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Wave–particle interactions in a resonant system of photons and ion-solvated water

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A B S T R A C T

We investigate a laser model for a resonant system of photons and ion cluster-solvated rotating water molecules in which ions in the cluster have very low, non-relativistic velocities and direction of motion along a static electric field induced in a single direction. This model combines Dicke superradiation with wave–particle interaction. As the result, we find that the equations of motion of the system are expressed in terms of a conventional free electron laser system. This result leads to a mechanism for dynamical coherence, induced by collective instability in the wave–particle interaction.

The mechanisms behind *coherence* in physical systems are mainly classified into two types. In the first type, coherence is due to long-range order; in the second type, it is due to the collective instability in a many-body system with a long-range interaction. In laser physics, Dicke superradiation belongs to the first type and the free electron laser (FEL) belongs to the second type [1–5]. In this Letter, we present a new laser model that incorporates both types of dynamical coherence mechanism from the aspect of wave–particle interactions [6–10]. The ingredients of this system are *photons*, *solvated ions* and *water molecules*.

In Ref. [11] the many-body system of electric dipoles of water molecules that interact with the electromagnetic field radiated from rotating water molecules was studied using a quantum field theoretical approach based on an analogy to an FEL [3,4,7–10,12]. In simplified settings, it was shown that, around an impurity that carries a sizeable electric dipole and induces a static electric field oriented in the z-direction, a permanent electric polarization of water molecules in the z-direction emerges in the limit cycle of the system, due to the coherent and collective interaction of the water molecules with the selected modes of the radiation field.

Using this result, in this Letter we consider an FEL-like model for a cluster of solvated ions with very low, non-relativistic velocities and a uniform direction of motion. The electric dipoles of the water molecules which solvate each moving ion behave as XY spins which interact directly only with the transverse electromagnetic field radiated from the rotating water molecules. On the particle side of the wave–particle (i.e., radiation field–particle) interactions, there are two kinds of elements: water molecules and ions around which the water molecules are trapped by a Lennard-Jones potential.

In the FEL model [3–5,7], each relativistic unbound electron has the phase degree of freedom of its XY spin-like direction on the x–y plane orthogonal to its direction of motion (called the z-direction). Due to the presence of an undulator, that is, a transverse magnetic field created by a periodic arrangement of magnets with alternating poles, the unbound electrons are accelerated, producing synchrotron radiation.

In our case, the variable corresponding to the phase coordinate of an unbound electron is the phase θ of the direction of the electric dipole vector $\vec{d}$ on the x–y plane for each water molecule. Classically, we have

$$d_x + i d_y = d_0 |\sin\varphi| e^{i\theta} \,, \tag{1}$$

where φ is the zenith angle and $d_0 = 2ed_e$ with $d_e \approx 0.2$ [Å] [13]; however, we consider it quantum mechanically. Here, we assume that ions move along the z-axis with velocity $v \ll c$[1] and a static electric field $\vec{E}_0$ is induced in the z-direction. Then, we can invoke the result of Ref. [11], that is, the emergence of a permanent electric polarization of the solvent and non-screening water[2] in the z-direction.

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[1] In this Letter, quantities of the order of $(v/c)^n$ $(n \geq 1)$ are ignored.

[2] The solvent water obeys an electrostatic ordering mechanism (see the explanation below Eq. (11)) [14].

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This Letter is based on the assertion that, *to a good approximation in our system, the radiation field exchanges energy with water molecules only through the excitation and de-excitation of water molecules between the two lowest levels of the internal rotation of the hydrogen atoms of each water molecule around its electric dipole axis. The energy difference between these two levels is* $\mathcal{E}$*, such that* $\mathcal{E}/(\hbar c) \approx$ 160 *[*cm^{-1}*]* [11,13,15]. To describe this *resonant* interaction, in the quantum mechanical regime, we introduce the *energy* spin variables of the j-th water molecule as[3]

