Adaptive Release Duration Modulation for Limited Molecule Production and Storage

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Abstract—The nature of molecular transmitter imposes some limitations on the molecule production process and its storage. As the molecules act the role of the information carriers, the limitations affect the transmission process and the system performance considerably. In this paper, we focus on the transmitter’s limitations, in particular, the limited molecule production rate and the finite storage capacity. We consider a time-slotted communication where the transmitter opens its outlets and releases the stored molecules for a specific time duration to send bit “1” and remains silent to send bit “0”. By changing the Release Duration (RD), we propose an adaptive RD modulation. The objective is to find the optimal transmission RD to minimize the probability of error. We characterize the properties of the optimal RD and use it to derive upper and lower bounds on the system performance. We see that the proposed modulation scheme improves the performance.

Index Terms—Molecular transmitter, transmitter’s limitations, production rate, storage capacity, release duration.

I. INTRODUCTION

MOLECULAR communication (MC) is a promising communication technique among machines in nanoscale and has important applications in medicine, health-care and environmental sciences, where the conditions of transfer such as transmission range, nature of the medium, and possible transmitted carriers are consistent with molecules [2]. MC has some similarities and differences with the classical communication. The main difference is that in MC, molecules are carriers of information instead of electromagnetic waves. Another important difference of MC with classical communication, which we focus on in this paper, is the transmitter’s limitations.

The most important transmitter’s limitation in MC is availability of molecules that must be released as information carriers. We study this problem for a common on/off keying (OOK) transmitter, where the transmitter releases a specific concentration of molecules into the medium in order to send bit “1” and remains silent in order to send bit “0”. The performance analysis of diffusion based MC with OOK modulation is studied in [3]. However, the amount of available molecules at the transmitter is subject to some constraints due to the molecule production process. In fact, the molecules are produced at the transmitter with a limited-rate process, such as chemical reactions [4]. In addition, the amount of molecules that can be stored in the transmitter is limited and thus we face a finite storage capacity. The objective in this paper is to study the effect of these limitations on the performance of an MC system, considering the probability of error at the receiver.

While molecular channels, receivers, and carriers are widely studied in the literature [4], [5], molecular transmitter’s limitations are relatively unexplored. An ideal transmitter is considered in most of the works, which is a point source that can release any desired number of molecules in a very short time at the beginning of a time-slot (i.e., an impulse function) [6], [7]. But the size of nano-machines is small and the storage capacity of molecules is limited [8]. In [8]–[10], an energy harvesting scheme called Simultaneous Molecular Information and Energy Transfer (SMIET) is introduced, where the receiver collects the received molecules and reuses them for its transmission as a relay. There are few works that study the transmitter’s challenges in MC [11]–[17]. The amplitude constraint on the number of transmitted molecules is studied in [11], [12]. In [13], the transmitter releases molecules in shape of a square or Gaussian pulse, where any number of molecules can be released during the pulse time (removing the impulse function). The authors in [14] consider a point source with instantaneous molecule release but with random symbol interval. The focus of [14] is on synchronization schemes.

A more realistic transmitter model is introduced in [16], which is a point source with limited molecule production rate, located at the center of a spherical cell with finite storage capacity. There are many ion channels on its surface that can be opened or closed by applying an electrical voltage or a ligand concentration. The MC transmitter with these two limitations is also studied in [17] from information theoretic perspective and some bounds on its capacity are derived. Although the transmitter’s limitations have been modeled in some of the above works, no specific transmission scheme has been proposed for these limitations. In our previous work [1], we have considered the transmitter’s limitations and proposed an adaptive modulation scheme, in the absence of inter-symbol interference (ISI). The hitting probabilities were considered independent of the Release Duration...
(RD). Important properties of the optimal RDs and some performance bounds have been provided.

In this paper, we focus on the transmitter’s limitations, i.e., the limited production rate and finite storage capacity. We consider diffusion-based mediums as an important class of MC. Our goal is to design a transmission scheme that has a good performance in terms of probability of error, considering the limitations. Our contribution and results are provided in two cases of the absence and the presence of ISI.

A. No-ISI Case

- **Transmitter Design:** We propose a coding scheme based on changing RDs (i.e., molecule RD). This lets the transmitter adapt the number of transmitted molecules to satisfy the transmitter constraints.
- **Optimal RDs:** We use the optimal maximum likelihood receiver, which has a threshold form with a fixed threshold, to formulate the optimal RDs as the solution of an optimization problem which does not have a closed form solution. However, we characterize its important properties.
- **Adaptive Receiver:** We propose a sub-optimal (ML like) receiver, which estimates the state of the system based on the previous decoded bits. Assuming a genie-aided receiver, we obtain the analytical probability of error, which is a lower bound on the performance of the sub-optimal receiver.
- **Sub-Optimal Decoder:** We design the optimal decoder that minimizes the probability of error. Thereby, we obtain an adaptive threshold based receiver for the optimal RDs.
- **Probability of Error Analysis:** We compute the probability of error. Using the properties of optimal RDs, we obtain upper and lower bounds on the probability of error.

These results were presented in our previous work [1]. In this paper, we also provide our results for the ISI case as follows.

B. ISI Case

- **Transmitter Design:** Similar to no-ISI case, we propose a coding scheme to adapt the number of transmitted molecules, satisfying the transmitter’s constraints.
- **Optimal RDs:** We formulate an optimization problem to find the optimal RDs for a fixed threshold receiver and obtain sub-optimal solutions for the channel with one and two-symbol ISI.
- **Adaptive Receiver:** Similar to no-ISI case, we propose a sub-optimal (ML like) receiver. The probability of error is obtained analytically for a genie-aided receiver, which is a lower bound on the performance of the sub-optimal receiver. Also, we show that the error probability of the sub-optimal receiver is at most two times of the error probability of the genie-aided receiver.
- **Sub-Optimal Decoder:** We design two sub-optimal decoders which use the sub-optimal RDs to minimize the probability of error for one and two-symbol ISI. Then, we derive the optimal adaptive threshold based receiver for the sub-optimal RDs.

Furthermore, we provide analytical and simulation results:
- **Storage Capacity:** We show that increasing the molecule storage capacity significantly improves the performance.

![Molecular transmitter, channel and receiver.](image)

- **Receiver:** It is shown that an adaptive threshold receiver performs better than a fixed threshold receiver as expected.
- **Memory:** While in our proposed adaptive strategy, the complexity of the system increases compared to non-adaptive strategy where RDs are fixed, we show that the required memories for the proposed transmitter and for the receiver are finite.
- **Simulation:** A Particle Based Simulation (PBS) is provided to validate the analytical results of proposed strategies, for both of sub-optimal and genie-aided receivers.

The rest of this paper is organized as follows. Section II presents a model for transmitter’s limitations. In Section III, a modulation scheme based on adaptive RD is proposed. Main results in no-ISI case and ISI case are provided in Sections IV and V, respectively. Numerical results are presented in Section VI. Finally, in Section VII, we conclude this paper.

II. SYSTEM MODEL

We consider a point-to-point MC system including a point source as a transmitter, a fluid medium as a three-dimensional channel, and a spherical receiver with radius \( r_R \). The transmitter is located at distance \( d \) from the surface of the receiver. We assume a time-slotted communication system with an OOK transmitter and the slot duration of \( T \), shown in Fig. 1. The transmitter releases a deterministic concentration of molecules into the channel. However, the transmitter has a limited molecule production rate \( \beta \) and a finite molecule storage capacity \( B_M \). The molecule storage is linearly recharged with a fixed rate \( \beta \) up to its storage capacity \( B_M \). We assume that the storage is fully charged in a duration less than slot duration \( (B_M < \beta T) \). Therefore, it is fully charged if a “0” is sent.

