Fiber optic probe of free electron evanescent fields in the optical frequency range

Jin-Kyu So,[1,a)] Kevin F. MacDonald,[1] and Nikolay I. Zheludev[1,2]
[1]*Optoelectronics Research Centre and Centre for Photonic Metamaterials, University of Southampton, Highfield, Southampton SO17 1BJ, United Kingdom*
[2]*Centre for Disruptive Photonic Technologies, Nanyang Technological University, Singapore 637371*

(Received 4 December 2013; accepted 24 February 2014; published online 19 May 2014)

We introduce an optical fiber platform which can be used to interrogate proximity interactions between free-electron evanescent fields and photonic nanostructures at optical frequencies in a manner similar to that in which optical evanescent fields are sampled using nanoscale aperture probes in scanning near-field microscopy. Conically profiled optical fiber tips functionalized with nano-gratings are employed to couple electron evanescent fields to light via the Smith-Purcell effect. We demonstrate the interrogation of medium energy (30–50 keV) electron fields with a lateral resolution of a few micrometers via the generation and detection of visible/UV radiation in the 700–300 nm (free-space) wavelength range. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4876395]

Light usually takes the form of propagating waves but can also exist in evanescent form under certain conditions. In near-field optical microscopy, imaging resolutions far beyond the diffraction limit are achieved by sampling these fields through nano-scale apertures. The technique is now in widespread use following its first demonstration almost thirty years ago[1] and continues itself to be the subject of research, with novel variants on the concept continuing to emerge.[2–4] Indeed, general interest in the physics of optical evanescent fields has grown substantially in recent years, most notably in the context of "perfect/super-lenses,"[5,6] optical forces,[7–11] and the entire field of plasmonics and photonic metamaterials.[12–15]

Moving free electrons also carry exponentially decaying "evanescent" fields[16–18] that may interact with nanostructures in the same manner as optical evanescent fields. The limited awareness of these electron fields is mainly derived from the fact that they decay on the sub-optical-wavelength scale (except in the case of highly relativistic electrons[19,20]), making experimental studies based on the visible/near-infrared frequency components of such fields extremely challenging. Indeed, while the nanoscale range of low/medium energy electron evanescent fields has provided for the emergence of electron-induced radiation emission (EIRE) and electron energy loss (EELS) spectroscopy techniques for probing and mapping the photonic and plasmonic properties of nanostructures,[18,21–24] it places constraints on the experimental geometry in such studies and on the utility of free-electron pumping in applications such as nanoscale tunable light sourcing.[25–27]

Here, we report on an experimental platform using a tapered optical fiber geometry (Fig. 1) akin to those employed for sampling evanescent light fields in near-field optical microscopy, which enables the study of proximity interactions between free electrons and nanophotonic structures mediated by the aforementioned exponentially decaying field at optical frequencies. As a proof-of-principle, in the present study, the evanescent fields of medium energy free electrons are diffractively decoupled into propagating fiber modes by a nano-grating. In the context of EIRE spectroscopy, such fiber probe tips may act simultaneously as a substrate for target nanostructures and a close-coupled (i.e., near-field) collector for the light generated, eliminating the need for independent light-collection optics, which are highly positionally sensitive (requiring precise alignment of optical and electron beam foci in three dimensions) and intrusive (often compromising electron beam working distance and therefore achievable spot size at sample).

A free electron moving at a velocity v carries a time-varying electromagnetic field that can be described using Maxwell's equations.[16–18] In cylindrical coordinates, the transverse and longitudinal components of this field are proportional to $K_1(\omega x/v\gamma)$ and $\gamma^{-1}K_0(\omega x/v\gamma)$ where K_0 and K_1 are the modified Bessel functions of the second kind, ω is the frequency of the electromagnetic field, x is the distance from the electron, and γ is the Lorentz factor.[18] The

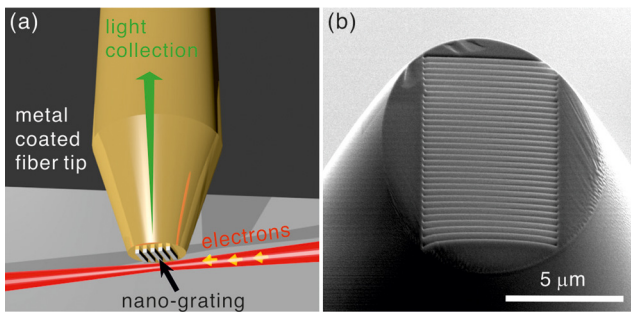

FIG. 1. (a) Artistic impression of an optical fiber probe for electron evanescent fields: light is generated via the Smith-Purcell effect as free-electrons pass a nano-grating on the end facet of a conically tapered, metal-coated fiber tip. (b) Electron microscope image of a 256 nm period grating (±6 nm as measured from electron microscope images) fabricated by focused ion beam milling on the end facet of a Cr-coated truncated conical fiber probe tip (cone angle = 78°).

