ON THE SUPERFLUIDITY OF A SYSTEM OF POLAR BOSE EXCITATIONS

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Submitted to JETP editor October 27, 1958

J. Exptl. Theoret. Phys. (U.S.S.R.) **36**, 1259-1266 (April, 1959)

We have investigated the possibility of a superconducting state in metals for which the elementary current excitations of the electronic system are (quasi-) bosons. This problem is solved, using Bogolyubov's method, in the framework of the "polar" many-electron model of a crystal. We have obtained criteria for a possible occurrence of a superconducting state in a system of charged bosons (low temperatures, low density of the quasi-particles, practically no "one-electron" transitions, and a negative sign of the exchange integral). The interaction between the current Bose particles, induced by the phonons, is attractive of character and opposes the appearance of superfluid properties of the latter. The critical temperature of a superconductor with boson current carriers depends differently on the isotopic mass of the crystal ions than for the case of metals with a Fermi electron spectrum. This difference can be used as the basis of an experimental method to distinguish superconductors of the "Fermi" and of the "Bose" type.

1. The latest developments in the microscopic theory of superconductivity in references 1 and 2 are connected with the assumption that the elementary current carriers obey Fermi statistics. An investigation of the interaction of these fermions with the phonons of the crystalline lattice made it possible to establish the presence of a state with the observed property of superfluidity below some critical temperature. It is, however, fully possible that there are cases where the current carriers in the system of interacting electrons in the crystal satisfy with sufficiently large accuracy Bose statistics.[3] It is in that connection of interest to study the presence of superconducting properties in a metal with a Bose-type energy spectrum of the current carriers. The idea itself of connecting the phenomenon of superconductivity with the Bose-Einstein condensation of a gas of charged bosons is not new, but a number of existing investigations in that direction (see, for instance, references 4 and 5) are, as a rule, methodic of character.[6] The reasons for this are, firstly, that the physical nature of the assumed charged bosons remains usually vague, and secondly that one considers a perfect boson gas, the thermodynamic properties of which differ from those observed in real superconductors (in particular, there is no phase transition of the second order in a perfect boson gas). These shortcomings are not present in the many-electron polar model[7,8] where, on the one hand, one does not postulate from the outset a gas of charged bosons, but obtains it in the appropriate limiting cases (predominance of the energy operator matrix elements corresponding to two-electron transitions[3]) through an analysis of the different kinds of interaction in the electron system, while, on the other hand, this gas turns out to be imperfect. It is therefore expedient to consider a system of charged polar Bose excitations, using the method applied by Bogolyubov in analyzing the properties of superfluidity of a weakly imperfect Bose-Einstein gas.[9]

2. In the case of a crystal where the energetically most favorable state is the one of maximum polarization[7,10] (the sites are either pairs or holes) while the matrix elements corresponding to two-electron transitions predominate over the matrix elements of the one-electron transitions ($|J_{qq'}| \gg |L_{qq'}|$, where $J_{qq'}$ is the integral of exchange between the orthonormal states θ_q and $\theta_{q'}$, and $L_{qq'}$ the corresponding transfer integral), the Hamiltonian of the polar model has in the second quantization representation according to (19) of reference 7 the following form:

$$\mathcal{H} = \sum_q G(q, q)\, n_q + sA + {}^1/_2 \sum_{q \neq q'} B_{qq'} n_q n_{q'} + \sum_{q \neq q'} J_{qq'} (\Phi_q \Psi_q^+ \Phi_{q'}^+ \Psi_{q'} - \Phi_q \Phi_q^+ \Phi_{q'} \Phi_{q'}^+), \quad (1)$$

where s is the number of pairs, A the energy eigenvalue of a pair, $B_{qq'}$ the integral of the mutual Coulomb repulsion of two valence elec-

trons in the states θ_q and $\theta_{q'}$, respectively, n_q the number of valence electrons in the state θ_q; G(q, q) the energy of a valence electron in an isolated atom, corrected for its interaction with the other atomic cores, Φ_q and Φ_q^+ the operators of the annihilation and the corresponding creation of a pair in the state θ_q, and Ψ_q and Ψ_q^+ the corresponding operators for holes, satisfying according to reference 3 the plus-minus quantization relations:

$$[\Phi_q\Phi_q^+]_+ = [\Psi_q\Psi_q^+]_+ = 1,$$
$$[\Phi_q\Phi_{q'}^+]_- = [\Psi_q\Psi_{q'}^+]_- = 0 \qquad (\text{if} \quad q \neq q'),$$
$$[\Phi_q\Phi_{q'}]_- = [\Psi_q\Psi_{q'}]_- = [\Phi_q^+\Phi_{q'}^+]_- = [\Psi_q^+\Psi_{q'}^+]_- = 0. \quad (2)$$

