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Stoustrup, Jakob; Schedletzky, O.; Glaser, S.J.; Griesinger, C.; Nielsen, N.C.; Sørensen, O.W.

Published in:
Physical Review Letters

DOI (link to publication from Publisher):
[10.1103/PhysRevLett.74.2921](https://doi.org/10.1103/PhysRevLett.74.2921)

Publication date:
1995

Document Version
Tidlig version også kaldet pre-print

[Link to publication from Aalborg University](#)

Citation for published version (APA):
Stoustrup, J., Schedletzky, O., Glaser, S. J., Griesinger, C., Nielsen, N. C., & Sørensen, O. W. (1995). A Generalized Bound on Quantum Dynamics: Efficiency of Unitary Transformations between Non-Hermitian States. *Physical Review Letters*, 2921-2924. <https://doi.org/10.1103/PhysRevLett.74.2921>

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Generalized Bound on Quantum Dynamics: Efficiency of Unitary Transformations between Non-Hermitian States

J. Stoustrup,¹ O. Schedletsky,² S. J. Glaser,² C. Griesinger,² N. C. Nielsen,^{3,*} and O. W. Sørensen^{4,*}

¹*Mathematical Institute, Technical University of Denmark, DK-2800 Lyngby, Denmark*

²*Institut für Organische Chemie, Universität Frankfurt, Marie-Curie Strasse 11, D-60439 Frankfurt, Germany*

³*Department of Chemistry, University of Aarhus, DK-8000 Aarhus C, Denmark*

⁴*Novo Nordisk A/S, DK-2880 Bagsværd, Denmark*

(Received 28 July 1994)

We report a general bound for the efficiency of polarization or coherence transfer between quantized states under unitary transformations. This bound is more general than the so-called universal bound on spin dynamics [O. W. Sørensen, *J. Magn. Reson.* **86**, 435 (1990)] that applies only to transformations between Hermitian states. The new bound on transfer efficiency under unitary transformations between general non-Hermitian operators depends exclusively on the singular values of the matrix representations of these operators.

PACS numbers: 33.25.+k, 42.50.-p, 76.60.-k, 76.70.-r

Design or evaluation of experimental schemes for transfer of coherence or polarization between states in quantized systems requires a clear understanding of which regions of operators in Liouville space are interconvertible by available propagators. This not only involves insight into upper limits for the efficiency of transfer from one state to another but is also related to conservation laws (or constants of motion) of the quantum dynamics. Furthermore, in experimental coherent spectroscopy it is important to have transformation of high efficiency because that is reflected in the sensitivity of the experiment. Recently, attempts in this direction have been undertaken in nuclear magnetic resonance (NMR) spectroscopy; however, attention has mainly been devoted to transformation between *Hermitian operators* in *nondissipative* systems using *unitary* transformations [1]. In many physical systems these simplifications fit the experimental circumstances because of the Hermiticity of physical observables, the different time scales applying for nondissipative and dissipative (e.g., relaxation) quantum evolution, and the fact that relevant Hamiltonians are Hermitian, leading to unitary propagators. Although so far addressed primarily in relation to NMR, determination of bounds on quantum dynamics is relevant in several disciplines of chemistry and physics [2] including quantum optics. We mention that progress in coherent optical spectroscopy [3] with ultrashort laser pulses [4] opens up for optical analogs to NMR multiple-pulse sequences to which bounds on coherence or population transfer are of interest. As another example, establishment of upper limits for polarization transfer efficiency is highly relevant for “hybrids” between optical and magnetic resonance spectroscopy [5]. Finally, bounds on quantum dynamics are also directly applicable in multidimensional microwave Fourier transform spectroscopy [6].

The so-called *universal bound on spin dynamics* and its multidimensional variants describe bounds on the regions of operators in Liouville space being interconvertible by unitary transformations [1]. For unitary transfor-

mation between *Hermitian operators* in finite spin systems it has been shown that (i) only states with identical eigenvalues can be interconverted completely, (ii) the accessible region is significantly smaller than predicted by considering conservation of the norm of the state vector as the only bound on spin dynamics, and (iii) from knowledge of maximum transfer efficiencies and transformation propagators it is possible to design optimized experimental schemes [1,7].

