ON CONSTRUCTION OF UPPER AND LOWER BOUNDS FOR THE HOMO-LUMO SPECTRAL GAP

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Abstract. In this paper we study spectral properties of graphs which are constructed from two given invertible graphs by bridging them over a bipartite graph. We analyze the so-called HOMO-LUMO spectral gap which is the difference between the smallest positive and largest negative eigenvalue of the adjacency matrix of a graph. We investigate its dependence on the bridging bipartite graph and we construct a mixed integer semidefinite program for maximization of the HOMO-LUMO gap with respect to the bridging bipartite graph. We also derive upper and lower bounds for the optimal HOMO-LUMO spectral graph by means of semidefinite relaxation techniques. Several computational examples are also presented in this paper.

1. **Introduction.** The spectrum $\sigma(G)$ of an undirected graph G consists of eigenvalues of its adjacency symmetric $n \times n$ matrix $\mathcal{A}(G)$, i.e. $\sigma(G) = \{\lambda_k(G), k = 1, \cdots, n, \ \lambda_k(G)$ is an eigenvalue of $\mathcal{A}(G)\}$, where $\lambda_1(G) \geq \cdots \geq \lambda_n(G)$ (cf. [6, 5]). If the spectrum does not contain zero there exists the inverse matrix A^{-1} of the adjacency matrix $A = \mathcal{A}(G)$, and the graph G_A is called invertible.

The concept of an inverse graph has been introduced by Godsil [10]. In addition to invertibility of the adjacency matrix it is required that A^{-1} is diagonally similar to a nonnegative or nonpositive integral matrix (cf. Godsil [10], Pavlíková and Ševčovič [23]). Notice that the least positive eigenvalue of a graph is the reciprocal value of the maximal eigenvalue of the inverse graph. Therefore properties of inverse graphs can be used in estimation of the least positive eigenvalue (cf. Pavlíková et al. [21, 22, 23]).

In many applied fields, e.g. theoretical chemistry, biology, or statistics, spectral indices and properties of graphs representing structure of chemical molecules or transition diagrams for finite Markov chains play an important role (cf. Cvetković

[6, 7], Brouwer and Haemers [5] and references therein). In the last decades, various graph energies and indices have been proposed and analyzed. For instance, the sum of absolute values of eigenvalues is referred to as the matching energy index (cf. Chen and Liu [16]), the maximum of the absolute values of the least positive and largest negative eigenvalue is known as the HOMO-LUMO index (see Mohar [19, 20], Li *et al.* [15], Jaklič *et al.* [13], Fowler *et al.* [9]), their difference is the HOMO-LUMO separation gap (cf. Gutman and Rouvray [11], Li *et al.* [15], Zhang and An [27], Fowler *et al.* [8]).

In computational chemistry, eigenvalues of a graph describing an organic molecule are related to energies of molecular orbitals. Following Hückel's molecular orbital method [12] (see also Pavlíková and Ševčovič [24]), the energies $E_k, k = 1, \cdots, n$, are the eigenvalues of the Hamiltonian matrix H and its eigenvectors are orbitals. The square symmetric matrix H has the following elements:

$H_{ii} = \alpha$ for the carbon C atom at the i-th vertex, and $H_{ii} = \alpha + h_A \beta$ for other atoms A, where $\alpha < 0$ is the Coulomb integral and $\beta < 0$ is the resonance integral;

$H_{ij} = \beta$ if both vertices i and j are carbon C atoms, $H_{ij} = k_{AB}\beta$ for other neighboring atoms A and B;

$H_{ij} = 0$ otherwise.

The atomic constants h_A, k_{AB} have to be specified ($h_C = k_{CC} = 0$). For instance, the molecule of pyridine contains one atom of nitrate N and five atoms of carbon C. Clearly, in the case of pure hydrocarbon we have $H = \alpha I + \beta A$ where I is the identity and A is the adjacency matrix of the molecular structural graph G. Hence $E_k = \alpha + \beta \lambda_k$. Now, the energy E_{HOMO} of the highest occupied molecular orbital (HOMO) corresponds to the eigenvalue $\lambda_{HOMO} = \lambda_k$ where $k = n/2$ for n even and $k = (n+1)/2$ for n odd. The energy E_{LUMO} of the lowest unoccupied molecular orbital (LUMO) corresponds to the subsequent eigenvalue $\lambda_{LUMO} = \lambda_{k+1}$ for n even, and $\lambda_{LUMO} = \lambda_k$ for n odd. The HOMO-LUMO separation gap is the difference between E_{LUMO} and E_{HOMO} energies, i.e. $E_{LUMO} - E_{HOMO} = -\beta(\lambda_{HOMO} - \lambda_{LUMO}) \geq 0$ because $\beta < 0$. The so-called properly closed shells have the property $\lambda_{HUMO} > 0 > \lambda_{LUMO}$ containing either zero or two electrons are called closed shells for which n is even (cf. Fowler and Pisanski [9]). For such orbital systems, the HOMO-LUMO separation gap is equal to the energy difference $E_{LUMO} - E_{HOMO} = -\beta \Lambda_{HL}(G_A)$ where

$$\Lambda_{HL}(G_A) = \check{\lambda}^+(G_A) - \hat{\lambda}^-(G_A). \tag{1}$$

Here $\check{\lambda}^+(G_A) = \lambda_k$ is the smallest positive eigenvalue, and $\hat{\lambda}^-(G_A) = \lambda_{k+1}$ is the largest negative eigenvalue of the adjacency matrix A of the structural molecular graph G_A (cf. [9]). According to Aihara [1, 2] the large HOMO-LUMO gap implies high kinetic stability and low chemical reactivity of the molecule, because it is energetically unfavorable to add electrons to a high-lying LUMO orbital. Notice that the HOMO-LUMO energy gap is generally decreasing with the size n of the structural graph (cf. Bacalis and Zdetsis [3]).

In this paper, our goal is to investigate extremal properties of the HOMO-LUMO spectral gap $\Lambda_{HL}(G_A)$. We show how to represent $\Lambda_{HL}(G_A)$ by means of the optimal solution to a convex semidefinite programming problem (Section 2). We study spectral properties of graphs which can be constructed from two given (not necessarily bipartite) graphs by bridging them over a bipartite graph (Section 3). We analyze their HOMO-LUMO spectral gap of such a bridged graph and its dependence

on the bridging bipartite graph. Finding an optimal bridging bipartite graph leads to a mixed integer nonconvex optimization problem with linear matrix inequality constraints (Section 4). We prove that the optimal HOMO-LUMO spectral gap can be obtained by solving a mixed integer semidefinite convex program. The optimization problem is, in general, NP hard (Section 5). This is why we also derive upper (Section 6) and lower (Section 7) bounds for the optimal HOMO-LUMO spectral graph by means of semidefinite relaxation techniques which can be solved in a fast and computationally efficient way. Various computational examples of construction of the optimal bridging graph are presented in Section 8.

