Viscosity effect on capillary waves at liquid interfaces

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Abstract. We discuss the viscosity damping effect on capillary waves in a binary-liquid system, using the linearized Navier–Stokes equation. The damping correction for the dispersion relation depends on the wave vector k as well as the interfacial tension. The calculated k-dependence of damping is characterized by a critical capillary-wave-number value k_c, which separates the regions of weak and strong damping. The surface and interfacial roughnesses of a binary liquid system with large liquid depths are calculated and compared to experiments. Although the analysis has been restricted to the classical, macroscopic level, we obtain a noticeable modification from earlier hydrodynamic results for capillary wave damping and liquid–liquid interfacial roughness.

1. Introduction

In one approach to describe the structure of a liquid surface or interface a continuous, inhomogeneous density profile (intrinsic density profile) is investigated using the mean field theory of van der Waals and Rayleigh [1]. Another approach, stimulated by Buff *et al* [2], is the capillary-wave model which presumes that the finite thickness of a fluid interfacial zone results from wavelike thermal excitations (capillary waves with wave vector k) on a flat abrupt interface. For quantitative interpretation of the reflectivity data from liquid surfaces and interfaces, the capillary wave theory has been extended [3, 4]. One of the modifications focuses on the high k cutoff of the capillary wave spectrum, which is often identified with the inverse of the correlation length or the molecular size [5, 6]. The molecular basis of capillary wave dispersion and damping has been explored using statistical mechanics, by Grant and Desai [7], but here we will focus on a macroscopic description.

We take the intrinsic density profile of a liquid surface or interface to be sharp and characterize the interfacial zone solely by capillary waves, an approach which is justified for immiscible liquids. Using the linearized Navier–Stokes equation we study the dynamic properties of waves on a liquid–liquid interface in thermal equilibrium. From the wave damping we determine a high-k cutoff for capillary waves, and show that the cutoff is essential for a quantitative description of interfacial roughness.

Although the hydrodynamic parameters, viscosity and surface/interface tension, can be related to descriptions on the molecular level [8], we use the phenomenological, hydrodynamic approach, assuming in particular that these parameters are independent of k. This is justified when the viscosity is not large [9, 10], and when the wavelengths $2\pi/k$ are much longer than the correlation length for the fluid [11]. The bending energy [12] for interface fluctuations is neglected, since for the small capillary wave amplitudes considered here the interface curvatures are small.

In spite of these classical simplifications, the dispersion relation for the interfacial waves is complicated and usually obtained under various approximations focusing on specific physical systems [13, 14], for instance, the neglect of air density and/or viscosity for an air–liquid interface [7, 15]. For a liquid–liquid interface, the dispersion relation corrected for small viscous damping has been given by Herpin *et al* [14, 16]. Following the approach used by Levich [17] we obtain an essentially different correction for the dispersion relation as shown below. The correction is, however, consistent with a result given by Loudon [18], and we will show that the modification is essential for the analysis of recent interface roughness data obtained by neutron reflection. We also derive an equation for the surface roughness for a single viscous fluid with small damping.

2. The viscous damping of capillary waves

We start from the linearized Navier–Stokes equation

$$\rho \frac{\partial \boldsymbol{v}}{\partial t} = -\boldsymbol{\nabla} p + \rho \boldsymbol{g} + \eta \boldsymbol{\nabla}^2 \boldsymbol{v} \tag{1}$$

for an incompressible fluid with density ρ, velocity $\boldsymbol{v}$, pressure p and the gravitational acceleration $\boldsymbol{g}$. For the small wavelike perturbations considered here, we can neglect the non-linear term $\rho(\boldsymbol{v} \cdot \boldsymbol{\nabla})\boldsymbol{v}$ in the general Navier–Stokes equation, but take into account finite viscosity $\eta = \rho \nu$, where ν is the kinematic viscosity. For simplicity, we discuss only the limiting case of *a binary-liquid system with large liquid depths*. It has been shown previously [19, 20] that for large depths surface and interfacial roughnesses are uncoupled, being generated independently by different modes, the surface and interfacial mode, respectively. We will focus first on the interfacial mode, and discuss the surface roughness in section 2.2.

