and 20—40°K and at 2% in the interval 5—20°K. The apparatus, the experimental procedure, and the sample preparation were described in detail in an earlier paper[12].

The electric conductivity of the investigated samples in the range 4.2—300°K was measured in an instrument used to obtain intermediate temperatures. It consisted of a massive copper block with the samples and thermometers. Liquid helium was poured into the block through a valve from a dewar. The copper block was placed in a vacuum jacket with a carbon sorption pump. The good vacuum made it possible to "break away" from the temperature of the external helium bath and to perform the measurements both in the region below 4.2°K and in the intermediate temperature region. The copper-block temperature was raised with a heater of constantan wire with resistance ~ 200 Ω. The temperature was maintained during the course of the measurements, depending on the temperature interval, accurate to 0.001—0.01°K with a temperature stabilizer that regulated the flow of current to the heater, and an (Au-Fe)-chromel thermocouple with sensitivity 10 μV/deg. The temperature was measured with germanium and platinum resistance thermometers. The resistances of the samples and of the thermometers were measured by a standard null method using an R-308 potentiometer.

To determine the temperature of the superconducting transition from the resistance we used a PDS-021M automatic x-y recorder. The voltage from the sample was fed through an F-18 photoamplifier to the Y input of the recorder, and the voltage from the thermometer was fed through the R-308 potentiometer to the X input. When the copper block was heated slowly, the recorder plotted the superconducting-transition curve. The block temperature could be maintained within 5—10% of the transition width. We measured simultaneously the thermometer resistance. When the copper block was cooled sufficiently slowly, the superconducting-transition curve was reproduced practically fully. The results were reduced graphically and the accuracy with which T_c was determined was ~ 0.02°K.

The high-temperature measurements of the electric conductivity at 300—1000°K were performed in a retort furnace with a hermetic quartz chamber through which helium evaporated from the dewar was pumped. The samples were rods 60 mm long with cross section 0.7 × 0.7 mm. Current and potential leads of nichrome of 0.2 mm diameter were welded to the samples. The leads were insulated with quartz capillaries. The samples were placed in individual quartz test tubes. The test tubes with the samples, together with the hot junction of the thermocouple placed between them, were located in the sealed end of the quartz tube. To eliminate temperature gradients at the samples, the exit port for the evaporating helium was 7 cm below the samples.

The sample temperature was measured with a chromel-alumel thermocouple whose cold junction was placed in melting ice. The resistance of the investigated samples and the thermal emf of the thermocouple were measured by a standard null method.

3. MEASUREMENTS RESULTS AND DISCUSSION

The measured temperature dependence of the specific heat C of the initial V and of its dilute alloys with Ta in a magnetic field ~ 18 kOe and without a field, in the range 1.2—8°K, are plotted in Fig. 1 in the coordinates C/T and T^2. The measurements in the field made it possible to determine the coefficient of the electronic specific heat γ and the Debye temperature Θ of the investigated alloys, while the measurements without the field yielded the superconducting transition temperature T_c, the jump $\Delta C/\gamma T_c$ of the specific heat at the transition, and the ratio of the electronic specific heats of the superconducting and normal states at T_c. These data are summarized in Table I, where the values of T_c determined from the specific heat and from the electric conductivity are also compared. The values of T_c obtained by these methods are in good agreement. The relatively small width of the transition to the superconducting state, T = 0.2—0.3°K, indicates that the samples were sufficiently homogeneous. Table I lists also the values of the mean free path l and of the coherence length ξ_0, calculated for V and its alloys with Ta from the experimental data on the electric resistance and the thermal conductivity. The condition $l < \xi_0$ is satisfied for all the samples. The initial V can therefore be regarded as an isotropic superconductor.

