

Metal-pad-roll instability theory for small-scale models of reduction cells

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We present a theoretical model for the metal-pad-roll instability in laboratory-scale experimental models of reduction cells consisting of two stably stratified liquid layers subjected to a vertical electric current and magnetic field. In contrast to most previous studies, we do not rely on the shallow-water approximation and can account for viscous and capillary effects that are prevalent in small-scale experiments. In small cells, dissipation effects are largely attributable to laminar viscous damping, which we calculate analytically using a perturbative formulation of the Stokes boundary layers forming at all container walls and on both sides of the liquid interface. The derived damping rates, which are also useful for modeling liquid-liquid sloshing in rectangular containers, are validated against direct numerical simulations conducted with the coupled level set and volume-of-fluid solver ARCHER. On this basis, we derive analytical solutions for the growth rates and instability onsets of the metal-pad-roll instability, which we compare with different theoretical descriptions and existing experiments. The presented solutions are intended to serve as a foundation for future benchmarking of multiphase solvers.

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I. INTRODUCTION

Aluminum reduction cells (ARCs) are known to be vulnerable to the metal-pad-roll (MPR) instability, and physical modeling can lead to strategies that help keep the instability under control [1,2]. In terms of hydrodynamics, ARCs can be regarded as stably stratified systems comprising two liquid layers: an upper layer of alumina dissolved in molten cryolite (Na_3AlF_6) and a lower layer of molten aluminum, both carrying a strong vertical electric current, see Fig. 1(a). Many studies have shown that the most critical instability results from the interaction of horizontal electrical currents in the aluminum layer and the vertical component of an external magnetic field generating Lorentz forces [2, Chap. 6.1]. As illustrated in Fig. 1(b), external magnetic fields are induced by the power supply lines in the aluminum smelter and cannot be avoided during operation. In this paper, we use the term MPR instability to refer to this particular scenario, in line with the majority of the literature. The MPR instability causes a rotating sloshing motion in the aluminum (metal pad) that must be strictly controlled throughout the electrolysis. As indicated in Fig. 1(a), the cells

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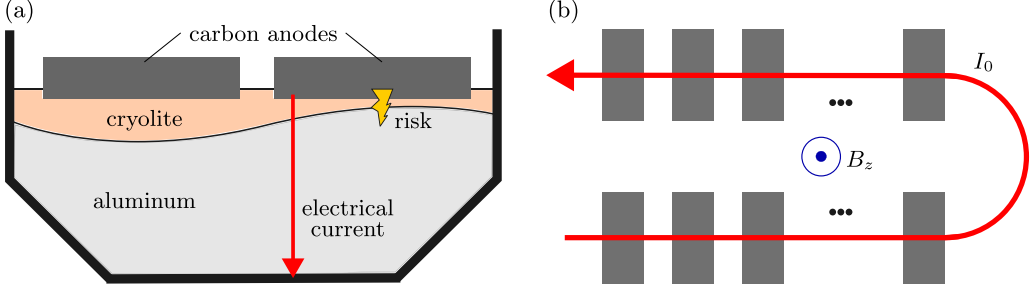


FIG. 1. (a) Side view of an aluminum reduction cell (ARC) with two superposed liquid layers. Metal-pad-roll instability destabilizes waves on the interface and can cause short-circuits. (b) ARCs are typically arranged around a central current loop in an industrial smelter, and this creates an ambient magnetic field B_z of typically a few mT.

can be short-circuited if the liquid aluminum layer makes contact with the upper carbon anodes. For ARCs to remain stable with respect to the MPR instability, the cryolite layer must not be thinner than about 3.5–4.5 cm [3]. This is, however, very unfavorable energetically as important Ohmic losses occur in the poorly conducting cryolite. About 40% of a cell's electrical energy does not contribute to aluminum reduction and is converted into waste heat. Keeping in mind that aluminum production consumes around 8% of the global industrial electricity consumption [4], the importance of understanding and controlling MPR instability is evident. Recently, there has also been interest regarding the fundamental aspects of the instability in the context of liquid metal batteries (LMBs): a promising technology for grid-scale energy storage applications, intended to accommodate fluctuations in the supply of energy from renewable sources. Flow instabilities occurring inside this molten metal-salt system can have a crucial impact on the cell's efficiency and operational safety [5–7].

The first successful attempts to explain the MPR instability mechanism go back to the studies of Urata [8] and Sele [9]. On the basis of some heuristic arguments, Sele succeeded in deducing an essential nondimensional parameter that controls the stability

$$\beta_S = \frac{I_0 B_z}{(\rho_a - \rho_c) g h_a h_c}, \quad (1)$$

today referred to as the Sele parameter. In this parameter, I_0 and B_z denote the total cell current and the vertical magnetic field, g is gravity, and ρ_a , ρ_c and h_a , h_c refer to the densities and heights of the aluminum and cryolite layers. According to Sele, ARCs are MPR-unstable when β_S exceeds a critical value, β_{crit} , which depends on the lateral cell geometry, but also on the viscous or turbulent damping. The scaling behavior $\beta_S \sim 1/h_c$ explains why we need a thick enough cryolite layer.

Much progress in the theoretical modeling of the MPR instability was made in the 1990s [10–12]. Perturbation techniques were applied to study the linear stability in the framework of the shallow-water approximation. It was revealed that the Lorentz force can couple different standing gravity wave numbers and destabilize them in pairs, much like in a parametric resonance of waves. The MPR instability is triggered when the natural frequencies ω and ω' of at least two transverse gravity wave numbers are sufficiently close to one another. The effect of the magnetohydrodynamic coupling is that ω and ω' are shifted towards each other for increasing $I_0 B_z$ until they coincide at $\beta_S = \beta_{\text{crit}}$. If $I_0 B_z$ is further increased ($\beta_S > \beta_{\text{crit}}$), the wave-pair becomes unstable and grows exponentially to become a rotating metal pad wave. The stability threshold β_{crit} scales with the difference between the squared eigenfrequencies $\beta_{\text{crit}} \sim |\omega^2 - \omega'^2|$ [12]. Cells in which pairs of gravity waves with identical natural frequencies $\omega = \omega'$ exist (e.g., this is the case for square and cylindrical cell geometries) are always unstable in the inviscid limit; the stability threshold in such cells only depends on the viscous or turbulent damping. An alternative view on the origin of the

MPR instability was given by Lukyanov *et al.* [13], who argued that the instability is the result of multiple self-amplifying wave reflections on the sidewalls. According to Lukyanov *et al.* [13], the MHD coupling mechanism is inherently connected to the lateral walls, explaining the vocabulary of *wall modes* introduced by Molokov *et al.* [14]. Jha *et al.* [15] studied the effect of horizontal and vertical magnetic fields on the destabilization of the metal pad, leading to the creation of standing waves. In addition to analytical studies, considerable effort was devoted to developing nonlinear numerical models [2,16–18] that can describe wave dynamics well beyond the stability threshold, e.g., for estimating saturation amplitudes. This has allowed ARC modeling to reach a certain level of maturity, and a great deal of effort has gone into the optimization of magnetic field and electric current distributions [19] to minimize the MPR instability.

Conducting well-controlled experiments on MPR instability in ARCs and LMBs is difficult due to the high temperature ($\sim 1000^\circ\text{C}$) and chemical aggressiveness. For this reason, several attempts have been made to develop downscaled model experiments that can synthesize or imitate MPR waves under ambient conditions. The pioneering experiment was devised by Pedcenko *et al.* [20,21], who replaced the cryolite layer with a grid of vertical thin rods imitating its electrical role. The aluminum layer was replaced by the eutectic alloy GaInSn that is liquid at room temperature. The experiment succeeded in demonstrating the MPR instability under a high imposed magnetic field, but also suffered some limitations. The steel rods were quite intrusive, and the instability was artificially intensified when the GaInSn detached from them under wave motion. This experiment also cannot be employed as a benchmark for MPR theories that consider two fluid layers. At this point, we would like to take the opportunity to highlight that another very sophisticated MPR experiment has been done using two immiscible transparent solutions of nitric acid ($\text{HNO}_{3(\text{aq})}$ at the bottom and $\text{HNO}_3 + \text{C}_5\text{H}_{12}\text{O}$ at the top). Unfortunately, this work of Borisov *et al.* [22] has only been published in a Russian-language journal, and therefore it has been overlooked. In this setup, both liquids with very similar densities were used to destabilize the interface with small currents in the presence of strong magnetic fields. This is the only study that has ever reported experimental stability onsets, including the associated unstable wave pairs as a function of the lateral aspect ratio of the cell. A more realistic MPR experiment setup using liquid gallium and aqueous electrolyte was conducted by Grants and Baranovskis [23], who observed a sustained rotating surface wave instead of a growing interfacial wave. The authors demonstrated that the horizontal rotation of the liquids significantly delayed the onset of the MPR instability. A similar two-layer setup using Ga-Hg liquid metal pairs was proposed [24], but no experiment has been done yet.

Recently, Hegde *et al.* [25] have proposed a new small-scale MPR experiment that works with one liquid layer of GaInSn. The MPR instability is synthesized through a feedback loop control that injects horizontal electrical currents in the GaInSn-layer, proportional to the measured slope. The experiment imitates the physics of the Sele-mechanism, but the feedback mechanism still needs to be carefully calibrated to imitate the MPR physics of two-layer cells. Another recent experiment is that of Eltishchev *et al.* [26,27], who destabilized rotating surface waves through a MHD forcing mechanism that is quite similar to that of the MPR instability.

Spurred on by these recent experimental advances and the rediscovery of Borisov's experiment, we present here a linear stability theory that is specially dedicated to MPR instability in small-scale experimental models. In small-scale setups, we must account for physical ingredients that are usually neglected in ARCs, and this requires a specific theory. We can have nonshallow layers, significant viscous damping, possibly magnetic damping (for high B_z), and low differences in electrical conductivity between the two liquid layers. Herreman *et al.* [28] proposed a linear stability theory for small cylindrical cells, whereas we present here a similar theory for small, rectangular cells. A perturbative approach was also used in Politis and Priede [29], but wave-damping there was modeled in a phenomenological way. In this study, we propose a laminar viscous damping model that has no adjustable parameters and can also be compared to previous work on viscous damping of waves in two-layer systems [30,31]. A linear stability model consisting of equations for the growth rate and the viscous damping rate will be essential in benchmarking numerical solvers and existing MPR experiments, as well as for developing new ones.

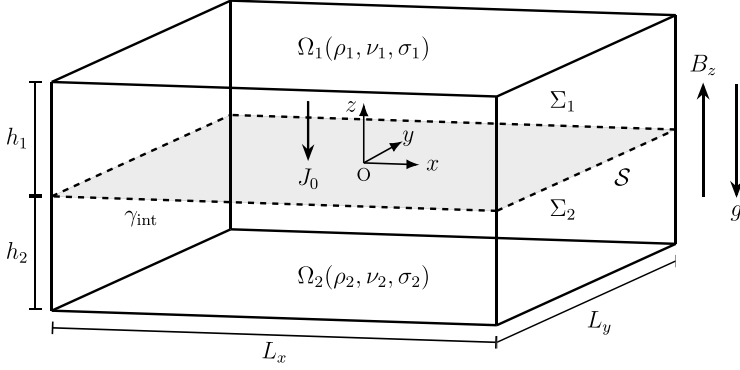


FIG. 2. Schematic of the cell at equilibrium consisting of two fluid layers Ω_1 (upper), Ω_2 (lower) characterized by their densities ρ_1, ρ_2 , kinematic viscosities ν_1, ν_2 , electrical conductivities σ_1, σ_2 , thicknesses h_1, h_2 , and bounded by rigid boundaries (container walls) Σ_1, Σ_2 , respectively. Gravity g acts in the negative z direction, stratifying the two media with an interface S located at $z = 0$ and characterized by interfacial tension γ_{int} . Both layers are traversed by a homogeneous vertical electrical current density J_0 and submitted to a homogeneous vertical magnetic field B_z .

The manuscript is organized as follows: In Sec. II we calculate the growth rate of the MPR instability in small cells using perturbation methods and including a model for viscous dissipation. In Sec. III we validate our new two-layer damping formula numerically with the multiphase DNS code ARCHER, and we also compare our damping rates to the experimental and numerical results of La Rocca *et al.* [31]. We then calculate the theoretical onsets of stability as a function of cell geometry and compare with the theory of Politis and Priede [29]. We also apply our stability theory to the two-layer experimental setup of Borisov *et al.* [22]. In Sec. IV we collate our results and discuss open questions.