[a)]Author to whom correspondence should be addressed. Electronic mail: js1m10@orc.soton.ac.uk.

dispersion of the field is given by $\omega = vq$ where q is the wave vector component in the propagation direction. This implies that the electron's field remains evanescent unless the speed of light in the surrounding medium is less than the electron velocity, in which case Cerenkov radiation will be generated,[28] or unless additional momentum is provided via scattering at an optical inhomogeneity, leading in the specific case of a grating to so-called diffraction[19] or Smith-Purcell (SP) emission.[20,29] The SP mechanism is well-understood and readily applied to medium-energy electrons using low-index optical media and is therefore employed in the present study as a controlled means of coupling electron evanescent fields to propagating guided modes in an optical fiber probe.

A grating with a period d provides additional parallel momentum in integer multiples (n) of its reciprocal lattice vector, $2\pi/d$, to incoming electromagnetic waves, leading to diffraction phenomena. The same applies to free electron evanescent fields, which have a modified dispersion $\omega = v[q + 2\pi n/d]$. Some portion of the electromagnetic energy can thus be decoupled into propagating waves when the parallel momentum is smaller than that of light in free space: $|\omega/v - 2\pi n/d| < \omega/c$. This condition leads to the angular dispersion of so-called Smith-Purcell emission

$$\lambda = \frac{d}{n}\left(\frac{1}{\beta} - \cos\theta\right), \tag{1}$$

where $\beta = v/c$ is the velocity of electron relative to the speed of light, and θ is the angle between the direction of light emission and the electron propagation direction.[29] Thus, the wavelength of light emitted in a given direction can be tuned by varying the grating period or the kinetic energy of incident electrons. Indeed, on the basis of its scalability and broad tunability (experimental observations extend from ultraviolet to microwave wavelengths[30,31]) the Smith-Purcell emission mechanism has been regarded as a promising basis for compact free-electron-lasers[31–33] and non-destructive diagnosis of the spatial coherence lengths of pico- or femtosecond electron bunches in accelerator facilities.[34,35]

In the present study, nano-gratings with a period of 256 nm were used to generate first- and second-order SP emission in the UV-visible part of electromagnetic spectrum from electrons with energies in the 30–50 keV range (the beam of a scanning electron microscope—SEM, operating in fixed-spot mode) passing the probe tip as illustrated in Fig. 1(a). Nano-gratings are formed on the end facets of optical fibers with a truncated conical profile, which provides for the probe tips to be positioned within the evanescent range of the electron beam waist without the bulk of the fiber clipping the incident beam, while emitted light is collected via the fiber.

Probes were prepared from standard multi-mode optical fiber (Thorlabs, SFS50/125 Y; 250–1200 nm; 0.22 NA) by laser cleaving, to form a conical tip, and focused ion beam (FIB) milling, to create a flat, smooth surface 5–10 μm in diameter. A 40 nm thick chromium film was then deposited on the fiber by electron-beam evaporation and patterned by FIB again to form a grating as shown in Fig. 1(b) (the grating lines being milled through the full depth of the Cr film). In the SEM, the fiber is aligned such that the trajectory of the electron beam (waist diameter ~50 nm; electron energy 30–50 keV; beam current 1–20 nA; dwell time 10 s) runs parallel to the end facet of the fiber probe and perpendicular to the grating lines as shown in Fig. 1(a). Light generated in the surface-normal direction is collected by the fiber and delivered to a spectrometer equipped with a liquid-nitrogen-cooled CCD array detector.

Figure 2(a) shows the emission spectrum for 38 keV electrons passing a nano-grating probe tip along a fixed trajectory with a grazing impact parameter (nominally equal to the beam radius, i.e., within available accuracy of SEM imaging and spot-mode beam targeting, as close as possible

