What the One Word “Luminescence” Hides Is a Factor of a Hundred Million in Time

— A fluorescence lifetime of 10 ns against a phosphorescence lifetime of 1 s is a ratio of 1.000000×10^{8} — the separator is whether the spin flips — [Paper 668]

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— *From thinking to syncing.* —

Abstract

Fluorescence and phosphorescence are both called luminescence. Between them lies a factor of a hundred million in lifetime.How that single difference shows up in brightness, in fragility, and in sensitivity to oxygen and temperature is counted.No new theorem or law is claimed. First, the gap is in the lifetime——a fluorescence lifetime of 10 ns against a phosphorescence lifetime of 1 s is a ratio of 1.000000×10^{8} (Section 2). **Second, and this is the core. The same number of molecules differ a hundred millionfold in initial brightness——with 10^{18} molecules excited at once, photons leave at 1.000000×10^{26} per second for fluorescence and 1.000000×10^{18} per second for phosphorescence. The total is the same; the way it is handed out is not** (Section 2). **Third, the share emitted in the first second differs too——fluorescence emits essentially all of it (1.000000×10^{18} photons), phosphorescence 6.321206×10^{17}, a share of 0.632121** (Section 2). **Fourth, a long lifetime is fragile——with a nonradiative path of 1000 per second, the quantum yield stays at 0.999990 for fluorescence and falls to 0.000999 for phosphorescence** (Section 3). **Fifth, oxygen and temperature also take only the long one——with dissolved oxygen the surviving share is 0.994431 for fluorescence against 1.785711×10^{-6} for phosphorescence, and at 298 K a barrier of 30 kJ/mol brings the phosphorescence yield to 1.813085×10^{-7}, while at 77 K it is essentially 1** (Section 3). **Sixth, the separator is whether the spin flips——transitions that keep the spin are fast and those that flip it are slow, and that difference in speed makes all four differences above** (Section 4). **Keywords**: fluorescence, phosphorescence, emission lifetime, intersystem crossing, quantum yield, oxygen quenching.

Scope of this paper (scope note): **No new theorem or law is claimed——allowed and forbidden transitions, the orders of magnitude of fluorescence and phosphorescence lifetimes, the definition of quantum yield, quenching by oxygen and the Stern--Volmer relation are all standard (Lakowicz 2006, Turro and colleagues 2010).Rate constants are not derived——where intersystem crossing and radiative rates come from is not the subject. Lifetimes are taken as given and everything downstream is counted.Representative values are used——lifetimes of 10 ns and 1 s, a nonradiative rate of 1000 per second, the oxygen concentration and quenching constant, and a barrier of 30 kJ/mol are representative, not measurements of a particular molecule.No material is designed——the design of persistent phosphors or of light-emitting devices, and which molecule to choose, are not the subject.Other emissions are not treated——thermally activated delayed fluorescence, triplet-triplet annihilation and chemiluminescence are not included.Relation to earlier papers**: **Paper 448 showed that the energy of a single photon decides whether a bond can be broken, and stated that it did not discuss mechanism, excited-state lifetimes, intersystem crossing or the role of oxygen——this paper takes that seat: where Paper 448 counted the entrance, the energy of one photon absorbed, this counts the exit, the time taken to emit.**What is added is **turning the ratio of lifetimes into a ratio of brightness**, **setting the effect of one and the same quencher on both yields side by side**, **putting numbers on oxygen and temperature taking only the long-lived side**, and **placing the separator on whether the spin flips**.

1. Notation and the Question

Setting: **let k_r be the rate at which an excited molecule emits light and k_{nr} the rate at which it returns without emitting. The lifetime and the quantum yield are**

$$\tau = \frac{1}{k_r + k_{nr}}, \qquad \Phi = \frac{k_r}{k_r + k_{nr}}$$

In fluorescence the molecule returns without the spin flipping; in phosphorescence it returns from a state in which the spin has flipped. The latter is forbidden, and its k_r is small.

Question: **under the one word “luminescence”, what differs between the two, and where does that difference appear?**

2. What Follows from a Hundred Million in Lifetime

Computation 2.1: **rates and emission times were derived from the lifetimes.**

	lifetime	half emitted by	99 per cent by
fluorescence	10 ns	6.931472×10^{-9} s	4.605170×10^{-8} s
phosphorescence	1 s	6.931472×10^{-1} s	4.605170 s

Every row stands in the ratio 1.000000×10^{8}.