2.1. Interfacial roughness of viscous fluids of large depths

2.1.1. Dispersion relation for the interfacial mode. We extend the analysis of small waves at the air–liquid interface of a single liquid by Levich [17] to the liquid–liquid interface of a binary-liquid system. For a binary-liquid system of large liquid depths (the criterion for 'large depths' is $kh \gg 1$ and $kh' \gg 1$, where $h(h')$ is the depth of the bottom (top) liquid), wavelike solutions of equation (1), for the bottom liquid, are of the form [17]

$$v_x = \sum_k (ik A_k \, e^{kz} - l C_k \, e^{lz}) \, e^{i(kx - \omega_k t)} \tag{2}$$

and

$$v_z = \sum_k (k A_k \, e^{kz} + ik C_k \, e^{lz}) \, e^{i(kx - \omega_k t)} \tag{3}$$

with

$$l = k\sqrt{1 - i\omega_k/(\nu k^2)} \tag{4}$$

where ω_k is the frequency of the mode with the wave number k. We use a coordinate system, where $z = 0$ defines the sharp interface and x is the direction of the wave propagation along the interface. Similarly, for the top liquid (using primed quantities),

$$v'_x = \sum_k (ik A'_k \, e^{-kz} + l' C'_k \, e^{-l'z}) \, e^{i(kx - \omega_k t)} \tag{5}$$

and

$$v'_z = \sum_k (-kA'_k \, \mathrm{e}^{-kz} + \mathrm{i}kC'_k \, \mathrm{e}^{-l'z}) \, \mathrm{e}^{\mathrm{i}(kx - \omega_k t)} \tag{6}$$

with $l' = k\sqrt{1 - \mathrm{i}\omega_k/(\nu' k^2)}$. In equations (2), (3), (5) and (6) the velocities are determined by two contributions: (i) the potential flow described by the terms with amplitudes A_k, A'_k, which correspond to the velocity potentials: $\phi = \sum_k A_k \, \mathrm{e}^{kz} \, \mathrm{e}^{\mathrm{i}(kx - \omega_k t)}$ and $\phi' = \sum_k A'_k \, \mathrm{e}^{-kz} \, \mathrm{e}^{\mathrm{i}(kx - \omega_k t)}$; (ii) the rotational flow represented by a vector potential with amplitudes C_k, C'_k, describing fluids of finite viscosity. The coefficients A_k, C_k, A'_k and C'_k, and the dispersion $\omega_k(k)$ are determined by the boundary conditions at the interface ($z = 0$), where the stress tensor and velocity must be continuous [21]:

(a) The continuity of the stress tensor in the vertical direction gives

$$\sigma_{zz} - \sigma'_{zz} = \alpha \frac{\partial^2 \zeta}{\partial x^2} \tag{7}$$

where $\sigma_{zz} = -p + 2\eta \partial v_z/\partial z$ and $\sigma'_{zz} = -p' + 2\eta' \partial v'_z/\partial z$ are corresponding stresses for the bottom and top liquid at the interface, and $\zeta = \sum_k \zeta_k \, \mathrm{e}^{\mathrm{i}(kx - \omega_k t)}$ is the instantaneous displacement of an interface point with horizontal coordinate x from the mean interfacial plane, with $\langle \zeta \rangle = 0$. The pressure is expressed as $p = -\rho g \zeta - \rho \partial \phi/\partial t$, $p' = -\rho' g \zeta - \rho' \partial \phi'/\partial t$ and the interfacial pressure $\alpha \partial^2 \zeta/\partial x^2$ is due to the interfacial tension α.

(b) The continuity of stress tensor components involving the horizontal direction gives $\sigma_{xz} = \sigma'_{xz}$, where $\sigma_{xz} = \eta(\partial v_x/\partial z + \partial v_z/\partial x)$ and $\sigma'_{xz} = \eta'(\partial v'_x/\partial z + \partial v'_z/\partial x)$ are the corresponding components for the bottom and top liquid, respectively.

(c) The continuity of vertical and horizontal velocity gives $v_z = v'_z$.

(d) $v_x = v'_x$.

