Negative resistance for colloids driven over two barriers in a microchannel

Urs Zimmermann,1 Hartmut Löwen,1 Christian Kreuter,2 Artur Erbe,3 Paul Leiderer,2 and Frank Smallenburg1

1Institut für Theoretische Physik II: Weiche Materie, Heinrich-Heine-Universität Düsseldorf, D-40225 Düsseldorf, Germany
2Fachbereich Physik, Universität Konstanz, D-78457 Konstanz, Germany
3Institut für Ionenstrahlphysik und Materialforschung, Helmholtz-Zentrum Dresden-Rossendorf, D-01328 Dresden, Germany

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Ohm’s law is one of the most central transport rules stating that the total resistance of sequential single resistances is additive. Here we test additivity of resistances in classical systems of interacting colloids driven over two energetic barriers in a microchannel, using real-space microscopy experiments, particle-resolved simulations, and dynamical density functional theory. If the barrier separation is comparable to the particle correlation length, the resistance is highly non-additive, such that the added resistance of the second barrier can be significantly higher or lower than that of the first. Surprisingly, for a barrier separation comparable to the particle interaction range, the second barrier can add a negative resistance, such that two identical barriers are easier to cross than a single one. We explain our results in terms of the structuring of particles trapped between the barriers.

One of the basic characteristics of any transport situation is the resistance, commonly known from electric circuits, which is in general defined as the ratio of the transport flux and the driving force, typically in the linear-response regime of small drives. For both electric circuits and classical transport, Ohm’s law states that when resistors are put in series, their resistances simply add up. However, this macroscopic law is expected to break down on the microscopic scale, in particular when the distance between the two obstacles approaches the correlation length of the transported particles.

In this Letter, we explore the additivity of resistances in mesoscopic colloidal suspensions driven through a microchannel.9 We first perform an experiment on repulsive colloidal particles confined to microchannels containing two step-like barriers on the substrate, and measure the current through the channels as a function of the strength of the gravitational driving force. Subsequently, we employ Brownian dynamics simulations and dynamical density functional theory to systematically explore the interplay between the two barriers. Our results show strong deviations from additivity for the resistance of two barriers when the separation between the two obstacles is comparable to the correlation length of the system, which is on the order of several interparticle spacings. Amazingly, if the barrier separation is comparable to the interaction range, we discover that the resistance contributed by the second barrier can even be negative. We explain this counterintuitive effect of negative resistance via the long-ranged particle interactions and the ordering of the particles trapped between the two barriers. When these particles are disordered, they exhibit spontaneous fluctuations which modulate their interactions with particles crossing the barriers, significantly enhancing barrier crossing rates10 11. This surprising phenomenon provides a route for tuning and enhancing particle flow over an obstacle by the inclusion of additional barriers, reminiscent of the use of geometric obstacles to assist e.g. the flow of panicked crowds12.

In our experiment, colloidal superparamagnetic particles are confined in a microchannel with obstacles, prepared using molds made via microlithography9 13, see Fig. 1. The experimental cell consists of two rectangular reservoirs, connected by multiple channels (to improve statistics). The colloidal particles are restricted to two-dimensional in-plane motion due to gravity. In each channel up to two U-shaped step-like barrier structures are
implemented perpendicular to the channel, for details see Supplemental Material [14]. A uniform external magnetic field $\mathbf{B}_{\text{ext}}$ is applied in the direction perpendicular to the plane in which the particles move. This magnetic field induces purely repulsive interactions between the colloidal particles.

We measure the particle current in the channel as a function of the gravitational driving force, controlled by the tilt angle of the setup, for channels with zero, one, and two barriers. In the absence of barriers, the current shows a trivial linear dependence on the driving force, shown by the blue line in Fig. 1. For a single barrier (green line in Fig. [1]), we observe a crossover from a zero-flow regime at small driving forces (where the driving force is too weak to push particles across the barrier) to an approximately linear regime for large driving forces [9]. Hence, the barrier provides a resistance to the flow, which reduces the particle current. Adding a second barrier to a channel results in a clear non-additivity of the resistance of the two barriers. In particular, for two barriers separated by approximately 2.5 times the typical interparticle distance (red line in Fig. 1a), the second barrier has a much stronger effect on the total particle current than the first one, indicating a higher effective resistance.

