

Eigenvalues of Two-Parameter Complex Hadamard Matrices of Order Six

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Abstract: Characterizing complex Hadamard matrices (CHMs) of order six is an open problem in both algebra and quantum information theory. Analyzing the eigenvalues of CHMs provides a novel approach to this problem. We construct a class of two-parameter CHMs of order six with two sorts of eigenvalues. All matrices in this class are H_2 -reducible, and the Diţă matrix, Haagerup matrices and Hermitian matrices are both included. This class turns out to be complex equivalent to the class of all CHMs in the dephased form whose eigenvalues are of the form $\sqrt{6}, -\sqrt{6}, \lambda_1, \lambda_1, \lambda_2, \lambda_2$ with λ_j of modulus $\sqrt{6}$ excluding Tao matrix $S_6^{(0)}$. We further show that some CHMs including all Hermitian CHMs in the former class have Schmidt rank at most three up to complex equivalence, and they are actually controlled unitary gates implementable in experiments. Our result demonstrates the feasibility of using eigenvalues for characterizing order-six CHMs and their application in quantum circuits.

Keywords: complex hadamard matrices; eigenvalues and eigenvectors; controlled unitary gates

1. Introduction

Characterizing complex Hadamard matrices of order n is a fundamental problem in quantum physics, as it is a way to find the complete mutually unbiased bases (MUBs). In general, k MUBs in Hilbert space $\mathbb{C}^n$ are k orthogonal bases, the inner product of any two vectors from different bases has a modulus of $1/\sqrt{n}$. When there are $n+1$ MUBs in $\mathbb{C}^n$, then they are referred to as complete MUBs. So far, it has been proven that complete MUBs exist in $\mathbb{C}^n$ when n is a prime power [1]. The complete classification of CHMs gives us a new way to find the maximal MUBs in the unsolved dimensions. In particular, the dimension six is the smallest one. Paper [2] investigated that the average distance between four bases in dimension six, providing a strong evidence against the existence of four MUBs in $\mathbb{C}^6$. Paper [3] showed that if complete MUBs in dimension six exist, it cannot include more than one product basis. The examination of the number of product vectors in a set of four MUBs in dimension six showed that each of the remaining three MUBs contains at most two product vectors [4]. The existence and limitations of MUB product bases in dimension six were explored in [5–7]. The skills such as Schmidt ranks, Lie algebra, semidefinite programming, entanglement theory and so on are used in [8–14].

The complete classification of 6×6 CHMs is also unsolved. Karlsson presented a three-parameter CHMs family in $\mathbb{C}^6$ [15], and named the family “the H_2 -reducible matrices”. As far as we know, most known 6×6 CHMs belong to the H_2 -reducible matrices such as the Haagerup matrix [16], but the Tao matrix is an exception [17]. The paper [18] presented a four-parameter 6×6 CHM family, though the analytic form of these matrices is still unknown. Another idea is analyzing the eigenvalues of the CHM. It was shown that CHMs of order six with dephased form have eigenvalues $\sqrt{6}, -\sqrt{6}$, and for the remaining four eigenvalues, at most two of them are the same [19]. More facts investigating the classification problem have been presented in [20–25].

CHMs in dimension six are also bipartite unitary matrices from the point of view in quantum information theory. Such matrices can be applied to create quantum entanglement and to implement quantum circuits and cryptography [26]. The so-called controlled-unitary (say controlled-NOT) gate, which is a fundamental bipartite unitary operator, serves as both the primary entanglement resource and a critical component of universal quantum computation [27]. Finding the decompositions by controlled-unitary gates of bipartite unitary operators generated by CHMs will provide a simple physical realization of complex operations. Schmidt rank of bipartite unitary matrices was shown to be closely connected to the existence of this decomposition [28,29].

In this paper, we investigate CHMs with certain type of eigenvalues. In Theorem 1 we prove that CHMs with only two sorts of eigenvalues must be complex equivalent to a family we will refer to as $K(d, x)$ in (8) with two parameters d, x . Lemma 1 gives the solutions of parameters in Theorem 1, and gives the suitable value range of the parameters. The main technical treatment in the proof of Theorem 1 is the construction in (A4), which exhausts the form of CHMs with only two sorts of eigenvalues. Next we analyze the CHMs in dephased form whose eigenvalues are $\sqrt{6}, -\sqrt{6}, \lambda_1, \lambda_1, \lambda_2, \lambda_2$. We give a complete classification of these CHMs, containing the Tao matrix in (3) and a family $K'(d, x)$ in (21) with parameters d, x . That is the result in Theorem 2. The key equation in the proof of Theorem 2 is shown in (A35), which gives the main restriction of this class of CHMs. In Corollary 1 we present the outcome of a special case of Theorem 2, we claim that if $\lambda_2 = -\lambda_1$, then such CHM must be complex equivalent to the Hermitian matrices. In Theorem 3 we show that a family of $K(d, x)$ has Schmidt rank at most three, and this family can be carried out as controlled unitary gates.

The rest of this paper will be presented as follows. In Section 2 we introduce some facts used in this paper, such as the definition of MUBs and CHMs, the eigenvalues of some famous CHMs. In Section 3, we classify all the CHMs with the specified eigenvalue structure. In Section 4, we find the application of some CHMs in implementable unitary gate. We conclude in Section 5.

2. Preliminaries

In this section, we present the definition of CHMs and MUBs, as well as the eigenvalues of some special CHMs.

Definition 1. Suppose $\mathcal{B}_1 = \{|\phi_{1j}\rangle\}_{j=1,\dots,d}, \dots, \mathcal{B}_n = \{|\phi_{nj}\rangle\}_{j=1,\dots,d}$ are n orthonormal basis in $\mathbb{C}^d$, if

$$|\langle \phi_{jl} | \phi_{km} \rangle| = \frac{1}{\sqrt{d}},$$

for any $j \neq k, j, k = 1, \dots, n; l, m = 1, \dots, d$, then $\mathcal{B}_1, \dots, \mathcal{B}_n$ form n MUBs.

Definition 2. If $H = [h_{jk}]$ is an $n \times n$ complex matrix satisfying

- (i). $|h_{jk}| = 1$ for any $j, k = 1, \dots, n$;
- (ii). $H \cdot H^\dagger = nI$,

then H is an $n \times n$ complex Hadamard matrix (CHM).

We also introduce the definition of complex equivalence. We refer to the monomial unitary matrix as a unitary matrix that every row and column have exactly one nonzero element. Then two matrices U and V are complex equivalent when $U = PVQ$ where P, Q are monomial unitary matrices.

Next we list some famous CHMs and their eigenvalues. The Haagerup matrices [16] are

$$H_6^q = \begin{bmatrix} 1 & 1 & 1 & 1 & 1 & 1 \\ 1 & -1 & i & i & -i & -i \\ 1 & i & -1 & -i & q & -q \\ 1 & i & -i & -1 & -q & q \\ 1 & -i & -\frac{1}{q} & \frac{1}{q} & -1 & i \\ 1 & -i & \frac{1}{q} & -\frac{1}{q} & i & -1 \end{bmatrix}, \quad (1)$$

where $|q| = 1$. The eigenvalues of H_6^q are $\sqrt{6}, -\sqrt{6}, -1 + \sqrt{5}i, -1 + \sqrt{5}i, -1 - \sqrt{5}i, -1 - \sqrt{5}i$.

Second, the Diţă matrix D_0 [30] is equivalent to H_6^q for $q = \pm i$. D_0 has the same eigenvalues as H_6^q .

$$D_0 = \begin{bmatrix} 1 & 1 & 1 & 1 & 1 & 1 \\ 1 & -1 & i & -i & -i & i \\ 1 & i & -1 & i & -i & -i \\ 1 & -i & i & -1 & i & -i \\ 1 & -i & -i & i & -1 & i \\ 1 & i & -i & -i & i & -1 \end{bmatrix}. \quad (2)$$

Third, the Tao matrix is isolated and belongs to the Buston matrices [31,32].

$$S_6^{(0)} = \begin{bmatrix} 1 & 1 & 1 & 1 & 1 & 1 \\ 1 & 1 & \omega & \omega & \omega^2 & \omega^2 \\ 1 & \omega & 1 & \omega^2 & \omega^2 & \omega \\ 1 & \omega & \omega^2 & 1 & \omega & \omega^2 \\ 1 & \omega^2 & \omega^2 & \omega & 1 & \omega \\ 1 & \omega^2 & \omega & \omega^2 & \omega & 1 \end{bmatrix}, \quad (3)$$

where $\omega = e^{\frac{2\pi i}{3}}$. The eigenvalues of $S_6^{(0)}$ are $\sqrt{6}, -\sqrt{6}, \frac{3+\sqrt{15}i}{2}, \frac{3+\sqrt{15}i}{2}, \frac{3-\sqrt{15}i}{2}, \frac{3-\sqrt{15}i}{2}$.