II. LINEAR STABILITY ANALYSIS

A. Formulation of the problem

We consider a system of two superposed immiscible, incompressible, and Newtonian liquid layers (Ω_1 and Ω_2) at equilibrium in a rectangular domain (x, y, z) of dimensions L_x, L_y and $H = h_1 + h_2$, as shown in Fig. 2. In this state, both the fluids are at rest. For both layers, the density ρ_i , kinematic viscosity ν_i , and electrical conductivity σ_i are taken into account. The subscripts $i = 1, 2$ refer to the upper and lower layers, respectively. The origin of the coordinate system O is located at the center of the interface. The rigid boundaries of each layer are denoted by Σ_i . We include interfacial tension γ_{int} in our model, which acts in addition to gravity g as a restoring force, but we neglect capillary effects at the side walls, i.e., we assume that the contact line slides freely along the side walls while maintaining a static contact angle of 90° . The entire cell is exposed to an external homogeneous magnetic field oriented vertically B_z , and gravity acts in the negative z direction $\mathbf{g} = -g\mathbf{e}_z$. At equilibrium, both layers carry a homogeneous vertical electrical current density $J_0 = -I_0/(L_x L_y)$, where I_0 denotes the total cell current. The equilibrium configuration is fully characterized by the hydrodynamic pressure $P_i = P_0 - \rho_i g z$, and the electrical potential $\Phi_i = \Phi_0 - J_0 z / \sigma_i$, where P_0 and Φ_0 are constants. In the perturbed state, the total current density $\mathbf{J}(x, y, z, t) = J_0 \mathbf{e}_z + \mathbf{j}_i(x, y, z, t)$ takes into account the excess currents $\mathbf{j}_i$ and the total magnetic field strength $\mathbf{B}(x, y, z, t) = B_z \mathbf{e}_z + \mathbf{b}_i(x, y, z, t)$ takes into account the induced magnetic fields $\mathbf{b}_i$. The perturbed interface S is located at $z = \eta(x, y, t)$.

B. Governing equations and boundary conditions

The quantities $(\mathbf{u}_i, p_i, \mathbf{j}_i, \varphi_i, \mathbf{b}_i)$, for the perturbations of velocity, pressure, current density, electrical potential, and induced magnetic field which modify the equilibrium state [28,32], are governed by the following equations:

$$\rho_i \partial_t \mathbf{u}_i + \nabla p_i = \mathbf{j}_i \times B_z \mathbf{e}_z + J_0 \mathbf{e}_z \times \mathbf{b}_i + \rho_i \nu_i \nabla^2 \mathbf{u}_i, \quad (2a)$$

$$\nabla \cdot \mathbf{u}_i = 0, \quad (2b)$$

$$\nabla \cdot \mathbf{j}_i = 0, \quad (2c)$$

$$\mathbf{j}_i = \sigma_i (-\nabla \varphi_i + \mathbf{u}_i \times \mathbf{B}), \quad (2d)$$

$$\nabla \times \mathbf{j}_i = -\sigma_i \frac{\partial \mathbf{b}_i}{\partial t} + \nabla \times \sigma_i (\mathbf{u}_i \times \mathbf{B}), \quad (2e)$$

$$\nabla \times \mathbf{b}_i = \mu_0 \mathbf{j}_i, \quad \nabla \cdot \mathbf{b}_i = 0. \quad (2f)$$

Equation (2a) is the linearized conservation of momentum relation with Lorentz forces $\mathbf{j}_i \times B_z \mathbf{e}_z$ and $J_0 \times \mathbf{b}_i$ as source terms; Eq. (2b) expresses the continuity of velocity and (2c) the excess current. Equation (2d) refers to the quasimagnetostatic approximation of Ohm's law [1]. Equations (2d) and (2e) (Faraday's law), and (2f) (Gauss's law) together give the following induction equation:

$$\frac{\partial \mathbf{b}_i}{\partial t} - \nabla \times (\mathbf{u}_i \times \mathbf{B}) - \frac{\nabla^2 \mathbf{b}_i}{\mu_0 \sigma_i} = 0, \quad (3)$$

where $1/(\mu_0 \sigma_i)$ is the magnetic diffusivity for a given material (μ_0 is the permeability of free space). To ascertain the limits of our theoretical model under the magnetostatic approximation, we introduce two dimensionless quantities to characterize the system, namely the Reynolds number Re and the magnetic Reynolds number R_m ,

$$\text{Re} = \omega L^2 / \nu, \quad \text{R}_m = \mu_0 \sigma \omega L^2. \quad (4)$$

The characteristic velocity can be written as a product of the interfacial wave frequency ω and the characteristic length of the geometry $L = \sqrt{L_x L_y}$. The Reynolds number Re is assumed to be within the limits $100 \leq \text{Re} < 10^4$, where irrotational waves are known to be linear and laminar. The magnetic Reynolds number R_m can be expressed as the ratio of the magnetic diffusion time $\tau_m = \mu_0 \sigma L^2$ and the flow time $\tau_f = \omega^{-1}$. At low $\text{R}_m \ll 1$, magnetic diffusion $\nabla^2 \mathbf{b}_i / \mu_0 \sigma_i$ is much faster than the advection of the magnetic field due to fluid velocity ($\nabla \times [\mathbf{u}_i \times \mathbf{B}]$). In this scenario, the field achieves equilibrium instantaneously relative to the global time scale. Therefore, we can neglect the terms $\partial \mathbf{b}_i / \partial t$ in Eq. (3). As $\mathbf{b}_i$ scales with R_m , they ensure that the induced fields are comparatively small, thereby reducing $\mathbf{u} \times \mathbf{B} \approx 0$. Previous studies based on shallow water theories have largely ignored the contribution of the induced magnetic field on the perturbed Lorentz force [11,12]. In a cylindrical system, it was shown that $J_0 \times \mathbf{b}_i$ does not contribute to the amplitude of the wave but rather to the frequency shift [28]. This is because the Lorentz force due to the horizontal component of the induced magnetic field $J_0 \mathbf{e}_z \times \mathbf{b}_i$ is out of phase with $\mathbf{j}_i \times B_z \mathbf{e}_z$ (the dominant contribution) and therefore cannot inject power into the wave. These assumptions effectively reduce the magnetic problem to a static case in both the liquid layers. Therefore, the conditions $\partial \mathbf{b}_i / \partial t \approx 0$, $\mathbf{u} \times \mathbf{B} \approx 0$, and $J_0 \times \mathbf{b}_i \approx 0$ allow us to impose the magnetostatic approximation on the system where the electric field only depends on the electric potential.

For the formulation of the boundary conditions at the tank walls, we account for the fact that the upper carbon anode in ARCs is a much better electrical conductor than the cryolite, and the bottom steel cathode often a much poorer conductor than aluminum. This is a standard assumption in MPD theories [2,33], and we then obtain the following linearized boundary conditions:

$$\mathbf{j}_1 \cdot \mathbf{e}_x|_{x=\pm L_x/2} = \mathbf{j}_2 \cdot \mathbf{e}_x|_{x=\pm L_x/2} = 0, \quad (5a)$$

$$\mathbf{j}_1 \cdot \mathbf{e}_y|_{y=\pm L_y/2} = \mathbf{j}_2 \cdot \mathbf{e}_y|_{y=\pm L_y/2} = 0, \quad (5b)$$

$$\varphi_1 = 0 \big|_{z=h_1}, \quad (5c)$$

$$\mathbf{j}_2 \cdot \mathbf{e}_z = 0 \big|_{z=-h_2}. \quad (5d)$$

Equations (5a) and (5b) ensure that no current can escape through the insulating side walls. The anode is modeled as an isopotential surface [Eq. (5c)] and the weakly conducting cathode is treated as a perfect insulator [Eq. (5d)] for the excess current component $\mathbf{j}$. In Sec. III D, we further consider a case where the cathode is modeled as a perfect conductor. In that respect, condition [Eq. (5d)] is not valid for all reduction cells and MPR model experiments. As for the kinematic boundary conditions, we simply apply impermeability at all walls,

$$\mathbf{u}_i \cdot \mathbf{n}_i \big|_{\Sigma_i} = 0, \quad (6)$$

for the inviscid formulation (no slip condition) in Sec. II D, to which we must add

$$\mathbf{u}_i \times \mathbf{n}_i \big|_{\Sigma_i} = 0 \quad (7)$$

to account for viscous effects, we address in Sec. II E. At the interface $\mathcal{S}$, we have to take into account the following linearized hydrodynamic and electrical boundary conditions:

$$\partial_t \eta = u_{1,z} \big|_{z=0} = u_{2,z} \big|_{z=0}, \quad (8a)$$

$$-\gamma_{\text{int}} \nabla^2 \eta + (\rho_2 - \rho_1) g \eta = p_2 \big|_{z=0} - p_1 \big|_{z=0}, \quad (8b)$$

$$\mathbf{j}_1 \cdot \mathbf{e}_z = \mathbf{j}_2 \cdot \mathbf{e}_z \big|_{z=0}, \quad (8c)$$

$$J_0 (\sigma_2^{-1} - \sigma_1^{-1}) \eta = \varphi_2 \big|_{z=0} - \varphi_1 \big|_{z=0}. \quad (8d)$$

Equations (8a) and (8b) represent the classic kinematic and dynamic boundary conditions for gravity-capillary waves. In Sec. II E, we state the corrections to Eq. (8b) which incorporate the continuity of the tangential velocities and viscous stresses in order to solve the Stokes velocities (for viscous damping). The electrical boundary conditions are derived from the basic principle that both the normal of the total current density $\mathbf{n} \cdot \mathbf{J}$ and the total electric potential ($\Phi_i + \varphi_i$) must remain continuous. This results in the linearized interfacial conditions [Eqs. (8c) and (8d)] for the perturbed quantities [34]. This jump condition is essential for the MPR instability as it describes how the current density redistributes in both layers as a response to an arbitrary (small) interface elevation η .

C. Interfacial waves

We intend to solve the linear stability problem using perturbation theory, supposing the Lorentz and viscous forces are small. At lowest order, in the unperturbed problem for $B_z = 0$ and $v_i = 0$, the hydrodynamic problem is decoupled from the electrical problem. We then have free gravity-capillary waves, and each different wave creates a deviation of electric current. The solutions for the classical gravity-capillary waves can be obtained by expressing the bulk velocities as gradients of scalar potentials $\mathbf{u}_i = \nabla \phi_i$ and solving the Laplace equation $\nabla^2 \phi_i = 0$ together with the no outflow boundary condition [Eq. (6)] and the interface conditions given in Eqs. (8a) and (8b) [35]. The flow potential in both layers is given by

$$\begin{bmatrix} \phi_1 \\ \phi_2 \end{bmatrix} = A_{mn} \begin{bmatrix} \frac{\omega}{k} \frac{\cosh[k(z-h_1)]}{\sinh[kh_1]} \\ -\frac{\omega}{k} \frac{\cosh[k(z+h_2)]}{\sinh[kh_2]} \end{bmatrix} \cos \left[\frac{m\pi}{L_x} \left(x + \frac{L_x}{2} \right) \right] \cos \left[\frac{n\pi}{L_y} \left(y + \frac{L_y}{2} \right) \right] e^{i\omega t}, \quad (9)$$

along with the interface elevation function reading

$$\eta = i A_{mn} \cos \left[\frac{m\pi}{L_x} \left(x + \frac{L_x}{2} \right) \right] \cos \left[\frac{n\pi}{L_y} \left(y + \frac{L_y}{2} \right) \right] e^{i\omega t}, \quad (10)$$

and the pressure $p_i = -\rho_i(i\omega)\phi_i$. Here, A_{mn} is a mode-dependent wave amplitude. The wave number k is a function of the mode indices (m, n) , which are the number of half-wavelengths along the x

and y axes given by

$$k = \pi \sqrt{\frac{m^2}{L_x^2} + \frac{n^2}{L_y^2}}. \quad (11)$$

The natural frequencies of the gravity-capillary waves are

$$\omega = \pm \sqrt{\frac{(\rho_2 - \rho_1)gk + \gamma_{\text{int}}k^3}{\rho_1 \coth[k h_1] + \rho_2 \coth[k h_2]}}. \quad (12)$$

The wave perturbs the interface and causes deviation of the electrical current. We calculate the current perturbations through Ohm's law, $\mathbf{j}_i = -\sigma_i \nabla \varphi_i$. The electric potentials must also fulfill the Laplace equation $\nabla^2 \varphi_i = 0$ with boundary conditions given in Eqs. (5a)–(5d), (8c), and (8d). We find the solutions to the electrical potential to be

$$\begin{bmatrix} \varphi_1 \\ \varphi_2 \end{bmatrix} = J_0 \Lambda \eta \begin{bmatrix} -\frac{\sinh[k(z-h_1)]}{\sigma_1 \cosh[kh_1]} \\ -\frac{\cosh[k(z+h_2)]}{\sigma_2 \sinh[kh_2]} \end{bmatrix}. \quad (13)$$

Here, we have introduced Λ , which can be interpreted as the conductivity jump parameter

$$\Lambda = \frac{(\sigma_1^{-1} - \sigma_2^{-1})}{\sigma_1^{-1} \tanh[kh_1] + \sigma_2^{-1} \coth[kh_2]}. \quad (14)$$

The parameter does not appear in most MPR models that assume $\sigma_2 \gg \sigma_1$ which gives $\Lambda \approx (kh_1)^{-1}$. This assumption is not always guaranteed in model MPR experiments [24], which is why we need to include the conductivity jump parameter in our calculations.

D. Perturbative calculation of the inviscid growth rate of the MPR instability

This perturbative approach is based on the (inviscid) potential flow solutions of free gravity-capillary interfacial waves that form a set of orthogonal functions in the absence of MHD effects ($\mathbf{B} = \mathbf{0}$) and without viscosity ($\nu = 0$). In this section, we calculate the inviscid growth rate. The effect of viscous dissipation due to the Stokes boundary layers forming at the container walls, and below and above the interface, are presented in Sec. II E.

