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REVIEW PAPER

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Photon-pair generation based on superconductivity

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Abstract: Superconductivity and optoelectronics have developed almost independently and had very rare interactions with each other in science and technologies. However recent interdisciplinary research opens up the potential for developing new optoelectronic devices. This review paper presents our recent theoretical and experimental demonstrations that superconductivity significantly modifies and accelerates photon generation processes. We have prepared superconducting light emitting diodes (LEDs) emitting at $\sim$ 1.6-μm optical-fiber communication band for the experimental demonstrations. This new-type LED operation is based on a unique physics related to Cooper-pair interband transition in a semiconductor, and further research leads to the solid-state simultaneously generated photon-pair sources for the potential application in quantum information and communication.

Keywords: photon pair, entanglement, superconductivity, Cooper pair, light emitting diode, quantum information

Classification: Superconducting electronics

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1 Introduction

With the development of information networks, information communication technology is being more and more important in the present information society. Especially the rapid growth of electronic commerce over the internet has led to an increasing demand for secure network communications. Cryptography has been widely used for this purpose and the Ron Rivest, Adi Shamir, and Lonard Adleman (RSA) algorithm is a representative public-key encryption [1]. Security of the RSA algorithm is based on the presumed difficulty of factoring large integers. However successful factorization of integers with larger decimal digit number has been reported with more and more improved performance of computers and the development of massively parallel computers [2]. Quantum information is expected to create robust highly secure information networks based on quantum mechanical principles [3, 4]. Quantum information or quantum bit is coded and transferred by the quantum states of photons. Since a photon is a single quantum of light, it cannot be cloned [5] and this prevents eavesdropping. Observation of a quantum state perturbs it and any attempt to eavesdrop the photon transmission should be instantaneously detectable.

BB84 is a quantum key distribution (QKD) scheme developed by Charles Bennett and Gilles Brassard in 1984 and is the first QKD protocol [6]. In addition to this single-photon-based protocol, protocols based on entangled photon pairs (EPPs) were proposed by A. Ekert (called as E91) [7] and by Charles Bennett, Gilles Brassard, and David Mermin (called as BBM92) [8]. Quantum entanglement occurs when a two-photon state is not expressed by the direct product of the underlying states. Its peculiar feature is that the two-photon states are correlated with each other but are not fixed before measurements. One-photon state in the photon pair is fixed only when the other photon state is fixed with a measurement. This holds true regardless of how far the two photons are separated in space. BBM92 and BB84 protocols works under the similar operations, but this feature makes it possible for the entanglement-based BBM92 pair detection scheme to double the QKD

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distance in principle [9]. Even longer communication distance is possible with entanglement swapping employing multiple EPP sources [10].

EPPs used for these demonstrations have been generated with parametric down conversion (PDC) [11], which is now a standard method in the relevant fields. Lasers are generally used for exciting EPPs and the directionality of the generated EPPs facilitates the application of EPPs in these demonstrations. However the coherence of the excitation laser sources results in the Poisson statistics of the generated photon-pair number states [12] and therefore leads to unregulated generation sequence of EPPs. This drawback has triggered the research toward "on-demand operation" of regulated EPP sources. Semiconductor quantum dots (QD) have been extensively studied on the biexciton and exciton cascaded photon-pair generation process and EPP generations have been reported [13, 14, 15, 16]. Photon pairs generated in this cascaded process have the time separation on the order of exciton lifetime, which is ~ 1 ns in semiconductor QDs. Therefore quantum entanglement is sensitively dependent on the coherence time of the exciton and biexciton emissions, which is generally shorter than the lifetime. It is also sensitively dependent on the exciton-state fine structure splitting [17], which significantly reduces the reproducible EPP generation. The biexciton-exciton cascaded processes also have a fundamental problem in generating ideal indistinguishable photon pairs. Because of the difference of the biexciton and exciton lifetimes, complete overlap of the two waveforms in time scale is difficult to achieve, which is important for various photon qubit operations.

Simultaneous generation of EPPs from semiconductors has been demonstrated with parametric scattering of biexciton polaritons in CuCl [18]. However this process also employs a laser excitation source and results in the Poisson distribution of the generated EPPs. In semiconductors two-photon absorption process has been widely studied and the reverse process of two-photon emission was employed to generate EPPs employing a GaInP/AlGaInP waveguide [19]. This is a spontaneous EPP generation and is a candidate for "on-demand" EPP source. The remaining problem is that this is the second-order process and the EPP generation probability is low on the

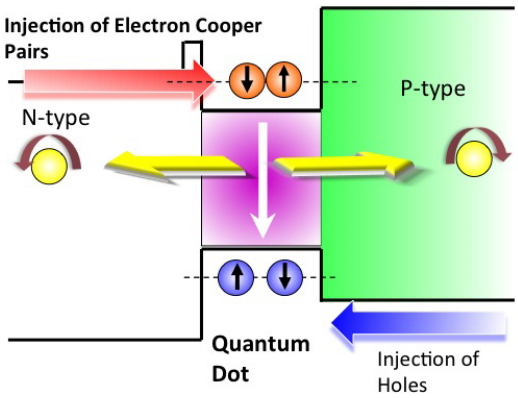

Fig. 1. Schematic band diagram and operation principle of superconducting LED.