Computation 2.2: with 10^{18} molecules excited at once, the initial brightness and the number of photons emitted in the first second were derived.

	initial brightness (photons per second)	photons in the first second
fluorescence	1.000000×10^{26}	1.000000×10^{18}
phosphorescence	1.000000×10^{18}	6.321206×10^{17}

Phosphorescence emits a share of 0.632121 in the first second, and the rest trails afterwards.

> **Observation 2.3**
>
> **this is the core: the total is the same and the handing out is not.Even when the same number of molecules emit the same number of photons, fluorescence packs them into 10 ns and phosphorescence spreads them thinly over 1 s——which is why one is dazzling for an instant and the other is dim but visible in the dark. Brightness and total are different quantities.**

3. A Long Lifetime Is Fragile

Computation 3.1: the quantum yield was derived with a nonradiative path of 1000 per second.

	without the nonradiative path	with 1000 per second
fluorescence	1	0.999990
phosphorescence	1	0.000999

One and the same obstacle leaves one almost untouched and takes three orders from the other.

Computation 3.2: with dissolved oxygen (concentration 2.8×10^{-4} M, quenching constant 2×10^{9} per molar per second), the surviving share is 0.994431 for fluorescence and 1.785711×10^{-6} for phosphorescence.

Computation 3.3: with a nonradiative process behind a barrier of 30 kJ/mol, its rate is 4.459015×10^{-9} per second at 77 K and 5.515462×10^{6} per second at 298 K. The phosphorescence yield is essentially 1 at 77 K and 1.813085×10^{-7} at 298 K, while fluorescence keeps 0.947728 even at 298 K.

> **Observation 3.4**
>
> the fragility comes from time, not from temperament.Phosphorescence is not sensitive to oxygen, or extinguished by warmth, because its molecules are especially fond of oxygen. It waits a hundred million times longer, so it has a hundred million times more opportunities to leave by another road——one difference in time shows up as three fragilities.

4. The Separator

> **Observation 4.1**
>
> the separator is whether the spin flips.A transition that returns with the spin unchanged is allowed and fast; one that returns from a flipped state is forbidden and slow——the one word “luminescence” divides at exactly this point, not by colour and not by brightness, but by whether the transition is allowed.

> **Observation 4.2**
>
> beside Paper 448.Paper 448 showed that the energy of a single incoming photon decides whether a bond can break (a 450 nm photon carries 265.8368 kJ/mol, a 550 nm one 217.5028)——there the entrance was counted; here the exit. The same molecules, and the quantity that acts is not energy but time.

> **Observation 4.3**
>
> what is not said.The origin of the rate constants is not derived and the lifetimes are representative; delayed fluorescence and triplet-triplet annihilation are not treated——what is shown is that one difference in time echoes, by the same factor, through brightness and fragility alike.

5. Roll Call and Closing

1. **lifetimes in the ratio 1.000000×10^{8}** (Computation 2.1): **10 ns against 1 s.**
2. **1.000000×10^{26} against 1.000000×10^{18}** (Computation 2.2): **the core.**
3. **0.632121 in the first second** (Computation 2.2): **the long one trails.**
4. **fluorescence yield 0.999990** (Computation 3.1): **the obstacle does not bite.**
5. **phosphorescence 0.000999** (Computation 3.1): **three orders, same obstacle.**
6. **1.785711×10^{-6} with oxygen** (Computation 3.2): **only the long one.**
7. **fluorescence keeps 0.994431** (Computation 3.2): **the same oxygen.**
8. **1.813085×10^{-7} at 298 K** (Computation 3.3): **essentially 1 at 77 K.**
9. **time makes the fragility** (Observation 3.4): **a hundred million chances.**
10. **whether the spin flips** (Observation 4.1): **the separator; Paper 448.**

Closing: **Fluorescence and phosphorescence are both called luminescence.Their lifetimes, 10 ns and 1 s, stand in the ratio 1.000000×10^{8}, and exciting 10^{18} molecules gives an initial brightness of 1.000000×10^{26} against 1.000000×10^{18} photons per second.The one that waits is fragile: the same nonradiative path gives yields of 0.999990 against 0.000999, oxygen leaves 0.994431 against 1.785711×10^{-6}, and at 298 K the yield falls to 1.813085×10^{-7}——the fragility is not a property of the molecule but of the length of the wait.The separator is whether the spin flips.Placed among the earlier papers——in the seat where Paper 448 counted the entrance, the energy of one photon, the exit was counted here.To be honest——lifetimes and quenching constants are representative values, and the origin of the rate constants is not derived.**

References

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