

FIG. 1. Variation in the x-ray  $K_{\beta 1}$ -line shifts of Eu (full circles) and Sm (open circles) with temperature. The continuous and dotted lines show the behavior of the heat capacities of Eu and Sm<sup>[1,2,7]</sup> respectively.

the atoms of Eu and Sm are respectively equal to  $\Delta E = -1450 \pm 40$  and  $-1455 \pm 40$  meV<sup>[8]</sup> it may be unambiguously concluded that none of the above phase transitions is accompanied by a change of more than  $\eta \approx \pm 0.07$  electron/atom<sup>2)</sup> in the number of 4f electrons in Eu and Sm.

The authors are grateful to Yu. I. Vasil'ev and N. M. Miftakhov for assistance in the measurements.

<sup>1)</sup>In view of the anomalously deep position (in the radial direction) of the 4f-electron in the atom its removal or excitation

leads to significant changes (shifts) in the energy of the K lines.  
<sup>2)</sup>An analogous result was obtained by us in studying the ferromagnetic-paramagnetic transition in metallic gadolinium ( $T_c = 290$  K).

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## Superconducting transition temperature, critical magnetic fields, and the structure of vanadium films

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The superconducting transition temperature  $T_c$ , the perpendicular critical magnetic field  $H_c^\perp$ , the electrical resistance, and the structure of vanadium films obtained by ionic evaporation in an ultrahigh vacuum apparatus were investigated. A carbon sublayer and coating of 15 Å thickness were applied in order to protect the samples against external influences. The critical fields were measured by the resistive method. Upon a decrease of the film thickness from 2900 to 60 Å a reduction of  $T_c$  from 5.1-5.2 to 2.4 K was observed, along with an increase of the residual resistivity  $\rho_n$  from 4.5 to 20 μohm-cm and an increase of the derivative  $|dH_c^\perp/dT|$  near  $T_c$  from 3.6 to 6.7 kOe/K. No superconductivity was observed in a film of 30 Å thickness down to 1.3 K. The type of crystal structure and the lattice constant for 125-1000 Å thick films, determined by the technique of electron diffraction, are the same as those for bulk samples of vanadium; the grain size in these films amounted to 500 to 800 Å. The electron density of states  $N(0)$ , calculated on the basis of the data concerning  $|dH_c^\perp/dT|$  and  $\rho_n$  for thick films (720 to 2900 Å), agrees with the value of  $N(0)$  for bulk vanadium (if it is assumed that the coefficient, which takes account of the correction due to strong coupling effects for  $H_{c2}$ , is given by  $\eta = 1.2$ ). Upon a reduction of the film thickness to 60 Å,  $N(0)$  is reduced by approximately a factor of two. The obtained results agree with the assumption that strong variations of  $N(0)$ , having an influence on  $T_c$ , can be observed in transition-metal films due to a reduction in the electron mean free path. The obtained quantitative information with regard to  $N(0)$  in thin vanadium films ( $d \leq 250$  Å) is sensitive, however, to the presence of a hypothetical surface transition layer and to its properties and thickness.

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### 1. INTRODUCTION

It follows from articles devoted to the investigation of the superconducting properties of vanadium films (a brief review of this problem is given in<sup>[1]</sup>) that:

- 1) These properties are very sensitive to impurities, and a transition temperature close to the  $T_c$  for pure, bulk vanadium (5.38 K) was obtained only for rather thick layers (thickness  $d \gtrsim 2000$  Å) and for a rather small ratio of the fluxes of residual gas and metal on

the substrate.

2) A new modification of vanadium having a high electrical resistance, associated with condensation on a cold substrate ( $T_{\text{subs}} \sim 4.2$  K), is observed<sup>[2]</sup> in films of thickness  $d < 200$  Å, the high electrical resistance being the reason to regard this new form as amorphous.<sup>[1]</sup>

3) The critical temperature of vanadium films drops as their thickness decreases for both the amorphous form and for the usual modification.<sup>[1]</sup>

Crow, Strongin *et al.*<sup>[3]</sup> expressed the conjecture that, in transition-metal films the change of  $T_c$  takes place primarily as a consequence of a change in the electron density of states  $N(0)$  at the Fermi surface. However, up to now no direct experiments, enabling one to uniquely establish for films the connection between changes of  $T_c$  and changes of the density of electron states, have been carried out.

