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THE ONE-DIMENSIONAL PLASMA MODEL

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1. Introduction

The mathematical description of unusual properties of a quasi-one-dimensional (q1d) solid, i.e., a solid whose microscopic structure consists of well-separated parallel chains, always begins with the introduction of a one-dimensional (1d) model Hamiltonian describing a single chain. It is introduced intuitively, there is no rigorous procedure based on exact analysis of the electron-ion system forming the chain for deriving this model Hamiltonian. The main purpose of this paper is to introduce a model Hamiltonian describing the 1d system of electrons and ions which interact through a Coulombic-type interaction.

There are two different theoretical methods of the mathematical description of the chain. In the first method only the electron system is considered. For the description of the chain along which the conductivity is almost metallic, the electron gas Hamiltonian with two-body interactions is used [1,2]. On the other hand, for the description of the nonconductive electron system the Hubbard Hamiltonian with strong intra-atomic correlations is employed [3,4]. The 1d electron models are used to explain effects arising from the variety of the electron pairing arrangements.

In the other method, the description of the chain begins with employing the 1d Fröhlich Hamiltonian [5 to 7] which describes the system of conduction electrons interacting with phonons. The intrachain electron-electron interaction is not included in the Hamiltonian. This model is used to explain the lattice distortion (Peierls phase) caused by the electron-phonon interaction.

There are, of course, model Hamiltonians in which both the electron-electron interaction and the electron-phonon interaction is included. However, the strengths of these interparticle interactions are
not specified by a consistent procedure (from first principles). Either
their explicit forms are not stated at all or they are parameterized by
constants [8 to 12] or at least the strength of the electron-electron
interaction is supposed to have the form of the three-dimensional (3d)
Fourier transform of the Coulombic electron-electron potential energy
[13 to 15]. One of the reasons for such a procedure is certainly the
impossibility to represent the Coulombic potential energy by the 1d
Fourier series, namely, the Fourier transform of the function $1/|z|$ is
logarithmically singular. To avoid this difficulty, Gutfreund and Little
[2] replaced the Coulombic potential energy between the two electrons
moving in the chain by the Coulombic-like potential energy:

$$\frac{e^2}{4\pi|z|} \rightarrow \frac{e^2}{4\pi(|z|+b_{ee})^3} ,$$

where $e$ is the elementary charge, $|z|$ is the distance between the two
electrons and the parameter $b_{ee}$ corresponds to the on-site electron-
electron potential energy. In this paper, the starting point of the
treatment of the 1d electron-ion system is the assumption that the
mutual potential energy $U(z)$ of any two particles not only of
electrons is of the following form

$$U(z) = \frac{q_1 q_2}{4\pi(|z|+b_{12})} ,$$

where $q_1$ and $q_2$ are the effective electrical charges of the particles
and $b_{12}$ is the parameter corresponding to the potential energy of the
two particles being situated at the same point of the chain. The further
treatment follows the well-known method of investigating the behaviour
of the 3d electron-ion system called the plasma model [16 to 18]. In
this method the investigation begins with the study of the system of
electrons and ions interacting according to the bare (unscreened)
Coulombic interaction. The normal modes of this system are the
corresponding plasma oscillations. The subsequent allowance for the
electronic screening allows to introduce ordinary (screened) phonons.

This method yields a clear-cut criterion of the lattice stability which
is formed from the requirement that the squares of the screened phonon
frequencies should not possess negative values. This stability criterion
is used for establishing the condition of appearing or not appearing the
lattice distortion. This paper is organized as follows: the classical
Hamiltonian in the momentum representation is stated in Section 2, the
stability conditions are analysed in Section 3, and Section 4 is devoted
to the discussion.

2. The Hamiltonian

As mentioned, the plasma model describes the electron-ion system whose
total Hamiltonian $H$ is

$$H = H_1 + H_e + H_{el} ,$$

where $H_1$, $H_e$, $H_{el}$ are the ion part, the electron part and the electron-
ion part of the total Hamiltonian, respectively.