The released molecules diffuse in the channel according to Fick’s second law of diffusion to reach the receiver, which results in a linear time-invariant channel. We consider an absorbing receiver which absorbs molecules hitting its surface, with probability \( p_k, k = 0, 1, \ldots, \) in the current and next \( k \) slots [18]. The reception process can be modeled as a Poisson process at the receiver [19]. In addition, we assume a Poisson background noise, \( N_{ch} \), with parameter \( \lambda \) as [20]. We consider a transmitter, located at \( r = 0 \), releases \( X_t \) molecules at \( i \)-th time-slot as a pulse train \( \sum_{k} X_t \delta(\tilde{r} = 0)\Pi(t - k\tau, t_\tau) \), where \( \Pi(t, t_\tau) \) is its pulse shape with the RD of \( t_\tau \) and thus, \( \Pi(t, t_\tau) = 0 \) for \( t \notin [0, t_\tau] \). If the shape of released pulse is Dirac delta \( \delta(t) \), the probability that a molecule hits the receiver before time \( t \), that is obtained in [21] as

\[
p_{hit}(t) = \frac{r_R}{r_R^2 + d^2} \frac{1}{4\pi D t} \frac{d}{\sqrt{4\pi D t}} \exp\left(-\frac{d^2}{4Dt}\right). \tag{1}
\]
We know that \( p_k = \int_{(k-1)T}^{kT} p_{hit}(t)dt \). If \( X_i \) is large and the \( p_k \)'s have small values, the number of received molecules is [22]

\[
Y_i \sim \text{Poisson} \left( \sum_{k=0}^{\infty} p_k X_{i-k} + \lambda \right).
\]

(2)

III. PROBLEM FORMULATION

A. No-ISI Case

First we study a simple case of no-ISI (i.e., if \( p_k^n \)'s are the hitting probabilities of no-ISI case, we have \( p_k^n = 0 \) for \( k \geq 1 \)). We focus on a specific transmission scheme, where to send bit “0”, the surface outlets of the transmitter are closed and no molecule is released. To send bit “1”, the outlets are opened for some specific time duration and the stored molecules and the produced molecules in this time duration are released. Then the outlets are closed and the storage starts recharging.

1) Non-Adaptive Communication Strategy: First, consider a simple strategy where the RD of \( T_M \) is fixed. We set \( T_M \) such that the storage is filled in time duration \( T - T_M \).

\[
T_M = \max_{B_M \geq \beta(T-t)} t = T - \frac{B_M}{\beta}.
\]

(3)

Thus, the storage is full at the beginning of each time-slot and \( B_M = \beta(T - T_M) \). In other words, \( T_M \) is chosen such that \( T - T_M \) is the minimum transmitted required time to refill the storage; otherwise, we lose some molecule production time. Assume that bit “1” is to be transmitted at slot \( t \). As soon as the storage fills up to its capacity \( B_M \), the outlets are opened for duration \( T_M \). \( B_M \) molecules released simultaneously, and \( \beta T_M \) molecules are also produced at this time and released while they are produced with rate \( \beta \). Therefore the total number of released molecules for bit “1” is \( X_i = B_M + \beta T_M = \beta T \). Therefore, there is not any limitation on the release rate of the molecules. Each molecule is released as soon as it is produced if the outlets are opened. If there are \( B_M \) molecules in the storage at \( t = 0 \) and the outlets are opened, the released pulse is as follows.

\[
\Pi(t, T_M) = \begin{cases} B_M \delta(t) + \beta, & 0 \leq t \leq T_M, \\ 0, & \text{otherwise}. \end{cases}
\]

For a continuous released pulse, the expected number of absorbed molecules at time \( t \) is obtained in [22] as

\[
N_{\text{abs}}(t) = \int_0^t \Pi(t, T_M) * p_{hit}(t)dt,
\]

where * indicates the convolution operator. Thus, we can normalize \( \Pi(t, T_M) \) to the number of molecules and obtain the hitting probabilities for non-adaptive strategy as

\[
p_k^N = \frac{1}{B_M + \beta T_M} (N_{\text{abs}}(k+1T) - N_{\text{abs}}(kT)).
\]

(4)

If we quantize the RD into very short intervals, substituting the above amount of \( X_i \) in (2) and using thinning property of Poisson distribution result in \( Y_i \sim \text{Poisson}(p_k^N \beta T + \lambda) \). At the receiver, we use the optimal maximum likelihood (ML) decoding approach. The ML receiver has a fixed threshold form. If we define \( M = \beta T \), the optimum threshold is derived as \( T_{th} = \frac{p_{th}^N M}{\ln(1 + p_{th}^N M)} \). Since \( p_{th}^N M \gg \lambda \), we have \( T_{th} < p_{th}^N M \). The receiver observes \( Y_i = y \) and decides \( \hat{X}_i = 1 \) if \( y \geq T_{th} \) and \( \hat{X}_i = 0 \) if \( y < T_{th} \). We assume that the bits “0” and “1” are equiprobable. Thus, the total probability of error is

\[
P_e = \frac{1}{2} \left( P_{e|0}(0) + P_{e|1}(p_{th}^N M) \right),
\]

(5)

where \( P_{e|1}(x) = 1 - P_{e|0}(x) = \sum_{y < T_{th}} e^{-(x+\lambda)} \frac{(x+\lambda)^y}{y!} \).

2) Adaptive Communication Strategy: Now, we propose an adaptive strategy. We improve the system performance in terms of probability of error at the receiver by making the RD adaptively variable. Thus, the storage may not be fully charged at the beginning of each time-slot. The duration of a time-slot is chosen such that transmitting a “0” fully charges the storage. As a result, the transmitter has to know the number of subsequent “1”s that has been transmitted prior to the current transmission. Thus, the system is state-dependent. We say that the system is in state \( s_j \) if the \( j \)-th subsequent “1”s are transmitted after the last transmitted “0”. In state \( s_j \), we change the RD of \( T_M \) to \( T_M + \tau_{j+1} \). We call \( \tau_i \in \mathbb{R} \) as RD increments for \( i = 1, 2, \ldots \). Note that it can be negative to decrease the RD. The release process in each slot begins when the storage is full, at \( kT + \tau_1 + \cdots + \tau_j \), in state \( s_j \) for \( k \)-th transmitted bit (see Fig. 3(a)). The transmitter state machine is shown in Fig. 2. The probability of state \( s_j \) is

\[
P(s_j) = \frac{1}{2^{j+1}}.
\]

(6)

Let \( \Delta_n = \beta \tau_n, n \in \mathbb{N} \) denotes the amount of the released molecules increment in state \( s_{n-1} \). If the hitting probabilities \( p_k \)'s are constant in different states, the probability of error is

\[
P_e = \frac{1}{2} \left( P_{e|0}(0) + \sum_{j=1}^{\infty} P(s_{j-1}) P_{e|1}(p_{th}(M + \Delta_j)) \right)
\]

\[
= \frac{1}{2} \left( P_{e|0}(0) + \sum_{j=1}^{\infty} \frac{1}{2^j} P_{e|1}(p_{th}(M + \Delta_j)) \right),
\]

(7)

where (a) follows from (6) and \( P_{e|1}(s_{j-1}) \) is the error probability of bit “1” in state \( s_{j-1} \).

Our goal is to design \( \tau_i \)'s in order to minimize the probability of error. Because \( P_{e|0} \) does not depend on \( \tau_i \), we only consider \( P_{e|1} \) and solve an optimization problem. Let us define

\[
F(\{\Delta_i\}_{i=1}^{\infty}) \triangleq \sum_{j=1}^{\infty} \frac{1}{2^j} P_{e|1}(p_{th}(M + \Delta_j)),
\]

(8)
storage to increase the released molecules in the slot (for bit "1"). From (7),

\[ P_i(t) = \frac{e^{-t} x + \lambda}{T_{th} x} < 0, \]

where \( x \) is a fraction of released molecules. As a result, if we increase RD for sending "1", then the number of released molecules increases and probability of error decreases.

As seen in Fig. 3(a), at state \( s_0 \), if bit "1" comes, we increase the RD by \( \tau_1 \). So for the next bit "1", the storage is not full at the beginning of time-slot. To compensate this reduction, we wait for time \( \tau_1 \) in order to the storage be filled. Then, the outlets are opened for RD of \( T_{th} + \tau_2 \). Note that we should have \( \tau_1 + \tau_2 \geq 0 \) not to waste molecule production time. For state \( s_i \), we wait for time \( \tau_1 + \cdots + \tau_i \) in order to the storage be filled and then open the outlets for duration \( T_{th} + \tau_{i+1} \).

Totally, we increase the transmitted molecule number in state \( s_i \) by \( \beta \tau_{i+1} \). If \( \tau_i, s \) are positive, the number of released molecules increases in states \( s_i \) for \( i = 1, \ldots, J \) and remains fixed (i.e., \( T_M \)) for \( i > J \).