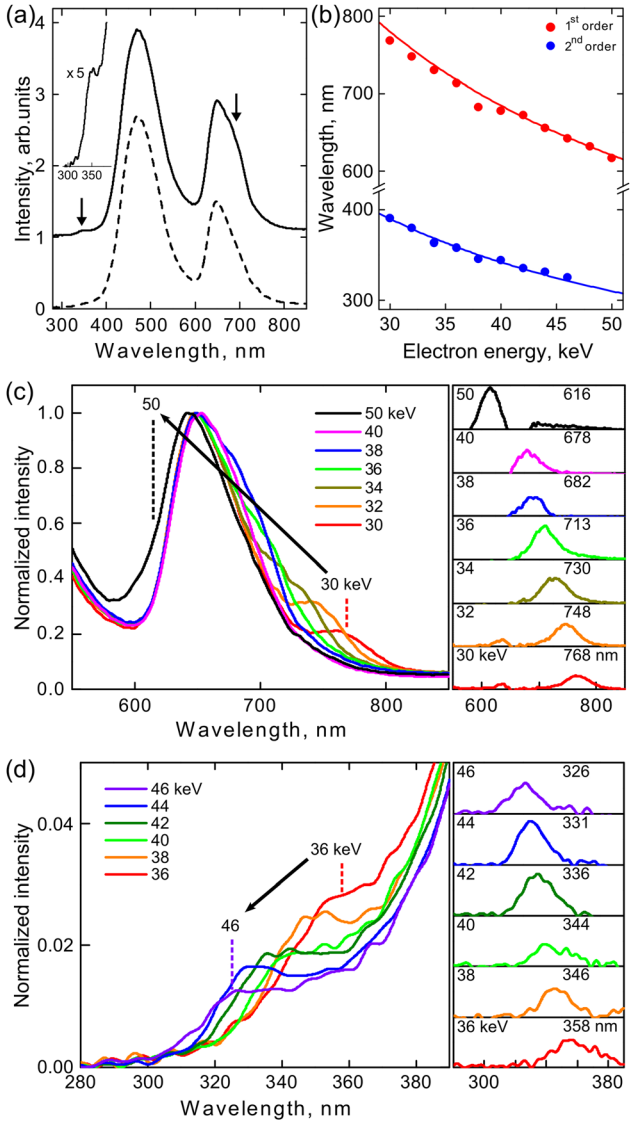

FIG. 2. (a) Emission spectra for 38 keV electrons (beam current 6.15 nA) moving along a grazing trajectory over a flat, unstructured, 40 nm thick chromium layer (dashed line) and a 256 nm period grating formed in an identical Cr film (solid line) on truncated conical fiber probe tips (for clarity the spectra are offset vertically by 1000 counts). The inset shows detail of the feature at ~350 nm in the grating spectrum. (b) First- and second-order SP emission wavelengths as a function of electron energy. Data points denote experimental measurements while the solid lines are derived from Eq. (1). (c) and (d) Detail of the first- (c) and second-order (d) SP emission features for a selection of electron energies in the range between 30 and 50 keV (grazing impact parameter; beam current 5–8 nA) in spectra normalized to the adjacent silica cathodoluminescence peak (main panels) and series of reduced spectra with the CL background subtracted (right, each labeled with beam energy and peak wavelength).

without impacting primarily on the side wall of the fiber) alongside the spectrum for an unstructured region of the Cr-coated fiber tip. The two major peaks at 480 and 650 nm in both spectra relate to cathodoluminescent (CL) emission of silica and their positions do not change with electron energy. However, the features seen only in the grating spectrum at around 680 and 350 nm correspond, respectively, in good agreement with Eq. (1), to first- and second-order Smith-Purcell emission in the surface normal ($\theta = 90°$) direction.

The spectral positions of these SP emission lines are found, again in accordance with Eq. (1), to depend on electron energy. Figures 2(c) and 2(d) show the evolution of the first- and second-order SP spectral features for electron energies between 30 and 50 keV, in each case in the form of a set of measured spectra normalized to the adjacent silica CL peak and reduced spectra (with the CL background subtracted) from which SP peak positions are derived via Gaussian fittings. These are plotted in Fig. 2(b) alongside theoretical curves for the two emission orders from Eq. (1). The expected blue shift with increasing electron energy and the excellent quantitative agreement between experiment and theory are clearly seen.

Figure 3 shows peak emission intensity as a function of electron beam current for representative first- and second-order SP peaks. The direct linear proportionality indicates that light is generated in the spontaneous (as opposed to super-radiant) SP emission regime.[32] Finally, measurements of emission intensity as a function of impact parameter, such as shown inset to Fig. 3, are found to be consistent with the exponentially decaying nature of the moving free electrons' evanescent electromagnetic field. The efficiency of emission into the fiber is estimated, based on the throughput and detection efficiencies of the light collection system, to be of order 10^{-6} photons per incident electron for first order SP radiation at 40 keV, and $\sim 10^{-7}$ photons/electron for the second order.