In contrast to most previous studies, which examine the interaction of infinitely many wave numbers and truncate the system *a posteriori* [10, 11], we calculate the two-wave interaction directly allowing us to derive exact analytical solutions for the growth rates and instability onsets. The disadvantage of this method is that the MPR instability can sometimes arise from three (or more) -wave interactions [11, 18], and these cases are not represented in our model. A standard two-wave ansatz is given as follows:

$$[\mathbf{u}_i, p_i, \eta, \mathbf{j}_i] = (C[\widehat{\mathbf{u}}_i, \widehat{p}_i, \widehat{\eta}, \widehat{\mathbf{j}}_i] + C'[\widetilde{\mathbf{u}}'_i, \widetilde{p}'_i, \widetilde{\eta}', \widetilde{\mathbf{j}}'_i] + [\widetilde{\mathbf{u}}_i, \widetilde{p}_i, \widetilde{\eta}, \widetilde{\mathbf{j}}_i]) e^{(\alpha + i\bar{\omega})t}. \quad (15)$$

In this notation, the tilded variables are small perturbations with respect to the hatted variables representing the spatial structure of the free waves. The bare and the dashed (') hatted variables denote the variables corresponding to the two wave modes (m, n) and (m', n') accordingly. The values C and C' are to be determined and α is the inviscid growth rate that we seek to derive. We assume that both waves are resonantly coupled by the Lorentz force $\mathbf{j}_i \times B_z \mathbf{e}_z$, and this requires that they both oscillate with the same mean frequency. We represent the mean natural frequency of the superposed wave as

$$\bar{\omega} = (\omega + \omega')/2, \quad (16a)$$

and the frequency difference as

$$\delta\omega = (\omega - \omega')/2. \quad (16b)$$

We suppose the frequency detuning $\delta\omega$ to be small. At perfect resonance, we have the frequency difference $\delta\omega = 0$.

We insert the ansatz [Eq. (15)] in the governing Eqs. (2a) and (2b), ignoring viscosity. After using the equations for the free gravity wave problem, we find

$$C(i\bar{\omega} + \alpha)\rho\hat{\mathbf{u}} + C\nabla\hat{p} + C'(i\bar{\omega} + \alpha)\rho\hat{\mathbf{u}}' + C'\nabla\hat{p}' + (i\bar{\omega})\rho\tilde{\mathbf{u}} + \nabla\tilde{p} + (i\bar{\omega})\rho\tilde{\mathbf{u}}' + \nabla\tilde{p}' \\ = [C(\hat{\mathbf{j}} \times B_z \mathbf{e}_z) + C'(\hat{\mathbf{j}}' \times B_z \mathbf{e}_z)], \quad (17a)$$

$$\nabla \cdot \tilde{\mathbf{u}}_i = 0. \quad (17b)$$

To find an equation for α , we have to state a solvability condition. For that we apply the Fredholm alternative in a manner analogous to Herreman *et al.* [28]. All technical calculations are provided in the Supplemental Material [36, Suppl. III]. The solvability condition results in an eigenvalue problem in the form of a 2×2 interaction matrix that can be expressed in the following form:

$$\begin{bmatrix} i\delta\omega - \alpha & \frac{I_{\eta\eta'}}{2K_{\eta\eta}} \\ \frac{I_{\eta'\eta}}{2K_{\eta'\eta'}} & -i\delta\omega - \alpha \end{bmatrix} \begin{bmatrix} C \\ C' \end{bmatrix} = 0. \quad (18)$$

The terms

$$I_{\eta\eta'} = \sum_{i=1,2} \int_{V_i} \hat{\mathbf{u}}_i^* \cdot (\hat{\mathbf{j}}_i \times B_z \mathbf{e}_z) dV = \sum_{i=1,2} \int_{V_i} \nabla \hat{\phi}_i^* \cdot (-\sigma_i \nabla \hat{\phi}_i' \times B_z \mathbf{e}_z) dV, \quad (19a)$$

$$I_{\eta'\eta} = \sum_{i=1,2} \int_{V_i} \hat{\mathbf{u}}_i'^* \cdot (\hat{\mathbf{j}}_i \times B_z \mathbf{e}_z) dV = \sum_{i=1,2} \int_{V_i} \nabla \hat{\phi}_i'^* \cdot (-\sigma_i \nabla \hat{\phi}_i \times B_z \mathbf{e}_z) dV, \quad (19b)$$

are associated with power transfers between waves mediated by Lorentz forces. The terms $K_{\eta\eta}$ and $K_{\eta'\eta'}$ are associated with the potential (or equivalently kinetic) energy of the respective wave mode (m, n) and are given by

$$K_{\eta\eta} = \int_S (\gamma_{\text{int}} |\nabla \hat{\eta}|^2 + (\rho_2 - \rho_1) g |\hat{\eta}|^2)|_{z=0} dS = \frac{L_x L_y}{2\delta_{m \times n}} [\gamma_{\text{int}} k^2 + (\rho_2 - \rho_1) g], \quad (20)$$

and similarly for $K_{\eta'\eta'}$ with m' , n' , and k' . For the sake of simplicity, we introduced an auxiliary function δ_r that is defined as follows:

$$\delta_r = \begin{cases} 1, & r = 0, \\ 2, & r > 0. \end{cases} \quad (21)$$

Finally, the secular equation of the matrix [Eq. (18)] yields the solutions for the inviscid α , one of the central results of our study given by

$$\alpha = \pm \sqrt{\frac{I_{\eta\eta'} I_{\eta'\eta}}{4K_{\eta\eta} K_{\eta'\eta'}} - \delta\omega^2}. \quad (22)$$

On solving the integrals in Eqs. (19a) and (19b), we obtain the explicit formulation

$$\alpha = \pm \sqrt{J_0^2 B_z^2 \Theta^2 (\bar{\omega}^2 - \delta\omega^2) \frac{\mathcal{T}\mathcal{T}'}{\mathcal{U}\mathcal{U}'}} - \delta\omega^2, \quad (23)$$

where

$$\Theta = \sqrt{\delta_{m \times m'} \delta_{n \times n'}} \frac{[(-1)^{m+m'} - 1][(-1)^{n+n'} - 1](m^2 n'^2 - m'^2 n^2)}{(m^2 - m'^2)(n^2 - n'^2)}. \quad (24a)$$

The term Θ is nonzero only if the parity conditions

$$m + m' = 2l + 1 \quad \text{and} \quad n + n' = 2l + 1, \quad l \in \mathbb{Z}, \quad (24b)$$

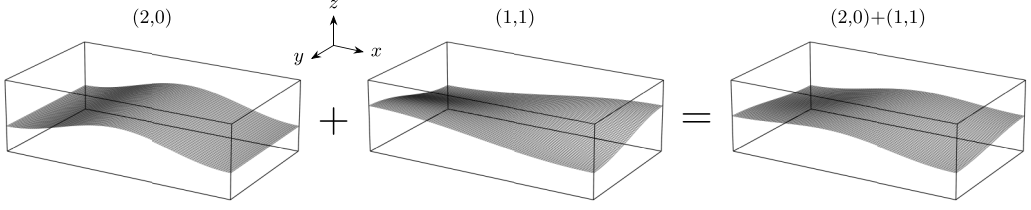


FIG. 3. An illustration of a linear interfacial wave in a rectangular system of lateral aspect ratio equal to 2. For example, we show the superposition of two standing waves of modes (2,0) and (1,1) to produce a rotating interfacial wave of mode (2, 0) + (1, 1).

are satisfied at the same time. These conditions allow for a preselection of wave pairs which can actually couple magnetohydrodynamically and thereby trigger the MPR instability. The terms

$$\mathcal{U} = \gamma_{\text{int}} k^2 + (\rho_2 - \rho_1)g, \quad \mathcal{U}' = \gamma_{\text{int}} k'^2 + (\rho_2 - \rho_1)g, \quad (24c)$$

are representations of the potential energy of the standing waves, and

$$\mathcal{T} = \frac{\Lambda}{k(k^2 - k'^2)L_x L_y} \left(\frac{k}{\tanh[k'h_2]} - \frac{k'}{\tanh[kh_2]} + \frac{k' \operatorname{sech}[k'h_1]}{\sinh[kh_1]} - \frac{k'}{\tanh[kh_1]} + k \tanh[k'h_1] \right), \quad (24d)$$

$$\mathcal{T}' = \frac{\Lambda'}{k'(k'^2 - k^2)L_x L_y} \left(\frac{k'}{\tanh[kh_2]} - \frac{k}{\tanh[k'h_2]} + \frac{k \operatorname{sech}[kh_1]}{\sinh[k'h_1]} - \frac{k}{\tanh[k'h_1]} + k' \tanh[kh_1] \right), \quad (24e)$$

arise from the power injected by the Lorentz force and contain the finite-depth dependencies of the cell currents redistributed by interface elevations. When $k' \rightarrow k$, these terms simplify to

$$\mathcal{T} = \mathcal{T}' = \frac{\Lambda}{4k^2 \sinh^2[kh_2]L_x L_y} \left(\frac{\sinh[k(h_1 + 2h_2)]}{\cosh[kh_1]} + 2kh_2 - \tanh[kh_1] \right). \quad (24f)$$

The perturbation analysis results in two distinct solutions for the growth rate α . The two solutions correspond to MPR waves that rotate either clockwise or counterclockwise (when looking at the cell from above), or equivalently, to the superposition of two standing-wave pairs $(\eta + \eta')$ with a phase shift, as shown in Fig. 3. One of the two wave solutions in Eq. (23) is always suppressed by the Lorentz force (when the induced Lorentz force $\mathbf{j} \times B_z \mathbf{e}_z$ opposes the rotating wave motion $\mathbf{u}$) and the other one is amplified in the unstable situations (when $\mathbf{j} \times B_z \mathbf{e}_z$ aligns with $\mathbf{u}$). In our configuration shown in Fig. 2, counterclockwise rotating waves are destabilized whenever the square root in Eq. (23) turns complex, fulfilled for

$$J_0^2 B_z^2 \Theta^2 (\bar{\omega}^2 - \delta\omega^2) \frac{\mathcal{T}\mathcal{T}'}{\mathcal{U}\mathcal{U}'} \geq \delta\omega^2. \quad (25)$$

From Eq. (25) one can readily derive the inviscid stability onset β_{crit} , which states that a system is unstable if

$$\beta_{\gamma_{\text{int}}} > \beta_{\text{crit}} = \frac{L_x L_y}{h_1 h_2 |\Theta|} \frac{\sqrt{\mathcal{U}\mathcal{U}'}}{\mathcal{U}^*} \sqrt{\frac{1}{\mathcal{T}\mathcal{T}'} \frac{\delta\omega^2}{(\bar{\omega}^2 - \delta\omega^2)}}, \quad (26)$$

where

$$\mathcal{U}^* = (\rho_2 - \rho_1)g + \gamma_{\text{int}} k k', \quad \text{and} \quad \beta_{\gamma_{\text{int}}} = \frac{I_0 B_z}{[(\rho_2 - \rho_1)g + \gamma_{\text{int}} k k'] h_1 h_2}, \quad (27)$$

is a modified Sele parameter, which takes into account interfacial tension γ_{int} in addition to gravity [6]. The stability criterion represented above in compact, dimensionless form suggests that all cells with matching frequency pairs ($\delta\omega = 0$) are always unstable, i.e., $\beta_{\text{crit}} = 0$ (when neglecting dissipation). In principle, such pairs can be found in all cell geometries, though often only for very large wave modes, which, when damping is taken into account, do not become destabilized in reality. In square cells, however, $\delta\omega = 0$ applies even to the first large-scale MPR wave mode $(1, 0) + (0, 1)$, which is why square cells are often referred to as inherently unstable.

E. The viscous correction of the growth rate

Up to this point, we have been looking at the MPR instability at the inviscid limit. In real cells, viscosity plays a crucial role in dissipating some of the energy injected in the system. To evaluate the stability of industrial ARCs, in which turbulent flows usually prevail, the damping rate due to viscous dissipation must be estimated or determined experimentally. In small-scale models, where flows remain largely laminar (at least in the boundaries or around the point of marginal instability), viscous damping rates can be calculated analytically. Here, viscosity is treated as a perturbation independent of the Lorentz force and the viscous growth rate α in the two-wave ansatz [Eq. (15)]. In concrete terms, we add small viscous corrections to the potential wave flows. To estimate the applicability and limits of this approach, we introduce two nondimensional parameters. First, it must be justified that viscous damping is the primary source of dissipation, i.e., magnetic damping effects can be neglected. This requires consideration of the Hartmann number

$$\text{Ha} = B_z L \sqrt{\frac{\sigma_i}{\rho \nu_i}}, \quad (28)$$

which compares the influence of Lorentz forces to the viscous forces. If the Ha number is sufficiently small, magnetic damping can be neglected. There is no fixed threshold in the literature, but an analysis of the magnetic damping theory by Sreenivasan *et al.* [37] reveals that, beginning at approximately $\text{Ha} \gtrsim 50$, magnetic dissipation contributes significantly to the overall dissipation. Industrial reduction cells operate at $\text{Ha} \approx 20$ [11], but the Ha numbers can be much higher in some MPR experiments, making it worthwhile for future studies to also take magnetic damping into account. Second, the approach of treating the boundary layer as small corrections to the irrotational bulk flow must be substantiated. We introduce a second dimensionless parameter Δ given by

$$\Delta = \frac{\sqrt{2\nu_i/\omega}}{L}, \quad L = \min(h_i, \sqrt{L_x L_y}), \quad (29)$$

which compares the width of the viscous boundary layer $\sim \sqrt{\nu_i/\omega}$ to the characteristic length of the domain. It is only when Δ is sufficiently small that the viscous boundary layers do not penetrate too much into the irrotational bulk flow. In such cases, the potential flow solutions remain valid in the leading order. Again, there is no strict threshold; it is sometimes stated in the literature that Δ must be clearly less than $\Delta < 0.1$. Our simulations in Sec. III A indicate that significant deviations occur when the ratio roughly exceeds $\Delta \gtrsim 0.05$.