The change of the phonon spectrum of V following the introduction of the Ta was determined by measuring the inelastic neutron scattering by V[11] and by measuring the low-temperature specific heat of V and of its dilute solutions with Ta[12], using the theoretical concepts

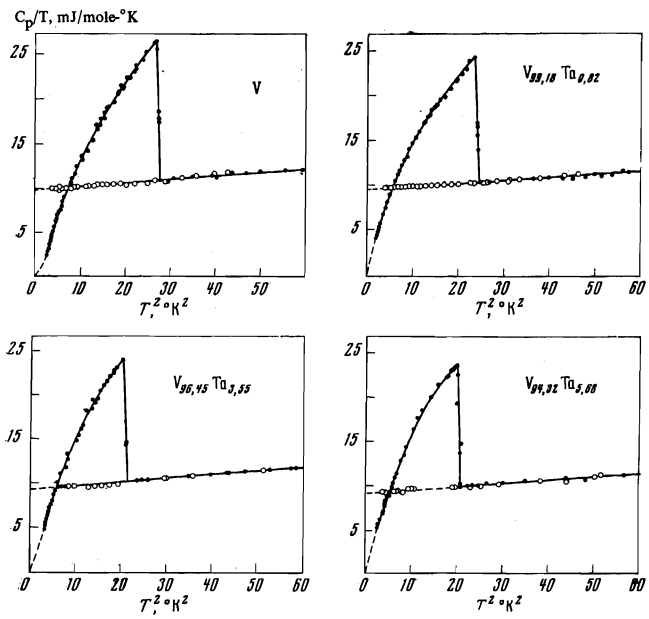

FIG. 1. Specific heat of pure V and of its alloys with Ta without a field and in a field ~18 kOe.

TABLE I

Composition	$\rho(300^\circ K)/\rho(5.2^\circ K)$	a, Å	T_c, °K: From specific heat	T_c, °K: From electric conductivity	mJ/mole-deg²	Θ, °K	$\frac{\Delta C}{\gamma T_c}$	$\left(\frac{C_{es}}{C_{en}}\right)_{T_c}$	l, Å	ξ_0, Å
V	24	3.029	5.24	5.21	9.80	373	1.40	2.52	315	470
$V_{99.18}Ta_{0.82}$	9.6	3.031	4.90	4.95	9.50	370	1.41	2.49	139	523
$V_{96.45}Ta_{3.55}$	5.6	3.037	4.55	4.57	9.25	359	1.42	2.48	68	581
$V_{94.32}Ta_{5.68}$	4.8	3.049	4.46	4.46	9.16	350	1.43	2.47	63	599

from [9]. Figure 2 shows the phonon spectrum $g_0(\omega/\omega_0)$ of the initial V, where ω_0 is the end-point frequency of the phonon spectrum in temperature units, equal to 460°K, and the phonon-spectrum deformation function $\Delta g(\omega/\omega_0)$, which is connected with the introduction of the Ta impurity into the V. The function $\Delta g(\omega/\omega_0)$ takes into account both the difference between the masses of the matrix and impurity atoms, and the change in the force constants of the interaction of the impurity atoms with the matrix atoms in comparison with the interaction in the initial matrix. In the investigated Ta concentration region, where the approximation linear in the concentration is valid, the phonon spectrum of the V-Ta alloy is a superposition of the functions $g_0(\omega/\omega_0)$ and $\Delta g(\omega/\omega_0)$ with allowance for the composition of the alloy. The phonon spectrum of the initial V, which is shown in Fig. 2, differs from the spectrum determined earlier [11] in that a quadratic law with an equivalent Debye temperature 370°K, determined experimentally in [12], was used in the low-frequency region instead of the 337°K used in [11]. In addition, the spectrum was normalized to unity by taking into account the factor $\omega^{-1/2}$, which was omitted in [11].

In MacMillan's theory [3,4], a relation is established between the properties of the metal or the alloy in the normal state, on the one hand, and T_c, on the other, for superconductors with strong coupling. Table II lists the calculated characteristics of the phonon spectra of the alloys, $\langle\omega\rangle$, $\langle\omega^2\rangle$, and ω_0, the concentration dependence of the electron-phonon interaction parameter λ in accordance with MacMillan's equation [3], and the concentration dependence of the quantities $N_{bs}(0)$, $\langle I^2\rangle$, and $M\langle\omega^2\rangle$, which enter in this equation, with $N_{bs}(0)$ calculated from the experimentally determined electronic specific heat coefficient γ and λ, while $M\langle\omega^2\rangle$ is the product of the ion mass by the mean-squared phonon frequency and is determined independently. The same table lists the electron-phonon interaction parameter λ^* calculated by means of the so-called generalized MacMillan equation [4].

