

# Superconductivity in Semiconductors

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A model of a degenerate polar semiconductor possessing a low-lying transverse optical phonon branch is considered. For impurity concentrations sufficient to obtain a high-density carrier gas, the semiconductor goes over into the superconducting state at sufficiently low temperatures. The peculiarly strong (with respect to the parameter  $\epsilon_0$ ) interaction between the electrons and the phonons belonging to the low-lying branch is specifically responsible for the superconductivity.

THE possibility of the transition of degenerate semiconductors into the superconducting state was theoretically predicted<sup>[1,2]</sup> long before such a transition was observed experimentally. As was first shown by Gurevich, Larkin, and Firsov,<sup>[1]</sup> in polar semiconductors the attraction between electrons near the Fermi surface may in principle exceed their Coulomb repulsion. In order for this to occur, the free electrons must form a degenerate gas of high density inside the semiconductor, and this gas interacts with the longitudinal long-wavelength optical phonons. A high density of the electron gas can be achieved at small impurity concentrations  $n \gtrsim 10^{19} \text{ cm}^{-3}$ , and the attainment of this situation is aided by the large value of the dielectric constant  $\epsilon_\infty$  and by the smallness of the effective mass in the filled band. It is assumed that the electron states fill the single valley at the center of the Brillouin zone. The threshold (with respect to the impurity concentration) for the appearance of the superconducting transition lies in a range of Fermi energies  $\epsilon_F \gtrsim \omega_l$  which is lower the higher the value of the static dielectric constant  $\epsilon_0$  (here  $\omega_l$  is the frequency of the longitudinal optical phonons). The superconducting transition temperature  $T_c$  falls as the impurity concentration increases.

Cohen proposed<sup>[2]</sup> and developed<sup>[3,4]</sup> an application of the BCS model to semiconductors having a many-valley band structure. It was proposed to describe the electron-phonon coupling in terms of deformation potentials. In this connection, numerical calculations have shown that the interaction of the electrons with all branches of the phonon spectrum, but not with any specific type of phonon, introduces a contribution to the kernel of the integral equation for the superconducting gap. As a consequence of such an interaction, the electrons are primarily scattered between different valleys. The effectiveness of the intervalley scattering is due to the large density of states and the relatively weak screening of scattering processes involving large momentum transfer.

In the present article we consider a model of a degenerate semiconductor under the conditions close to the ferroelectric phase transition. The structure of the electron spectrum is not very important for what follows, and for simplicity we confine ourselves to a single valley. In such "pseudo-ferroelectrics" the dielectric constant may increase to very large values as the temperature decreases. In this connection, there is a transverse branch in the optical phonon spectrum which has an anomalously small gap  $\omega_0$  at low temperatures.<sup>[5]</sup>

Under these conditions the mechanism responsible for the superconductivity is not the polarization interaction of the electrons with longitudinal phonons, as it was in<sup>[1]</sup>, but the peculiarly strong (with respect to the parameter  $\epsilon_0$ ) scattering by phonons belonging to the low-lying transverse optical branch.

## 1. EFFECTIVE ELECTRON INTERACTION

Let us discuss the specific properties of the electron-phonon interaction close to the ferroelectric transition, and let us derive the effective potential of the two-particle interaction. We shall express the coupling constant in terms of observable macroscopic quantities. In order to be definite, we shall discuss the case of conduction electrons, but all of the results are equally applicable to p-semiconductors.

In the field of a lattice of point ions the potential energy  $\varphi(\mathbf{r})$  of a conduction electron is of the form

$$\varphi(\mathbf{r}) = -\frac{4\pi e i}{\sqrt{N}} \sum_{j\mathbf{K}} \frac{P_\alpha^j(\mathbf{q})(q_\alpha + K_\alpha)}{|\mathbf{q} + \mathbf{K}|^2} \xi_j(\mathbf{q}) \exp\{i(\mathbf{q} + \mathbf{K})\mathbf{r}\}, \quad (1)$$

where the  $\xi_j(\mathbf{q})$  are the normal coordinates of the  $j$ -th branch of the lattice vibrations with wave vector  $\mathbf{q}$ ,  $N$  is the number of elementary cells in the crystal volume  $V$ ,  $e$  is the electron charge, and  $\mathbf{K}$  is the reciprocal lattice vector. The polarization amplitude  $\mathbf{P}^j(\mathbf{q})$  of the branch  $j$  is related to the eigenvectors  $\mathbf{e}_j$  by the harmonic dynamical matrix:

$$\mathbf{P}^j(\mathbf{q}) = v_0^{-1} \sum_s \frac{z_s}{m_s^{1/2}} \mathbf{e}_j(s, \mathbf{q}). \quad (2)$$