2. **Semidefinite programming representation of the HOMO-LUMO spectral gap.** The HOMO-LUMO spectral gap of a graph G_C is defined as follows:

$$\Lambda_{HL}(G_C) = \check{\lambda}^+(G_C) - \hat{\lambda}^-(G_C),$$

where $\check{\lambda}^+(G_C) \geq 0$ is the smallest nonnegative eigenvalue, and $\hat{\lambda}^-(G_C) \leq 0$ is the largest nonpositive eigenvalue of the adjacency matrix C. Notice that the spectrum $\sigma(G_C) = \sigma(C)$ of a nontrivial graph G_C without loops must contain negative as well as positive eigenvalues because the trace $Tr(C) = \sum_{\lambda \in \sigma(C)} \lambda = 0$. Clearly, if the graph G_C is invertible then $\check{\lambda}^+(G_C) > 0$ and $\hat{\lambda}^-(G_C) < 0$ and so $\Lambda_{HL}(G_C) > 0$, otherwise $\Lambda_{HL}(G_C) = 0$.

2.1. **Semidefinite representation of the HOMO-LUMO gap.** Suppose that a graph G_C is invertible. Following [23] the smallest positive and largest negative eigenvalues of G_C can be expressed as follows:

$$\check{\lambda}^+(G_C) = \frac{1}{\lambda_{max}(C^{-1})}, \qquad \hat{\lambda}^-(G_C) = \frac{1}{\lambda_{min}(C^{-1})},$$

where $\lambda_{max}(C^{-1}) > 0$ and $\lambda_{min}(C^{-1}) = -\lambda_{max}(-C^{-1}) < 0$ are the maximum and minimum eigenvalues of the inverse matrix C^{-1}, respectively. We denote by $\preceq$ the Löwner partial ordering on symmetric matrices, i.e. $A \preceq B$ iff the matrix $B - A$ is a positive semidefinite matrix, that is $B - A \succeq 0$. The maximal and minimal eigenvalues of C^{-1} can be expressed as follows:

$$0 < \lambda_{max}(C^{-1}) = \min_{C^{-1} \preceq tI} t, \qquad 0 > \lambda_{min}(C^{-1}) = \max_{sI \preceq C^{-1}} s,$$

(see e.g. [4], [7]). Since $\{t, \ C^{-1} \preceq tI\} \subset (0, \infty)$ and $\{s, \ sI \preceq C^{-1}\} \subset (-\infty, 0)$ then, by using the substitution $\mu = 1/t, \eta = -1/s$, we obtain the following characterization of the lowest positive and largest negative eigenvalues of the graph G_C:

$$\check{\lambda}^+(G_C) = \max_{\mu C^{-1} \preceq I} \mu, \qquad \hat{\lambda}^-(G_C) = - \max_{-\eta C^{-1} \preceq I} \eta. \tag{2}$$

As a consequence, we obtain the following semidefinite representation of the HOMO-LUMO spectral gap for a vertex labeled invertible graph G_C without loops. Then the HOMO-LUMO spectral gap $\Lambda_{HL}(G_C)$ of the graph G_C is the optimal value of the following semidefinite programming problem:

$$\begin{aligned} \Lambda_{HL}(G_C) \quad = \quad & \max_{\mu, \eta \geq 0} \quad \mu + \eta \\ & s.t. \quad \mu C^{-1} \preceq I, \\ & \qquad -\eta C^{-1} \preceq I. \end{aligned} \tag{3}$$

(cf. Pavlíková and Ševčovič [24]).

3. **Graphs bridged over a bipartite graph.** In this section we introduce a notion of a graph which is constructed from two given graphs G_A and G_B by bridging vertices of G_A to vertices of G_B. More, precisely, let G_A and G_B be two undirected vertex-labeled graphs on n and m vertices without loops, respectively. In general, we do not assume that G_A and G_B are bipartite graphs. Let G_K be a (n, m)-bipartite graph on $n + m$ vertices with the adjacency matrix:

$$\mathcal{A}(G_K) = \begin{pmatrix} 0 & K \\ K^T & 0 \end{pmatrix}, \tag{4}$$

where K is an $n \times m$ matrix containing $\{0, 1\}$-elements only.
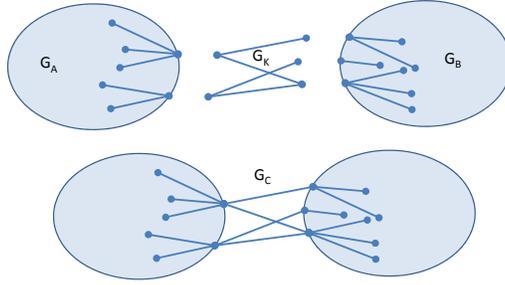

FIGURE 1.   A bridged graph $G_C = \mathcal{B}_K(G_A, G_B)$ through a bipartite graph G_K.

By $\mathcal{B}_K(G_A, G_B)$ we shall denote the graph G_C on $n+m$ vertices which is obtained by bridging the vertices of the graph G_A to the vertices of G_B through the (n, m)-bipartite graph G_K, i.e. its adjacency matrix $C = \mathcal{A}(G_C)$ of the graph G_C has the form:

$$C = \begin{pmatrix} A & K \\ K^T & B \end{pmatrix}, \tag{5}$$

In what follows, we will assume that the adjacency matrices A and B are symmetric $n \times n$ and $m \times m$ invertible matrices, respectively.

Theorem 3.1. *Let G_A and G_B be two undirected vertex-labeled invertible graphs on n and m vertices, respectively. Let G_K be a (n, m)-bipartite graph. Let $G_C = \mathcal{B}_K(G_A, G_B)$ be the graph which is constructed by bridging the graphs G_A and G_B through the bipartite graph G_K.*

Then the graph G_C is invertible if and only if the $n \times n$ matrix $S = A - KB^{-1}K^T$ is invertible. In this case we have

$$\begin{aligned} C^{-1} &= \begin{pmatrix} A & K \\ K^T & B \end{pmatrix}^{-1} = \begin{pmatrix} S^{-1} & -S^{-1}KB^{-1} \\ -B^{-1}K^TS^{-1} & B^{-1} + B^{-1}K^TS^{-1}KB^{-1} \end{pmatrix} \cdot \\ &= Q^T \begin{pmatrix} S^{-1} & 0 \\ 0 & B^{-1} \end{pmatrix} Q, \end{aligned} \tag{6}$$

where Q is an invertible matrix with the inverse $Z = Q^{-1}$ given by:

$$Q = \begin{pmatrix} I & -KB^{-1} \\ 0 & I \end{pmatrix}, \qquad Z = \begin{pmatrix} I & KB^{-1} \\ 0 & I \end{pmatrix}.$$

P r o o f. The proof is a direct consequence of the Schur complement theorem (see e. g. [18, Theorem A.6]). Indeed, $C \begin{pmatrix} x \\ y \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}$ if and only if $Ax + Ky = 0$ and $K^T x + By = 0$, that is, $Sx = (A - KB^{-1}K^T)x = 0$. As $x \neq 0 \Leftrightarrow y \neq 0$ we have C is invertible if and only if S is invertible. The rest of the proof is a straightforward verification of the form of the inverse matrix C^{-1}. $\diamond$

3.1. **Semidefinite representation of the HOMO-LUMO gap for a bridged graph.** Now, let $G_C = \mathcal{B}_K(G_A, G_B)$ be the graph obtained from graphs G_A and G_B by bridging them through a bipartite graph G_K with adjacency matrix K (4).

