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Photon Bunching in Cathodoluminescence

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We have measured the second order correlation function $[g^{(2)}(\tau)]$ of the cathodoluminescence intensity resulting from the excitation by fast electrons of defect centers in wide band-gap semiconductor nanocrystals of diamond and hexagonal boron nitride. We show that the cathodoluminescence second order correlation function $g^{(2)}(\tau)$ of multiple defect centers is dominated by a large, nanosecond zero-delay bunching ($g^{(2)}(0) > 30$), in stark contrast to their flat photoluminescence $g^{(2)}(\tau)$ function. We have developed a model showing that this bunching can be attributed to the synchronized emission from several defect centers excited by the same electron through the deexcitation of a bulk plasmon into few electron-hole pairs.

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In recent years, the emergence of nanoparticles and heterostructures with original optical properties has coincided with the development of new optical microscopy and spectroscopy techniques with subwavelength resolutions.

In this field, techniques such as scanning (transmission) electron microscopy—S(T)EM—and associated spectroscopies, are of particular interest. They benefit from the very high spatial resolution allowed by the picometer-range electron wavelength and ultimately from the spectral resolution arising through laser monochromaticity [1]. Indeed, cathodoluminescence (CL) [2] and electron energy-loss spectroscopy (EELS) [3,4] have emerged as major techniques for nanoplasmonic studies; understanding the similarities and differences between these methods and pure optical ones has led to an extremely fertile field of research [5,6]. On the other hand, CL in a scanning electron microscope (SEM) has long been a key technique for studying semiconducting materials [7]. Increasingly higher resolution SEMs and the emerging use of STEM-CL [8] makes CL a particularly suitable technique for nanophotonics applications [9–12]. However, contrary to the case of nanoplasmonics, the comparison between electron and photon based spectroscopies is almost unexplored in the field of nanostructured semiconductors.

When a semiconductor material is excited by a photon, the energy transfer immediately creates an electron-hole (e-h) pair that decays to the energy level of the most probable radiative transition in the material. Electron-matter interactions are more complex because they give rise to a range of primary excitations, essentially plasmons and hot e-h pairs, with varying energies. The number of primary excitations per incoming electron depends on the thickness of the sample and the energy of the electron. In typical STEM conditions, this number is close or even less than 1 and can be measured experimentally [13], making STEM-CL an appealing technique for unveiling the electron-matter interaction physics.

Various theories have been proposed to account for the chain of events leading from the electron-matter interaction to e-h creation [14,15] with the inclusion of plasmons, by far the dominant source of primary electron energy loss [16,17], as intermediate states [18,19]. This highlights major differences between photoluminescence (PL) and CL excitation mechanisms, although they seem to have no influence on the emission spectra, which are almost identical between both techniques [20]. Thus, the impact of plasmon decay mechanisms on measured optical quantities has been unclear up to now.

In this Letter, we show experimental evidence for the fundamental difference in the creation of e-h pairs between CL and PL signals. We have developed a new method relying on the measurement of the time correlation between the cathodoluminescence emission events from a sample excited by a low intensity fast electrons beam (energy ≈ 60 keV) in a STEM. We apply it to diamond particles containing a high density of defect centers (nitrogen vacancy, NV). We observed that at low electron currents ($I \leq 100$ pA), the second order auto-correlation function $[g^{(2)}_{\mathrm{CL}}(\tau) = \langle I(t)I(t+\tau)\rangle / \langle I(t)\rangle^2]$ of the CL signal intensity $I(t)$ displays a large, nanosecond-range peak at zero delay (bunching), which depends on the incoming electron current. This behavior strongly departs from the PL $g^{(2)}_{\mathrm{PL}}(\tau)$ function which is flat when multiple independent emitters are excited. We show that a model in which plasmons induce a synchronized emission from several emitters can account for this current-intensity-dependent bunching effect. The universality of the phenomemon is exemplified by the observation of $g^{(2)}(0)$ larger than 35 in CL from defect centers in hexagonal boron nitride (h-BN).