The Hermitian matrices, by exchanging rows and columns, can take the following form [33].

$$H(t) = \begin{bmatrix} 1 & 1 & 1 & 1 & 1 & 1 \\ 1 & -1 & \frac{1}{x} & -y & -\frac{1}{x} & y \\ 1 & x & -1 & t & -t & -x \\ 1 & -\frac{1}{y} & \frac{1}{t} & -1 & \frac{1}{y} & -\frac{1}{t} \\ 1 & -x & -\frac{1}{t} & y & 1 & \frac{1}{z} \\ 1 & \frac{1}{y} & -\frac{1}{x} & -t & z & 1 \end{bmatrix}, \quad (4)$$

where $\theta \in [-\pi, -\arccos(\frac{-1+\sqrt{3}}{2})] \cup [\arccos(\frac{-1+\sqrt{3}}{2}), \pi]$, and the parameters x, y, t, z are given by

$$y = e^{i\theta}, z = \frac{1+2y-y^2}{y(-1+2y+y^2)}; \quad (5)$$

$$x = \frac{1+2y+y^2-\sqrt{2(1+2y+2y^3+y^4)}}{1+2y-y^2}; \quad (6)$$

$$t = \frac{1+2y+y^2-\sqrt{2(1+2y+2y^3+y^4)}}{-1+2y+y^2}. \quad (7)$$

The eigenvalues of the Hermitian matrix $H(t)$ are $\sqrt{6}, \sqrt{6}, \sqrt{6}, -\sqrt{6}, -\sqrt{6}, -\sqrt{6}$.

3. Result

In this section, we will present our results in this paper. We will discuss CHMs with special type of eigenvalues up to complex equivalence. Though eigenvalues are not invariant with respect to the equivalence relation, we can simplify the result by this relation and at the same time, no results will be omitted. We first consider a family of CHMs with exactly two sorts of eigenvalues. Paper [19] claimed that there is no 6×6 CHM with four identical eigenvalues. So the two distinct eigenvalues must be of algebraic multiplicity three. The following theorem shows the complete classification of such CHMs.

Theorem 1. *The order-six CHM with exactly two sorts of eigenvalues is complex equivalent to the following CHM.*

$$K(d, x) = \begin{bmatrix} d & 1 & 1 & 1 & 1 & 1 \\ 1 & -\bar{d} & x & \bar{y} & -x & -\bar{y} \\ 1 & \bar{x} & -\bar{d} & z & -z & -\bar{x} \\ 1 & y & \bar{z} & -\bar{d} & -y & -\bar{z} \\ 1 & -\bar{x} & -\bar{z} & -\bar{y} & d & -\overline{xyz} \\ 1 & -y & -x & -z & -xyz & d \end{bmatrix}. \quad (8)$$

Here the parameters d, x, y, z are all of modulus one and satisfy

$$-2\operatorname{Re}[d] + x + y + z + xyz = 0. \quad (9)$$

If $d = e^{i\theta}$, then the eigenvalues of $K(d, x)$ are

$$\lambda_1 = -\sqrt{6 - \sin^2 \theta} + i \sin \theta, \quad \lambda_2 = \sqrt{6 - \sin^2 \theta} + i \sin \theta. \quad (10)$$

The proof of Theorem 1 will be present in Appendix A. The restriction in (9) can be solved by the following lemma. Note that if $|d| = 1$, then $-2\operatorname{Re}[d] \in [-2, 2]$.

Lemma 1. For a real number $r \in [-2, 2]$, suppose the complex numbers x, y, z of modulus one satisfy the following equation,

$$r + x + y + z + xyz = 0. \quad (11)$$

We denote $x = e^{i\phi_1}, y = e^{i\phi_2}, z = e^{i\phi_3}$, then x, y, z can be solved as follows.

(i) If $r = 0$, then $\{x, y\} = \{1, -1\}$ or $\{y, z\} = \{1, -1\}$ or $\{x, z\} = \{1, -1\}$.

(ii) If $\phi_1 = -\phi_2$, then $\cos \phi_1 = \frac{-r-2}{2}, \phi_3 = 0$ or $\cos \phi_1 = \frac{-r+2}{2}, \phi_3 = \pm\pi$.

(iii) If $\phi_1 - \phi_2 = \pm\pi$, i.e. $x = -y$, then $z + \bar{z} = -r, \{x, y\} = \{i\bar{z}, -i\bar{z}\}$.

(iv) For other cases, a, b, c can be solved by

$$y = \frac{rx^2 + r^2x + r \pm \sqrt{\Delta}}{2(x^2 - rx - 1)}, \quad (12)$$

$$c = -\frac{r + x + y}{1 + xy}, \quad (13)$$

where

$$\Delta = (r^2 + 4)x^4 + 2r^3x^3 + (r^4 - 2r^2 - 8)x^2 + 2r^3x + r^2 + 4, \quad (14)$$

and the range of ϕ_1 is

$$\frac{-r^3 - 4\sqrt{2}\sqrt{r^2 + 2}}{2(r^2 + 4)} \leq \cos \phi_1 \leq \frac{-r^3 + 4\sqrt{2}\sqrt{r^2 + 2}}{2(r^2 + 4)}. \quad (15)$$

□

The proof of Lemma 1 will be presented in Appendix B. Noting that the solutions of (9) can be expressed by parameter ϕ_1 below (11) or x . So the notation $K(d, x)$ means a class of two-parameter CHMs, here d controls the eigenvalues, d and x control the solutions of restriction (9).

Recall that the CHM of order-six with dephased form has eigenvalues $\sqrt{6}, -\sqrt{6}$ [19]. If the remaining four eigenvalues have at most two distinct elements, then [19] claims that the remaining four eigenvalues are $\lambda_3, \lambda_3, \lambda_4, \lambda_4$ with $\lambda_3 \neq \lambda_4$. Next we present another theorem to classify such CHMs.

Theorem 2. Suppose that an order-six CHM H with dephased form has eigenvalues $\sqrt{6}, -\sqrt{6}, \lambda_3, \lambda_3, \lambda_4, \lambda_4$. Then H is complex equivalent to the Tao matrix $S_6^{(0)}$ in (3) or the following two-parameter H_2 -reducible CHM

$$K'(d, x) = \begin{bmatrix} 1 & 1 & 1 & 1 & 1 & 1 \\ 1 & -1 & xd & \bar{y}d & -xd & -\bar{y}d \\ 1 & \bar{x}d & -1 & zd & -zd & -\bar{x}d \\ 1 & yd & \bar{z}d & -1 & -yd & -\bar{z}d \\ 1 & -\bar{x}d & -\bar{z}d & -\bar{y}d & d^2 & -\bar{x}y\bar{z}d \\ 1 & -yd & -xd & -zd & -xyzd & d^2 \end{bmatrix}, \quad (16)$$

where

$$\lambda_3 = d\lambda_1, \quad \lambda_4 = d\lambda_2 \quad (17)$$

with d, λ_1, λ_2 in (10).

In particular, if $d = \pm i$, then $K'(\pm i, x)$ is complex equivalent to Haagerup matrices in (1). If $d = \pm 1$, then $K'(\pm 1, x)$ in (16) is complex equivalent to Hermitian matrices in (4). □

We present the brief version of proof of Theorem 2 below. More details can be found in Appendix C. The proof shows the connection between Theorems 1 and 2. In fact, $K(d, x)$ and $K'(d, x)$ are equivalent by the equality

$$K'(d, x) = d \cdot P \cdot K(d, x) \cdot P \quad (18)$$

where

$$P = \begin{bmatrix} \bar{d} & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \end{bmatrix}. \quad (19)$$

Proof. For a given H in dephased form with eigenvalues $\sqrt{6}, -\sqrt{6}, \lambda_3, \lambda_3, \lambda_4, \lambda_4$, we choose

$$|u_1\rangle = \frac{1}{\sqrt{12+2\sqrt{6}}} \begin{bmatrix} 1+\sqrt{6} \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \end{bmatrix}, \quad |u_2\rangle = \frac{1}{\sqrt{12-2\sqrt{6}}} \begin{bmatrix} 1-\sqrt{6} \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \end{bmatrix}. \quad (20)$$

as the eigenvectors of $\sqrt{6}, -\sqrt{6}$, respectively. We have

$$H = H_2 + \lambda_3 \sum_{j=3}^4 |u_j\rangle\langle u_j| + \lambda_4 \sum_{j=5}^6 |u_j\rangle\langle u_j|. \quad (21)$$

Here

$$H_2 = \sqrt{6}|u_1\rangle\langle u_1| - \sqrt{6}|u_2\rangle\langle u_2| = \frac{1}{5} \begin{bmatrix} 5 & 5 & 5 & 5 & 5 & 5 \\ 5 & -1 & -1 & -1 & -1 & -1 \\ 5 & -1 & -1 & -1 & -1 & -1 \\ 5 & -1 & -1 & -1 & -1 & -1 \\ 5 & -1 & -1 & -1 & -1 & -1 \\ 5 & -1 & -1 & -1 & -1 & -1 \end{bmatrix}. \quad (22)$$

The equation (21) can be rewritten as follows.

