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OPTICAL PROPERTIES AND ELECTRON CHARACTERISTICS OF VANADIUM-GALLIUM ALLOYS OBTAINED BY EVAPORATION IN VACUUM

A. I. GOLOVASHKIN, I. S. LEVCHENKO, and G. P. MOTULEVICH

P. N. Lebedev Physics Institute, USSR Academy of Sciences

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A new method for treating experimental optical data on alloys is proposed in which the contribution of virtual interband transitions to the dielectric constant is taken into account. A method of obtaining vanadium-gallium compounds is developed in which each metal is evaporated in vacuum from a separate evaporator. The optical constants in the spectral range between 0.45 and 10 μ are measured at $T = 293^\circ$K and in the 0.45–2.6 μ range at $T = 78^\circ$K. The density, electric, and superconducting properties of the alloys are also measured. It is found that in a broad range of concentrations the critical temperature of the superconducting transition ($\sim 14^\circ$K) practically does not depend on the composition. The observed dependence differs sharply from that of fused samples. The following electron characteristics of the vanadium-gallium alloy have been obtained: the concentration of conduction electrons, the frequency of electron collisions, the velocity of the electrons on the Fermi surface, the total area of the Fermi surface, the frequency of collisions between electrons and phonons or defects, and the pseudopotential Fourier components. The contribution of interband transitions to the dielectric constant and to the conductivity is determined. The contribution of virtual interband transitions is significant in the long-wave region.

1. AN OPTICAL METHOD OF DETERMINING THE ELECTRON CHARACTERISTICS IN ALLOYS[1]

THE investigation of the optical properties of metals makes it possible to obtain their main electron characteristics.[1] Analogous investigations in alloys can also be utilized for the same purpose. However, the relationship between the electron characteristics and optical constants in alloys has a series of special features. As a result of this, a simple transfer of the processing methods of the experimental data used in the case of metals to alloys can lead to considerable errors.

In order to obtain the characteristics of the conduction electrons, it is usual to make use of measurements in the long-wave region of the spectrum where there are no real interband transitions. However, as has been shown in[2], the contribution of virtual interband transitions to the dielectric constant of metals and alloys in this spectral range is 10–100. In the common metals the contribution of the conduction electrons to the dielectric constant ϵ exceeds this value considerably; for this reason one need not take into account virtual transitions. Because of the high effective frequency of electron collisions ν, in alloys the contribution of conduction electrons to ϵ is of the same order of magnitude as the contribution of the virtual interband transitions and must not be neglected. The large value of ν gives rise to the normal skin effect. As a result of this, the formulas which relate the optical constants to the microcharacteristics are of the form

$$\varepsilon(\omega) \equiv n^2 - \varkappa^2 = 1 + \tilde{\varepsilon}(\omega) + \varepsilon_e(\omega), \tag{1}$$

$$\sigma(\omega) \equiv \frac{n\varkappa\omega}{2\pi} = \tilde{\sigma}(\omega) + \sigma_e(\omega), \tag{2}$$

$$\varepsilon_e = -\frac{4\pi e^2}{m}\frac{N}{\omega^2+\nu^2}, \quad \sigma_e = \frac{e^2}{m}\frac{N\nu}{\omega^2+\nu^2}. \tag{3}$$

Here ϵ and σ are the dielectric constant and the conductivity at a frequency ω; $\tilde{\epsilon}$ and $\tilde{\sigma}$ are the contributions of the interband transitions to the dielectric constant and to the conductivity; ϵ_e and σ_e are the corresponding contributions of the conduction electrons; $n - i\kappa$ is the complex refractive index; N is the concentration of conduction electrons; e and m are the charge and mass of a free electron.

In the region in which there are no real interband transitions $\tilde{\sigma} \ll \sigma_e$ and the contribution of such transitions to σ is negligible. Therefore formula (2) takes on the form

$$\sigma(\omega) = \frac{e^2}{m}\frac{N\nu}{\omega^2+\nu^2}, \tag{4}$$

and it can be used to determine N and ν. Relation (1) in which $\tilde{\epsilon}$ is neglected is usually also used. As has already been pointed out, in alloys this can lead to considerable errors.