In Eq. (2) the summation runs over the lattice basis,  $z_s$  and  $m_s$  denote, respectively, the effective charge and mass of the ion in the  $s$ -th sublattice, and  $v_0$  denotes the volume of an elementary cell.

At the temperature  $T$  the effective electron-electron interaction is described by the two-particle potential

$$D(\mathbf{r} - \mathbf{r}', \tau) = V_c(\mathbf{r} - \mathbf{r}') + \sum_{\mathbf{Q}} \Phi(\mathbf{Q}, \tau) e^{i\mathbf{Q}(\mathbf{r} - \mathbf{r}')}, \quad (3)$$

where we have introduced the function

$$\Phi(\mathbf{Q}, \tau) = -\frac{16\pi^2 e^2}{N} \sum_j P_\alpha^j(\mathbf{Q}) P_\beta^j(-\mathbf{Q}) \frac{Q_\alpha Q_\beta}{|Q|^4} \langle \hat{T} \xi_j(\mathbf{Q}, \tau) \xi_j(-\mathbf{Q}, \tau) \rangle \quad (4)$$

of the variables  $\mathbf{Q} = \mathbf{q} + \mathbf{K}$  and the imaginary time  $\tau$ , which appears in the temperature diagram technique;<sup>[6]</sup> here  $V_c(\mathbf{r})$  denotes the Coulomb interaction energy of the electrons. In what follows it is sufficient to assume<sup>[1]</sup> that the contribution of the free electrons to the dielectric constant leads to screening at the Debye radius  $\kappa^{-1}$ .

For simplicity let us confine our attention to the case of a diatomic cubic crystal and let us assume that, to a high degree of accuracy, one can divide the lattice vibrations into longitudinal and transverse.

As is well-known,<sup>[7]</sup> the dipole-dipole interaction leads to the result that the tensor  $\chi_{\alpha\beta}^{-1}(\mathbf{q}) + 4\pi q_{\alpha}q_{\beta}/q^2$  plays the role of the reciprocal of the dielectric permittivity of the lattice in the long wavelength region. With this fact taken into account, we arrive at the following relations by expressing the dielectric constant  $\epsilon(\mathbf{q}, \omega)$  of a cubic crystal in terms of the root-mean-square displacements of the ions:

$$\begin{aligned} \frac{P_{\alpha}^{\prime}(\mathbf{q})P_{\beta}^{\prime}(-\mathbf{q})}{\omega_i^2(\mathbf{q}) - \omega^2} &= \frac{\delta_{\alpha\beta}}{4\pi\nu_0} (\epsilon_{\infty}^{-1} - \epsilon^{-1}(\mathbf{q}, \omega)), \\ \frac{P_{\alpha}^{\prime}(\mathbf{q})P_{\beta}^{\prime}(-\mathbf{q})}{\omega_i^2(\mathbf{q}) - \omega^2} &= \frac{\delta_{\alpha\beta}}{4\pi\nu_0} [\epsilon(\mathbf{q}, \omega) - \epsilon_{\infty}]. \end{aligned} \quad (5)$$

Hence

$$\begin{aligned} P_{\alpha}^{\prime}(\mathbf{q})P_{\beta}^{\prime}(-\mathbf{q}) &= \frac{\delta_{\alpha\beta}}{4\pi\nu_0} \omega_i^2(\mathbf{q}) [\epsilon_{\infty}^{-1} - \epsilon_0^{-1}(\mathbf{q})], \\ P_{\alpha}^{\prime}(\mathbf{q})P_{\beta}^{\prime}(-\mathbf{q}) &= \frac{\delta_{\alpha\beta}}{4\pi\nu_0} \omega_i^2(\mathbf{q}) [\epsilon_0(\mathbf{q}) - \epsilon_{\infty}]. \end{aligned} \quad (6)$$

The representation (6) for the polarization amplitudes in terms of observable macroscopic characteristics was utilized in<sup>[8]</sup> in the problem of the electron mobility in the neighborhood of the ferroelectric transition.