With both these limits defined, we now model the viscous damping rate by calculating the total dissipation produced in the Stokes boundary layers, which are created solely by wave motions. We follow the approach by Keulegan [30], who has calculated damping rates of free-surface gravity waves in rectangular containers. We extend his formulation to two liquid layers, which is not straightforward as viscous boundary layers are formed above and below the interface in two-liquid systems that do not develop on free surfaces and must be calculated separately. In the absence of MHD effects, the linear governing Eqs. (2a) and (2b) reduce to

$$\left. \begin{aligned} \rho_i \partial_t \mathbf{u}_i + \nabla p_i &= \rho_i \nu_i \nabla^2 \mathbf{u}_i \\ \nabla \cdot \mathbf{u}_i &= 0 \end{aligned} \right\}. \quad (30)$$

For the upper and lower layer ($i = 1, 2$), the no-slip condition at the side wall (represented as rigid boundary Σ_i) is

$$\mathbf{u}_i|_{\Sigma_i} = 0, \quad (31a)$$

and the tangential velocities (denoted by $\perp$) must be equal at the interface:

$$\mathbf{u}_{1,\perp}|_S = \mathbf{u}_{2,\perp}|_S. \quad (31b)$$

The dynamic boundary condition [Eq. (8b)] under the consideration of viscous effects now changes to

$$p_2|_S - p_1|_S = (\rho_2 - \rho_1)g\eta - \gamma_{\text{int}}\nabla^2\eta + 2(\rho_2\nu_2\partial_z u_{2,z} - \rho_1\nu_1\partial_z u_{1,z}), \quad (31c)$$

while the continuity of the tangential stresses is

$$\rho_1\nu_1(\partial_z u_{1,x} + \partial_x u_{1,z})|_S = \rho_2\nu_2(\partial_z u_{2,x} + \partial_x u_{2,z})|_S, \quad (31d)$$

$$\rho_1\nu_1(\partial_z u_{1,y} + \partial_y u_{1,z})|_S = \rho_2\nu_2(\partial_z u_{2,y} + \partial_y u_{2,z})|_S. \quad (31e)$$

In a stratified two-layer system, consider an interfacial gravity wave mode of (m, n) of amplitude A_{mn} oscillating at angular frequency ω that undergoes viscous decay over time at a rate of λ . The fluid velocity taking into account the Stokes boundary layer corrections can be formulated as

$$\mathbf{u}_i = C[\widehat{\mathbf{u}}_i + \widehat{\bar{\mathbf{u}}}_i]e^{(i\omega - \lambda)t}, \quad (32)$$

where the $\widehat{\mathbf{u}}_i$ are the potential flow velocities, and the barred term $\widehat{\bar{\mathbf{u}}}_i$ are the viscous corrections to be calculated. We introduce wall-normal coordinates ζ that tend to decay the viscous boundary layer corrections to zero as they traverse inwards in the bulk. We define ζ as follows:

$$\zeta = \begin{cases} h_1 - z, & \text{near } z = h_1 \\ \frac{L_x}{2} - x, & \text{near } x = L_x/2 \\ \frac{L_y}{2} - y, & \text{near } y = L_y/2 \\ z, & \text{near } z = 0 \end{cases}, \quad \zeta = \begin{cases} z + h_2, & \text{near } z = -h_2 \\ x + \frac{L_x}{2}, & \text{near } x = -L_x/2 \\ y + \frac{L_y}{2}, & \text{near } y = -L_y/2 \\ -z, & \text{near } z = 0 \end{cases}. \quad (33)$$

The effect of the diffusive boundary layer tends to diminish in the bulk region. Inserting the ansatz [Eq. (32)] into Eq. (30), we obtain $\widehat{\bar{\mathbf{u}}}_i \cdot \mathbf{n}_i = 0$ and $\widehat{\bar{p}}_i = 0$, i.e., the wall-normal boundary layer correction for velocity and the leading-order boundary layer correction for pressure, both equal to zero. We compute the Stokes boundary layer at the domain walls (with no-slip condition) as

$$i\omega\widehat{\bar{\mathbf{u}}}_{i,\perp} \approx \nu_i\partial_\zeta^2\widehat{\bar{\mathbf{u}}}_{i,\perp}. \quad (34)$$

The mode-dependent velocity at the solid walls is

$$\widehat{\bar{\mathbf{u}}}_{i,\perp} = -\widehat{\mathbf{u}}_{i,\perp}|_{\Sigma_i}e^{-\Gamma\sqrt{|\omega|/\nu_i}\zeta}, \quad \{\Gamma = (1+i)/\sqrt{2}\}. \quad (35)$$

From Eq. (35) we can obtain the characteristic length of the viscous boundary layer of each medium as $\sqrt{\nu_i/|\omega|}$. At the interface, from Eqs. (31d) and (31e) we deduce

$$\widehat{\mathbf{u}}_{1,\perp} + \widehat{\bar{\mathbf{u}}}_{1,\perp}|_S \approx \widehat{\mathbf{u}}_{2,\perp} + \widehat{\bar{\mathbf{u}}}_{2,\perp}|_S \quad \begin{cases} \rho_1\nu_1\partial_z\widehat{u}_{1,x}|_S \approx \rho_2\nu_2\partial_z\widehat{u}_{2,x}|_S \\ \rho_1\nu_1\partial_z\widehat{u}_{1,y}|_S \approx \rho_2\nu_2\partial_z\widehat{u}_{2,y}|_S \end{cases}, \quad (36)$$

$$\widehat{\bar{\mathbf{u}}}_{1,\perp}|_{m,n} = \frac{\Lambda_v}{\rho_1\sqrt{\nu_1}} \frac{\omega}{k} \left[\begin{aligned} & \left[\frac{m\pi}{L_x} \sin\left[\frac{m\pi}{L_x}\left(x + \frac{L_x}{2}\right)\right] \cos\left[\frac{n\pi}{L_y}\left(y + \frac{L_y}{2}\right)\right] \mathbf{e}_x \right. \\ & \quad \left. + \right. \\ & \left. \left[\frac{n\pi}{L_y} \cos\left[\frac{m\pi}{L_x}\left(x + \frac{L_x}{2}\right)\right] \sin\left[\frac{n\pi}{L_y}\left(y + \frac{L_y}{2}\right)\right] \mathbf{e}_y \right] \right] e^{-\Gamma\sqrt{|\omega|/\nu_1}\zeta}, \quad (37) \end{aligned}$$

$$\hat{\mathbf{u}}_{2,\perp}|_{m,n} = \frac{-\Lambda_v}{\rho_2\sqrt{v_2}} \frac{\omega}{k} \left[\begin{aligned} &\frac{m\pi}{L_x} \sin\left[\frac{m\pi}{L_x}\left(x + \frac{L_x}{2}\right)\right] \cos\left[\frac{n\pi}{L_y}\left(y + \frac{L_y}{2}\right)\right] \mathbf{e}_x \\ &+ \\ &\frac{n\pi}{L_y} \cos\left[\frac{m\pi}{L_x}\left(x + \frac{L_x}{2}\right)\right] \sin\left[\frac{n\pi}{L_y}\left(y + \frac{L_y}{2}\right)\right] \mathbf{e}_y \end{aligned} \right] e^{\Gamma\sqrt{\frac{|\omega|}{v_2}}z}, \quad (38)$$

$$\text{with, } \Lambda_v = \frac{(\coth[kh_1] + \coth[kh_2])}{1/(\rho_1\sqrt{v_1}) + 1/(\rho_2\sqrt{v_2})}.$$

The energy balance [Eq. (39)] for viscous dissipation can be equated to the rate of change of kinetic and potential energy. It is given as

$$\begin{aligned} &\underbrace{\frac{d}{dt} \left(\sum_{i=1,2} \frac{1}{2} \int_{V_i} \rho_i |\mathbf{u}_i|^2 dV \right)}_{E_{\text{kin}}} + \underbrace{\frac{d}{dt} \left(\int_S \left[\frac{1}{2} (\rho_2 - \rho_1) g \eta^2 + \gamma_{\text{int}} \sqrt{1 + |\nabla \eta|^2} \right] dS \right)}_{E_{\text{pot}}} \\ &= - \underbrace{\sum_{i=1,2} 2\rho_i v_i \int_{V_i} \boldsymbol{\varepsilon}_i : \boldsymbol{\varepsilon}_i dV}_{\mathcal{D}}. \end{aligned} \quad (39)$$

We adapt the dissipation function $\mathcal{D}$ from Lamb [35], in which the strain tensor is denoted by $\boldsymbol{\varepsilon}_i$. The solution to the balance Eq. (39) provides the damping rates due to the side wall (λ_{wall}), interface (λ_{int}), and irrotational viscous stresses (λ_{irr}). The total damping rate for a wave of mode (m, n) is composed as

$$\lambda = \lambda_{\text{wall}} + \lambda_{\text{int}} + \lambda_{\text{irr}}, \quad (40)$$

and is assumed to have a much larger time scale than the natural frequency of the rotating wave, i.e., $\lambda^{-1} \gg |\omega|^{-1}$. All calculations are given in detail in the Supplemental Material [36, Suppl. IV]. The solutions of λ_{wall} , λ_{int} , and λ_{irr} are

$$\begin{aligned} \lambda_{\text{wall}}(m, n) = \sum_{i=1,2} \frac{\frac{1}{\sqrt{2}} \frac{\rho_i \sqrt{|\omega| v_i}}{k L_x L_y}}{\left[\frac{\rho_1}{\tanh[kh_1]} + \frac{\rho_2}{\tanh[kh_2]} \right]} &\left(\left[\frac{L_x L_y k^2}{2 \sinh^2[kh_i]} + \frac{\delta_m h_i (n^2 \pi^2 - k^2 L_y^2)}{2 L_y \sinh^2[kh_i]} \right. \right. \\ &\left. \left. + \frac{\delta_m (n^2 \pi^2 + k^2 L_y^2)}{2 L_y k \tanh[kh_i]} + \frac{\delta_n h_i (m^2 \pi^2 - k^2 L_x^2)}{2 L_x \sinh^2[kh_i]} + \frac{\delta_n (m^2 \pi^2 + k^2 L_x^2)}{2 L_x k \tanh[kh_i]} \right] \right), \end{aligned} \quad (41)$$

$$\lambda_{\text{int}}(m, n) = \frac{k\sqrt{|\omega|}}{2\sqrt{2}} \frac{(\coth[kh_1] + \coth[kh_2])^2}{\left(\frac{1}{\rho_1\sqrt{v_1}} + \frac{1}{\rho_2\sqrt{v_2}} \right) \left[\frac{\rho_1}{\tanh[kh_1]} + \frac{\rho_2}{\tanh[kh_2]} \right]}, \quad (42)$$

$$\begin{aligned} \lambda_{\text{irr}}(m, n) = \frac{2k^2 \left(\frac{1}{\rho_1\sqrt{v_1}} + \frac{1}{\rho_2\sqrt{v_2}} \right)^{-1}}{\rho_1 \coth[kh_1] + \rho_2 \coth[kh_2]} &\left(\left[\frac{\rho_2 \sqrt{v_1 v_2} + \rho_1 v_1}{\rho_2 \sqrt{v_2} \tanh[kh_1]} + \frac{\rho_1 \sqrt{v_1 v_2} + \rho_2 v_2}{\rho_1 \sqrt{v_1} \tanh[kh_2]} \right. \right. \\ &\left. \left. - (\coth[kh_1] + \coth[kh_2])(\sqrt{v_1} + \sqrt{v_2}) \right] \right). \end{aligned} \quad (43)$$

Equation (41) is the mode-dependent damping rate originating from the bounding walls. The first term of the five-part sum corresponds to the damping at the top and bottom walls. The remaining terms stem from viscous dissipation in the lateral walls [see Eq. (21) for δ_m and δ_n functions]. In model ARCs, which have smaller lateral and longitudinal surface areas than the upper and lower ones, dissipation at the top and bottom surfaces tend to outweigh the contribution from the sidewalls. This is contrary to LMBs, which, in general, have a larger sidewall surface area than the upper/lower surface area. The damping at the interface [Eq. (42)] is the same as in cylindrical cells [28]. Classically, when considering free-surface waves, the irrotational term in Eq. (43) considers

the dissipation in the irrotational bulk flow. In our formulation, it also involves some leading-order contributions at the interface, which do not appear in gas-liquid free-surface systems. It should be noted that the presented contributions are only applicable when we assume no contact angle hysteresis and no-slip boundary condition at the bounding walls.

