Quantum Nonreciprocity of Nanoscale Antenna Arrays in Timed Dicke States

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We predict a linear nonreciprocal effect that is based on the timed Dicke states in an ensemble of dipole-dipole coupled oscillators. This effect is examined on a nanoscale antenna array comprising two-level identical emitters. The studied nonreciprocity, which has no analogs in classical antennas, manifests itself in strong characteristic asymmetry of the radiation pattern, even for a single-photon laser pumping. Promising applications of our results for remotely tunable nanoantennas and nanocircuit elements are discussed.

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Introduction.—The Lorentz reciprocity theorem is one of the fundamental principles of electrodynamics. It states that fields $\mathbf{E}_{1,2}(\mathbf{r})$ induced by dipoles $\mathbf{p}_{1,2}$ located at $\mathbf{r}_{1,2}$, respectively, are related by the reciprocity relation, $\mathbf{E}_1(\mathbf{r}_2)\mathbf{p}_2 = \mathbf{E}_2(\mathbf{r}_1)\mathbf{p}_1$ [1]. One consequence of the reciprocity theorem is the symmetry of the scattering matrices at any given frequency. Because of the reciprocity theorem the transmitting and receiving field patterns of a reciprocal antenna are identical [2]. The reciprocity theorem is a consequence of the symmetry of kinetic coefficients (Onsager principle) [3]. Violation of the reciprocity theorem is possible in the cases of broken time reversal symmetry, when the Onsager principle in its ordinary form becomes invalid (for example, for uniformly rotating bodies and magnetized ferrites [3]). Nonreciprocity offers a number of means to control the electromagnetic field propagation (e.g., for separation of signals that travel in opposite directions). The classical mechanisms of nonreciprocity are of macroscopic nature, and therefore cannot be used in nanooptics and nanoelectronics. Thus, the search for new nonreciprocal phenomena, which are manifested at the nanometric scale is of fundamental physical interest. Such mechanisms have been discussed in earlier works: for example, Rabi-waves propagation in the quantum dots chains [4,5] and a "moving" photonic crystal generated in a three-level electromagnetically induced transparency medium [6]. However, they are predicted only for the regime of strong coupling of condensed matter with electromagnetic field. Nonreciprocal phenomena in the chains of plasmonic nanoparticles were considered in [7,8]. Here, the nonreciprocity manifests itself under simultaneous magnetization and geometrical chirality of the chain.

The coherent states of two-level quantum emitters coupled by the dipole-dipole (d-d) interaction have been extensively discussed by Scully's group in a series of recent publications [9–13]. This concept leads to the generalization of the usual Dicke states [14] for the case where atoms at different positions are excited at different times (so called, timed Dicke states). To that end, it was stated "... that the timing of the atomic excitation is the key physical process behind the present directional spontaneous emission; i.e., 'timing is everything'" [9]. The main result of this Letter is that such timing even in the linear regime leads to the appearance of nonreciprocity, which is not associated with magneto-optic or bianisotropic media and doesn't have any analogs in the classical electrodynamics. This effect is demonstrated by the example of a quantum nanoantenna array, because such devices are of great interest in modern nanooptics. Various types of antennas working in the optical regime at nanometer scale have been demonstrated [7,8,15–24], and thereafter the language of microwave circuits and antennas was successfully adopted for optical antennas [16,22], single two-level emitter [23], and the pair of d-d interacting two-level dipoles [24]. The majority of nanoantennas that have been studied so far manifested the reciprocal behavior, with the exception of the magnetized chiral chains of plasmonic ellipsoids [7,8]. Dynamic control of the optical field in these nanoantennas is performed by the bias magnetic field. The nonreciprocity mechanism presented in this Letter appears promising for the optical scanning of the radiation patterns by a variation of the laser pump-beam direction.

