

Electron-hole liquid in biological tissues under ultra high dose rate ionizing radiation

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We develop a quantitative model of ionization processes in biological tissues under Ultra High Dose Rate (UHDR) radiation. The underlying conjecture is that of electron-hole liquid (EHL) forming in water based substances of biological tissues. Unlike the earlier known EHL in semiconductor crystals, the charge carriers here are low mobile due to strong interactions with the background (solvated electrons, etc.); hence, EHL resembling ionic melts. Similar to all ionic systems, the Coulomb coupling makes that EHL energetically favorable that leads to recombination barriers suppressing subsequent structural transformations. In particular, generation of secondary reactive species in such EHL becomes limited translating into reduction of biological damages and tissue sparing effect. We show how these processes are sensitive to the tissue quality and frequency dispersion of the dielectric permittivity. Equations for dose and dose rate defining the sparing thresholds are derived.

I. INTRODUCTION

A seminal paper of 2014 [1] triggered intensive quest into understanding the Ultra High Dose Rate (UHDR) radiation effects in biological tissues. Nowadays, these effects are considered a new modality in radiation treatment of cancer known as FLASH RT. Its unique feature is a reduced damage to surrounding healthy tissues (sparing effect) not compromising the integral antitumor effectiveness. As evidenced by many reviews, [2–8] the underlying UHDR mechanisms remain elusive. Here we develop a quantitative physical model of UHDR effects. This work is limited to basic physics ignoring biological processes which may be oversimplification.

Our motivation here is driven by the observations that healthy tissues remain perceptive to radiation damages for dose rates below a certain threshold $\dot{D}_c$. However, they become much less responsive when dose rate $\dot{D} > \dot{D}_c$. That observation can be phenomenologically interpreted as originating from some collective behavior characteristic of materials with high concentration of plasma (positive and negative charges). The only such phenomenon known to us is that of the electron-hole liquid (EHL), previously known for perfect semiconductor crystals at low temperatures [9–18] and never discussed for objects like water at room temperature.

The nontrivial features of EHL in the preceding research are essentially quantum implying the non-localized electrons and holes in their corresponding bands of ideal crystalline structures. For such a ‘quantum EHL’, there exists a critical temperature T_c , below which EHL remains stable. With a few exceptions, $T_c \sim 100$ K. The ‘quantum EHL’ cases reveal themselves in the observed optical spectra, magnetism, and electrical conduction. For example, [15] EHL photoluminescence emerges much later than that of stand-alone charge carrier excitations, which reflects the kinetics of EHL condensation.

Here, we extend the conjecture of EHL to biological tissues. Our thinking is based on a simple intuitive argument illustrated in Fig. 1 where increase in dose rates leads to qualitative changes in the ionized radiation product towards denser e-h conglomerate, through excitons, then biexcitons, to EHL.

Our paper is organized as follows. In Section II we discuss general property of our EHL and its dose related criterion assuming zero electron-hole recombination. The recombination effects are introduced in Section III along with the corresponding EHL kinetics. Section IV discusses EHL effects related to the frequency dispersion of dielectric permittivity that are typical of water based substances and appear important for here introduced EHL. Finally, Section V summarizes our results and their correspondence to the available observations.

II. EHL WITHOUT RECOMBINATION

Here, we extend the EHL conjecture over the domain of biological materials. Unlike the ‘quantum EHL’, charge

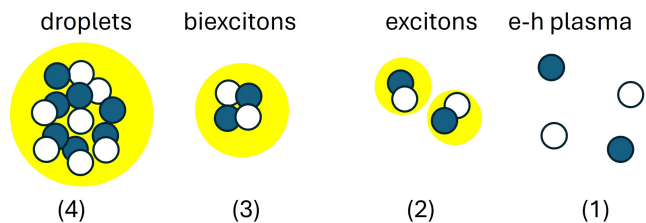


FIG. 1. Radiation induced ionization depending on dose rate and energy. Dark (white) circles represent electrons (holes). From right to left: (1) plasma of weakly interacting electrons and holes generated with low dose rates high energy radiation; (2) excitons, i. e. coupled electron - hole pairs generated with higher dose rates and close to the absorption edge radiation; (3) biexcitons which are coupled pairs of excitons forming with increase of exciton concentration; (4) a droplet of electron hole liquid under high dose rate of well absorbed radiation.