In summary, we have demonstrated that interactions between the optical frequency components of free electron evanescent fields and photonic nanostructures can be interrogated using a fiber-optic probe reminiscent of those employed to sample evanescent light fields in scanning near-field optical microscopy—a conically tapered tip with a sampling aperture 5–10 μm in diameter, functionalized, in the present case, with a nano-grating that couples electron evanescent fields to UV/visible light in the fiber. This fiber-based geometry can serve as a flexible platform for nanophotonic EIRE spectroscopy, providing advantages over existing geometries by integrating samples (in place of the nano-grating) directly with a photonic waveguide for the collection of light generated via any form of sample-beam interaction. The fiber platform also allows for straightforward integration of external light sources, providing the possibility of extended light/matter/free-electron interaction lengths applicable, for example, to novel geometries for electron energy loss/gain spectroscopies[36,37] and the acceleration of electrons with intense optical fields.[38,39] One may also envisage new forms of compact, tunable, fiber-coupled UV-VIS light sources, even single photon sources,[40] driven by chip-scale free-electron emitters.[41,42]

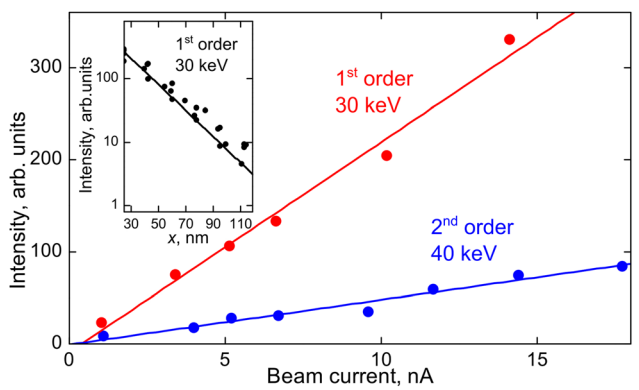

FIG. 3. SP emission intensity as a function of electron beam current for the first order at 30 keV (peak wavelength 767 nm) and the second order at 40 keV (344 nm) with grazing impact parameters. The inset shows the 30 keV first order SP emission intensity as a function of impact parameter x (beam current = 9.77 nA). An exponential fit of the form $A\exp(-4\pi x/\beta\gamma\lambda)$ is shown, where $A = 853.33$.

This work was supported by the UK's Engineering and Physical Sciences Research Council [EP/G060363/1] (all authors), The Royal Society and the Singapore Ministry of Education [Grant MOE2011-T3-1-005] (N.I.Z.). The authors would like to thank Giorgio Adamo and Jun-Yu Ou, respectively, for constructive discussions on EIRE spectroscopy and FIB processing.

[1]D. W. Pohl, W. Denk, and M. Lanz, Appl. Phys. Lett. **44**, 651 (1984).
[2]M. Burresi, D. van Oosten, T. Kampfrath, H. Schoenmaker, R. Heideman, A. Leinse, and L. Kuipers, Science **326**, 550 (2009).
[3]M. Mivelle, I. A. Ibrahim, F. Baida, G. W. Burr, D. Nedeljkovic, D. Charraut, J.-Y. Rauch, R. Salut, and T. Grosjean, Opt. Express **18**, 15964 (2010).
[4]P. Bharadwaj, R. Beams, and L. Novotny, Chem. Sci. **2**, 136 (2011).
[5]J. B. Pendry, Phys. Rev. Lett. **85**, 3966 (2000).
[6]N. Fang, H. Lee, C. Sun, and X. Zhang, Science **308**, 534 (2005).
[7]K. Dholakia and T. Čižmár, Nat. Photonics **5**, 335 (2011).
[8]M. L. Juan, M. Righini, and R. Quidant, Nat. Photonics **5**, 349 (2011).
[9]J. Zhang, K. F. MacDonald, and N. I. Zheludev, Phys. Rev. B **85**, 205123 (2012).
[10]D. Van Thourhout and J. Roels, Nat. Photonics **4**, 211 (2010).
[11]J. Zhang, K. F. MacDonald, and N. I. Zheludev, Light: Sci. Appl. **2**, e96 (2013).
[12]S. A. Maier, *Plasmonics—Fundamentals and Applications* (Springer, New York, 2007).
[13]J. A. Schuller, E. S. Barnard, W. Cai, Y. C. Jun, J. S. White, and M. L. Brongersma, Nature Mater. **9**, 193 (2010).
[14]E. Ozbay, Opt. Photonics News **19**, 22 (2008).
[15]N. I. Zheludev and Y. S. Kivshar, Nature Mater. **11**, 917 (2012).
[16]G. T. Di Francia, Il Nuovo Cimento **16**, 61 (1960).
[17]P. Van den Berg, J. Opt. Soc. Am. **63**, 1588 (1973).
[18]F. J. García de Abajo, Rev. Mod. Phys. **82**, 209 (2010).
[19]T. Muto, S. Araki, R. Hamatsu, H. Hayano, T. Hirose, P. Karataev, G. Naumenko, A. Potylitsyn, and J. Urakawa, Phys. Rev. Lett. **90**, 104801 (2003).
[20]G. Kube, H. Backe, H. Euteneuer, A. Grendel, F. Hagenbuck, H. Hartmann, K. H. Kaiser, W. Lauth, H. Schöpe, G. Wagner, T. Walcher, and M. Kretzschmar, Phys. Rev. E **65**, 056501 (2002).
[21]M. V. Bashevoy, F. Jonsson, K. F. MacDonald, Y. Chen, and N. I. Zheludev, Opt. Express **15**, 11313 (2007).
[22]M. Bosman, V. J. Keast, M. Watanabe, A. I. Maaroof, and M. B. Cortie, Nanotechnology **18**, 165505 (2007).
[23]V. Myroshnychenko, J. Nelayah, G. Adamo, N. Geuquet, J. Rodríguez-Fernández, I. Pastoriza-Santos, K. F. MacDonald, L. Henrard, L. M. Liz-Marzán, N. I. Zheludev, M. Kociak, and F. J. García de Abajo, Nano Lett. **12**, 4172–4180 (2012).