Since in the case under consideration the condition of maximum polarization:

$$\Phi_q\Phi_q^+ + \Psi_q\Psi_q^+ = 1 \quad (3)$$

is satisfied, it is expedient to introduce instead of the operators Φ_q, Φ_q^+, Ψ_q, Ψ_q^+ the operators:

$$b_q = \Psi_q\Phi_q^+, \; b_q^+ = \Phi_q\Psi_q^+, \quad (4)$$

which because of condition (3) also satisfy the commutation relations (2). The Hamiltonian (1) has in the new operators (4) the following form

$$\mathcal{H} = sA_1 + \sum_{q \neq q'} J_{qq'} b_q^+ b_{q'} + \sum_{q \neq q'} C_{qq'} b_q^+ b_q b_{q'}^+ b_{q'}, \quad (5)$$

where

$$A_1 = A + 2G(q, q) \text{ and } C_{qq'} = 2B_{qq'} - J_{qq'}. \quad (6)$$

Making in (5) the transition to wave vector space by means of the Fourier transformation

$$b_q = N^{-1/2} \sum_{\mathbf{k}} b_{\mathbf{k}} e^{i\mathbf{k}\mathbf{R}_q}, \quad b_q^+ = N^{-1/2} \sum_{\mathbf{k}} b_{\mathbf{k}}^+ e^{-i\mathbf{k}\mathbf{R}_q}, \quad (7)$$

where N is the number of lattice sites, we get

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1, \quad (8)$$

$$\mathcal{H}_0 = \sum_{\mathbf{k}} (A_1 + \sum_{\mathbf{h}} J(h) e^{i\mathbf{k}\mathbf{h}}) b_{\mathbf{k}}^+ b_{\mathbf{k}} = \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} b_{\mathbf{k}}^+ b_{\mathbf{k}}, \quad (8_1)$$

$$\mathcal{H}_1 = \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4} \delta(\mathbf{k}_2 - \mathbf{k}_1 + \mathbf{k}_4 - \mathbf{k}_3) \sum_{\mathbf{h}} C(h) e^{i\mathbf{h}(\mathbf{k}_4 - \mathbf{k}_3)} \times b_{\mathbf{k}_1}^+ b_{\mathbf{k}_2} b_{\mathbf{k}_3}^+ b_{\mathbf{k}_4}. \quad (8_2)$$

Here $\sum_{\mathbf{h}}$ denotes a sum over the radius vectors $\mathbf{h}$ performed from a fixed lattice site to the remaining lattice sites. According to reference 3, the operators $b_{\mathbf{k}}$ satisfy, when the ratio s/N is small, the commutation relations for Bose-operators approximately. If we restrict ourselves accordingly to the case* s/N ≪ 1 ("poor" metal[11]) we must interpret $\mathcal{H}_0$ as the energy operator of a system of non-interacting quasi-bosons,* and $\mathcal{H}_1$ as the energy operator of their interaction. The Hamiltonian (8), (8_1), (8_2) has basically the same form as the Hamiltonian (3.63), (3.64) considered in reference 9. Following, accordingly, the reasoning given there we find in the first perturbation theory approximation

$$E\ldots n_{\mathbf{k}} \ldots = s\left[A_1 - \frac{1}{N}\sum_h C(h)\right] + \frac{s^2}{N}\sum_h C(h) + \sum_{\mathbf{k}} n_{\mathbf{k}} \sum_h J(h) e^{i\mathbf{k}\mathbf{h}} + \frac{1}{N} \sum_{\mathbf{k}_1 \neq \mathbf{k}_2} n_{\mathbf{k}_1} n_{\mathbf{k}_2} \sum_h C(h) e^{i\mathbf{h}(\mathbf{k}_1 - \mathbf{k}_2)}, \quad (9)$$

