

OPEN ACCESS

A Cooper-Pair Light-Emitting Diode: Temperature Dependence of Both Quantum Efficiency and Radiative Recombination Lifetime

To cite this article: Ikuo Suemune *et al* 2010 *Appl. Phys. Express* **3** 054001

View the [article online](#) for updates and enhancements.

You may also like

- [Recombination dynamics in GaInN/GaN quantum wells](#)
Andreas Hangleiter
- [Analysis for non-radiative recombination and resistance loss in chalcopyrite and kesterite solar cells](#)
Masafumi Yamaguchi, Hitoshi Tampo, Hajime Shibata et al.
- [Review—Carrier Lifetime Spectroscopy for Defect Characterization in Semiconductor Materials and Devices](#)
E. Gaubas, E. Simoen and J. Vanhellemont

A Cooper-Pair Light-Emitting Diode: Temperature Dependence of Both Quantum Efficiency and Radiative Recombination Lifetime

Ikuo Suemune^{1,2*}, Yujiro Hayashi¹, Shuhei Kuramitsu¹, Kazunori Tanaka^{2,3}, Tatsushi Akazaki^{2,4}, Hiroataka Sasakura^{1,2}, Ryotaro Inoue^{2,5}, Hideaki Takayanagi^{2,5}, Yasuhiro Asano⁶, Eiichi Hanamura⁷, Satoru Odashima^{1,2}, and Hidekazu Kumano^{1,2}

¹Research Institute for Electronic Science, Hokkaido University, Sapporo 001-0020, Japan

²CREST, Japan Science and Technology Agency, Kawaguchi, Saitama 332-0012, Japan

³Central Research Laboratory, Hamamatsu Photonics, Hamamatsu 434-8601, Japan

⁴NTT Basic Research Laboratories, Atsugi, Kanagawa 243-0198, Japan

⁵Department of Applied Physics, Tokyo University of Science, Shinjuku, Tokyo 162-8601, Japan

⁶Graduate School of Engineering, Hokkaido University, Sapporo 060-8628, Japan

⁷Japan Science and Technology Agency, Kawaguchi, Saitama 332-0012, Japan

Received February 18, 2010; accepted April 13, 2010; published online May 14, 2010

A light-emitting diode (LED) in the optical-fiber communication band showed special features after replacing the n-type electrode with niobium (Nb) superconducting metal. Nb electrodes prepared on an InGaAs-based semiconductor surface formed a superconductor/semiconductor/superconductor junction, and the current-voltage characteristics exhibited both DC and AC Josephson junction properties. This was a result of the injection of electron Cooper-pairs into the n-InGaAs active layer of an LED. The drastic enhancement of the electroluminescence output observed below the Nb superconducting critical temperature, T_c , demonstrates the active role of electron Cooper-pairs in radiative recombination. Lifetime measurements of this LED and accurate evaluation of the luminescence output made it possible to estimate the radiative recombination lifetimes. A theoretical formula derived for the Cooper-pair radiative recombination accurately describes both the measured steep reduction of the radiative recombination lifetime and the observed enhancement of the internal quantum efficiency below T_c . This work will assist the development of interdisciplinary physics and new applications in superconductivity and optoelectronics. © 2010 The Japan Society of Applied Physics

DOI: 10.1143/APEX.3.054001

In conventional light-emitting diodes (LEDs), electrons and holes are incoherently injected by drift or diffusion. This leads to incoherent radiative recombination of electron-hole pairs at injection currents below the lasing threshold. When electrons instead form Cooper pairs, the injection occurs in the condensed coherent state and a remarkable change takes place.¹⁾ An LED with superconducting Nb electrodes exhibited a drastic enhancement of electroluminescence (EL) by more than one order of magnitude below the Nb superconducting critical temperature, T_c .²⁾ This demonstrated the possibility of a new LED operation principle: Cooper-pair-based radiative recombination. The “Cooper-pair LED” can generate entangled photon pairs for quantum information applications.³⁾ However, no quantitative examination of the quantum efficiency related to the observed EL intensity enhancement was given in the previous report, and there was no detailed discussion of the recombination processes.

