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Photocurrents induced by stimulated absorption of light

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ABSTRACT

In organic materials absorption of light results in creation of the Frenkel excitons, Coulomb -bound electron-hole pairs having an integer spin. Being composite bosons, Frenkel excitons may accumulate in significant quantities in a single quantum state. The probability of photon absorption by such a state increases as (N+1) where N is the occupation number of the state. This enhancement is due to the stimulated absorption of light, which is a final-state stimulated process analogous to the well-known stimulated emission of light. We propose using the stimulated absorption for creation of solar cells of a record quantum efficiency. We are going to use the organic microspheres to accumulate the Frenkel excitons at the discrete frequencies corresponding to the photonic whispering gallery modes of the sphere. The dissociation of accumulated Frenkel excitons will be effectuated periodically using the transparent carbon contacts on a piezoelectric mechnical support.

Keywords: SolarCell; Photovoltaics; Exciton; Bose-EinsteinCondensation

1. INTRODUCTION

Development of the renewable sources of energy is a strategic economic priority nowadays. The solar energy is especially important for the tropical countries due to their favorable climate conditions. At present, the commercially available solar cells are based on silicon monocrystals which are relatively expensive and can hardly compete with the conventional energy sources therefore. Organic materials are widely discussed now as a valuable alternative to silicon but several obstacles still block the full-scale development of organic photovoltaics. First, unlike silicon, absorption of light in organics does not produce the free electron-hole pairs, but bound electrically neutral states known as Frenkel excitons. A significant external voltage needs to be applied to help dissociating the Frenkel excitons with formation of the unbound electrons and holes which contribute to the photocurrent [1]. Second, the metallic contacts used at present absorb light themselves which reduces significantly the quantum efficiency of photocells. The revolutionary improvement of the performance of the organic photocells due to the use of new materials and new physical effects is in the focus of this Letter. The starting idea is to take advantage of the bosonic properties of Frenkel excitons and use them to achieve the stimulated absorption of light. The photocells based on stimulated absorption are expected to posses a dramatically enhanced quantum efficiency with respect to traditional photocells based on the spontaneous absorption of light. The expected difference is as huge as the difference between a light emitting diode (producing light by spontaneous emission) and a laser source (producing light by stimulated emission). Furthermore, we take advantage of the experience accumulated in the recent years in production of the optically transparent electric contacts based on the diamond-like-carbon (DLC) with embedded carbon nanotubes and to operate with the organic microspheres characterized by the photonic whispering gallery modes of extremely high quality factors. We emphasize the important role of photonic confinement in our photocells, which allows for resonant generation of macroscopic number of Frenkel excitons in the same quantum state during the life-time of the corresponding photonic mode. In turn, once being created, the macroscopic population of Frenkel excitons reproduces itself by stimulated absorption of new solar photons. We propose a new technology based on a new physics, new structures and new materials. The targeted outcome is a new generation of photocells with a drastically improved quantum efficiency.

In 1917 Einstein has proposed the theory of radiation of atoms which accounted for three essential processes of light-matter interaction: the spontaneous emission of light, the spontaneous absorption of light by atoms and the stimulated emission of light. This latter process has found its use in the new coherent sources of light (lasers, from Light Amplification by Stimulated Emission of Radiation, which we frequently meet in the everyday life now). In lasers, generation of new photons by matter is stimulated by existing population of the photon mode in a laser cavity. The fourth essential process of light-matter coupling is the stimulated absorption of light. It may only take place in the systems where the absorbed photons generate bosonic quasiparticles, e.g. excitons. Excitons are Coulomb correlated electron-hole pairs, a direct analogy of hydrogen atoms, created inside inorganic semiconductor crystals due to resonant absorption of light at the energies slightly below the bandgap (Wannier-Mott excitons [2]) or in organic crystals at the energy below the HOMO-LUMO transition (Frenkel excitons [3]). While electrons and holes are fermions, the excitons possess integer spins and are composite bosons therefore. As such, the excitons can be accumulated in large quantities in a single quantum state, forming the Bose-Einstein condensate (BEC). Recently, multiple experimental evidences for the BEC of excitons coupled to light modes in semiconductor microcavities have been reported [4-6]. An exciton coherently coupled to a confined photon in a microcavity forms a so-called exciton-polariton. A new generation of semiconductor light sources called "polariton lasers" and based on the Bose-Einstein condensation of exciton-polaritons has been suggested and experimentally realized very recently [7,8].