As seen from Table II, introduction of Ta impurity atoms in V, which is accompanied by the onset of quasi-local oscillations in the spectrum of the crystal with impurity, leads to a softening of the phonon spectrum, whereby $\langle\omega\rangle$, $\langle\omega^2\rangle$ and ω_0 decrease with increasing Ta concentration. However, since the constants of the interaction between the impurity and the neighbors are larger than the constants of the interaction in the initial lattice [12], the product $M\langle\omega^2\rangle$ increases with increasing Ta concentration, owing to the rapid growth of the average mass M. This tends to decrease λ. A similar tendency is produced by the change of the density of the electronic states on the Fermi surface. With increasing Ta concentration, the value of $N_\gamma(0)$ decreases, obviously indicating a transition from a metal with a narrow d-band to a metal with a broad d-band. As a result of the action of these factors, the electron-phonon interaction parameter λ also decreases with increasing Ta concentration. MacMillan's hypothesis [3] that the product $N_{bs}(0)\langle I^2\rangle$ is constant for isostructural systems does not hold in fact for the investigated alloys, as seen from Table II. With increasing impurity content, this product increases, so that the decrease of λ with increasing Ta concentration slows down.

Thus, within the framework of the theory of superconductors with strong coupling [3], as applied to the dilute solutions of Ta in V investigated in the present study, we can conclude that the rapid decrease of T_c following introduction of a relatively small amount of the isoelectronic impurity Ta into V is due to the decrease in the density of the electronic states on the Fermi surface with increasing product $M\langle\omega^2\rangle$, which leads to a lowering of the value of λ and to a decrease in the preexponential factor ω_0 in the MacMillan equation [8].

An obvious shortcoming of the foregoing analysis of the obtained data on the basis of the theory of superconductors with strong coupling [3] is that the quantity $\langle I^2\rangle$, which enters in the MacMillan equation [3], remains unknown. It is desirable to compare the experimental data with theoretical equations that contain no free parameters. Such a possibility is afforded by the use of the equation for T_c of a superconductor with weak coupling, either in the form of the equation of the BCS theory

$$T_c = 1.14\Theta \exp\{-1/N_\gamma(0)V_{ph}\} \quad (1)$$

or in the form of the MacMillan equation [3] under the assumption that $\lambda = N_\gamma(0)V_{ph}$. In this case, measurement of the temperature dependence of the resistance of V and of its alloys with Ta at high temperatures makes it possible to determine the character of the concentration dependence of the electron-phonon interaction potential V_{ph}. Indeed [14], for transition metals at $T \geq 2\Theta$ we can write approximately

$$V_{ph} \approx \text{const} \cdot \frac{d\rho}{dT} \frac{1}{1+5N_\gamma(0)}. \quad (2)$$

Such an approach to the analysis of the concentration dependence of T_c for a V – Ta alloy at high concentrations of Ta was used by Rapp [15].

Figure 3 shows the experimental data on the temperature dependence of the resistivity of the investigated samples in the temperature interval from 5 to 1000°K, while Table III lists the results of the calculation of $d\rho/dT$, V_{ph}, and T_c on the basis of these experimental data. It should be noted that curves 1–4 of Fig. 3 were obtained from results of reproducible measurements in the entire investigated temperature range. The value of T_c for the alloys was calculated with the data renormalized to T_c of the initial V. The data of Table III show that the value of V_{ph} remains practically unchanged in the investigated interval of the concentrations of Ta in V, in contradiction to Rapp's hypothesis [15]. The dependence of T_c on the concentration of the Ta and V is determined by the change in the density of states on the

TABLE II

Composition	$\frac{\langle\omega\rangle}{\omega_0}$	$\frac{\langle\omega^2\rangle}{\omega_0^2}$	ω_0	M_{ionic}	$M\langle\omega^2\rangle/\omega_0^2$	$N_\gamma(0)$, (eV-atom)$^{-1}$	$N_{bs}(0)$, (eV-atom)$^{-1}$	λ after BCS	$N_{bs}(0)\langle I^2\rangle$, eV/Å² atom	$\langle I^2\rangle$, (eV/Å)²	λ* [4]
V	0,559	0,358	460	50,94	18.21	2,08	1.38	0.50	9,14	6.60	0.72_4
V from [13]	—	—	—	50.94	—	2.00	1,33	0.50	9.12	6,85	—
$V_{99.18}Ta_{0.82}$	0,556	0,354	456	52.00	18.40	2,02	1,35	0.49_6	9.13	6.76	0.71_3
$V_{96.45}Ta_{3.55}$	0.546	0,343	444	55.56	19,07	1,96	1.31	0.49_2	9.38	7.14	0.70_6
$V_{94.32}Ta_{5.68}$	0,535	0,333	435	58,33	19,40	1.94	1.30	0.49_0	9,51	7.30	0.71_3