The ion part of the Hamiltonian is of the form

$$H_1 = \sum_{\alpha=1}^{N_i} \frac{p^2_\alpha}{2M} + \sum_{\alpha=1}^{N_i} \sum_{\beta=1}^{N_i} \frac{Z^\alpha \cdot Z^\beta}{4\pi\epsilon(|\alpha - \beta| + b_{11})} = \sum_{\alpha=1}^{N_i} \frac{p^2_\alpha}{2M} + \sum_{k=0}^{L} \sum_{\alpha=1}^{N_i} \frac{1}{2\pi} w(k) e^{ik(\alpha - \beta)} ,$$

where $P_\alpha$ and $Z_\alpha$ are the momentum and the position of the $\alpha$-th ion,
respectively, $M$ is the ion mass, $N_i$ is the number of the ions in the
system, $Z^\alpha$ is the effective valence ($0 < Z^\alpha < 2$), $\epsilon$ is the dielectric
constant of the organic media surrounding the chain (typical values of
the relative dielectric constant are in the region $2 < \epsilon < 3$), $L$ is the
length of the chain, $k=2\pi n/L$ is the wave vector, $n$ is an integer. The
strength of the ion-ion interaction is

$$w(k) = \frac{Z^\alpha \cdot Z^\beta}{2\pi\epsilon} \gamma(b_{11}|k|) ,$$
The electron part of the total Hamiltonian has the form

\[ H_e = \sum_{j=1}^{N_e} \left( \frac{p_j^2}{2m} + \frac{e^2}{4\pi \varepsilon_0} \sum_{j=1}^{N_e} \frac{1}{|z_j - z_j'|} \right) \]

where \( p_j \) and \( z_j \) are the momentum and the position of the \( j \)-th electron, respectively, \( m \) is the electron mass, \( N_e \) is the number of the electrons in the system and the strength of the electron-electron interaction is

\[ u(k) = \frac{e^2}{2\varepsilon_0} \rho_{ee}(|k|) \]

The electron-ion interaction is supposed to consist of the Coulomb-like attraction and the short range delta-function repulsion [19]:

\[ H_{el} = \sum_{a=1}^{N_i} \sum_{j=1}^{N_e} \left( \frac{Z^* e^2}{4\pi \varepsilon_0 |z_j - a_j'|} + \beta^* \delta(z_j - a_j') \right) \]

\[ = -\sum_{k=0}^{L} \sum_{a=1}^{N_i} \frac{1}{L} \sum_{j=1}^{N_e} u(k) e^{ik(z_j - a_j)} \]

where \( \beta^* \) is the strength of the repulsion, \( \delta(x) \) is the Dirac function of the argument \( x \) and the strength of the electron-ion interaction is

\[ u(k) = -\frac{Z^* e^2}{2\varepsilon_0} \rho_{el}(|k|) + \beta^* \]

The term with \( k=0 \) does not appear in the Hamiltonian because of the electrical neutrality of the system. As the whole charge of the electrical neutral system is zero, the equation \( Z^* N_i = N_e \) is valid. It is obvious that \( N_i = L/a \) and \( e = 2\pi k_F /a \), where \( a \) is the interionic spacing and \( k_F \) is the Fermi wave vector. The combination of the previous equations gives

\[ k_F = \frac{Z^* e}{2a} \]

Because \( x/a \) is the 1d Debye wave vector, the parameter \( Z^* e /2 \) expresses the degree of the band filling. Like in the 3d plasma model [18], the condition of the electrical neutrality can be represented as well as by the relation

\[ \lim_{k \to 0} \frac{u^2(k)}{w(k)} = 1 \]

The function \( y(x) \) which gives the Fourier transform of the potential energy is shown in Fig. 1. In the limit \( x \to 0 \), the following expansion holds

\[ y(x) = -\ln(x) + \ldots \]

Hence, the Fourier transform of the potential energy diverges logarithmically either as the parameter \( b \) or the wave vector \( k \) approaches zero. The singularity at \( b=0 \) expresses the logarithmical singularity of the 1d Fourier transform of the Coulomb potential energy. The singularity at \( k=0 \) is the consequence of the long-range nature of the interaction.
In the limit $x \to \infty$, the following expansion is valid

$$\gamma(x) = \frac{1}{x^2} - \frac{6}{x^4} + \ldots$$

Therefore, in the limit of the large wave vector $k$, one gets

$$v(k) = \frac{1}{2\pi \hbar k} \left[ \frac{e^2}{\varepsilon_0 \varepsilon_k^2} \right] + \ldots$$

The expression in the square brackets is the well-known form of the 3D Fourier transform of the Coulombic electron-electron potential energy. It is often substituted for the strength of the electron-electron interaction in the 1D electron-phonon Hamiltonian [13 to 15].