Only the optical branches of the phonon spectrum are taken into consideration in Eqs. (5) and (6). The contribution of the acoustic degrees of freedom to the sum of the oscillator strengths is small for  $q \ll K$ . This smallness is related to the electroneutrality of the cell, as a consequence of which the terms of zero order in the expansion of the acoustic polarization amplitudes in powers of  $q$  turn out to vanish. We note that the well-known Lyddane-Sachs-Teller relation follows from Eqs. (5) and (6)

$$\omega_i^2(\mathbf{q}) / \omega_t^2(\mathbf{q}) = \epsilon_0(\mathbf{q}) / \epsilon_{\infty}. \quad (7)$$

In accordance with formulas (6), let us represent the function (4) in the form

$$\begin{aligned} \Phi(\mathbf{Q}, \tau) &= \Phi_l(\mathbf{Q}, \tau) + \Phi_t(\mathbf{Q}, \tau), \\ \Phi_l(\mathbf{Q}, \tau) &= -\frac{4\pi e^2}{V} \frac{\omega_i^2}{|\mathbf{Q}|^2} [\epsilon_{\infty}^{-1} - \epsilon_0^{-1}(\mathbf{q})] \langle \hat{T} \xi_i(\mathbf{q}, \tau) \xi_i(-\mathbf{q}, \tau) \rangle, \\ \Phi_t(\mathbf{Q}, \tau) &= -\frac{4\pi e^2}{V} \frac{\omega_i^2(\mathbf{q})}{|\mathbf{Q}|^2} [\epsilon_0(\mathbf{q}) - \epsilon_{\infty}] \\ &\quad \times \langle \hat{T} \xi_i(\mathbf{q}, \tau) \xi_i(-\mathbf{q}, \tau) \rangle (1 - \delta_{\mathbf{q}, 0}). \end{aligned} \quad (8)$$

Here one can, as usual, regard the longitudinal optical phonons as dispersionless. The dependence of the transverse phonon frequency  $\omega_t$  and of the dielectric constant  $\epsilon_0$  on the wave vector should, in general, be taken into account, keeping in mind the anomalous smallness of  $\omega_0$  in the dispersion law:

$$\omega_i^2(\mathbf{q}) = \omega_0^2 + sq^2. \quad (9)$$

The principal term in the longitudinal part of the potential (3) is the one containing the function  $\Phi_l$  with  $\mathbf{K} = 0$ . It describes the Fröhlich interaction of an electron with the macroscopic polarization field of the lattice. The terms with  $\mathbf{K} \neq 0$  represent small corrections and can be neglected.

The transverse degrees of freedom do not give any contribution to the macroscopic field. Therefore  $\Phi_t$  always corresponds to scattering with  $\mathbf{K} \neq 0$ . Such a

deformation contribution is usually small in comparison with the scattering by the longitudinal polarization potential. However, in the ferroelectric case, when the value of  $\epsilon_0$  is large, it is precisely the scattering of electrons by the deformations created by the vibrations in the low-lying transverse optical branch which is the major effect.

Let us define the momentum representation of the two-particle potential  $D$  on electron Bloch functions  $|k\rangle$  of the conduction band (one can neglect the interband matrix elements of  $D$ ). The interaction vertex, at which two electron lines and a single  $D$ -line come together, corresponds to the matrix element

$$C_{\mathbf{k}, \mathbf{k}_1}(\mathbf{K}) = \langle k | \exp \{i(\mathbf{k} - \mathbf{k}_1 + \mathbf{K})\mathbf{r}\} | \mathbf{k}_1 \rangle, \quad (10)$$

which, for values of  $\mathbf{k}, \mathbf{k}_1 \sim \mathbf{p}_0$  ( $\mathbf{p}_0$  is the Fermi momentum), we shall assume to be an isotropic constant smaller than unity. The values of these constants are different for polarization ( $\mathbf{K} = 0$ ) and deformation ( $\mathbf{K} \neq 0$ ) processes. Let us denote these values by  $C_a$  and  $C_b$ , respectively.