We propose a sub-optimal (ML-like) receiver, which uses the estimated states (\( \hat{s} \)) for its decision. Because of different delays in different states, the hitting probabilities may change based on the system state. To tackle this problem, in the estimated state \( \hat{s} = s_k \) and \( r \)-th slot, the receiver uses the absorbed molecules in time interval \( [\theta_1(r + i, i_r), \theta_2(r + i, i_r)] = [rT + \sum_{j=1}^{i-1} \tau_j, rT + T_M + \sum_{j=1}^{i-1} \tau_j] \) for its decision (see Fig. 3(b)). The beginning of each interval equals the related delay of the pulse in state \( s_j \) and the end of each interval equals the end of the related pulse. These limitations are applied to the duration of receiving intervals in order not to enter the next slot. If the distance of the transmitter and the receiver is small enough, most of the molecules are quickly received in this interval, after they are released. Thus, most of them hit the receiver and they are absorbed in the mentioned interval after they are released. Thus, for \( k = 0, 1, \ldots \), the hitting probabilities are

\[
p_k = \frac{1}{B_M + \beta T_M} \int_{0}^{T_M} \Pi(t, T_M + \tau_{i_r}) * \phi_{hit}(t) dt,
\]

where (a) follows from a variable change. If \( \tau_i, s \) are much smaller than the slot duration \( T \), we can approximately assume that the hitting probabilities do not depend on \( \tau_i \). Therefore, we can formulate the problem by substituting \( \tau_i = 0 \) in the above equation, which results in the following hitting probabilities

\[
p_k = \frac{1}{B_M + \beta T_M} \frac{kT + T_M}{J} \Pi(t, T_M) * \phi_{hit}(t) dt.
\]

Obtaining an analytical expression for the error probability of the sub-optimal (ML-like) receiver is not feasible. We find this error probability by performing a particle-based simulation in Section VI. Furthermore, we assume a genie-aided receiver (\( \hat{s} = s \)) and we obtain its probability of error analytically and also via simulations. It gives us a lower bound on the performance of the proposed sub-optimal (ML like) receiver. We now show that the error probability of the sub-optimal receiver is at most twice that of the genie-aided receiver as follows. It is proved in Section IV-A2.

**B. ISI Case**

Similar to no-ISI case, our goal is to minimize \( P_e \) subject to the constraints in (10) and (11). If the channel memory is \( K \) slots (i.e., \( p_k = 0, k > K \) in (2)), and if we denote the increment of molecule number to send "1" in state \( s_i \) by \( \Delta_{i+1} \), \( P_e \) for a fixed threshold receiver can be written as the following form.

\[
P_e = \frac{1}{2} \sum_{j=0}^{\infty} P(s_j) \left( P_{e|0,s_j} + P_{e|1,s_j} \right)
\]
where $w_0(\cdot)$ and $w_1(\cdot)$ are the number of received molecules when the current bit is “0” and “1”, respectively.

The $c_i$ and $d_i$s are weighted sum of $M$ and $p_i$s, depending on the current and $K$ previous states and bits. We explain these weights through an example. Assume that $K = 4$ and bit sequence $(B_1, \ldots, B_6) = (011101101)$ has been transmitted. Now, we send $B_{10} \in \{0, 1\}$ and we have

$w_0 = (p_1 + p_3 + p_4) M + p_1 \Delta_1 + p_2 \times 0 + p_3 \Delta_2 + p_4 \Delta_1,$

$w_1 = w_0 + p_0 (M + \Delta_2),

= (p_0 + p_1 + p_3 + p_4) M + (p_1 + p_4) \Delta_1 + (p_0 + p_3) \Delta_2.$

### IV. MAIN RESULTS IN NO-ISI CASE

1) **Optimal Solution:** It can be easily shown that the objective function $F(\cdot)$ in (8), is convex (see Appendix A) and the domain of optimization problem in (9) is compact ($\Delta_i \in [0, \beta(T - T_M)]$ for $i = 1, 2, \ldots$). Thus, the global minimum exists. We start to find the solution using the Lagrangian method. We have the KKT conditions at the optimal point (shown as $\{\Delta^*_i\}_{i=1}^\infty$), that the regularity condition is Linear Independence Constraint Qualification (LICQ). As a result, for the active conditions, we have

$$\nabla F(\{\Delta^*_i\}_{i=1}^\infty) = \sum_{i=1}^{\infty} \mu^*_1 \nabla C_1 + \sum_{i=1}^{\infty} \mu^*_2 \nabla C_2,$$

where $\nabla$ denotes the vector differential operator ($\nabla = (\frac{\partial}{\partial \Delta_1}, \frac{\partial}{\partial \Delta_2}, \ldots)$). $\mu^*_1$ and $\mu^*_2$ are non-negative Lagrange multipliers for $i = 1, 2, \ldots$, and $C_{ki}, k = 1, 2$ have been defined in (10) and (11). The above equation results in the following three properties for the optimal solution, stated in Lemmas 1, 2 and 3. The proofs of these lemmas are provided in Appendices B, C and D, respectively.

**Lemma 1:** The sequence of $\{\Delta^*_i\}_{i=1}^\infty$ is decreasing and nonnegative.

**Lemma 2:** There exists an index $J$ such that $\Delta^*_j = 0$, for $j > J$. This means that a finite number of RD increments are positive (i.e., non-zero).

**Remark 1:** The above lemma concludes that we only need to change the RDs for a finite number of states. Hence, the transmitter needs a limited memory for saving the optimal values of these RD increments. An upper bound on the number of RD increments is derived in Section IV-A3.

Note that from (12), $P_{e|0}$ is a decreasing function. Now, let $p_0(M + a_j)$ be the point where $\frac{dx}{dx} P_{e|1}(x)$ reaches half of its value at $x = p_0 M$ and similarly $p_0(M + a_i)$ for $i = 1, 2, \ldots, J + 1$ be the point where

$$\frac{dx}{dx} P_{e|1}(x) \bigg|_{p_0(M + a_i)} = \frac{1}{2} \frac{dx}{dx} P_{e|1}(x) \bigg|_{p_0 M}.$$  

(16)

These points determine the boundaries of the optimal solution in the following lemma.

**Lemma 3:** The optimal $\Delta^*_i$ belongs to the interval $[a_{i+1}, a_i]$ for $i = 1, 2, \ldots$.

**Remark 2:** The above property helps us to provide upper and lower bounds on the minimum probability of error (see Section IV-A1).

**Remark 3:** Substituting the optimal $\Delta^*_i$s in (7) gives us the minimum probability of error for a fixed threshold receiver.

#### A. Performance Bounds

In this section, we first derive the upper and lower bounds on the probability of error, and then we characterize an upper bound on the number of RD increments.

1) **Upper and Lower Bounds on the Minimum Probability of Error:** We provide upper and lower bounds on the probability of error using Lemma 3. Let $J$ be the number of positive RD increments. Recall that $\Delta^*_i \in [a_{i+1}, a_i]$. We consider active constraints in (36), which result in a local minimum of the error probability. That is also the global minimum. Using $\Delta^*_i \in [a_{i+1}, a_i]$ and (36), we obtain $\sum_{j=1}^{J^*+1} a_j < \beta(T - T_M) \leq \sum_{j=1}^{J^*} a_j$. First, we derive an upper bound. From (7), we know that $P_{e|0}$ does not depend on $\Delta_i$, and from (12) we note that $P_{e|1}$ is a decreasing function of the number of transmitted molecules. Based on Lemma 3, we have $\Delta^*_i > a_{i+1}$ (see Fig. 4 for $n = 3$). Thus, substituting $\Delta_i = a_{i+1}$, instead of $\Delta_i = \Delta^*_i$ in (7) provides an upper bound on the minimum probability of error. We note that $\Delta^*_i < a_i$. Similarly we obtain a lower bound on the minimum probability of error by substituting $\Delta_i = a_i$, instead of $\Delta_i = \Delta^*_i$ in (7).

2) **An Upper Bound on the Error Probability of the Sub-Optimal Receiver:** For the sub-optimal receiver, it is possible that the previous bits were decoded incorrectly and we may have error propagation. This problem is ignored for the genie-aided receiver. Here, we investigate this problem and obtain an upper bound on the performance of the sub-optimal receiver.
In the following, we obtain the error probability of the system, considering the sub-optimal receiver which uses the estimated states for its decision. If the estimated state is incorrect, the receiver uses the wrong receiving interval for the next slot. Therefore, if the next bit is “1”, the molecules are probably missed, which results in a propagation error. Note that if the next bit is “0”, no molecule was transmitted and thus the receiving interval for the next slot is limited to 1.

Because of $1 - P_{eg} > 0$, we have $(1 - P_{eg})^{j+1} \geq 1 - (j + 1)P_{eg}$.

By using this inequality and substituting (19) in (18), we have

$$P(\tilde{s} \neq s) \leq \sum_{j=0}^{\infty} \frac{1}{2^{j+1}} (j + 1)P_{eg} = 2P_{eg}. \tag{17}$$

The above equation and (17) result in $\tilde{P}_e \leq 2P_{eg}$. Also, we find the error probability for the sub-optimal receiver by performing a particle-based simulation, which confirms this result.