2. STIMULATED ABSORPTION OF LIGHT BY EXCITONS

Creation of a new exciton in a quantum state already occupied by N excitons has a probability (N+1) times higher than creation of an exciton in an empty state. This is why the stimulated absorption of light by excitonic condensates is expected to be extremely efficient. Several conditions need to be met to ensure stimulated absorption: 1) the seed exciton population exceeding 1 must be created; 2) the life-time of the exciton state (accounting for its radiative and non-radiative life-times) must be longer than the generation rate of excitons. The first condition is readily satisfied due to the intensity fluctuations in the solar light: statistically, simultaneous excitation of two or more excitons is always possible within a sufficiently long time interval. The second condition can be satisfied if the intensity of incident light exceeds some critical intensity dependent on the exciton-life time. In this case the amplification of the seed population takes place, the exciton condensates stabilizes itself and the rate of stimulated absorption keeps steadily growing up to the saturation value dependent on the spectral density of incident light and the Mott transition threshold for the exciton system. Making use of the stimulated absorption of light would be a fundamental breakthrough in photovoltaics which may be compared with invention of lasers in optical lighting. The research in this direction is timely and relevant given the overall high interest to organics materials for photovoltaic applications in the world.

The qualitative analysis shows that the most important condition to be satisfied to ensure the stimulated absorption going on is a sufficiently long life-time of an exciton state of interest. Sufficiently long means about 100 ps according to our estimations based on the average intensity of the day-time light in the Mediterranian region. Another important practical issue is linked with a necessity to have a significant absorption within the large frequency range of the visible and near ultraviolet light, in order to avoid losses of the solar energy. This is partly resolved because of the natural inhomogeneous broadening of Frenkel exciton resonances in the most part of organic materials, which is of the order or several hundreds meV, typically. In order to further extend the frequency range of efficient stimulated absorption and to control the life-time of the concerned exciton states we propose using the organic microspheres as a main element of the solar cell. In is well known that dielectric or semiconductor microspheres are characterized by discrete optical spectra composed by a multitude of sharp resonances. These resonances are associated with so-called whispering gallery modes, i.e. the light modes confined near the surface of the microsphere (**Figure 1**). The whispering gallery modes of high orbital and magnetic indices may have a quality factor (ratio of the mode frequency to its linewidth) of the order of 10^5-10^6 which ensures the photonic life-time of 100 ps or more. In the same time, the finesse of these modes (ratio of the frequency splitting between different modes to the linewidth of the mode) remains quite low (of the order of several units). This is why a microsphere filled by an absorbing aggregate would absorb light at the huge multitude of discrete frequencies corresponding to the whispering gallery modes. The life-time of excitons coupled to each of these modes would be long enough to ensure stimulated absorption. Between these selected frequencies the absorption of light would be still spontaneous.

3. DEVICE CONCEPT

In order to cover the most part of the visible spectrum of light we propose to operate with the microspheres of different radii ranging from several microns to several tens of microns. Furthermore, in order to ensure efficient collection of the charge carriers accumulated in the microsphere due to stimulated absorption, we propose using the transparent contacts made of a diamond-like-carbon (DLC). Fabrication of such contacts which may serve as Fresnel lenses as well is now a registered know-how in Europe. The DSC cover layer would pro-