In the present article we investigate the superconducting properties and structure of vanadium layers of varying thickness, obtained by the method of ionic evaporation in an ultrahigh vacuum apparatus,<sup>[4]</sup> and an attempt is made to observe a correlation between changes of  $T_c$  and  $N(0)$  in thin film samples of vanadium. In order to obtain information about  $N(0)$ , the perpendicular critical magnetic fields  $H_c^\perp$  and the electrical resistivity were measured. In addition, the parallel critical magnetic fields  $H_c^\parallel$  were measured. Together with these measurements, electron microscope and electron diffraction studies of the structure of the samples were carried out.

## 2. PREPARATION OF THE SAMPLES AND MEASUREMENT PROCEDURE

The technique for preparation of the film samples by ionic evaporation has been described by us in<sup>[4]</sup>. The deposition was carried out in an ultrahigh vacuum apparatus under a hydrogen pressure  $\leq 10^{-9}$  Torr, and the pressure of the other residual gases was  $< 10^{-10}$  Torr. The pressure of krypton, which was employed as the working gas, amounted to  $5 \times 10^{-6}$  Torr. The rate of vanadium deposition in different experiments varied from 4.5 to 24.5 Å/min depending on the intensity of the discharge current. Vanadium twice smelted with an electron-beam, of purity better than 99.94%, was used as the cathodes.

Glass substrates with soldered platinum leads were mainly used, but also Pyroceram substrates. In order to eliminate the effect of possible imperfections and impurities of the substrates on the properties of the sample, an artificial diamond layer<sup>[5]</sup> of thickness 10–15 Å was deposited on the substrate before deposition of the vanadium film. After the completion of its deposition, the vanadium film was covered by the same diamond layer in order to protect it from oxidation. As the measurements have shown, the resistance and superconducting properties of samples thus protected do not change noticeably during the course of a year.

All measurements of critical magnetic fields were carried out on films, whose edges were cut by a diamond

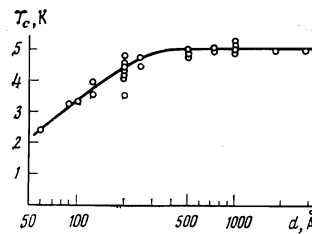


FIG. 1. Dependence of the superconducting transition temperature of vanadium films on thickness.

microcutter, in order to eliminate edge effects. The method which we used to measure the critical fields according to resistance was described in detail in a previous article.<sup>[6]</sup> Above 4.2 K the transition curves were plotted for fixed magnetic fields and a slow variation of the temperature; below 4.2 K the temperature was held constant and the magnetic field was varied.

The structure of the samples was investigated by transillumination in a BS-513 electron microscope.

## 3. RESULTS

Films in a range of thicknesses from 30 to 2900 Å were investigated. The transition curves in a zero magnetic field are rather sharp; the width of the transition varied from 0.005–0.02 K for films of thickness  $d = 725$ –2900 Å up to 0.18 K for films of thickness  $d = 60$  Å. The temperature at which  $R = 0.5 R_n$ , where  $R_n$  denotes the sample's resistance after the transition into the normal state, was taken as  $T_c$ .

The dependence of  $T_c$  on  $d$  is given in Fig. 1. The critical temperature of thick films ( $d \sim 500$  to 2900 Å) amounts to 5.04–5.21 K. The proximity of these values, even for quite thin films (500 Å), to  $T_c$  for pure, bulk vanadium (5.38 K for a sample with  $R_{300}/R_n = 140$ <sup>[7]</sup> and 5.21–5.24 K for a sample with  $R_{300}/R_n = 24$ <sup>[8]</sup>) indicates the high quality of the prepared films. The rather high value of the ratio of the resistances,  $R_{300}/R_n = 5.5$  to 6.6 for films in this range of thicknesses, also says something about this point. A lowering of  $T_c$  begins upon a reduction of the thickness below 500 Å. Superconductivity is not observed in a film of thickness  $d = 30$  Å down to a temperature of 1.5 K.

Ten samples were utilized for the measurements of the critical magnetic fields and resistivity; the data concerning these measurements is presented in Table I. The critical fields were determined with respect to the middle of the transition ( $R = 0.5 R_n$ ). The dependences  $H_{c1}(T)$  and  $H_{c1}^2(T)$  are linear near  $T_c$ . For each given sample the width and shape of the transition curves do not change significantly with increasing magnetic field, so that the slopes of the straight lines  $H_{c1}(T)$  and  $H_{c1}^2(T)$  are essentially independent of the method of determining  $H_c$ —according to the middle or the end of the transition curve (in the latter case, the value of the field, obtained by extrapolation of the middle, straight-line segment of the transition curve to  $R = R_n$ , is important); the difference amounts to no more than 2–3%. The width of the transition in a transverse field varies from  $\sim 0.14$  kOe for films with  $d = 720$ –2900 Å to  $\sim 1.25$  kOe for a film with  $d = 60$  Å.