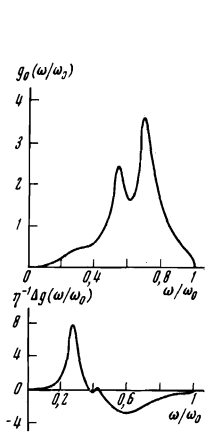

FIG. 2

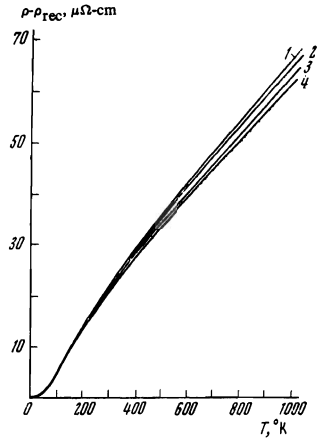

FIG. 3

FIG. 2. Phonon spectrum of V(top) and correction to the phonon spectrum of vanadium (bottom) due to the introduction of the Ta impurity atoms ($\epsilon = -2.55$, $\tau = 1.2$).

FIG. 3. Temperature dependence of the resistivity of pure V and its alloys with Ta in the range 5–1000°K: 1–pure V, 2–$V_{99.18}Ta_{0.82}$, 3–$V_{96.45}Ta_{3.55}$, 4–$V_{94.42}Ta_{5.68}$.

TABLE III

Composition	$\left(\frac{d\rho}{dT}\right)\cdot 10^3 \pm 0.5\%$, μΩ-cm/deg	$\frac{V_{ph}}{\text{const}}\cdot 10^3$	$\lambda = N(0)V_{ph}$ (const = 22,48)	T_c, °K calculation
V	5.7_1	5.0_1	0.234	5.24
$V_{99,18}Ta_{0,82}$	5.6_6	5.1_1	0.232	4.97
$V_{96,45}Ta_{3,55}$	5.4_7	5.0_6	0.223	4.05
$V_{94,32}Ta_{5,68}$	5.2_8	4.9_3	0.215	3.35

Fermi surface $N_\gamma(0)$ and by the change of the characteristic temperature Θ. It turns out here that the calculated values of T_c decrease with Ta concentration more rapidly than the experimental values.

So far, the experimental data on the concentration dependence of T_c in dilute solutions of Ta in V were evaluated on the basis of theoretical concepts that are valid for regular crystals. The investigated alloys are disordered, and to describe the interaction in the electron-ion system of these alloys it is more appropriate to use an approach based on the correlation-function formalism [7-9]. This approach makes it possible to redefine the electron-phonon interaction parameter λ in terms of the correlation function S(k, ω)[10]:

$$\lambda = \frac{(2\pi)^2 N(0)}{m^2 k_F^2}\int_0^{2k_F} k\,dk \int_0^{\infty} S(k,\omega)\frac{d\omega}{\omega}. \qquad (3)$$

When an impurity is introduced into a metal, a change takes place in its phonon and electron spectra. In the approximation linear in the concentration, this change is simply reflected by a change in the correlation function

$$S(k,\omega) = S_0(k,\omega) + \Delta S(k,\omega),$$

where $S_0(k, \omega)$ is the correlation function of the initial crystal, and $\Delta S(k, \omega)$ is the change produced in the correlation function by introducing the impurity and takes into account the dynamic character of the defects. Since the high-temperature resistivity of the metal due to single-phonon scattering is expressed in terms of the correlation function in a manner similar to (3):

$$\rho = \frac{3\pi(2\pi)^2 v_F^2 k_F^4 T}{2e^2 m^2}\int_0^{2k_F} k^3\,dk \int_0^{\infty} S(k,\omega)\frac{d\omega}{\omega} \qquad (4)$$

(the notation is standard), the change of the electron-phonon interaction parameter λ following the introduction of the impurity can be expressed in terms of the change of the high-temperature resistivity [10]:

$$\frac{\Delta\lambda}{\lambda_0} \approx \frac{\Delta\rho}{\rho_0}; \qquad (5)$$

here λ_0 and ρ_0 pertain to the initial metal and only the temperature-dependent part of the resistivity is taken into account.