In the single-layer limit $\rho_1 = 0$, interface damping λ_{int} reduces to zero. Considering unidirectional wave modes $(m, 0)$, Eqs. (41) and (43) simplify to

$$\lambda = \sqrt{\frac{\omega v_2}{2}} \frac{1}{L_y} \left[1 + \frac{L_y}{L_x} \left(1 + m\pi \frac{L_x - 2h_2}{L_x \sinh[2m\pi h_2/L_x]} \right) \right] + 2v_2 k^2. \quad (44)$$

This single-layer wave limit is fully equivalent to the well-known free-surface damping rate derived by Keulegan [30] as presented in [38, chapter 6]. Now that the mode-dependent viscous damping rate has been established, we solve the two-wave ansatz [Eq. (15)] considering viscous terms which appear through the boundary conditions of small perturbative quantities. The 2×2 interaction matrix [Eq. (18)] changes to

$$\begin{bmatrix} i\delta\omega - \alpha - \lambda & \frac{I_{\eta\eta'}}{2K_{\eta\eta}} \\ \frac{I_{\eta'\eta}}{2K_{\eta'\eta'}} & -i\delta\omega - \alpha - \lambda' \end{bmatrix} \begin{bmatrix} C \\ C' \end{bmatrix} = 0, \quad (45)$$

and the viscous growth rate α is

$$\alpha = \pm \sqrt{J_0^2 B_z^2 \Theta^2 (\bar{\omega}^2 - \delta\omega^2) \frac{\mathcal{T}\mathcal{T}'}{\mathcal{U}\mathcal{U}'} + [i\delta\omega - \delta\lambda]^2} - \bar{\lambda}, \quad \text{with} \quad \bar{\lambda} = \frac{\lambda + \lambda'}{2}, \quad \delta\lambda = \frac{\lambda - \lambda'}{2}. \quad (46)$$

Interesting physical differences arise from the introduction of damping terms with respect to inviscid stability criterion (23). First of all, it is clear that for $\delta\omega = 0$ cases, the square root is no longer necessarily real, with the result that even for inherently unstable cell configurations, finite Sele parameters $\beta_{\text{crit}} > 0$ are required to trigger MPR instability. Perhaps even more interesting, however, is that a completely new stability regime emerges. For the cells to become unstable, it is no longer sufficient for the square root to be real; rather, its real part must also be greater than the mean damping $\bar{\lambda}$. If the real part is smaller than $\bar{\lambda}$, the cell remains MPR-stable, but the decay rates are effectively reduced because the damping and growth rates partially cancel each other. This applies at least to one direction of rotation (positive branch) of the coupled wave motion; the other direction of rotation (opposing the Lorentz force) is accordingly damped out more quickly. This regime of magnetohydrodynamically deferred (or amplified) relaxation of wave disturbances does not exist in the dissipationless descriptions and can be demonstrated in experiments with relative ease, without necessarily having to achieve large Sele parameters β_S . To find an analytical criterion for instability, we now compare the real part of the positive branch to the mean damping $\bar{\lambda}$, yielding

$$\beta_{\gamma_{\text{int}}} > \beta_{\text{crit}} = \frac{L_x L_y}{h_1 h_2 |\Theta|} \frac{\sqrt{\mathcal{U}\mathcal{U}'}}{\mathcal{U}^*} \sqrt{\frac{1}{\mathcal{T}\mathcal{T}'} \left(\frac{\bar{\lambda}^2 - \delta\lambda^2}{\bar{\omega}^2 - \delta\omega^2} \right) \left(1 + \frac{\delta\omega^2}{\bar{\lambda}^2} \right)}. \quad (47)$$

For the critical wave pairs with degenerate eigenfrequencies $\delta\omega = 0$, β_{crit} simplifies to

$$\beta_{\text{crit}} = \frac{L_x L_y}{h_1 h_2 |\Theta|} \frac{\sqrt{\mathcal{U}\mathcal{U}'}}{\mathcal{U}^*} \frac{1}{\bar{\omega}} \sqrt{\frac{\lambda\lambda'}{\mathcal{T}\mathcal{T}'}}. \quad (48)$$

This expression helps us understand why it is so difficult to identify the most unstable wave pair for each aspect ratio L_x/L_y . In principle, wave mode combinations satisfying $\delta\omega = 0$ can be found for any geometry, but these often have a high wave number k . But small-scale wave modes are subject to much stronger viscous damping than large-scale modes with small wave numbers, which is why, in many cases, wave pairs with $\delta\omega \neq 0$ (but smaller λ) determine the smallest onset values β_{crit} . This effect is further exacerbated by the term containing the interfacial tension $\gamma_{\text{int}} k k'$, which, for

large wave numbers, outweighs the gravitational term and effectively reduces the Sele parameter β_{int} . A notable exception is the family of cells with aspect ratios of $L_x/L_y = \sqrt{1}, \sqrt{3}, \sqrt{5}, \dots$, see Sec. III C. These cells satisfy $\delta\omega = 0$ for large-scale waves, and instability onsets can be obtained from Eq. (48) alone. These geometries are generally most unstable and must be avoided in industrial applications.

F. MPR instability in the shallow water limit

To analyze the stability of large-scale aluminum reduction cells, the criterion can be further simplified. We reformulate Eq. (47) in the shallow-water limit and neglect interfacial tension. Then the interfacial waves become nondispersive: we have $\omega = ck$ and $\omega' = ck'$, with wavespeed

$$c = \sqrt{\frac{(\rho_2 - \rho_1)g}{\rho_1/h_1 + \rho_2/h_2}}. \quad (49)$$

Moreover, the nonshallow height dependencies $\mathcal{T}\mathcal{T}'$ reduce to

$$\mathcal{T}\mathcal{T}' = \frac{1}{k^4 k'^4 h_1^2 h_2^2 L_x^2 L_y^2}. \quad (50)$$

First, we consider the dissipationless inviscid limit. For resonantly coupled waves ($\delta\omega = 0$), we have a wave number equality in the shallow water limit:

$$k = \pi \sqrt{\frac{m^2}{L_x^2} + \frac{n^2}{L_y^2}} = \pi \sqrt{\frac{m'^2}{L_x^2} + \frac{n'^2}{L_y^2}} = k' \implies \frac{L_x^2}{L_y^2} = \frac{m'^2 - m^2}{n^2 - n'^2} = \frac{(m' - m)(m' + m)}{(n - n')(n + n')}. \quad (51)$$

From Eq. (24a), the function Θ is nonzero only if both wave modes (m, m') and (n, n') are of opposite parity. This implies that $(m \pm m')$ and $(n \pm n')$ are both odd integers. We write

$$\frac{(m' - m)(m' + m)}{(n - n')(n + n')} = \frac{M}{N}, \quad (52)$$

where M and N are arbitrary odd numbers. As a result, resonant wave coupling occurs for aspect ratios $L_x/L_y = \sqrt{M/N}$. This condition was previously found by Politis and Priede [29] and implies a fractality in the distribution of resonant aspect ratios: for any choice of L_x/L_y we can find a ratio of odd numbers M/N that is arbitrarily close to L_x^2/L_y^2 . Following [29], we focus on the most unstable wave pairs that have $(m - m') = (n - n') = 1$. From Eq. (52), we then derive that the wave modes have to be

$$m = \frac{M - 1}{2}, \quad m' = \frac{M + 1}{2}, \quad n = \frac{N + 1}{2}, \quad n' = \frac{N - 1}{2}. \quad (53)$$

By inserting these wave modes into the inviscid growth rate formula of Eq. (23) and applying all the mentioned limits, we obtain

$$\alpha = \pm \beta_S \frac{c}{\sqrt{L_x L_y}} \frac{8}{\pi^3} \sqrt{\frac{\delta_{M-1} \delta_{N-1}}{(M + N)(MN + 1)\sqrt{MN}}}. \quad (54)$$

In shallow cells with resonant aspect ratios ($L_x/L_y = \sqrt{M/N}$), the inviscid growth rate is proportional to the Sele number β_S and the ratio $c/\sqrt{L_x L_y}$. This is the dimensional form of the growth rate solution [Eq. (3.28)] of Politis and Priede [29], which shows that our model is fully compatible.

When we also consider viscous effects in the shallow limit, the terms in the viscous damping rate pertaining to sidewall damping and bulk irrotational damping disappear. That leaves us with

$$\lambda_{\text{wall}}(m, n) = \frac{\sqrt{ck} (\rho_1 h_2^2 \sqrt{v_1} + \rho_2 h_1^2 \sqrt{v_2})}{2\sqrt{2} h_1 h_2 (\rho_1 h_2 + \rho_2 h_1)}, \quad (55)$$

the wall damping due to the top and bottom walls, and

$$\lambda_{\text{int}}(m, n) = \frac{\sqrt{ck} \rho_1 \rho_2 \sqrt{v_1 v_2} (h_1 + h_2)^2}{2\sqrt{2} h_1 h_2 (\rho_1 h_2 + \rho_2 h_1) (\rho_1 \sqrt{v_1} + \rho_2 \sqrt{v_2})}, \quad (56)$$

which is the interfacial damping. These formulas are valid as long as $\sqrt{vk}/c \ll kh \ll 1$. In the shallow water limit, Eq. (47) reduced to

$$\beta_{\text{crit}} = \frac{L_x^2 L_y^2}{c |\Theta|} \sqrt{k^3 k'^3 (\bar{\lambda}^2 - \delta \lambda^2) \left(1 + \frac{\delta \omega^2}{\bar{\lambda}^2}\right)}. \quad (57)$$

This stability criterion is equivalent to the numerical solution obtained by Politis and Priede [29] through the Chebyshev collocation method, see Sec. III C.

In square cells, in which the wave pair $(m = 1, n = 0) + (m = 0, n = 1)$ satisfying $\delta\omega = \delta\lambda = 0$ is most unstable, Eq. (57) reduces to

$$\beta_{\text{crit}} = \frac{\pi^3 L}{4c} \lambda. \quad (58)$$

As expected, the critical Sele parameter in this case is not dictated by the difference in natural frequencies anymore but solely by the damping parameter λ , as is true for all infinitely many inherently unstable aspect ratios [Eq. (52)]. Square cell configurations are of academic interest and have been implemented in various MPR model experiments [20,22,23,25].

Further, it is interesting to note that the shallow-water criterion (57) can be linked to classic inviscid MPR models. Bojarevics and Romero [11] have analytically derived different linear stability criteria, but with the additional limitation that the natural frequencies ω and ω' must be very close to one another. In this limit $\delta\omega \ll 1 \text{ s}^{-1}$, we can approximate

$$\delta\omega \approx \frac{\omega^2 - \omega'^2}{4\sqrt{\omega\omega'}}, \quad (59)$$

and we can construct an explicit expression for β_{crit} as a function only of the mode numbers and lateral dimensions. We set $\lambda, \lambda' = 0$ in Eq. (57) and consider only unstable wave pairs fulfilling $m + m' = \text{odd}$ and $n + n' = \text{odd}$. Then β_{crit} translates to

$$\beta_{\text{crit}} = \frac{\pi^4}{16} \left| \frac{\sqrt{\left(\frac{m'^2}{L_x^2} + \frac{n'^2}{L_y^2}\right) \left(\frac{m^2}{L_x^2} + \frac{n^2}{L_y^2}\right) ((m'^2 - m^2)L_y^2 + (n'^2 - n^2)L_x^2) (m^2 - m'^2)(n^2 - n'^2)}}{\sqrt{\delta_{m \times m'} \delta_{n \times n'} (m^2 n'^2 - m'^2 n^2)}} \right|. \quad (60)$$

This expression is algebraically equivalent to the linear stability relation (4.22) presented in Bojarevics and Romero [11]. The critical Sele parameter becomes much simpler when considering the subset of one-dimensional wave modes $(m, 0) + (0, n')$:

$$\beta_{\text{crit}} = \frac{\pi^4}{16} mn' \left| m^2 \frac{L_y}{L_x} - n'^2 \frac{L_x}{L_y} \right| = \frac{\pi^2 mn' L_x L_y}{16c^2} |\omega^2 - \omega'^2|. \quad (61)$$

This is the well-known classical representation of the MPR instability threshold, which demonstrates that the cell stability is largely determined by the separation of the natural wave frequencies. Equation (61) is equivalent to Eq. (4.20) in Bojarevics and Romero [11] and also closely resembles the stability criterion that can be obtained from the simplified pendulum analog model introduced by Davidson and Lindsay [12].

TABLE I. Cell parameters and material properties from the different studies, for comparison with the theoretical results. The interfacial tension $\gamma_{\text{int}} = 0 \text{ N m}^{-1}$ in all studies.