Then, for any $\mu \geq 0$, we have $\mu C^{-1} \preceq I$ if and only if $\mu Z^T C^{-1} Z \preceq Z^T Z$, i.e.,

$$\mu \begin{pmatrix} S^{-1} & 0 \\ 0 & B^{-1} \end{pmatrix} \preceq Z^T Z = \begin{pmatrix} I & KB^{-1} \\ B^{-1}K^T & I + B^{-1}K^T K B^{-1} \end{pmatrix}.$$

Therefore,

$$\mu C^{-1} \preceq I \quad \Leftrightarrow \quad \begin{pmatrix} I - \mu S^{-1} & KB^{-1} \\ B^{-1}K^T & I - \mu B^{-1} + B^{-1}K^T K B^{-1} \end{pmatrix} \succeq 0. \quad (7)$$

Similarly,

$$-\eta C^{-1} \preceq I \quad \Leftrightarrow \quad \begin{pmatrix} I + \eta S^{-1} & KB^{-1} \\ B^{-1}K^T & I + \eta B^{-1} + B^{-1}K^T K B^{-1} \end{pmatrix} \succeq 0. \quad (8)$$

With regard to (4) we obtain the following representation of the HOMO-LUMO spectral gap $\Lambda_{HL}(G_C)$ for a the bridged graph:

$$\Lambda_{HL}(G_C) \quad = \quad \max_{\mu, \eta \geq 0} \quad \mu + \eta \quad (9)$$

$$s.t. \quad \begin{pmatrix} I - \mu S^{-1} & KB^{-1} \\ B^{-1}K^T & I - \mu B^{-1} + B^{-1}K^T K B^{-1} \end{pmatrix} \succeq 0,$$

$$\begin{pmatrix} I + \eta S^{-1} & KB^{-1} \\ B^{-1}K^T & I + \eta B^{-1} + B^{-1}K^T K B^{-1} \end{pmatrix} \succeq 0.$$

Since for the Schur complement we have $S = A - KB^{-1}K^T$ then the matrix inequality constraints appearing in (9) represent, in general, nonconvex constraints with respect to the matrix K. To overcome this difficulty we further restrict the class of bipartite graphs G_K bridging G_A to G_B to those turning (9) to a convex semidefinite program in the K variable.

Definition 3.2. [23] Let G_B be an undirected vertex-labeled graph on m vertices with an invertible adjacency matrix B. We say that G_B is arbitrarily bridgeable over the first $\{1, \cdots, k_B\}$ vertices of G_B if the $k_B \times k_B$ upper principal sub-matrix of B^{-1} is a null matrix, i.e. $EB^{-1}E^T = 0$ where $E = (I, 0)$ is a $k_B \times m$ block matrix and I is a $k_B \times k_B$ identity matrix.

A graph G_B is said to be arbitrarily bridgeable over the subset $\{i_1, \cdots, i_{k_B}\}$ of vertices of G_B if there exists a permutation P of its vertices such that $i_k \mapsto k, k = 1, \cdots, k_B$, and $E\tilde{B}^{-1}E^T = 0$ where $\tilde{B} = P^T B P$.

Notice that if G_B is arbitrarily bridgeable then $k_B \leq m/2$ because there is no regular $m \times m$ matrix B^{-1} such that $EB^{-1}E^T = 0$ for $k_B > m/2$.

Using the notion of an arbirtarily bridgeable graph we conclude the following theorem:

Theorem 3.3. *Let G_A and G_B be undirected vertex-labeled invertible graphs on n and m vertices without loops, respectively. Assume that G_B is arbitrarily bridgeable over the first $\{1, \cdots, k_B\}$ vertices of G_B. If the $n \times m$ matrix K has zero last $m-k_B$ columns, i.e. $K_{ij} = 0$ for $j = k_B + 1, \cdots m$, then $KB^{-1}K^T = 0$, and, consequently, for the Schur complement S we have $S = A - KB^{-1}K^T = A$, and $S^{-1} = A^{-1}$.*

Moreover, the HOMO-LUMO spectral gap $\Lambda_{HL}(G_C)$ for the bridged graph $G_C = \mathcal{B}_K(G_A, G_B)$ through the bipartite graph G_K is the optimal value of the following semidefinite programming problem:

$$\Lambda_{HL}(G_C) \quad = \quad \max_{\mu, \eta \, \geq \, 0} \quad \mu + \eta \tag{10}$$

$$s.t. \quad \begin{pmatrix} I - \mu A^{-1} & KB^{-1} \\ B^{-1}K^T & I - \mu B^{-1} + B^{-1}K^T K B^{-1} \end{pmatrix} \succeq 0,$$

$$\begin{pmatrix} I + \eta A^{-1} & KB^{-1} \\ B^{-1}K^T & I + \eta B^{-1} + B^{-1}K^T K B^{-1} \end{pmatrix} \succeq 0.$$

4. **Construction of an optimal bridging bipartite graph by means of a mixed integer nonlinear programming problem.** In this section we focus our attention on extremal properties of the HOMO-LUMO spectral gap for bridged graphs. Given an invertible graph G_A and arbitrarily bridgeable invertible graph G_B, over the first $\{1, \cdots, k_B\}$ vertices of G_B, our goal is to find an optimal bridging graph G_K (see (4)) such that $K_{ij} = 0$ for $j = k_B + 1, \cdots, m$ and the HOMO-LUMO spectral gap $\Lambda_{HL}(G_C)$ is maximal, where $G_C = \mathcal{B}_K(G_A, G_B)$.

Using representation of $\Lambda_{HL}(G_C)$ for the graph $G_C = \mathcal{B}_K(G_A, G_B)$ (see Theorem 3.3), the maximal HOMO-LUMO gap $\Lambda_{HL}^{opt} = \Lambda_{HL}^{opt}(G_A, G_B)$ with respect to a bipartite matrix K is given as the optimal value of the following mixed integer nonlinear optimization problem:

$$\Lambda_{HL}^{opt} \quad = \quad \max_{\substack{\mu, \eta \, \geq \, 0 \\ K, W}} \quad \mu + \eta \tag{11}$$

$$s.t. \quad \begin{pmatrix} I - \mu A^{-1} & KB^{-1} \\ B^{-1}K^T & I - \mu B^{-1} + B^{-1}W B^{-1} \end{pmatrix} \succeq 0,$$

$$\begin{pmatrix} I + \eta A^{-1} & KB^{-1} \\ B^{-1}K^T & I + \eta B^{-1} + B^{-1}W B^{-1} \end{pmatrix} \succeq 0,$$

$$W = K^T K, \quad K_{ij} \in \{0, 1\} \quad \text{for all } i, j, \quad \sum_{k,l} K_{kl} \geq 1,$$

$$K_{ij} = 0 \ \text{for } j = k_B + 1, \cdots, m, \ \ i = 1, \cdots, n.$$

Notice that the condition $K \neq 0$ for a binary matrix K is equivalent to the condition $\sum_{k,l} K_{kl} \geq 1$. The objective function as well as the first two matrix inequality constraints in the optimization problem (11) are linear[1] in the variables μ, η, K, W. However, the last two constraints in (11) make the problem considerably harder to solve because of the nonconvex constraint $W = K^T K$ and the binary constraint

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[1] Convex semidefinite problems with linear matrix inequality constraints can be solved by means of computational Matlab toolboxes available for semidefinite programming, e.g. SeDuMi solver developed by J. Sturm [26] with Yalmip Matlab programming framework due to J. Löfberg [17].

$K_{ij} \in \{0, 1\}$. It means that (11) is a mixed integer nonconvex programming problem which is, in general, NP-hard to solve.