$$H = \sqrt{6}|u_1\rangle\langle u_1| - \sqrt{6}|u_2\rangle\langle u_2| + \lambda_3 \sum_{i=3}^4 |u_i\rangle\langle u_i| + \lambda_4 \sum_{j=5}^6 |u_j\rangle\langle u_j| \quad (23)$$

$$= \sqrt{6}|u_1\rangle\langle u_1| - \sqrt{6}|u_2\rangle\langle u_2| + \lambda_3(I - |u_1\rangle\langle u_1| - |u_2\rangle\langle u_2|) + (\lambda_4 - \lambda_3) \sum_{j=5}^6 |u_j\rangle\langle u_j| \quad (24)$$

$$= \frac{1}{5} \begin{bmatrix} 5 & 5 & 5 & 5 & 5 & 5 \\ 5 & -1+4\lambda_3 & -1-\lambda_3 & -1-\lambda_3 & -1-\lambda_3 & -1-\lambda_3 \\ 5 & -1-\lambda_3 & -1+4\lambda_3 & -1-\lambda_3 & -1-\lambda_3 & -1-\lambda_3 \\ 5 & -1-\lambda_3 & -1-\lambda_3 & -1+4\lambda_3 & -1-\lambda_3 & -1-\lambda_3 \\ 5 & -1-\lambda_3 & -1-\lambda_3 & -1-\lambda_3 & -1+4\lambda_3 & -1-\lambda_3 \\ 5 & -1-\lambda_3 & -1-\lambda_3 & -1-\lambda_3 & -1-\lambda_3 & -1+4\lambda_3 \end{bmatrix} + (\lambda_4 - \lambda_3) \sum_{j=5}^6 |u_j\rangle\langle u_j|. \quad (25)$$

Let the vectors $|u_j\rangle = [g_1^j, \dots, g_6^j]^T, j = 5, 6$, and we can deduce the expressions of all elements in H . The details will be present in Appendix C.

By discussing three distinct cases, we obtain the relations of λ_3, λ_4 . Further construction shows that for given CHM in dephased form with eigenvalues $\sqrt{6}, -\sqrt{6}, \lambda_3, \lambda_3, \lambda_4, \lambda_4$, we will deduce the Tao matrix in (3) or a CHM with two sorts of eigenvalues, that is $K(d, x)$. By Theorem 1, we have finished the proof of Theorem 2. The details will be present in Appendix C. $\square$

Corollary 1. Suppose H is an order-six CHM in dephased form with the first two eigenvalues $\sqrt{6}$ and $-\sqrt{6}$.

(i) If H has the remaining four eigenvalues $\lambda, \lambda, -\lambda, -\lambda$, then H is the Hermitian matrix in (4).

(ii) If H has the remaining four eigenvalues of at most two distinct elements, then H has been completely studied in [19] and Theorem 2.

Proof. (i) By using Theorem 1, we substitute $\lambda_3 = -\lambda_4$ into (10). We know that $d = \pm 1$, and thus $\{\lambda_3, \lambda_4\} = \{\sqrt{6}, -\sqrt{6}\}$. Hence H is the Hermitian matrix in (4). (ii) If the four remaining eigenvalues satisfy $\lambda_3 = \lambda_4 = \lambda_5$, then by the result of Theorem 5 in paper [19] we deduce the contradiction. If $\lambda_3 = \lambda_4$ and $\lambda_5 = \lambda_6$, then Theorem 2 has done the complete classification of this case. $\square$

4. Application

In this section, we introduce an application of our results. For a bipartite Hilbert space $\mathcal{H}_A \otimes \mathcal{H}_B$, a controlled unitary gate up to local unitary equivalence takes the form

$$U = \sum_{i=1}^{d_A} |i\rangle\langle i|_A \otimes U_B^{(i)}. \quad (26)$$

where $\{|i\rangle_A\}$ is an orthonormal basis for subsystem A and $U_B^{(i)}$ are unitary operators acting on subsystem B . d_A is the dimension of subsystem A .

It is known that every bipartite unitary matrix can be regarded a non-local operation from the viewpoint of quantum information. For example, the so-called controlled-NOT and other types of controlled unitary gates play a key role in quantum computing and so on, as every bipartite unitary gate can be decomposed into the product of some controlled unitary gates [34].

Further, it has been proven that a bipartite unitary gate of Schmidt rank at most three is actually a controlled unitary gate [28], which is more easily implementable in experiments. Here, for a bipartite unitary matrix $U \in \mathcal{H}_A \otimes \mathcal{H}_B$, the Schmidt rank is the smallest integer r such that

$$U = \sum_{i=1}^r \lambda_i M_i^{(A)} \otimes N_i^{(B)} \quad (27)$$

with linearly independent sets $\{M_i\}$ and $\{N_i\}$ and all $\lambda_i > 0$.

In the following, we show that a family of CHMs $K(d, x)$ are complex equivalent to some CHMs of Schmidt rank three. In particular, all order-six Hermitian CHMs are included in this family. Hence, such CHMs can be actually carried out as a controlled unitary gate, and they can be applied in experiments more easily. Our result help realize more non-local operations in quantum system with less coherence time and lower error rates.

Theorem 3. For the CHM $K(d, x)$ in (8), if $x = d$ or $x = \bar{d}$ and $\{y, z\} = \{i\bar{x}, -i\bar{x}\}$, then $K(d, x)$ is complex equivalent to a CHM whose Schmidt rank is at most three.

Proof. We consider the monomial unitary matrix

$$Q = \begin{bmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -\bar{y} \\ 0 & 0 & 0 & 0 & -z & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \end{bmatrix}, \quad (28)$$

then we have

$$M = Q^\dagger \cdot K(d, x) \cdot Q = \begin{bmatrix} d & 1 & 1 & 1 & -z & -\bar{y} \\ 1 & d & -\overline{xyz} & -\bar{y} & 1 & \overline{xy} \\ 1 & -xyz & d & -z & xz & 1 \\ 1 & -y & -\bar{z} & -\bar{d} & -1 & -1 \\ -\bar{z} & 1 & \overline{xz} & -1 & -\bar{d} & \overline{xyz} \\ -y & xy & 1 & -1 & xyz & -\bar{d} \end{bmatrix}. \quad (29)$$

We denote $M = \begin{bmatrix} A & B \\ C & D \end{bmatrix}$ and A, B, C, D are four 3×3 submatrices of M . Then we have

$$A + D = (d - \bar{d})I_3, \quad (30)$$

and

$$B + C = \begin{bmatrix} 2 & -y - z & -\bar{y} - \bar{z} \\ -\bar{y} - \bar{z} & 2 & \bar{x}(y + z) \\ -y - z & x(y + z) & 2 \end{bmatrix}. \quad (31)$$

If $y = -z$, then $B + C = 2I_3$, and $A + D, B + C$ are linear dependent. It means that the CHM M has Schmidt rank at most three. Using the Lemma 1 we know that if $y = -z$, then $x = d$ or $x = \bar{d}$ and $\{y, z\} = \{i\bar{x}, -i\bar{x}\}$. Hence we have finished the proof. $\square$

For example, if $d = 1$, then $x = 1$ and $K(d, x)$ is indeed Hermitian. We shall take $y = i, z = -i$, then M in (29) turns to be

$$\begin{bmatrix} 1 & 1 & 1 & 1 & i & i \\ 1 & 1 & -1 & i & 1 & -i \\ 1 & -1 & 1 & i & -i & 1 \\ 1 & -i & -i & -1 & -1 & -1 \\ -i & 1 & i & -1 & -1 & 1 \\ -i & i & 1 & -1 & 1 & -1 \end{bmatrix} \quad (32)$$

and we can rewrite M as follows:

$$M = \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \otimes \begin{bmatrix} 1 & 1 & 1 \\ 1 & 1 & -1 \\ 1 & -1 & 1 \end{bmatrix} + \begin{bmatrix} 0 & 1 \\ 0 & 0 \end{bmatrix} \otimes \begin{bmatrix} 1 & i & i \\ i & 1 & -i \\ i & -i & 1 \end{bmatrix} + \begin{bmatrix} 0 & 0 \\ 1 & 0 \end{bmatrix} \otimes \begin{bmatrix} 1 & -i & -i \\ -i & 1 & i \\ -i & i & 1 \end{bmatrix} \quad (33)$$

We take

$$Q_1 = I_2 \otimes \frac{1}{\sqrt{6}} \begin{bmatrix} \sqrt{2} & \sqrt{3} & 1 \\ -\sqrt{2} & \sqrt{3} & -1 \\ -\sqrt{2} & 0 & 2 \end{bmatrix}. \quad (34)$$

Then

$$Q_1^\dagger M Q_1 = \begin{bmatrix} -1 & 1-2i \\ 1+2i & 1 \end{bmatrix} \otimes |0\rangle\langle 0| + \begin{bmatrix} 2 & 1+i \\ 1-i & -2 \end{bmatrix} \otimes (|1\rangle\langle 1| + |2\rangle\langle 2|). \quad (35)$$

Where $|0\rangle = [1, 0, 0]^T$, $|1\rangle = [0, 1, 0]^T$ and $|2\rangle = [0, 0, 1]^T$. So we know that up to local unitary equivalence, M is a controlled unitary gate. Thus it can be applied in experiments more easily.