[24]R. Sapienza, T. Coenen, J. Renger, M. Kuttge, N. F. van Hulst, and A. Polman, Nature Mater. **11**, 781 (2012).
[25]G. Adamo, K. F. MacDonald, Y. H. Fu, C.-M. Wang, D. P. Tsai, F. J. García de Abajo, and N. I. Zheludev, Phys. Rev. Lett. **103**, 113901 (2009).
[26]S. Liu, M. Hu, Y. Zhang, W. Liu, P. Zhang, and J. Zhou, Phys. Rev. E **83**, 066609 (2011).
[27]P. J. M. van der Slot, T. Denis, J. H. H. Lee, M. W. van Dijk, and K. J. Boller, IEEE Photonics J. **4**, 570 (2012).
[28]J. V. Jelley, *Čerenkov Radiation and its Applications* (Pergamon Press, London, 1958).
[29]S. J. Smith and E. M. Purcell, Phys. Rev. **92**, 1069 (1953).
[30]Y. Neo, H. Shimawaki, T. Matsumoto, and H. Mimura, J. Vac. Sci. Technol., B: Microelectron. Nanometer Struct. **24**, 924 (2006).
[31]J. Gardelle, P. Modin, and J. Donohue, Phys. Rev. Lett. **105**, 224801 (2010).
[32]J. Urata, M. Goldstein, M. F. Kimmitt, A. Naumov, C. Platt, and J. E. Walsh, Phys. Rev. Lett. **80**, 516 (1998).
[33]S. Korbly, A. Kesar, J. Sirigiri, and R. Temkin, Phys. Rev. Lett. **94**, 054803 (2005).
[34]S. E. Korbly, A. S. Kesar, R. J. Temkin, and J. H. Brownell, Phys. Rev. Spec. Top.–Accel. Beams **9**, 022802 (2006).
[35]G. Doucas, V. Blackmore, B. Ottewell, C. Perry, P. G. Huggard, E. Castro-Camus, M. B. Johnston, J. L. Hughes, M. F. Kimmitt, B. Redlich, and A. van der Meer, Phys. Rev. Spec. Top.–Accel. Beams **9**, 092801 (2006).
[36]F. G. de Abajo and M. Kociak, New J. Phys. **10**, 073035 (2008).
[37]B. Barwick, D. J. Flannigan, and A. H. Zewail, Nature **462**, 902 (2009).
[38]J. Breuer and P. Hommelhoff, Phys. Rev. Lett. **111**, 134803 (2013).
[39]E. Peralta, K. Soong, R. England, E. Colby, Z. Wu, B. Montazeri, C. McGuinness, J. McNeur, K. Leedle, and D. Walz, Nature **503**, 91 (2013).
[40]X. Bendana, A. Polman, and F. J. García de Abajo, Nano Lett. **11**, 5099 (2011).
[41]P. C. Fletcher, V. K. R. Mangalam, L. W. Martin, and W. P. King, J. Vac. Sci. Technol., B **31**, 021805 (2013).
[42]J. Y. Lin, S. H. Wang, T. T. Chen, C. Y. Chen, L. J. Chou, J. C. Hwang, and C. S. Kou, J. Electrochem. Soc. **158**, D426 (2011).