To explore this non-additivity in detail, we make use of overdamped Brownian dynamics simulations and dynamical density functional theory (DDFT) calculations. We consider a two-dimensional system with periodic boundary conditions along the channel ($x$-direction), containing $N$ particles interacting via a dipolar repulsion

$$\beta V_{\text{int}}(r) = \Gamma \left(\frac{a}{r}\right)^3,$$

where $\beta = 1/k_B T$ with $k_B$ Boltzmann’s constant and $T$ the temperature, $\Gamma$ is the dimensionless interaction strength, and $a = \rho_0^{-1/2}$ sets the length scale of a typical interparticle spacing of a given mean number density $\rho_0$. The particles additionally experience a constant driving force $F \mathbf{x}$ pushing the particles along the channel.

The confining channel and barriers are modeled as an external potential $V_{\text{ext}}(x, y) = V_{\text{channel}}(y) + V_{\text{barrier}}(x)$. The first term here is a steep repulsive wall potential confining the particles in one direction. $V_{\text{barrier}}$ represents one or two parabola-shaped potential barriers with width $a$ and height $V_0 = 10 k_B T$, see Fig. 2a inset and Supplemental Material [14]. We choose the channel width $L_y = 4.65a$, and the channel length $L_x$ such that the total number density $\rho_0 = N/(L_x L_y) = 1/a^2$ for a given particle number $N$.

In our DDFT calculations [15, 16], we choose the Ramakrishnan–Yussouff functional [17] to model interacting particles in a fluid state ($\Gamma = 5$). In addition to DDFT, we perform Brownian Dynamics simulations of particles experiencing the same potentials and external driving force. As a reference we provide an analytical solution for non-interacting particles ($\Gamma = 0$). See Supplemental Material [14] for details.

Using both DDFT and simulations, we explore the relation between the total steady-state particle current $J$ along the channel, the driving force $F$ on the particles, and the distance $\Delta x$ between the two barriers. The ratio of the driving force and current characterizes the total resistance of the system, $R^{\text{tot}} = J/F$. In a channel without barriers, the particles trivially adopt the average drift current $J_0 = F/\rho_0 L_y$, and the external field strength was 0.6 mT. The separation between the two barriers was 30 µm.

![FIG. 1. a) Schematic setup of the experiment: two particle reservoirs are connected by a microfluidic channel through which particles are flowing due to gravity. b) Top view of the experimental system. c) Snapshots of a two barrier system for different times. The position of the barriers is indicated by a red vertical line. Two particles are highlighted in red and green. d) Flux as a function of tilt angle in a system with no barriers (blue line), single barrier (green line) and double barriers (red line). The initial density was $\rho_0 = (7.23 \pm 0.5) \times 10^{-3} \mu m^{-2}$, and the external field strength was 0.6 mT. The separation between the two barriers was 30 µm.](image-url)
extracted from the total resistance \( R_{s}^{\text{tot}} = R_{bg} + R_{1} \) by measuring the single-barrier current \( J_{a} \):

\[
R_{1} = R_{s}^{\text{tot}} - R_{bg} = F \left( \frac{1}{J_{s}} - \frac{1}{J_{0}} \right).
\]

Similarly, in a double-barrier system (with current \( J_{d} \), the total resistance is \( R_{d}^{\text{tot}} = R_{bg} + R_{1} + R_{2} \), and the effective resistance of the second barrier \( R_{2} \) can be written as

\[
R_{2} = F \left( \frac{1}{J_{d}} - \frac{1}{J_{s}} \right).
\]

In the case of additivity, the resistance \( R_{2} \) of the second barrier will be equal to \( R_{1} \) (the resistance of the first barrier), while deviations from this rule will indicate non-additivity.

In Fig. 2 we plot \( R_{2}/R_{1} \) for a range of barrier separations \( \Delta x \) at different driving forces \( F \), as obtained from analytical theory [14] (a), DDFT calculations (b), and computer simulations (c). For non-interacting particles \( R_{2} \) is lowest when the two barriers are touching (\( \Delta x = a \)) and converges exponentially to \( R_{1} \) for larger distances. In contrast, for interacting particles and for all investigated \( F \), the resistance of the second barrier is highest at \( \Delta x = a \). At this separation the resistance added by the second barrier can be many times higher than \( R_{1} \), signaling strong non-additivity. More interestingly, for slightly larger separations (\( \Delta x \approx 1.5a \)), \( R_{2} \) becomes smaller than \( R_{1} \), and even negative for sufficiently weak driving forces. In this regime, the addition of the second barrier reduces the overall resistance in the channel. At larger \( \Delta x \), \( R_{2} \) shows decaying oscillations, converging towards the additive case (\( R_{2} = R_{1} \)), as expected at sufficiently large distances.