The Green's function of the electrons in the momentum representation can now be determined according to the standard rules of the diagram technique,<sup>[9]</sup> where the line representing the two-particle interaction corresponds to the function

$$\begin{aligned} D(\mathbf{q}, \omega_n) &= \frac{4\pi e^2}{\epsilon_{\infty}} \frac{|C_a|^2}{q^2 + \kappa^2} \left[ 1 - (\epsilon_{\infty}^{-1} - \epsilon_0^{-1}(\mathbf{q})) \frac{\omega_i^2}{\omega_i^2 + \omega_n^2} \right] \\ &\quad - \frac{4\pi e^2}{K^2} |C_b|^2 \epsilon_0(\mathbf{q}) \frac{\omega_i^2(\mathbf{q})}{\omega_i^2(\mathbf{q}) + \omega_n^2}. \end{aligned} \quad (11)$$

Here  $\omega_n = 2\pi nT$  are the Bose frequencies which appear in the temperature technique. The first nonvanishing terms of the expansion in powers of the reciprocal lattice vectors have been kept on the right-hand side of Eq. (11). It has also been taken into consideration that  $\epsilon_{\infty}$  will be very much smaller than  $\epsilon_0(\mathbf{q})$  for  $q \ll K$ , on account of the Lyddane-Sachs-Teller relationship, provided  $\omega_t(\mathbf{q}) \ll \omega_l$ .

## 2. THE WIDTH OF THE SUPERCONDUCTING GAP

At  $T = 0$  the magnitude of the superconducting gap is determined by solving Eliashberg's integral equation<sup>[9]</sup> with the kernel given by expression (11):

$$\begin{aligned} \Delta(\omega) &= -\frac{m}{4\pi^2 p_0} \int_{\Delta}^{\infty} d\omega_1 \frac{\Delta(\omega_1)}{\sqrt{\omega_1^2 - \Delta^2}} \int_0^{2p_0} q dq \left\{ \frac{4\pi e^2}{\epsilon_{\infty}} \frac{|C_a|^2}{q^2 + \kappa^2} \right. \\ &\quad \times \left[ 1 - (\epsilon_{\infty}^{-1} - \epsilon_0^{-1}(\mathbf{q})) \frac{\omega_i}{2} \left( \frac{1}{\omega_1 + \omega_i - \omega - i\delta} \right. \right. \\ &\quad \left. \left. + \frac{1}{\omega_1 + \omega_i + \omega - i\delta} \right) \right] - \frac{4\pi e^2}{K^2} |C_b|^2 \epsilon_0(\mathbf{q}) \frac{\omega_i(\mathbf{q})}{2} \\ &\quad \left. \times \left( \frac{1}{\omega_1 + \omega_i(\mathbf{q}) - \omega - i\delta} + \frac{1}{\omega_1 + \omega_i(\mathbf{q}) + \omega - i\delta} \right) \right\}. \end{aligned} \quad (12)$$

First let us consider the case of small impurity concentrations when

$$sp_0^2 \ll \omega_0^2. \quad (13)$$

As the estimates show for a substance having the parameters characteristic, for example, of SnTe, the gas parameter  $r_g = (9\pi/4)^{1/3} e^2 / \epsilon_{\infty} v_F$  becomes small even in the region (13). This implies that it is possible to obtain a free electron gas of high density in this range of concentrations. Neglecting the dispersion of  $\omega_t$  and  $\epsilon_0$  over the entire range of integration over  $q$ , correct to within

terms of logarithmic order one can write Eq. (12) in the form

$$\begin{aligned} \Delta(\omega) = & -\alpha \int_{\Delta}^{\epsilon_F} \Delta(\omega_i) \frac{d\omega_i}{\omega_i} + \alpha \frac{\epsilon_\infty}{\epsilon_c} \int_{\Delta}^{\omega_i} \Delta(\omega_i) \frac{\omega_i}{2\omega_i} \left[ \frac{1}{\omega_i + \omega_i - \omega} \right. \\ & \left. + \frac{1}{\omega_i + \omega_i + \omega} \right] d\omega_i + \alpha_i \int_{\Delta}^{\omega_i} \Delta(\omega_i) \frac{\omega_i}{2\omega_i} \\ & \times \left[ \frac{1}{\omega_i + \omega_0 - \omega} + \frac{1}{\omega_i + \omega_0 + \omega} \right] d\omega_i, \end{aligned} \quad (14)$$