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carriers in biological substances strongly interact with atomic subsystems creating local atomic rearrangements, forming polarons or solvated electrons in a matter of picoseconds. [19–22] Because they carry clouds of atomic deformations, their diffusivities are extremely low, making them qualitatively similar to ionic melts. Following the standard description of ionic systems, the binding energy in such biological EHL is given by,

$$U = \frac{\mathfrak{M}e^2}{\kappa r}. \quad (1)$$

Here, κ is the dielectric permittivity, r is the nearest neighbor distance, and $\mathfrak{M}$ is the Madelung constant [23–25] accounting for interactions with charges beyond the nearest neighbors; typically, $\mathfrak{M} \sim 2$ varying between different structures.

To establish the EHL criterion, it is natural to introduce a dimensionless parameter

$$L = \frac{U}{k_B T} = \frac{\mathfrak{M}e^2}{\kappa r k_B T} \quad (2)$$

where k_B is the Boltzmann's constant and T is the absolute temperature; $k_B T = 0.026$ eV for room temperature $T = 300$ K. Depending on its L value, a e-h system can behave as a low concentration plasma ($L \ll 1$), or EHL ($L \gtrsim 1$), or ionic crystal ($L \gg 1$).

The significance of EHL concept here is that the binding energy barrier U suppresses recombination between the negative and positive components (say, between hydroxide ions and hydronium cations) by making it necessary to first decouple such a pair from the rest of EHL before they can recombine. The probability of such extraction process is estimated in terms of the Boltzmann's exponent,

$$p = \exp(-U/k_B T) = \exp(-L). \quad (3)$$

Such a coupling is consistent with the observation of bound pairs of solvated electrons and hydronium cations. [26]

To estimate L we start with the integral radiation dose D . Assuming that ionized charge carriers do not recombine yields their ultimate concentration,

$$n = 2D\rho/I, \quad (4)$$

in units $1/\text{cm}^3$ where $\rho \approx 1 \text{ g/cm}^3$ is the density of water as the main tissue component, I is the energy of e-h pair creation, and the coefficient 2 accounts for two charge carriers per pair. (Effects of recombination are described next.) The particle concentration and the average distance r between them are related through the standard rigid sphere approximation,

$$n(4\pi r^3/3) = 1. \quad (5)$$

Substituting here the above n yields

$$r = \left(\frac{3I}{8\pi D\rho} \right)^{1/3}. \quad (6)$$

As a numerical example, the average nearest neighbor distance in EHL is estimated as 7 nm assuming $D = 30$ Gy (escalated over the standard regime towards UHDR values).

A comment is in order with regards to the tacitly assumed ionization events randomly distributed in space. Given the typical energies of secondary electrons (in excess of 1 keV), and average linear energy transfers (LET, ranging from approximately 5 to 9 keV/ μm), these dipoles' lengths r_{dipole} exceed many microns, i. e. $r_{\text{dipole}} \gg r$. Therefore, interactions in gemini $e-h$ pairs created in the same ionization event can be neglected resulting in uniformly distributed EHL particles.

Using the latter r in Eq. (1) yields the value of parameter L . Setting then $L > 1$ defines a condition on integral dose under which EHL can form,

$$D > D_{\min} = \left(\frac{\kappa k_B T}{\mathfrak{M}e^2} \right)^3 \frac{3I}{8\pi\rho}. \quad (7)$$

For numerical estimates we set $I = 10$ eV, and $\kappa \approx 5$ based on the published data for fat in human tissues [27] and in bacterial cell. [28] The choice for κ will be discussed more in detail in Sec. IV below. Substituting these values yields $D_{\min} \sim 10$ G, in the ballpark of conventional radiation treatment doses.[2–7]

III. EHL WITH RECOMBINATION

While the criterion of Eq. (7) is given in terms of dose, it is a general perception that the UHDR effects are defined by the dose rates $\dot{D}$. Our resolution here is that the dose of Eq. (7) was derived assuming zero recombination. In reality, some recombination does take place, which may impose additional restrictions on the UHDR criterion. One relevant restriction is readily established based on qualitative arguments as follows.