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order of 10^{-5} of the first-order single photon process [19].

We have proposed another scheme to generate EPPs employing superconductivity [20]. This is fundamentally a light emitting diode (LED) but the n-type electrode is replaced with a superconductor. Cooper pairs induced in the superconductor are injected into the n-type semiconductor through the Fermi level in the conduction band with the proximity effect [21] as shown in Fig. 1. Radiative recombination of a Cooper pair with two holes is expected to generate EPP. When the radiative recombination takes place in a QD and the number of holes per excitation is limited definitely to two by the Pauli's exclusion principle on the ground-state energy level of the QD, then single EPP generation is realized per excitation pulse. Since Cooper pairs collapse with energy relaxation, the Cooper-pair injection into the QD conduction band should be either resonant injection into a QD energy level with a type-I QD as shown in Fig. 1 or evanescent penetration with a type-II QD [20]. In this review paper, the theoretical treatment of the Cooper-pair radiative recombination is described and is compared with our experimental results. Future prospects of our proposal are discussed.

2 Theoretical description of luminescence from a Cooper pair

In usual p–n junctions of semiconductors, an n-type carrier with wavenumber $\boldsymbol{k}$ and energy $\varepsilon_{\rm n}(\boldsymbol{k}) = (\hbar k)^2/(2m) + E_{\rm C}$ recombines with a p-type carrier with wavenumber $\boldsymbol{k}-\boldsymbol{q}$ and energy $\varepsilon_{\rm p}(\boldsymbol{k}-\boldsymbol{q}) = (\hbar(k-q))^2/(2{\rm m}) + E_{\rm V}$, which emits a photon with wavenumber $\boldsymbol{q}$ and energy $\hbar\omega_{\rm q}$. The energy of the conduction band $E_{\rm C}$ and that of the valence band $E_{\rm V}$ is measured from a common origin within the gap as shown in Fig. 2 (a). The energy conservation in such a process implies $\varepsilon_{\rm n}(\boldsymbol{k}) + \varepsilon_{\rm p}(\boldsymbol{k}-\boldsymbol{q}) = \hbar\omega_{\rm q}$. Since the speed of light is much larger than the Fermi velocity, a relation $|\boldsymbol{q}| \ll |\boldsymbol{k}|$ usually holds. The emission spectra per unit time are described well by the Fermi's golden rule within the first-order expansion with respect to the dipole interaction between an electron and a photon,

$$n_1(\omega_{\boldsymbol{q}}) = \frac{2\pi}{\hbar}\sum_{\boldsymbol{k}} |B|^2 \delta\left(\Omega_{\boldsymbol{q}} - \xi_{\boldsymbol{k}} - \xi_{\boldsymbol{k}-\boldsymbol{q}}\right), \tag{1}$$

$$\Omega_{\boldsymbol{q}} = \hbar\omega_{\boldsymbol{q}} - (E_{\rm C} + E_{\rm V} + 2\mu). \tag{2}$$

For simplicity of description, we have assumed that $\mu_{\rm n} = \mu_{\rm p} = \mu$ in this article. The constant B is the amplitude of the dipole interaction and is usually much smaller than the Fermi energy μ and the band gap energy $E_{\rm C}+E_{\rm V}$. We also assume that n- and p-type carriers are independent in their equilibrium characterized by μ. The energy of carrier $\xi_{\boldsymbol{k}} = (\hbar k)^2/(2m) - \mu$ is measured from the Fermi level. The δ-function in Eq. (1) implies the energy conservation law in the recombination process and gives the photon emitting condition. The result in Eq. (1) is derived based on the perturbation expansion theory [22]. The rough estimation of the expansion parameter of the phenomenon is as follows. The emission spectra are proportional to a ratio of $|B|/(\Omega_{\boldsymbol{q}} - \xi_{\boldsymbol{k}} - \xi_{\boldsymbol{k}-\boldsymbol{q}} + i\delta)$ which can be estimated as $|B|\delta(\Omega_{\boldsymbol{q}} - 2\xi_{\boldsymbol{k}}) \propto$

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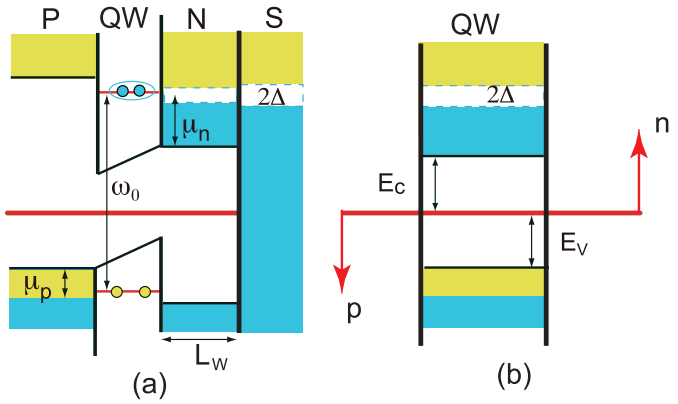

Fig. 2. A schematic picture of the superconducting p-n junction.