where

$$\alpha = \frac{e^2 |C_a|^2}{2\pi\epsilon_\infty v_F} \ln \left( \frac{2p_0}{\kappa} \right)^2, \quad \alpha_i = \frac{e^2 |C_b|^2}{2\pi v_F} \left( \frac{2p_0}{K} \right)^2 \epsilon_0, \quad (15)$$

$$\epsilon_c^{-1} = \epsilon_\infty^{-1} - \epsilon_0^{-1}.$$

Let us seek the solution of Eq. (14) in the form

$$\begin{aligned} \Delta(\omega) = & -\Delta_1 \alpha_i \ln \left( \frac{\omega_i}{\omega_0} \right) \frac{\omega_0^2}{\omega_0^2 - \omega^2} + \Delta_2 \left[ 1 - \left( 1 + \alpha \ln \frac{\epsilon_F}{\omega_i} \right) \right. \\ & \left. \times \left( \frac{\epsilon_\infty}{\epsilon_c} \frac{\omega_i^2}{\omega_i^2 - \omega^2} + \frac{\alpha_i}{\alpha} \frac{\omega_0^2}{\omega_0^2 - \omega^2} \right) \right]. \end{aligned} \quad (16)$$

Within the frequency intervals  $\Delta < \omega < \omega_0$ ,  $\omega_0 < \omega < \omega_l$ , and  $\omega_l < \omega < \epsilon_F$  one can set  $\Delta(\omega)$  equal (correct to within terms of logarithmic order) respectively to the constants  $\Delta$ ,  $\Delta_1$ , and  $\Delta_2$  given by the following equations:

$$\begin{aligned} \Delta = & -\Delta_1 \alpha_i \ln \frac{\omega_i}{\omega_0} + \Delta_2 \left[ 1 - \left( \frac{\epsilon_\infty}{\epsilon_c} + \frac{\alpha_i}{\alpha} \right) \left( 1 + \alpha \ln \frac{\epsilon_F}{\omega_i} \right) \right], \\ \Delta_1 = & \Delta_2 \left[ 1 - \frac{\epsilon_\infty}{\epsilon_c} \left( 1 + \alpha \ln \frac{\epsilon_F}{\omega_i} \right) \right], \\ \Delta_2 = & -\alpha \left( \Delta \ln \frac{\omega_0}{\Delta} + \Delta_1 \ln \frac{\omega_i}{\omega_0} + \Delta_2 \ln \frac{\epsilon_F}{\omega_0} \right). \end{aligned} \quad (17)$$

The condition for the solvability of the system of equations (17) leads to the relation

$$\Delta = \omega_0 \exp \{-1 / (\alpha_i + \alpha)\}, \quad (18)$$

where

$$\alpha_i = \alpha \frac{\epsilon_\infty / \epsilon_c - (1 + \alpha \ln \epsilon_F / \omega_i)^{-1}}{1 - [\epsilon_\infty / \epsilon_c - (1 + \alpha \ln \epsilon_F / \omega_i)^{-1}] \ln(\omega_i / \omega_0)}. \quad (19)$$

In "pseudo-ferroelectrics"  $\alpha_i \gg \alpha_l$  and the superconductivity is due to the strong coupling between the electrons and the transverse optical phonons.

In the case of large impurity concentrations when

$$sp_0^2 \gg \omega_0^2, \quad (20)$$

it is necessary to take the dispersion of the phonon spectrum into consideration in Eq. (12). As a result  $\Delta$  turns out to be given by

$$\Delta = 2s^{1/2} p_0 \exp(-1 / \beta_i), \quad (21)$$

where

$$\beta_i = \alpha_i \left( \frac{\omega_0^2}{2s^{1/2} p_0} \right)^2 \ln \left( \frac{2s^{1/2} p_0}{\omega_0} \right)^2 \ll \alpha_i. \quad (22)$$

Thus, the inclusion of dispersion effectively reduces the coupling constant. Therefore, on the curve  $T_c(n)$  of the transition temperature as a function of the impurity concentration there is an ascending branch corresponding to densities  $r_s \ll 1$ ,  $sp_0^2 \ll \omega_0^2$ , a maximum in the region of concentrations  $n$  corresponding to  $p_0 \sim \omega_0 s^{-1/2}$ , and a descending part for large values of  $n$ . Actually condition (20) is apparently satisfied for such impurity concentrations at which the properties of the alloy have already very little in common with the properties of the solvent material.