where

$$n_{\mathbf{k}} = b_{\mathbf{k}}^+ b_{\mathbf{k}}.$$

We find now the condition that the lowest level is the one with occupation numbers

$$n_{\mathbf{k}} = \begin{cases} s & \text{if} \quad \mathbf{k} = 0 \\ 0 & \text{if} \quad \mathbf{k} \neq 0, \end{cases} \quad (10)$$

i.e., the level

$$E_0 = s\left[A_1 - \frac{1}{N}\sum_h C(h)\right] + \frac{s^2}{N}\sum_h C(h) + s\sum_h J(h). \quad (11)$$

From (9) and (11) we find, if we use the nearest neighbor approximation and also restrict ourselves to considering the low temperature region, where ka ≪ 1,[11]

$$E\ldots n_{\mathbf{k}} \ldots - E_0 = -J \sum_{\mathbf{k}} n_{\mathbf{k}} (ka)^2 + \frac{C}{N} \sum_{\mathbf{k}_1 \neq \mathbf{k}_2} n_{\mathbf{k}_1} n_{\mathbf{k}_2} [6 - (\mathbf{k}_1 - \mathbf{k}_2)^2 a^2]. \quad (12)$$

The expression within the square brackets is here positive by virtue of the condition ka ≪ 1. Moreover, C = 2B − J is also positive since B does not contain overlaps of the functions θ_q, while J contains two such overlaps. The second sum on the right hand side of (12) is thus positive. The first term on the right hand side of (12) is positive, if J < 0. If, however, J > 0, this term is negative and this coupled with the fact that in the case under consideration the interaction energy is assumed to be small compared to the kinetic energy, means that the right hand side of (12) will be negative. It is thus necessary that the exchange integral J be negative, if the energetically lowest state is to be the one for which all quasiparticles have k = 0, i.e., one for which condensation takes place.

*If there are only pairs and holes, (i.e., when the polarization is a maximum) the condition s/N ≪ 1 can be satisfied if the number of electrons 2s is less than the number of lattice sites N, which shall be assumed to be the case in the following.

*We use in this case the term "quasi-bosons" to emphasize once again that for s/N ≪ 1 the quasiparticles behave with a high degree of accuracy, but even so still only approximately, as bosons.

Restricting ourselves, therefore, in the following to the case $J < 0$, we have from (8_1)

$$\varepsilon_k = A_1 - 6|J| + p_k^2/2m_{\text{eff}}, \tag{13}$$

where

$$m_{\text{eff}} = \hbar^2/2|J|a^2. \tag{14}$$

We find now the energy necessary to take one quasi-particle from the system of quasi-particles moving all with the same wave vector $\mathbf{k}$. To do this, we use (9) and consider the difference ΔE between the energy $E_{\mathbf{k}_0}$ of the system moving with the wave vector $\mathbf{k}_0$ and the energy $E_{\mathbf{k}_0-\mathbf{g}}$ of a system of $(s-1)$ particles with wave vector $\mathbf{k}_0$ and one particle with wave vector $\mathbf{k}_0 - \mathbf{g}$:

$$\Delta E = E_{\mathbf{k}_0-\mathbf{g}} - E_{\mathbf{k}_0} \approx \sum_{\mathbf{h}} |J(h)| e^{i\mathbf{k}_0\mathbf{h}} - \sum_{\mathbf{h}} |J(h)| e^{i(\mathbf{k}_0-\mathbf{g})\mathbf{h}} + \frac{2s}{N}\sum_{\mathbf{h}} C(h)\cos(\mathbf{g},\mathbf{h}). \tag{15}$$

In the last term of (15) we have dropped 1 compared to s. Using the nearest neighbor approximation and (14), we can transform (15) to the form

$$\Delta E = E(\mathbf{g}) - (\mathbf{p}_g\mathbf{v}_0), \tag{16}$$

where

$$E(\mathbf{g}) = p_g^2/2m_{\text{eff}} + (2s/N)\sum_{\mathbf{h}} C(h)\cos(\mathbf{g},\mathbf{h}); \tag{17}$$

$$\mathbf{v}_0 = \mathbf{p}_0/m_{\text{eff}} \tag{18}$$

is the average velocity of the condensate corresponding to a quasi-momentum $\mathbf{p}_0 = \hbar\mathbf{k}_0$. In the nearest-neighbor approximation the ratio $E(\mathbf{g})/g$ has, according to (17), a minimum at the point:

$$(g_0a)^2 = 1/(N|J|/12sC - 1/6). \tag{19}$$

In order that this minimum occur in the region where the low temperature condition $ga \ll 1$ is satisfied, the inequality

$$s/N \ll |J|/2C, \tag{20}$$

must thus be satisfied, and this is the case, if the condition $s/N \ll 1$ is obeyed, which is used, anyway, in the present paper.