Inserting the velocities (equations (2), (3), (5) and (6)) and the velocity potentials into the four boundary equations in (a)–(d), and using the relation $v_z = \partial \zeta/\partial t$ at the interface, we obtain four linear homogeneous equations:

$$\sum_j N_{ij} S_j = 0 \tag{8}$$

where $i, j = 1$ to 4, and S_j stands for A_k, C_k, A'_k and C'_k for $j = 1, 2, 3, 4$, respectively. The coefficients N_{ij} form a 4×4 matrix N. Then dispersion relation $\omega_k(k)$ is obtained from the condition $\det[N] = 0$, which can be written

$$[\rho(k - l') + \rho'(k - l)](\rho + \rho') \left(\omega_k^2 - \omega_0^2 \frac{\Delta\rho}{\rho + \rho'} \right) - 4k[\rho\omega_k + \mathrm{i}(\eta - \eta')k(k - l)]$$
$$\times [\rho'\omega_k - \mathrm{i}(\eta - \eta')k(k - l')] = 0 \tag{9}$$

with $\omega_0^2 = gk + \alpha k^3/\Delta\rho$ and $\Delta\rho = \rho - \rho'$. It can be checked that for $\eta = \eta' = 0$ we obtain the same result $\omega_k = \omega_{k0} = (\Delta\rho/(\rho + \rho'))^{1/2}\omega_0$ as for the interfacial mode of ideal liquids. It is difficult to obtain a general solution for ω_k, since l and l' are functions of ω_k (see equations (4) and following (6)), but one can see that ω_k is a complex number whose imaginary part describes the viscous damping of the capillary waves. To shed more light on the general nature of the solutions to (9) we have converted it into a ninth-order polynomial equation for a quantity D related to ω_k by $D = \eta' + \eta\sqrt{1 - (\mathrm{i}\omega_k)/(\nu k^2)}$. Since in converting (9) the signs of the square roots (for l and l') were lost, not all the solutions to the converted equation correspond to solutions of the original equation (9). A numerical study in a wide k-range shows that for any k only one solution exists, and this solution assumes the asymptotic forms given below in the low- and high-viscosity limits.

We discuss the dispersion relation $\omega_k(k)$ and the interfacial roughness in these two cases only: (1) $|\omega_k| \gg \nu k^2$ and $|\omega_k| \gg \nu' k^2$, the weak damping approximation, and (2) $|\omega_k| \ll \nu k^2$ and $|\omega_k| \ll \nu' k^2$, the strong damping approximation.

2.1.2. Dispersion relation and interfacial roughness in the weak damping approximation.

Under the weak damping approximation the dispersion relation ω_k differs from the dispersion $\omega_{k0} = (\Delta\rho/(\rho + \rho'))^{1/2}\omega_0$ for ideal liquids only by a small quantity γ_k, and can be approximated by $\omega_k = \omega_{k0} - i\gamma_k$ as for a damped oscillator. Using this approximation for ω_k, l and l' in (9), we obtain

$$\gamma_k = (1 - i)\frac{\rho\rho'\sqrt{2\nu\nu'k^2/\omega_{k0}}}{(\rho + \rho')(\rho\sqrt{\nu} + \rho'\sqrt{\nu'})}\omega_{k0} \tag{10}$$

which is consistent with the expression given by Loudon [18]. The imaginary part of γ_k gives a small correction to the real part of ω_k. Inserting ω_{k0} in (10) we notice that the damping coefficient γ_k is not a constant for capillary waves, but increases with k as $k^{7/4}$.

Equation (10) differs essentially from the expression $\gamma_k = 2[(\eta + \eta')/(\rho + \rho')]k^2$ often used [14]. Moreover, when the top fluid density is set equal to zero equation (10) does not reduce to the damping coefficient $\gamma_k = 2\nu k^2$ [14, 15] for the air–liquid interfacial fluctuations obtained by neglecting the density of the top fluid from the beginning, as shown below in section 2.2.1.

Using the dispersion relation $\omega_k = \omega_{k0} - i\gamma_k$ and equation (8), we obtain the following relations for the coefficients:

(a) $C_k' = i(2k/l')A_k - (l/l')C_k$
(b) $A_k' = -(1 + 2k/l')A_k - i(1 + l/l')C_k$
(c) $C_k = i[(2\rho'k/l')/(\rho + \rho'l/l')]A_k$.