In this Letter we present a generalization of this theory to include unitary transformations between operators which are not restricted to be Hermitian. The practical usefulness of such a bound is a consequence of the fact that a pair of off-diagonal elements of the (Hermitian) density matrix σ_{ij} and σ_{ji} evolve with eigenfrequencies of opposite sign under the free-precession Hamiltonian and rotations about the symmetry axis z . Hence such components can be separated by a complex Fourier transformation or appropriate z rotations like phase cycles or series of pulsed field gradients [8,9]. In other words, the new bound describes the limits of unitary transformations of individual non-Hermitian operators of the Hermitian density operator. Within, for example, NMR this bound is of considerable practical value, e.g., for optimization of multidimensional liquid-state NMR experiments employing pulsed field gradients [9], useful for the suppression of disturbing solvent signals [10].

A general entry to the problem is given by the transformation

$$UBU^\dagger = aA + Q, \quad \text{Tr}\{A^\dagger Q\} = 0, \quad (1)$$

where U is a unitary propagator (i.e., $U^{-1} = U^\dagger$) serving transformation between the initial and desired final operators B and A , respectively. Obviously, the coefficient a is given by

$$a = \frac{\text{Tr}\{UB^\dagger U^\dagger A\}}{\text{Tr}\{A^\dagger A\}}. \quad (2)$$

The problem of transfer efficiency is equivalent to determining the range of a .

For transformation between *Hermitian operators*, the limits for transfer of polarization or coherence from spin state B to spin state A depend on the eigenvalues Λ_i of these operators through the relations [1]

$$a_{\max}^{\text{Herm}} = \max_U \left[\frac{\text{Tr}\{UB^\dagger U^\dagger A\}}{\text{Tr}\{A^\dagger A\}} \right] = \frac{\sum_{i=1}^n \Lambda_i^A \Lambda_i^B}{\sum_{i=1}^n (\Lambda_i^A)^2}, \quad (3a)$$

$$a_{\min}^{\text{Herm}} = \min_U \left[\frac{\text{Tr}\{UB^\dagger U^\dagger A\}}{\text{Tr}\{A^\dagger A\}} \right] = \frac{\sum_{i=1}^n \Lambda_{n-i+1}^A \Lambda_i^B}{\sum_{i=1}^n (\Lambda_i^A)^2}, \quad (3b)$$

with the eigenvalues sorted in descending (or ascending) order; e.g., $\Lambda_1^A \geq \Lambda_2^A \geq \dots \geq \Lambda_n^A$ and $\Lambda_1^B \geq \Lambda_2^B \geq \dots \geq \Lambda_n^B$. The upper limit may readily be proven by rewriting the trace according to

$$\begin{aligned} \text{Tr}\{UB^\dagger U^\dagger A\} &= (\Lambda_1^A \Lambda_2^A \dots \Lambda_n^A) \\ &\times \begin{pmatrix} |U'_{11}|^2 & \dots & |U'_{1n}|^2 \\ \vdots & \ddots & \vdots \\ |U'_{n1}|^2 & \dots & |U'_{nn}|^2 \end{pmatrix} \begin{pmatrix} \Lambda_1^B \\ \Lambda_2^B \\ \vdots \\ \Lambda_n^B \end{pmatrix}, \end{aligned} \quad (4)$$

where we introduced the diagonalizations and eigenvalue orderings $A = U_A \Lambda^A U_A^\dagger$ and $B = U_B \Lambda^B U_B^\dagger$ along with $U' = U_A^\dagger U U_B$ (all matrices are unitary). Since U' is unitary the $n \times n$ matrix with elements $|U'_{ij}|^2$ in Eq. (4) is doubly stochastic. (A square matrix S is said to be stochastic if it has non-negative entries and all its row sums are equal to 1; S is denoted doubly stochastic if S and S^\dagger are stochastic.) Any doubly stochastic matrix S other than the unity matrix $\mathbb{1}$ will contain two nonzero entries s_{ij} and s_{kl} with $i > k$ and $j < l$. Hence, it is possible to define another doubly stochastic matrix S_δ by

$$S_\delta = \begin{pmatrix} s_{11} & \dots & s_{1j} & \dots & s_{1l} & \dots & s_{1n} \\ \vdots & & \vdots & & \vdots & & \vdots \\ s_{k1} & \dots & s_{kj} + \delta & \dots & s_{kl} - \delta & \dots & s_{kn} \\ \vdots & & \vdots & & \vdots & & \vdots \\ s_{i1} & \dots & s_{ij} - \delta & \dots & s_{il} + \delta & \dots & s_{in} \\ \vdots & & \vdots & & \vdots & & \vdots \\ s_{n1} & \dots & s_{nj} & \dots & s_{nl} & \dots & s_{nn} \end{pmatrix}, \quad (5)$$