3. The Stability Conditions

In the harmonic approximation [16,20], the ion part of the Hamiltonian takes the form

$$H_1 = E_0 + \frac{1}{2} \sum_g \left[ P(g)P(-g) + \frac{\gamma^2}{\varepsilon_0} Q(g)Q(-g) \right], \quad (5)$$

where

$$E_0 = \frac{1}{2} \sum_{k \neq 0} \sum_{n=1}^{N} w(k) e^{i k a (n-\beta)} = \frac{2\varepsilon_0^2}{4\pi \varepsilon_0} \sum_{k \neq 0} \left[ \frac{\varepsilon_0^2}{4\pi \varepsilon_0} - \frac{\gamma}{2\varepsilon_0} \right]$$

is the equilibrium position ion-ion potential energy, $g$ is the wave vector from the first Brillouin zone defined by $-a < g < a$ and $\epsilon_0 = \varepsilon_0/a$ is the reciprocal lattice wave vector, $P(g)$ is the momentum conjugate to the normal mode coordinate of the ion oscillations $Q(g)$ which is implicitly defined by the relation

$$Z_0 = \varepsilon_0 + \sum_g \frac{Q(g)}{(N_0\varepsilon_0)^{1/2}} e^{i g x}$$

The unrenormalized (unscreened) phonon frequency at the wave vector $g$ is

$$\gamma_{\text{ph}}^2(g) = \gamma_0^2(g) + \sum_{n=0}^{N_0} \left[ \gamma_n^2 (g\epsilon_0^2) - \gamma_0^2 (g) \right], \quad (6)$$

where

$$\gamma_0^2(k) = \frac{k^2 w(k)}{a M}$$

It can easily be shown using the expansion of the function $\gamma(x)$ at large values of $x$ that the series in (6) converges.

The unrenormalized phonon frequency $\gamma_{\text{ph}}(g)$ as the function of the wave vector $g$ is shown in Fig.2. In this paper, the following numerical values of the parameters are used: $Z^* = 5/3$, $a = 3.4 \times 10^{-10}$ m, $\varepsilon_r = 2.6$ ($\varepsilon_0 = \varepsilon_r$).

![Fig 2 The unrenormalized phonon frequency $\gamma_{\text{ph}}(g)$ as the function of the wave vector $g$.](image)

At high temperatures $T > \frac{\hbar \omega_{\text{ph}}}{k_B}$ where $k_B$ is the Boltzmann constant, $\hbar$ is the Planck constant, the effects of the anharmonic (cubic, biquadratic, etc.) terms in the ion displacements may become important. As known [21], the anharmonic terms are of no importance as far as the temperature satisfies the condition $T \ll \frac{\hbar \omega_{\text{ph}}}{k_B}$. The plot of the function $\varepsilon_0 = \varepsilon_0 / N_0$ versus the parameter $b$ is shown in Fig.3 for the numerical values of the parameter $Z^*$, $a$, $\varepsilon_r$ given above ($Z^* = (\varepsilon_0^2 / (4\pi \varepsilon_0 a^2 \varepsilon_0^2)) = 2.1 \times 10^5$ K).

There is still another possible interpretation of the inequality settled for the temperature, namely, as the stability criterion against melting. According to the idea of Lindemann [22], a solid melts as the vibrations of ions about their equilibrium positions become too large.
In the 3d system, the ion vibrations are considered to be large when the mean-square amplitude of the vibrations is comparable to the square of the interionic spacing. This criterion of the large vibrations is not applicable to the 1d system as the 1d mean-square amplitude of the ion vibrations as well as many other 1d fluctuation quantities yields a divergent result for the small wave vector. The ion vibrations can also be regarded large as far as the energy of the vibrations (i.e., \( N_k k_B T \) at high temperatures) is comparable with the absolute value of the equilibrium position ion-ion interaction energy \( E_{\text{eq}} \). Hence, the melting temperature is \( T_m = \gamma H_1 \), where \( \gamma H_1 \) is a dimensionless parameter which may depend on the parameter \( b_{11} \) and \( T \equiv [E_0]/(N k_B) \). However, the real destruction of the system may happen at the temperature \( T_D \) lower than the melting temperature \( T_m \) owing to the chemical decomposition of the organic media surrounding the chain.