Relations (a)–(c) determine the velocities at the interface ($z = 0$) as follows:

$$v_x = v_x' = \sum_k ikA_k\left(1 - \frac{2\rho'\sqrt{\nu'}}{\rho\sqrt{\nu} + \rho'\sqrt{\nu'}}\right)e^{i(kx-(\omega_{k0}-i\gamma_k)t)} \tag{11}$$

and

$$v_z = v_z' = \sum_k kA_k\left(1 - \frac{k}{l}\frac{2\rho'\sqrt{\nu'}}{\rho\sqrt{\nu} + \rho'\sqrt{\nu'}}\right)e^{i(kx-(\omega_{k0}-i\gamma_k)t)}. \tag{12}$$

This result shows: (1) The horizontal velocity at the interface (equation (11)) is continuous (no slip), which differs significantly from that for ideal liquids where $v_x = -v_x' \neq 0$. (2) The vertical velocity (equation (12)) is affected only slightly by the viscosity in the weak damping region where $|k/l| \ll 1$.

The multiplying factor, A_k, for the fluid velocities in (11) and (12) can be determined from the wave energy. In thermal equilibrium the mean kinetic energy of each k wave in the system is related to the thermal energy $k_B T$ through

$$\varepsilon_k = \frac{\rho}{2}\int_{-\infty}^{0}(|v_{xk}|^2 + |v_{zk}|^2)\,dz + \frac{\rho'}{2}\int_{0}^{\infty}(|v_{xk}'|^2 + |v_{zk}'|^2)\,dz = \frac{1}{2}k_B T \tag{13}$$

where v_{xk}, v_{zk}, v_{xk}' and v_{zk}' are the velocity components of the k wave in (2), (3), (5) and (6).

Integrating (13) and using the relation $v_{zk} = \partial\zeta_k/\partial t$ at the interface, we obtain the mean square interfacial roughness contributed by the k wave:

$$\langle\zeta_k^2\rangle \approx \frac{k_B T\xi}{[(\rho + \rho')/k]\omega_{k0}^2}. \tag{14}$$

We have introduced here a factor

$$\xi = 1 - \pi\sqrt{\frac{2\nu k^2}{\omega_{k0}}}\frac{\rho\rho'\sqrt{\nu'}}{(\rho+\rho')(\rho\sqrt{\nu}+\rho'\sqrt{\nu'})}$$

which takes into account that for the damped oscillator a fraction $(1-\xi)$ of thermal energy is consumed by dissipation (and fed back into the thermal reservoir). This fraction is estimated to be $(\pi\gamma_k)/\omega_{k0}$, the same loss fraction as for a classical damped oscillator which cannot convert all its kinetic energy at oscillation amplitude zero to potential energy at the next reversal point.

In the weak damping limit, (14) reduces to the result for ideal liquids: $\langle\zeta_k^2\rangle \approx k_B T/[(\Delta\rho/k)(gk + \alpha k^3/\Delta\rho)]$, indicating that the ideal liquid assumption is valid, since $\omega_{k0} \gg \nu k^2$ and $\omega_{k0} \gg \nu'k^2$. Inserting in these inequalities $\omega_{k0}^2 = (\Delta\rho/(\rho + \rho'))(gk + \alpha k^3/\Delta\rho)$, we express the criterion for weak damping as:

$$k \ll \frac{\alpha}{4(\rho + \rho')\nu^2} \equiv k_c \tag{15}$$

where ν in the denominator should be replaced by ν' when $\nu < \nu'$. Here, we have neglected the gravity effect (which is justified except for extremely small $\alpha \ll 0.1$ dyn cm^{-1}).

In (15), k_c defines the wave number for critical damping of capillary waves at the interface of two viscous fluids. For weak damping: $k \ll k_c$, and for strong damping: $k \gg k_c$. The latter limit is discussed now.