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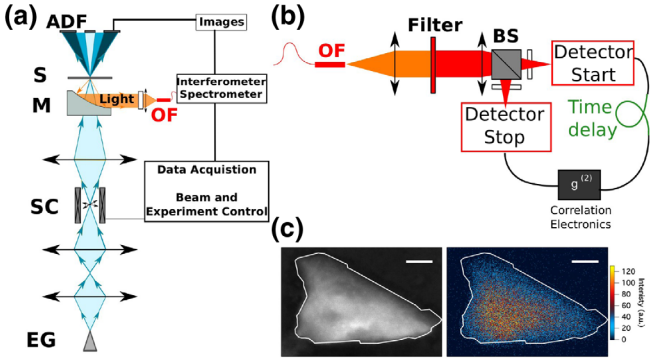

FIG. 1 (color online). CL-STEM setup. (a) A parabolic mirror (M) with a high numerical aperture has been incorporated inside the STEM to collect the CL signal efficiently, forming a collimated beam which is then coupled to a multimode optical fiber (OF). The fiber output beam is sent either to an imaging spectrograph to record the emission spectrum for each pixel, or to a Hanbury Brown–Twiss interferometer for $g^{(2)}_{\mathrm{CL}}(\tau)$ measurement shown in (b). Annular dark field (ADF) detector, sample (S), scanning coils (SC), electron gun (EG). (c) Simultaneously recorded ADF image (left) and filtered CL maps (right) of the nanodiamond studied in Fig. 2 (scale bar 50 nm).

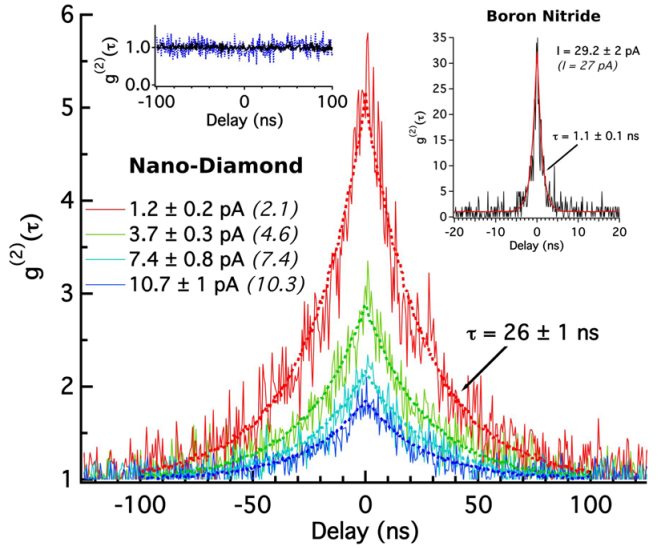

FIG. 2 (color online). $g^{(2)}_{\mathrm{CL}}(\tau)$ measurement (continuous lines) for intensities I ranging from 1.2 to 11 pA for an invididual ND, 60 nm thick. Each measurement lasted 5–20 min. The data were fitted with the Monte Carlo simulations explained in the text (dashed line). The lifetime τ_{NV} was retrieved using an exponential fit of the bunching curves, leading to $\tau_{\mathrm{NV}} \approx 26 \pm 1$ ns. All the parameters of the Monte Carlo simulation were kept fixed except the current, which was changed until a good agreement between the experiment and the simulation was found. The resulting fitted currents are written in parenthesis next to the measured experimental values. Inset, left: PL $g^{(2)}_{PL}(\tau)$ measurements performed at two different laser excitation powers on another ND from the same batch. Inset, right: $g^{(2)}_{\mathrm{CL}}(\tau)$ measurement and Monte Carlo simulation for defect centers in h-BN.

We measured $g^{(2)}_{\mathrm{CL}}(\tau)$ from several diamond nanocrystals [21] (nanodiamond, ND, with a mean size ≈100 nm) each containing a large number $N \approx 900$ of NV centers [22] using the same setup [Fig. 1(a)] as in Ref. [23]. Briefly, it allows one to measure the $g^{(2)}_{\mathrm{CL}}(\tau)$ with a Hanbury Brown–Twiss (HBT) interferometer [Fig. 1(b)] when individual emitters are excited with a nanometric 60 keV electron beam, in conjunction with various other signals, like filtered CL images, CL spectra or the morphological (ADF) images [Fig. 1(c)]. We used a bandpass filter (wavelength range 570–720 nm) to collect preferentially the emission of the neutral NV^0 center. We chose NV centers because their photophysical properties are well known, but the observed zero-delay photon bunching is universal, as exemplified on defect centers in h-BN (see Supplemental Material for details on these centers [24]).