Parameters from La Rocca <i>et al.</i> [31]		
Dimensions of cell ($L_x = L_y$)	0.5 m	
Density (ρ_1, ρ_2)	Ω_1 850 kg m ⁻³	Ω_2 1000 kg m ⁻³
Kinematic viscosity (ν_1, ν_2)	$3.6 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$	$1.0 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$
Layer thickness (h_1, h_2)	0.06 m	0.08 m
Parameters from Politis and Priede [29]		
Length of cell (L_x)	6.325 m	
Width of cell (L_y)	6.325, . . . , 2.108 m	
Density (ρ_1, ρ_2)	Cryolite (Ω_1) 2130 kg m ⁻³	Aluminum (Ω_2) 2330 kg m ⁻³
Kinematic viscosity (ν_1, ν_2)	$4.4 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$	$8.8 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$
Electrical conductivity (σ_1, σ_2)	210 S m ⁻¹	$3.3 \times 10^6 \text{ S m}^{-1}$
Layer thickness (h_1, h_2)	0.05 m	0.25 m
Parameters from Borisov <i>et al.</i> [22]		
Cross-section area of cell (S)	0.002 m ²	
Density (ρ_1, ρ_2) at 35 °C	Nitric acid (amyl alcohol) (Ω_1) 859 kg m ⁻³	Nitric acid (aq) (Ω_2) 1050 kg m ⁻³
at 45 °C	847 kg m ⁻³	1046 kg m ⁻³
at 55 °C	834 kg m ⁻³	1041 kg m ⁻³
Kinematic viscosity (ν_1, ν_2) at 35 °C	$2.9 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$	$7.4 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$
at 45 °C	$2.2 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$	$6.1 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$
at 55 °C	$1.8 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$	$5.2 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$
Electrical conductivity (σ_1, σ_2)	0.67 S m ⁻¹	47.5 S m ⁻¹
Layer thickness ($h_1 = h_2$)	0.014 m	

III. COMPARISON AND VALIDATION

In this section, we compare our theory for the viscous damping rate of MPR instability to existing results. Table I summarizes all of the physical parameters and material properties taken from the respective studies that are required to reproduce all the results presented in this section.

A. Viscous damping: Comparison with direct numerical simulations

We have verified that the viscous damping rate λ agrees with the damping rate of Eq. (44) derived by Keulegan [30] in the single-layer limit for free-surface waves. However, interfacial waves in two-layer systems are physically different because sharp viscous boundary layers form above and below the interface. The literature on two-layer interfacial wave damping is limited. Therefore, as a first test, we start by comparing our analytical damping rate formulas of Eqs. (41)–(43) to damping rates that are measured in direct numerical simulations of decaying waves. We use ARCHER, a multiphase numerical solver that applies the coupled level-set and volume of fluid method on Cartesian grids [39,40].

In our numerical simulations, we consider the cryolite-aluminum (from [29] in Table I) system in small centimeter-scale cells. We initialized a standing wave of mode (m, n) with a small amplitude of $H/500$, where $H = h_1 + h_2$, and simulated its decay over time. Three different cell aspect ratios ($L_x/L_y = 1, 2$, and 3) with $L_x = 4$ and 18 cm were simulated. We fix the layer heights to $h_1 = h_2 = L_x/8$. Capillary effects are ignored in the simulations, i.e., we set $\gamma_{\text{int}} = 0$.

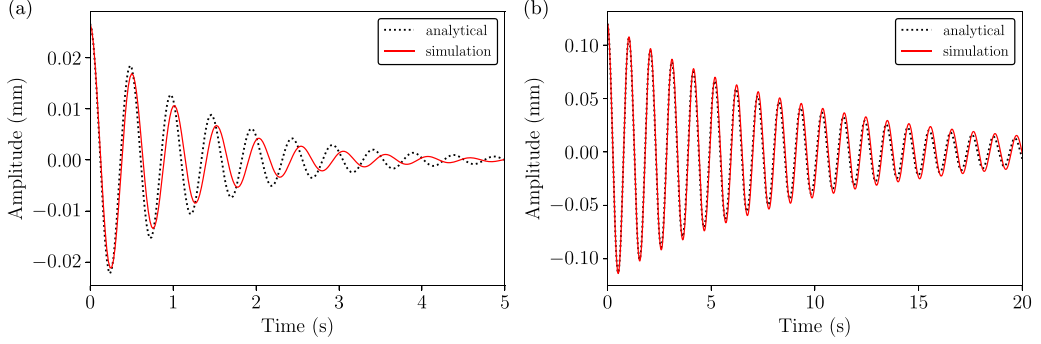


FIG. 4. The theoretical and simulated wave amplitude decay for mode (4,1) in a cell of length (a) $L_x = 4$ cm and (b) $L_x = 18$ cm and aspect ratio $L_x/L_y = 3$. The initial amplitude was set as $H/500$, where $H = h_1 + h_2$. The two-layer viscous damping equation succeeds in capturing the wave frequency and decay rate in the larger cell with a larger bulk flow domain.

Figure 4 shows the simulated amplitude in comparison with the theoretical profile, for the particular case of a decaying wave mode (4,1). The damping rates of the waves were obtained from fitting an exponential curve to the envelope of the numerical curve. A detailed explanation regarding the post-processing of the decay rates and the mesh convergence study is provided in Appendix. The damping rates obtained for all wave modes pertaining to geometries $L_x = 4$ and $L_x = 18$ cm are presented in Fig. 5. For the latter case ($L_x = 18$ cm), we found good agreement between the simulated and theoretical values of the damping rates, with a net relative deviation of 9.8%. The values of the damping rate in the cell with $L_x = 4$ cm showed a larger disagreement with the theory, with an average error of 22%, and we also notice a visible deviation in wave frequency as shown in Fig. 4(a), for smaller cells. The discrepancy in wave frequency and damping rates in the smaller cells can be attributed to the fact that the width of the viscous boundary layer $\sqrt{2\nu_i/\omega}$ is no longer very small compared to the layer heights. The parameter Δ [Eq. (29)] in this case was $\Delta > 0.05$. Here, our boundary layer approach begins to lose its applicability [41,42]. Similar

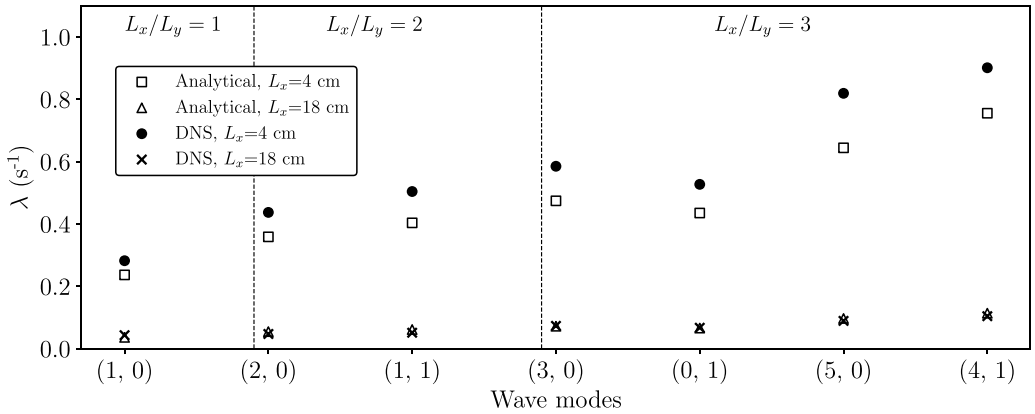


FIG. 5. The comparison of the viscous damping rates for a two-layer cryolite-aluminum system in a rectangular geometry with the analytical viscous decay rates. The respective dimensions of these systems are as follows: length $L_x = 4$ cm and layer heights $h_1, h_2 = 0.5$ cm and length $L_x = 18$ cm and layer heights $h_1, h_2 = 2.25$ cm. The simulations were conducted in the most refined meshes obtained from grid convergence tests (see Appendix).

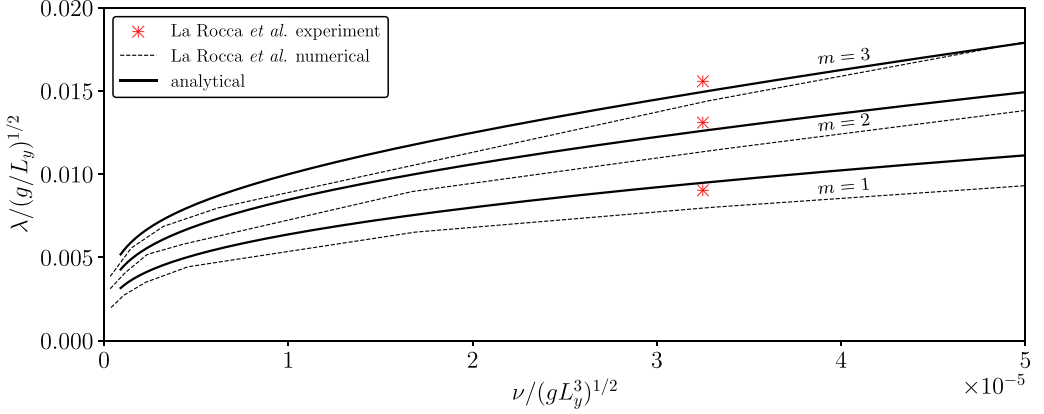


FIG. 6. Nondimensionalized viscous damping rate as a function of the nondimensionalized kinematic viscosity for three different unidirectional wave modes $(m, 0)$, $m = 1, 2, 3$ at a constant density ratio of $\rho_1/\rho_2 = 0.85$. Additional measurement points are included for the first three modes $m = 1, 2, 3$.

comparisons of the damping rates for other cell geometries $L_x = 6, 12$ cm are provided in Table III, Appendix.

B. Viscous damping: Comparison with La Rocca *et al.* [31]

La Rocca *et al.* [31] conducted interfacial sloshing experiments in an enclosed square tank filled with two liquid layers. The authors measured decay rates of different waves. A theoretical model for viscous dissipation was also developed using the Rayleigh dissipation method using Lagrange equations, but no analytical expression for the damping rates was given. We compare our viscous damping rates to their results.

In Fig. 6 we compare Eq. (40) with La Rocca’s results limited to cases where the authors provide both numerical and experimental data. Figure 6 shows the nondimensionalized viscous damping rate as a function of the nondimensionalized kinematic viscosity for three different wave modes $(m, 0)$, $m = 1, 2, 3$. Our linear damping theory slightly overshoots La Rocca’s predictions, but globally, we observe the same trends. Our model agrees better with the three experimental measurements carried out at $\nu_1/(gL_y^3)^{1/2} = 3.25 \times 10^{-5}$ and $\rho_1/\rho_2 = 0.85$. We conclude from this comparison that our viscous damping theory is compatible with the model and the experiments of La Rocca *et al.* [31]. We suspect that the differences between the damping rate profiles arise from the higher-order terms of the rotational stresses at the boundary and irrotational stresses in the bulk, which are not considered by La Rocca *et al.* [31].

C. MPR instability in shallow cells: Comparison with Politis and Priede [29]

We now consider the MPR instability and show that MPR growth rate and stability onset predictions by Politis and Priede [29] are fully contained in our model in the shallow limit for $h_1, h_2 \ll L_x, L_y$, supposing high conductivity jumps ($\sigma_1 \ll \sigma_2$), and ignoring surface tension ($\gamma_{\text{int}} = 0$). The authors derive the analytical MPR growth rate and the stability criterion by solving a linear eigenvalue problem for two shallow layers at the inviscid limit. While they do not explicitly derive a viscous correction to the growth rate, they solve the eigenvalue problem using two different spectral methods: the Galerkin expansion in gravity-wave modes and the Chebyshev collocation scheme, where they model viscous damping using a dimensionless phenomenological friction coefficient γ . The relation between the nondimensional friction coefficient and viscous damping rates λ and λ' is

