BIO Web of Conferences **129**, 32012 (2024) https://doi.org/10.1051/bioconf/202412932012
*EMC 2024*

# Analysis of photon bunching in coherent cathodoluminescence

Mr Sotatsu Yanagimoto[1], Dr Naoki Yamamoto[1], Dr Takumi Sannomiya[1], Dr Keiichirou Akiba[1,2]
[1]Tokyo Institute of Technology, Nagatsuta, Midoriku, Yokohama,, Japan, [2]National Institutes for Quantum Science and Technology, Watanuki, Takasaki, Japan

Background

Cathodoluminescence (CL) microscopy, utilizing an electron beam of an electron microscope, is a powerful approach for accessing nanoscale optical information of materials and devices. Furthermore, the time correlation measurement of CL by the Hanbury-Brown=Twiss (HBT) interferometer enables lifetime analysis without pulsing the electron beam by utilizing the photon bunching features [1]. This CL-HBT microscopy has been applied in various ways, such as lifetime measurements of semiconductors and dielectrics [2] and evaluations of the Purcell effect [3]. The origin of the bunching in CL has been investigated through a semiclassical description [4] and descriptions starting from quantum master equation [5] and is understood to be due to the random time modulation of excitation event by the fast electrons. The CL process can be classified into two classes depending on their excitation mechanisms, namely coherent CL (CCL) and incoherent CL (ICL). In the CCL process, fast electrons directly generate photons, e.g., transition radiation or Cherenkov radiation. On the other hand, in the ICL, photons are indirectly excited through mediator particles, such as bulk plasmons or secondary electrons, thus involving relaxation processes. Examples of the ICL are emission from defects and semiconductors. It is theoretically expected that the photon bunching can be observed even in CCL as far as emission events of two or more photons are included. However, most discussions on photon bunching using CL-HBT focused on ICL, and few studies have been reported regarding CCL. In this study, we experimentally confirmed the bunching phenomenon in CCL. By formulating the correlation function, we propose a method to extract the photon statistics in individual excitation events, excluding the bunching enhancement effect due to the excitation timing modulation.

Methods

Considering the semiclassical model, where n-photons are excited by a single electron, split by a half-beam splitter, and then detected by two single-photon detectors, the correlation function can be formulated. The analytical formula includes the parameters of electron beam current, correlation time and inherent correlation factor which reflects the fluctuation of the number of photons excited by a single electron. In CCL, it is expected that the photon statistics follow the Poisson distribution, and the inherent correlation factor should equal to unity. We performed this statistical analysis for localized surface plasmon (LSP) radiation, which is one of the typical examples of CCL, using a silver nanosphere with a diameter of approximately 200 nm, as schematically shown in Fig.1 (a).

We utilized a scanning transmission electron microscope (STEM) equipped with a CL detection unit and an HBT interferometer [3]. The light emitted from the sample was guided out of the STEM column by a parabolic mirror to the HBT measurement system. We performed the measurement at room temperature at an accelerating voltage of 160 kV.

Results

From the HBT measurement of the LSP radiation, we observed photon bunching, as shown in Fig. 1(b). The correlation time is approximately 400 ps, which reflects only the temporal resolution of our instrument, indicating that the lifetime of LSP is extremely short and cannot be resolved. We can observe a trend where the bunching height decreases with an increase in the electron beam current. Figure 1(c) shows an inversely proportional dependence of the correlation function on the electron beam current; such a tendency is described from our analytical formula. On the other hand, from Fig.1 (d), the values of the inherent correlation factors are approximately 1 for all the beam currents. This suggests that the photon statistics of LSP radiation follow the Poisson distribution, which is consistent with the theoretical predictions. Additionally, the independence of the inherent correlation factor on the electron beam current indicates that the pure photon statistics by a single electron excitation is effectively extracted by removing the effect of the time modulation of the excitation event by fast electrons.

Conclusions
Through the analysis of the correlation function obtained from the HBT measurement of CL, we successfully extracted the coefficient which reflects the fluctuation of the number of photons excited by a single electron. We believe that this method is useful to investigate the elementary processes of photon generation by fast electrons.

**Graphic:**

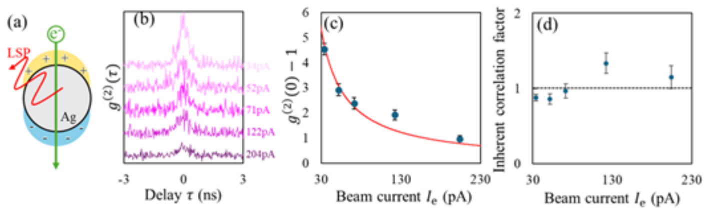


Fig.1 (a) Diagram of the LSP radiation from silver nano-sphere. (b) Correlation curves obtained with the varying electron beam current. (c), (d) Electron beam current dependency of the correlation function and the inherent correlation factor, respectively. The red solid line in (c) indicates the fitting curve with the function of inverse proportion.

**Keywords:**

Cathodoluminescence, Photon statistics

**Reference:**

[1] S. Meuret, et.al., Photon Bunching in Cathodoluminescence, Phys Rev Lett 114, (2015).
[2] S. Meuret, et.al., Lifetime Measurements Well below the Optical Diffraction Limit, ACS Photonics 3, 1157 (2016).
[3] S. Yanagimoto, et.al., Purcell Effect of Nitrogen-Vacancy Centers in Nanodiamond Coupled to Propagating and Localized Surface Plasmons Revealed by Photon-Correlation Cathodoluminescence, Phys Rev B 103, 205418 (2021).
[4] M. Sola-Garcia, et.al., Photon Statistics of Incoherent Cathodoluminescence with Continuous and Pulsed Electron Beams, ACS Photonics 8, 916 (2021).
[5] T. Yuge, et.al., PHYSICAL REVIEW B 107 , 165303 ( 2023 ) Superbunching in Cathodoluminescence : A Master Equation Approach, Phys Rev B 107, 165303 (2023).