

## Electron-Beam-Driven Collective-Mode Metamaterial Light Source

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We demonstrate experimentally that the energy from a highly localized free-electron-beam excitation can be converted via a planar plasmonic metamaterial to a low-divergence free-space light beam. This emission, which emanates from a collectively oscillating coupled metamolecule nanoantenna ensemble much larger in size than the initial excitation, is distinctly different from cathodoluminescence and bears some similarity with laser light. It offers a novel, flexible paradigm for the development of scalable, threshold-free light sources.

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The recent explosion of interest in nanophotonics, plasmonics, and metamaterials has led to the pursuit of nanoscale light and plasmon sources driven by optical [1–5], current [6–11], and free-electron [12–16] excitations. We experimentally demonstrate here that a scalable, low-divergence, threshold-free optical source can be constructed on the basis of a collectively oscillating optical nanoantenna array driven by the localized injection of free electrons. In marked contrast to conventional (microwave) antenna arrays where coherence is ensured by the feed, the emission of the plasmonic array is determined by strong collective interactions among the metamolecules.

The emission of an electron-beam-driven metamaterial source is radically different from that of Smith-Purcell [17] or “Light-well” [14] sources based on spatially extended interactions between electrons and periodically structured media in that its resonant frequency is not a function of electron energy, but rather corresponds to a plasmonic mode of the metamaterial. The emission also differs from that of singular nanostructures (particles, rods, nanoantennae, etc.) [16,18–20] or single emitters in nanostructured environments that control output field profiles [21–23] as it involves the collective excitation [24] of and emission from a large number of plasmonic metamolecule oscillators. Indeed, the light generated by collective mode metamaterial sources bears some comparison with laser light in the sense that it shows directionality, spatial coherence, and spectral narrowing.

The concepts behind this new type of light source are illustrated in Fig. 1: it is found that in certain types of metamaterial (previously dubbed “coherent” for their collective absorption properties [25,26]) a *localized* excitation can generate a *delocalized* response across a large number of metamolecules. Following an initially highly confined excitation the coherent metamaterial system will, as a

result of strong interactions among metamolecules over a much larger spatial area than the initial energy injection spot, rapidly achieve a steady state dominated by weakly radiative modes [24] (see Supplemental Material [27]). The dominant ( $M_{11}$ ) mode is of particular interest: it is frequency-matched to the metamaterial’s absorption resonance; has a highly uniform phase profile; couples to (i.e., may launch) a plane wave propagating perpendicular to the metamaterial plane; and, depending on the level of losses in the system, may assume a dominant proportion of the collective excitation energy following stimulation of a single metamolecule. We report here on the observation of such emission in the optical part of spectrum using

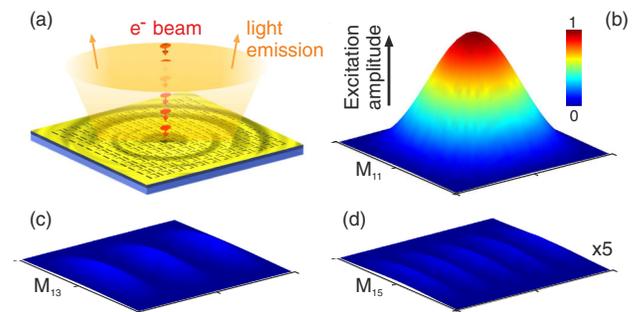


FIG. 1 (color online). Electron-beam-driven collective metamaterial light source concept. (a) Highly localized electron-beam excitation at the center of a metamaterial array leads, via the strong coupling among metamolecules, to the collective oscillation of many cells and thereby the emission of a free-space light beam. (b)–(d) Numerically calculated amplitude profiles of the three strongest collective eigenmodes  $M_{pq}$  of a center-driven finite ( $32 \times 32$  cell) array. A low-divergence free-space wave is launched in the surface normal direction by the dominant  $M_{11}$  mode.

a plasmonic metamaterial: the localized injection of electrons into the array is indeed found to drive directed light emission from a large ensemble of metamolecules. Furthermore, we show that the emission spectrum narrows, i.e. the temporal coherence of the emitted radiation improves, with increasing array size.