5. **Construction of upper bounds for the HOMO-LUMO spectral gap by semidefinite relaxation techniques.** In the field of solving mixed integer nonconvex problems various techniques have been developed in the last decades. We refer the reader to the book [4] by Boyd and Vanderberghe on recent developments on semidefinite relaxation methods for solving nonconvex and mixed integer nonlinear optimization problems. In general, semidefinite relaxations of an original nonconvex problem can be constructed by means of the second Lagrangian dual problem which is already a convex semidefinite problem (see e.g. Ševčovič and Trnovská [25]).

5.1. **Mixed semidefinite-integer relaxation.** In order to construct a suitable convex programming relaxation of (11) we have to enlarge the domain of variables μ, η, K, W. Notice that the integer constraint $K_{ij} \in \{0, 1\}$ is equivalent to the equality: $K_{ij} = K_{ij}^2$. Moreover, from the constraint $W = K^T K$ we deduce $W_{ij} \in \mathbb{N}_0^+$ and $W_{jj} = \sum_l K_{lj}^2 = \sum_l K_{lj}$. The nonconvex constraint $W = K^T K$ can be relaxed by a convex matrix inequality constraint $W \succeq K^T K$. Using the Schur complement theorem (cf. [18]), it can be rewritten as a linear matrix inequality constraint:

$$W \succeq K^T K \quad \Leftrightarrow \quad \begin{pmatrix} W & K^T \\ K & I \end{pmatrix} \succeq 0.$$

Hence the nonconvex-integer programming problem (11) can be relaxed by means of the following mixed integer semidefinite programming problem with linear matrix inequality constraints and integer constraints for the upper bound approximation $\overline{\Lambda}_{HL}^{sir} = \overline{\Lambda}_{HL}^{sir}(G_A, G_B)$:

$$
\begin{aligned}
\overline{\Lambda}_{HL}^{sir} \quad = \quad & \max_{\substack{\mu, \eta \geq 0 \\ K, W}} \quad \mu + \eta \\
s.t. \quad & \begin{pmatrix} I - \mu A^{-1} & K B^{-1} \\ B^{-1} K^T & I - \mu B^{-1} + B^{-1} W B^{-1} \end{pmatrix} \succeq 0, \\
& \begin{pmatrix} I + \eta A^{-1} & K B^{-1} \\ B^{-1} K^T & I + \eta B^{-1} + B^{-1} W B^{-1} \end{pmatrix} \succeq 0, \\
& \begin{pmatrix} W & K^T \\ K & I \end{pmatrix} \succeq 0, \\
& K_{ij} \in \{0, 1\}, \quad W_{ij} \in \mathbb{N}_0^+, \quad W_{jj} = \sum_l K_{lj} \quad \text{for all } i, j, \ \sum_{k,l} K_{kl} \geq 1. \\
& K_{ij} = 0 \ \text{for } j = k_B + 1, \cdots, m, \ \ i = 1, \cdots, n.
\end{aligned}
$$
(12)

It is worth noting that if $(\hat{\mu}, \hat{\eta}, \hat{K}, \hat{W})$ is the optimal solution to the mixed integer semidefinite programming problem (12) then $(\hat{\mu}, \hat{\eta}, \hat{K}, \hat{W})$ is also feasible for (11) because $\hat{W} = \hat{K}^T \hat{K}$. Indeed, if we denote $L = \hat{W} - \hat{K}^T \hat{K}$ then $L \succeq 0$ and $L_{jj} = \hat{W}_{jj} - \sum_l \hat{K}_{lj}^2 = \hat{W}_{jj} - \sum_l \hat{K}_{lj} = 0$. Hence $diag(L) = 0$ and so $L = 0$, as claimed. Consequently, the HOMO-LUMO gap $\Lambda_{HL}(\mathcal{B}_{\hat{K}}(G_A, G_B)) = \overline{\Lambda}_{HL}^{sir}(G_A, G_B)$. Hence

$$\Lambda_{HL}^{opt}(G_A, G_B) = \overline{\Lambda}_{HL}^{sir}(G_A, G_B).$$

Next we present a sample code for solving the mixed integer semidefinite programming problem (12) for construction of the optimal bridging for maximal HOMO-LUMO spectral gap $\overline{\Lambda}_{HL}^{sir}(G_A, G_B) = \overline{\Lambda}_{HL}^{opt}(G_A, G_B)$. We employed the Matlab programming environment Yalmip which is capable of solving mixed integer problems with semidefinite linear matrix inequality constraints due to Löfberg [17]). The structure of the code is shown in Table 5.1. After declaring classes of variables and setting the constraints, then the main solver routine `solvsdp` is executed. It is designed for solving minimization problem. It employs SeDuMi semidefinite programming solver (cf. Sturm [26]) as the lower solver and branch and bound integer rounding solver as the upper solver.

TABLE 1. A sample Matlab code for computing mixed integer semidefinite programming problem (12). The output of the program is the optimal value $\Lambda_{HL}^{opt}(G_A, G_B) = \overline{\Lambda}_{HL}^{sir}(G_A, G_B)$.

```
mu=sdpvar(1); eta=sdpvar(1); W=intvar(m,m); K=binvar(n,m);
ops=sdpsettings('solver','bnb','bnb.maxiter', bnbmaxiter);

Fconstraints=[...
    [[W, K'];
    [K, eye(n,n)]
    ]>=0, ...
    mu>=0, eta>=0, ...
    [[eye(n,n) - mu*inv(A), K*inv(B)];
    [inv(B)*K', eye(m,m) - mu*inv(B) + inv(B)*W*inv(B)]
    ] >= 0, ...
    [[eye(n,n) + eta*inv(A), K*inv(B)];
        [inv(B)*K', eye(m,m) + eta*inv(B) + inv(B)*W*inv(B)]
    ] >= 0, ...
    sum(K(:,:))==diag(W)', sum(K(:))>=1, ...
    vec(W(:))>=0, 0<=vec(K(:))<=1, ...
    sum([[A, K]; [K', B] ])<=maxdegree*ones(1,n+m), ...,
    K*[zeros(kB,m-kB); eye(m-kB,m-kB)] == zeros(n, m-kB), ...
    ];

solvesdp(Fconstraints, -mu-eta, ops)

LambdaSIR = double(mu + eta)
```

5.2. **Full semidefinite relaxation.** Next, we further relax the binary and integer constraints appearing in (12). The integer constraint $K_{ij} \in \{0,1\}$ can be relaxed by the box convex inequality constraints: $0 \le K_{ij} \le 1$ for all i,j. Clearly, such a relaxation may lead to a non-integer optimal matrix K. The maximization problem for the full semidefinite relaxation of the HOMO-LUMO spectral gap $\overline{\Lambda}_{HL}^{sdp} = \overline{\Lambda}_{HL}^{sdp}(G_A, G_B)$ can be formulated as follows:

$$
\overline{\Lambda}_{HL}^{sdp} = \max_{\substack{\mu, \eta \ge 0 \\ K, W}} \mu + \eta
$$

$$
s.t. \quad \begin{pmatrix} I - \mu A^{-1} & K B^{-1} \\ B^{-1} K^T & I - \mu B^{-1} + B^{-1} W B^{-1} \end{pmatrix} \succeq 0,
$$

$$\begin{pmatrix} I + \eta A^{-1} & KB^{-1} \\ B^{-1}K^T & I + \eta B^{-1} + B^{-1}WB^{-1} \end{pmatrix} \succeq 0, \tag{13}$$

$$\begin{pmatrix} W & K^T \\ K & I \end{pmatrix} \succeq 0,$$

$$0 \le K_{ij} \le 1, \quad W_{jj} = \sum_l K_{lj}, \quad W_{ij} \ge 0 \text{ for all } i,j, \quad \sum_{k,l} K_{kl} \ge 1,$$

$$K_{ij} = 0 \quad \text{for } j = k_B + 1, \cdots, m, \quad i = 1, \cdots, n. \tag{14}$$

In order to compute $\overline{\Lambda}_{HL}^{sdp}(G_A, G_B)$ the full semidefinite relaxation (13) we have to change the specification of real variables, i.e. `W=sdpvar(m,m); K=sdpvar(n,m)` and add the box constraint `0<=vec(K(:))<=1` in the code shown in Table 5.1.

Remark 1. Following the recent paper by Kim, Kojima and Toh [14], the box constraint $0 \le K_{ij} \le 1$ can be further enhanced by introducing a slack variable $\tilde{K}$ where $\tilde{K}_{ij} = 1 - K_{ij}$. Then $K_{ij} \in \{0,1\}$ if and only if $K_{ij}\tilde{K}_{ij} = 0$ for all i,j. It is equivalent to the condition $V_{jj} = 0$ for each j, where $V = \tilde{K}^T K$. Next, the nonconvex matrix constraints $W = K^T K, \tilde{W} = \tilde{K}^T \tilde{K}$, can be relaxed in the form of the following linear matrix inequality:

$$\begin{pmatrix} W & V^T \\ V & \tilde{W} \end{pmatrix} \succeq \begin{pmatrix} K^T \\ \tilde{K}^T \end{pmatrix} \begin{pmatrix} K & \tilde{K} \end{pmatrix} \quad \Longleftrightarrow \quad \begin{pmatrix} W & V^T & K^T \\ V & \tilde{W} & \tilde{K}^T \\ K & \tilde{K} & I \end{pmatrix} \succeq 0,$$

$W_{ij}, \tilde{W}_{ij}, V_{ij} \ge 0, \ V_{jj} = 0$ for all i,j.

Theorem 5.1. *Let G_A and G_B be undirected vertex-labeled invertible graphs on n and m vertices without loops, respectively. Assume G_B is arbitrarily bridgeable over the first k_B vertices $\{1, \cdots, k_B\}$. Then*

$$\Lambda_{HL}(G_C) \le \Lambda_{HL}^{opt}(G_A, G_B) \equiv \overline{\Lambda}_{HL}^{sir}(G_A, G_B) \le \overline{\Lambda}_{HL}^{sdp}(G_A, G_B) \le \Lambda_{HL}(G_A),$$

for any graph $G_C = \mathcal{B}_K(G_A, G_B)$ which is constructed from graphs G_A, G_B by bridging the vertices of G_A to the first k_B vertices of G_B through an (n,m)-bipartite graph G_K such that $K_{ij} = 0$ for $j = k_B + 1, \cdots, m$.

P r o o f. The set

$$\{(K,W), \ K_{ij} \in \{0,1\}, \quad W_{ij} \in \mathbb{N}_0^+, \quad W_{jj} = \sum_l K_{lj} \quad \text{for all } i,j,$$

$$\sum_{k,l} K_{kl} \ge 1, W \succeq K^T K\}$$

of feasible integer matrices K, W for (12) is a subset of the set:

$$\{(K,W), \ 0 \le K_{ij} \le 1, \quad W_{ij} \ge 0, \quad W_{jj} = \sum_l K_{lj}, \quad \text{for all } i,j,$$

$$\sum_{k,l} K_{kl} \ge 1, W \succeq K^T K\},$$

of real matrices K, W that are feasible for (13). From this fact we conclude the inequality $\overline{\Lambda}_{HL}^{sir}(G_A, G_B) \le \overline{\Lambda}_{HL}^{sdp}(G_A, G_B)$. The inequality $\overline{\Lambda}_{HL}^{sdp}(G_A, G_B) \le \Lambda_{HL}(G_A)$ follows from the fact that

$$\begin{pmatrix} I - \mu A^{-1} & KB^{-1} \\ B^{-1}K^T & I - \mu B^{-1} + B^{-1}WB^{-1} \end{pmatrix} \succeq 0 \quad \Longrightarrow I - \mu A^{-1} \succeq 0,$$

that is $1/\mu \geq \lambda_{max}(A^{-1})$ and so $\mu \leq \check{\lambda}^+(G_A)$. Similarly, we obtain $I + \eta A^{-1} \succeq 0$ and, consequently, $\eta \leq -\hat{\lambda}^-(G_A)$. Therefore, $\mu + \eta \leq \Lambda_{HL}(G_A)$, as claimed. $\diamondsuit$
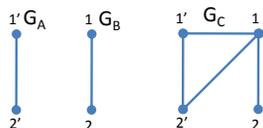

FIGURE 2. Simple graphs G_A and G_B (left) and the bridged graph G_C with the maximal HOMO-LUMO spectral gap which can be constructed by bridging G_A and G_B over the vertex 1 of G_B ($k_B = 1$) to the vertices of G_A (right).

Example 1. In Figure 2 (left) we show two simple graphs G_A and G_B having the spectrum $\sigma(G_A) = \sigma(G_B) = \{1, -1\}$, i.e. $\Lambda_{HL}(G_A) = 2$. The graph G_B is arbitrarily bridgeable over the vertex 1. The optimal bipartite graph G_K bridging G_B to G_A with $k_B = 1$ has the adjacency matrix $K = (1, 1)^T$. The optimal bridged graph G_C is shown in Figure 2 (right) and it has the spectrum $\sigma(G_C) = \{2.1701, 0.3111, -1, -1.4812\}$, i.e. $\Lambda_{HL}^{opt}(G_A, G_B) = \overline{\Lambda}_{HL}^{sir}(G_A, G_B) = 1.3111$. On the other hand, it turns out that $\overline{\Lambda}_{HL}^{sdp}(G_A, G_B) = 1.67597$. Hence we have the strict inequalities

$$\Lambda_{HL}^{opt}(G_A, G_B) \equiv \overline{\Lambda}_{HL}^{sir}(G_A, G_B) < \overline{\Lambda}_{HL}^{sdp}(G_A, G_B) < \Lambda_{HL}(G_A),$$
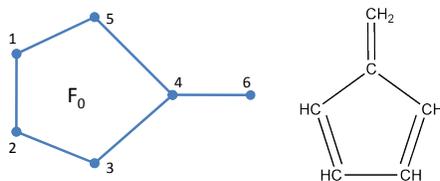
in this example.

FIGURE 3. An example of an invertible graph F_0 (left) representing the chemical organic molecule of fulvene (right).

In Figure 3 (left) we show the graph F_0 on 6 vertices representing the fulvene organic molecule (5-methylidenecyclopenta-1,3-diene) (right). The spectrum consists of the following eigenvalues:

$$\sigma(F_0) = \{2.1149, 1, 1/q, -0.2541, -q, -1.8608\},$$

where $q = (\sqrt{5} + 1)/2$ is the golden ratio. The HOMO-LUMO spectral gap $\Lambda_{HL}(F_0) = 0.872134$. It is easy to verify that the graph $G_B \equiv F_0$ is arbitrarily bridgeable over the following subsets of vertices: $\{5\}, \{4\}, \{3\}, \{2\}, \{1\}$ for $k_B = 1$, $\{4, 5\}, \{2, 5\}, \{3, 4\}, \{2, 4\}, \{1, 4\}, \{1, 3\}, \{1, 2\}$ for $k_B = 2$, and $\{2, 4, 5\}, \{1, 3, 4\}$, $\{1, 2, 4\}$ for $k_B = 3$ (cf. Pavlíková and Ševčovič [23]).