$|B|N(\Omega_{\boldsymbol{q}}) \approx |B|/\mu \ll 1$, where $i\delta$ is the small imaginary part and $N(\varepsilon)$ is the joint density of states (DOS) of n- and p-type carriers. Therefore contributions of the higher-order terms to the emission spectra are usually negligible. The total emitted photon number N_{ph} is calculated from

$$N_{ph}^{(1)} = \sum_{q} n_1(\omega_q). \tag{3}$$

The combined expansion parameter (λ_N) for $N_{ph}^{(1)}$ should include the DOS of photon at $E = 1\,\mathrm{eV}$ (Z_0) and the Einstein's A-coefficient ($A_E \approx 10^{10}\,[\mathrm{s}^{-1}]$) and is given by $\lambda_N = (\hbar/(2\pi))A_E/\mu$. By using $\mu \approx 100\,[\mathrm{meV}]$, we find $\lambda_N \approx 10^{-4}$. The small value of λ_N can be roughly understood by the above argument. A schematic picture of the energy diagram of a superconducting p-n junction is shown in Fig. 2 (a). The superconductor is attached to the n-type semiconductor. A quantum well (QW) was assumed as the luminescent area in this treatment. A Cooper pair penetrates into the QW due to the proximity effect [21] and recombines with two p-type carriers there. As a result, a pair of photons is emitted at the QW. This process clearly belongs to the second-order expansion in the perturbation theory. In the following, we try to draw a rough physical picture for the enhancement of such higher-order terms in superconducting p-n junctions. We assume the superconducting order parameter at the n-type region in the QW as shown in Fig. 2 (b).

We first qualitatively discuss the anomalous emission process for a pair of photons. The formation of Cooper pairs is of the essence in superconducting phenomena [23]. A Cooper pair consists of two electrons that have the wavenumber $\boldsymbol{k}$ with spin $\uparrow$ and the wavenumber $-\boldsymbol{k}$ with spin $\downarrow$. The number of Cooper pairs is unfixed in superconducting states, which leads to the coherent nature of superconducting condensate. This can be understood by the quantum mechanical uncertainty relationship between the fluctuations in number of pairs δN and those in macroscopic phase of superconducting states $\delta\varphi$, (i.e., $\delta N \delta\varphi \approx 1$). The superconducting state has properties similar to the classical coherent state of photons [24]. This fact plays an important role in the enhancement of emission spectra. Since the number of Cooper pairs is unfixed, the superconducting ground state remains almost unchanged even

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if a pair is removed for emitting a pair of photons. In other wards, the final state of a superconductor after emitting a pair of photons is very similar to the initial state before emitting. This is the background of resonant-like emission process of a pair of photons. In contrast to the superconducting state, the number of carriers is fixed in the normal state. When we remove two carriers from the N-carrier state, the number of carriers in the final state after emitting a pair of photons is $N-2$. The N-carrier state and $N-2$-carrier one are orthogonal to each other in the sense of quantum mechanics. Thus resonant-like emitting process for two photons is absent in the normal state. As a result, the contribution of such a higher-order process to the emission spectra is negligible.

To discuss the anomalous recombination quantitatively, we calculate the emission spectra based on the mean-field theory of superconductivity [25]. The elementary excitation from the superconducting ground state is described by

$$H = \sum_{\sigma=\uparrow,\downarrow} \sum_{\boldsymbol{k}} E_{\boldsymbol{k}} \gamma^{+}_{\boldsymbol{k},\sigma} \gamma_{\boldsymbol{k},\sigma}, \tag{4}$$

with $E_{\boldsymbol{k}} = \sqrt{\xi^2_{\boldsymbol{k}} + \Delta^2}$ being the excitation energy measured from the Fermi level, where $|\Delta|$ represents the superconducting gap and $\gamma^{+}_{\boldsymbol{k},\sigma}(\gamma_{\boldsymbol{k},\sigma})$ is the creation (annihilation) operator of the so-called *Bogoliubov quasiparticle* with (k,σ). The mean-field theory is the effective theory for low-energy excitation around the Fermi level. The energy $E_{\boldsymbol{k}}$ represents the gapped excitation spectra of quasiparticle. We note that the quasiparticle is not equal to the n-type electron. The relation between the electron and quasiparticle is defined by so-called Bogoliubov transformation.

$$c_{\boldsymbol{k},\sigma} = u_{\boldsymbol{k}} \gamma_{\boldsymbol{k},\sigma} - s v_{\boldsymbol{k}} \gamma^{+}_{-\boldsymbol{k},\bar{\sigma}}, \tag{5}$$

$$u_{\boldsymbol{k}}(v_{\boldsymbol{k}}) = \sqrt{\frac{1}{2}\left(1 + (-)\frac{\xi_{\boldsymbol{k}}}{E_{\boldsymbol{k}}}\right)}, \tag{6}$$

where $\bar{\sigma}$ denotes the opposite spin of σ and $s = 1$ and -1 for $\sigma = \uparrow$ and $\sigma = \downarrow$, respectively. The annihilation of an electron $(c_{\boldsymbol{k},\uparrow})$ is described by the linear combination of the annihilation of a quasiparticle $(\gamma_{\boldsymbol{k},\uparrow})$ and the creation of a quasiparticle $(\gamma^{+}_{-\boldsymbol{k},\downarrow})$. This relationship is a consequence of unfixed number of Cooper pairs. The annihilation of a Cooper pair includes the following terms

$$c_{\boldsymbol{k},\uparrow} c_{-\boldsymbol{k},\downarrow} \quad \rightarrow \quad -u_{\boldsymbol{k}} v_{\boldsymbol{k}} \gamma^{+}_{-\boldsymbol{k},\downarrow} \gamma_{-\boldsymbol{k},\downarrow} + u_{\boldsymbol{k}} v_{\boldsymbol{k}} \gamma_{\boldsymbol{k},\uparrow} \gamma^{+}_{\boldsymbol{k},\uparrow}. \tag{7}$$