Figure 2 displays a representative experimental data set. It shows $g^{(2)}_{\mathrm{CL}}(\tau)$ measured at different excitation current intensities (from $I = 1$ to $I = 10$ pA; curves with higher excitation currents display the same tendency, see Supplemental Material [24]) with a typical 20 mn integration time. The $g^{(2)}_{\mathrm{CL}}$ function is inferred from the normalization of the time-delay histogram to 1 at long time delays. A bunching [$g^{(2)}_{\mathrm{CL}}(0) > 1$] peak is observed at all intensities for the studied NDs. The $g^{(2)}_{\mathrm{CL}}(0)$ value increases upon decrease of the electron beam current. In sharp contrast, a $g^{(2)}_{\mathrm{PL}}(\tau)$ taken well below the saturation power in a confocal geometry results in a flat $g^{(2)}_{\mathrm{PL}}(0) \approx 1$ (inset of Fig. 2), as expected, because for a system made up of N emitters, $g^{(2)}_{\mathrm{PL}}(0) = 1 - 1/N$ [32]. $g^{(2)}_{\mathrm{CL}}(0) \approx 1$ is also retrieved at high excitation current (typically 200 pA, see Supplemental Material Fig. 2 [24]). The initial bunching effect with the same peak intensity value can be retrieved by diminishing the electron excitation current again, indicating that the latter effect is reversible. The characteristic time (extracted from an exponential fit, see below) lay between 10 and 30 ns in all the nearly 100 investigated NDs (26 ns in Fig. 2), which is compatible with the NV^0 center excitation lifetime in NDs [33], while for h-BN it lay between 1 and 3 ns, which is also compatible with known values [34].

Photon bunching is well known in PL. However, zero-delay bunching appears at the pico-or femtosecond time scales for thermal light [35], amplified spontaneous sources [36], and semiconductor microcavities in the strong coupling regime [37]; it has been also observed at the nanosecond time scale in specific cases (quantum dots in cavities [38] or coupled to plasmons [39], superradiant emitters in cavities [40]). Bunching is also observed in single-emitter fluorescence (organic dyes in particular) due to intersystem crossing from the excited state to metastable states, and in fluorescence correlation spectroscopy due to the fluctuations of the number of emitters in the excitation volume. However, these photon bunchings take place on nonzero delay time scales [41]. Finally, experimental zero-delay bunching larger than 3 has never been reported

to our knowledge. In summary, all previous reports of photon bunching depart from our observations, which therefore require a different interpretation.

Synchronizing the emission from multiple emitters may lead to bunching. Synchronized emission can happen when two or more centers are excited at the same time. It is worth noting that even for current values corresponding to less than 1 incoming electron per defect excited-state lifetime (typically $I < 24$ pA for NVs), the bunching effect is present. This clearly indicates that the photon correlations are not related to incoming electron correlations, but are the result of a single electron excitation mechanism.

In the case of a photon exciting a semiconductor, the excitation energy is only slightly larger than that of the emission. A single *e-h* pair is created, exciting at most one center. In contrast, an electron creates bulk plasmons of high energy ($E_p \approx 30$ eV in diamond) [13]. From this, there are two possibilities to explain the bunching. One is the creation of multiple *e-h* pairs originating from the same plasmon decay, an event made possible thanks to the high energy of the plasmon compared to the gap energy ($E_g \approx 5$ eV). The second is the creation of multiple e-h pairs that may stem from two different plasmons. Whatever the mechanism, all these *e-h* pairs may excite several centers at once. If at least 2 centers are excited by the recombinations of *e-h* pairs coming from the same single electron, there is a high probability that at least two photons will be emitted in a given time window. Therefore, each electron produces a packet of photons. If the electrons are sufficiently separated in time, the HBT interferometer will receive well-separated packets of photons, leading to bunching. As the incoming electron current increases, packets of photons produced by different electrons will arrive closer and closer in time until they become indistinguishable. Thus, increasing I will blur the bunching effect. As the electron arrival time statistics are Poissonian, at high I, we have $g^{(2)}_{\mathrm{CL}}(\tau) \approx g^{(2)}_{\mathrm{PL}}(\tau)$. The time scale of a packet of photons produced by one electron is approximately the lifetime of the emitters; thus, the width of the bunching peak is related to the emitter lifetime.

Based on this analysis, we developed a Monte Carlo method to calculate the photon emission probability as a function of time for an emitter excited by electrons. We then retrieved the $g^{(2)}_{\mathrm{CL}}(\tau)$ function by calculating the delay between emitted photons. Because the exact number per plasmon and the mechanism of *e-h* pair production vary in the literature [17,42], we evaluated several models in which we changed the plasmon decay mechanism, with either exactly n *e-h* pairs produced per plasmon or a Poissonian distribution of *e-h* pairs with mean value n. Taking into account Refs. [17,42] we present results with $n = 1$ or 3, but we obtained similar conclusions for other n values.

In all cases, we define a chain of events made up of (i) the creation of plasmons by an electron and their femtosecond decay into *e-h* pairs, with probability P^n_{el} [13]:

$$P^n_{\mathrm{el}}(L/\lambda_{\mathrm{e}}) = \frac{1}{n!}\left(\frac{L}{\lambda_{\mathrm{e}}}\right)^n e^{-L/\lambda_{\mathrm{e}}}, \tag{1}$$

where L/λ_{e} is the ratio of the thickness traversed by the electron to its mean free path for plasmon excitation, (ii) the excitation of centers by the *e-h* pairs happening a few picoseconds later with probability $P_{\mathrm{int}}(N, l_d, \sigma)$, with l_d the diffusion length, ρ the defect density considering the material as homogeneous, and σ the absorption cross section:

$$P_{\mathrm{int}}(N, l_d, \sigma) = 1 - e^{-l_d \sigma \rho}, \tag{2}$$

and (iii) the radiative relaxation, at time t_{rad} for a center (lifetime τ_e) excited at time t_0, with probability P_{rad}:

$$P_{\mathrm{rad}}(t_{\mathrm{rad}}, t_0) = \frac{1}{\tau_e} e^{-(t_{\mathrm{rad}} - t_0)/\tau_e}. \tag{3}$$

In the simulations we consider that the number N of centers likely to absorb an *e-h* pair is constant, because the number of excited centers is always negligible compared to N.

Whatever the parameters, the simulations exhibit a bunching profile converging to 1 at large delays, proving that multiple excitations by a single electron are indeed responsible for the bunching effect. Moreover, they show that the curves can be fitted with an exponential curve, the decay parameter of which is precisely equal to τ_e. Finally, they show the negligible influence of the value of P_{int} over several orders of magnitude, on the value of $g^{(2)}(0)$ (see Supplemental Material [24]), and therefore that the precise values of the parameters ρ, σ, and l_d are not needed to determine τ_e and $g^{(2)}(0)$.

In order to compare the accuracy of the different models (Poissonian or not) we proceeded as follows. First, τ_e was determined by fitting an exponential to an experimental $g^{(2)}_{\mathrm{CL}}(\tau)$ at a given I. τ_e is then kept fixed for the curves acquired for other I. L/λ_e is directly measured from EELS data (see Supplemental Material [24]). The current is then determined as the only free parameter (we took ρ from Ref. [22], $\sigma \approx 95$ nm^2 [43], and $l_d \approx 50$ nm for NDs [44]) by comparing a series of simulations to experimental curves. The accuracy of the different models can thus be inferred from the comparison between the fitted and simulated results.

As seen in Fig. 3, all the simulations reproduce the bunching effect, again confirming that multiple excitations by a single electron are responsible for the effect. Models in which the number of *e-h* pairs per plasmon is constant underestimate the bunching effect. Models assuming a Poissonian distribution for the plasmon decay seem to work better, highlighting the complexity of plasmon decay in the materials, but without giving any reason or insight into it. It may be due to stochastic phonon–*e-h* interaction, or to the creation of Auger and secondary electrons for example. Further studies, outside the scope of the present Letter, would be needed to clarify the exact plasmon decay mechanisms. In contrast with PL, the bunching effect is thus intrinsic to CL physics and needs to be taken into account when performing HBT experiments in STEM.

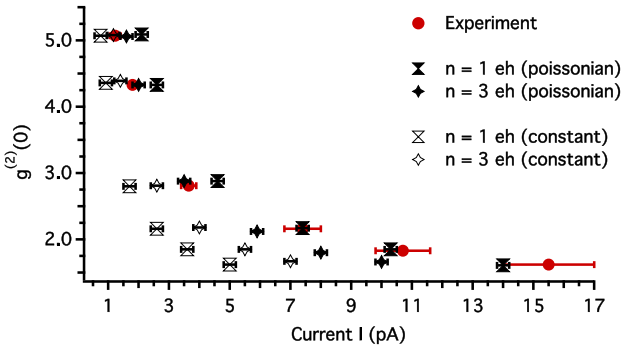

FIG. 3 (color online). $g^{(2)}_{\mathrm{CL}}(0)$ calculated from Monte Carlo simulations within the two different models for the decay mechanism (see text) compared to experimental data points extracted from Fig. 2. For all the simulations, all parameters are fixed except the current. Error bars in the simulations are generated by introducing a ± 1 ns error in the estimate of τ_e and a very conservative $\pm 10\%$ error on L/λ_e. The bunching effect is qualitatively reproduced by both models.