We can understand this observation by considering the interactions between the particles. Since these are dipolar in nature, they are sufficiently long-ranged to span across the barrier. Hence, a particle on top of the barrier experiences forces from particles between the two barriers, which depend on the density and structuring of those particles. In Fig. 3 we plot the density profile of the particles \( \rho_{s}(x) \), projected onto the long axis of the channel, for various barrier separations \( \Delta x \), as well as for a single barrier. In the single-barrier case, we always observe a high density peak in front of the barrier, and a slightly lower peak just after the barrier (see Fig. 3a). In the two-barrier cases, the additional peaks in between the two barriers vary in height based on \( \Delta x \). For very small separations (Fig. 3b), where the resistance of the second barrier is high (\( R_{2} > R_{1} \)), we find a single sharp density peak between the barriers, which is significantly higher than the peak observed after a single barrier. Here, particles between the barriers are arranged in a single line with little room for fluctuations, and hence provide a strong and relatively constant force on particles crossing the first barrier, pushing them back. In the regime where \( R_{2} < R_{1} \) (Fig. 3c), we instead see two much lower peaks, indicating a structure with two layers and significantly larger fluctuations. These larger fluctuations not only provide space for particles entering via the first barrier, but also modulate the force exerted on particles crossing the barriers, resulting in a fluctuating effective barrier height. For weak driving forces, barrier crossings are rare events, whose rate depends exponentially on the barrier height. Fluctuations in barrier height are known to lead to significantly higher crossing rates [10] [11] and hence higher currents. Finally, for larger separations, where \( R_{2} > R_{1} \) again, we observe two higher peaks, indicating a more structured pair of layers between the barriers.

We confirm this intuitive picture by plotting in Fig. 4 the relative height of the first peak after the first barrier \( \delta \rho_{\text{peak}} = \rho_{d}^{\text{peak}}/\rho_{s}^{\text{peak}} \), where \( \rho_{s}^{\text{peak}} \) is the height of the first peak after a single barrier, and \( \rho_{d}^{\text{peak}} \) is the height of the first peak after the first of two barriers. When plotted as a function of \( \Delta x \), the peak height (blue in

FIG. 2. (Color online) Effective resistance \( R_{2} \) of the second barrier relative to the resistance \( R_{1} \) of the first barrier, as a function of the barrier spacing \( \Delta x \), at different driving forces. The dashed lines highlight special values of \( R_{2} \): the gray line shows Ohmic additivity and the red line marks the onset of negative effective resistance. Results are shown for analytical theory [14] at \( \Gamma = 0 \) (a), DDFT at \( \Gamma = 5 \) (b), and simulations at \( \Gamma = 5 \) (c). A sketch of the barrier configuration is shown in inset b.
The sensitivity of the resistance to the barrier separation and microscopic particle interactions provide a method to tailor and control flow through channels or porous media. Interestingly, geometric obstacles have similarly been shown to enhance flow, as applied in e.g. the design of emergency exits. However, in these cases, enhanced flow rate is typically observed when the added obstacle is placed before the bottleneck, rather than behind it.

The possibility of mitigating a flow-resisting barrier by placing another barrier behind it might have important implications in microfluidic devices. Moreover, the specificity of this approach to relatively long-ranged interactions...
tions suggests an opportunity for separating different particle species, or enhanced flow control via external fields modifying the interactions. Further applications include the directed transport of strongly charged dust particles in a plasma \[26\] and congestions in granulates \[27\], as well as jammed flow situations of colloids \[28\], or agents through constrictions \[29\]. In particular, a jammed situation near an obstacle may be avoided by adding further obstacles. An interesting question for future research is whether the effective total resistance could be further tuned by using a combination of three, four, or an infinite number of obstacles \[30\] (forming e.g. a ratchet \[31, 32\]), or by using barriers of differing heights.

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