This paper demonstrates electron Cooper-pair injection into the LED active layer by the proximity effect⁴⁾ during junction field-effect transistor (JFET) operation of the LED.⁵⁾ It also presents lifetime measurements in the LED structure, together with simultaneous precise measurements of the luminescence output intensities. This demonstrates a steep reduction of the radiative recombination lifetime and the enhancement of the internal quantum efficiencies below the superconducting critical temperature. A formula developed on the basis of a recent theoretical study on Cooper-pair radiative recombination^{6,7)} was in good agreement with the measured temperature dependences of the radiative recombination lifetimes and internal quantum efficiencies.

Figure 1(a) shows a schematic of the LED device. The n-InGaAs layers grown on a p-InP substrate consisted of a 10-nm-thick $\text{In}_{0.6}\text{Ga}_{0.4}\text{As}$ quantum well (QW) layer ($n \sim 1 \times$

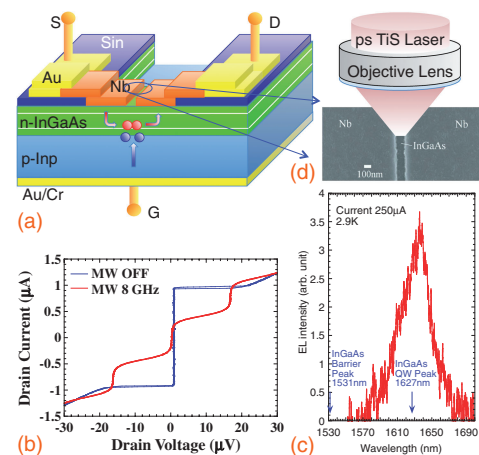


Fig. 1. (a) Schematic structure of a Cooper-pair LED. (b) S-D I - V characteristics measured at 30 mK with no gate bias. Under microwave (MW) irradiation, Shapiro steps were observed. (c) Measured EL spectrum. (d) 110-nm-wide slit dry-etched in a Nb electrode, and a schematic of a laser beam focused with an objective lens ($\times 50$ magnification and a numerical aperture of 0.42) for TR-PL measurement.

10^{18} cm^{-3}), a 20-nm-thick $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ barrier layer ($n \sim 5 \times 10^{18} \text{ cm}^{-3}$), and a 10-nm-thick $\text{n-In}_{0.7}\text{Ga}_{0.3}\text{As}$ contact layer ($n \sim 5 \times 10^{18} \text{ cm}^{-3}$). The thin contact layer with a higher In concentration improved the ohmic contact to the Nb metal without thermal annealing, because of the low Schottky barrier height.⁸⁾ The Nb electrode formed on the surface was $50 \mu\text{m}$ wide and 80 nm thick and was split into two parts with a 110-nm-wide slit formed at the center [Fig. 1(d)]. The p-InP substrate had an Au/Cr electrode at the bottom. The Cooper-pair injection was examined during JFET operation of the device, employing the two Nb electrodes as a source (S) and a drain (D) and the Au/Cr electrode as a junction gate (G). Gate-voltage control of the drain current flowing through the n-InGaAs channel layers

*E-mail address: isuemune@es.hokudai.ac.jp

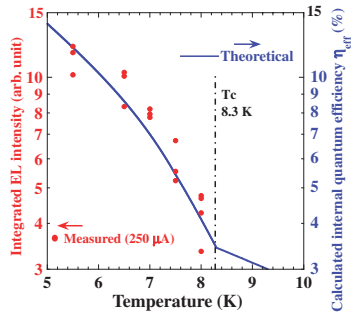


Fig. 2. Temperature dependence of integrated EL intensity and calculated internal quantum efficiency.

was previously demonstrated in a device with essentially the same structure.⁵⁾

The S–D current–voltage (I – V) characteristic measured at 30 mK is shown in Fig. 1(b) (indicated by “MW OFF”). The current flowing at zero drain voltage demonstrates the super-current flow through the n-InGaAs layers, and the voltage jump at a current of $\sim 1 \mu\text{A}$ to the normal state accompanied by slight hysteresis indicates the critical super-current, I_c . This is the DC Josephson junction property. Upon illumination by a microwave signal, Shapiro steps⁹⁾ were observed. An example, measured at 8 GHz, is shown in Fig. 1(b). The voltage step changed with the irradiation frequency f , following the relation $\Delta V/f = h/2e = 2.1 \mu\text{V}/\text{GHz}$, where h is the Planck constant and e is the electron charge. This combination of AC and DC Josephson junction properties demonstrates successful Cooper-pair injection into n-InGaAs semiconductor layers. No gate bias was applied in the case of Fig. 1(b), but the I_c value increased from ~ 1 to $\sim 2 \mu\text{A}$ with an increase of the forward bias to 0.8 V (not shown). This gate-bias modulation indicates that the super-current flows very close to the depletion layer at the p–n junction. This shows that Cooper-pair injection into the LED active layer is possible at a higher forward gate bias.