Model and generalized susceptibility formalism.—We begin by considering a one-dimensional (1D) array of N identical two-level emitters (for example, semiconductor quantum dots, or cold atom chains in optical lattices) coupled by d-d interactions. Emitters are positioned along the z axis at points $\mathbf{R}_m$, $m = 1, 2, \ldots, N$ with separation a. Every emitter is characterized by the ground state $|\downarrow_n\rangle$, excited state $|\uparrow_n\rangle$, and the energy of resonant transition $\hbar\omega_0$. Let us assume that the system works in a "transmitting regime." It means that the antenna transforms the near field induced by the currents in the input (feed) terminals to the propagating far field. The near field throughout the Letter will be named an external field and denoted as $\mathbf{E}^{NF}(\mathbf{R}_n, t) = \mathrm{Re}[\mathbf{E}^{NF}_\omega(\mathbf{R}_n)e^{-i\omega t}]$. It corresponds to the classical value and has a quasistatic structure.

Antenna array radiation is described by the total Hamiltonian $\hat{H} = \hat{H}_0 + \hat{H}_I$, where $\hat{H}_0$ represents the antenna array Hamiltonian in the absence of the external field (it includes the internal energy of all emitters and the energy of d-d interactions). Hamiltonian $\hat{H}_I$ corresponds to the interaction of the antenna array with external field and therefore reads

$$\hat{H}_I = -\mu \sum_{n=1}^{N} \mathbf{E}^{\rm NF}(\mathbf{R}_n, t)\mathbf{e}_\mu(\hat{S}_n^+ + \hat{S}_n^-), \quad (1)$$

where $\hat{S}_n^+ = |\uparrow_n\rangle\langle\downarrow_n|$, $\hat{S}_n^- = |\downarrow_n\rangle\langle\uparrow_n|$ are, respectively, creation-annihilation operators of excitation in the nth emitter, μ is the dipole moment, and $\mathbf{e}_\mu$ is the unit vector of the dipole orientation. Let us assume that the system is initially prepared in an arbitrary equilibrium state and it is perturbed by the external field. This field induces the observable polarization in the nth emitter, whose complex amplitude is expressed as $\mathbf{P}_{m\omega} = \mu\Sigma_{n=1}^N \alpha_{mn}(\omega) \times (\mathbf{e}_\mu \otimes \mathbf{e}_\mu)\mathbf{E}_\omega^{NF}(\mathbf{R}_n)$, where $\alpha_{mn}(\omega)$ is a partial generalized susceptibility of the mth emitter to the near field in the nth emitter. This polarization produces a propagating far field $\mathbf{E}_\omega^{\rm FF}(\mathbf{r})$ that is naturally regarded as the system response to the "generalized force", whose role is assumed by the near (external) field. Note also that the polarization vanishes if the external field tends to zero. The antenna response to the generalized force is described by the radiation pattern, which plays the role of the "generalized susceptibility" and can be expressed using the general Kubo formula [25].

Let us present the far field in the spherical coordinate system $\{r, \theta, \varphi\}$ with the origin centered at the first emitter ($n = 1$). Hereafter, we will follow the conventional terminology of the antenna theory [2]. Thus, the total field radiated by an N-element array is equal to the field of a single element positioned at the origin (the 1st one in our notation) multiplied by a factor $(\mathrm{AF})_N$, which is referred to as an array factor. Thus, the far field of the N-element array reads (see Supplemental Material [26])

$$\mathbf{E}_\omega^{\rm FF}(\mathbf{r}) = \mathbf{E}_{0\omega}^{\rm FF}(\mathbf{r})(\mathrm{AF})_N, \quad (2)$$

where

$$\mathbf{E}_{0\omega}^{\rm FF}(\mathbf{r}) = \frac{k^2\mu}{r} e^{i(kr-\omega t)}\alpha_{11}(\omega)\left(\overleftrightarrow{\mathbf{I}} - \frac{\mathbf{r}\otimes\mathbf{r}}{r^2}\right) \times (\mathbf{e}_\mu \otimes \mathbf{e}_\mu)\mathbf{E}_\omega^{\rm NF}(\mathbf{R}_1) \quad (3)$$