TABLE I. Data characterizing the films utilized for measurements of critical magnetic fields and resistivities.

Sample	d, Å	Rate of deposition, Å/min	T <sub>c</sub> , K	R <sub>295</sub> /R <sub>n</sub>	ρ <sub>n</sub> , μohm-cm	R <sub>295</sub> - ρ <sub>n</sub> , μohm-cm	dH <sub>c1</sub> /dT , kOe/K	Δ(H <sub>c1</sub> )/ΔT , kOe/K	l/ξ <sub>0</sub> , Å	l/ξ <sub>0</sub>	ν = 0.88 ξ <sub>0</sub> /l	νF = const		νF = const N(0)	
												η = N(0)/N <sub>m</sub> (0)	N(0)/N <sub>m</sub> (0)	ν = N(0)/N <sub>m</sub> (0)	N(0)/N <sub>m</sub> (0)
1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
E3	2900	24.5	5.08	5.55	4.47	20.3	3.57	3.78	78	0.15	5.5	1.20	1.00	1.23	1.00
D1	1800	9.0	5.04	6.63	3.77	21.2	2.93	6.60	93	0.18	4.6	1.11	1.00	1.13	1.00
B6	1000	9.0	5.14	5.87	4.18	20.4	3.28	—	84	0.17	5.2	1.15	1.00	1.17	1.00
B8	1000	4.5	5.14	5.90	4.02	19.7	3.28	12.5	87	0.17	5.2	1.20	1.00	1.22	1.00
A8	720	24.5	5.05	5.50	4.53	20.4	3.62	30.8	77	0.15	5.5	1.21	1.00	1.22	1.00
B1	720	24.5	5.18	5.55	4.47	20.3	3.57	—	78	0.16	5.5	1.10	1.00	1.21	1.00
A6	250	18.0	4.67	4.53	6.48	22.9	3.97	415	54	0.10	8.0	0.96	0.83	1.04	0.87
C1	200	18.0	4.42	4.31	7.93	26.2	4.57	647	44	0.07	8.8	0.94	0.81	1.00	0.83
A5	125	18.0	3.91	3.53	10.6	26.8	5.19	1760	33	0.05	13	0.82	0.69	0.88	0.73
A3	60	18.0	2.42	2.34	19.7	26.4	6.70	1700	18	0.02	44	0.60	0.51	0.64	0.53

It is clear from the table that, just as in the case of T<sub>c</sub>, the deviation of the quantities R<sub>295</sub>/R<sub>n</sub>, the residual resistivity ρ<sub>n</sub>, and |dH<sub>c1</sub>/dT| from the values characteristic of thick films begins for d < 500 Å.

As a result of the electron microscope and electron diffraction investigations of samples of thickness 125–1000 Å, it is established that the films represent polycrystals. The grains have a planar shape, and as a rule the superposition of one grain on another is observed (a moiré pattern). Hence one can conclude that the average size of a grain in a direction perpendicular to the plane of the film is apparently several times smaller than the sample thickness. The average size of the grain in the plane of the film amounts to ~ 500 Å in all samples with the exception of certain films of thickness 1000 Å in which an average grain size ~ 800 Å was observed. The protective layers of diamond do not have any effect on the structure and lattice constant of the samples. The parameter (3.03 ± 0.01 Å) and the crystal lattice type (bcc) coincide in the investigated films and in bulk vanadium. Electron diffraction patterns and electron microphotographs for films of different thicknesses are shown in Fig. 2.

4. DISCUSSION OF THE RESULTS

The analysis of the results is based on the microscopic theory of weakly bound superconductors in a magnetic field, developed by Gor'kov.<sup>[9]</sup> From this theory it follows that

$$|dH_{c1}/dT|_{T=T_c} = (2\pi^2 k^2 c / \hbar e) (T_c / v_F^2) F(\nu)^{-1}, \tag{1}$$

where k is the Boltzmann constant, c is the velocity of light in vacuum, ħ is Planck's constant, e is the electron charge, v<sub>F</sub> is the Fermi velocity, and

$$F(\nu) = (1/2) \left\{ \sum_{n=0}^{\infty} [(2n+1)^2 |2n+1+\nu|^{-1}] \right\}^{-1}, \tag{2}$$

$$\nu = \hbar v_F / 2\pi k T_c l = 0.88 \xi_0 / l. \tag{3}$$

l denotes the mean free path of the electrons, and ξ<sub>0</sub> denotes the coherence length:

$$\xi_0 = 0.18 \hbar v_F / k T_c. \tag{4}$$

In the case of strong or intermediate coupling,<sup>[12-16]</sup> it is necessary to introduce a coefficient η of the order of