Proceeding along the well-known route followed for the 3d model [16,201, one easily obtains the expression for the squared renormalized (screened) phonon frequency at the wave vector \( g \) and the temperature \( T \):

\[
\mathcal{G}_{\text{ph,1d}}(g,k) = \mathcal{G}_{\text{1d}}^2 + \sum_{n=0}^{\infty} \frac{\mathcal{G}_{\text{1d}}^2(g+n)}{\mathcal{G}_{\text{1d}}^2}.
\]  

where

\[
\mathcal{G}_{\text{1d}}^2 = \frac{2}{\pi k_B T} \ln \frac{2k_B T}{\hbar^2 k}.
\]

and \( \epsilon_{\text{r}}(k,0) \) is the static dielectric function of the 1d electron system.

In the RPA, it is given by

\[
\epsilon_{\text{r}}(k,0) = 1 - v(k)\Lambda_t(k,0),
\]

where \( \Lambda_t(k,0) \) is the 1d static Lindhard function

\[
\Lambda_t(k,0) = \frac{1}{\pi} \sum_{k'} \frac{f(k') - f(k+k')}{E(k') - E(k+k')}.
\]

\( f(k) \) is the Fermi-Dirac distribution function and \( E(k) = \hbar^2 k^2/(2m) \).

At the zero temperature, the 1d Lindhard function contains the well-known logarithmic singularity at \( k = 2k_r \):

\[
\Lambda_0(k,0) = \frac{2\pi}{\hbar^2 k} \ln \frac{2k_r}{\hbar^2 k}.
\]

This logarithmic singularity manifests itself as well as in a phonon anomaly of the present model of the electron-ion system.

Neglecting the effects of the periodicity in the expression \( (7) \), one gets the following phonon dispersion relation at the zero temperature

\[
\mathcal{G}_{\text{ph,0}}^2(k) = \mathcal{G}_{\text{0}}^2(k) - \mathcal{G}_{\text{1d}}^2(k) + \frac{\mathcal{G}_{\text{1d}}^2}{2\pi k_B T} \ln \frac{2k_r}{\hbar^2 k}.
\]

Evidently, the expression \( (8) \) contains a sharp logarithmic anomaly at \( k = 2k_r \). If \( \mathcal{G}_{\text{0}}^2(2k_r) > \mathcal{G}_{\text{1d}}^2(2k_r) \), i.e., \( \mathcal{G}_{\text{0}}^2(2k_r) > \mathcal{G}_{\text{1d}}^2(2k_r) \) it even leads to imaginary phonon frequencies expressing the lattice instability. In terms of the parameters of the present model, this inequality has the following form \( (2e^2/mc)^{k_{1d}} < (Z e^2/mc) \).

\[
\mathcal{G}_{\text{ph,0}}^2(k) = \mathcal{G}_{\text{0}}^2(k) - \mathcal{G}_{\text{1d}}^2(k) + \frac{\mathcal{G}_{\text{1d}}^2}{2\pi k_B T} \ln \frac{2k_r}{\hbar^2 k}.
\]
As a matter of fact, the effects of the periodicity in the phonon dispersion relation can be neglected only as far as the wave vector \( k \) is small: \( k \approx 2k_f \). Hence, one has to use the dispersion relation (7) while examining the 2\( k_f \)-phonon anomaly. Fig.4 shows the plot of the squared renormalized phonon spectrum at the zero temperature calculated with the use of the dispersion relation (7) for the numerical values of the parameters \( Z^*, a, \epsilon_f, b \) given above and \( \beta^* = 0 \). There is a giant anomaly in the phonon spectrum at the wave vector \( k \) such that \( k \approx 2k_f \).

![Graph of squared renormalized phonon frequency at zero temperature](image)

Fig. 4 The squared renormalized phonon frequency at the zero temperature \( \chi^2_{ph,0}(k) \) as the function of the wave vector \( g \).