is the field of a single element, $\overleftrightarrow{\mathbf{I}}$ is the unit tensor, and

$$(\mathrm{AF})_N = C\sum_{n=1}^{N}\sum_{m=1}^{N} (\mathbf{e}_\mu \mathbf{E}_\omega^{\rm NF}(\mathbf{R}_n))\alpha_{mn}(\omega)e^{i(m-1)ka\cos\theta} \quad (4)$$

is the array factor with $C = [(\mathbf{e}_\mu\mathbf{E}_\omega^{\rm NF}(\mathbf{R}_1))\alpha_{11}(\omega)]^{-1}$ being the normalization coefficient. According Kubo relation [25], the partial generalized susceptibility can be expressed in terms of Heisenberg operators $\hat{S}_n(t) = \hat{S}_n^+(t) + \hat{S}_n^-(t)$ as

$$\alpha_{mn}(\omega) = \frac{i}{\hbar}\int_0^\infty e^{i\omega\tau}\langle\hat{S}_n(\tau)\hat{S}_m(0) - \hat{S}_m(0)\hat{S}_n(\tau)\rangle d\tau, \quad (5)$$

where $\langle\cdot\rangle$ means averaging over the given quantum state of the system.

Timed Dicke state in nanoantenna array: Effect of nonreciprocity.—For antenna array analysis, it is convenient to use so-called timed Dicke basis [13], for which the first excited state in the notation of [13] reads

$$|\Psi\rangle = |B_0\rangle = \frac{1}{\sqrt{N}}\sum_{m=1}^{N} |\downarrow\downarrow\downarrow\ldots\uparrow_m\ldots\downarrow_N\rangle e^{i(m-1)\Phi}, \quad (6)$$

where Φ is a given phase shift (timing factor). As has been shown by Scully [13], the probability amplitude $|\beta_0\rangle$ for $|B_0\rangle$ state for large N obeys the equation $\dot{\beta}_0 = -(\Gamma + \Gamma_N + i\Im_N)\beta_0$, where Γ is the single emitter decay rate, Γ_N is the collective decay rate of the array, while $\Im_N$ is the collective Lamb shift [13]. This expression allows one to consider $|B_0\rangle$ as a quasistationary eigenstate of the antenna array, where its eigenvalue can be found as a complex eigenvalue of the operator of the d-d interactions given in [26,27]. Let us note that this operator corresponds to the presence of the counterrotating states with two excited atoms and one virtual photon with negative energy, which must be taken into account for the correct description of the d-d interactions [28]. For detailed calculations of collective decay rate and collective Lamb shift for antenna array, see [26]. Principles of preparing the $|B_0\rangle$ state in the single photon laser experiments were discussed in [9,13].

The averaging of general relation (5) with respect to the timed Dicke state $|B_0\rangle$ reads

$$\alpha_{mn}(\omega, \Phi) = \frac{1}{\hbar N}\left(\frac{e^{i(n-m)\Phi}}{\omega - \omega_0 - \Im_N + i(\Gamma + \Gamma_N)} - \frac{e^{-i(n-m)\Phi}}{\omega + \omega_0 + \Im_N + i(\Gamma + \Gamma_N)}\right), \quad (7)$$

(here we take into account that for the $|B_0\rangle$ state $\langle\hat{S}_m^+\hat{S}_n^+\rangle = \langle\hat{S}_m^-\hat{S}_n^-\rangle = 0$). Originally, the factor $e^{-\eta\tau}$ with infinitesimal positive parameter η has been included in the Kubo relation to force the response decay in time when $\tau \to \infty$. In the last stage of the calculation therefore the limit $\eta \to +0$ was taken (stationary state). We relate the Kubo formula to the $|B_0\rangle$ state which is a quasistationary one, following the concept of the resonance on the quasidiscrete level [29]. Thus, we keep in the final result the decay factor equal to the collective spontaneous emission decay rate. Let us assume that the near field varies along the array as in a traveling wave, thus $\mathbf{E}_\omega^{\rm NF}(\mathbf{R}_n) = \mathbf{E}_\omega^{\rm NF}(\mathbf{R}_1)e^{i(n-1)\beta}$, where β is an arbitrary phase shift. Thus, the array factor (4) may be represented as [30]