Tacitly assuming zero recombination in deriving Eq. (7) implies that, in the very beginning of radiation exposure, the recombination time τ_0 is long enough to allow charge accumulation rather than depletion under charge generation rate g related to the dose rate as $g = 2\rho\dot{D}/I$. Following the standard kinetic analysis we present the recombination driven decrease in carrier concentration as $n = n_0 \exp(-t/\tau_0)$, i. e. recombination rate

$$(\partial n/\partial t)_{\text{recomb}} = -n/\tau_0. \quad (8)$$

Including both the generation and recombination processes yields, $dn/dt = g - n/\tau_0$. We thus require $dn/dt > 0$, i.e. $g > n/\tau_0$. Using the above expression for g and $n = 2\rho D/I$, the latter inequality yields the criterion

$$\frac{\dot{D}}{D} > \frac{1}{\tau_0}, \quad (9)$$

which should be applied along with the previously derived criterion $D > D_{\min}$ of Eq. (7). In a rough approximation, $\dot{D}/D$ expresses a reciprocal of the time τ_0

of dose deposition; hence, the latter criterion states that the entire dose $D > D_{\min}$ must be delivered during a short time not exceeding the initial recombination time in healthy tissues.

For numerical comparison, we assume water as a major component and, from independent study of electrolysis, take $\tau_0 \sim 0.01 - 1$ s. [29] Also, as in the above, we set $I = 10$ eV, and $\kappa \approx 5$. Combining the above yields the following criteria for UHDR effects:

$$D_{\min} \sim 10\text{Gy}, \quad \dot{D}_{\min} = \frac{D_{\min}}{\tau_0} \sim 10 - 10^3 \frac{\text{Gy}}{\text{s}}. \quad (10)$$

A drawback of Eq. (8) based approach is that it ignores the charge carrier binding with EHL. Taking the latter into account dictates renormalization $\tau_0 \rightarrow p\tau_0$ with p from Eq. (3) and r expressed through n according to Eq. (5). As a result, Eq. (8) takes the form of a kinetic equation,

$$\frac{dn}{dt} = -\frac{n}{\tau_0} \exp \left[-\frac{\mathfrak{M}e^2}{\kappa k_B T} \left(\frac{4\pi n}{3} \right)^{1/3} \right] + g \quad (11)$$

where we have added the charge generation rate g .

According to our theory, the reactive species are generated by charge carriers that overcome the EHL binding. Their concentration (R) is given by

$$R(t) = \int_0^t \frac{n}{\tau_0} \exp \left[-\frac{\mathfrak{M}e^2}{\kappa k_B T} \left(\frac{4\pi n}{3} \right)^{1/3} \right] dt. \quad (12)$$

Depicted in Fig. 2 are the rates of EHL concentration dn/dt and the concentration dR/dt of charges departed from EHL that are capable of forming reactive secondary species (radicals, etc.) mediating biological damage. We observe that $dR(t)/dt$ first increases as EHL accumulates, and then decreases when EHL binding becomes strong enough to suppress particle departure from EHL. (Note parenthetically that the concentration of re-

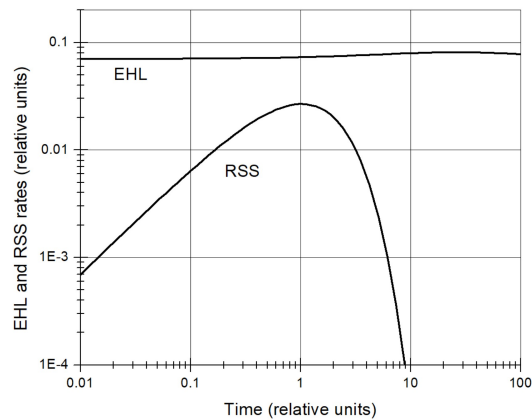


FIG. 2. Typical temporal dependencies of EHL ($n(t)$) and RSS ($R(t)$) rates assuming strong enough binding ($L > 2$).

active secondary species can be much lower than R when

the charges departing from EHL take part in subsequent transformations.)