It is so giant that the squares of the renormalized phonon frequencies possess negative values. This violation of the criterion of the lattice stability indicates the tendency of the system to undergo the transition to the state with the periodic lattice distortion. Several equi-(squared renormalized phonon) frequency lines are shown in Fig. 5 for the wave vector \( 2k_f \) whereby the squared frequency unit is \( (Z \cdot a \cdot M)/(\hbar k_f)^2/(\text{amu})^2 \).

The area bordered by the dotted lines defines the numerical values of the parameters \( b \) and \( \beta^* \) describing the 1d electron-ion system with the parameters \( Z = 5/3, a = 3.4 \times 10^{-10} \) m, \( \epsilon_f = 2.6 \) that is stable against the lattice distortion.

With increasing the temperature, the logarithmal singularity of \( \chi^2_{ph,0}(k) \) is smoothed out by smearing of the Fermi-Dirac distribution function. This results only in a sharp peak of \( \chi^2_{ph,0}(k) \) which progressively weakens with increasing the temperature. Consequently, the phonon anomaly becomes less giant and above some temperature \( T_p \) (the transition temperature), the squares of the renormalized phonon frequencies do not possess negative values any more. Hence, the crucial point of the calculation of the transition temperature which is given by the solution of the equation

\[
\chi^2_{ph,k,0}(2k_f) = 0
\]

is the determination of the dependence of \( \chi^2_{ph,k,0}(2k_f) \) on the temperature. Unfortunately, there exists only a very rough estimate of \( \chi^2_{ph,k,0}(2k_f) \), namely [6]:

\[
\chi^2_{ph,k,0}(2k_f) = -\frac{n}{\hbar^2 k_f^2} \ln \left( \frac{\sqrt{n} k_f}{\pi k_B T} \right),
\]

where \( \ln = 0.5772 \ldots \) is the Euler constant. With the help of the expression (10), the equation (9) can be rewritten as

\[
T_p = \frac{\sqrt{n} k_f^2}{\pi k_B} \exp \left[ -1/\lambda_{ph} \right],
\]

where the dimensionless electron-phonon interaction parameter of the 1d electron-ion system at the temperature \( T \) is given by

\[
\lambda_{ph} = \frac{n}{\hbar^2 k_f^2} \ln \left( \frac{\sqrt{n} k_f}{\pi k_B T} \right),
\]

Fig. 5 The equi-(squared renormalized phonon) frequency lines at the wave vector \( 2k_f \).
As the dielectric function at the wave vector \( k_t \) slightly varies with the temperature, \( \lambda_T \) in the equation (11) can be replaced by \( \lambda_0 \), the electron-phonon interaction parameter at the zero temperature:

\[
\frac{1}{\lambda_T} \approx - \frac{\hbar^2 k_f}{m v(2k_f)} \left[ 1 + \sum_{G_n=0}^\infty \left[ \frac{\tilde{\alpha}^2(2k_f) - \tilde{\alpha}^2(2k_f,G_n)}{\tilde{\alpha}^2(2k_f) + \tilde{\alpha}^2(2k_f,G_n)} \right] \right]
\]

As the dielectric function at the wave vector \( k=2k_f \) slightly varies with the temperature, \( \lambda_T \) in the equation (11) can be replaced by \( \lambda_0 \), the electron-phonon interaction parameter at the zero temperature:

\[
\lambda_0 = - \frac{m v(2k_f)}{\hbar^2 k_f} \left\{ \frac{\tilde{\beta}^2(2k_f,G_n)}{\tilde{\beta}^2(2k_f,G_n) + \tilde{\beta}^2(2k_f,G_n)} \right\}
\]

To obtain a more reliable value of the transition temperature, one should refer to the equation (9) and solve it by numerical methods. It will be still only a mean-field value of the transition temperature, i.e. a value calculated without taking into account fluctuations which shift the phase transition of any 1D system to the zero temperature. However, it is known [23,24] that a small degree of the interchain coupling is sufficient to bring the actual transition temperature close to its mean-field value.

Evidently, if the transition temperature \( T_p \) is greater than the temperature \( T_D \), the system is in the state with the periodic lattice distortion as far as the temperature is lower than \( T_D \) and at \( T_D \) it is destroyed by melting or the chemical decomposition. In the opposite case, if \( T_p \) is lower than \( T_D \), the system is in the metallic state as far as the temperature is greater than \( T_p \) and lower than \( T_D \) and at \( T_p \) the lattice is periodically distorted.