$$\widehat{s}^1_j = \frac{1}{2}[|e\rangle\langle g| + |g\rangle\langle e|]_j \;, \tag{2}$$

$$\widehat{s}^2_j = \frac{1}{2i}[|e\rangle\langle g| - |g\rangle\langle e|]_j \;, \tag{3}$$

$$\widehat{s}^3_j = \frac{1}{2}[|e\rangle\langle e| - |g\rangle\langle g|]_j \;, \tag{4}$$

where $|g\rangle_j$ and $|e\rangle_j$ are, respectively, the low-lying energy ground state and the excitation energy state of the j-th water molecule in the two-level approximation, and the superscripts of the energy spins represent fictitious dimensions [15,16]. We introduce an electric dipole moment operator $\hat{\vec{d}}_j$ for the j-th water molecule such that its third axis is the quantization axis of the rotating water molecule. One of the off-diagonal matrix elements $(\langle e|\hat{\vec{d}}|g\rangle)_j = \overline{(\langle g|\hat{\vec{d}}|e\rangle)_j}$ of the electric dipole moment operator $\hat{\vec{d}}_j$ of the j-th water molecule is given by

$$d_0 \int_{-1}^{1} d(\cos\varphi) \int_{0}^{2\pi} d\theta (\bar{Y}_{1,1}(\theta,\varphi)(\vec{e}_1 \sin\varphi \cos\theta + \vec{e}_2 \sin\varphi \sin\theta$$
$$+ \vec{e}_3 \cos\varphi)_j Y_{0,0}(\theta,\varphi)) = d_0 \sqrt{\frac{1}{6}}(-\vec{e}_1 + i\vec{e}_2)_j \;. \tag{5}$$

Then, the electric dipole moment operator can be represented as an off-diagonal matrix in the two-dimensional energy state space [17]:

$$\hat{\vec{d}}_j = \frac{\tilde{d}_0}{2}(-\vec{e}_1(2\widehat{s}^1) - \vec{e}_2(2\widehat{s}^2))_j \;, \tag{6}$$

where we define

$$\tilde{d}_0 = d_0\sqrt{\frac{2}{3}} \approx 0.82 \cdot d_0 \;. \tag{7}$$

The Hamiltonian for the interaction between ion-solvated water and the transversal radiation vector field $\hat{\vec{A}}$ (i.e., the electromagnetic field in the radiation gauge) can be written using the total relevant electric charge current $\hat{\vec{j}}$ of the solvated ions and water molecules as

$$\widehat{H}^{(p-i-w)}_{\text{int}} = -\sum_{I=1}^{N}\sum_{i=0}^{n_I} \hat{\vec{A}} \cdot \hat{\vec{j}}_{I,i} \;, \tag{8}$$

where the natural number n_I is the number of water molecules solvating the I-th ion and is observed to be 20 to 40 [18–21]. The total relevant electric polarization current $\hat{\vec{j}}$ is the sum of the ions' electric charge currents $\hat{\vec{j}}_I$ and the $1,2$-components of the water molecules' electric polarization currents $\dot{\hat{\vec{d}}}_{I,i}$:

$$\hat{\vec{j}}_{I,0} = \hat{\vec{j}}_I \;, \tag{9}$$

$$\hat{\vec{j}}_{I,i} = (\vec{e}_1\dot{\hat{d}}_1 + \vec{e}_2\dot{\hat{d}}_2)_{I,i} \quad (i \neq 0) \;, \tag{10}$$

where the time derivative $\dot{\hat{\vec{d}}}_{I,i}$ is taken in the interaction picture. In the interaction Hamiltonian Eq. (8), the odd-parity operator $\hat{\vec{j}}_{I,i}$ $(i \neq 0)$ has only off-diagonal matrix elements in the representation where the water molecule's free Hamiltonian in the two-level approximation $\mathcal{E}\widehat{s}^3_{I,i}$ is diagonal [15].

It is a crucial point that the photon-water molecule part of Eq. (8) is the Dicke interaction Hamiltonian for superradiance that works via the long-range order of electric dipoles in the system [1].