An EL spectrum measured with a gate current of $250 \mu\text{A}$ is shown in Fig. 1(c). The area of the InGaAs surface in direct contact with the Nb electrode was $50 \times 50 \mu\text{m}^2$. The EL output was observed from the 110-nm-wide slit between the S and D Nb electrodes. During this LED operation, a common bias was applied to the S and D. The emission peak at 1627 nm was close to the QW electron–hole ground-state energy difference.²⁾ T_c of the Nb electrode of this specific device was 8.3 K, and a drastic enhancement of the integrated EL intensity was observed in the temperature range below T_c , as shown by the solid circles in Fig. 2. To evaluate the relevant internal quantum efficiency, lifetime measurements were performed.

The EL decay of the LED was examined using electrical pulsed measurements. The EL decay was much faster than the time resolution of $\sim 2 \text{ ns}$, which was limited by the CR time delay of the measurement setup. Therefore, time-resolved photoluminescence (TR-PL) measurements were performed with a mode-locked Ti:sapphire laser with a pulse width of $\sim 5 \text{ ps}$. The repetition frequency and excitation wavelength were 76 MHz and 750 nm, respectively. The excitation laser spot was focused to $\sim 3 \mu\text{m}$ on the Nb slit by an objective lens (OL) [Fig. 1(d)], and the luminescence data was collected through the same OL (this measurement

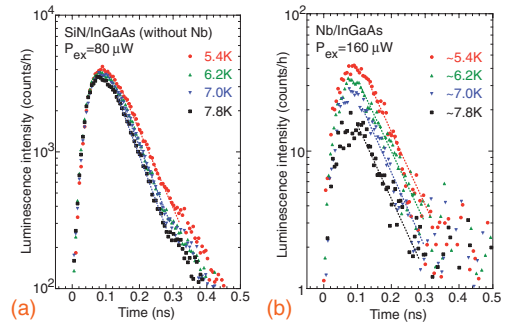


Fig. 3. Transient PL decay of InGaAs QW measured on (a) SiN passivated surface and (b) Nb with a slit opening.

is hereafter called Nb/InGaAs). It is also well-known that photo-excited Cooper pairs in superconductors recover themselves within $\sim 30 \text{ ps}$,¹⁰⁾ and the photo-excitation of Nb will not influence superconductivity. Similar measurements were performed on the InGaAs surface, which was covered with a SiN passivation layer, on the same LED device shown in Fig. 1(a) (called as SiN/InGaAs hereafter). The TR-PL measurements indicated that the gate was auto-biased at a photo-voltage over 0.8 V even with a weak average laser power of $0.1 \mu\text{W}$. A streak camera (Hamamatsu Photonics C9510) was used to detect photons around the wavelength of 1600 nm using a photon counting technique, and the time resolution was $\sim 20 \text{ ps}$.

The TR-PL decay measured on SiN/InGaAs is shown in Fig. 3(a). The spectrally integrated PL decay rate and the transient maximum PL intensities indicated a weak temperature dependence. Similar measurements of Nb/InGaAs showed a remarkable difference. The maximum PL intensity showed a distinct increase with decreasing temperature, as shown in Fig. 3(b). The measured time-integrated PL intensities and recombination lifetimes derived from Fig. 3(b) are summarized in Figs. 4(a) and 4(b), respectively. The measured lifetimes remained the similar level in both cases. The integrated PL intensity measured as the photon count rate, expressed as I_{output} , is related to the excitation laser power, P_{excite} , by the equation $I_{\text{output}} = a_{\text{eff}} \cdot \eta_{\text{eff}} \cdot [P_{\text{excite}} / \hbar\omega_{\text{excite}}]$. $\hbar\omega_{\text{excite}}$ is the incident laser photon energy, and η_{eff} is the internal quantum efficiency. The constant a_{eff} expresses the optical coupling efficiencies, such as the detection efficiency, the transmission efficiency of the optical measurement setup, and the excitation and photon extraction efficiencies related to the sample surface geometry. The former two factors are common to SiN/InGaAs and Nb/InGaAs. The third factor is influenced by the presence of the Nb electrodes for Nb/InGaAs, which acts as photo-shielding masks. The dependence of the output PL intensity on the photo-excitation power was examined in detail on the two cases. It was found that above T_c the two cases showed a good agreement with each other by setting the a_{eff} factor ratio as $a_{\text{eff,SiN}}/a_{\text{eff,Nb}} \approx 500$. This value is reasonable when the surface geometry and the carrier diffusion from the slit area are taken into account. The laser-beam irradiation diameter of $3 \mu\text{m}$ and the excitation through the ~ 100 -nm-wide Nb slit result in an excitation-area ratio of ~ 24 . The diffusion of photo-generated carriers from the slit area will reduce the carrier concentration by ~ 20 times employing the diffusion length of $\sim 1 \mu\text{m}$ in n-InGaAs.¹¹⁾ The total ratio of