4. Discussion

Models used to interpret experimental data do not involve all complex processes that occur in real 1D solids. Though they are very simplified, their solutions give some guidance about what is going on. The plasma model presented in this paper also describes the 1D electron-ion system in the simplified way. In general, the interaction between ions is not well represented by a potential when the coupling between closed-shell electrons on different ions plays an important role. In the expression for the electron-ion interaction energy, the fact that ions possess a structure (core electrons) has again been neglected. As soon as the Pauli principle becomes important in the interaction between the conduction electrons and the core electrons, the interaction cannot be represented by a potential as well. The electron-ion interaction has also been assumed not to be affected by the ion displacement (the rigid-ion approximation). However, the restriction to the interparticle interactions represented by the Coulombic-like potential brings along a compensating possibility of obtaining the Hamiltonian which is explicitly defined. On the other hand, this turns to be the main advantage of the present model over others which resides in the explicitly stated dependence of the strengths of all the interparticle interactions on the wave vector. In the same consistent way, the model can be extended to describe a 1D system by including the interchain Coulombic interaction in the Hamiltonian.

Comparing with the other models of the 1D electron-ion system, one also finds out that the present model yields another view on the role of the interparticle interactions and the periodicity of the system (the Umklapp processes) in arising the lattice instability. Within the framework of the Fröhlich model, the lattice instability arises at any values of the strength of the electron-phonon interaction [5 to 7]. The present model whose Hamiltonian includes both the electron-ion interaction and the electron-electron interaction yields a fully different result. Without taking into account the effects of the
periodicity, the lattice instability appears if the squared strength of the electron-ion interaction is greater than the product of the strengths of the ion-ion interaction and the electron-electron interaction taken at the wave vector \( k = 2k_F \). A model of the 1d electron-ion system fulfilling the criterion of the lattice instability in this form was studied, e.g., by Nakane and Takada [15].

However, it follows from the present treatment of the 1d system that the effects of the periodicity significantly make for the lattice instability. From the phonon dispersion relation which neglects the effects of the periodicity one finds out that the lattice of the system described by the present model should be stable as far as

\[
\beta_N < \frac{2}{\pi} \frac{m_e}{m_i} \frac{2}{\pi} \frac{\hbar^2}{\kappa a}.
\]

This inequality defines incorrect values of the parameters \( b \) and \( \beta_N \) for which the lattice is stable. The real values calculated from the phonon dispersion relation taking into account the effects of the periodicity can be obtained from Fig. 5. Evidently, they are fully different from those defined by the above-given inequality. Hence, one can also conclude that the effects of the periodicity (the Umklapp processes) tend to strengthen the transition to the state with the lattice distortion. This conclusion is in the opposition with that of Gasser [14] who asserted that the Umklapp processes tend to suppress the lattice instability. It is also necessary to remark that a better approximation for the dielectric function which goes beyond the RPA may give a little different region of the values of the parameters \( b \) and \( \beta_N \) for which the lattice is stable. Evidently, the appreciation of the influence of various effects (the Umklapp processes, the self-energy and exchange contributions to the dielectric function, etc.) on the lattice instability is possible only within the framework of such a model in which the strengths of all the interparticle interactions are explicitly stated.

In dependence on the values of the parameters, the Hamiltonian stated in this paper describes the 1d electron-ion system in which the periodic lattice distortion either appears or does not. The lattice distortion is generally connected with the formation of a charge-density-wave. On the other hand, the system which is stable against the lattice distortion can exhibit various effects originating in miscellaneous electron pairing arrangements. Thus the plasma model allows to study all primary phenomena exhibiting in the 1d electron-ion system by the application of only one Hamiltonian. It is hope that further study will lead to the description of other 1d phenomena within the framework of the plasma model.

References

The One-Dimensional Plasma Model

The Hamiltonian of the one-dimensional electron-ion system is proposed. The relevant forces, electrons and ions interact via, are of the Coulombic-type. Besides, the repulsive pseudopotential in the form of the delta-function is included in the electron-ion interaction. The stability of the system against the lattice distortion is examined. The effects of the periodicity appear to strengthen the transition to the state with lattice distortion. It is defined the region of the values of the parameters describing the system stable against the lattice distortion.

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