5. Conclusion

In this paper, we have presented the complete classification of CHMs with two sorts of eigenvalues λ_1, λ_2 , these CHMs are included in a two-parameter family $K(d, x)$ in (8). Moreover, we have classified all CHMs with dephased

form whose eigenvalues are exactly $\sqrt{6}, -\sqrt{6}, \lambda_1, \lambda_1, \lambda_2, \lambda_2$, this class consists of $K'(d, x)$ in (21) and Tao matrix in (3), here $K'(d, x)$ is complex equivalent to $K(d, x)$ for the same d, x . One interesting thing is that some known CHMs such as Diţă matrix, Haagerup matrices and Hermitian matrices, are included in the class $K'(d, x)$ and all CHMs in this class are H_2 -reducible. We have shown that the Schmidt rank of some CHMs including all Hermitian CHMs in $K(d, x)$ is at most three up to complex equivalence. Our future research will focus on the Schmidt rank of every CHM in $K(d, x)$ and more CHMs with three or more sorts of eigenvalues to find more connections between the classification of CHMs and their eigenvalues.

Author Contributions

Authors equally contribute to the paper. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest

No conflict.

Data Availability Statement

No data.

Appendix A. The proof of Theorem 1

Proof. We consider the spectral decomposition of a CHM H whose eigenvalues are $\lambda_1, \lambda_1, \lambda_1, \lambda_2, \lambda_2, \lambda_2$, that is, $H = \lambda_1 H_1 + \lambda_2 H_2$, with projection matrices H_1, H_2 . It implies that $H_j \cdot H_k^\dagger = 0$ for $j \neq k$ and $H_j \cdot H_j^\dagger = H_j$ and $H_j^\dagger = H_j$ for $j = 1, 2$. We have $H_1 = \sum_{j=1}^3 |u_j\rangle\langle u_j|$ and $H_2 = \sum_{j=4}^6 |u_j\rangle\langle u_j|$, such that $H_1 + H_2 = I_6$. If we consider the matrix $M = \frac{\sqrt{6}}{\lambda_1} H$, then $M = \sqrt{6} H_1 + \lambda H_2$ with $\lambda = \frac{\sqrt{6}\lambda_2}{\lambda_1}$. So we obtain that $M = \sqrt{6} I_6 + (\lambda - \sqrt{6}) H_2$. The point of our proof is to design the projection matrix H_2 such that M is indeed a CHM for a given λ . Since all the entries of M have modulus one, we consider the diagonal elements and non-diagonal elements of M respectively. If we denote $M = [h_{jk}]$ and $H_2 = [g_{jk}]$, then the diagonal elements are $h_{jj} = \sqrt{6} + (\lambda - \sqrt{6})g_{jj}$ and the non-diagonal elements can be expressed by $h_{jk} = (\lambda - \sqrt{6})g_{jk}$ for $j \neq k$. So we have

$$|\sqrt{6} + (\lambda - \sqrt{6})g_{jj}| = 1, \quad |(\lambda - \sqrt{6})g_{jk}| = 1, \quad j, k \in \{1, \dots, 6\}; j \neq k. \quad (\text{A1})$$

Recall that $H_2 = \sum_{j=4}^6 |u_j\rangle\langle u_j|$, we see that g_{jj} is real for all $j \in \{1, \dots, 6\}$. The first equation shows that

$$[\sqrt{6} + (\lambda - \sqrt{6})g_{jj}] \cdot [\sqrt{6} + (\bar{\lambda} - \sqrt{6})g_{jj}] = 1, \quad (\text{A2})$$

which means $(12 - \sqrt{6}(\lambda + \bar{\lambda}))g_{jj}^2 - (12 - \sqrt{6}(\lambda + \bar{\lambda}))g_{jj} + 5 = 0$. If we denote $m = \frac{\text{Re}[\lambda]}{\sqrt{6}}$, then the solution will be expressed by $g_{jj} = \frac{1}{2} \pm \sqrt{\frac{3m+2}{12(m-1)}}$. The second equation in (A1) shows that $|g_{jk}| = \frac{1}{|\lambda - \sqrt{6}|} = \frac{1}{\sqrt{12-12m}}$. The range of m is in $[-1, 1]$, and the above g_{jj} and $|g_{jk}|$ are both real. So $\frac{3m+2}{12(m-1)} \geq 0$ and $12 - 12m > 0$, hence the range of m should be restricted in $[-1, -\frac{2}{3}]$.

We then rewrite the projection matrix H_2 by $H_2 = \frac{1}{2} I_6 + kM(a)$, and once we take $k = \frac{1}{\sqrt{12-12m}}$, then the non-diagonal elements of $M(a)$ have modulus one. The diagonal elements have modulus $\sqrt{-3m-2}$. We denote $a = \sqrt{-3m-2}$, then $k = \frac{1}{\sqrt{20+4a^2}}$ and we have already constructed the following form of Hermitian matrices $M(a)$, here a is real, $0 \leq a \leq 1$. For $M(a) = [f_{jk}]_{6 \times 6}$, the entries of $M(a)$ satisfies the following conditions: For $j \neq k$, we have $|f_{jk}| = 1, f_{jk} = \bar{f}_{kj}$, and $f_{jj} = \pm a$. Since the trace of M is $3(\sqrt{6} + \lambda)$ and the trace of M can also be $6\sqrt{6} + 6 \times \frac{1}{2}(\lambda - \sqrt{6}) + \sum_{j=1}^6 g_{jj} = 3(\sqrt{6} + \lambda) + \sum_{j=1}^6 g_{jj}$, we must have $\sum_{j=1}^6 g_{jj} = 0$, so $\sum_{j=1}^6 f_{jj} = 0$. We shall assume $f_{11} = f_{55} = f_{66} = a$ and $f_{22} = f_{33} = f_{44} = -a$, then $M(a)$ takes the following form.

$$\begin{bmatrix} a & 1 & 1 & 1 & 1 & 1 \\ 1 & -a & & & & \\ 1 & & -a & & & \\ 1 & & & -a & & \\ 1 & & & & a & \\ 1 & & & & & a \end{bmatrix} \quad (\text{A3})$$

Now we recall that M is a CHM, then $M \cdot M^\dagger = 6I_6$, and

$$M = \sqrt{6}I_6 + (\lambda - \sqrt{6})\left(\frac{1}{2}I_6 + \frac{1}{\sqrt{20+4a^2}}M(a)\right), \quad (\text{A4})$$

it means that $M(a) \cdot M(a)^\dagger = M(a)^2 = (5+a^2)I_6$.

We analyze the orthogonality of row 1 and rows 2,3,4 of $M(a)$ in (A3), and we assume the entries of $M(a)$ to be

$$M(a) = \begin{bmatrix} a & 1 & 1 & 1 & 1 & 1 \\ 1 & -a & h_{23} & h_{24} & h_{25} & h_{26} \\ 1 & \bar{h}_{23} & -a & h_{34} & h_{35} & h_{36} \\ 1 & \bar{h}_{24} & \bar{h}_{34} & -a & h_{45} & h_{46} \\ 1 & \bar{h}_{25} & \bar{h}_{35} & \bar{h}_{45} & a & h_{56} \\ 1 & \bar{h}_{26} & \bar{h}_{36} & \bar{h}_{46} & \bar{h}_{56} & a \end{bmatrix} \quad (\text{A5})$$

The orthogonality of rows 1,2 shows that $h_{23} + h_{24} + h_{25} + h_{26} = 0$. By permuting rows and columns, we divide into two cases namely Case A, B, that is $h_{23} = -h_{24}$ or $h_{23} = -h_{25}$.

Case A. $h_{23} = -h_{24}$, we denote $x_1 = h_{23}$, $x_2 = h_{25}$, then $M(a)$ become

$$\begin{bmatrix} a & 1 & 1 & 1 & 1 & 1 \\ 1 & -a & x_1 & -x_1 & x_2 & -x_2 \\ 1 & \bar{x}_1 & -a & h_{34} & h_{35} & h_{36} \\ 1 & -\bar{x}_1 & \bar{h}_{34} & -a & h_{45} & h_{46} \\ 1 & \bar{x}_2 & -x_1 & \bar{h}_{45} & a & h_{56} \\ 1 & -\bar{x}_2 & -\bar{h}_{34} & \bar{h}_{46} & \bar{h}_{56} & a \end{bmatrix} \quad (\text{A6})$$

Considering the orthogonality of rows 1,3, we have $\bar{x}_1 + h_{34} + h_{35} + h_{36} = 0$, then we discuss the following two cases by permuting columns 5,6, namely Case A.1 and A.2, that is $h_{34} = -\bar{x}_1$ or $h_{35} = -\bar{x}_1$.