The results of the experimental determination of the change of the resistivity of the alloys at $T \geq 2\Theta$ and of the comparison of this change with the change in the electron-phonon interaction parameter are listed in Table IV. This table gives also the calculated changes of T_c of the alloy with increasing Ta concentration, based on the equation

$$\frac{\Delta T}{T_c^0} \approx \frac{\Delta\rho}{\rho_0}\ln\left(\frac{\omega_0}{T_c^0}\right), \qquad (6)$$

derived by Maksimov [10] neglecting the Coulomb repulsion for disordered superconductors of non-transition metals in the model with weak coupling.

As seen from the data of Table IV, there is clear-cut correlation between the change of the high-temperature resistivity following introduction of the Ta impurity into V, on the one hand, and the change of the electron-phonon interaction parameter, on the other. However, it follows from the changes in the resistivity that the values of $\Delta\lambda/\lambda_0$ and $\Delta T_c/T_c^0$ should change more strongly with increasing Ta concentration, something not observed in the experiment. In view of the approximate character of (5) and (6), especially when applied to transition metals, the agreement between the calculations based on these equations with the experimental data can be regarded as satisfactory, and Maksimov's approach itself [10] can be regarded as useful but calling for further development.

It should be noted that the estimate of the sign and of the scale of the variation of the parameter λ can be made also directly from a calculation of $\Delta S(k, \omega)$, which is determined approximately by the change of the electron-ion scattering amplitude following introduction of the impurity (this quantity can be obtained from the residual resistivity of the alloy) and the change of the force constants of the interaction of the impurity with the neighbors in comparison with the interaction in the initial lattice [12]. It then turns out[1)] that

$$|\Delta\lambda/\lambda_0| \approx -0{,}7\eta, \qquad (7)$$

where η is the content of the Ta impurity atoms in at.%. This is in even better agreement with the experimental data than the results of calculations with Eqs. (5) and (6) (see Table IV). Thus, within the framework of the theory of superconductivity of nonideal crystals [10], the decrease of T_c of V following the introduction of the isoelectronic Ta impurity atoms is due primarily because the introduction of the Ta is accompanied by a negative change in the electron scattering amplitude and by a negative change (increase) of the force constants of the

TABLE IV

Composition	$(\Delta\rho/\rho_0)\cdot 10^2$ $T = 700°$ K	800° K	900° K	1000° K	Average	$\left(\frac{\Delta\lambda}{\lambda_0}\right)\cdot 10^3$ after BCS	$\left(\frac{\Delta\lambda^*}{\lambda_0^*}\right)\cdot 10^3$ from [4]	$\left(\frac{\Delta\lambda}{\lambda}\right)\cdot 10^3$ from (7)	T_c^0, °K from (6)
$V_{99,18}Ta_{0,82}$	−1,2	−1,3	−1,0	−1,1	−1,1	−0.8	−1,6	−0,9	4.96
$V_{96,45}Ta_{3,55}$	−4.7	−4,6	−4,5	−4.6	−4.6	−1.6	−3.3	−3,0	4,16
$V_{94,32}Ta_{5,68}$	−7.0	−7,2	−7,0	−6.4	−6.9	−2,0	−3.3	−4,8	3.80

interaction between the impurity and the environment, the contribution of the latter process being decisive.

Geĭlikman and Kresin[16] advanced the hypothesis that the introduction of heavy impurity atoms into a superconductor should be accompanied by its transition to the strong-coupling limit, since Θ decreases, and an additional attraction due to the interaction with the electrons on the impurity levels is possible between the conduction electrons. For the V – Ta system investigated in the present paper, the value of C_{es}/C_{en} at the transition point T_c approaches with increasing concentration the value 2.43 corresponding to the weak-coupling limit (see Table I). This indicates that this additional pairing mechanism is apparently not realized in dilute solutions of Ta in V, and no transition into a superconductor with strong coupling takes place.

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