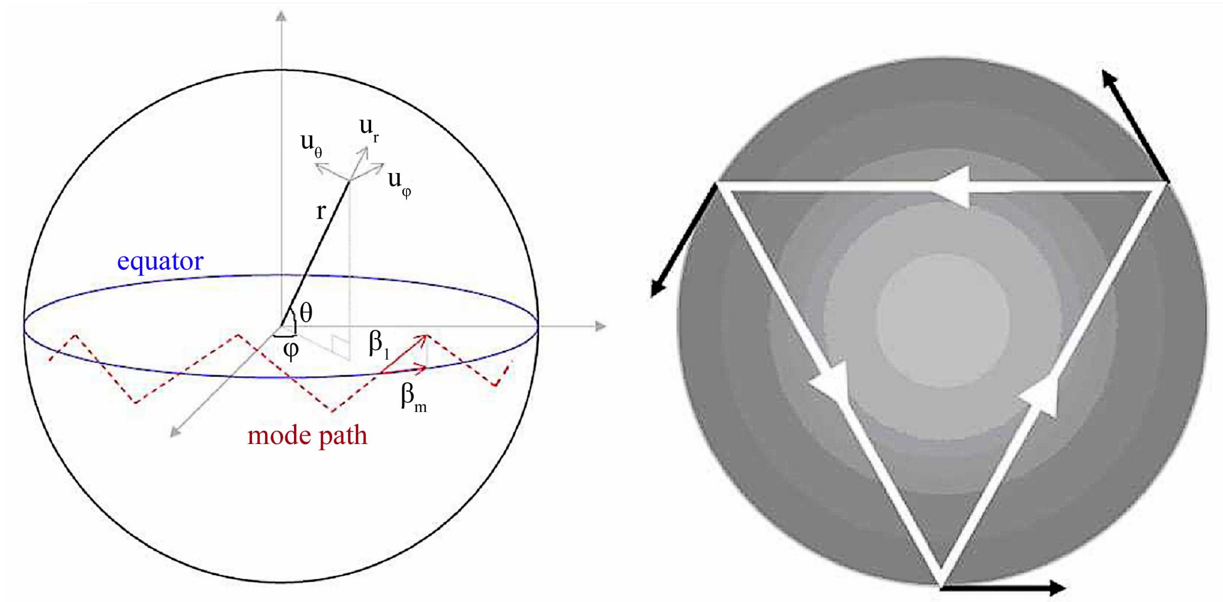

Figure 1. A dielectric sphere and the path of light in the whispering gallery mode (left); localization of light in a sphere due to multiple internal reflections (right).

tect the organic solar cell from chemical and mechanical damage, ensure focusing of light on the microsphere and play a role of a non-absorbing upper contact (see the scheme in **Figure 2**). In order to allow for efficient accumulation of Frenkel excitons inside the organic microsphere we plan discharging it periodically which could be achieved by placing the upper contact on a piezoelectric support which would extend and contract

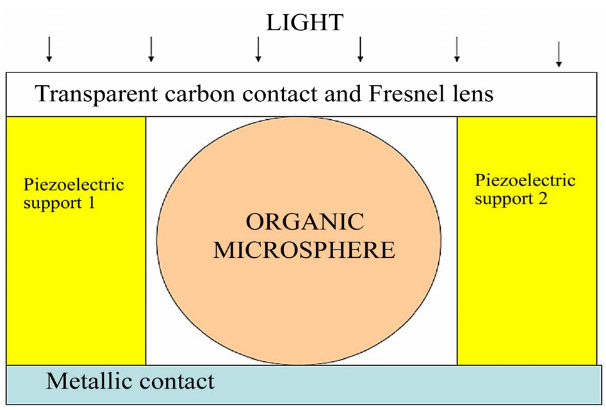

Figure 2. Scheme of the proposed photovoltaic device structure composed by an organic microsphere, where the Bose-Einstein condensate of exciton polaritons will be created by resonant optical pumping, a lower metallic contact and an upper transparent carbon contact supported by two piezoelectric pillars operating in a THz modulating regime: when the voltage between two contacts drops due to the current passing through the sphere the piezoelectric supports extend which leads to the break of current. As soon as the current is broken, the supports contract again, which allows for the new discharge of the microsphere etc.

with a Tera-Hertz frequency.

The proof-of-concept demonstration device would be a standard porphirine microsphere of the radius from 10 to 100 microns connected to two platinum microcontacts. One would measure a photocurrent generated by an accordable laser as a function of the frequency and the intensity of the laser light. A threshold-like superlinear dependence of the photocurrent as a function of the laser intensity at the discrete frequencies corresponding to the whispering gallery modes of the microsphere would be a "smoking gun" for the stimulated absorption. In the next phase the optimized microspheres would be assembled with the metallic and carbon contacts on the piezoelectric support. To improve the collection of solar light, the organic Fresnel lenses covered by a DSC protective film would be assembled on the top of the structure. The main risk at this stage is linked with the proposed discharge mechanism for the microspheres which has not yet been tested experimentally.

4. CONCLUSIONS

In conclusion, the recent advances in the Bose-Einstein condensation of excitons and exciton-polaritons open new horizons for the organic photovoltaics. A revolutionary concept of stimulated absorption of light may allow for a dramatic increase of the quantum efficiency of organic solar cells. In this Letter a device concept suitable for demonstration of photocurrents generated by stimulated absorption of light is proposed.

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