It follows from (4) that

$$\frac{1}{4\pi\sigma} \equiv \frac{1}{2n\varkappa\omega} = A\omega^2 + B, \tag{5}$$

where

$$A = \frac{m}{4\pi e^2}\frac{1}{N\nu}, \quad B = \frac{m}{4\pi e^2}\frac{\nu}{N}. \tag{6}$$

Thus, in the spectral range in which the quantity $1/4\pi\sigma$ is a linear function of ω^2 we obtain from the slope A of the straight line (5) and from the intercept B of this straight line on the ordinate axis

$$N = \frac{m}{4\pi e^2}\frac{1}{\sqrt{AB}}, \quad \nu = \sqrt{\frac{B}{A}}. \tag{7}$$

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[1] The method considered below is also applicable to metals in which the normal skin effect takes place (for example, transition metals at room temperature).

Using the obtained values of N and ν, one can find $\tilde{\varepsilon}(\omega)$ from formula (1). The method of processing experimental data indicated below was used to obtain the electron characteristics of vanadium-gallium alloys.

2. THE OPTICAL AND SUPERCONDUCTING PROPERTIES OF VANADIUM-GALLIUM ALLOYS

The superconducting compounds of vanadium and gallium are of great interest because they have high values of T_c, j_c, and H_c (here T_c is the transition temperature to the superconducting state, j_c is the critical current density, and H_c is the critical magnetic field). In this paper we investigate both the optical and superconducting properties of these alloys and determine their electron characteristics.

1. Samples. We have worked out a method of obtaining vanadium-gallium compounds based on simultaneous vacuum evaporation of vanadium and gallium from separate evaporators. This method made it possible to obtain samples with T_c up to 14°K and j_c up to 2×10^5 A/cm^2 with a good optical surface. The evaporation was carried out in a vacuum of 2×10^{-6}–2×10^{-5} torr on polished substrates prepared of sapphire or ruby. The substrate temperature was about 500°C. The substrate material was specially chosen so that it would not produce compounds with either vanadium or gallium. The evaporation of the vanadium was carried out thermally from a tungsten coil or by an electron beam.[3] The gallium was evaporated thermally from tantalum boats. The substrates were placed at a distance of ~120 mm from the evaporators. Such a geometry ensured a sufficiently good homogeneity of the obtained samples. The samples were annealed at a pressure of 2×10^{-6} torr and a temperature of ~800°C for 1–2 hours directly after evaporation without letting air into the vacuum chamber.

The percentage composition of the obtained alloy was determined by weighing. We had previously prepared experimental calibration curves which made it possible to determine from the weight of evaporated vanadium or gallium the weight of the corresponding element deposited on our substrate. The use of calibration curves made it possible to determine readily the percentage composition of the alloy. A chemical analysis of the prepared alloys carried out in a number of cases as a check confirmed the results obtained with the calibration curves. Samples of various compositions were prepared by this method.

Table I. Composition dependence of the characteristics of the investigated samples

Weight content of Ga, percent	ρ, g/cm^3	$\sigma_0 \cdot 10^{-16}$, cgs esu	R_{res}/R_r
80	—	—	0.12±0.01
61	—	—	0.24±0.02
51	—	—	0.50±0.08
44	—	—	0.45±0.02
37	6.1±0.1	0.82±0.1	0.51±0.01
33	6.3±0.1	1.2±0.1	0.56±0.02
31	—	—	0.84±0.02
20	—	1.1±0.1	0.60±0.02
11	—	1.0±0.1	0.65±0.02
6	5.6±0.3	0.78±0.1	0.56±0.02
3	—	—	0.84±0.02

2. Characteristics of the samples. The statistical characteristics averaged over several depositions are given in Table I. The density ρ at room temperature, the static conductivity at room temperature σ_0, and the ratio of the residual resistance to that at room temperature R_{res}/R_r were measured by the usual methods. We obtained an electron diffraction pattern for the alloy containing 33 weight percent gallium. Using the rings of the Debye pattern of the various indices of the atomic planes, we found a lattice constant a = 5.0 ± 0.05 Å. This value is close to the lattice constant of fused samples[2] of V_3Ga for which a = 4.82 Å.[4] It is seen from Table I that the density of the samples containing 33 percent gallium is also close to the density of fused V_3Ga samples which is 6.6 g/cm^3.

Figure 1 shows the dependence of the temperature of the transition to the superconducting state T_c on the composition of the alloy[3]. For comparison we show the analogous dependence taken from[4] for fused samples. It is seen from Fig. 1 that the maximum values of T_c for deposited and fused samples are close to one another. However, the dependence of T_c on the composition differs sharply. It turned out that for alloys prepared by this method the high value of T_c is retained in a very wide range of gallium concentrations (from ~6 to ~40 percent). On decreasing the gallium content below 6 percent T_c decreases sharply. A rather sharp change of T_c with concentration was also observed on increasing the gallium concentration above 50 percent. Alloys containing 60 percent gallium remained in the normal state even at liquid helium temperature. Such a dependence of T_c on the concentration of the obtained alloy is possibly connected with the fact that the compound V_3Ga is readily produced under our conditions. An excess of vanadium or gallium has little effect on the superconducting properties.

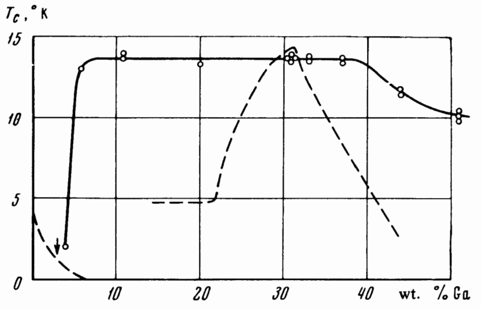

FIG. 1. Dependence of the temperature of the transition to the superconducting state on the composition of the vanadium-gallium alloy. Solid curve–deposited samples (this work), dashed curve–fused samples. [4] The arrow in the Figure indicates that deposited samples containing 3 percent gallium did not go over to the superconducting state down to a temperature of 2°K.

The superconducting transition for the obtained samples was rather sharp. Its width ΔT_c was between

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[2] The stoichiometric composition of the V_3Ga alloy is 31.3 wt. percent gallium.

[3] T_c was measured from the temperature dependence of the resistance. T_c was taken to be the temperature corresponding to a resistance equal to one half of R_{res}.

0.2 and 1°. The critical current density of the investigated samples was of the same order of magnitude as in the case of fused V_3Ga samples investigated in[5].

Thus, the method of preparation presented makes it possible to obtain readily alloys of vanadium and gallium with high critical parameters.

3. Optical properties. The optical constants n and κ were determined by the polarization method on setups described in[6,7]. Fourfold reflection of light from the investigated mirrors was employed. The results obtained on the different setups coincided in the overlapping spectral range.

Optical parameters are cited for samples containing 37, 33, and 6 percent gallium for T = 293°K and T = 78°K. At room temperature the measurements were carried out in the spectral range of 0.45—10 μ, and at nitrogen temperature between 0.45—2.6 μ. The spectral width of the slit for the 0.5—0.7 μ region was less than 0.01 μ, for $\lambda \approx 1$ μ—about 0.05 μ, for $\lambda \approx 3$ μ—about 0.06 μ, and for the 7—10 μ region—about 1 μ. The optical constants referring to samples of various compositions turned out to be close to one another. Therefore, we present in Table II and in Figs. 2 and 3 the values of n and κ averaged over the indicated compositions. The deviation of the values referring to the various compositions from the average amounted in the short-wave spectral region (0.45—2.6 μ) to 1—2 percent for κ and 2—3 percent for n; in the long-wave spectral region (3—8 μ) it was 1.5—3 percent for κ and 4—5 percent for n. At the very end of the investigated spectral range (9—10 μ) the deviation for both constants amounts to 5—8 percent. The errors in the measurements referring to a single composition are smaller than the indicated deviations.

It is seen from Figs. 2 and 3 and from Table II that for the investigated vanadium-gallium alloys $\kappa > n$ and $\epsilon = n^2 - \kappa^2 < 0$ as in the case of the usual metals. However, $|\epsilon|$ and κ are considerably smaller than in the usual metals. With increasing wavelength n and κ increase monotonically but their λ dependence is weaker than in good metals.

No new features appear in the n(λ) and $\kappa(\lambda)$ dependences on decreasing the temperature to 78°K. The values of κ for 78°K coincide practically with the values of κ for 293°K, and the values of n for 78°K are somewhat lower than the values of n for 293°K; however this difference is small and the optical constants at 78°K are therefore not presented in Figs. 2 and 3.