with $\delta = \min[s_{ij}, s_{kl}] > 0$. For the two real vectors $\Phi = (\phi_1 \phi_2 \dots \phi_n)$ and $\Pi = (\pi_1 \pi_2 \dots \pi_n)$, with the orderings $\phi_1 \geq \phi_2 \geq \dots \geq \phi_n$ and $\pi_1 \geq \pi_2 \geq \dots \geq \pi_n$, one obtains

$$\Phi S_\delta \Pi^\dagger = \Phi S \Pi^\dagger + \delta(\phi_i - \phi_k)(\pi_l - \pi_j) > \Phi S \Pi^\dagger. \quad (6)$$

This shows that

$$\max_S [\Phi S \Pi^\dagger] = \sum_{i=1}^n \phi_i \pi_i, \quad (7)$$

where the maximum is obtained using $S = \mathbb{1}$. With

Eq. (7) at hand it follows from Eq. (4) that

$$\max_U [\text{Tr}\{UB^\dagger U^\dagger A\}] = \sum_{i=1}^n \Lambda_i^A \Lambda_i^B. \quad (8a)$$

The maximum in Eq. (8a) is assumed when $U' = \mathbb{1}$ (i.e., $U = U_A U_B^\dagger$), thereby establishing the proof of Eq. (3a). For the lower limit a_{\min}^{Herm} the proof is analogous and

$$\min_U [\text{Tr}\{UB^\dagger U^\dagger A\}] = \sum_{i=1}^n \Lambda_{n-i+1}^A \Lambda_i^B. \quad (8b)$$

It holds that $a_{\min}^{\text{Herm}} = -a_{\max}^{\text{Herm}}$ when for the ordered eigenvalues $\Lambda_i^A = \Lambda_i^{-A}$ or $\Lambda_i^B = \Lambda_i^{-B}$ for all i .

As stated in the introduction, it is of considerable interest to establish similar bounds on the quantum dynamics associated with transformation between *non-Hermitian operators*. In this case singularity of the initial and/or desired final states prevents the use of similarity transformations to determine their eigenvalues. However, for any complex $n \times m$ matrix M there exists unitary matrices T and V of dimensions $n \times n$ and $m \times m$, respectively, and a real diagonal $n \times m$ matrix Σ ,

$$\Sigma = \begin{pmatrix} \tilde{\Sigma} & 0 \\ 0 & 0 \end{pmatrix}, \quad \tilde{\Sigma} = \begin{pmatrix} \sigma_1 & 0 & \dots & 0 \\ 0 & \sigma_2 & \dots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \dots & \sigma_k \end{pmatrix}, \quad (9)$$

with $\sigma_1 \geq \sigma_2 \geq \dots \geq \sigma_k > 0$ such that

$$M = T \Sigma V^\dagger. \quad (10)$$

Equation (10) is known as the *singular value decomposition* (SVD) of M [11].

Using the singular value decompositions $A = T_A \Sigma^A V_A^\dagger$ and $B = T_B \Sigma^B V_B^\dagger$ for the general complex $n \times n$ matrices A and B one readily obtains

$$\text{Tr}\{UB^\dagger U^\dagger A\} = \text{Tr}\{(V_A^\dagger U V_B) \Sigma^B (T_A^\dagger U T_B)^\dagger \Sigma^A\}, \quad (11)$$

where the traces can be complex. It follows that

$$\max_U [\text{Tr}\{UB^\dagger U^\dagger A\}] \leq \max_{X,Y} [\text{Tr}\{X \Sigma^B Y^\dagger \Sigma^A\}], \quad (12)$$

where X and Y are unitary matrices. The inequality holds because the right-hand side has an additional degree of freedom compared to the expression in Eq. (11). Equation (12) can be evaluated in the following way:

$$\begin{aligned} \text{Tr}\{(X - Y) \Sigma^B (X - Y)^\dagger \Sigma^A\} &= \\ \text{Tr}\{X \Sigma^B X^\dagger \Sigma^A\} + \text{Tr}\{Y \Sigma^B Y^\dagger \Sigma^A\} & \\ - 2 \text{Re}(\text{Tr}\{X \Sigma^B Y^\dagger \Sigma^A\}) &\geq 0, \end{aligned} \quad (13)$$

