Pulse-coupled BZ oscillators with unequal coupling strengths

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Electronic Supplementary Information (ESI)

ESI consists of: (1) a detailed description of the way in which we modified and augmented the model equations (1)-(4) in order to account for the long-term effects of inhibitory perturbations, and (2) figures that provide more detail about the results of simulating inhibitory perturbation.

(1) Modifications to the Model of Lavrova and Vanag [12]

To describe the long-term effects of repeated inhibitory perturbations, we introduce an additional variable, *p*, which increases by $[Br^-]_0$ at the instant of perturbation and then decays to produce bromide (*y*). This new variable and its kinetic terms were added, and the differential equation for the perturbed variable *y* was modified ($+k_{11}p$ term, k_{11} = 2x10^{-2} s^{-1}).

$$\frac{dy}{dt} = -k_1hxy - k_2h^2ay + k_9vz + k_{11}p - k_0y \tag{S1}$$

$$\frac{dp}{dt} = -k_{11}p - k_0p \tag{S2}$$

In order to model the long-term effect of perturbations, instead of assuming constant *a* ($[BrO_3^-]$), we allowed it to be an additional variable:

$$\frac{da}{dt} = -k_2h^2ay - k_4hax\frac{(z_t - z)}{z_t - z + c_{min}} + (a_0 - a)k_0 \tag{S3}$$

The cycle length depends strongly on $[BrO_3^-]$ and $[H^+]$. The latter was maintained constant in the experiments, while all other species may experience small decreases due to the dilution caused by the finite volume of the pulse perturbation. In the model, when an inhibitory pulse occurs, the integration of the differential equations is interrupted, the concentration of *p* is increased by $[KBr]_{inj}$ and only the value of *a* is decreased according to the following equation.

$$a^{'} = a \times (0.8198 - 0.015 \times \ln [KBr]_{inj,1}) \qquad \text{(S4)}$$

The long-term effect was chosen to have a logarithmic dependence on $[KBr]_{inj}$ because the value of $[KBr]_{inj}$ may vary over two or more orders of magnitude. The function multiplying *a* has a maximum value of 1 at $[KBr]_{inj}$ = 6x10^{-6} M and at 5x10^{-4} M it equals 0.9338. The adjustment of *a* ensures that the cycle lengths increase slightly over time and reach a plateau when repetitive perturbations occur. The total effect remains small compared to the immediate effect of a single inhibitory pulse, which itself can even double the cycle length at high $[KBr]_{inj}$. We measured how the decrease in *a* affects the cycle length using simulations in which a single oscillator was perturbed 40 s after each peak. The cycle length increase relative to the initial cycle length: $\Delta T_r = (T_{10} - T_0)/T_0$ was calculated for the 10th perturbed cycle. When $[KBr]_{inj}$ = 5x10^{-4} M was used and perturbations were implemented so that *p* increased and *a* decreased at the instant of a perturbation, ΔT_r was 0.51. When only *p* was increased, ΔT_r was 0.36 at the same $[KBr]_{inj}$. Based on these results the overall contribution to the cycle length increase of the decrease of variable *a* is about 15% of the natural cycle length, which is comparable to our experimental observations. Lavrova and Vanag showed that a higher difference must be present for most of the N:M patterns [12]. For instance, 1:2 patterns were observed with reciprocal inhibitory coupling when T_2/T_1 was about 0.5 before the coupling was turned on. Ours is a relatively small change, and we therefore do not consider it to be the primary source of the patterns that arise when the two BZ oscillators are coupled.

(2) Simulations of inhibitory perturbation

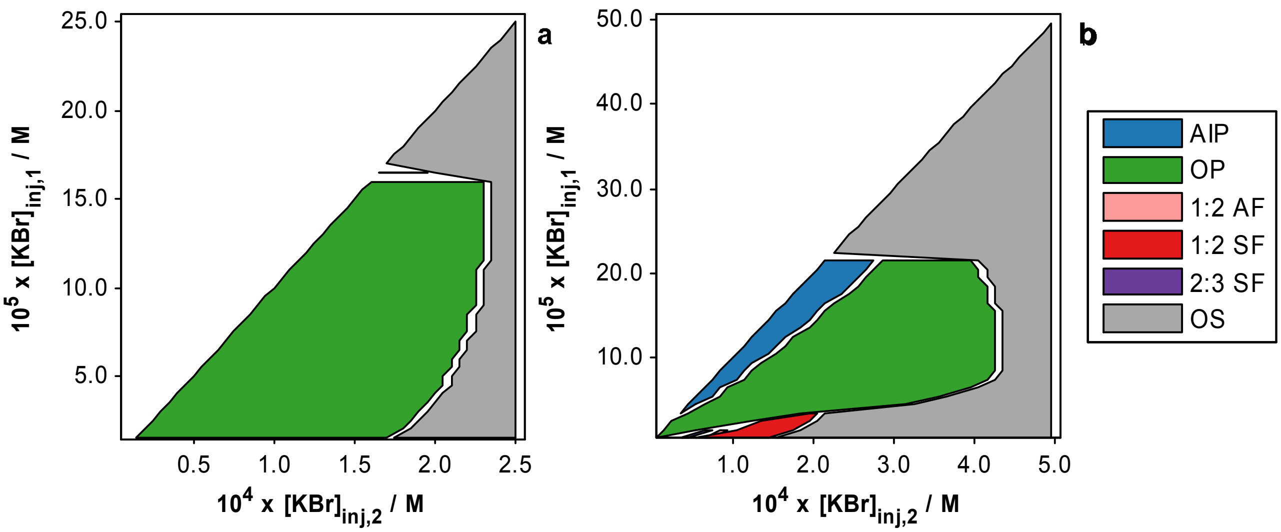

Fig S1 Temporal patterns found in numerical simulations

in the $[KBr]_{inj,1}$–$[KBr]_{inj,2}$ phase plane, a) without time delay, b) with 30 s time delay. $\Delta\varphi_0 = 0.5$. Note the difference in the scales.

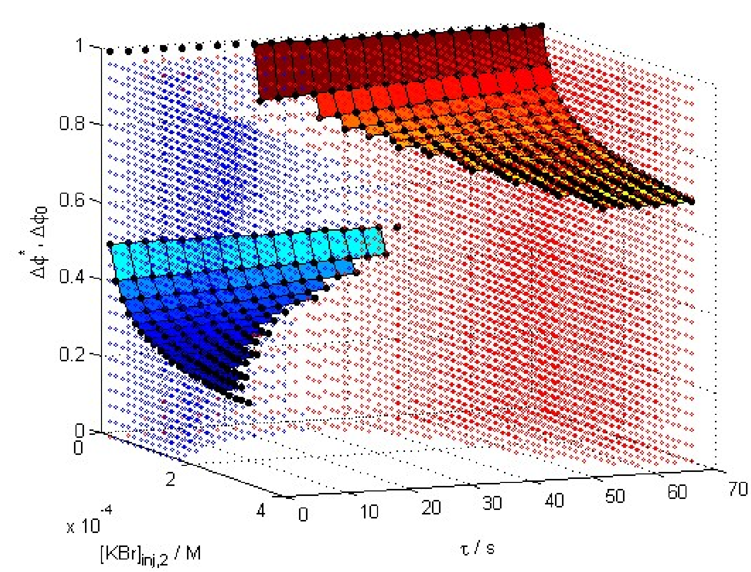

Fig S2 Stable lobes ($\Delta\varphi^*$ values) of 1:1 OP (blue) and 1:1 AIP (red) oscillations in the $[KBr]_{inj,2}$–τ–$\Delta\varphi_0$ phase space. Overlaid points: initial phase differences leading to asymptotically stable AP-like oscillations (blue) and those leading to IP-like oscillations (red).