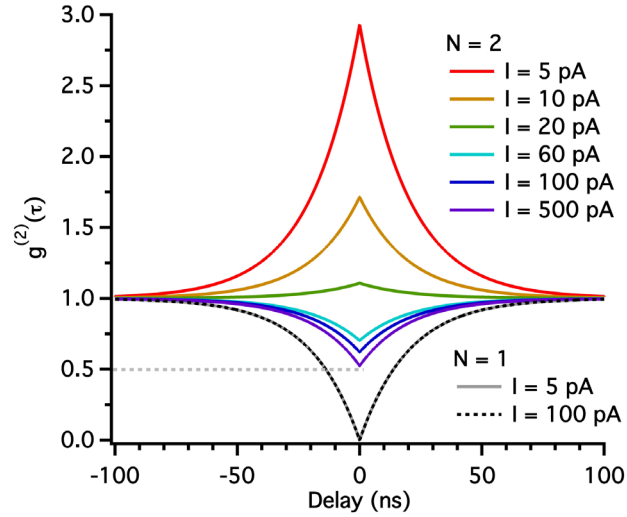

FIG. 4 (color online). Simulation for a ND with $L = 30$ nm and $\tau_e = 20$ ns. For $N = 1$ the $g^{(2)}_{\mathrm{CL}}(\tau)$ is independent of the probe current. For $N > 1$ (here $N = 2$) the expected antibunching behavior is retrieved at high current but not at low current.

Finally, we turn to the low N regime, in which antibunching [23] and bunching compete. We modeled phenomenologically the behavior of the $g^{(2)}_{\mathrm{CL}}(\tau)$ function for a small number of centers ($N < 5$) and a thickness of 30 nm, which means that the number of plasmons per electron can be considered to be equal to 1. The equation after simplification is given by (see details in the Supplemental Material [24])

$$g^{(2)}(\tau) = \frac{I_0}{I \times P^1_{\mathrm{el}}}\left(1 - \frac{1}{N}\right)e^{-\tau/\tau_e} + \left(1 - \frac{1}{N}e^{-\tau/\tau_e}\right), \quad (4)$$

where I_0 is the incoming electron current required to have one electron per lifetime. The first term in Eq. (4) represents correlations due to electron excitation and thus is responsible for the bunching effect. The second term, identical to the $g^{(2)}_{PL}(\tau)$ expression, represents the correlation due to the sample emission and is responsible for the antibunching behavior for single photon emitters. We can clearly see that when the current increases, the bunching part is blurred, leading to a $g^{(2)}(\tau)$ function similar to that for PL [45]. We also note that the expression formally diverges at zero current. However, at such a value, the bunch of photons is emitted with an infinite delay, preventing such a diverging limit from being reached experimentally. Also, Eq. (4) indicates the possibility of getting large $g^{(2)}(0)$ for short lifetimes (large I_0), as clearly demonstrated for defect centers in h-BN (Fig. 2). The $g^{(2)}_{\mathrm{CL}}(\tau)$ for $N = 1$ and $N = 2$ are shown in Fig. 4 for different currents. An individual NV center in a particle can only be excited by one e-h pair at a time, leading to antibunching with $g^{(2)}_{\mathrm{CL}}(0) = 0$ for all currents. If more than one center is present, bunching effects will appear superimposed on the antibunching behavior. For $N = 2$, a transition from bunching to antibunching is expected upon increasing I. This shows that the law $g^{(2)}(0) = 1 - 1/N$ is difficult to observe in CL-STEM. The transition between the bunching and antibunching is also difficult to observe experimentally, as the necessary integration time to obtain a meaningful $g^{(2)}_{\mathrm{CL}}(\tau)$ (typically 5 min) is large enough to induce radiation damage for a small number of defect centers.

In conclusion, we have shown that $g^{(2)}_{\mathrm{CL}}(\tau)$ of defect centers in two examples of wide band-gap semiconductors exhibits a bunching behavior depending on the excitation current and the lifetime of the emitter. This phenomenon has been shown to arise from the synchronization of multiple excitations via primary excitation decay into multiple e-h pairs and should be observable in any luminescent system by CL. Moreover, we believe that the autocorrelation shape with a single peak and a large $g^{(2)}(0) \gg 2$ could be recovered with an optical excitation, if emitters are excited with intense short laser pulses which are randomly separated in time. The ability to model the emitter lifetime with a simple exponential fit opens the way to easy and quantitative measurement of any emitter lifetime with a spatial resolution only limited by e-h pair diffusion lengths [46] and with a high signal-to-noise ratio. Finally, the high observed $g^{(2)}(0)$ value is independent of the emission wavelength (tunable by material choice), meaning such photon sources could be of interest in two-photon excitation fluorescence [47], where the amplitude of the effect depends linearly on $g^{(2)}(0)$.

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