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

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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 the 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

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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 recombination6,7) 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 In0.53Ga0.47As barrier layer (n \( \sim \) 5 \( \times \) 10^{18} \text{cm}^{-3}), a 20-nm-thick In0.53Ga0.47As barrier layer (n \( \sim \) 5 \( \times \) 10^{18} \text{cm}^{-3}), and a 10-nm-thick n-In0.7Ga0.3As 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
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 supercurrent flow through the n-InGaAs layers, and the voltage jump at a current of ~1 µA to the normal state accompanied by slight hysteresis indicates the critical super-current, Ic. This is the DC Josephson junction property. Upon illumination by a microwave signal, Shapiro steps5) 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 ΔV/f = h/2e = 2.1 µV/GHz, where h is the Plank 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 Ic value increased from ~1 to ~2 µA with an increase of the forward bias to 0.8 V (not shown). This gate-bias modulation indicates that the supercurrent 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 µA is shown in Fig. 1(c). The area of the InGaAs surface in direct contact with the Nb electrode was 50 × 50 µm². 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. 20) Tc 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 Tc, 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 ~2 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 ~5 ps. The repetition frequency and excitation wavelength were 76 MHz and 750 nm, respectively. The excitation laser spot was focused to ~3 µm on the Nb slit by an objective lens (OL) [Fig. 1(d)], and the luminescence data was collected through the same OL, the pulsewidth of which was limited by the microscope objective.

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 Ioutput, is related to the excitation laser power, Pexcite, by the equation

\[ I_{\text{output}} = \alpha_{\text{eff}} \times \eta_{\text{eff}} \times \frac{P_{\text{excite}}}{h \omega_{\text{excite}}} \]  

where \( \alpha_{\text{eff}} \) and \( \eta_{\text{eff}} \) express 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 photoshielding 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 Tc the two cases showed a good agreement with each other by setting the \( \alpha_{\text{eff}} \) factor ratio as \( \alpha_{\text{eff,SiN}} / \alpha_{\text{eff,Nb}} \approx 500 \). 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 µm 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 ~1 µm in n-InGaAs. 11) The total ratio of
~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 \( \eta_{\text{eff}} \) is related to the radiative and nonradiative recombination lifetimes of \( \tau_{\text{rad}} \) and \( \tau_{\text{nonrad}} \), respectively, by the equation \( \eta_{\text{eff}} = \tau_{\text{total}} / \tau_{\text{rad}} \), where \( \tau_{\text{total}} = (1 / \tau_{\text{rad}} + 1 / \tau_{\text{nonrad}})^{-1} \) is the total recombination lifetime. In an LED with a device structure similar to the present one, the radiative recombination lifetime \( \tau_{\text{rad}} \) value at around 9 K (above \( T_c \)) was measured to be 2.25 ns.\(^5\) Rationality of this value can be checked with the well-known formula \( \tau_{\text{rad}, \text{without Nb}} = 1 / (BN) \). 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 \( \times 10^{18} \) cm\(^{-3}\). This value is higher than 1 \( \times 10^{18} \) cm\(^{-3}\) in the QW, but it is reasonable by considering the modulation doping effect from the neighboring barrier with the density of 5 \( \times 10^{18} \) cm\(^{-3}\). Once \( a_{\text{eff}} \) is fixed employing this \( \tau_{\text{rad}} \) value at 9 K, the temperature dependence of \( \eta_{\text{eff}} \) and \( \tau_{\text{rad}} \) 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.\(^{50}\) 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,\(^{6,7}\) \( W = A[\Delta_0^2(T) / T] \exp[-2L / \xi_{\text{c}}(T)] \), where \( A \) is a proportionality constant which includes the dipole moment for the optical transition, \( \Delta_0 \) is the Nb superconducting gap, the square of which represents a quantity proportional to the Cooper pair number. \( \xi_{\text{c}} \) is the Cooper-pair coherent length in the n-InGaAs layers, and is much longer than the layer thickness \( L \) of \( \sim 40 \) nm in the present device.\(^7\) Electrons in the energy band of \( \Delta_0 \) around the conduction-band Fermi level will dominantly contribute to the condensation into Cooper pairs, and their concentration was roughly estimated to be \( 5 \times 10^{16} \) 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 \( \tau_{\text{rad}} = (1 / \tau_{\text{rad}, \text{without Nb}} + W)^{-1} \), 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 \( \Delta_0 \),\(^4\) 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.

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