$$(\mathrm{AF})_N = A_-(\omega, \beta, \Phi)\frac{\sin\Psi_-}{N\sin(\frac{\Psi_-}{N})} + A_+(\omega, \beta, \Phi)\frac{\sin\Psi_+}{N\sin(\frac{\Psi_+}{N})}, \quad (8)$$

where

$$A_{\pm}(\omega, \beta, \Phi) = \sin\left(\frac{N(\beta \pm \Phi)}{2}\right)\left[\sin\left(\frac{\beta \pm \Phi}{2}\right) \times \alpha_{11}(\omega)(\omega \mp \omega_0 \mp \Im_N + i(\Gamma + \Gamma_N)\right]^{-1},$$

$\Psi_{\mp} = Nka(\cos\theta \mp \xi)/2$, $\xi = \Phi/ka$ (see Supplemental Material [26]).

Analysis of the Eq. (8) shows that the array in the considered $|B_0\rangle$ state corresponds to a system of two macroscopic arrays with different linear phase progressions [31]: the total array factor is presented as a superposition of two partial array factors $f_{\pm}(\theta) = \sin\Psi_{\pm}/N\sin(\Psi_{\pm}/N)$. These factors correspond to single-beam arrays with phase shifts $\mp\Phi$ and appear in (8) with the amplitude factors $A_{\pm}(\omega, \beta, \Phi)$, respectively.

It is easy to see from (7) that for the $|B_0\rangle$ state the symmetry relations for generalized susceptibilities satisfy $\alpha_{mn}(\omega, \Phi) = \alpha_{nm}(\omega, -\Phi)$. This situation is analogous to the Onsager relation for kinetic coefficients for uniformly rotating body and for bodies in an external magnetic field [32] (in this case phase Φ plays the role of rotation velocity, or magnetic field induction). It means that array factor $(\mathrm{AF})_N$ is not invariant under the exchange $\mathbf{E}_{\omega}^{\mathrm{NF}}(\mathbf{R}_n) \to \mathbf{E}_{\omega}^{\mathrm{NF}}(\mathbf{R}_{N-n+1})$. Therefore, we have the transformation

$$(\mathrm{AF})_N(\theta, \beta, \omega, \Phi) = (\mathrm{AF})_N(\pi - \theta, -\beta, \omega, -\Phi) \quad (9)$$

but not $(\mathrm{AF})_N(\theta, \beta, \omega, \Phi) = (\mathrm{AF})_N(\pi - \theta, -\beta, \omega, \Phi)$. Expression (9) shows that with the spatial rotation of the antenna array by 180° one must replace $\Phi \to -\Phi$ to keep the physical state (like the sign of the angular velocity or of magnetic field is changed in [32]).

Thus, the antenna array manifests the nonreciprocal properties in the $|B_0\rangle$-state, which are illustrated by numerical results presented in Figs. 1–3. If $|\xi| \leq 1$, each term in (8) creates a main lobe, which represents a conical wave. Their direction of propagation is determined by $\cos\theta_0 = \pm\xi$, and therefore depends on the phase shift Φ. Thus, the variation of phase Φ allows one to perform spatial scanning by the radiation pattern of the array in the $|B_0\rangle$ state (for a detailed picture of spatial scanning, see figures in [26]). The nonreciprocity manifests itself in the different peak field magnitudes in the two lobes oriented at angles θ_0 and $\pi - \theta_0$. Two different pictures of nonreciprocity are possible. In the first one which may be called "weak nonreciprocity", the orientation of the primary (strongest) main lobes, that are normalized to unity, is invariant with respect to the array rotation by 180 degrees. Array rotation affects only the peak field magnitude in the secondary main lobe (see Fig. 1). In the second nonreciprocity manifestation, which may be called "strong nonreciprocity", the array rotation is accompanied by the exchange between the primary and secondary main lobe orientations (for details, see numerical results in [26]). For specific values of ξ the antenna main lobe direction is not affected by the sign of β, while the secondary main lobes are strongly suppressed.