Table II. Optical constants

λ, μ	T = 293°K		T = 78°K		λ, μ	T = 293°K		T = 78°K	
	n	ϰ	n	ϰ		n	ϰ	n	ϰ
0.45	2.09	3.09	1.96	3.09	1.90	—	—	4.22	5.71
0.50	2.33	3.37	2.24	3.36	2.0	4.70	6.04	4.47	6.16
0.55	2.45	3.42	2.35	3.42	2.1	—	—	4.48	6.32
0.60	2.60	3.60	2.45	3.60	2.2	—	—	4.58	6.56
0.65	2.76	3.68	2.65	3.68	2.25	4.93	6.61	—	—
0.70	2.90	3.93	2.82	3.99	2.3	—	—	4.61	6.86
0.74	3.12	3.98	3.06	3.90	2.4	—	—	4.74	6.66
0.76	—	—	3.12	3.98	2.5	5.2	6.9	4.79	6.78
0.80	3.29	4.06	3.26	4.00	2.6	—	—	4.95	7.10
0.85	3.33	4.07	3.27	4.01	2.75	5.4	7.9	—	—
0.90	3.52	4.18	3.45	4.16	3.0	5.7	8.4	—	—
0.95	—	—	3.50	4.16	3.5	6.3	9.6	—	—
0.98	—	—	3.49	4.24	4.0	6.9	10.4	—	—
1.0	3.58	4.34	3.55	4.33	4.5	7.7	11.4	—	—
1.04	—	—	3.58	4.31	5.0	8.4	12.2	—	—
1.07	—	—	3.60	4.34	5.5	9.0	12.8	—	—
1.10	—	—	3.63	4.37	6.0	9.6	13.2	—	—
1.20	4.04	4.67	3.94	4.61	6.5	10.2	14.0	—	—
1.30	—	—	4.16	4.67	7.0	10.8	14.1	—	—
1.40	—	—	4.35	4.76	7.5	11.8	14.8	—	—
1.50	4.32	5.06	4.30	4.94	8.0	12.3	15.3	—	—
1.60	—	—	4.35	5.16	9.0	13.2	16.6	—	—
1.70	4.48	5.50	4.44	5.28	10.0	13.6	16.8	—	—
1.80	—	—	4.32	5.24	—	—	—	—	—

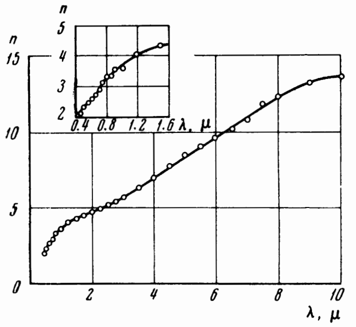

FIG. 2. Wavelength dependence of the refractive index for T = 293°K.

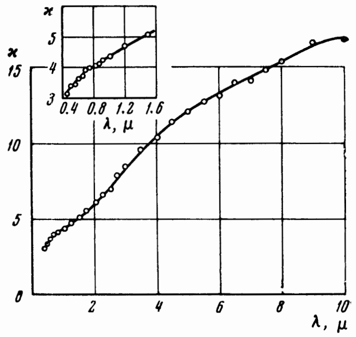

FIG. 3. Wavelength dependence of the absorption index for T = 293°K.

3. THE MICROCHARACTERISTICS OF VANADIUM-GALLIUM COMPOUNDS

1. The dependence of $1/4\pi\sigma$ on ω^2 for the investigated compounds is shown in Fig. 4. It is seen from this Figure that in the 3—10 μ spectral range this dependence is linear. The straight line drawn in the figure was obtained by the method of least squares. Utilizing the slope of the straight line and its intercept on the ordinate axis, we obtain values of N and ν cited below:

$$N = 2.4\cdot 10^{22}\ \text{cm}^{-3}, \quad N_a = 6.40\cdot 10^{22}\ \text{cm}^{-3}, \qquad N/N_{\text{val}} = 0.08, \quad N/N_a = 0.38; \tag{8}$$

$$\nu = 8.2\cdot 10^{14}\ \text{sec}^{-1}, \qquad \nu_{ep} = 4.6\cdot 10^{14}\ \text{sec}^{-1}, \quad \nu_{ed} = 3.6\cdot 10^{14}\ \text{sec}^{-1}. \tag{9}$$

The deviation of the values referring to the various compositions from the average amounts to 7 percent for N and 1.5 percent for ν. The measurement error of these quantities in each series is 4 percent. In (8) we also cite the concentration of atoms N_a and the ratios N/N_{val} and N/N_a where N_{val} is the valence electron concentration (the values of N_a and N_{val} were obtained for a lattice constant a = 5.0 Å).