We note that the left-hand side of the above equation also gives another terms like $\gamma^{+}\gamma^{+}$ and $\gamma\gamma$. However such terms do not give large contribution to the emission spectra. The remaining terms in Eq. (7) are diagonal of Bogoliubov quasiparticle. This means that removing a Cooper pair from the Bardeen Cooper Schrieffer (BCS) ground state does not disturb the ground state so much.

The recombination process of Cooper pairs is described by two steps. We count the energy of emission process of the first term in Eq. (7). Although we remove two electrons on the left hand side of Eq. (7), remaining terms on the

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right hand side create a quasiparticle and annihilate a quasiparticle. In the first step, an n-type carrier with $(-\boldsymbol{k},\downarrow)$ and a p-type carrier with $(-\boldsymbol{k}+\boldsymbol{q},\downarrow)$ recombine and emit a photon with $-\boldsymbol{q}$. This process annihilates a Bogoliubov quasiparticle with $(-\boldsymbol{k},\downarrow)$ as shown in the first term in Eq. (7). The energy of this state is $E_1 = \Omega_{\boldsymbol{q}} - E_{\boldsymbol{k}} - \xi_{\boldsymbol{k}-\boldsymbol{q}}$. In the second step, an n-type carrier with $(\boldsymbol{k},\uparrow)$ and a p-type carrier with $(\boldsymbol{k}-\boldsymbol{q},\uparrow)$ recombine and emit a photon with $\boldsymbol{q}$. In contrast to the first process, the second process creates a Bogoliubov quasiparticle with $(-\boldsymbol{k},\downarrow)$. Thus the energy of this states is $E_2 = \Omega_{\boldsymbol{q}} + E_{\boldsymbol{k}} - \xi_{\boldsymbol{k}-\boldsymbol{q}}$. The perturbation theory requires the energy conservation between the initial and the final state (i.e., $E_1+E_2=0$). Namely we obtain $\Omega_{\boldsymbol{q}} - \xi_{\boldsymbol{k}-\boldsymbol{q}} = 0$, which is the emitting condition of photons. The expansion parameters are $|B|/(\Omega_{\boldsymbol{q}} - E_{\boldsymbol{k}} - \xi_{\boldsymbol{k}-\boldsymbol{q}} + i\delta)$ from the first step and $|B|/(\Omega_{\boldsymbol{q}} + E_{\boldsymbol{k}} - \xi_{\boldsymbol{k}-\boldsymbol{q}} - i\delta)$ from the second step. Due to the emitting condition derived from the energy conservation law, the large energy term in the denominator is completely removed. As a result, the expansion parameter in the two successive emission process approximately becomes $(|B|/E_{\boldsymbol{k}})^2 \approx |B/\Delta|^2$ which is no longer a small value. After considering contributions of the similar terms, the emission spectra become [25]

$$n_2(\omega_{\boldsymbol{q}}) = \frac{2\pi}{\hbar^2}\sum_{\boldsymbol{k}} |B|^4 \delta(\Omega_{\boldsymbol{q}} - \xi_{\boldsymbol{k}-\boldsymbol{q}}) \frac{\Delta^2}{E_{\boldsymbol{k}}^4}. \tag{8}$$

In the same way as in Eq. (3), the emission intensity $N_{ph}^{(2)}$ is obtained by summation over q in Eq. (8). The ratio of the first- and second-order terms $\lambda_S = N_{ph}^{(2)}/N_{ph}^{(1)}$ can be estimated as $\hbar A_E N_0/(Z_0\Delta)$, where N_0 is the density of states for n-type carrier at the Fermi level. By putting reasonable values into the ratio, we find λ_S is of the order of unity. The width of emission spectra in Eq. (8) is given by the bandwidth of p-type carrier. This is because the recombination of Cooper pairs is possible only at the Fermi level of the n-type carrier.

To apply the above theoretical analysis to realistic junctions as shown in Fig. 2 (a), we take into account junction characters phenomenologically. The proximity effect in n-type semiconductor is considered through a factor $e^{-2L_{\rm W}/\xi_{\rm N}(T)}$ [26], where $\xi_{\rm N}(T) = \sqrt{\hbar D/2\pi T}$ is the coherence length in n-type region with $D = v_{\rm F}^2\tau/3$ and τ being the diffusion constant and the elastic-scattering relaxation time due to impurity scattering, respectively. $L_{\rm W}$ is the n-type semiconductor thickness. The n-type energy level in the QW stays at the Fermi energy of n-type semiconductor. When the p-type energy level is approximated with a simple Lorentzian lineshape characterized by Γ, we reach the phenomenological expression of emission spectra [25]

$$n_2(\omega_{\rm q}) \approx |B|^2 N_0 \tau^2 \frac{\Delta^2}{T} e^{-2L_{\rm W}/\xi_{\rm N}(T)} \frac{\Gamma}{(\omega_{\rm q}-\omega_0)^2 + \Gamma^2}, \tag{9}$$

where the emission spectral peak was given as ω_0.