6. **Lower bounds for the optimal HOMO-LUMO spectral gap.** In this section, our aim is to derive lower bounds for the optimal HOMO-LUMO separation gap $\Lambda_{HL}^{opt}(G_A, G_B)$. Similarly as in derivation of upper bounds we will construct the lower bound by means of a solution to a certain nonlinear optimization problem.

The idea is based on construction of upper bounds for the maximal eigenvalues $\lambda_{max}(\pm C^{-1})$ of the inverse matrices C^{-1} and $-C^{-1}$. Here C is the adjacency matrix of the bridged graph $G_C = \mathcal{B}_K(G_A, G_B)$. This way we obtain a lower bound for the first positive and negative eigenvalues of C yielding the HOMO-LUMO spectral gap for G_C.

The maximal eigenvalue $\lambda_{max}(C^{-1})$ can be expressed by means of the Rayleigh quotient, and, consequently, it can be estimated as follows:

$$
\begin{aligned}
\lambda_{max}(C^{-1}) &= \max_{\|z\|^2=1} z^T C^{-1} z = (Qz)^T \begin{pmatrix} A^{-1} & 0 \\ 0 & B^{-1} \end{pmatrix} Qz \\
&= (x - KB^{-1}y)^T A^{-1}(x - KB^{-1}y) + y^T B^{-1}y \\
&\leq \lambda_{max}(A^{-1})\|x - KB^{-1}\|^2 + \lambda_{max}(B^{-1})\|y\|^2,
\end{aligned}
$$

where $z = (x, y) \in \mathbb{R}^n \times \mathbb{R}^m$ and the matrix Q is given as in (6). Analogously,

$$
\lambda_{max}(-C^{-1}) \leq \lambda_{max}(-A^{-1})\|x - KB^{-1}\|^2 + \lambda_{max}(-B^{-1})\|y\|^2.
$$

To estimate the right hand side of the estimate for $\lambda_{max}(\pm C^{-1})$ we apply the following auxiliary lemma proved in [23].

Lemma 6.1. [23, Lemma 1] *Assume that D is an $n \times m$ matrix and $\alpha, \beta > 0$ are positive constants. Then, for the optimal value γ^* of the following constrained optimization problem:*

$$
\begin{aligned}
\gamma^* = \max \quad & \alpha\|x - Dy\|^2 + \beta\|y\|^2 \\
\text{s.t.} \quad & \|x\|^2 + \|y\|^2 = 1, \quad x \in \mathbb{R}^n, y \in \mathbb{R}^m,
\end{aligned}
\tag{15}
$$

we have the explicit expression:

$$
\begin{aligned}
\gamma^* &= \max\left\{\gamma, \; \frac{(\gamma - \alpha)(\gamma - \beta)}{\alpha\gamma} \in \sigma(D^T D)\right\} \\
&= \frac{\alpha(\omega^* + 1) + \beta + \sqrt{(\alpha(\omega^* + 1) + \beta)^2 - 4\alpha\beta}}{2},
\end{aligned}
$$

where $\omega^* = \max\{\sigma(D^T D)\}$ is the maximal eigenvalue of the matrix $D^T D$.

With help of the previous lemma we obtain the upper estimate:

$$
\lambda_{max}(\pm C^{-1}) \leq \frac{\alpha^{\pm}(\omega^* + 1) + \beta^{\pm} + \sqrt{(\alpha^{\pm}(\omega^* + 1) + \beta^{\pm})^2 - 4\alpha^{\pm}\beta^{\pm}}}{2}
$$

where $\alpha^{\pm} = \lambda_{max}(\pm A^{-1})$, $\beta^{\pm} = \lambda_{max}(\pm B^{-1})$, and,

$$
\omega^* = \max \sigma(B^{-1}K^T KB^{-1}).
$$

Indeed, for the matrix $D = KB^{-1}$ we have $D^T D = B^{-1}K^T KB^{-1}$. The maximal eigenvalue of the matrix $B^{-1}K^T KB^{-1}$ can be expressed by means of a solution to the semidefinite programming problem:

$$
\begin{aligned}
\omega^* &= \max \sigma(B^{-1}K^T KB^{-1}) = \min_{B^{-1}K^T KB^{-1} \preceq \omega I} \omega \\
&= \min_{\omega} \quad \omega \\
&\quad \text{s.t.} \quad \begin{pmatrix} \omega I & B^{-1}K^T \\ KB^{-1} & I \end{pmatrix} \succeq 0.
\end{aligned}
\tag{16}
$$

Since

$$
\begin{aligned}
\Lambda_{HL}(G_C) &= \Lambda_{HL}(\mathcal{B}_K(G_A, G_B)) \\
&= \check{\lambda}^+(G_C) - \hat{\lambda}^-(G_C) = \frac{1}{\lambda_{max}(C^{-1})} + \frac{1}{\lambda_{max}(-C^{-1})},
\end{aligned}
$$

and the optimal value γ^* is an increasing function of ω^* we obtain the following lower bound $\underline{\Lambda}_{HL}^{sir}(G_A, G_B) \leq \Lambda_{HL}^{opt}(G_A, G_B)$ for the optimal HOMO-LUMO spectral gap $\Lambda_{HL}^{opt}(G_A, G_B)$, where

$$
\begin{aligned}
\underline{\Lambda}_{HL}^{sir}(G_A, G_B) &= \frac{2}{\alpha^+(\omega^*+1) + \beta^+ + \sqrt{(\alpha^+(\omega^*+1)+\beta^+)^2 - 4\alpha^+\beta^+}} \\
&+ \frac{2}{\alpha^-(\omega^*+1) + \beta^- + \sqrt{(\alpha^-(\omega^*+1)+\beta^-)^2 - 4\alpha^-\beta^-}},
\end{aligned}
$$

$$
\text{where} \qquad \omega^* = \min_{\omega, K} \ \omega
$$

$$
s.t. \ \begin{pmatrix} \omega I & B^{-1}K^T \\ KB^{-1} & I \end{pmatrix} \succeq 0. \tag{17}
$$

$$
K_{i,j} \in \{0,1\}, \quad \text{for each } i,j, \quad \sum_{k,l} K_{kl} \geq 1.
$$

Similarly, as in the construction of the upper bound, we can relax the condition $K_{i,j} \in \{0,1\}$ by the box constraint

$$
0 \leq K_{i,j} \leq 1, \quad \text{for each } i,j, \tag{18}
$$

to construct the full semidefinite relaxation for the lower bound $\underline{\Lambda}_{HL}^{sdp}(G_A, G_B)$.