2. Using the relations[8]

$$v_F / v_F^0 \simeq S_F / S_F^0 \simeq \sqrt{N / N_{\text{val}}}, \tag{10}$$

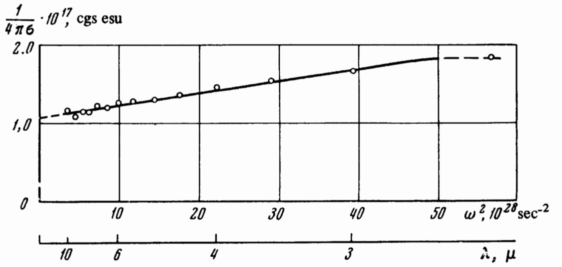

FIG. 4. Dependence of the quantity $1/4\pi\sigma$ on ω^2.

we obtain the average velocity of the electrons on the Fermi surface and the total area of the Fermi surface S_F:

$$v_F = 0.68\cdot 10^8\ \text{cm/sec} \qquad v_F / v_F^0 = 0.29;$$
$$S_F = 1.7\cdot 10^{-37}\ \text{g}^2\text{-cm}^2/\text{sec}^2 \quad S_F / S_F^0 = 0.29. \qquad (11)$$

The values v_F^0 and S_F^0 refer to free electrons at a concentration equal to the valence concentration[4)].

3. Relations (8) of [8], as well as the static conductivity and the ratio of R_{res}/R_r of our samples, made it possible to obtain the collision frequencies of the electrons with phonons ν_{ep} and with defects ν_{ed}. They are cited in (9).

4. The following depth of the skin layer δ (for $\lambda \approx 6\ \mu$) and electron mean free path l were obtained:

$$l = 0.08\cdot 10^{-6}\ \text{cm}$$
$$\delta = 7.26\cdot 10^{-6}\ \text{cm}.$$

It is seen that the normal skin effect appears in fact in the investigated vanadium-gallium alloys, i.e. $l \ll \delta$ and $v_F \ll c/n$; the formulas for processing cited in Sec. 1 are therefore valid.

Using the values of N and ν obtained in the long-wave region, we calculated the interband dielectric constant $\tilde{\epsilon}$ and the interband conductivity $\tilde{\sigma}$ according to formulas (1)—(3). The $\tilde{\sigma}(\omega)$ and $\tilde{\epsilon}(\omega)$ dependences are shown in Figs. 5 and 6. It is seen from Fig. 5 that there exists a broad band connected with interband transitions in the region $\hbar\omega > 0.5$ eV. Maxima appear at 1.68 and 2.48 eV. An additional band is apparently superimposed in the region of 1 eV. For $\hbar\omega \lesssim 0.5$ eV one can neglect the effect of interband transition on σ.

It is seen from Fig. 6 that anomalous dispersion is observed in the region $\hbar\omega > 0.5$ eV. For $\hbar\omega \approx 2.1$ eV the permittivity $\tilde{\epsilon}$ vanishes and for $\hbar\omega > 2.1$ eV it becomes negative. For $\hbar\omega < 0.5$ eV normal dispersion is observed. For $\hbar\omega \lesssim 0.25$ eV $\tilde{\epsilon}$ practically ceases to depend on the frequency and takes on a value of about +14 ± 5. The scatter of points in this region is somewhat larger than in the region of large ω since the sought quantity is determined as the difference of two large quantities. A dashed averaged curve is therefore drawn in Fig. 6. Thus, even in the long-wave region $\tilde{\epsilon}$ amounts to 15—20 percent of ϵ and one must not neglect $\tilde{\epsilon}$.

4. DISCUSSION OF THE RESULTS

1. The cited results [see (8), (9), and (11)] show that the absolute values of the concentration of the conduction electrons are approximately by a factor of two smaller than in metals of the type of lead or tin.[6-7] The ratio of this concentration to the concentration of valence electrons amounts in all to 0.08, whereas in lead or tin this ratio is about 0.3.

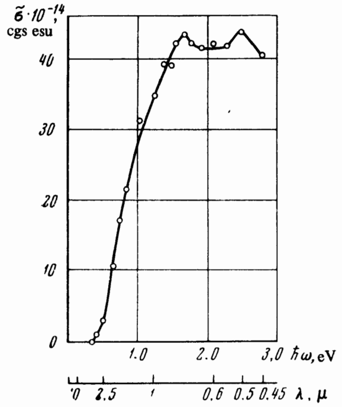

FIG. 5. Dependence of the interband conductivity $\tilde{\sigma}$ on ħω.