If  $\Delta_0$  denotes the value of the gap at the impurity concentration  $n_0$ , then by assuming parabolic bands and

with the aid of expressions (15) and (18) one can determine the gap  $\Delta$  at the concentration  $n$ :

$$\ln \Delta = (n_0/n)^{1/2} \ln \Delta_0. \quad (23)$$

One can write down an analogous expression for  $T_c$ .

### 3. CONCLUSION

At the present time it has been firmly established by experiment that the three semiconducting compounds SnTe,<sup>[10]</sup> SrTiO<sub>3</sub>,<sup>[11]</sup> and GeTe<sup>[12]</sup> can be regarded as possessing superconducting properties. The first two substances belong to the class of "pseudo-ferroelectrics." Thus, for example, for SnTe we have  $\omega_0 \approx 20^\circ\text{K}$  at  $T = 0^\circ\text{K}$ . At temperatures below  $670^\circ\text{K}$  the compound GeTe is apparently ferroelectric.<sup>[13]</sup> At the temperature of absolute zero, here  $\omega_0$  is still smaller than the characteristic Debye frequency.

The superconducting properties of SnTe and GeTe are extremely complicated. In the narrow range of carrier density between  $8 \times 10^{20}$  and  $1.6 \times 10^{21} \text{ cm}^{-3}$  their transition temperature  $T_c$  increases from 0.1 to  $0.4^\circ\text{K}$ . The ratio  $2\Delta(0)/T_c$  has been measured<sup>[14]</sup> for GeTe, where  $\Delta(0)$  is the value of the superconducting gap at  $T = 0$ . It turns out to be equal to 4.3, which is not in agreement with the law of corresponding states. Superconductivity has been observed in SrTiO<sub>3</sub> over a much wider range of densities, from  $10^{18}$  to  $10^{21} \text{ cm}^{-3}$ . In this region  $T_c$  varies from  $0.1^\circ\text{K}$ , passes through a maximum ( $0.5^\circ\text{K}$ ) at  $n \approx 10^{20} \text{ cm}^{-3}$ , and then decreases to  $0.1^\circ\text{K}$  for large values of  $n$ .

For SrTiO<sub>3</sub> relation (23) describes the ascending branch of the  $T_c(n)$  curve and agrees with the observable data to within the limits of experimental accuracy. For SnTe and GeTe the superconducting transition temperature  $T_c$  increases with the concentration more rapidly than Eq. (23) implies, a result which is related to the strongly nonparabolic nature of the electron (hole) spectra of these substances. The centers of the L-valleys in these semiconductors do not correspond to extrema, but rather to saddle points at which two valence bands intersect.<sup>[15]</sup> Therefore, the superconductivity begins here only after filling the upper band of "light" holes, when a maximum appears in the density of states.

As follows from the present investigation, the superconductivity is certainly not at any time connected with the many-valley structure of the electron spectrum of a degenerate semiconductor, as it is in Cohen's model. On the other hand, the single-valley mechanism of Gurevich, Larkin, and Firsov enables us to predict the phenomenon only in principle. Here the very origin of superconductivity is due to such delicate relationships between the set of parameters appearing in the theory, that the effective coupling turns out to be weak.

The deviation from the law of corresponding states indicates that the Cooper logarithm does not have a sufficiently large value. Therefore, we need to take account of optical frequencies smaller than the Debye frequency, which is done in the present work. The specific mechanism considered above enables us to qualitatively explain the strong coupling in polar semiconductors possessing a low-lying optical phonon branch. In this connection  $T_c$  is bounded from above by the small frequency  $\omega_0$ , but it may approach arbitrarily near to this value for not too small values of the matrix elements

$C_b$ . In order to explain the transition temperature of the known semiconductors it is necessary to assume  $C_b \sim 1/10$ .

In order to clarify the actual role of the optical degrees of freedom of the lattice, it would be desirable to measure the isotope effect in a diatomic crystal such as, for example, GeTe. Since the heavy atom mainly oscillates in the acoustic branch, then the isotope effect should be more weakly expressed by the atoms of  $Te^{128}$  than by the atoms of  $Ge^{73}$ .

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