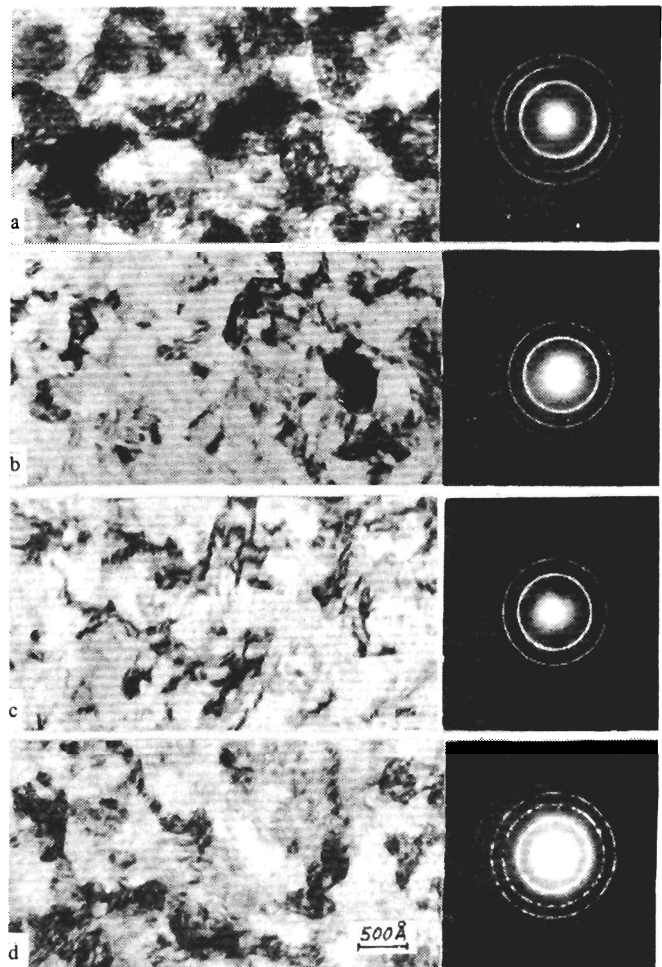


FIG. 2. Electron microphotographs and electron diffraction patterns for vanadium film samples of varying thickness (×250,000): a—d=1000 Å, b—d=500 Å, c—d=200 Å, d—d=125 Å.

unity on the right-hand side of Eq. (1):

$$|dH_{c2}/dT|_{T=T_c} = (2\pi^2 k^2 c / \hbar e) (\eta T_c / v_F^2) F(\nu). \quad (5)$$

The inequality  $0.02 < l/\xi_0 < 0.19$ ,<sup>2)</sup> which corresponds to  $4.6 < \nu < 44$  (see columns 11 and 12 of the table), is satisfied for our samples. In this range of variation of  $\nu$ , the linear approximation  $F(\nu)$  leads to the following formula which is valid to 2% accuracy:

$$|dH_{c2}/dT|_{T=T_c} = (0.386\pi^2 kc/e) (\eta/lv_F) (1.04 + 1.61l/\xi_0). \quad (6)$$

Utilizing relations (6), (4), and

$$1/\rho_n = (2/3) e^2 N(0) l v_F \quad (7)$$

and taking into consideration that  $H_{c1} = H_{c2}$ ,<sup>[17]</sup> we obtain the following formula which is convenient for analysis of our results:

$$\eta N(0) = \rho_n^{-1} [ |dH_{c1}/dT|_{T=T_c} / 2.57 k e c - (13.2 k / \hbar e^2) (\eta T_c / v_F^2) ]. \quad (8)$$

The second term on the right-hand side of formula (8) plays the role of the correction; its relative contribution to  $\eta N(0)$  amounts to  $1.6l/\xi_0$ .