We introduce now the notation

$$E(\mathbf{g}_0)/g_0 = \hbar u, \tag{21}$$

whence

$$E(\mathbf{g}) \geqslant \hbar u g. \tag{22}$$

From (16) and (22) we find

$$\Delta E \geqslant p_g[u - v_0\cos(\mathbf{p}_g, \mathbf{v}_0)], \tag{23}$$

or, since the maximum of cosine $(\mathbf{p_g}, \mathbf{v}_0) = 1$, we have

$$\Delta E \geqslant (u - v_0)p_g. \tag{24}$$

From this it follows that if $v_0 < u$ an excitation of the moving condensate of pairs is energetically unfavorable.

Applying Bogolyubov's method to the real Bose gas of the quasi-particles of the polar model we are thus able to establish the occurrence of the property of superfluidity of the electron system of the crystal, if the following conditions are satisfied: low temperatures $(ka \ll 1)$, poor metals $(s/N \ll 1)$, practically no one-electron transitions, and a negative exchange integral (positive effective mass).

3. The applicability of the results of the preceding section are, strictly speaking, restricted to the region of sufficiently large k (and satisfying at the same time the condition $ka \ll 1$),* when it has sense to use perturbation theory, assuming the interaction energy to be small compared to the kinetic energy. One can convince oneself of this by applying to the problem considered above the method of approximate second quantization.[9] Taking into account, namely, the fact that the main part of the quasi-particles is in the $k = 0$ state, and dropping accordingly in (8_2) the terms with triple and quadruple products of the b_k (for $k \neq 0$) one can transform the Hamiltonian of the system under consideration to the form:

$$\mathcal{H} = (s^2/N)\sum_{\mathbf{h}} C(h) + (A_1 - 6|J|)s + \sum_{\mathbf{k}} (|J|k^2a^2 + \frac{2n_0}{N}\sum_{\mathbf{h}} C(h)e^{i\mathbf{hk}})b_{\mathbf{k}}^+b_{\mathbf{k}} + (b_0^2/N) \times \sum_{\mathbf{k},\mathbf{h}} C(h)e^{i\mathbf{hk}}b_{\mathbf{k}}^+b_{-\mathbf{k}}^+ + (b_0^{+2}/N)\sum_{\mathbf{k},\mathbf{h}} C(h)e^{i\mathbf{hk}}b_{\mathbf{k}}b_{-\mathbf{k}}, \tag{25}$$

where n_0 is the occupation number of the $k = 0$ state.

Now making the transition to the new Bose operators[9]

$$\beta_{\mathbf{k}} = b_0^+n_0^{-1/2}b_{\mathbf{k}},\quad \beta_{\mathbf{k}}^+ = b_0n_0^{-1/2}b_{\mathbf{k}}^+ \tag{26}$$

and carrying out the canonical transformation[9]

$$\beta_{\mathbf{k}} = (\xi_{\mathbf{k}} + A_k\xi_{-\mathbf{k}}^+)/\sqrt{1 - A_k^2},$$
$$\beta_{\mathbf{k}}^+ = (\xi_{\mathbf{k}}^+ + A_{\mathbf{k}}\xi_{-\mathbf{k}})/\sqrt{1 - A_{\mathbf{k}}^2}, \tag{27}$$

*From (29) it is clear that one can consider k, to be "large," Σ if it satisfies the condition $ka \gg 2(n_0/N|J|\sum_{\mathbf{h}} C(h)e^{i\mathbf{kh}})^{1/2}$. The region of "large" k which do not violate the condition $ka \ll 1$ satisfies thus the inequalities $1 \gg ka \gg 2(n_0/N|J|\sum_{\mathbf{h}} C(h)e^{i\mathbf{kh}})^{1/4}$. Since $n_0/N < s/N \ll 1$, and $\sum_{\mathbf{h}} C(h)e^{i\mathbf{kh}}$ is of the same order of magnitude as $|J|$, such a region clearly exists.