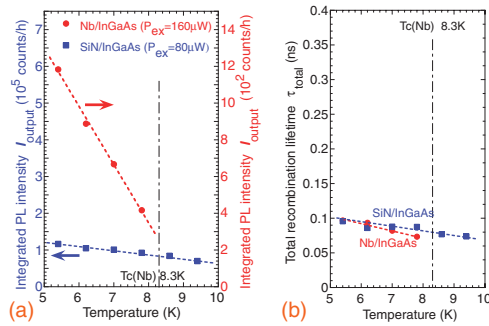


Fig. 4. Temperature dependence of (a) integrated PL intensity and (b) total recombination lifetime of Nb/InGaAs (closed circles) and SiN/InGaAs (closed squares).

~ 480 is close to the estimated ratio of 500. The average density of electron-hole pairs photo-generated in the Nb slit area in the present experiment is thus estimated to be $\sim 1.9 \times 10^{16} \text{ cm}^{-3}$. The vertical axes for the two cases in Fig. 4(a) are scaled considering this ratio. It will be clear that the EL enhancement below T_c shown in Fig. 2 is well reproduced with the present TR-PL measurements.

The internal quantum efficiency η_{eff} is related to the radiative and nonradiative recombination lifetimes of τ_{rad} and τ_{nonrad} , respectively, by the equation $\eta_{\text{eff}} = \tau_{\text{total}}/\tau_{\text{rad}}$, where $\tau_{\text{total}} = (\tau_{\text{rad}}^{-1} + \tau_{\text{nonrad}}^{-1})^{-1}$ is the total recombination lifetime. In an LED with a device structure similar to the present one, the radiative recombination lifetime τ_{rad} value at around 9 K (above T_c) was measured to be 2.25 ns.⁷⁾ Rationality of this value can be checked with the well-known formula $\tau_{\text{rad,without Nb}} = 1/(BN)$. Employing the B coefficient of $1.43 \times 10^{-10} \text{ cm}^{-3} \text{ s}^{-1}$ determined from a series of n- and p-type InGaAs measurements,¹²⁾ the corresponding majority-carrier (electron) density is calculated to be $3.1 \times 10^{18} \text{ cm}^{-3}$. This value is higher than $1 \times 10^{18} \text{ cm}^{-3}$ in the QW, but it is reasonable by considering the modulation doping effect from the neighboring barrier with the density of $5 \times 10^{18} \text{ cm}^{-3}$. Once a_{eff} is fixed employing this τ_{rad} value at 9 K, the temperature dependence of η_{eff} and τ_{rad} is directly calculated using the above-given formulae. The results are summarized in Fig. 5. It is found that the increase in internal quantum efficiency below T_c is strongly correlated with the steep reduction of the radiative recombination lifetime in Nb/InGaAs.