3) An Upper Bound on the Number of Positive RD Increments: As the required memory at the transmitter is proportional to the number of positive RD increments, we are interested in bounding it.

Theorem 1: The number of positive RD increments is upper bounded as $N < \min_{m \in \{1, \ldots, J\}} \left\{ m + \frac{B_M}{a_J - m} \right\}$. 

Proof: Lemma 1 shows that RD increments are decreasing. Remind that $C_{1J}$ is an active condition of (10) (see Appendix B). Thus, from (10) and Lemma 1, we have

$$\left\{ \sum_{i=1}^{j} \Delta_i^* = B_M, \right. \left. \Delta_1^* > \Delta_2^* > \ldots > \Delta_{j-1}^* > \Delta_j^* > 0 \right\},$$

which results in $J < 1 + \frac{B_M}{a_J - 1}$. By following a similar approach and considering $\sum_{i=1}^{j-m} \Delta_i^*$ from $C_{1J}$, we obtain $N < m + \frac{B_M}{a_J - m} < m + \frac{B_M}{a_J - m + 1}$. Note that $\Delta_{j-m}^* > a_J - m + 1$. Further, from (16), the following equation can be easily obtained.

$$a_i = \frac{-T_{th}}{P_0} \left( -\tau W \left( \frac{T_{th}}{\sqrt{2(\Delta_1^* + \Delta_2^*)P_{eg}(x)}} \right) p_0 \right) - \frac{\lambda}{P_0} - M, \tag{20}$$

where $W(\cdot)$ is the Omega Function defined as $W(ze^z) = z$. ■

B. Adaptive Threshold Receiver

In no-ISI case, we want the receiver to adjust its threshold such that $P_e$ decreases. To do so, we need to know the state of the system and the number of transmitted molecules in that state. This threshold detector is sub-optimal which gives us an upper bound on the performance of the system with optimal detector, because the knowledge of different RDs (side information) may improve the performance. Since $P_e$ is low, we ignore error propagation effects. Thus, the receiver uses its estimation instead of the system state and faces a hypothesis testing problem in each state. Using the ML decoding rule in state $s_j$, results in $P_{eg}(s_j|x) = \frac{B_j}{P_0}$. Combining this inequality and (2), we obtain the sub-optimal threshold as $T_{th} = \frac{p_0(M + \Delta_i^*)}{\ln(1 + \frac{p_0(M + \Delta_i^*)}{p_0})}$. Note that the number of positive $\Delta_i^*$s is limited to $J$. So the number of different thresholds is also limited and for states $s_i$, $i = J + 1, \ldots$, the threshold is fixed.

As a result, the receiver needs a limited memory to save these
thresholds. Considering a genie-aided receiver, from (7) error probability of adaptive threshold receiver is
\[
\hat{P}_e = \sum_{y \geq T_{th}^j} e^{-\lambda} \frac{\Delta^y}{y!} + \sum_{j=1}^{\infty} \sum_{y < T_{th}^j} e^{-\left(p_0(M + \Delta_j) + \lambda\right)} \frac{\left(p_0(M + \Delta_j) + \lambda\right)^y}{y!}.
\]

Remark 4: We can optimize the RD increments and thresholds simultaneously in each state by solving the following optimization problem.
\[
\min_{\{\Delta_i\}_{i=1}^\infty} \hat{P}_e
\]
\[
\text{s.t.: } \left\{\begin{array}{l}
0 \leq \sum_{j=1}^i \Delta_j \leq B_M, \ \forall i, \\
0 < T_{th}^i < p_0(M + \Delta_i), \ \forall i
\end{array}\right.
\]

V. MAIN RESULTS IN ISI CASE

In this section, we consider the problem in the presence of ISI. First, we assume one-symbol ISI and find the optimal solution for RD increments to minimize the probability of error. Then, we consider two-symbol ISI.

A. One-Symbol ISI

If the ISI is one symbol, the received molecules at the receiver have a Poisson distribution whose parameter is a function of the current bit and the previous bit. So if a bit “0” is transmitted, the ISI is erased and received signal is only a function of the current bit. As a result, if we know the current state, we know the previous bit and we can obtain the probability of error. From (2), we have: \(Y_i \sim \text{Poisson}(p_0X_i + p_1X_i - 1 + \lambda)\). We are going to find the conditional probability of error. If we use \(P_{e|j}\) for error probability where the current bit is “0” and the previous bit is “1”, we have \(P_{e|B} = \frac{1}{2}(P_{e|0B} + P_{e|1B})\), for \(B = 0, 1\). The previous bit has two cases:

I. The previous bit is “0”: ISI is zero and we have
\[P_{e|0B} = P_{e|1B}(Bp_0(M + \Delta_1)), \text{ for } B = 0, 1.\]

II. The previous bit is “1”: ISI gets different amounts on the previous state. If the previous state is \(s_{j-1}\), the released molecule increment is \(\Delta_j\). Since the previous bit is “1”, the current state is \(s_j\) and the current released molecule increment is \(\Delta_{j+1}\). Thus, the average amount of \(P_{e|1B}, B \in \{0, 1\}\) is
\[P_{e|1B} = \sum_{j=1}^{\infty} P(s_{j-1})P_{e|s_{j-1}} = \sum_{j=1}^{\infty} \frac{1}{2} P_{e|B}(Bp_0(M + \Delta_{j+1}) + p_1(M + \Delta_j)).\]

Our goal is finding \(\Delta_i\)s to minimize \(P_e\).
\[
\min_{\{\Delta_i\}_{i=1}^\infty} P_e = \frac{1}{2} \left(P_{e|0} + P_{e|1}\right),
\]

TABLE I

| State | ISI value |
|-------|-----------|
| \(s_0 = 0\) | \(v_1 = 0\) |
| \(s_1 = 1\) | \(v_2 = p_1(M + \Delta_1)\) |
| \(s_2 = 01\) | \(v_3 = p_1(M + \Delta_2)\) |
| \(s_3 = 011\) | \(v_4 = p_1(M + \Delta_3)\) |
| \(\vdots\) | \(\vdots\) |

s.t.: \(C_1(\{\Delta_i\}_{i=1}^\infty) = B_M - \sum_{j=1}^{i} \Delta_j \geq 0, \ \forall i,\)
\[
C_{2i}(\{\Delta_i\}_{i=1}^\infty) = \sum_{j=1}^{i} \Delta_j \geq 0, \ \forall i.
\]

In Appendix E, for the general case of finite memory channel (i.e., \(\exists K \in \mathbb{N} : p_k = 0, \text{ for } i > K + 1\)), we show that the objective function in (22) is convex and therefore the optimal solution exists. As a result, this function has a minimum in the compact domain of \(\Delta_i\)s, which is the optimal solution. The Lagrangian method results in a problem, which is difficult to solve in a closed form. To simplify this problem, we write the probability of error in (22), as a function of ISI value in each state.

In no-ISI case, we defined the states as the number of transmitted bits “1” after the last bit “0”. For the case of one symbol ISI, using the same definition of states, we can determine the value of ISI, \(v_i\) for \(i = 1, 2, \ldots, \) in state \(s_{i-1}\), as given in TABLE I. Therefore, the previous definition of states is used in one symbol ISI case, with the probabilities of states given in (6), i.e., \(P(v_i) = P(s_{i-1}) = \frac{1}{2^i}\). As a result, in state \(s_{i-1}\), we choose the number of released molecules, \(l_i\), as a function of ISI value (i.e., \(l_i = M + \Delta_i = f(v_i)\)), similar to [12]. Thus, the probability of error can be written as
\[
P_e = \frac{1}{2} \mathbb{E}_I \left[P_{e|0}(I) + P_{e|1}(p_0f(I) + I)\right],
\]
where \(\mathbb{E}_I[X] = \sum_{j=1}^{\infty} P(s_{i-1})P(X|I = v_i)\). Now, we minimize the probability of error, subject to
\[
C_{1i}(\{l_i\}_{i=1}^\infty) = B_M - \sum_{j=1}^{i} (l_j - M) \geq 0, \ \forall i,\)
\[
C_{2i}(\{l_i\}_{i=1}^\infty) = \sum_{j=1}^{i} (l_j - M) \geq 0, \ \forall i.
\]