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3 Experiments on superconducting light emitting diodes

For the experimental examination of the theoretical indication, one key issue is the reproducible injection of Cooper pairs into a semiconductor. The main factor to prevent the injection was the Schottky barrier at a metal-semiconductor interface. Conventional GaAs shows the barrier height of $\sim$ 900 meV and the reproducible Cooper pair injection is difficult. In the case of InGaAs, the Schottky barrier height is reduced to nearly zero for the In concentration of $\sim$ 80% [27] and the reproducible Cooper pair injection is possible. $In_{0.53}Ga_{0.47}As$ layers lattice matched to InP substrates emit at near 1.55-μm optical-fiber communication band and are advantageous to the potential future applications. Among metals showing superconductivity, intrinsic niobium (Nb) has relatively high transition critical temperature (T_C) of 9.26 K and is good for measuring a temperature-dependence property.

Cooper-pair injection was examined by measuring the Josephson junction (J-J) characteristics with the Nb/n-InGaAs/Nb superconductor(SC)/semiconductor/SC junctions formed on n-InGaAs/p-InP as shown schematically in Fig. 3 (a). Details of the device fabrication are given in Ref. [28]. The device was set in a ^{3}He closed-cycle cryostat. At the temperature of 30 mK, clear DC J-J characteristics was observed with the current (I)-voltage (V) measurement as shown in Fig. 3 (b). The slit width separating the two Nb contacts was 110 nm in this device and the measured critical supercurrent (I_C) was $\sim$ 1 μA. When the slit was reduced to 80 nm at later stage, I_C increased to $\sim$ 50 μA and the DC J-J characteristics was observable up to $\sim$ 7 K [29], and therefore the properties discussed here are not limited to 30 mK. With the irradiation of 8 GHz microwave, the Shapiro steps were observed as shown in Fig. 3 (b), demonstrating the AC J-J characteristics [30]. These observations demonstrate the successful injection of Cooper pairs into n-InGaAs layers. The next step is to confirm that Cooper pairs are flowing into the area close to the p-n junction. The device shown in Fig. 3 (a) functions as junction field-effect transistor (J-FET) at room temperature [31], that is, the depletion layer width at the p-n junction can be controlled with the bias at the back p-gate electrode. The I_C increased with the forward bias and decreased with the reverse bias, demonstrating that the Cooper pairs are flowing the area adjacent to the p-n junction [28, 32].

In order to investigate the Cooper-pair-based photon generation process, the device was forward biased with the back p-gate electrode for the LED operation. Generated photons, i.e., the electroluminescence (EL) output were observed through the slit formed on the surface between the two Nb electrodes. The LED I-V characteristic and the EL spectrum measured with the injection current of 250 μA at 4 K are shown in Fig. 4 (a). The superconductivity effect on the LED operation will be most critically evident at the temperature around T_C, and therefore the temperature dependence of the EL decay was examined. The decay time constant was measured with stepwise decrease from the constant current of 250 μA to the bias of 600 mV. This offset bias was determined to keep close to the flat-band condition of

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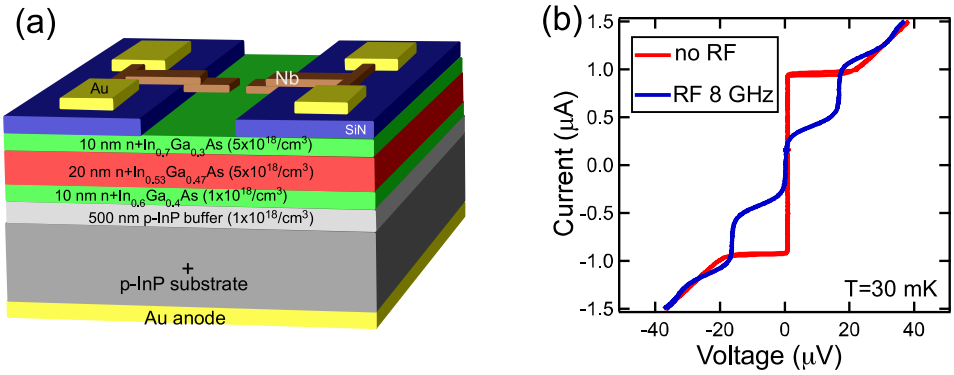

Fig. 3. (a) Schematic illustration of the superconducting LED. (b) Current-voltage characteristics of the Nb/n-InGaAs/Nb junction formed on the LED surface measured at 30 mK with [solid (red) curve] and without [solid (blue) curve] RF illumination.

the p-n junction with minimal injection current.