Case A.1. $h_{34} = -\bar{x}_1$, then $h_{36} = -h_{35}$ and the orthogonality of rows 3,4 shows that $a(x_1 + \bar{x}_1) + h_{35}(\bar{h}_{45} - \bar{h}_{46}) = 0$. The orthogonality of rows 2,4 shows that $a(x_1 + \bar{x}_1) + x_2(\bar{h}_{45} - \bar{h}_{46}) = 0$. So $h_{35} = x_2$ or $h_{45} = h_{46}$. If $h_{35} = x_2$, then the orthogonality of rows 2,3 shows $3 - a(x_1 + \bar{x}_1) + x_1^2 = 0$, the equation has no solution, which is a contradiction. If $h_{45} = h_{46}$, then $a = 0$ or $x_1 = \pm i$. Now if $a = 0$, then the inner product of rows 2,3 is $1 + x_1^2 + 2x_2\bar{h}_{35} = 0$. Then $x_1^2 = 1$ and $x_2\bar{h}_{35} = -1$, then $x_1 = \pm 1$, $h_{35} = -x_2$, $M(a)$ becomes

$$\begin{bmatrix} 0 & 1 & 1 & 1 & 1 & 1 \\ 1 & 0 & x_1 & -x_1 & x_2 & -x_2 \\ 1 & x_1 & 0 & -x_1 & -x_2 & x_2 \\ 1 & -x_1 & -x_1 & 0 & x_1 & x_1 \\ 1 & \bar{x}_2 & -\bar{x}_2 & x_1 & 0 & h_{56} \\ 1 & -\bar{x}_2 & \bar{x}_2 & x_1 & \bar{h}_{56} & 0 \end{bmatrix} \quad (\text{A7})$$

The orthogonality of rows 4,5 shows $h_{56} = -x_1$, then for any x_2 , $M(a)$ indeed satisfies that $M(a)^2 = 5I_6$. For $x_1 = 1$ we obtain $M(0)_1$, for $x_1 = -1$ we obtain $M(0)_2$. The expression of M in (A4) deduced by $M(0)_1$ or $M(0)_2$ will be shown in the latter case.

If $x_1 = \pm i$, then the inner product of rows 2,3 is $-2ax_1 + 2x_2\bar{h}_{35} = 0$, so $a = 1$. We then find that $M(a)$ is indeed a self-adjoint CHM, which means $M(a)$ is complex equivalent to Hermitian matrices in (4). Moreover, $m = -1$ and $\lambda = -\sqrt{6}$. It is known that CHM with eigenvalues $\sqrt{6}, \sqrt{6}, \sqrt{6}, -\sqrt{6}, -\sqrt{6}, -\sqrt{6}$ is complex equivalent to Hermitian matrices in (4).

Case A.2. $h_{35} = -\bar{x}_1$, then $h_{36} = -h_{34}$, and the row 5 turns to $[1, \bar{x}_2, -x_1, \bar{h}_{45}, a, h_{56}]$, the orthogonality of rows 2,5 shows that $-x_1 h_{45} - x_2 \bar{h}_{56} = 0$. And the orthogonality of rows 3,5 and shows that $1 + \bar{x}_1 x_2 + h_{34}(h_{45} - \bar{h}_{56}) = 0$, then $(1 + \bar{x}_1 x_2)(1 - h_{34} h_{56}) = 0$, then $h_{34} = h_{56}$ or $x_2 = -x_1$. If $h_{34} = h_{56}$, then the orthogonality of rows 1,6 shows $1 - \bar{x}_2 + \bar{h}_{46} + a = 0$, since we let $0 \leq a \leq 1$, then $h_{46} = -\bar{x}_2$. The orthogonality of rows 2,6 and rows 4,5 show that $1 + x_1 \bar{x}_2 + h_{34}(x_2 - x_1) = 0$ and $1 - x_1 \bar{x}_2 - h_{34}(x_1 + x_2) = 0$. Then $h_{34} = \bar{x}_1$, then $h_{45} = -x_2$. The inner product of rows 1,5 is $2a + \bar{x}_1 - x_1 = 0$, which shows x_1 is real and $a = 0$. The orthogonality of rows 1,4 shows that $h_{46} = x_2$, so $M(a)$ takes the following form.

$$\begin{bmatrix} 0 & 1 & 1 & 1 & 1 & 1 \\ 1 & 0 & x_1 & -x_1 & x_2 & -x_2 \\ 1 & x_1 & 0 & x_1 & -x_1 & -x_1 \\ 1 & -x_1 & x_1 & 0 & -x_2 & x_2 \\ 1 & \bar{x}_2 & -x_1 & -\bar{x}_2 & 0 & x_1 \\ 1 & -\bar{x}_2 & -x_1 & \bar{x}_2 & x_1 & 0 \end{bmatrix} \quad (\text{A8})$$

We denote this matrix by $M(0)_3$. The expression of M in (A4) deduced by $M(0)_3$ will be shown in the latter case.

If $x_2 = -x_1$, then rows 2 and 4 are respectively $[1, -a, x_1, -x_1, -x_1, x_1]$ and $[1, -\bar{x}_1, x_1, -a, -x_1, \bar{x}_1]$, the orthogonality of rows 2,4 shows that $3 + a(x_1 + \bar{x}_1) + x_1^2 = 0$, which has no solution.

Case B. $h_{23} = -h_{25}$, then by permuting rows and columns, the first four rows of $M(a)$ must be

$$\begin{bmatrix} a & 1 & 1 & 1 & 1 & 1 \\ 1 & -a & x_1 & x_2 & -x_1 & -x_2 \\ 1 & \bar{x}_1 & -a & x_3 & h_{35} & h_{36} \\ 1 & \bar{x}_2 & \bar{x}_3 & -a & h_{45} & h_{46} \end{bmatrix} \quad (\text{A9})$$

Here $x_1 \neq -x_2$, $x_3 \neq -\bar{x}_1$, $x_3 \neq -x_2$, otherwise Case B will degenerate into Case A. By permuting rows and columns and redefining x_1, x_2, x_3 , there are only two distinct cases to discuss, that is $h_{35} = -\bar{x}_1, h_{36} = -x_3, h_{45} = -\bar{x}_2, h_{46} = -\bar{x}_3$ or $h_{35} = -x_3, h_{36} = -\bar{x}_1, h_{45} = -\bar{x}_2, h_{46} = -\bar{x}_3$. We denote the two cases by Case B.1 and Case B.2.

Case B.1. Now $M(a)$ is

$$\begin{bmatrix} a & 1 & 1 & 1 & 1 & 1 \\ 1 & -a & x_1 & x_2 & -x_1 & -x_2 \\ 1 & \bar{x}_1 & -a & x_3 & -\bar{x}_1 & -x_3 \\ 1 & \bar{x}_2 & \bar{x}_3 & -a & -\bar{x}_2 & -\bar{x}_3 \\ 1 & -\bar{x}_1 & -x_1 & -x_2 & a & h_{56} \\ 1 & -\bar{x}_2 & -\bar{x}_3 & -x_3 & \bar{h}_{56} & a \end{bmatrix} \quad (\text{A10})$$

The orthogonality of rows 3,5 shows that $h_{56} = -x_2$. The orthogonality of rows 1,5 and 1,6 show that x_2 is real and either $x_1 = x_3$ or $x_1 = \bar{x}_3$. The orthogonality of rows 4,5 implies that $x_1 = \bar{x}_3$. By solving the equation $M(a)^2 = (5 + a^2)I_6$, we obtain the only restriction, that is

$$-2ax_1 + 2x_1x_2 + x_1^2 + 1 = 0. \quad (\text{A11})$$

We substitute $x = x_1, y = x_3 = \bar{x}, z = x_2$, the above equation turns to $-2a + 2z + x + y = 0$, i.e. $-2a + x + y + z + xyz = 0$. This case corresponds to the case $\phi_1 = -\phi_2$ in Lemma 1. The solutions and the expression of M in (A4) will be shown in the latter case.

Case B.2. Now $M(a)$ is

$$\begin{bmatrix} a & 1 & 1 & 1 & 1 & 1 \\ 1 & -a & x_1 & x_2 & -x_1 & -x_2 \\ 1 & \bar{x}_1 & -a & x_3 & -x_3 & -\bar{x}_1 \\ 1 & \bar{x}_2 & \bar{x}_3 & -a & -\bar{x}_2 & -\bar{x}_3 \\ 1 & -\bar{x}_1 & -\bar{x}_3 & -x_2 & a & h_{56} \\ 1 & -\bar{x}_2 & -x_1 & -x_3 & \bar{h}_{56} & a \end{bmatrix} \quad (\text{A12})$$

The orthogonality of rows 3,5 shows that $h_{56} = -\bar{x}_1 x_2 \bar{x}_3$. Computing $M(a)^2$, we denote $M(a)^2 = [f_{jk}]_{6 \times 6}$, then $f_{11} = f_{22} = \dots = f_{66} = 5 + a^2$, and $|f_{15}| = |2a - x_1 - x_3 - \bar{x}_2 - x_1 \bar{x}_2 x_3|$. To satisfy $M(a)^2 = (5 + a^2)I_6$, we must have

$$x_1 + \bar{x}_2 + x_3 + x_1 \bar{x}_2 x_3 = 2a. \quad (\text{A13})$$

The equation above is the only restriction such that $M(a)^2 = (5 + a^2)I_6$ in Case B.2. To solve this equation, we substitute $x = x_1, y = \bar{x}_2, z = x_3$, then equation A13 becomes

$$x + y + z + xyz = 2a. \quad (\text{A14})$$

By Lemma 1 we can solve the above equation. In fact, this equation takes the same form as equation (9).