The characteristic length for the long-range order of water molecules, that is, the *coherence length* (i.e., the wavelength of a resonant photon), denoted by l_c, is estimated to be the inverse of the wavenumber of a resonant photon $\mathcal{E}/(\hbar c)$. As $\mathcal{E}/(\hbar c) \approx 160$ [cm^{-1}], $l_c \approx 63$ [μm] [13]. In our model, we assume that the x–y dimensions of the system fall within this length l_c.

In the semi-classical treatment, the generic form of the Hamiltonian of the water molecule system relevant to our mechanism consists of the rotational kinetic energy of water molecules with average moment of inertia $I^{(w)} = 2m_p d_g^2$ (where m_p is the proton mass and $d_g \approx 0.82$ [Å]) [13], the water solvation potential of ions and the interaction Hamiltonian of water molecules with the radiation field and the static electric field:

$$\widehat{H}^{(w)} = \sum_{I=1}^{N}\sum_{i=1}^{n_I} \frac{1}{2I^{(w)}}\widehat{L}^2_{I,i} + \sum_{I=1}^{N}\sum_{i=1}^{n_I} v^{(i-w)}_{I,i} + \sum_{I=1}^{N}\sum_{i=1}^{n_I} v^{(LJ)}_{I,i}$$
$$- \sum_{I=1}^{N}\sum_{i=1}^{n_I}\left\{\vec{A} \cdot \hat{\vec{j}}_{I,i} + \vec{E}_0 \cdot \vec{d}_{I,i}\right\} , \tag{11}$$

where $\vec{d}_{I,i} = (\vec{e}_3)_{I,i} d_0$ and $v^{(i-w)}_{I,i}$ is the screened Coulomb potential between the I-th ion and the charges on a water molecule. The Lennard-Jones potential $v^{(LJ)}_{I,i} = 4\epsilon_{LJ}[(\sigma_{LJ}/r_{I,i})^{12} - (\sigma_{LJ}/r_{I,i})^6]$ causes the I-th ion be solvated by n_I water molecules. It is a function of the distance $r_{I,i}$ between the I-th ion and the i-th water molecule and is attractive for $r_{I,i} > \sigma_{LJ}$ and repulsive for $\sigma_{LJ} > r_{I,i}$, where σ_{LJ} is a very short distance of the order of 1 [Å] [14]. When there is no external electromagnetic field, $v^{(i-w)}_{I,i}$ determines the configuration of electric dipoles of water molecules that are in contact with an ion. The solvation potentials are translationally invariant with respect to phase coordinates $\theta_{I,i}$. It is a significant point that, in our system, the bulk water molecules that screen the charges of ions do not form part of the laser mechanism, due to thermal noise that prevents ordered motion.

Now, due to the FEL-like mechanism, the collective and coherent behavior of the x–y phase coordinates of water molecules follows. This mechanism consists of two interlocked parts in a positive feedback cycle. The first part is the long-range collective ordering of dipole vectors of water molecules that solvate each ion moving along the z-axis. As a consequence, the radiation from the clusters of ordered rotating water molecules is almost monochromatic and the time-dependent process of the radiation field and order of water molecules approximates a coherent wave amplified along the z-axis. These approximations are improved by positive feedback in the second part of the mechanism: that is, the FEL-like wave–particle interaction process where an exponential instability of the fluctuation around the dynamic equilibrium state (i.e., our ready state) accompanies both the magnification of the radiation intensity and the water molecule's XY-phase bunching that produces long-range ordering [4,22]. Finally, we will find that this positive feedback cycle leads to laser radiation.

Under the assumption of monochromaticity of the radiation, the details of the first part of the mechanism are as follows. A part of the classical radiation field is a transverse wave in the x–y plane, which can be written as

$$A_x + iA_y = A_0 e^{-i\phi_0} \;, \tag{12}$$

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[3] In this Letter, a hat indicates that a variable is a quantum mechanical operator.

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