To simplify the problem, similar to [12, p. 247], we assume that \(I\) is a random variable, which takes values \(v_1, v_2, \ldots, \) in each state. Considering this assumption, the first term of (25) is fixed and only the second term depends on \(l_i\)s. Therefore, we simplify the optimization problem and obtain a sub-optimal solution by minimizing only the second term of (25) as follows.
\[
\min_{\{l_i\}_{i=0}^\infty} \sum_{j=1}^{\infty} \frac{1}{2^j} \sum_{y < T_{th}^j} e^{-(p_0l_i + v_i + \lambda)} \frac{(p_0l_i + v_i + \lambda)^y}{y!},
\]
TABLE II
STATES AND ISI VALUES FOR TWO-SYMBOL ISI CASE

| State | ISI value |
|-------|------------|
| s₀ = 0 | v₁ = 0 |
| s₁ = 1 | v₁ = p₁(M + Δ₁) |
| s₂ = 0 | v₂ = p₁(M + Δ₁) + p₂(M + Δ₂) |
| s₃ = 1 | v₂ = p₁(M + Δ₂) + p₂(M + Δ₃) |
| s₄ = 0111 | v₅ = p₁(M + Δ₃) + p₂(M + Δ₃) |

Fig. 5. Transition diagram of two-symbol ISI.

subject to (26) and (27). Using Lagrangian method, at the optimal solution, we have

$$\nabla \mathbb{E}_f \left( P_{e|1}(l_i^*, v_i^*) \right) = \sum_{k=1}^{\infty} \sum_{i=1}^{\infty} \mu_{k_i} \nabla C_{k_i}(\{l_i^*\}_{i=1}^{\infty}),$$

where \(\mu_{k_i}^*\) are Lagrangian multipliers. Due to KKT conditions, we have \(\mu_{k_i}^* C_{k_i} = 0, \forall i = 1, \ldots\). In Appendix F, we obtain a local minimum for (28), which is a global minimum (because \(P_{e|1}\) is a convex function and the domain of \(l_i^*\)s in conditions (26) and (27) is compact). Since \(l_i^* = M + \Delta_i^*\), the \(\Delta_i^*\)s are obtained from (44) as

$$\Delta_i^* = \frac{1}{p_0} \begin{ cases } p_i M + (p_0 + p_1) \Delta_{i-1}^* - p_1 \Delta_i^* & , 2 \leq i \leq J; p_i M + (p_0 + p_1 + p_2) \Delta_{i-1}^* & , i = 1; p_2 M + p_i M + (p_0 + p_1 + p_2) \Delta_{i-2}^* - p_1 \Delta_i^* - p_2 \Delta_i^* & , 2 < i \leq J, i > J. \end{ cases }$$

B. Two-Symbol ISI

We know the storage is filled when a bit “0” comes. We use different RD for each bit “1” after bit “0”. For example, if the number of transmitted “1”s after the last “0” is \(i\) bits, the released molecule increment for the next bit “1” is \(\Delta_i+1\). We update the definition of state diagram to match two-symbol ISI. We define the states as \(01 \ldots 1\) or \(01 \ldots 10\), where \(i = 1, 2, \ldots\), as shown in Fig. 5. These states show the sequence of previous transmitted bits (at least two bits) and include sufficient information about the ISI values. TABLE II shows ISI values for these states. For example, if the system is in state \(\tilde{s}_3 = 010\) and bit “1” comes, ISI takes value of \(p_1(M + \Delta_1)\) and the system goes to state \(\tilde{s}_2 = 011\).

The transition matrix of Fig. 5 is \(\tilde{\Pi} = [\pi_{ij}]\), where

$$\pi_{ij} = \begin{ cases } 1/2, & (i, j) \in C_π; 0, & \text{otherwise.} \end{ cases }$$

and \(C_π = \{(2k - 1, 1), (2k - 1, 2), (2k, 2k + 1), (2k, 2k + 2) | \forall k \in \mathbb{N}\}\).

The steady-state distribution \(P_\tilde{s} = [P_{s_1}, P_{s_2}, \ldots]\) is derived as \(P_\tilde{s} = P_\tilde{s}\tilde{\Pi}\), which results in

$$P_\tilde{s} = [1/4, 1/4, 1/8, 1/8, 1/16, 1/16, \ldots].$$

As shown in Fig. 5, in states \(\tilde{s}_1, \tilde{s}_3, \tilde{s}_5, \ldots\), the previous transmitted bit is “0”; Thus, the storage is full and we choose released molecule increment \(\Delta_1\) in these states (if a bit “1” comes in these states, the released number of molecules are \(l_1 = l_2 = l_3 = \ldots = M + \Delta_1\)). In states \(\tilde{s}_2, \tilde{s}_4, \tilde{s}_6, \ldots\), the number of transmitted bits “1” after the last bit “0” are respectively \(1, 2, 3, \ldots\); Thus, we choose released molecule increments \(\Delta_2, \Delta_3, \Delta_4, \ldots\) in these states, respectively (i.e., \(l_2 = M + \Delta_2, l_4 = M + \Delta_3, l_6 = M + \Delta_4, \ldots\)). Similar to one-symbol ISI case, again we assume that \(I\) is a random variable, which takes values \(v_1, v_2, \ldots\), in states \(\tilde{s}_1, \tilde{s}_2, \ldots\), and released number of molecules in these states are a function of ISI (i.e., \(l_i = f(v_i)\)). Using these assumptions, we write the probability of error as (25) and only minimize the second term as:

$$\mathbb{E}_f \left( P_{e|1}, I \right) = \sum_{i=1}^{\infty} \sum_{j=1}^{\infty} P_{s_i} P_{e|1}(l_i^*, v_i^*)$$

$$= \sum_{i=1}^{\infty} \left( P_{s_{2i-1}} P_{e|1}(l_i^*, v_{2i-1}) + P_{s_{2i}} P_{e|1}(l_i^*, v_{2i}) \right).$$

C. Adaptive Threshold Receiver in the Presence of ISI

So far, we consider a fixed threshold receiver and obtain the optimal RDs in each state. These optimal RDs can be used to update the threshold in each state. If we assume a genie-aided receiver, which correctly decodes the received bits, we can obtain the best threshold in state \(s_i\). If the next state after coming a bit “1” is \(s_j\), then we have \(P(y|s_i) \geq 0\), and thus

$$\frac{p_0 l_j + v_i + \lambda}{v_i + \lambda} T_{th}^i \geq e^{p_0 l_j}. \text{ Therefore, threshold is}$$

$$T_{th}^i = \left( \frac{p_0 l_j}{\ln(1 + \frac{p_0 l_j}{v_i + \lambda})} \right).$$

The receiver which save these adaptive thresholds in its memory and uses \(T_{th}^i\) in state \(s_i\), is called adaptive threshold receiver.
VI. NUMERICAL AND SIMULATION RESULTS

In this section, we provide numerical and simulation results to show the performance improvement achieved by the proposed adaptive RD scheme. We choose system parameters as $r_R = 3.33 \times 10^{-7} \text{ m}$, $d = 10^{-5} \text{ m}$, $D = 9.31 \times 10^{-9} \text{ m}^2/\text{s}$, $\beta = 50 \text{ molecule/sec}$, and $T_M = 4 \text{ s}$, which are the radius of the receiver, distance of the transmitter and the surface of the receiver, diffusion coefficient, the molecule production rate, and non-adaptive RD, respectively. We assume slot duration for no-ISI case as $T = 45$, for one-symbol ISI case as $T = 25$ and for two-symbol ISI case as $T = 20$. First, we compare the performance of four different strategies in terms of probability of error for no-ISI case.

- **Strategy 1 (non-adaptive):** RD is fixed and the receiver uses a fixed threshold (probability of error is derived in (5)).
- **Strategy 2:** RDs are optimal and the receiver uses a fixed threshold (probability of error is discussed in Remark 3).
- **Strategy 3:** RDs are optimal and the receiver uses these optimal RDs to obtain adaptive thresholds in one iteration such that $P_e$ decreases (probability of error is derived in (21)).
- **Strategy 4:** RDs and thresholds are optimized simultaneously, i.e., in several iterations such that the probability of error converges (probability of error is discussed in Remark 4).

For the particle based simulation, we follow an algorithm similar to [23]. For each amount of $\lambda$, we generate $N$ random bits, where $N \approx \frac{100}{\lambda}$. In each state, the transmitter releases molecules with pulse $\Pi(t, T_M + \tau_1)$, where $\tau_1$ has been obtained analytically. For each molecule, we use the hitting probability in (1). Then, the receiver counts the number of received molecules, during its receiving intervals with time step size of $dt = 10^{-4}$.