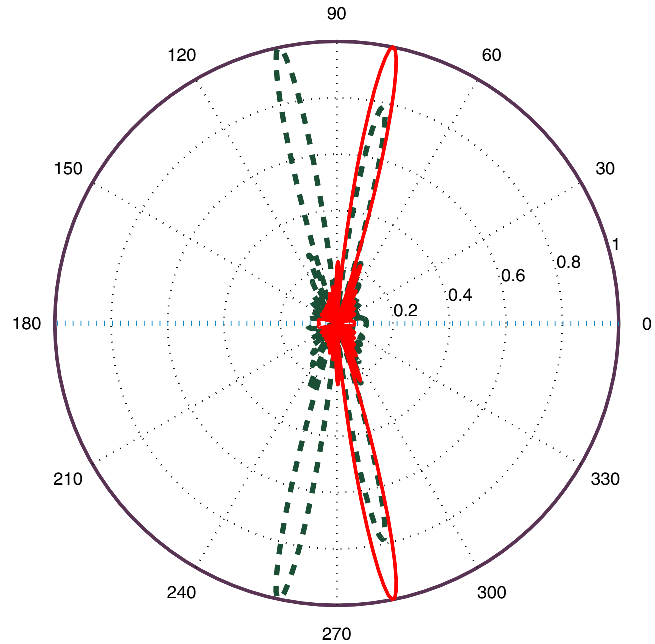

FIG. 1 (color online). Radiation patterns in the regime of weak nonreciprocity: antenna array comprises $N = 16$ elemental dipoles with $ka = \pi$, $k_0 a = 1.2\pi$, and $\xi = 1.8$: red line—$\beta = \pi/4$, dashed green line—$\beta = -\pi/4$. Dotted blue line denotes the array axis. The inversion of the sign of β leads to the symmetric turn of the main lobes with respect to the axis $\theta = \pi/2$ accompanied by a strong variation of secondary main lobe magnitude.

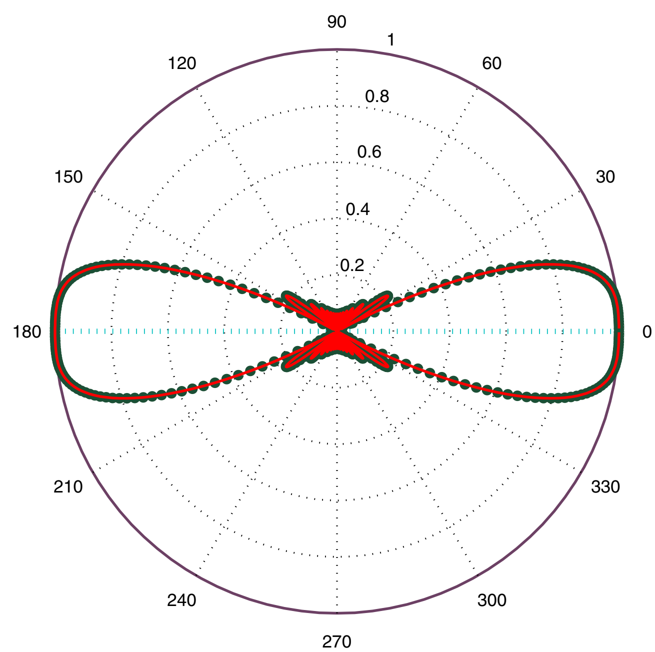

FIG. 2 (color online). Radiation patterns of a linear array in the axial radiation regime: $\xi = 1.0$. All other parameters are the same as in Fig. 1. The pattern symmetry corresponds to the array reciprocity.