where Re denotes the real part. This implies that

$$\max_{X,Y} [\text{Re}(\text{Tr}\{X \Sigma^B Y^\dagger \Sigma^A\})] \leq \max_Z [\text{Tr}\{Z \Sigma^B Z^\dagger \Sigma^A\}] \quad (14a)$$

or

$$\max_{X,Y} [\text{Tr}\{X \Sigma^B Y^\dagger \Sigma^A\}] \leq \max_Z [\text{Tr}\{Z \Sigma^B Z^\dagger \Sigma^A\}], \quad (14b)$$

where Z is unitary. The equivalence of Eqs. (14a) and (14b) is due to the fact that all diagonal elements of the

product matrix through phase adjustments in X can be made real and positive. In fact, the equal sign in Eq. (14) holds because the trace on the right-hand side is always positive and a special case of the one on the left-hand side. It corresponds to the Hermitian case, and in analogy to Eq. (3) it is now possible to establish a *generalized bound* in terms of Eqs. (1), (2), and (9),

$$|a| \leq \frac{\sum_{i=1}^{k'} \sigma_i^A \sigma_i^B}{\sum_{i=1}^k (\sigma_i^A)^2}, \quad (15)$$

where k' denote the smallest of the dimensions for the ordered singular value matrices $\tilde{\Sigma}_A$ and $\tilde{\Sigma}_B$ and k the dimension of $\tilde{\Sigma}_A$. In the following we let the equal sign in Eq. (15) denote the SVD bound on spin dynamics, i.e.,

$$a_{\max}^{\text{SVD}} = \frac{\sum_{i=1}^{k'} \sigma_i^A \sigma_i^B}{\sum_{i=1}^k (\sigma_i^A)^2}, \quad (16a)$$

$$a_{\min}^{\text{SVD}} = -a_{\max}^{\text{SVD}}. \quad (16b)$$

In contrast to the bounds on unitary transformation between Hermitian operators where the boundaries can always be achieved there is no guarantee that the boundaries in Eq. (16) can be achieved. Applied to Hermitian operators the general bound in Eq. (16) yields the same result as the Hermitian bound provided the numerators in Eq. (3) are invariant to absolute-value formation of the individual eigenvalues and reordering in descending (or ascending) order.

The following coherence transfer processes in heteronuclear $I_N S$ spin- $\frac{1}{2}$ systems with $N = 1, 2,$ and 3 illustrate all aspects of the spin dynamics bounds discussed:

$$U2F_z S_x U^\dagger = aF_x + Q, \quad (17)$$

$$U2F_z S_- U^\dagger = aF_- + Q, \quad (18)$$

$$US_x U^\dagger = aF_x + Q, \quad (19)$$

$$US_- U^\dagger = aF_- + Q, \quad (20)$$

where $F_p = \sum_{i=1}^N I_{ip}$ and S_p describe angular momentum operators ($I_\pm = I_x \pm iI_y$, $S_\pm = S_x \pm iS_y$) for the I and S spins, respectively. For these four examples all $a_{\max} = -a_{\min}$ so we only discuss a_{\max} . In addition, a_{\max} and a_{\min} are invariant to the substitution of any x by y or z and any $-$ by $+$.

Table I contains a_{\max}^{Herm} and a_{\max}^{SVD} for the transfer processes in Eqs. (17)–(20) supplemented by the norm bound $a_{\max}^{\text{norm}} = \sqrt{\text{Tr}\{B^\dagger B\}/\text{Tr}\{A^\dagger A\}}$ predicted by conservation of the norm of the state vector alone [i.e., $\text{Tr}\{(UBU^\dagger)^\dagger(UBU^\dagger)\}$ is independent of U]. Table I confirms the common finding that the norm limit is achievable for two-spin IS systems or when the eigenvalues of the two (Hermitian) operators in question are identical. In the other cases the norm limit is unachievable. Even the SVD bound can predict too high an efficiency of transfer between non-Hermitian operators as is the case for the $I_2 S$ and $I_3 S$ processes of Eq. (20). That follows from multiple application of the Hermitian bound to transfers between the Hermitian linear combinations $(B + B^\dagger)$, $i(B - B^\dagger)$ and $(A + A^\dagger)$, $i(A - A^\dagger)$ of operators A and B :