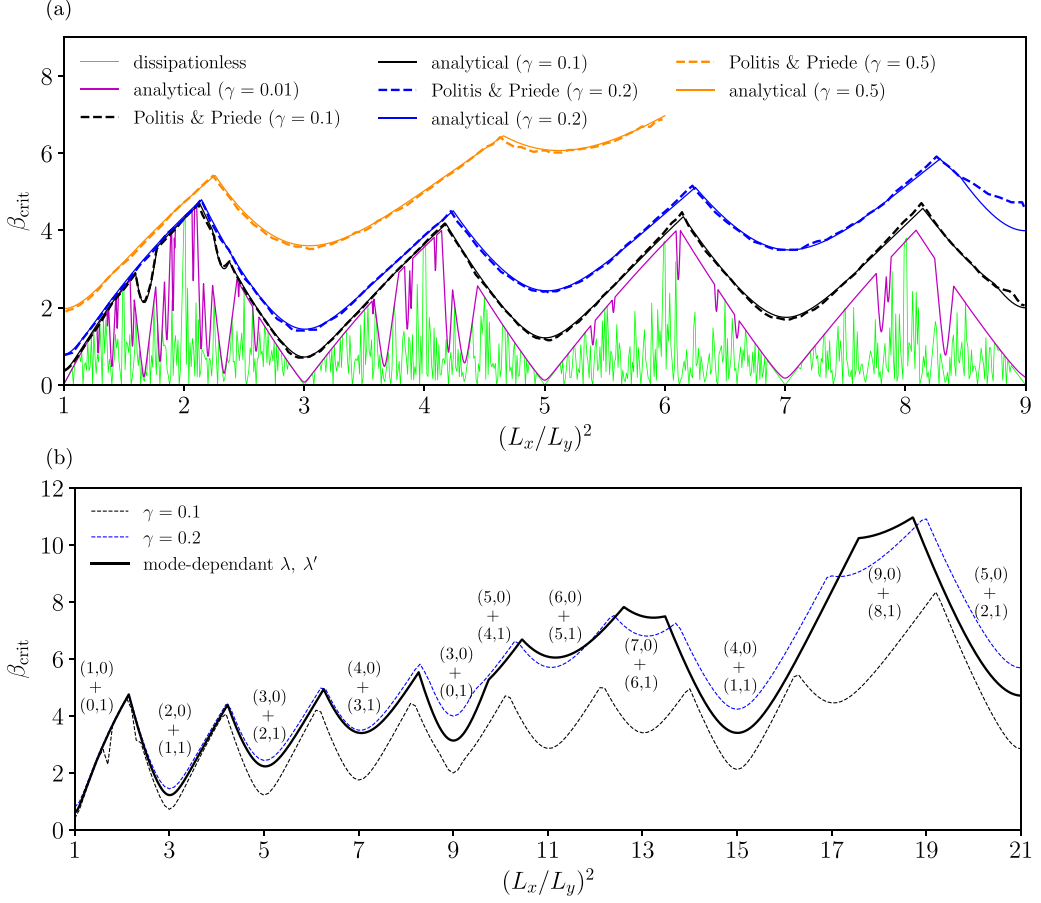


FIG. 7. Critical Sele parameters β_{crit} from Politis and Priede [29] and our analytical prediction [Eq. (47)] as a function of the aspect ratio squared $(L_x/L_y)^2$, with and without considering the effects of damping here modeled by (a) arbitrary constant damping coefficients γ . The nondimensional coefficients γ were dimensionalized as $\lambda = 0.5\gamma c/L_x$ and $\lambda' = 0.5\gamma c/L_y$. In (b) the instability onsets are plotted for mode-dependent friction coefficients calculated from damping rates in Eq. (40) for aspect ratios squared extending to 21. The solutions obtained from fixed coefficients are plotted for reference.

given by

$$\lambda = \frac{\gamma c}{2L_x}, \quad \lambda' = \frac{\gamma c}{2L_y}, \quad (62)$$

where c is the wave speed [Eq. (49)]. These damping rates based on the friction coefficient are mode-independent and different from the mode-dependent damping rates that we have calculated, see Eqs. (55) and (56).

Although we have derived an explicit analytical solution for β_{crit} , for each given mode pair (m, n) and (m', n') , see Eq. (47), it is not evident *a priori* which mode pair is first to destabilize the metal pad, and has the minimal β_{crit} . Outside the shallow approximation, the first unstable mode pair cannot be determined analytically. Our strategy for handling this difficulty is to calculate β_{crit} for all possible combinations of wave pairs and then to find the minimal β_{crit} . In practice, we see that large wave modes are less unstable, so that only wave modes up to about $m, n \lesssim 10$ must be examined.

Figure 7(a) shows the minimal onset values β_{crit} , refer to Eq. (47), as a function of the squared lateral aspect ratio $(L_x/L_y)^2$. In this graph, we use the same damping rate model as Politis and Priede [29], meaning that Eq. (62) is used to determine λ and λ' . We compare our results with the numerical solutions of Politis and Priede [29] for different friction coefficients γ . All geometrical parameters and material properties of the considered cell are given in Table I. The pattern of the green fractal curve ($\gamma = 0$) is determined solely by the chosen resolution and hence is not physical. For all other values of friction coefficients $\gamma = 0.1, 0.2$ and 0.5 , we find that our analytical model is in quasiperfect agreement with the curves of Politis and Priede [29].

For certain aspect ratios ($L_x^2/L_y^2 = 1, 3, 5, \dots$), a simple approximate solution for β_{crit} can be derived from our theory that is valid in the vicinity of these most unstable aspect ratios. At these aspect ratios, the integers M and N defined by Eq. (52) satisfy $L_x^2/L_y^2 = M/N$. Setting $M = 1, 3, 5, \dots$ and $N = 1$, neglecting the term involving $i\delta\omega\delta\lambda$, and inserting Eqs. (53) and (62) into (57), we obtain β_{crit} as a function of arbitrary aspect ratio L_x^*/L_y^* :

$$\beta_{\text{crit}}^M(L_x^*, L_y^*) = \frac{\pi^4 \sqrt[4]{M}(M+1)}{16\sqrt{\delta_{M-1}}} \sqrt{\frac{\gamma^2}{\pi^2} + \frac{L_y^*}{2L_x^*}(M^2+1) + \frac{L_x^*}{L_y^*} - \sqrt{\frac{(M^2-1)^2}{4} \left(\frac{L_y^*}{L_x^*}\right)^2 + (M+1)^2}} \quad (63)$$

for $M = 1, 3, 5, \dots$, where L_x^*/L_y^* must be close to the critical aspect ratios $|L_x^*/L_y^* - \sqrt{M}| \ll 1$ with $M = L_x^2/L_y^2$. From this equation, one can further derive a useful expression for β_{crit} of the most unstable aspect ratios (minimum local β_{crit}) by assigning $L_x^*/L_y^* = \sqrt{M}$, yielding

$$\beta_{\text{crit}}^M = \frac{\pi^3 \sqrt[4]{M}(M+1)}{16\sqrt{\delta_{M-1}}} \gamma. \quad (64)$$

In square cells defined by $M = 1$, Eq. (64) reduces to Eq. (58) in its dimensional form.

Figure 7(b) provides the analytical prediction from Eq. (47) taking into account wave-number-dependent viscous damping from Eq. (40), highlighting all the unstable wave modes between the inflection points. It also shows that modeling viscous damping to calculate the stability criterion provides more realistic onsets in the linear regime compared to the arbitrary coefficients. Moreover, extending the graph until the aspect ratio square of 21 shows us why most industrial ARCs operate at $(L_x/L_y)^2 = 17-18$, due to the presence of a larger stability buffer [43]. Using Fig. 7(b), it is possible to explain the small deviation emerging between our respective theories at $\gamma = 0.2$ for $(L_x/L_y)^2 > 8.45$. The curve from Politis and Priede [29] shows here β_{crit} for the mode pair $(5, 0) + (4, 1)$. However, our solution predicts that the pair $(3, 0) + (0, 1)$ becomes unstable first, and accordingly, our calculated curve is slightly lower. Apart from this point, however, the first unstable wave mode pair is always correctly predicted by the wave mode tuples [Eq. (52)]. It is important to note that the absolute values of the β_{crit} are not very significant in large cells. However, by using the mode-dependent viscous damping formulas, the change in the critical beta between different wave numbers can be more accurately described.

In the previous discussion, we have focused on the first destabilized mode, but it is important to remember that many other modes are also destabilized above the threshold. The first unstable wave pair (smallest β_{crit}) is not always the wave pair that is most unstable (highest α). Beyond the threshold, i.e., $\beta_S > \beta_{\text{crit}}$, the exact pattern of MPR waves can be difficult to predict from linear theory. At some point $\beta_S > \beta_{\text{crit}}$, we always find the primary rotating mode pair $(1, 0) + (0, 1)$ as the most unstable mode in all cell configurations, as already noted by Munger and Vincent [44].

D. MPR instability in nonshallow cells: Comparison with Borisov *et al.* [22]

We apply our theory to the little-known model experiment of Borisov *et al.* [22], who succeeded in destabilizing MPR waves in a two-layer experimental cell [nitric acid ($\text{HNO}_3(\text{aq})$) at the bottom and $\text{HNO}_3 + \text{C}_5\text{H}_{12}\text{O}$ at the top]. Both liquids have small but sufficiently contrasting electrical

conductivities ($\sigma_1 = 0.67 \text{ S m}^{-1}$ and $\sigma_2 = 47.5 \text{ S m}^{-1}$). Due to the small difference in density of the two liquids, small cell currents $I_0 \sim 2 - 4 \text{ A}$, in combination with a strong external magnetic field $B_z = 0.25 \text{ T}$, allowed us to reach large enough Sele parameters $\beta_S \sim 2$ to destabilize MPD waves.

In this experiment, Borisov *et al.* [22] could also change the electrical boundary conditions at the bottom electrode, using a removable insert made of porous material that was impregnated with an aqueous nitric acid. This is done to mimic the boundary conditions of an ARC, i.e., that the bottom electrode be a worse electrical conductor than the lower liquid. Unfortunately, the authors have only published measurements without the insert, for which the ground electrode can be regarded as a perfect electrical conductor. To cover this case of a perfectly conducting bottom plate, replace the boundary condition of Eq. (5d) with $\varphi_2 = 0|_{z=-h_2}$. On deriving the stability onset once again, we find it has the same form as before:

$$\beta_{\text{crit}} = \frac{L_x L_y}{h_1 h_2 |\Theta|} \frac{\sqrt{U U'}}{U^*} \sqrt{\frac{1}{\mathcal{T}_c \mathcal{T}_c'} \left(\frac{\bar{\lambda}^2 - \delta \lambda^2}{\bar{\omega}^2 - \delta \omega^2} \right) \left(1 + \frac{\delta \omega^2}{\bar{\lambda}^2} \right)}, \quad (65)$$

but with other terms. Mainly,

$$\begin{aligned} \mathcal{T}_c &= \frac{\Lambda_c}{k(k^2 - k'^2) L_x L_y} \left[\frac{-k'}{\tanh[kh_1]} + \frac{k' \operatorname{sech}[k'h_1]}{\sinh[kh_1]} + \frac{k}{\coth[k'h_1]} + \frac{-k'}{\tanh[kh_2]} \right. \\ &\quad \left. + \frac{k' \operatorname{sech}[k'h_2]}{\sinh[kh_2]} + \frac{k}{\coth[k'h_2]} \right], \\ \mathcal{T}_c' &= \frac{\Lambda_c'}{k'(k'^2 - k^2) L_x L_y} \left[\frac{-k}{\tanh[k'h_1]} + \frac{k \operatorname{sech}[kh_1]}{\sinh[k'h_1]} + \frac{k'}{\coth[kh_1]} + \frac{-k}{\tanh[k'h_2]} \right. \\ &\quad \left. + \frac{k \operatorname{sech}[kh_2]}{\sinh[k'h_2]} + \frac{k'}{\coth[kh_2]} \right], \end{aligned}$$

and a different conductivity jump parameter is

$$\Lambda_c = \frac{\sigma_1^{-1} - \sigma_2^{-1}}{\sigma_1^{-1} \tanh[kh_1] + \sigma_2^{-1} \tanh[kh_2]}.$$

We use the subscript c to clearly differentiate from the jump parameter given in Eq. (14). Borisov *et al.* [22] measured the critical onsets using a different dimensionless parameter $W = I_0 B_z / [(\rho_2 - \rho_1) g L_x L_y]$ that is related to β_S through $\beta_S = W L_x L_y h_1^{-1} h_2^{-1}$. In Fig. 8(a), the measured critical Sele parameters are shown together with the first unstable wave pairs $(m, n) + (m', n')$ in nine different cells of different aspect ratios L_x/L_y . For a better understanding, Fig. 8(b) further shows our continuous solution [Eq. (65)] compared with the linear stability prediction of Borisov *et al.* [45] (Galerkin method solved numerically).

During their experiments, Borisov *et al.* [22] measured strong temperature fluctuations (between 35 and 55 °C) due to Joule heating occurring in both liquids. This creates an uncertainty in the material parameters which is quantified using error bars in Fig. 8(a). At all examined aspect ratios, the unstable wave pairs reported by Borisov *et al.* [22] coincide with the unstable pairs predicted by our model. Quantitatively, we also find fair agreement, but there are some considerable deviations, especially for the mode pair $(2, 2) + (1, 3)$. Considering an arbitrary interfacial tension ($\gamma_{\text{int}} = 0.005 \text{ Nm}^{-1}$ for alcohol-water systems) vastly improves the agreement between our theory and the experimental measurements. Although interfacial tension physically exists, the study does not discuss it, therefore we do not want to make any assumptions.