where

$$A_k = (N/2n_0 \sum_h C(h) e^{ikh}) [E(k) - |J| k^2 a^2 + \frac{2n_0}{N} \sum_h C(h) e^{ikh}], \quad (28)$$

one can verify that the Hamiltonian (25) is diagonalized, while one obtains for the energy of the quasi-particles the expression:

$$E(k) = \left[\frac{4n_0}{N} \sum_h C(h) e^{ikh} |J| k^2 a^2 + J^2 k^4 a^4\right]^{1/2}. \quad (29)$$

One sees from this easily that the result (17), obtained in Sec. 2 by perturbation-theory methods, is obtained for large values of k.

In the region of small k it follows from (29):

$$E(k) \approx 2\left[\frac{n_0}{N} \sum_h C(h) e^{ikh} |J|\right]^{1/2} ak, \quad (30)$$

i.e., one obtains phonon-type ($E \sim k$) excitations which leads also, as is well known, to the presence of the superfluidity property in a system of bosons.[9,12]

4. Since the method of approximate second quantization and the application of perturbation theory lead to the identical result for large k, we can take expression (21) for the critical velocity u.* Since the left hand side of (21) depends, according to (17) and (19), only on J, C, a, and s/N, it follows from this that in the case under consideration the critical velocity u does not depend on the isotopic mass M (if we neglect the possible, very weak dependence of the lattice constant a on M). In that respect a gas of bosons differs from a gas of fermions where the critical velocity of superfluidity is determined from the relation $u = \tilde{\omega} e^{-1/\rho} k_F^{-1}$, according to reference 1, and thus depends on the ionic mass according to a $u \sim M^{1/2}$ law, through $\tilde{\omega}$. It is in this connection of interest to elucidate in how far the results obtained above are influenced by taking the interaction of the gas of bosons with the vibrations of the crystalline lattice into account. Putting

$$J_{qq'} = J^0_{qq'} + (\Delta R_q - \Delta R_{q'}) (\text{grad } J_{qq'})_0 + \ldots, \quad (31)$$

where the index 0 indicates the equilibrium values of quantities, and expanding the displacement ΔR_q of the g-th lattice site, caused by the temperature vibrations, from its equilibrium position in plane waves

$$\Delta R_q = \frac{1}{\sqrt{N}} \sum_{f_j} p_{f_j} [\xi_{f_j} e^{i(f_j R_q)} + \xi^+_{f_j} e^{-i(f_j R_q)}], \quad (32)$$

*The arguments given below are also applicable, if we begin the determination of u with (30).

where p_{f_j} is the unit polarization vector and f_j the wave vector, one can separate from (5) the following part of the Hamiltonian, which describes the interaction of the bosons with the lattice vibrations:[11]

$$H_{ph} = g \sum_{(k', f \atop k'-k=f)} i \sqrt{\frac{\hbar\omega_f}{2V}} b^+_k b_{k'} \alpha^+_f - g \sum_{(k, f \atop k-k'=f)} i \sqrt{\frac{\hbar\omega_f}{2V}} b^+_k b_{k''} \alpha_f, \quad (33)$$

where the α_f are the Bose operators for the phonons, V the volume of the system, and where the coupling constant $g = 2\pi J a^{3/2}/3v\sqrt{M}$ does not depend on M, since the sound velocity $v \sim M^{-1/2}$.

If we now apply Fröhlich's canonical transformation[13] we can separate a boson-boson interaction caused by the phonons of the form:

$$H_{b(ph)} = -\frac{1}{2} g^2 \sum_{f, k, k'} \frac{\hbar\omega_f}{2V} \frac{(1 + \Delta(k, f))(1 - \Delta(k, f))}{\varepsilon_{k'-f} - \varepsilon_{k'} + \hbar\omega_f} b^+_k b_{k-f} b^+_{k'-f} b_{k'} + \text{c.c.}, \quad (34)$$

where Δ has the same meaning as in reference 13. We note there that although an expression of the form (34) was obtained by Fröhlich* for fermions, it retains its validity also for bosons, since in deriving (34) the commutation relations for the quasi-particle operators were in reference 13 only used to obtain the relation (2.26):

$$a^+_k a_l a^+_q a_r - a^+_q a_r a^+_k a_l = \delta_{lq} a^+_k a_r - \delta_{kr} a^+_q a_l,$$

and this relation retains its validity, as can easily be checked, also for Bose operators.