At the n-InGaAs/p-InP heterojunction, radiative process takes place in the n-InGaAs layer and the recombination rate is limited by the injection of the minority carriers of holes. The population dynamics of holes can be described by a simple rate equation, $\mathrm{d}p(t)/\mathrm{d}t = G(t) - p(t)/\tau_{\mathrm{LED}}$, where $p(t)$ and τ_{LED} are the number of holes and the EL decay time constant, respectively. $G(t)$ is the injection term and is equal to p_0/τ_{LED} under the steady-state condition, where p_0 is the steady-state number of holes. Since the bias circuit has the system response with the capacitance-resistance time constant (τ_{CR}), the stepwise bias reduction at $t = 0$ is expressed as $G(t) = p_0/\tau_{\mathrm{LED}} \cdot e^{-t/\tau_{\mathrm{CR}}}$ for $t \geq 0$). Then the measured transient EL intensity is given by [33]

$$I_{\mathrm{EL}}(t) \propto \frac{p(t)}{\tau_{\mathrm{rad}}} = \frac{p_0}{\tau_{\mathrm{LED}}}\eta_{\mathrm{int}}\left\{e^{-\frac{t}{\tau_{\mathrm{LED}}}} + \left(1 - \frac{\tau_{\mathrm{LED}}}{\tau_{\mathrm{CR}}}\right)^{-1}\left(-e^{-t/\tau_{\mathrm{LED}}} + e^{-t/\tau_{\mathrm{CR}}}\right)\right\}. \tag{10}$$

η_{int} is the internal quantum efficiency and is given by the balance of radiative and non-radiative recombination lifetimes, τ_{rad} and τ_{nonrad}, respectively, as follows: $\eta_{\mathrm{int}} = 1/\tau_{\mathrm{rad}}/(1/\tau_{\mathrm{rad}} + 1/\tau_{\mathrm{nonrad}}) = \tau_{\mathrm{LED}}/\tau_{\mathrm{rad}}$. The time-dependent term in Eq. (10) is simplified to the single exponential form of $e^{-t/\tau_{\mathrm{CR}}}$ under the condition of $\tau_{\mathrm{LED}}/\tau_{\mathrm{CR}} \ll 1$. This condition is realized by employing the internal-electric-field effect under the zero or reverse bias [34], which results in the shortening of τ_{nonrad} due to barrier leakage and the elongation of τ_{rad} due to spatial separation of electrons and holes by the Stark effect. τ_{CR} is thus experimentally determined to be 2.70 ns [28] and τ_{LED} is determined with Eq. (10) from measurements.

Firstly a reference LED with Au electrodes was studied. The measured decay time constants were almost temperature independent and the average is given as $\left\langle \tau_{\mathrm{LED}}^{\mathrm{normal}} \right\rangle = 2.25$ ns. On the other hand, the superconducting LED showed an abrupt change of τ_{LED} at the temperature around 7.5 K. The resistivity of the Nb electrode was measured employing the two neighboring

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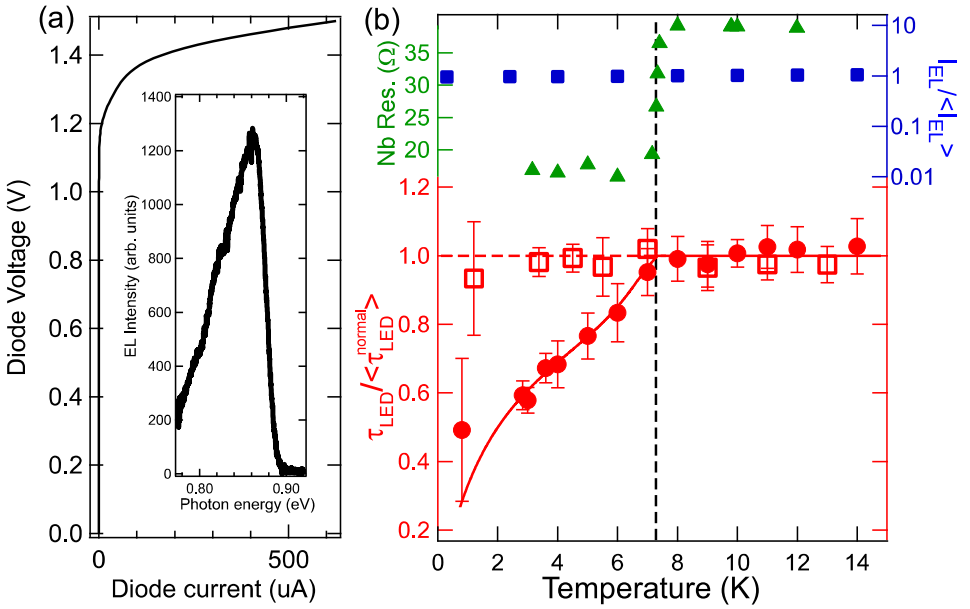

Fig. 4. (a) Diode characteristic at 4 K. Inset: EL spectrum measured at 4 K with constant injection current of 250 μA. (b) Temperature dependence of the normalized decay time. Solid (red) circles and open (red) boxes show the data on the LED with Nb electrodes and Au electrodes, respectively. The resistance [solid (green) triangles] measured along the Nb electrode indicates $T_C = 7.3$ K. Solid (blue) boxes show the temperature dependence of the steady-state integrated EL intensity. The solid (red) line is the fit with Eq. (11).