Once EHL is formed, it persists being almost intact for a certain time interval after the radiation is turned off (making $g = 0$ in Eq. (11)) because the EHL binding remains about the same during several τ_0 . Such a process is presented in Fig. 3 as the integral of Eq. (11) with $g = 0$. The decay of Fig. 3 reveals first a degree of EHL

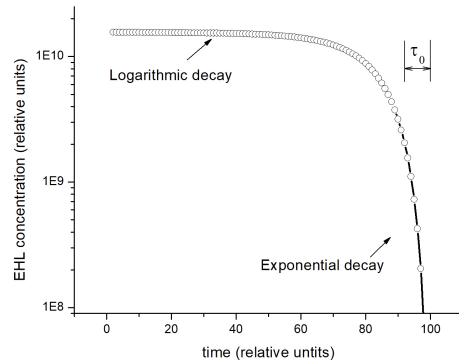


FIG. 3. A typical temporal dependence of EHL concentration with initial strong binding ($L > 2$) upon switching off the radiation source.

stability due to the mutual binding of charges in EHL resulting in a relatively weak temporal dependence close to logarithmic. However that decay abruptly accelerates becoming exponential after the EHL concentration decreases enough to considerably weaken the binding. The latter prediction can be useful for identifying EHL effects experimentally, through optical spectra triggered by UHDR exposure, similar to the experiments identifying solvated electrons. [30, 31]

Also, following Eq. (11) we were able to verify the biological sparing effect, according to which radiation inflicted damages decrease with the dose rate. Such evidence of the sparing effect is shown in Fig. 4 presenting $R(t)$ from Eq. (12) corresponding to an order of magnitude different generation rates, g and $10g$. These curves are qualitatively similar to recently published experimental data [32, 33].

IV. FREQUENCY DISPERSION EFFECTS

There is yet another constraint defining the minimum dose rate: the time (frequency) dispersion of dielectric permittivity, which, in the low frequency region, is known as α -dispersion. [34–36] It originates from the relatively slow processes of molecular movements, protein reorientations, etc. determining the low frequency dielectric permittivity of water and related substances. These slow relaxation processes make dielectric permittivity very high, $\kappa \approx 80$ for water after long enough time.

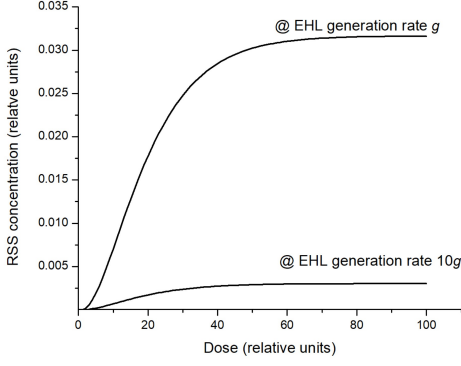


FIG. 4. A calculated sketch of sparing effect showing how irradiation created modifications *decrease* with the dose rate.

With such dielectric constants as $\kappa \approx 80$, our parameter L in Eq. (2) decreases by more than order of magnitude effectively eliminating the EHL coupling. To retain the latter, $D_{\min}$ of Eq. (7) must increase by almost three orders of magnitude vs estimated in the above. However, as pointed out in many cited papers, [34] the slow atomic processes start showing up only at times exceeding 1-10 ms. At shorter time intervals, they do not contribute much, and dielectric permittivity is mostly of electronic nature, about the same as in the above mentioned fat tissues (we note that the dielectric permittivity of ice is close to 3.17. [35]). Coincidentally, the molecular movement time constraint is of almost the same order of magnitude as our first culprit of recombination, i.e. $\sim 1 - 10$ ms. Therefore, our estimate in Eq. (10) remains qualitatively the same.

Because the ‘frozen’ low frequency molecular dynamic can be an important part of UHDR effects, we provide in this section its analysis based on Eq. (11) with κ being a time dependent variable. It is convenient to use a simple model of its time dependence in the form,

$$z(t) \equiv \kappa^{-1} = \kappa_0^{-1} \exp(t/\tau_\kappa) \quad (13)$$

where $\tau_\kappa \sim 1 - 10$ ms plays the role of a material parameter and κ_0 will not show up in our calculations.