Fig. 6 shows the probability of error versus the noise parameter for the mentioned strategies. Since finding optimal RD increments needs to solve the optimization problem in (9), upper and lower bounds in Section IV-A1 respectively are plotted without solving this optimization problem and only by using the interval of optimal solution. It can be seen that these two bounds are very close to each other. As one can see, strategy 1, which is the simplest technique for transmission and reception, can be improved by changing the RDs and/or thresholds. Because more molecules are used in the adaptive strategy. Though the complexity of the system increases compared to non-adaptive strategy, we have shown that the transmitter and receiver only need limited memories to save the amounts of RD increments and adaptive thresholds. For example, there is twelve positive optimal RD increments that the transmitter saves these numbers in its memory. Also the number of adaptive threshold that the receiver saves in its memory is twelve numbers.

Fig. 7 shows the probability of error versus the storage capacity for the above four strategies. As one can see, in strategy 1, the storage capacity has no effect on the system performance while for three other methods, increasing storage capacity improves the system performance. As shown in Fig. 7, for strategies 3 and 4 (which we change both RDs and thresholds), with increasing storage capacity, $P_e$ decreases exponentially. Fig. 7 shows that changing the RDs and thresholds (strategies 3 and 4) significantly improve the probability of error.

In Fig. 8, we show the upper bound on the number of positive RD increments versus storage capacity.
limitation. It shows that the optimal transmission rates are chosen such that the received signal for bit “1” has a fixed rate for almost all values of ISI. For a transmitter with limited molecule production rate and storage capacity, when ISI is zero ($I = 0$), the received rate cannot be more than $p_0 M$. Thus, the maximum fixed rate of molecules that can be received is

$$p_0 X_i + I = p_0 M. \quad (35)$$

We simplify this method for our transmitter with limitations in one-symbol ISI case and find a closed form for the transmission rates (see Appendix H). Now, we compare strategies 1, 4 and the following strategies:

- **Strategy 5**: The method proposed in [12]; RDs are such that the received signal has a fixed rate and the receiver uses a fixed threshold.

- **Strategy 6 (sub-optimum strategy)**: RDs are sub-optimum as in (30) and (33) for one and two-bit ISI, respectively. The receiver uses adaptive thresholds in (34).

Fig. 9 shows the probability of error versus the noise parameter for these four strategies in a channel with one-symbol ISI. For strategy 5, we plot the upper and lower bounds on the probability of error (derived in (48) and (49), respectively). It can be seen that the performance of strategy 1 and strategy 5 is almost the same. Because, strategy 5 forces the transmission scheme to have a fixed rate in (35) at the receiver. Thus, it forces the transmitter to decrease release rate (i.e., $X_i < M$) when ISI is not zero. It means that the transmitter uses less number of molecules to send bit “1” (it reduces both signal and ISI), and the probability of error does not change considerably. Our proposed strategy in both optimum (strategy 4) and sub-optimum cases (strategy 6) significantly outperform strategies 1 and 5, because strategy 5 is not proposed for a system with transmitter’s limitations (limited molecule production rate and finite storage capacity). Therefore, considering these limitations in designing the modulation scheme, we can significantly improve the performance. In this figure, the particle based simulation is also provided for strategies 1 and 4, which confirms the analytical results.

**VII. Conclusion**

In this paper, the transmitter’s limitations are studied in MC, in particular the limited molecule production rate and finite storage capacity. By changing the duration in which molecules are released, an adaptive RD modulation is proposed. The objective is finding the optimal transmission RDs to minimize the probability of error. Important properties of the optimal solution are proved and it is shown that optimal RDs are decreasing. Using these properties, upper and lower bounds are derived on the minimum probability of error. It is shown that increasing the storage capacity significantly improves the system performance in terms of probability of error. It is also shown with analytical and simulation results, that an adaptive threshold receiver performs better than a fixed threshold receiver. The proposed modulation scheme also performs better than transmission method proposed in [12]. Because this method is not proposed for a system with transmitter’s limitations. While in our proposed strategy, the complexity of the system is increased compared to non-adaptive strategy, we show that the required memories for the proposed transmitter and receiver are limited.

**APPENDIX A**

**Convexity of Probability of Error in (8)**

Consider $F(\cdot)$ in (8). We have $\frac{d^2}{d\Delta_i d\Delta_j} F(\Delta_1, \Delta_2, \ldots) = 0$ for $i \neq j$ and $\frac{d^2}{d\Delta_i^2} F(\Delta_i) = \sum_{i=1}^{M} \frac{p_0}{2} g(\Delta_i)$, where $g(\cdot)$ is
defined as
\[ g(x) = e^{-y} y^{T_{th} - 1} \frac{(y - T_{th})}{T_{th}} \bigg|_{y=p_0(M+x)+\lambda}. \]

Since \( p_0(M+x)+\lambda > T_{th} \), \( g(\Delta_j) > 0 \), for \( i = 1, \ldots, J \). Thus, \( F(\cdot) \) is convex. Therefore, \( P_{e1} \) is also a convex function.

**APPENDIX B**

**PROOF OF LEMMA 1**

We propose a solution for (9) that satisfies all conditions in (10) and (11). Assume only \( C_{1i} = 0 \), for \( i \geq J \) (\( C_{2i} \neq 0 \)) and \( \mu^*_{1i} = 0 \) for \( i > J \). Thus, the conditions result in

\[
\begin{cases}
\beta(T - T_{M}) - \sum_{j=1}^{J} \Delta_j^* > 0, & \text{for } i < J, \\
\beta(T - T_{M}) - \sum_{j=1}^{J-1} \Delta_j^* = 0, & \text{for } j > J.
\end{cases}
\]

(36)

First, note that a solution subject to the conditions in (36) satisfies the conditions in (10) and (11). The above assumptions and (15) result in

\[
\left( \frac{\partial F}{\partial \Delta_1^*}, \ldots, \frac{\partial F}{\partial \Delta_J^*} \right)^T = \mu^*_{1J} \langle -1, \ldots, -1, 0, \ldots \rangle^T,
\]

(37)

where \( \mu^*_{1J} > 0 \) due to KKT conditions. Since the objective function in (9) is convex and solution of (36) satisfies the conditions in (10) and (11), this solution is the global minimum.

From (37), we obtain

\[
\frac{d}{d \Delta_k} P_{e1} = \frac{d}{d \Delta_k} P_{e1} \bigg|_{p_0(M+\Delta_k^*)} = 2^{k-h} \frac{d}{d \Delta_k} P_{e1} \bigg|_{p_0(M+\Delta_k^*)}. 
\]

(38)

Since \( P_{e1} \) is a convex function, for \( k > h \), (38) results in \( \Delta_k^* < \beta_k^* \), which means that the optimal RD increments are decreasing. As a result if one of them (\( \Delta_m^* \)) is negative, then the \( \{ \Delta_j^* \}_{m,J} \) are negative too. (36) results in \( \beta(T - T_{M}) - \sum_{j=1}^{J-1} \Delta_j^* = \Delta_J^* < 0 \), which is in contradiction with (36). Thus, all \( \Delta_i^* \)s are nonnegative.

**APPENDIX C**

**PROOF OF LEMMA 2**

According to Lemma 1, the optimal RD increments are decreasing and the optimal solution satisfies (36). Assume that \( C_{1J} \) is active. Thus \( \sum_{j=1}^{J} \Delta_j^* = \beta(T - T_{M}) \), and \( \Delta_j^* = 0 \), for \( j > J \). If the number of positive \( \Delta_j^* \)s (J) is unlimited, (36) forces

\[ \lim_{j \to \infty} \Delta_j^* = 0, \]

(39)

and \( \Delta_{j-1}^* > 0 \). From (8) and (37), we obtain

\[
e^{-p_0 \Delta_{j-1}} \frac{P_{e1}(p_0(M+\Delta_{j-1}^*) + \lambda)^{T_{th}}}{T_{th}!} \]

\[ = \frac{e^{-p_0 \Delta_{j-1}}}{2} \frac{P_{e1}(p_0(M+\Delta_{j-1}^*) + \lambda)^{T_{th}}}{T_{th}!}. \]

From (39) and the above equation, we have \( e^{-\Delta_{j-1}} = \frac{1}{2} \frac{P_{e1}(p_0(M+\Delta_{j-1}^*) + \lambda)^{T_{th}}}{T_{th}!} < \frac{1}{2} \), which means that \( \Delta_{j-1} > \ln 2 \). Thus, \( \lim_{j \to \infty} \Delta_{j-1}^* \neq 0 \). This contradicts (39) and completes the proof.

**APPENDIX D**

**PROOF OF LEMMA 3**

Consider the smallest optimal positive released molecule increment as \( \Delta_j^* \). Remind that \( \Delta_j^* = 0 \) for \( j > J \). We prove that \( \Delta_j^* \in [0, a_j] \). We use contradiction. Suppose \( \Delta_j^* \notin [0, a_j] \), i.e., \( \Delta_j^* > a_j \). This scheme is referred as scheme A.