In order to calculate  $\eta N(0)$ <sup>3)</sup> according to formula (8), we shall utilize the fact that, although  $\eta$  and  $v_F$ , appearing in the correction term, are not accurately known for vanadium, the ratio  $\eta/v_F^2$  can be found from data on the critical magnetic field of pure, bulk samples with the aid of Eq. (5), where  $F(\nu) = 1.43$  as  $\nu \rightarrow 0$ . From the known experimental results we obtain  $\eta/v_F^2 = 0.319 \times 10^{-14}$ ,<sup>[18]</sup>  $0.330 \times 10^{-14}$ ,<sup>[19]</sup> and  $0.358 \times 10^{-14}$  cm<sup>-2</sup>-sec<sup>2</sup>.<sup>[20]</sup> We used the average of these values,  $\eta/v_F^2 = 0.336 \times 10^{-14}$  cm<sup>-2</sup>-sec<sup>2</sup>. On the basis of<sup>[12-16]</sup>, it is difficult to anticipate strong variations of  $\eta$  in our samples upon a reduction of the thickness; therefore, we shall assume that  $\eta$  is constant. As for  $v_F$ , we carried out the calculations for two cases:  $v_F = \text{const}$  and  $v_F = \text{const}/N(0)$ . Since  $v_F$  only enters into the correction term in formula (8), the end result is modified very little even under the assumption of strong variations of  $v_F$ .

Now let us consider the results for thick films ( $d \geq 720$  Å). First of all we note that the quantity  $\eta N(0)/N_m(0)$ ,<sup>[4]</sup> calculated according to formula (8), correlates with  $T_c(d)$  (see Figs. 3 and 1 and the table). For films of thickness  $d \geq 720$  Å, the ratio  $\eta N(0)/N_m(0)$  and the value of  $T_c$  are approximately constant, and furthermore the transition temperature is close to the value for bulk vanadium. In this range of thicknesses the difference  $\rho_{295} - \rho_n$  (see column 7 of the table) is also close to the value characteristic of bulk samples ( $19.9 \mu\text{ohm-cm}$ <sup>[21]</sup>). These facts, and also the fact that according to the electron diffraction data for films with  $d \geq 125$  Å the crystal structure and the lattice parameter coincide with the values for bulk samples, provide the foundation for the assumption that  $N(0) = N_m(0)$  for  $d \geq 720$  Å. From the values of  $\eta N(0)/N_m(0)$  found for thick films, we obtain  $\eta = 1.20 \pm 0.10$  (the average value for films of thickness  $d \geq 720$  Å).<sup>5)</sup> For purposes of comparison, we point out that a theoretical estimate  $\eta = 1.16$  is presented in

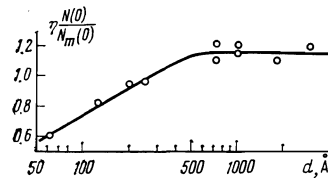


FIG. 3. Dependence of the product of the electron density of states  $N(0)$  times the strong coupling coefficient  $\eta$  on the thickness of the vanadium film (calculated according to formula (8) with  $v_F = \text{const}$ ).

article<sup>[20]</sup> by Williamson, but an estimate made according to the theoretical work of Geilikman and Kresin<sup>[12]</sup> gives  $\eta \approx 1.03$ .

Now let us go on to a discussion of the results for thin films ( $d \leq 250$  Å). By division of the obtained values  $\eta N(0)/N_m(0)$  by  $\eta = 1.20$ , we obtain the values for  $N(0)/N_m(0)$  cited in the table (columns 14 and 16). The electron density of states decreases as the thickness of the films is reduced and correlates with  $T_c$  (this is also clear from Fig. 3, if one keeps in mind that  $\eta = \text{const}$  (1.20). Such a conclusion agrees qualitatively with the assumption<sup>[3]</sup> that, strong variations of  $N(0)$ , having an influence on  $T_c$ , can be observed in transition-metal films due to a reduction in the electron mean free path. A possible mechanism,<sup>[3]</sup> associated with a narrowing of the electron energy levels upon a decrease of the mean free path, which should lead to a smoothing out of the curve  $N(\epsilon)$  which has a number of sharp maxima in the case of the transition metals, lies at the foundation of this assumption. It should be noted that the quantitative information which was found with regard to  $N(0)$  in thin films is obtained under the assumption of homogeneity of the vanadium layer of thickness  $d$ . However, a transition layer may exist near the boundaries, this layer being enriched by carbon impurities which penetrate from the protective covering. The specific resistance of such a hypothetical transition layer is probably larger than in the depths of the film. Taking account of the corrections associated with this would lead to higher values of  $N(0)$  in thin films. We still do not have available the information concerning the distribution of impurities over distances on the scale of tens of angstroms from the boundary, which is the type of information necessary to take this correction into account.