Physics of Cooper-pair-based radiative recombination was recently clarified with the second-order perturbation theory considering effects of superconductivity through the Bogoliubov transformation.⁶⁾ This theory demonstrated that the recombination of a Cooper pair with two p-type carriers (holes) induces the enhancement of luminescence intensity. The following analytical formula was derived for the Cooper-pair-based inter-band radiative recombination rate:^{6,7)} $W = A[\Delta_0^2(T)/T] \exp[-2L/\xi_N(T)]$, where A is a proportionality constant which includes the dipole moment for the optical transition, Δ_0 is the Nb superconducting gap, the square of which represents a quantity proportional to the Cooper pair number. ξ_N is the Cooper-pair coherent length in the n-InGaAs layers, and is much longer than the layer thickness L of $\sim 40 \text{ nm}$ in the present device.⁷⁾ Electrons in the energy band of Δ_0 around the conduction-band Fermi level will

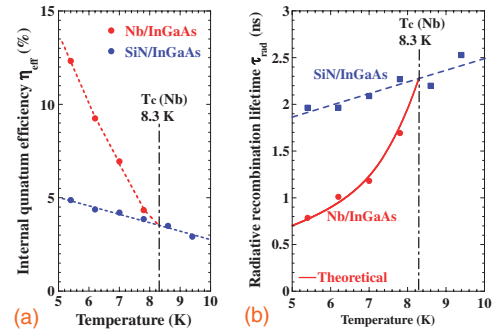


Fig. 5. Temperature dependence of (a) internal quantum efficiency and (b) radiative recombination lifetime. Solid line in (b) represents the theoretical calculation.

dominantly contribute to the condensation into Cooper pairs, and their concentration was roughly estimated to be $5 \times 10^{16} \text{ cm}^{-3}$ using the energy integral. The estimated injected minority carrier (normal hole) concentration was lower than this value in the present experiment. The radiative recombination lifetime including the normal component is given by $\tau_{\text{rad}} = (1/\tau_{\text{rad,without Nb}} + W)^{-1}$, and the calculated result is shown by the solid line in Fig. 5(b). This shows a very close fit to the observed steep reduction in the radiative recombination lifetime. The temperature dependence was dominated by the intrinsic property Δ_0 ,⁴⁾ and the only-one fitting parameter A merely changes the contribution of the Cooper-pair radiative recombination relative to the normal-carrier radiative recombination. The measured ratio of the Cooper-pair-based radiative recombination rate relative to the normal component increased from 0 at 8.3 K (T_c) to 1.50 at 5.4 K. The LED internal quantum efficiency was calculated with the assumption that the nonradiative recombination rate remained the same as during the TR-PL measurements, and this is shown by the solid line in Fig. 2. This fits the observed EL enhancement quite well and quantitatively converts the measurements into the internal quantum efficiency.

- 1) E. Hanamura: *Phys. Status Solidi B* **234** (2002) 166.
- 2) Y. Hayashi, K. Tanaka, T. Akazaki, M. Jo, H. Kumano, and I. Suemune: *Appl. Phys. Express* **1** (2008) 011701.
- 3) I. Suemune, T. Akazaki, K. Tanaka, M. Jo, K. Uesugi, M. Endo, H. Kumano, E. Hanamura, H. Takayanagi, M. Yamanishi, and H. Kan: *Jpn. J. Appl. Phys.* **45** (2006) 9264.
- 4) P. G. de Gennes: *Superconductivity of Metals and Alloys* (Westview Press, Oxford, U.K., 1999) p. 227.
- 5) Y. Hayashi, K. Tanaka, T. Akazaki, M. Jo, H. Kumano, and I. Suemune: *Phys. Status Solidi C* **5** (2008) 2816.
- 6) Y. Asano, I. Suemune, H. Takayanagi, and E. Hanamura: *Phys. Rev. Lett.* **103** (2009) 187001.
- 7) H. Sasakura, S. Kuramitsu, Y. Hayashi, K. Tanaka, T. Akazaki, E. Hanamura, R. Inoue, H. Takayanagi, Y. Asano, H. Kumano, and I. Suemune: arXiv:0910.5161v1.
- 8) K. Kajiyama, Y. Mizushima, and S. Sakata: *Appl. Phys. Lett.* **23** (1973) 458.
- 9) T. van Duzer and C. W. Turner: *Principles of Superconductive Devices and Circuits* (Prentice Hall, Upper Saddle River, NJ, 1998).
- 10) R. Sobolewski, A. Vervekin, G. N. Gol'tsman, A. Lipatov, and K. Wilsher: *IEEE Trans. Appl. Supercond.* **13** (2003) 1151.
- 11) E. Kuphal, K. Mause, K. Miethe, A. Eisenbach, F. Fiedler, and A. Corbet: *Solid-State Electron.* **38** (1995) 795.
- 12) R. K. Ahrenkiel, R. Ellingson, S. Johnston, and M. Wanlass: *Appl. Phys. Lett.* **72** (1998) 3470.