In fact, If we take $a = 0$, the equation (A13) has solutions $\{x_1, x_2\} = \{1, -1\}$ or $\{x_1, x_3\} = \{1, -1\}$ or $\{x_2, x_3\} = \{1, -1\}$. If we take $\{x_1, x_3\} = \{1, -1\}$, then the matrix in (A12) gives $M(0)_1$ and $M(0)_2$ in Case A.1 and $M(0)_3$ in Case A.2 up to complex equivalence. Moreover, if we take $y = \bar{x}$, then by the result in Lemma 1 (ii) we can solve the case B.1.

In conclusion, $M(a)$ must be complex equivalent to

$$\begin{bmatrix} a & 1 & 1 & 1 & 1 & 1 \\ 1 & -a & x & \bar{y} & -x & -\bar{y} \\ 1 & \bar{x} & -a & z & -z & -\bar{x} \\ 1 & y & \bar{z} & -a & -y & -\bar{z} \\ 1 & -\bar{x} & -\bar{z} & -\bar{y} & a & -\bar{x}y\bar{z} \\ 1 & -y & -x & -z & -xyz & a \end{bmatrix} \quad (\text{A15})$$

Then

$$M = \frac{\lambda - \sqrt{6}}{\sqrt{20 + 4a^2}} \begin{bmatrix} d & 1 & 1 & 1 & 1 & 1 \\ 1 & -\bar{d} & x & \bar{y} & -x & -\bar{y} \\ 1 & \bar{x} & -\bar{d} & z & -z & -\bar{x} \\ 1 & y & \bar{z} & -\bar{d} & -y & -\bar{z} \\ 1 & -\bar{x} & -\bar{z} & -\bar{y} & d & -\bar{x}y\bar{z} \\ 1 & -y & -x & -z & -xyz & d \end{bmatrix} \quad (\text{A16})$$

Here $d = \frac{\sqrt{20 + 4a^2}}{\lambda - \sqrt{6}} \cdot \left[\sqrt{6} + (\lambda - \sqrt{6}) \left(\frac{1}{2} + \frac{a}{\sqrt{4a^2 + 20}} \right) \right]$. We know that $\text{Re}[\lambda] = \sqrt{6}m = -\frac{\sqrt{6}}{3}(2 + a^2)$, and we may assume $\text{Im}[\lambda] = \sqrt{\frac{10 - 8a^2 - 2a^4}{3}}$, then $d = 2\sqrt{5 + a^2}(a - \sqrt{1 - a^2}i)$. The eigenvalues of M are $\sqrt{6}, \sqrt{6}, \sqrt{6}, \lambda, \lambda, \lambda$. For the simplicity of the form, we denote

$$K(d, x) = \begin{bmatrix} d & 1 & 1 & 1 & 1 & 1 \\ 1 & -\bar{d} & x & \bar{y} & -x & -\bar{y} \\ 1 & \bar{x} & -\bar{d} & z & -z & -\bar{x} \\ 1 & y & \bar{z} & -\bar{d} & -y & -\bar{z} \\ 1 & -\bar{x} & -\bar{z} & -\bar{y} & d & -\bar{x}y\bar{z} \\ 1 & -y & -x & -z & -xyz & d \end{bmatrix} \quad (\text{A17})$$

In conclusion, CHMs with two sorts of eigenvalues must be complex equivalent to two-parameter matrices $K(d, x)$. The meaning of parameter d, x will be explained in detail in Lemma 1. The trace of $K(d, x)$ is $6\text{Im}[d]$, we denote the two sorts of eigenvalues by

$$\lambda_1 = \frac{\sqrt{20+4a^2}}{\lambda - \sqrt{6}} \cdot \sqrt{6}, \quad \lambda_2 = \frac{\sqrt{20+4a^2}}{\lambda - \sqrt{6}} \cdot \lambda, \quad (\text{A18})$$

and rewrite $d = e^{i\theta}$, then by computation we know that

$$\lambda_1 = \sqrt{6 - \sin^2 \theta} + i \sin \theta, \quad \lambda_2 = -\sqrt{6 - \sin^2 \theta} + i \sin \theta. \quad (\text{A19})$$

Hence we have finished the proof of Theorem 1. $\square$

Appendix B. The Proof of Lemma 1

Proof. If $r = 0$ then we have $x = -y$ or $x = -z$ or $y = -z$. If $x = -y$, then equation (11) turns to $z - y^2 z = 0$. Hence $y = \pm 1$, and $\{x, y\} = \{1, -1\}$. For the other two cases, we can deduce $\{x, z\} = \{1, -1\}$ or $\{y, z\} = \{1, -1\}$ similarly.

If $r \neq 0$, then $r \in [-2, 0) \cup (0, 2]$. Now if $1 + xy = 0$, then $y = -\bar{x}$, then (11) turns to $r + x - \bar{x} = 0$, which means $x \in \mathbb{R}$ and $r = 0$, hence the contradiction. So we obtain that $z = -\frac{r+x+y}{1+xy}$.

To satisfy $|z| = 1$, we have

$$(1 + \bar{x}\bar{y})(1 + xy) = (r + x + y)(r + \bar{x} + \bar{y}). \quad (\text{A20})$$

Then

$$(x - \bar{x} - r)y^2 - r(x + \bar{x} + r)y + (\bar{x} - x - r) = 0. \quad (\text{A21})$$

We obtain that

$$y = \frac{rx^2 + r^2x + r \pm \sqrt{\Delta}}{2(x^2 - rx - 1)}. \quad (\text{A22})$$

where

$$\Delta = (r^2 + 4)x^4 + 2r^3x^3 + (r^4 - 2r^2 - 8)x^2 + 2r^3x + r^2 + 4. \quad (\text{A23})$$

here the square root denotes the principal value of the complex power function $z \rightarrow z^{\frac{1}{2}}$. Next we discuss the condition that keeps $|y| = 1$.

We assume $x = e^{i\phi_1}, y = e^{i\phi_2}, z = e^{i\phi_3}$, then

$$\begin{cases} \cos \phi_1 + \cos \phi_2 + \cos \phi_3 + \cos(\phi_1 + \phi_2 + \phi_3) = -r; \\ \sin \phi_1 + \sin \phi_2 + \sin \phi_3 + \sin(\phi_1 + \phi_2 + \phi_3) = 0. \end{cases} \quad (\text{A24})$$

If $\phi_1 = -\phi_2$, then (A24) becomes

$$\begin{cases} 2 \cos \phi_1 + 2 \cos \phi_3 = -r; \\ 2 \sin \phi_3 = 0. \end{cases} \quad (\text{A25})$$

Once λ_1, λ_2 are given, then r is also given, we can solve x, y, z due to (A25). That is, $\cos \phi_1 = \frac{-r-2}{2}, \phi_3 = 0$ or $\cos \phi_1 = \frac{-r+2}{2}, \phi_3 = \pm\pi$.

If $\phi_1 + \phi_2 = \pi$, then $x = -\bar{y}$, and (A24) becomes

$$\begin{cases} 0 = -r; \\ 2 \sin \phi_1 = 0. \end{cases} \quad (\text{A26})$$

Hence $r = 0$, which is a contradiction.

If $\phi_1 - \phi_2 = \pm\pi$, then $x = -y$, (11) becomes $r + z - y^2 z = 0$, then $y = \pm 1$ or $y^2 z^2 = -1$. $y = \pm 1$ violates the restriction; $y^2 z^2 = -1$ means $z + \bar{z} = r$, $\{x, y\} = \{i\bar{z}, -i\bar{z}\}$.

For other cases, we have

$$\begin{aligned}\sin \phi_3 &= \frac{r + \cos \phi_1 + \cos \phi_2 + (1 + \cos(\phi_1 + \phi_2)) \cos \phi_3}{\sin(\phi_1 + \phi_2)} \\ &= -\frac{\sin \phi_1 + \sin \phi_2 + \sin(\phi_1 + \phi_2) \cos \phi_3}{1 + \cos(\phi_1 + \phi_2)}.\end{aligned}\quad (\text{A27})$$

We square both sides of equations of (A24). Their sum is

$$\cos \phi_3 = \frac{\frac{r^2-4}{4} - \cos \phi_1 \cos \phi_2}{\cos \phi_1 + \cos \phi_2}.\quad (\text{A28})$$

Substituting the value of $\cos \phi_3$ in (A28) to (A27), we have

$$\frac{1}{2} \cos \frac{\phi_1 + \phi_2}{2} \sec \frac{\phi_1 - \phi_2}{2} \left(r^2 + 2r(\cos \phi_1 + \cos \phi_2) + 4 \sin \phi_1 \sin \phi_2 \right) = 0.\quad (\text{A29})$$

Then we have

$$-r^2 - 2r \cos \phi_1 = 2r \cos \phi_2 + 4 \sin \phi_1 \sin \phi_2.\quad (\text{A30})$$

The right side of (A30) can be written as $\sqrt{4r^2 + 16 \sin^2 \phi_1} \sin(\phi_2 + \psi)$, where $\tan \psi = \frac{r}{2 \sin \phi_1}$. For the case that $\sin \phi_1 = 0$, we stipulate that $\psi = \frac{\pi}{2}$. Since $\sin(\phi_2 + \psi) \in [-1, 1]$, we deduce the value condition of ϕ_1 below.