2.1.3. Strong damping approximation ($k \gg k_c$). The strong damping approximation is defined by $l \approx k$ and $l' \approx k$ (or $|\omega_k| \ll \nu k^2$ and $|\omega_k| \ll \nu'k^2$). In this case, $l/k \approx 1 - i\omega_k/(2\nu k^2)$ and $l'/k \approx 1 - i\omega_k/(2\nu'k^2)$. Inserting these approximations into (9), we obtain two solutions for the dispersion relation: $\omega_k = 0$ and a purely imaginary $\omega_k = -i\omega_{k0}^2(\rho+\rho')/[2k^2(\eta+\eta')]$. The value $\omega_k = 0$ is formally an exact solution of (9), but it follows from (8), (2), (3), (5) and (6) that v, v' would vanish, and a finite static distortion of the interface does not satisfy balance of forces. The remaining purely imaginary solution for ω_k is overdamped, and contributes very little to the interfacial roughness. Thus, a good approximation to roughness is achieved by integrating the expression for $\langle\zeta_k^2\rangle$, obtained neglecting viscosity, from its minimum value $k_{min} \approx 10^{-8}$ Å^{-1} (determined by the sample size) not up to $k_{max} \approx 1$ Å^{-1} (as usually suggested), but only up to k_c (or up to k_c' for the single liquid case, discussed later in the paper). In this way, all effects of viscosity are approximated by neglect of the high k region $k > k_c$ (or $k > k_c'$).

2.1.4. Interfacial roughness of an oil–water–surfactant system. We apply (14) to the liquid–liquid interface of an octane–water–surfactant system, which has (at 20 °C) $\rho' = 0.7$ g cm^{-3} and $\nu' = 0.0077$ cm^2 s^{-1} for octane; $\rho = 1.0$ g cm^{-3}, $\nu = 0.01$ cm^2 s^{-1} for water; and a very small interfacial tension $\alpha = 0.1$ dyn cm^{-1}. Using these parameters with k-cutoffs of $k_{min} \approx 10^{-8}$ Å^{-1} and $k_{max} = k_c = 1.5 \times 10^{-6}$ Å^{-1} (much smaller than the value obtained from the molecular size), we obtain a much smaller interfacial roughness, $\langle\zeta^2\rangle^{1/2} \approx 19$ Å, than that, 90 Å, predicted by the standard model [22, 23]. This result is not only consistent with our previous experimental neutron reflection data [19, 20] but also with our previous analysis based on the potential flow assumption [19]. Now we have obtained the lower estimate of interfacial roughness without invoking the artificial low k cutoff due to slip. The slip is an artifact of the potential flow assumption and vanishes in viscous liquids.

We have neglected all effects of the surfactant monolayer at the oil–water interface other than reduction of interfacial tension. It has been shown in [17] that the fluctuation

of surfactant concentration in this monolayer due to interface fluctuations gives rise to wave damping. We assume that this effect is small when the monolayer coexists with an abundant supply of surfactant in the solution, which tends to counteract a change of surfactant concentration [24, 25] at the interface.

We have discussed viscous damping of interfacial roughness only for the limiting case of infinite liquid depths. If the top film is very thin, we expect additional suppression of interfacial roughness due to the coupling between waves at the surface and interface, as shown previously [19, 20].

2.2. Surface roughness of a single layer of viscous fluid of large depth

In [19] and [20] we have shown that the surface roughness of a binary liquid system of infinite depths is due solely to the surface mode, and is the same as that for a single liquid of infinite depth. Therefore, in the following we briefly analyse the surface roughness of a single liquid layer of density ρ', and show that for weak damping, we obtain the results of [14] and [15].

2.2.1. Dispersion relation. We use again expansions (2) and (3) for wavelike solutions to the linearized Navier–Stokes equation (1), changing to primed quantities.

These expressions have to satisfy continuity of the stress tensor at the air–liquid interface $z = 0$ for the vertical and horizontal directions, implying

$$(\omega_k + i2\nu'k^2)^2 + (2\nu'k^2)^2(l'/k) - \omega_0'^2 = 0 \tag{16}$$

and

$$C_k' = \frac{2i(k/l')^2}{1 + (k/l')^2} A_k'. \tag{17}$$

In (16), $l' = k\sqrt{1 - (i\omega_k)/(\nu'k^2)}$, and $\omega_0'^2 = gk + \alpha'k^3/\rho'$ is the dispersion relation for an ideal liquid of infinite depth. A general solution for ω_k is difficult to obtain from (16). For simplicity, we discuss only the cases of weak and strong damping, as before.

(A) *Weak damping* $(|\omega_k| \gg \nu'k^2)$. For weak damping we use $l'/k \approx (1-i)\sqrt{\omega_k/(2\nu'k^2)}$, and $\omega_k = \omega_0' - i\gamma_k'$, where $\omega_0' \gg \gamma_k'$, to obtain $\gamma_k' = 2\nu'k^2$ as in [14] and [15]. As expected, the results deviate little from those for an ideal liquid.