Theorem 6.2. *Let G_A and G_B be undirected vertex-labeled invertible graphs on n and m vertices without loops, respectively. Assume G_B is arbitrarily bridgeable over the first k_B vertices $\{1, \cdots, k_B\}$. Then*

$$
\underline{\Lambda}_{HL}^{sdp}(G_A, G_B) \leq \underline{\Lambda}_{HL}^{sir}(G_A, G_B) \leq \Lambda_{HL}^{opt}(G_A, G_B).
$$

7. **Additional constraints imposed on the bridging bipartite graph.** In practical applications one may impose additional constraints on the bridging bipartite graph G_K. For example, in computational chemistry the so-called chemical molecules play important role. The structural graph G of a chemical molecule has all vertices of the degree less or equal to 3. If the goal is to construct a bridged graph $G_C = \mathcal{B}_K(G_A, G_B)$ representing a chemical molecule with the maximal degree M_d, we can add additional constraint:

$$
\sum_k C_{ik} \leq M_d, \quad \text{for all } i, \quad \text{where} \quad C = \begin{pmatrix} A & K \\ K^T & B \end{pmatrix}. \tag{19}
$$

The inequality (19) is linear in the K variable and it can be easily added to all nonlinear optimization problems (11), (12), (13), (17), (18). The computational results of construction of graphs with the maximal degree $M_d = 3$ are presented in the next section.

Another useful constraint imposed on the bridging graph G_K is the min-max box constraints:

$$
\underline{L}_i^A \leq \sum_k K_{ik} \leq \overline{L}_i^A, \quad \text{for all } i = 1, \cdots, n, \tag{20}
$$

$$\underline{L}_j^B \leq \sum_k K_{kj} \leq \overline{L}_j^B, \quad \text{for all } j = 1, \cdots, k_B, \tag{21}$$

representing the box constraints for minimal and maximal number of edges in the bridging graph G_K pointing from the graph G_A to G_B. Again, such a box constraint can be easily added to (11), (12), (13), (17), (18).
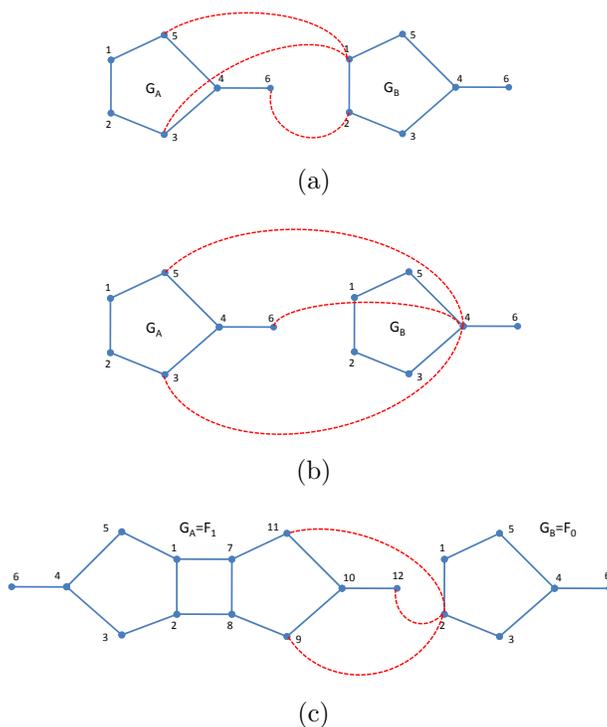

(a)

(b)

(c)

FIGURE 4. Results of optimal bridging of the fulvene graph $G_B = F_0$ through the vertices $\{1, 2\}$ to $G_A = F_0$ a); through the vertices $\{1, 4\}$ to $G_A = F_0$ b); and through the vertices $\{1, 2\}$ to $G_A = F_1$ c).

8. **Computational results.** In this section we present computational results. In Table 2 we present results of construction of the optimal bridging by a bipartite graph for various sets of bridged graphs G_A and G_B. First, we chose the fulvene graph F_0 as the graph G_B and set $k_B = 2$. The graph $G_B \equiv F_0$ is arbitrarily bridgeable through the pairs vertices $\{1, 2\}, \{1, 3\}, \{1, 4\}$ (cf. [23]). We show the results of the optimal value $\Lambda_{HL}^{opt} = \overline{\Lambda}_{HL}^{sir}$ for target graphs $G_A = F_0$ and $G_A = F_1$ (see Figure 4). We also presented upper and lower bounds obtained by means of the full semidefinite relaxation. Among the tested examples the maximal HOMO-LUMO gap was attained in the case when $G_B = F_0$ was bridged to $G_A = F_0$ through vertices $\{1, 4\}$. Solving mixed integer semidefinite program (12) is time consuming (see Table 2). On the other hand, we provided upper and lower bounds which had

TABLE 2. The computational results and comparison of various semidefinite relaxations. The first two columns describe the graph G_A and G_B with the chosen set of bridging vertices. The optimal value $\Lambda_{HL}^{opt} = \overline{\Lambda}_{HL}^{sir}$ is shown in bold in the middle column. The upper $\overline{\Lambda}_{HL}^{sdp}$ and lower bounds $\underline{\Lambda}_{HL}^{sdp}$, $\underline{\Lambda}_{HL}^{sir}$ are also presented together with computational times in seconds computed on Quad core Intel 1.5GHz CPU with 4 GB of memory.

G_A	G_B	$\underline{\Lambda}_{HL}^{sdp}$	$\underline{\Lambda}_{HL}^{sir}$	$\Lambda_{HL}^{opt} = \overline{\Lambda}_{HL}^{sir}$	$\overline{\Lambda}_{HL}^{sdp}$	bridging $G_B \mapsto G_A$
F_0	F_0	0.233688	0.531664	**0.74947**	0.87214	$1 \mapsto 3,5;\ \ 2 \mapsto 6$
	$(1,2)$	$(0.27s)$	$(3.38s)$	$(83s)$	$(2.2s)$	
F_0	F_0	0.333126	0.72678	**0.85828**	0.87214	$1 \mapsto \emptyset;\ \ 4 \mapsto 3,5,6$
	$(1,4)$	$(0.31s)$	$(4.75s)$	$(36s)$	$(2.2s)$	
F_0	F_0	0.333126	0.719668	**0.81389**	0.87214	$1 \mapsto 4;\ \ 3 \mapsto 4$
	$(1,3)$	$(0.31s)$	$(4.27s)$	$(75s)$	$(2.2s)$	
F_1	F_0	0.163626	0.450022	**0.56655**	0.56666	$1 \mapsto \emptyset;\ \ 2 \mapsto 9,11,12$
	$(1,2)$	$(0.28s)$	$(7.65s)$	$(12470s)$	$(2.2s)$	
P_4	P_4	0.472136	0.86953	**1.06418**	1.23607	$2 \mapsto 2,4;\ \ 3 \mapsto 1,3$
	$(2,3)$	$(0.27s)$	$(2.18s)$	$(12.6s)$	$(2.2s)$	
P_6	P_4	0.367365	0.811369	**0.87366**	0.89008	$1 \mapsto 4,6;\ \ 3 \mapsto 4,6$
	$(1,3)$	$(0.26s)$	$(4.6s)$	$(59s)$	$(2.1s)$	
P_6	P_4	0.367365	0.737641	**0.87321**	0.89008	$2 \mapsto 4,6;\ \ 3 \mapsto 1,3$
	$(2,3)$	$(0.26s)$	$(3.41s)$	$(57s)$	$(2.1s)$	
P_{10}	P_4	0.252282	0.523808	**0.56837**	0.56926	$2 \mapsto 8,10;\ \ 3 \mapsto \emptyset$
	$(2,3)$	$(0.26s)$	$(6.32s)$	$(4109s)$	$(2.6s)$	
T_4	P_4	0.38832	0.73094	**0.93258**	0.95452	$2 \mapsto 3,8$
	(2)	$(0.31s)$	$(1.57s)$	$(12s)$	$(2.31s)$	