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<sup>1)</sup>We note that formula (1) is obtained under the assumption of scattering by point defects. However, the applicability of this formula can also be proved for scattering by film boundaries,<sup>[10]</sup> and for the case of fine grains, separated by thin layers of dielectric, in the limit  $l \ll \xi_0$ .<sup>[11]</sup> We shall assume that it is also applicable in the case of scattering by grain boundaries.

<sup>2)</sup>The value of  $l$  (column 10 of Table I) was estimated from the specific resistance,  $\xi_0$  was calculated according to formula (4) for  $v_F = 1.89 \times 10^7$  cm/sec. This value of  $v_F$  is obtained

from data concerning the critical magnetic field (see below).  
<sup>3</sup>Below we shall utilize the normalized quantity  $\eta N(0)/N_m(0)$ , where  $N_m(0) = 0.94 \times 10^{35}$  states-erg<sup>-1</sup>-cm<sup>-3</sup> denotes the density of electron states in pure, bulk vanadium, corresponding to a coefficient of electronic heat capacity  $\gamma_m = 9.80$  mJ-mole<sup>-1</sup>-K<sup>-2</sup>.<sup>[8]</sup>  
<sup>4</sup>The accuracy of the determination of  $\eta N(0)/N_m(0)$  amounts to  $\sim 8\%$ .  
<sup>5</sup>From  $\eta = 1.20$  and  $\eta/v_F^2 = 0.336 \times 10^{14}$  cm<sup>-2</sup> we obtain the value  $v_F = 1.89 \times 10^7$  cm/sec, which was utilized by us above in order to determine  $\xi_0$ ; see footnote 2.

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## Hydrodynamics of the A phase of superfluid <sup>3</sup>He

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Starting from the kinetic equation for the quasiparticle distribution function and from considerations of gauge invariance, a complete system of linear hydrodynamic equations for the A phase of superfluid <sup>3</sup>He is obtained. All the thermodynamic quantities and (in the  $\tau$ -approximation) kinetic coefficients appearing in the equations are found and expressed in terms of the Fermi-liquid parameters of the Landau theory. It is shown that in the range of applicability of hydrodynamics ( $\omega\tau \ll 1$ ) there exist two regions of frequency, in which the equations of motion for the orbital angular-momentum vector have a fundamentally different character. At low frequencies this equation is a diffusion equation, while at higher frequencies it describes the propagation of weakly damped orbital waves with a linear dispersion law.

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### 1. INTRODUCTION

A number of experiments carried out recently (measurements of the fourth-sound velocity and heat flows, and experiments with a vibrating wire in the liquid; cf.<sup>[1]</sup>) prove incontrovertibly that the A and B phases of liquid <sup>3</sup>He are superfluid liquids. On the other hand, magnetic-susceptibility measurements and linear and pulsed NMR make it possible to identify these phases with the so-called Anderson-Brinkman-Morel and Balian-Werthamer states (cf. the review<sup>[2]</sup>) for Cooper pairing of <sup>3</sup>He atoms in a state with angular momentum  $l=1$  and spin  $S=1$ .

The Anderson-Brinkman-Morel state, corresponding to the A phase, is characterized by the following order parameter:

$$\hat{\Delta} = i(\hat{\sigma}\mathbf{V})\hat{\sigma}_y(\mathbf{n}\Delta), \quad (1.1)$$

where  $\hat{\sigma}$  are the Pauli matrices,  $\mathbf{V}$  is a unit vector,  $\mathbf{n} = \mathbf{k}/|\mathbf{k}|$ , and  $\Delta = \Delta' + i\Delta''$  where  $\Delta'$  and  $\Delta''$  are real vectors, equal in magnitude ( $|\Delta'| = |\Delta''| = \Delta(T)$ ) and mutually perpendicular. Their vector product  $\Delta' \times \Delta'' = \Delta^2(T)\mathbf{l}$  defines the direction (common for all pairs) of the orbital angular momentum of a pair in the absence of walls or of external fields. Thus, the coordinate (or orbital) part of the order parameter is specified by the position of the triad of vectors  $\Delta'$ ,  $\Delta''$ ,  $\mathbf{l}$ . We shall assume that in equilibrium the vectors  $\Delta'$ ,  $\Delta''$ ,  $\mathbf{l}$  are directed along the axes  $X$ ,  $Y$ ,  $Z$  respectively.

We shall describe small deviations from the equilibrium position as a rotation (specified by a vector  $\theta$  equal