- **Scheme A:** Choose the smallest optimal positive \( \Delta_J = \Delta_J^* \) in state \( s_J \) which \( \Delta_J > a_J \) and \( \Delta_J = 0 \) in state \( s_j \) for \( j > J \). So condition \( C_{1J} \) is active: \( \sum_{j=1}^{J} \Delta_j^* + \Delta_J = \beta(T - T_{M}) \). The probability of error for scheme A is

\[
P_{e1} = \frac{1}{2} \left( \frac{P_{e1}}{p_0(M+\Delta_j^*)} \right) + \frac{1}{2} \left( \frac{P_{e1}}{p_0(M+\Delta_J^*)} \right).
\]

Now, we show that there is a better scheme of transmission which has a less probability of error.

- **Scheme B:** Consider real numbers \( b_1, b_2 \in \mathbb{R} \) (will be derived later in text). Choose \( \Delta_J = b_2 \) in state \( s_J \), \( \Delta_{J-1} = b_1 \) in state \( s_{J-1} \), and \( \Delta_J = 0 \) in state \( s_j \) for \( j > J + 1 \). So condition \( C_{1J+1} \) is active: \( \sum_{j=1}^{J} \Delta_j^* + b_1 + b_2 = \beta(T - T_{M}) \). The probability of error for scheme B is

\[
P_{eB} = \frac{1}{2} P_{e1} + \frac{1}{2} \left( \frac{P_{e1}}{p_0(M+\Delta_j^*)} \right) + \frac{1}{2} P_{e1} + \frac{1}{2} \left( \frac{P_{e1}}{p_0(M+\Delta_J^*)} \right)
\]

It is shown in Appendix D-A that positive numbers \( b_1 \) and \( b_2 \) exist, which satisfy \( \Delta_j^* = b_1 + b_2 \) and the following property (see Fig. 11).

\[
\frac{d}{dx} P_{e1} \bigg|_{p_0(M+b_1)} = \frac{2}{2} \frac{d}{dx} P_{e1} \bigg|_{p_0(M+b_1)}.
\]

(40)

The result of subtraction is

\[
P_{eA} - P_{eB} = \frac{1}{2} \left( \frac{P_{e1}}{p_0(M+\Delta_j^*)} - \frac{P_{e1}}{p_0(M+b_1)} \right) + \frac{1}{2} \left( \frac{P_{e1}}{p_0(M+\Delta_J^*)} - \frac{P_{e1}}{p_0(M+b_1)} \right)
\]

\[ > \frac{1}{2} \left( \frac{P_{e1}}{p_0(M+\Delta_j^*)} - \frac{P_{e1}}{p_0(M+b_1)} \right) + \frac{2}{2} \left( \frac{P_{e1}}{p_0(M+\Delta_J^*)} - \frac{P_{e1}}{p_0(M+b_1)} \right), \]

where (a) follows from the convexity of the probability of error (see Appendix A) and its decreasing property (see (12)). Combining (40) and the above equation, results in \( P_{eA} - P_{eB} > 0 \), which means the error probability of scheme B is less than scheme A. It means scheme A cannot be optimal. So, \( \Delta_j^* \notin [0, a_j] \) and from (37), \( \Delta_j^* \notin [a_j+1, a_j] \) for \( j = 1, \ldots, J - 1 \).
A. Proof of Existence of Points $b_1$ and $b_2$

Suppose that $\Delta^* > a_J$. Let us define

$$h(x) = \frac{d}{dx} P_e[x] \bigg|_{x_0(\Delta^*-a_J)} - \frac{1}{2} \frac{d^2}{dx^2} P_e[x] \bigg|_{x_0(\Delta^*+a_J)}.$$ 

We want to show that $h(x)$ has a root in the interval $[0, \Delta^*-a_J]$. Consider two points $x_1 = 0$ and $x_2 = \Delta^*-a_J$.

$$h(x_1) = \frac{d}{dx} P_e[x] \bigg|_{x_0(\Delta^*-x_0)} - \frac{1}{2} \frac{d^2}{dx^2} P_e[x] \bigg|_{x_0(\Delta^*+x_0)}.$$ 

Combining (12), (16) and the above equation results in $h(x_1) > 0$. Similarly, we obtain $h(x_2) < 0$. These inequalities, say that sign of the continuous function $h(x)$ at the sides of interval $[0, \Delta^*-a_J]$ changes. According to the Bolzano’s theorem, $\exists 0 < x_0 < \Delta^*-a_J : h(x_0) = 0$, the above equation means

$$\frac{d}{dx} P_e[x] \bigg|_{x_0(\Delta^*-x_0)} - \frac{1}{2} \frac{d^2}{dx^2} P_e[x] \bigg|_{x_0(\Delta^*+x_0)} = 0.$$ 

Using the above equation, we define $b_1 = x_0$, $b_2 = \Delta^*-x_0$.

APPENDIX E

CONVEXITY OF $P_e$ IN THE PRESENCE OF ISI

To show that $P_e$ is a convex function of its arguments, we obtain the Hessian of $P_e$ and check whether or not it is positive semidefinite (PSD). Considering the definition of $s_t$, as the number of “1”s after the last bit “0”, we obtain

$$H(P_e) = \sum_{t=1}^{K} \frac{1}{2^{t+1}} (H(P_{e|s=0}) + H(P_{e|s=1})).$$ 

(41)

where $H(.)$ is the Hessian of the function. In the following, we show that both $H(P_{e|s=0})$ and $H(P_{e|s=1})$ are PSD. Therefore, $H(P_e)$ is PSD too. To do this, we have $H(P_{e|B}) = \frac{d^2}{d \Delta_i d \Delta_j} P_{e|B}$, $i,j=1,\ldots,K+1$, for $B = 0, 1$. If the number of positive $\Delta_i$s equal to $K$, we have

$$\frac{d^2}{d \Delta_i d \Delta_j} P_{e|s=0} = \sum_{t=1}^{K} \frac{1}{2^{t+1}} \frac{d^2}{d \Delta_i d \Delta_j} P_{e|s=1} + \frac{d^2}{d \Delta_i d \Delta_j} P_{e|s=1},$$ 

where from (14) we have

$$\frac{d^2}{d \Delta_i d \Delta_j} P_{e|s=1} = c_i c_j e^{-(c_0 M + \sum_{k=1}^{K} c_i \Delta_i + \lambda)} \times \frac{T_h - (c_0 M + \sum_{k=1}^{K} c_i \Delta_i + \lambda)}{T_h!}.$$ 

For $w_0$, we have $c_0 M + \cdots + c_K \Delta_K + \lambda < T_h$ and $c_i, c_j \geq 0$. So

$$h_{ij} = \frac{d^2}{d \Delta_i d \Delta_j} P_{e|s=0} \bigg|_{i,j=1} = c_i c_j P^+_{u_0}(w_0) \geq 0,$$

where $F^+_{u_0}(w_0)$ is a positive function. The Hessian matrix of $P_{e|s=0}$ is $H(P_{e|s=0}) = [h_{ij}]_{i,j=1,\ldots,K}$, where $h_{ij}$s are defined in the above equation. We can see that $X^T H(P_{e|s=0}) X = P^+_{u_0}(w_0)((c_1 X_1 + \cdots + c_K X_K)^2) \geq 0$. As a result, $H(P_{e|s=0})$ is a PSD matrix. Similarly, we can show that $H(P_{e|s=1})$ is also PSD. From (41), $H(P_e)$ is a sum of PSD matrices. Thus, it is also a PSD matrix. Therefore, $P_e$ is a convex function of $\{\Delta_i\}_{i=1}^{K}$.

APPENDIX F

SUB-OPTIMAL SOLUTION FOR ONE-SYMBOL ISI

A local minimum of (28) can be found if $C_{1i}$ is active for $i = J, J+1, \ldots$, (J is the number mentioned in Lemma 2), and $\mu^*_{1i} = \mu^*_{2i} \geq 0$ for $i = J+1, \ldots$. Therefore, $C_{2i}$ is active for $i = J+1, \ldots$, i.e., $l_i = M$ and

$$v_i = p_i M = v_{j+1}, \text{ for } i = J+1, \ldots$$

(42)

Thus, $\mu^*_{1i} = 0$ for $i = 1, \ldots, J$ and $\mu^*_{2i} = 0$ for $i = 1, \ldots, J$.