Our solution is particularly sensitive to temperature fluctuations, which leads to large error bars. Also, the measurement point that corresponds to $(1, 2) + (0, 3)$ is very close to the region where the mode pair $(1, 3) + (0, 4)$ is most unstable. When taking this neighboring pair instead

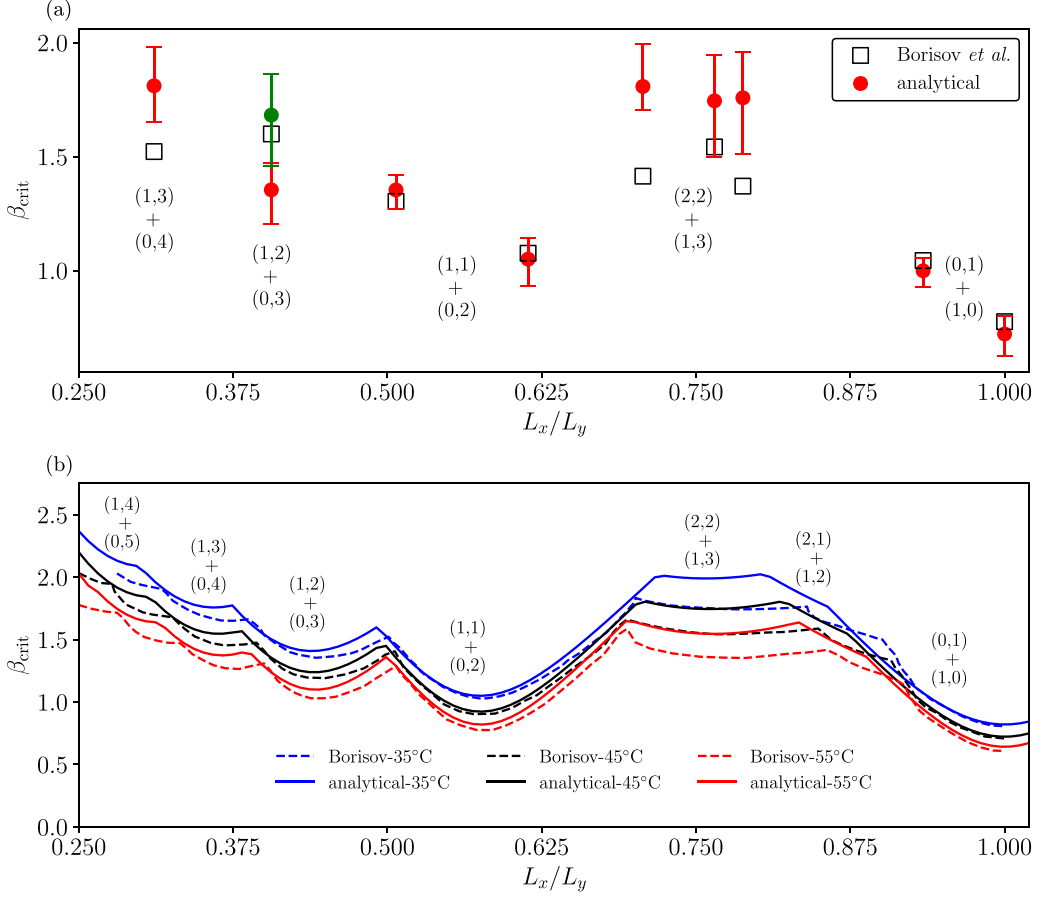


FIG. 8. (a) Comparison with the experimentally observed instability onsets at nine different cell geometries. The error bars cover a temperature range of 35 to 55 °C. The green data point corresponds to the neighboring mode pair (1, 3) + (0, 4). Panel (b) shows the analytical stability onset comparisons along with the critical wave modes observed at different cell aspect ratios.

(green data point), our model agrees well with the experimental observation. This shows that temperature changes in regions of close-lying modes may have led to spontaneous mode transitions. The comparison of both linear stability theories reveals that our model always yields slightly larger β_{crit} . The reason for this discrepancy is difficult to identify, but we noticed that Borisov *et al.* [22] have neglected sidewall damping. From our damping rates in Eqs. (41)–(43), we find that sidewall damping accounts for more than a third of the total damping, resulting in a higher β_{crit} in our theory.

IV. CONCLUDING REMARKS

We have presented a perturbative formulation of the linear metal-pad-roll (MPR) instability in small-scale rectangular cells. The theory is motivated by the existence of several laboratory-scale experiments [20,22,23,25], for which we aim to provide accurate theoretical benchmarks.

We calculated the magnetohydrodynamic coupling of interfacial waves pairwise, which made it possible to derive explicit analytical solutions for the stability limits. Unlike previous models, our formulation is not limited to shallow cells, and it also incorporates viscous damping and interfacial tension. With the incorporation of damping, the model can also make stability predictions for degenerate wave modes with coinciding natural frequencies. To simplify the calculations for the

reader, we have gathered all the essential equations with explanations in the Supplemental Material [36], along with a Python module.

The main result of our study is provided by Eq. (46), which predicts MPD growth rates for any given pair of standing-wave modes. This equation recovers the linear stability predictions of Politis and Priede [29] in the shallow-water limit and, for nearly degenerate eigenfrequencies, also reduces to the classical MPD stability theories [11]. For further validation, we also compared our stability predictions with experimental measurements from the nonshallow model experiment devised by Borisov *et al.* [22], which has received little attention to date. For many aspect ratios, we found good quantitative agreement, and for each measurement, our model correctly predicts the most unstable wave pair.

Our formulas for the viscous damping rate of waves in two-layer systems, which include the dissipation at all tank walls and the boundary layers above and below the interface, can be of use in other problems such as two-layer sloshing. To validate the damping rates independently of the MPD stability problem, we conducted 3D numerical simulations using the hydrodynamic, multiphase solver ARCHER and found good agreement in all cases, where the viscous boundary layers did not penetrate too far into the bulk flow. Our damping rate predictions further compare well to the interface damping experiments by La Rocca *et al.* [31] and, in the one-layer limit, algebraically reduce to the classical viscous damping rates of standing surface waves [30].

The two-wave formulation of the MPD instability problem can be adapted to the case of the recent single-layer MPD experiment [25]. In this experiment, an electrical current proportional to the wave slope is injected at the sidewalls, and this imitates the MPD instability mechanism. Using this setup, we intend to study the MPD instability in its nonlinear regime, but the setup needs to be calibrated to be representative of two-layer cells. Moreover, our theory is well-positioned for augmenting the cell current (or external magnetic field) with alternating components, with a view to studying active MPD suppression techniques that have recently proved promising [46]. Horstmann *et al.* [47] showed that the alternating current MPD suppression rests on a special form of parametric antiresonance. However, the analysis relied on a highly idealized pendulum model with only two degrees of freedom. The ideal AC frequencies for minimally invasive MPD suppression predicted in [47] still need to be confirmed by continuous MPD theories.

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

APPENDIX: ADDITIONAL DETAILS REGARDING THE NUMERICAL ANALYSIS

Table II provides the mesh sizes adopted for all the cases simulated. Grid sizes of 2^n and its multiples were considered for grid independence tests. The mesh behavior regarding damping rates and resonant frequency for different cell sizes of aspect ratio 1 is shown in Figs. 9(a) and 9(b).

TABLE II. Grid sizes adopted for different simulation cases in ARCHER. Cell sizes were kept uniform throughout the domain.

Aspect ratio	Length of cell L_x (cm)	Width of cell L_y (cm)	Layer height $h_1 = h_2$ (cm)	Grid size ($n_x \times n_y \times n_z$)
1	4,6,12 & 18	4,6,12, and 18	$L_x/8$	$64 \times 64 \times 16$
				$128 \times 128 \times 32$
				$256 \times 256 \times 64$
2		2,3,6, and 9	$L_x/8$	$64 \times 32 \times 16$
				$128 \times 64 \times 32$
3		1.33,2,4, and 6	$L_x/8$	$192 \times 64 \times 48$

The values for all cases were found to converge at $n_z=64$, 32, and 48 for aspect ratios 1, 2, and 3, respectively. Moreover, the same cases were repeated for different initial wave amplitudes as shown in Fig. 9(c), to verify that the amplitude was in no way influencing the resultant damping rates, similar to the theoretical values. The simulations were carried out with an initial amplitude of $H/500$. The damping rates remained unchanged when the cases were simulated with and without the advection term $\nabla \cdot (\rho \mathbf{u} \otimes \mathbf{u})$, confirming that the wave motion was within the scope of Stokes's flow.

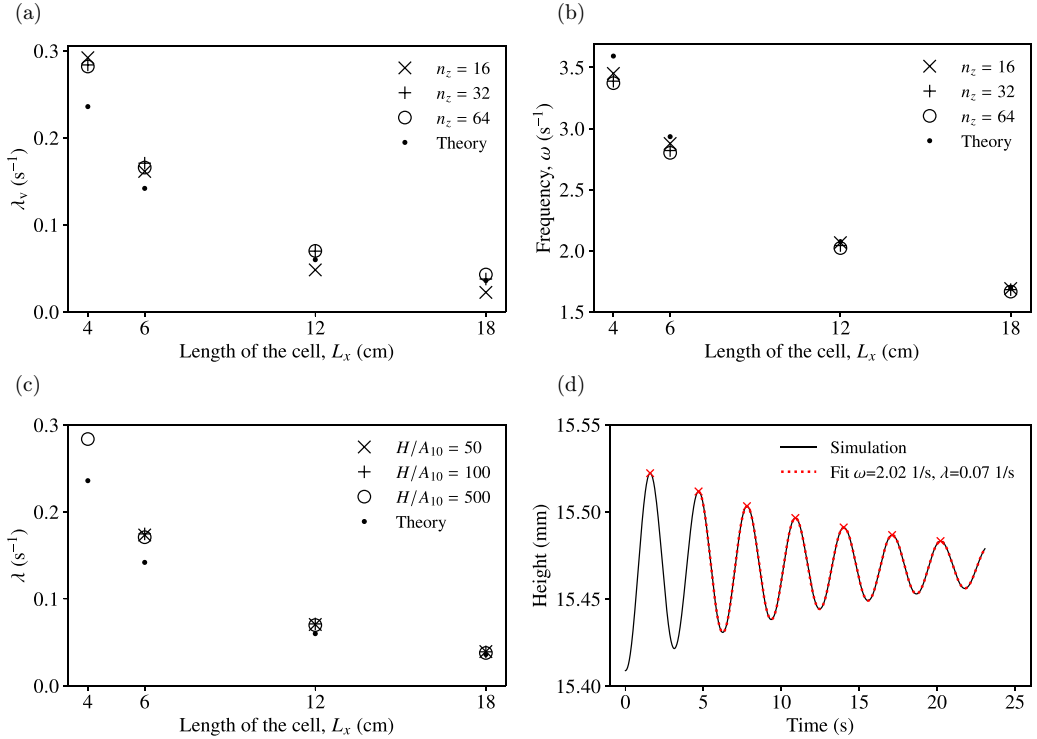


FIG. 9. The grid independence test observing the (a) viscous damping rate and (b) wave frequency for different cell sizes for aspect ratio 1 and wave mode (1,0). (c) The same case was simulated with different initial wave amplitudes to certify the independence of initial wave amplitude on the damping rate. (d) The postprocessing was performed with a cell size of $12 \times 12 \times 3$ cm, and the exponentially decaying oscillatory fit of the amplitude signal at the local point $-L_x/2, -L_y/2$ was used to determine the wave frequency and damping rate.

TABLE III. The detailed comparison of theoretical values of two-layer wave frequency and viscous damping with the numerical simulations of ARCHER code, for different cell dimensions and resultant wave modes. The two layers considered are cryolite and aluminum (see Table I). The heights of the respective layers are equal, i.e., $h_1 = h_2$.

Aspect ratio	Cell dimensions $L_x \times L_y \times [h_1 + h_2]$ (cm)	Wave mode (m,n)	Wave frequency ω (s ⁻¹)		Viscous damping rate λ (s ⁻¹)	
			Theory	ARCHER	Theory	ARCHER
1.0	4×4×1		3.593	3.372	0.236	0.282
	6×6×1.5		2.934	2.801	0.142	0.166
	12×12×3	(1,0)	2.075	2.022	0.060	0.070
	18×18×4.5		1.694	1.666	0.036	0.043
	4×2×1	(2,0)	6.732	6.453	0.359	0.437
2.0		(1,1)	7.383	7.078	0.403	0.504
	6×3×1.5	(2,0)	5.496	5.357	0.216	0.260
		(1,1)	6.028	5.879	0.243	0.296
	12×6×3	(2,0)	3.887	3.858	0.090	0.095
		(1,1)	4.262	4.233	0.102	0.104
	18×9×4.5	(2,0)	3.173	3.164	0.054	0.047
		(1,1)	3.480	3.471	0.061	0.051
		(3,0)	9.258	8.875	0.474	0.585
	4×1.33×1	(0,1)	9.258	8.929	0.435	0.527
		(5,0)	12.89	12.44	0.644	0.819
3.0		(4,1)	12.89	12.33	0.755	0.901
		(3,0)	7.559	7.347	0.285	0.347
	6×2×1.5	(0,1)	7.559	7.379	0.261	0.315
		(5,0)	10.52	10.28	0.385	0.485
		(4,1)	10.52	10.23	0.452	0.548
		(3,0)	5.345	5.288	0.119	0.137
	12×4×3	(0,1)	5.345	5.296	0.109	0.126
		(5,0)	7.440	7.384	0.161	0.177
		(4,1)	7.440	7.373	0.189	0.207
		(3,0)	4.364	4.343	0.072	0.073
	18×6×4.5	(0,1)	4.364	4.346	0.066	0.067
		(5,0)	6.075	6.054	0.097	0.089
		(4,1)	6.075	6.051	0.114	0.104

The total time of the simulation was at least 20 times the theoretical period of the oscillations. The wave frequencies and decay rates were extracted as shown in Fig. 9(d): by tracking the elevation of the interface at a specific point $(-L_x/2, -L_y/2)$, giving the local amplitude profile. After removing the first period, the resulting profile was fitted to an exponentially decaying oscillation function. It can also be obtained by taking the volume integral of the kinetic energy $\sum_{i=1,2} \int_{V_i} 0.5 \rho_i \mathbf{u}_i^2 dV$, whose envelope decays twice as fast as the amplitude. The complete list of the frequencies and damping rates obtained from the simulations, in comparison with their respective theoretical values, are given in Table III. Overall, we found good agreement with the theoretical values, especially in larger cells ($L_x = 12, 18$ cm).

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