Our experiments employed coherent metamaterial designs comprising square arrays of asymmetrically split rings (ASRs) [28] (see inset to Fig. 2(b)), which possess a Fano-like plasmonic resonance [29] derived from the interaction of large numbers of metamolecules in the array, as theory indicates is required for the observation of collective light emission.

Samples comprising  $20 \mu\text{m} \times 20 \mu\text{m}$  arrays of ASRs with identical unit cell geometries but different cell sizes between 220 and 280 nm were manufactured by focused ion beam milling in a 50 nm evaporated gold film supported on a 100 nm silicon nitride membrane. Their electron-induced emission spectra were recorded in a scanning electron microscope equipped with achromatic reflective optics to direct light radiated by samples in response to electron-beam excitation (spot diameter 200 nm; electron energy 30 keV; beam current  $\sim 50$  nA) to a spectrometer with a liquid nitrogen cooled charge-coupled device array detector. These spectra [Fig. 2(a)], each with the cathodoluminescent contribution of gold

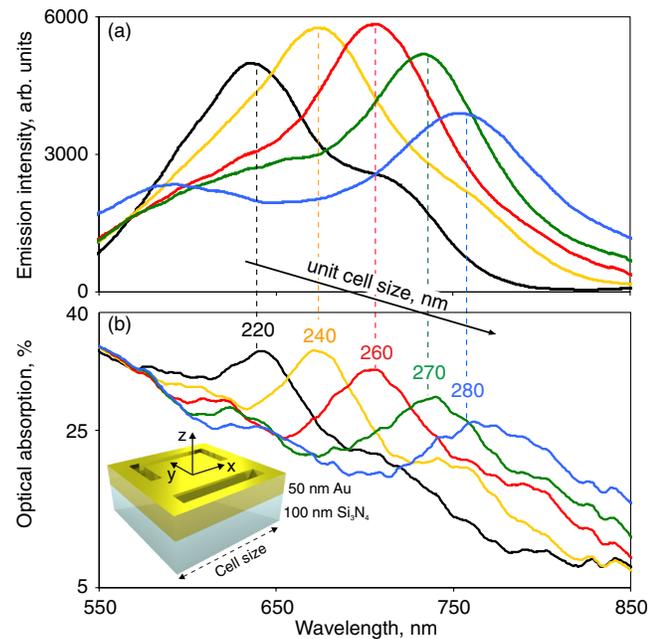


FIG. 2 (color online). Electron-induced emission vs optical absorption in a plasmonic metamaterial. (a) Electron-beam-induced metamaterial light emission spectra for arrays with different metamolecule cell sizes (as labeled). (b) Corresponding optical absorption spectra (derived from microspectrophotometric measurements of normal incidence transmission and reflection). The inset to (b) shows a schematic of the asymmetric split ring metamolecule.

(measured separately on an unstructured part of the sample) removed, reveal a direct correlation between a metamaterial's peak emission wavelength and the spectral position of its plasmonic absorption peak [Fig. 2(b)], across all unit cell sizes. We therefore conclude that the metamaterial component of emission is linked directly to the collective mode known to underpin the plasmonic absorption resonance [26,30].

Emission directionality was studied by projecting the light collected from the sample directly onto the charge-coupled device array [31] as illustrated in Fig. 3(a). These

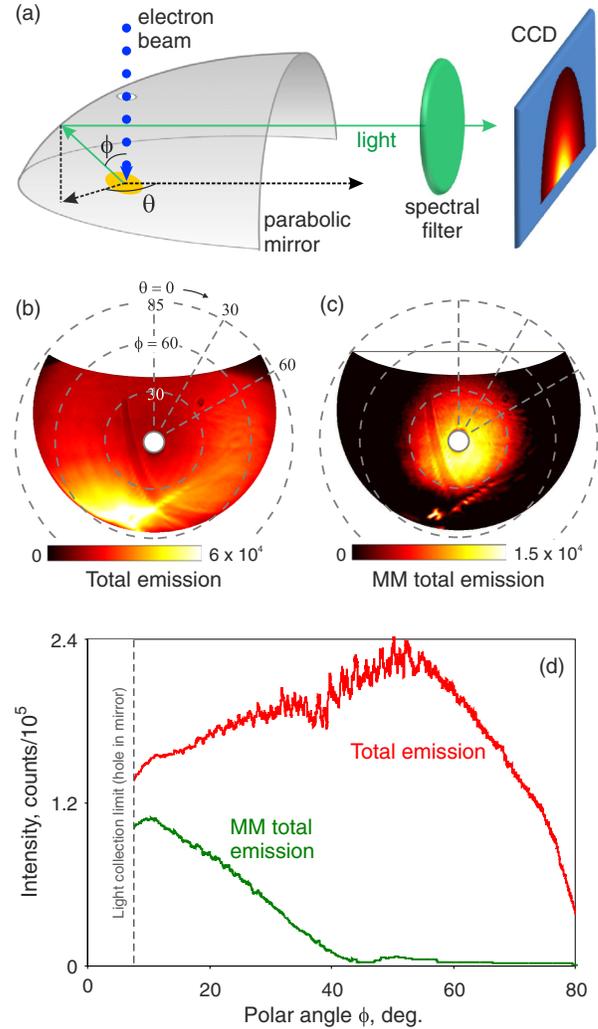


FIG. 3 (color online). Directional resonant light emission from a plasmonic metamaterial. (a) Experimental configuration for angle-resolved detection of electron-induced light emission. (b),(c) Angular intensity distributions of total electron-induced light emission from a metamaterial array with a 220 nm unit cell, integrated over all wavelengths from 400 to 1100 nm (b), and the metamaterial-specific component of this spectrally integrated emission [(c), background contribution from unstructured gold removed]. (d) Emission intensity as a function of polar angle (integrated over azimuth angle) for the two cases shown in (b) and (c).

measurements clearly demonstrate that the resonant metamaterial emission is directional while the total broadband emission (including gold cathodoluminescence) is not; the broad angular distribution of the total emission, integrated over all wavelengths between 400 and 1100 nm, is shown in Fig. 3(b); Figure 3(c) shows the metamaterial component of this emission, which is confined to low polar angles around the surface-normal direction; Fig. 3(d) presents, for these two cases, emission intensity integrated over all azimuth angles as a function of the polar angle. We consider the low divergence of the resonant metamaterial radiation to be indicative of the collective nature of the underlying excitation mode and suggestive of a degree of spatial coherence across a large number of metamolecule emitters. Indeed, numerical analysis (see the Supplemental Material [27]) illustrates that the angular profile of the metamaterial emission is markedly narrower than the emission profile of a single dipole (and therefore incoherent ensembles of dipoles) either in free space or in close proximity to perfectly conducting or dielectric substrates. Simulations of the collective effects of the ASR array reveal that in a steady-state response to the localized excitation the collective magnetic mode  $M_{11}$  [Fig. 1], in which all the metamolecules oscillate in phase, can assume the dominant proportion of the total excitation energy, resulting in a very high degree of spatial coherence (see [27]). Moreover, the mode  $M_{11}$  exhibits a highly directed emission pattern and a narrow radiative linewidth [30].

Finally, variations in the electron-induced emission spectrum with the number of metamolecules engaged in the emission process were studied. It has recently been shown that the width of the optical absorption peak in a finite ASR metamaterial array collapses with increasing array size and this behavior is a reflection of the collective, coherent nature of the plasmonic mode underpinning the resonant absorption [26,30]. One may therefore expect a similar collapse in the emission spectrum as the number of contributing metamolecules increases and experimental results demonstrate exactly that. