which switches from homogeneous to heterogeneous at $t = 150$ with the removal the sublink inside the pink node in Fig. S1(a).

Appendix S2: Example of *AISync* in undirected networks

The simplest example of undirected network exhibiting *AISync* is the network of two nodes, coupled bidirectionally. Consider a system with this network structure in which node 1 has subnodes $1'$ and $1''$, and node 2 has subnodes $2'$ and $2''$, and the network of sublinks (all with weight equal to one) forms a directed chain connecting the four subnodes as $2'' \to 1' \to 1'' \to 2'$. This network, when described at the node level, is indeed undirected because the pattern of sublink connections from node 1 to 2 is identical to the pattern of connections from node 2 to 1. The nodes are heterogeneous because subnode $1'$ is connected to $1''$, while subnode $2'$ is not connected to $2''$. Since the directed chain is an optimal network with $\lambda_2 = \lambda_3 = \lambda_4 = 1$, this heterogeneous system is more synchronizable than any combination of internal sublink configurations that leads to a homogeneous system. Thus, this undirected network exhibits *AISync*.

Appendix S3: Supplementary Tables

For each of the symmetric networks that can be derived from the network diagrams in Table I for $N = 3$ and 4, we identify all possible two-layer optimal heterogeneous systems with that symmetric network structure. Tables S1 and S2 show all these systems in the monolayer representation for $N = 3$ and $N = 4$, respectively

TABLE S1. The 9 optimal heterogeneous systems with $N = 3$ and $L = 2$.

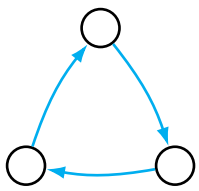
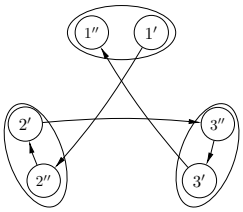
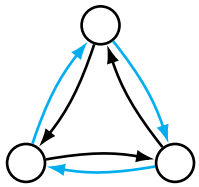
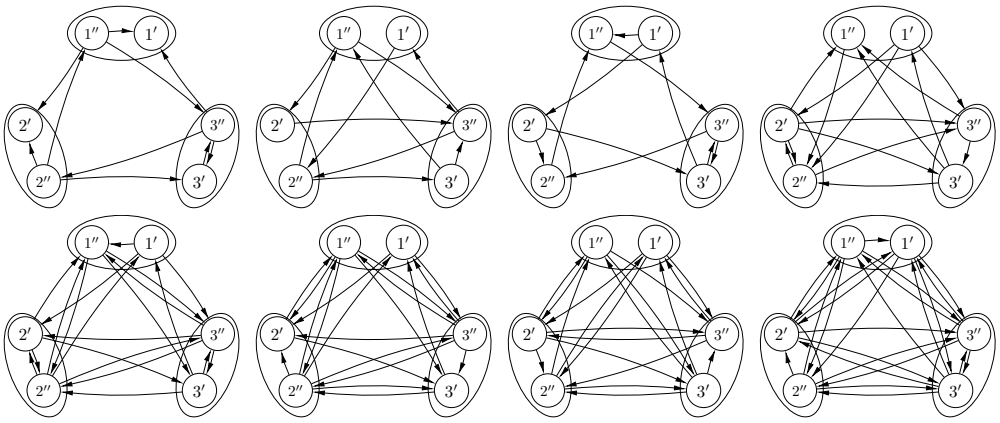
symmetric network	optimal heterogeneous systems
	
	

TABLE S2. The 14 optimal heterogeneous systems with $N = 4$ and $L = 2$.

symmetric network	optimal heterogeneous systems