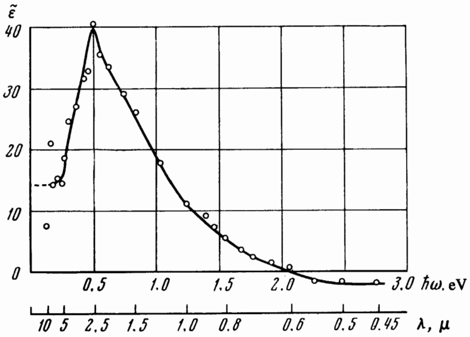

FIG. 6. Dependence of the interband dielectric constant $\tilde{\epsilon}$ on ħω.

2. The small value of N/N_{val} is in qualitative agreement with the presence of a large number of Bragg planes with a nonzero structure factor which intersect the free-electron sphere.[9] The radius of the free-electron sphere[9] is in our case $p_F = 1.63\,(2\pi\hbar/a)$. The compound V_3Ga has the structure of β-tungsten. The indices $n_1n_2n_3$ of the Bragg planes with nonzero structure factors which intersect the free-electron sphere for this alloy are listed in Table III. The same table also lists the distances $p_{n_1n_2n_3}$ of the corresponding planes to the center of the zone Γ, the number of physically equivalent planes $n_{n_1n_2n_3}$ and $|F_{n_1n_2n_3}|$. It is seen from Table III that the free-electron sphere is intersected by 12 {110} planes, 6 {200} planes, 24 {210} planes, 24 {211} planes, 12 {220} planes, and 24 {310} planes. With such a number of Bragg planes N, S_F, and v_F should be considerably smaller than N_{val}, S_F^0, and v_F^0, even for small values of the Fourier components of the pseudopotential. In fact, for this alloy $N/N_{val} = 0.08$, $v_F/v_F^0 \approx S_F/S_F^0 \approx 0.29$. For lead[7] $N/N_{val} = 0.31$ and $v_F/v_F^0 \approx S_F/S_F^0 \approx 0.56$. The difference between the characteristics of our alloy and those of lead are particularly appreciable if it is borne in mind that the volume included in the Fermi surface for the indicated alloy is larger by a factor of two than that for lead.

[4)] For our alloy $V_F^0 = 2.36 \times 10^8$ cm/sec, $S_F^0 = 5.83 \times 10^{-37}$ g^2-cm^2/sec^2, and $E_F^0 = 15.9$ eV where E_F^0 is the Fermi energy.

Table III. Bragg planes that intersect the free-electron sphere and have non-zero structure factors

Indices $n_1n_2n_3$ of the planes	Distance from the center of the zone $p_{n_1n_2n_3}$ $(2\pi\hbar/a)^{-1}$	Number of physically equivalent planes n_g	Structure factor $\lvert F_{n_1n_2n_3}\rvert$
110	0.707	12	0.70
200	1.00	6	4.70
210	1.118	24	4.00
211	1.225	24	4.70
220	1.414	12	0.70
310	1.581	24	0.70

3. Such a large number of important Bragg planes with various indices should lead to a complicated situation for the interband conductivity $\tilde{\sigma}$. The experimentally observed band in $\tilde{\sigma}$ consists apparently of at most three superimposed bands. This is indicated by the fact that a decrease of the temperature to 78°K has almost no effect on the shape of the $\tilde{\sigma}$ band.

Taking into account the results of[2], the position of the maxima of $\tilde{\sigma}$ makes it possible to determine three values of the Fourier components of the pseudopotential. They turned out to be $|V_1| = 1.0$ eV, $|V_2| = 0.65$ eV, and $|V_3| = 0.4$ eV. It is so far impossible to identify these values with specific Bragg planes. The presence of Bragg planes with a small structure factor can lead to the situation in which the $\tilde{\sigma}$ bands due to them will be located in the far infrared region.

4. As follows from the results presented above, the frequency of electron-phonon collisions ν_{ep} is of the same order of magnitude as that of metals of the type of lead[10] for which $\nu_{ep} = 3.05 \times 10^{14}$ sec^{-1}. The frequency of collisions with defects ν_{ed} for the investigated alloys is high. It is of the same order as ν_{ep}. It is at the same time natural that a decrease of the temperature has little effect on $\nu = \nu_{ep} + \nu_{ed}$.

5. The results of this work confirm the point of view that a large number of Bragg planes leads to high values of T_c and j_c. Similar views have already been expressed in the literature, for example in[11]. In connection with what has been said above it would be interesting to carry out analogous work for fused V_3Ga alloys as well as for other compounds with high T_c and H_c.

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