Au pads shown in Fig. 3 (a) and the result is shown in Fig. 4 (b). Its stepwise reduction at the temperature of 7.3 K indicates $T_C = 7.3$ K in this superconducting LED. Above T_C, τ_{LED} did not depend on the temperature and its average value was equal to $\left\langle \tau_{LED}^{normal} \right\rangle$. However it decreased abruptly below T_C. Figure 4 (b) shows the temperature dependence of $\tau_{LED} / \left\langle \tau_{LED}^{normal} \right\rangle$. At 0.8 K, $\tau_{LED} / \left\langle \tau_{LED}^{normal} \right\rangle$ is decreased down to ~ 0.5. This agreement of the transition temperature with T_C suggests the major role of the injected Cooper-pairs in the recombination process of the LED.

In spite of the significant change of the measured decay time constants below T_C, the integrated EL intensities in the steady state showed very weak temperature dependence as shown with the solid (blue) boxes in Fig. 4 (b). Since the integrated EL intensity is proportional to η_{int}, this shows that the internal quantum efficiency is constant but τ_{LED} is shortened below T_C. This uniquely determines τ_{LED} and η_{int} so that $\tau_{LED} \approx \tau_{rad} \ll \tau_{nonrad}$ and $\eta_{int} \approx 1$ [28, 33]. Therefore the drastic reduction of $\tau_{LED} / \left\langle \tau_{LED}^{normal} \right\rangle$ below T_C is the indication of the enhanced radiative process with superconductivity.

The observed radiative recombination enhancement is examined with the theoretical model described in the previous section. The total radiative pro-

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cess is the sum of the normal process and the one based on superconductivity and is given by

$$\frac{1}{\tau_{\rm rad}(T)} = \frac{1}{\tau_{\rm rad}^{\rm normal}} + A\frac{\Delta^2(T)}{T}e^{-2L_{\rm W}/\xi_{\rm N}(T)}. \quad (11)$$

The second term based on superconductivity on the right-hand side is given by the spectral integration of Eq. (9). The prefactors related to the dipole interaction and the DOS are summarized to the single prefactor A. The solid line in Fig. 4 (b) is the fit of Eq. (11) to the measurements. The coherence length of the electron Cooper-pairs penetrating into the n-InGaAs layer is estimated to be $\xi_{\rm N}(T) = 1570\,[K^{\frac{1}{2}}]/\sqrt{T}\,[{\rm nm}]$ in the dirty limit and is much longer than $L_{\rm W}$, the thickness of the n-type InGaAs layer, which is 40 nm in this device. Therefore the exponential term in Eq. (11) does not contribute to the temperature dependence. $\Delta(T)$ is the superconducting gap but is also called pair potential and the squared value is proportional to the Cooper-pair number in the superconductor. This factor appears because the number of the electron Cooper-pairs penetrating into the recombination region is proportional to this factor in the superconductor. The measured temperature dependence of the radiative lifetime is well reproduced with the fator A as a single fitting parameter. The temperature dependence below $T_{\rm C}$ is dominated by the factor $\Delta^2(T)/T$.

In our first trial of the superconducting LED, integrated EL intensities showed drastic enhancement below $T_{\rm C}$ [35], demonstrating the superconductivity effect on radiative process in semiconductors. However the decay time constants measured were nearly temperature insensitive [30]. This is attributed to the low internal quantum efficiency of the measured LED [33]. From the comparison of the two types of LEDs with low [35] and high [28] internal quantum efficiencies, we conclude that the increase of the luminescence intensity by the modification of the internal radiative processes manifests the low internal quantum efficiency of the measured device.

4 Future prospect

The agreement of the theory and experiment shown in Fig. 4 (b) demonstrates that photon pairs are generated with the radiative processes based on superconductivity. From the temperature dependence of the measured lifetimes on the superconducting LED, the ratio of the photon pairs generated from Copper pairs to classical photons generated from normal electrons is definitely calculated [33]. At 0.8 K in Fig. 4 (b), the ratio is nearly 50% in each process. With the further optimization of the LED structure such as the n-type doping levels in the n-type semiconductor, further increase of the photon-pair generation ratio is expected. Considering such mixed photon states in realistic solid-state photon sources, quality evaluation of the generated photon states is of great interest. In general, full knowlege of the two-photon states can be obtained by quantum-state tomography [36] measured under 16 polarization configurations. Since the two-photon states from the Cooper-pair-based radiative recombination can be expected to be a convex combination of the

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polarization-entangled photon pairs from the Cooper-pairs' contribution and totally mixed (uncorrelated) signal via normal current, the two-photon states can be expressed by the Werner state described by [37],

$$\rho_w = p\rho_{Bell} + (1-p)\rho_{mixed} \quad (0 \le p \le 1), \tag{12}$$

where p is a singlet weight, ρ_{Bell} is the density matrix for one of the maximally entangled Bell states, and ρ_{mixed} is the one of the totally mixed uncorrelated state. Therefore, the two-photon states can be evaluated under eight detection configurations, which could be further reduced in handling the temporally uncorrelated totally mixed state which is likely for solid-state quantum photon sources [38].