$$-1 \leq -\frac{r^2 + 2r \cos \phi_1}{\sqrt{4r^2 + 16 \sin^2 \phi_1}} \leq 1.\quad (\text{A31})$$

So we have

$$\frac{-r^3 - 4\sqrt{2}\sqrt{r^2 + 2}}{2(r^2 + 4)} \leq \cos \phi_1 \leq \frac{-r^3 + 4\sqrt{2}\sqrt{r^2 + 2}}{2(r^2 + 4)}.\quad (\text{A32})$$

This equation ensures that $|y| = 1$ and determines the range of ϕ_1 . Hence we have finished the proof. $\square$

Appendix C. The Proof of Theorem 2

Proof. We assume $|u_5\rangle = [0, g_2, g_3 e^{is_3}, g_4 e^{is_4}, g_5 e^{is_5}, g_6 e^{is_6}]^T$, $|u_6\rangle = [0, h_2, h_3 e^{it_3}, h_4 e^{it_4}, h_5 e^{it_5}, h_6 e^{it_6}]^T$ and denote $s_2 = t_2 = 0$ and $\lambda_3 = \sqrt{6}e^{i\theta_1}$, $\lambda_4 = \sqrt{6}e^{i\theta_2}$. We have

$$h_{jk} = -\frac{1}{5} - \frac{1}{5}\lambda_3 + (\lambda_4 - \lambda_3)(g_j g_k e^{i(s_j - s_k)} + h_j h_k e^{i(t_j - t_k)}), \quad j, k \geq 2, j \neq k,\quad (\text{A33})$$

$$h_{jj} = -\frac{1}{5} + \frac{4}{5}\lambda_3 + (\lambda_4 - \lambda_3)(g_j^2 + h_j^2), \quad j \geq 2,\quad (\text{A34})$$

$$\begin{aligned}||h_{jk}|^2 - |h_{kj}|^2| &= -\frac{8}{5}\sqrt{6}\left(\cos \frac{\theta_1 - \theta_2}{2} + \cos \frac{\theta_1 + \theta_2}{2}\right) \cdot \sin \frac{\theta_1 - \theta_2}{2}. \\ g_j g_k \sin(s_j - s_k) + h_j h_k \sin(t_j - t_k) &= 0.\end{aligned}\quad (\text{A35})$$

To satisfy the condition in (A35), we divide our discussion into three cases, namely **Case A**, **B** and **C**.

Case A. If $\sin \frac{\theta_1 - \theta_2}{2} = 0$, then $\theta_1 = \theta_2$, and $\lambda_3 = \lambda_4$. From the first sentence below (19), we see that $K'(d, x)$ has four identical eigenvalues. However the CHM with four identical eigenvalues does not exist due to Theorem 6 in [19].

Case B. If $g_j g_k \sin(s_j - s_k) + h_j h_k \sin(t_j - t_k) = 0$, then for any $j, k \geq 2$, $f_{jk} := g_j g_k e^{i(s_j - s_k)} + h_j h_k e^{i(t_j - t_k)}$ is real. Now $h_{jk} = -\frac{1}{5} - \frac{1}{5}\lambda_3 + f_{jk}(\lambda_4 - \lambda_3)$ for $j \neq k$, so we have $h_{jk} = h_{kj}$ and H is symmetric. Note that $|h_{jk}| = 1$, if λ_3, λ_4 are given, then $-\frac{1}{5} - \frac{1}{5}\lambda_3$ and $\lambda_4 - \lambda_3$ are both given. Using geometry properties, we see that f_{jk} takes only two distinct values. It means for all $j, k \geq 2$ and $j \neq k$, h_{jk} can only take two distinct values. We denote them by a_1, a_2 . For h_{jj} , $j \geq 2$, by a similar discussion, we obtain that h_{jj} can take at most two distinct values b_1, b_2 . In particular if h_{jj} can only take one value, then we take $b_1 = b_2$.

We consider the orthogonality of rows 1,2, that is, $1 + \sum_{k=2}^6 h_{2k} = 0$. If three of $h_{23}, \dots, h_{26}$ are the same, we shall assume $1 + h_{22} + 3h_{23} + h_{26} = 0$. Then $h_{22} = h_{26} = 1$ and $h_{23} = -1$, so λ_3, λ_4 are both real, which means for any $j, k \geq 2$, h_{jk} is real, too. So H contains only real elements, i.e. $\{1, -1\}$. Such a CHM is known to be non-existent. So two of $h_{23}, \dots, h_{26}$ are the same, then $1 + h_{22} + 2a_1 + 2a_2 = 0$. By simple vector theory we know that the vector $a_1 + a_2$ is orthogonal to $\lambda_4 - \lambda_3$.

Then $1 + h_{22}$ must be orthogonal to $\lambda_4 - \lambda_3$. If $b_1 = b_2$ then h_{22} is orthogonal to $\lambda_4 - \lambda_3$, so $b_1 = b_2 = 1$. If $b_1 \neq b_2$, we shall assume $h_{22} = b_1$. Since $b_1 + b_2$ is orthogonal to $\lambda_4 - \lambda_3$, then we have $b_2 = 1$. So in both cases, we have $b_2 = 1$.

Since $h_{jk} = h_{kj}$, if b_1 appears no less than three times in $h_{22}, \dots, h_{66}$, then we select three rows that contain b_1 , and by permuting columns, the three rows should be

$$\begin{bmatrix} 1 & b_1 & a_1 & a_1 & a_2 & a_2 \\ 1 & a_1 & b_1 & x_1 & x_2 & x_3 \\ 1 & a_1 & x_1 & b_1 & x_4 & x_5 \end{bmatrix}$$

Here $\{x_1, x_2, x_3\} = \{x_1, x_4, x_5\} = \{a_1, a_2, a_2\}$. If $x_1 = a_1$, then the orthogonality of rows 2,3 never holds. Then $x_1 = a_2$, then $x_4 = x_3, x_5 = x_2$, the inner products of the three rows shows that $2 + b_1 \bar{a}_1 + a_1 \bar{b}_1 + a_1 \bar{a}_2 + a_2 \bar{a}_1 = 0$ and $2 + b_1 \bar{a}_2 + a_1 \bar{b}_2 + a_1 \bar{a}_2 + a_2 \bar{a}_1 = 0$. Then we know that $-b_1^2 |a_2 - a_1|^2 = (a_1 - a_2)^2$. Then b_1 is orthogonal to $a_1 - a_2$. By simple geometry property we know that $b_1 - 1$ is parallel to $a_1 - a_2$, then $b_1 = 1$ or $a_1 = a_2$. The latter case violates the orthogonal of the three rows, then $b_1 = 1$. And by solving $2 + a_2 + \bar{a}_2 + a_1 \bar{a}_2 + a_2 \bar{a}_1 = 0$, we know that $\{a_1, a_2\} = \{\omega, \omega^2\}$ with $\omega = e^{\frac{2\pi i}{3}}$. Other rows contains the same elements, so H is complex equivalent to Tao matrix $S_6^{(0)}$. If b_2 appears no less than three times, we can also solve that $\{a_1, a_2\} = \{\omega, \omega^2\}$, and we then deduce that $b_1 = 1$.

Case C. If $\sqrt{6} \cos \frac{\theta_1 - \theta_2}{2} + \cos \frac{\theta_1 + \theta_2}{2} = 0$, then $\tan \frac{\theta_1}{2} \tan \frac{\theta_2}{2} = -\frac{7+2\sqrt{6}}{5}$. Since $\lambda_3 \neq \lambda_4$, we denote $a' = -\frac{1+\lambda_3}{5(\lambda_4 - \lambda_3)}$, $b' = g_j g_k e^{i(s_j - s_k)} + h_j h_k e^{i(t_j - t_k)}$, then $|h_{jk}| = |h_{kj}| = 1$ shows that $|a' + b'| = |a' + \bar{b}'|$, where $\bar{b}'$ is the conjugate of b' . Since a' is a given complex number, the equality shows that either a' is real or b' is always real whatever j, k are. The latter case was discussed in Case B, it is just the case that $g_1 g_2 \sin(s_1 - s_2) + h_1 h_2 \sin(t_1 - t_2) = 0$.

Now a' is real, so there exists a real number k such that $a' = k$, then $-\frac{1}{5} - \frac{1}{5} \lambda_3 = k(\lambda_4 - \lambda_3)$. So

$$\lambda_4 = -\frac{1}{5k} + (1 - \frac{1}{5k})\lambda_3. \quad (\text{A36})$$

Compute the real and imaginary part of two sides, we obtain the following system of equations.

$$\begin{cases} \sqrt{6} \cos \theta_2 = -\frac{1}{5k} + \sqrt{6}(1 - \frac{1}{5k}) \cos \theta_1; \\ \sin \theta_2 = (1 - \frac{1}{5k}) \sin \theta_1. \end{cases} \quad (\text{A37})$$

We compute

$$|\lambda_4 - \lambda_3| = \sqrt{(\lambda_4 - \lambda_3)(\bar{\lambda}_4 - \bar{\lambda}_3)} = \frac{1}{\sqrt{k - 5k^2}}. \quad (\text{A38})$$

We claim that the following equality holds:

$$1 + \lambda_3 + \lambda_4 = \frac{\lambda_4 - \lambda_3}{\bar{\lambda}_4 - \bar{\lambda}_3}. \quad (\text{A39})$$

From (A36), the equality turns to be

$$(1 - \frac{1}{5k} + (2 - \frac{1}{5k})\lambda_3)(1 + \bar{\lambda}_3) = 1 + \lambda_3. \quad (\text{A40})$$

Using $|\lambda_3| = \sqrt{6}$, it turns to be

$$(1 - \frac{1}{5k})\lambda_3^2 + (12 - \frac{7}{5k})\lambda_3 + 6(1 - \frac{1}{5k}) = 0. \quad (\text{A41})$$

From (A36) and $|\lambda_4|^2 = \lambda_4 \cdot \bar{\lambda}_4 = 6$ we deduce the same equation as (A41). So we have proven the equality (A39).