If we define $U_i = [u_i(1), u_i(2), \ldots, u_i(M)]^T$, where

$$u_i(j) = \begin{cases} 1, & j \leq i, \\ 0 & j > i, \end{cases}$$

then, (29) results in $\nabla v_i^T [P_{e|1}(p_i l_i^* + v_i^*)] = \mu^*_{i,j} U_i$.

Combining (29) and the above equation results in

$$\frac{1}{M^2} \sum_{j=1}^{M} \frac{d}{dv_i} P_{e|1}(p_i l_i^* + v_i^*) = \mu^*_{i,j} \frac{U_i}{p_i},$$

(43)

Form (42), we have $P(I = v_{j+1}) = \sum_{j=J+1}^{J+1} P(s_{J+1} = \frac{1}{2^J})$. As a result, ISI takes $J + 1$ different values. Comparing (43) with (37), we conclude that these problems have same solution, which is the optimal solution of no-ISI case. If we denote the hitting probabilities and optimal released molecule increments in no-ISI case by $p_{i,\text{no-ISI}}$ and $\Delta^*_{i,\text{no-ISI}}$, respectively, the solution of (43) will be $p_{0i}^* + v_{i}^* = p_{0,\text{no-ISI}}(M + \Delta^*_{i,\text{no-ISI}})$, for $i = 1, \ldots, J$. The number of received molecules in these two cases must be equal. Thus, $p_{0,\text{no-ISI}} = p_{0} + p_{1}, p_{i,\text{no-ISI}} = 0, i \geq 1$. Recall that $v_i$ is the ISI value in state $s_0$ (the previous bit is “0”). Thus, $v_i^* = 0$ and from the above equation, $l_i^* = (1 + \frac{p_i}{p_0})(M + \Delta^*_{i,\text{no-ISI}}).$ For $i \geq J + 1$, we have $l_i = M$ and for $2 \leq i \leq J$, we have $v_i^* = p_i l_i^* - 1$ and $l_i^*$ is recursively obtained as

$$l_i^* = \begin{cases} 1 + \frac{p_i}{p_0}(M + \Delta^*_{i,\text{no-ISI}}) & i = 2, \ldots, J, \end{cases}$$

(44)

APPENDIX G

SUB-OPTIMAL SOLUTION FOR TWO-SYMBOL ISI

If we assume that $v_{2i-1}$s for $i = 1, 2, \ldots$ are equal ($v_{2i-1} = v_1$ for $i = 1, 2, \ldots$), and substitute their average value in non-adaptive strategy instead of $v_1$, we can simplify (32) and obtain a sub-optimal solution ($l_i^*$s), $v_1 = \frac{\sum_{i=1}^{\infty} v_{2i-1} P_{2i-1}^*}{\sum_{i=1}^{\infty} P_{2i-1}^*} = \frac{p_0 M}{2}$.
the above equation and (31) in (32) results in the following minimization problem:

$$\min_{\{l_i\}_{i=1}^{\infty}} E_I \left[ P_{e|1, I} \right] = \frac{1}{2} P_{e|1} (p_0 l_1 + v_2 l_{i-1})$$

$$+ \sum_{i=1}^{\infty} \frac{1}{2} l_{i+1} P_{e|1} (p_0 l_{2i} + v_2 l_{i}),$$

subject to (26) and (27). Using Lagrangian method, results in

$$\nabla E_I \left[ P_{e|1, I} \right] = \frac{2}{\lambda} \sum_{k=1}^{\infty} \mu_{k}^* \nabla C_{k} \left( \{ l_{i}^* \}_{i=1}^{\infty} \right).$$

(45)

where $\mu_{k}^*$s are Lagrangian multipliers. Due to KKT conditions, we have $\mu_{k}^* C_{k} = 0$, $\forall i$.

A local minimum of (28) is obtained if $C_{k} = 0$ for $i \geq J$ (the number mentioned in Lemma 2), and $\mu_{k}^* = \mu_{k}^* \geq 0$ for $i > J + 1$. Therefore, $C_{k} = 0$ is active for $i > J + 1$ (i.e., $l_i = M$ and $v_i = p_1 M$, for $i > J+1$). So $\mu_{k}^* = 0$ for $i = 1, \ldots, J$ and $\mu_{k}^* = 0$ for $i = 1, \ldots, J$. Thus, (45) results in the same equation stated in (43). Comparing (43) and (37), we conclude that these equations have the same solution which is the optimal solution of no-ISI case. Therefore, the solution of two-bit ISI case is

$$p_0 l_i^* + v_i^* = p_0, n-o-SI \left( M + \Delta_{i, n-o-SI} \right),$$

for $i = 1, \ldots, J$,

where $p_0, n-o-SI = p_0 + p_1 + p_2$. Recall that $v_i$ is the ISI value in state $s_i$ (the previous bits are "00"). Thus, $v_i^* = p_2 M/2$ and from the above equation, $l_i^* = (1 + (p_1 + p_2)) (M + \Delta_{i, n-o-SI})$, $\Delta_{i, n-o-SI} = \frac{p_2}{p_0} M$. For $2 \leq i \leq J$, we have $v_i^* = p_1 l_{i-1} + p_2 l_{i-2}$ and $l_i^* = l_{i-1}^* + p_2 l_{i-2}^*$ is recursively obtained as

$$l_i^* = \left( 1 + \frac{p_1 + p_2}{p_0} \right) \left( M + \Delta_{i, n-o-SI} \right) - \frac{p_1 l_{i-1}^*}{p_0} - \frac{p_2}{p_0} l_{i-2}^*.$$  

(46)

**APPENDIX H**

**CLOSED FORM RATES OF ONE-SYMBOL ISI IN [12]**

In [12], if the transmitter starts with a “0” bit, $X_1 = 0$ (state $s_0$) and $X_2 = B_2 M$ (state $s_{B_2}$). For $i \geq 3$ if $B_i = 0$, the ISI resents (state $s_0$) and if $B_i = 1$ (state $s_1$), we have $X_i = M - \frac{p_1}{p_0} X_{i-1}$. We simplify $X_i$ in state $s_j$ recursively as $X_{i,j} = B_i M \sum_{k=0}^{j-1} (-\frac{p_1}{p_0})^k$. The conditional probability of error is $P_{e|1, s_j} = P_{e|1} (p_0 M), \forall j \in \mathbb{N}$, and we have $P_e|0, s_j = P_e|0 (p_1 X_{i,j})$. Using the probability of states in (6) causes $P_e|0 = \sum_{i=0}^{\infty} \left( \frac{1}{2} \right)^{i+1} (P_e|0 (p_1 X_{i,j})).$ $P_{e|1}$ is a constant function but $P_{e|0}$ is an increasing function of its argument. Because $\frac{d}{dx} P_{e|0} (x) = e^{-(x+\lambda)} \frac{\lambda^2 x}{e^{2x}} > 0$, we use this result to update and lower bounds on $P_{e|0}$.

**I. Upper Bound:** if $B_1 = 1$, we have

$$X_{i,j} < M \left( 1 + \cdots + (-\frac{p_1}{p_0})^{2k} \right) = 2k, \quad 0 < 2k < j, k \in \mathbb{N}$$

According to the above inequality, other terms ($j > 2k$) are less than an even term. Therefore, we have

$$P_e = \frac{1}{2} \left( P_{e|00} + P_{e|10} + P_{e|1} \right)$$

$$= \frac{1}{2} \left( \frac{1}{2} \left( P_{e|00} (0) + \sum_{j=1}^{2} \frac{1}{2^j} P_{e|00} (p_1 X_{i,j}) \right) \right) + \frac{1}{2} P_{e|1}$$

$$< \frac{1}{4} P_{e|00} (0) + \sum_{j=1}^{2k} \frac{1}{2j+2} P_{e|00} (p_1 X_{i,j})$$

$$+ \frac{1}{2} P_{e|1}$$

$$\left( p_1 X_{2k} \right) + \frac{1}{2} P_{e|1} = P_{e, 2k}.$$ 

(47)

**II. Lower Bound:** if $B_1 = 1$, we have

$$X_{i,j} > M \left( 1 + \cdots + (-\frac{p_1}{p_0})^{2k-1} \right)$$

$$= X_{2k-1}, \quad 0 < 2k < j, k \in \mathbb{N}.$$ 

According to the above inequality, other terms ($j > 2k - 1$) are more than an odd term. Therefore, using (47) we have

$$P_e > \frac{1}{4} P_{e|00} (0) + \sum_{j=1}^{2k-1} \frac{1}{2j+2} P_{e|00} (p_1 X_{i,j})$$

$$+ \frac{1}{2} P_{e|1} = P_{e, 2k-1}.$$ 

(49)

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