Any two-photon states can be analyzed by the degree of entanglement and mixedness, with tangle (T) [39] and linear entropy (S_{L}) [40], respectively. They are mapped onto the linear entropy (S_{L}) – Tangle (T) plane. For the Werner states, it follows the solid curve displayed in Fig. 5, and the position is determined by the singlet weight p. If the measured intensity correlation function can be divided unambiguously into entangled and mixed contributions, p can be directly determined. Under the horizontal-vertical (HV) detection polarization for example, the contribution of the entangled state to the coincidence count is given by $\mathrm{C}^{(\mathrm{Bell})}(p) = p\langle \mathrm{HV}|\rho_{\mathrm{Bell}}|\mathrm{HV}\rangle = p/2$, while the totally mixed state gives $\mathrm{C}^{(\mathrm{TM})}(p) = (1-p)\langle \mathrm{HV}|\rho_{\mathrm{TM}}|\mathrm{HV}\rangle = (1-p)/4$. Therefore the singlet weight p is given from a coincidence histogram as $\mathrm{C}^{(\mathrm{Bell})}/(\mathrm{C}^{(\mathrm{Bell})} + 2\mathrm{C}^{(\mathrm{TM})})$ and can be plotted on the Werner line in Fig. 5. As discussed in Ref. [37], the singlet weight p is related to the degree of entanglement, mixedness, and also Bell's S parameters. As for the degree of entanglement, $T = \left[\max\left(\frac{3p-1}{2}, 0\right)\right]^2$ holds for the Werner states and $p > 1/3$ is required for polarization- entangled state (red line region in Fig. 5)

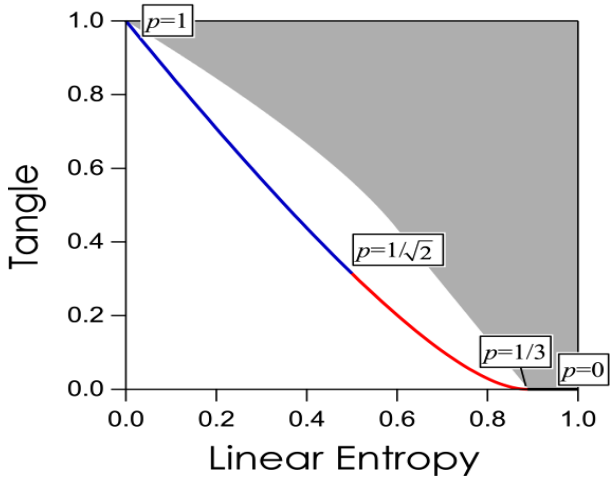

Fig. 5. Two-photon state mapping in the linear entropy (S_{L}) – Tangle (T) plane. Solid line indicates the Werner states, and the gray area corresponds to the states physically not realized. Singlet weights p for state boundaries are also shown.

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and $p > 1/\sqrt{2}$ for violating the Clauser, Horne, Shimony, and Holt (CHSH) inequality (blue line region) [41]. This criterion is expected to assist the optimization of the superconducting LED toward realizing high-efficiency EPP sources.

There still remain several challenging issues. The infra-red photon-pair emission is attractive for optical-fiber based communication, but high-quality single-photon detectors (SPDs) have been missing in this wavelength range. Conventional InGaAs avalanche photo diodes encounter high dark counts and the after-pulse effect. This has prevented our direct confirmation of the photon pair generation from the superconducting LED. However the performance of superconducting SPD is improving rapidly in recent years and the situation is improving. Our LED operation at present is based on QW structures and the incorporation of QDs is necessary for single EPP generation per excitation and the preparation and trials are under way. The LED structure presented in this article deals with electron Cooper pairs. However hole Cooper pair injection is also possible [42]. We are also trying to fabricate a LED structure with superconducting contacts to both n-type and p-type semiconductors [29]. When radiative recombination of both electron and hole Cooper pairs is realized, superradiance based on superconductivity will be realized [43] and ultra-bright EPP sources will be possible.

5 Conclusion

We have reviewed our recent research on photon-pair generation based on superconductivity. A theoretical model to deal with the radiative recombination of an electron Cooper pair with two holes was developed with the second-order perturbation theory. We found a peculiar second-order terms related to superconductivity, which showed drastic enhancement of the radiative process. Superconducting LEDs were fabricated and the electron Cooper-pair injection into n-InGaAs semiconducting layers was demonstrated with the observation of DC and AC J-J characteristics, and the observation of the gate-bias dependence of the critical supercurrent demonstrated that the injected Cooper pairs are flowing through the proximity area to the p-n junction. With the forward bias of the LED, drastic enhancement of the radiative process based on superconductivity was confirmed with the observation of the radiative lifetime reduction below T_C. The experimental observations were very well reproduced with the theory, demonstrating the generation of EPP in the superconducting LED. We have also developed the Werner state model that correlates the degree of entanglement of the generated EPP with the measured coincidence histograms of the paired photon states and the classical mixed states, which will be an optimization standard for realisitic solid-state photon sources.

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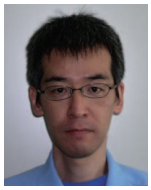

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