(B) *Strong damping* $(|\omega_k| \ll \nu'k^2)$. This region is defined by $l' \approx k$. Using the approximation $l'/k \approx 1 - i\omega_k/(2\nu'k^2)$, we obtain from (16) the solution $\omega_k = -i\omega_0'^2/(2\nu'k^2)$ satisfying the criterion $|\omega_k| \ll \nu'k^2$ [17].

In the region $k > k_c'$ there exists also a second solution to (16), $\omega_k \approx -i0.9\nu'k^2 + (0.2i\omega_0'^2)/(\nu'k^2)$. It is overdamped even more strongly than the first solution, and both can, therefore, be neglected in roughness calculations. Numerical studies confirmed that these are the only solutions in the high k region.

2.2.2. Roughness. Using the same procedure for the surface roughness as was used for the interfacial roughness in section 2.1.2, we obtain for weak damping

$$\langle \zeta_k'^2 \rangle = \frac{k_B T}{(\rho'/k)(\omega_0')^2} \left(1 - 2\pi \frac{\nu'k^2}{\omega_0'}\right) \tag{18}$$

for the k contribution to mean-square roughness. For $\omega_0' \gg \nu' k^2$ equation (18) reduces to $\langle \zeta_k'^2 \rangle \approx k_B T/[(\rho'/k)(\omega_0')^2]$, the well known result for an ideal liquid. Using $\omega_0'^2 = gk + \alpha' k^3/\rho'$, the criterion for validity of the ideal-liquid approximation can be written

$$k \ll \alpha'/(4\rho'\nu'^2) = k_c' \tag{19}$$

where k_c' is the critical damping wave vector for capillary waves of a viscous fluid. k_c' plays the same role at the surface as k_c plays at the interface (see (15)).

2.2.3. Surface roughness of water. We apply (18) to a water surface, using the parameters $\alpha' = 72$ dyn cm^{-1}, $\rho' = 1$ g cm^{-3} and $\nu' = 0.01$ cm^2 s^{-1} at 20 °C, and the k-cutoffs: $k_{max} = k_c' \approx 1.8 \times 10^{-3}$ Å^{-1} and $k_{min} \approx 10^{-8}$ Å^{-1}. The result $\langle \zeta'^2 \rangle^{1/2} \approx 2.3$ Å agrees with the x-ray reflectometry data [26] but is somewhat lower than the value 3.8 Å obtained using the standard capillary wave theory with the usual cutoffs: $k_{max} \approx 0.5$ Å^{-1} and $k_{min} \approx 10^{-8}$ Å^{-1} [27].

3. Summary and discussion

We have discussed the viscosity damping effect on capillary waves using the linearized Navier–Stokes equation. The main result of this analysis is a high k cutoff determined directly from the k-dependence of viscosity damping. The transition to extremely large damping can be characterized by the critical damping wave number k_c for the interfacial roughness (and by k_c' for the surface roughness). The roughness contribution in the high k region, where $k > k_c$ (or $k > k_c'$), can be neglected. When the interfacial tension is low, as in an oil–water–surfactant system, or the fluid viscosity is large, as in melted diblock-copolymer films [28], k_c is so low that most of the capillary wave spectrum is strongly damped, thus leading to a small interfacial roughness. This result is consistent with an analysis based on potential flow (ideal liquids), only if in this approximation a phenomenological low k cutoff is introduced, as in [19] and [20]. (In [19] and [20], this cutoff was physically justified by the requirement of limited horizontal slip at the interface.)

Our conclusion on the roughness of a liquid–liquid interface of low interfacial tension disagrees with several previous studies using x-ray [22] and neutron reflectometry [23], which have reported interfacial roughness as large as about 100 Å. The measured roughnesses in our neutron reflectometry experiments [19, 20] agree with the calculations presented here. It seems possible that the wider transition regions between the fluids observed in other studies are due mainly to the presence of a physically wider mixed interfacial layer, rather than to large capillary wave fluctuations.

We have also shown that the damping at a liquid–liquid interface must be characterized by the individual hydrodynamic parameters of both liquids, and cannot simply be deduced, as has been done in some cases [23], from the single-liquid result by replacing the density of the liquid by the difference in density and the surface tension by the interface tension.

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