$$\begin{aligned} 4\text{Tr}\{UB^\dagger U^\dagger A\} &= \text{Tr}\{U(B^\dagger + B)U^\dagger(A + A^\dagger)\} \\ &\quad - i\text{Tr}\{U(B^\dagger + B)U^\dagger i(A - A^\dagger)\} \\ &\quad - i\text{Tr}\{Ui(B^\dagger - B)U^\dagger(A + A^\dagger)\} \\ &\quad - \text{Tr}\{Ui(B^\dagger - B)U^\dagger i(A - A^\dagger)\}, \end{aligned} \quad (21)$$

implying

$$\begin{aligned} 4 \max_U [|\text{Tr}\{UB^\dagger U^\dagger A\}|] &\leq \max_V [([\text{Tr}\{V(B^\dagger + B)V^\dagger(A + A^\dagger)\}]^2 + [\text{Tr}\{V(B^\dagger + B)V^\dagger i(A - A^\dagger)\}]^2)^{1/2}] \\ &\quad + \max_W [([\text{Tr}\{Wi(B^\dagger - B)W^\dagger(A + A^\dagger)\}]^2 + [\text{Tr}\{Wi(B^\dagger - B)W^\dagger i(A - A^\dagger)\}]^2)^{1/2}]. \end{aligned} \quad (22)$$

TABLE I. Maximum transfer coefficients a_{\max} for typical $UBU^\dagger = aA + Q$ coherence transfer processes in heteronuclear $I_N S$ spin systems ($N = 1, 2,$ and 3) determined by the Hermitian (a_{\max}^{Herm}), the SVD (a_{\max}^{SVD}), and the norm (a_{\max}^{norm}) bound. For transfer between non-Hermitian operators the upper limit of Eq. (23) is included in brackets (see text).

Eq.	B	A	IS			$I_2 S$			$I_3 S$		
			a_{\max}^{Herm}	a_{\max}^{SVD}	a_{\max}^{norm}	a_{\max}^{Herm}	a_{\max}^{SVD}	a_{\max}^{norm}	a_{\max}^{Herm}	a_{\max}^{SVD}	a_{\max}^{norm}
(17)	$2F_z S_x$	F_x	1	1	1	1	1	1	1	1	1
(18)	$2F_z S_-$	F_-	[1]	1	1	[1]	$\frac{1}{\sqrt{2}}$	1	[1]	$\frac{7+2\sqrt{3}}{12}$	1
(19)	S_x	F_x	1	1	1	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{\sqrt{2}}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{\sqrt{3}}$
(20)	S_-	F_-	[1]	1	1	$[\frac{1}{2}]$	$\frac{1}{\sqrt{2}}$	$\frac{1}{\sqrt{2}}$	$[\frac{1}{2}]$	$\frac{3+2\sqrt{3}}{12}$	$\frac{1}{\sqrt{3}}$

When the operators $(A + A^\dagger)$, $i(A - A^\dagger)$, and $[(A + A^\dagger), i(A - A^\dagger)]$ fulfill the same commutator relations as the angular momentum operators I_x , I_y , and I_z , linear combinations $(A + A^\dagger) \cos\theta + i(A - A^\dagger) \sin\theta$ have the same eigenvalues independent of θ . With these stipulations Eq. (22) can be simplified,

$$4 \max_U [\text{Tr}\{UB^\dagger U^\dagger A\}] \leq \max_V [\text{Tr}\{V(B^\dagger + B)V^\dagger(A + A^\dagger)\}] + \max_W [\text{Tr}\{Wi(B^\dagger - B)W^\dagger(A + A^\dagger)\}], \quad (23)$$

where the two terms on the right-hand side are identical when also $(B + B^\dagger)$ and $i(B - B^\dagger)$ have the same eigenvalues. The bound of Eq. (23) is included in brackets in the Hermitian column of Table I for the transfer processes of Eqs. (18) and (20). A final comment on Table I is that the SVD limit for Eq. (18), in fact, has been achieved for $N = 1$ and $N = 2$ and appropriate pulse experiments developed in heteronuclear liquid-state NMR using pulsed field gradients [9].

To summarize, we have generalized the universal bound on spin dynamics to include unitary transformations between non-Hermitian operators in Liouville space. This extension will have important consequences for design and evaluation of experimental techniques in wide ranges of coherent spectroscopy. In particular, it is foreseen that the generalized bounds will find immediate application for the design of sensitivity-enhanced NMR experiments using pulsed field gradients for artifact and solvent signal suppression.

*Author to whom correspondence should be addressed.

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