It is clear from (34) that if $|\varepsilon_{k'-f} - \varepsilon_{k'}| < \hbar\omega_f$ the interaction induced by the phonons is attractive in nature and impedes thus the establishment of the superfluidity property in a system of bosons. H_b (ph) does here not depend on M, since in (34) there is a cancellation of ω in the numerator and the denominator.

In this respect there exists a similarity with the case of fermions where the interaction induced by the phonons is also by itself independent of M.[2] As far as the dependence on M of the critical velocity and of the critical temperature (isotope effect) is concerned, they occur in the case of fermions because the region of the effective attraction induced by the phonons is restricted to

*The interaction (34) was introduced by Fröhlich[13] without taking Coulomb forces into account, but it is also valid when they are taken into account, if we understand by g the "screened" coupling constant.[14]

the region $\hbar\omega$. In the case of bosons, however, when it is essential for the occurrence of superfluidity that the repulsion dominates over the attraction, a restriction of the consideration to the region of the effective phonon attraction is clearly not necessary. It follows that, although in this case the attraction induced by the phonons decreases the magnitude of u (through the effective decrease of the magnitude of C which, by the way, improves the applicability of perturbation theory methods, expounded in Sec. 2), the dependence of u on M is, apparently, in the case of bosons appreciably weaker than in the case of fermions.

To conclude this section we note that the above-mentioned restriction on the region of the effectiveness of the attraction induced by the phonons leads in the case of fermions not only to a dependence of different characteristics of the superconducting state on M, but also introduces according to Bogolyubov's theory[1] (in contradistinction to the theory of Bardeen, Cooper, and Schrieffer[2]) a dependence of the criterion for superconductivity itself on M. The Bogolyubov-Tolmachev criterion, $\rho > \rho_C [1 + \rho_C \ln(\tilde{E}_F/\tilde{\omega})]^{-1}$ depends, namely, on $\tilde{\omega}$ and thus on M, while the Bardeen, Cooper, and Schrieffer criterion $\rho > \rho_C$ does not depend on M. Thus, if one isotope of some metal is superconductive, all other isotopes of this metal will, according to the Bardeen, Cooper, and Schrieffer criterion, also possess the superconducting property. According to the Bogolyubov-Tolmachev criterion, however, it is possible in principle that the heavy isotopes of a given metal possess the superconducting property, while its light isotopes (having a relatively larger value of $\tilde{\omega}$) will not possess this property. It is accordingly possible that when we go to lighter isotopes of superconducting metals there is displayed a disappearance of the superconducting property itself, instead of an increase of T_{cr} following from the $T_{cr} \sim M^{-1/2}$ dependence. Such an effect, if it does occur, will be superficially analogous to the disappearance of the superfluidity observed when one goes from He_4 to the light isotope He_3. It is necessary, however, to note that in this case the disappearance of superfluidity is usually explained not through the lowering of M, but through the different statistics (He_4: bosons; He_3: fermions).

5. What we have stated above leads to the conclusion that the occurrence in a given metal of the superconducting property is apparently not yet a sufficient indication for a purely Fermi character of the energy spectrum of its electron system. It is fully possible that there are superconductors where the current carriers behave with sufficient accuracy as bosons. In the latter case, the dependence of the critical quantities on M must be appreciably weaker than in the case of fermions. Apart from that, the temperature dependence of the transport coefficients in the ground state of a metal with Bose current carriers must be different from that of a metal with Fermi current carriers.[11] These differences between Bose and Fermi conductors enable us to expect that a combined study of different properties of superconductors both above and below the critical temperature ($T_{cr} \sim M^{-1/2}$ dependence) should give us the possibility to solve experimentally the problem of whether the energy spectrum of every real superconductor belongs to the Bose or to the Fermi type.

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Translated by D. ter Haar
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