been obtained efficiently by means of the full semidefinite relaxation technique. A graphical presentation of optimal bridging of fulvene graphs can be seen in Figure 4.
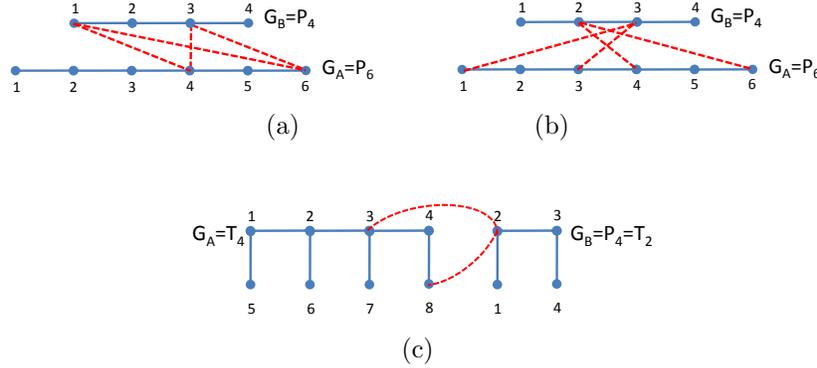

FIGURE 5. Results of optimal bridging of the graph $G_B = P_4$ through the vertices $\{1,3\}$ to $G_A = P_6$ a); through the vertices $\{2,3\}$ to $G_A = P_6$ b); and through the vertex $\{2\}$ to $G_A = T_4$ c).

The next set of examples consists of bridging a simple path $G_B = P_m, m = 4$ to the path $G_A = P_n, n = 4, 6$. An illustration of optimal bridging of P_4 to P_6 over various pairs of vertices is shown in Figure 5.

The last example is the optimal bridging of $G_B = P_4$ to the graph $G_A = T_{2k}$, where T_{2k} is the graph consisting of the simple path P_k with attached pendant vertices to each vertex of P_4. In this case solving the optimal bridging problem yields the bridged graph G_C containing a circle C_4 (see Figure 5, c)).

TABLE 3. The computational results and comparison of various relaxations. The chosen graphs and description of columns is the same as in Table 2. In this table we present results of optimization when additional constraint of the maximal degree 3 has been imposed.

G_A	G_B	$\underline{\Lambda}_{HL}^{sdp}$	$\underline{\Lambda}_{HL}^{sir}$	$\Lambda_{HL}^{opt} = \overline{\Lambda}_{HL}^{sir}$	$\overline{\Lambda}_{HL}^{sdp}$	bridging $G_B \mapsto G_A$
F_0	F_0 $(1,2)$	0.233688 $(0.31s)$	0.507678 $(2.73s)$	**0.720830** $(7.1s)$	0.87214 $(2.9s)$	$1 \mapsto \emptyset;\ 2 \mapsto 6$
F_0	F_0 $(1,4)$	0.233688 $(0.31s)$	0.468053 $(1.1s)$	**0.720830** $(2.33s)$	0.87214 $(2.85s)$	$1 \mapsto 6; 4 \mapsto \emptyset$
F_0	F_0 $(1,3)$	0.333126 $(0.35s)$	0.706635 $(2.45s)$	**0.776875** $(8.4s)$	0.87214 $(2.82s)$	$1 \mapsto 6; 3 \mapsto 6$
F_1	F_0 $(1,2)$	0.163626 $(0.38s)$	0.389941 $(3.67s)$	**0.493727** $(13.4s)$	0.566658 $(2.83s)$	$1 \mapsto 6; 2 \mapsto \emptyset$
P_4	P_4 $(2,3)$	0.472136 $(0.31s)$	0.869530 $(1.86s)$	**0.954520** $(7.8s)$	1.23607 $(2.86s)$	$3 \mapsto \emptyset; 2 \mapsto 2$
P_6	P_4 $(1,3)$	0.367365 $(0.36s)$	0.811369 $(3.35s)$	**0.828427** $(22.9s)$	0.89008 $(2.83s)$	$1 \mapsto 4, 6; 3 \mapsto 2$
P_6	P_4 $(2,3)$	0.367365 (0.33)	0.737641 $(2.73s)$	**0.820751** $(9.21s)$	0.89008 $(2.87s)$	$2 \mapsto 5; 3 \mapsto 2$
P_{10}	P_4 $(2,3)$	0.252282 $(0.33s)$	0.523808 $(4.78s)$	**0.559046** $(13.87s)$	0.56926 $(2.86s)$	$2 \mapsto \emptyset; 3 \mapsto 11$
T_4	P_4 (2)	0.38832 $(0.31s)$	0.692266 $(0.88s)$	**0.890084** $(1.5s)$	0.95452 $(2.11s)$	$2 \mapsto 4$

In Section 7 we discussed additional constraints imposed on the bridging graph G_K. In what follows, we present results of computing the optimal HOMO-LUMO gap and its upper and lower bound under the constraint that the resulting graph G_C represents a chemical molecule with the maximal vertex degree $M_d = 3$. The results are summarized in Table 3 and illustrative examples are shown in Figure 6. In Figure 6, c), we confirmed the well known fact that the comb graph T_{2k} has the maximal HOMO-LUMO gap among all trees on $2k$ vertices with perfect matchings. It was first proved by Krč and Pavlíková [21, Theorem 7] (see also Zhang and An [28]). Interestingly enough, adding additional constraint on maximal degree of vertices considerably reduced computational time for solving the mixed integer semidefinite problem (12).
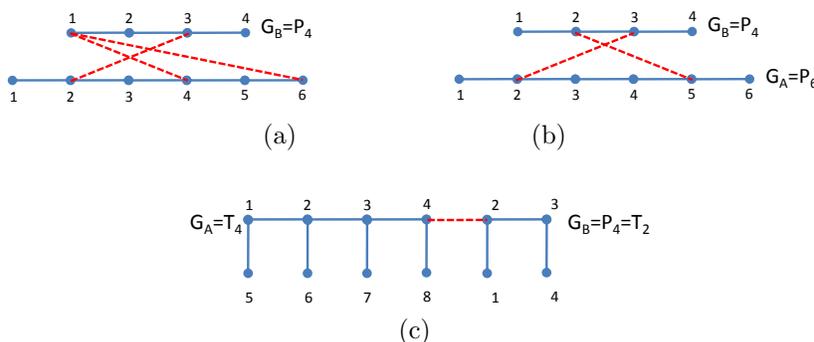

FIGURE 6. Results of optimal bridging of the graph $G_B = P_4$ through the vertices $\{1,3\}$ to $G_A = P_6$ a); through the vertices $\{2,3\}$ to $G_A = P_6$ b); and through the vertex $\{2\}$ to $G_A = T_4$ c); with the constraint of maximal degree equal to 3.

Conclusions. We analyzed spectral properties of graphs which are constructed from two given invertible graphs by bridging them over a bipartite graph. We showed how the HOMO-LUMO spectral gap can be computed by means of a solution to mixed integer semidefinite programming problem. We investigated the optimization problem in which we constructed a bridging graph maximizing the HOMO-LUMO spectral gap. We also provided upper and lower bounds to the optimal value, again expressed as solution to relaxed semidefinite programming problems. Various computational examples were presented in this paper.

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