Now we denote

$$d = \frac{\lambda_4 - \lambda_3}{|\lambda_4 - \lambda_3|}. \quad (\text{A42})$$

So from (A39) we know that

$$d^2 = \frac{(\lambda_4 - \lambda_3)^2}{(\lambda_4 - \lambda_3)(\bar{\lambda}_4 - \bar{\lambda}_3)} = \lambda_3 + \lambda_4 + 1. \quad (\text{A43})$$

We select

$$|v_1\rangle = [\sqrt{5k}, -\sqrt{\frac{1-5k}{5}}, -\sqrt{\frac{1-5k}{5}}, -\sqrt{\frac{1-5k}{5}}, -\sqrt{\frac{1-5k}{5}}, -\sqrt{\frac{1-5k}{5}}]^T, \quad (\text{A44})$$

$$|v_2\rangle = [\sqrt{1-5k}, \sqrt{k}, \sqrt{k}, \sqrt{k}, \sqrt{k}, \sqrt{k}]^T. \quad (\text{A45})$$

If we denote $\lambda_{43} = \lambda_4 - \lambda_3$, then

$$\lambda_3|v_1\rangle\langle v_1| + \lambda_4|v_2\rangle\langle v_2| = \begin{bmatrix} A_1 & A_2 & A_2 & A_2 & A_2 & A_2 \\ A_2 & A_3 & A_3 & A_3 & A_3 & A_3 \\ A_2 & A_3 & A_3 & A_3 & A_3 & A_3 \\ A_2 & A_3 & A_3 & A_3 & A_3 & A_3 \\ A_2 & A_3 & A_3 & A_3 & A_3 & A_3 \\ A_2 & A_3 & A_3 & A_3 & A_3 & A_3 \end{bmatrix} \quad (\text{A46})$$

with $A_1 = \lambda_4 - 5k\lambda_{43}$, $A_2 = \sqrt{k-5k^2}\lambda_{43}$, $A_3 = \frac{1}{5}\lambda_3 + k\lambda_{43}$.

Substituting the expression of λ_3, λ_4 discussed in (A36)(A38)(A42)(A43), we have

$$A_1 = \lambda_4 - 5k\lambda_{43} = (1-5k)\lambda_{43} + \lambda_3 = 1 + \lambda_4 + \lambda_3 = d^2, \quad (\text{A47})$$

$$A_2 = \sqrt{k-5k^2}\lambda_{43} = \frac{\lambda_4 - \lambda_3}{|\lambda_4 - \lambda_3|} = d, \quad (\text{A48})$$

$$A_3 = \frac{1}{5}\lambda_3 + k\lambda_{43} = \frac{1}{5}\lambda_3 + k\left[-\frac{1}{5k} + (1 - \frac{1}{5k})\lambda_3 - \lambda_3\right] = -\frac{1}{5}. \quad (\text{A49})$$

And

$$\langle v_1|v_2\rangle = \sqrt{5k(1-5k)} - 5\sqrt{\frac{k(1-5k)}{5}} = 0. \quad (\text{A50})$$

So $|v_1\rangle, |v_2\rangle$ are orthogonal. From (A46)(A47)(A48)(A49) we know that

$$\lambda_3|v_1\rangle\langle v_1| + \lambda_4|v_2\rangle\langle v_2| = \frac{1}{5} \begin{bmatrix} 5d^2 & 5d & 5d & 5d & 5d & 5d \\ 5d & -1 & -1 & -1 & -1 & -1 \\ 5d & -1 & -1 & -1 & -1 & -1 \\ 5d & -1 & -1 & -1 & -1 & -1 \\ 5d & -1 & -1 & -1 & -1 & -1 \\ 5d & -1 & -1 & -1 & -1 & -1 \end{bmatrix}. \quad (\text{A51})$$

For the vector $|u_j\rangle = [g_1^j, \dots, g_6^j]^T, j = 3, 4, 5, 6$, we have $\langle u_1|u_j\rangle = \langle u_2|u_j\rangle = 0$. Then $\langle u_1 - u_2|u_j\rangle = 0$, from (20) we know that $g_1^j = 0$ for $j = 3, 4, 5, 6$. So once we take the matrix P in (19) and set the parameter d to be in (A42), we have $P^\dagger \cdot \langle u_j|u_j\rangle \cdot P^\dagger = \langle u_j|u_j\rangle$ for $j = 3, 4, 5, 6$. And

$$P^\dagger \cdot H \cdot P^\dagger = P^\dagger H_2 P^\dagger + \lambda_3 \sum_{j=3}^4 |u_j\rangle\langle u_j| + \lambda_4 \sum_{j=5}^6 |u_j\rangle\langle u_j|, \quad (\text{A52})$$

here H_2 was defined in (22) and

$$P^\dagger H_2 P^\dagger = \frac{1}{5} \begin{bmatrix} 5d^2 & 5d & 5d & 5d & 5d & 5d \\ 5d & -1 & -1 & -1 & -1 & -1 \\ 5d & -1 & -1 & -1 & -1 & -1 \\ 5d & -1 & -1 & -1 & -1 & -1 \\ 5d & -1 & -1 & -1 & -1 & -1 \\ 5d & -1 & -1 & -1 & -1 & -1 \end{bmatrix}. \quad (\text{A53})$$

Then we find that

$$\lambda_3|v_1\rangle\langle v_1| + \lambda_4|v_2\rangle\langle v_2| = P^\dagger H_2 P^\dagger. \quad (\text{A54})$$

We shall assume $|u_k\rangle = [0, x_1, x_2, x_3, x_4, x_5]^T$ for any $k = 3, 4, 5, 6$, from (20) and the orthogonality of $|u_1\rangle$ and $|u_k\rangle$, we know that

$$x_1 + x_2 + x_3 + x_4 + x_5 = 0. \quad (\text{A55})$$

From (A44)(A45) we know that $\langle v_j|u_k\rangle = 0$ for any $j = 1, 2$. So we obtain a orthonormal basis $\{|v_1\rangle, |v_2\rangle, |u_3\rangle, |u_4\rangle, |u_5\rangle, |u_6\rangle\}$ and from (A52)(A54) we know that

$$P^\dagger H P^\dagger = \lambda_3|v_1\rangle\langle v_1| + \lambda_4|v_2\rangle\langle v_2| + \lambda_3 \sum_{j=3}^4 |u_j\rangle\langle u_j| + \lambda_4 \sum_{j=5}^6 |u_j\rangle\langle u_j|. \quad (\text{A56})$$

So $P^\dagger H P^\dagger$ exactly has eigenvalues $\lambda_3, \lambda_3, \lambda_3, \lambda_4, \lambda_4, \lambda_4$. Hence given a CHM H in dephased form with eigenvalues $\sqrt{6}, -\sqrt{6}, \lambda_3, \lambda_3, \lambda_4, \lambda_4$ excluding the Tao matrix (which has been discussed in Case B) in (3), we will deduce a CHM with two sorts of eigenvalues, the latter one must be complex equivalent to $K(d, x)$ due to Theorem 1. So H must be complex equivalent to $P \cdot K(d, x) \cdot P$ with P in (19). We take $K'(d, x) = d \cdot P \cdot K(d, x) \cdot P$, now $K'(d, x)$ here is in dephased form. And if we assume the eigenvalues of $K'(d, x)$ are $\sqrt{6}, -\sqrt{6}, \lambda_3, \lambda_3, \lambda_4, \lambda_4$, we obtain that $\{\lambda_3, \lambda_4\} = \{d\lambda_1, d\lambda_2\}$ with λ_1, λ_2 in (10). Hence we deduce the expression of $K'(d, x)$ in (21).

Now if $d = \pm i$, then by the result of Lemma 1 (i), we obtain that $\{x, y\}$ or $\{y, z\}$ or $\{x, z\}$ is equal to $\{1, -1\}$. It is easy to verify that in all these cases, $K'(d, x)$ must be complex equivalent to Haagerup matrices in (1).

If $d = \pm 1$, then from (10) we know that $\lambda_1 = \sqrt{6}, \lambda_2 = -\sqrt{6}$. So $\{\lambda_3, \lambda_4\} = \{\sqrt{6}, -\sqrt{6}\}$, then $K'(d, x)$ is complex equivalent to Hermitian matrices in (4). Hence we have finished the proof of Theorem 2. $\square$

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