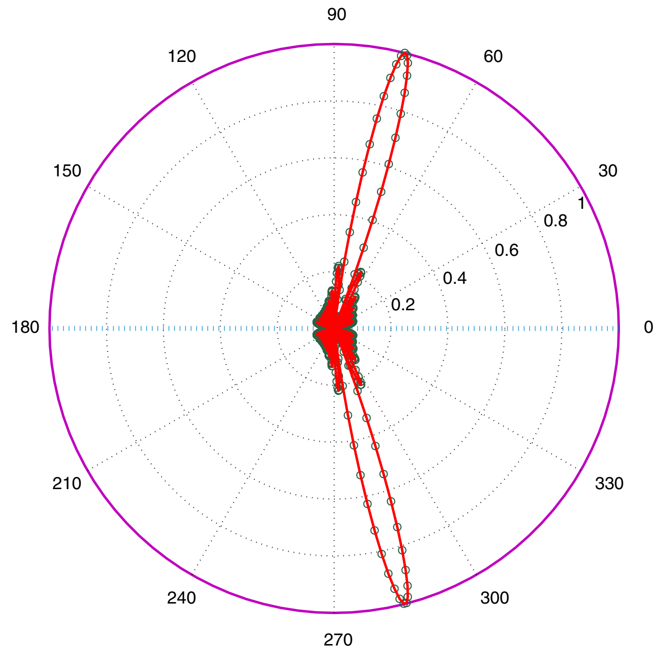

FIG. 3 (color online). Radiation patterns in the regime of extreme nonreciprocity: $\xi = 1.75$. The inversion of the sign of β does not affect the direction of the primary main lobe (while the secondary main lobes are strongly suppressed). All other parameters are the same as in Fig. 1.

Such behavior of the radiation pattern may be called "extreme nonreciprocity" (see Fig. 3). For $|\xi| \to 1$ two main lobes are combined into a single one (Fig. 2), which corresponds to the axial radiation regime and disappearance of the nonreciprocity. For $ka \geq 2\pi(N-1)/N(1+|\xi|)$, the number of main lobes increases, and the directivity of the antenna is reduced.

For an in-phase excitation of the antenna array ($\beta = 0$), we have a symmetry relation $(\mathrm{AF})_N(\theta, 0, \omega, \Phi) = (\mathrm{AF})_N(\pi - \theta, 0, \omega, \Phi)$, which leads to the reciprocity. To understand the role of quantum coherence in the appearance of nonreciprocity, it is instructive to consider the process of array radiation in the ground state, which in spinlike notation reads $|C_0\rangle = |\downarrow\downarrow\ldots\downarrow_N\rangle$. It is easy to see that it corresponds to the time-reversal symmetry and therefore reciprocity property because of $(\mathrm{AF})_N(\theta, \beta) = (\mathrm{AF})_N(\pi - \theta, -\beta)$ (for details, see [26]).

Conclusion.—We predict the linear effect of nonreciprocal light emission based on the use of emitters in the timed Dicke states and demonstrated on nanoantenna arrays. This effect is manifested in a characteristic asymmetry of the radiation pattern; thus, a nanoantenna is well suited for its experimental observation. This nonreciprocity appears without magneto-optic or bianisotropic media due to the time reversal asymmetry, which is stipulated by the directional single-photon resonant pumping. A similar mechanism of nonreciprocity will be observed in nanoplasmonics: an ensemble of emitters with *d*-*d* interactions placed on the boundary between vacuum and noble metal will produce an asymmetry of the dispersion characteristics of the surface plasmon with respect to the direction of propagation along the timing axis.

The effect predicted in this Letter looks promising for various future applications. Since timed Dicke states may be prepared by resonant laser pumping in the weak coupling regime [9], nonreciprocal elements working in the optical regime at nanometer scale can be driven with low-intensity light. For example, dependence of the nonreciprocity on the direction of pump pulse propagation opens up possibilities for the remote scanning of optical nanoantennas.

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