We then describe the kinetics in terms of independent variable z noting that

$$\frac{dn}{dt} = \left(\frac{z}{\tau_\kappa} \right) \frac{dn}{dz}. \quad (14)$$

Introducing a new function

$$u = z \left[\frac{\mathfrak{M}e^2}{\kappa k_B T} \left(\frac{4\pi n}{3} \right)^{1/3} \right], \quad (15)$$

Eq. (11) takes the form

$$\frac{3}{\tau_\kappa} \left(\frac{du}{dz} - \frac{u}{z} \right) = \frac{u}{z\tau_0} \exp(-u). \quad (16)$$

Its solution can be represented as,

$$\int_{u_0}^u \frac{\tau_0 \exp(u) du}{u[3\tau_0 \exp(u) + \tau_\kappa]} = \ln \left[\frac{z(t)}{z(0)} \right] + C \quad (17)$$

with integration constant C determined by the initial condition $u(t=0) = u_0$.

Assuming the temporal model of Eq. (13), the right-hand-side in Eq. (17) reduces to simply t/τ_κ . Its left-hand-side while cannot be expressed in elementary functions, is offering a quick numerical solution illustrated in Fig. 5. Comparing Figs. 3 and 5 it is obvious that the frequency dispersion of dielectric permittivity can significantly affect the temporal dependence of EHL decay.

V. DISCUSSION AND CONCLUSIONS

Our theory is limited to physical processes thus predicting UHDR effects at the cellular and subcellular levels of hierarchy, much below that of organisms. In that connection, we would like to mention a recent publication [32] experimentally evaluating dose rate effects on oxidative damage to cellular culture. It was found that the higher dose rates resulted in less damage, although no underlying physical mechanism was determined. Similarly, a recent publication [33] explored the molecular basis of the electron UHDR effect in healthy human bronchial epithelial cells with the conclusion that it resulted in lower RSS production. Phenomenologically our mechanism of sparing correlates with the “free radical hypothesis” [8] and hypothetical scavenger effects, [37] according to which some undefined processes diminish the concentration of free radicals. This paper attributes such processes to EHL binding.

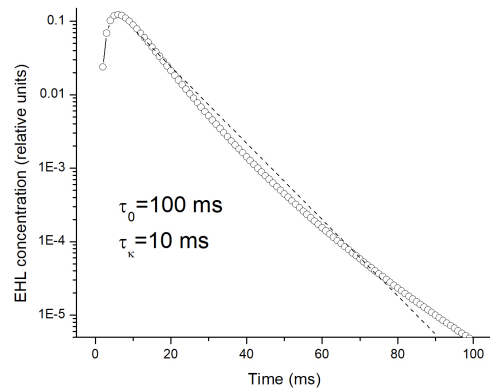


FIG. 5. A temporal dependence of EHL concentration n for recombination and dielectric permittivity relaxation processes acting simultaneously. The corresponding times are shown in the figure will serve as our example. The maximum point reflects increase in EHL concentration before the dielectric relaxation effects starts. Dashed line represents an approximation of the purely exponential decay $\exp(-t/\tau_\kappa)$.

We note that our ‘kinetic’ results presented in figures 2, 3, 4, and 5 can be rather sensitive to parameter choice. The depicted trends are aimed at illustrating the major trends only; more accurate tuning (possibly taking into account some other kinetic processes) will be needed to compare our predictions with actual data.

Our theory predicts that EHL effects are not expected within cancer morphologies where their inherent disorder shortens recombination times by many orders of magnitude [38, 39] killing EHL. Lacking EHL, free radical generation and cancer killing effects are expected to prevail.

Finally, we would like to point at a related problem

not explicitly addressed here. It concerns the dispersion/relaxation of dielectric permittivity under the condition of multiple charge centers created at random instances rather than simultaneously at $t = 0$ as implicitly suggested in Sec. IV. That problem will be considered elsewhere.

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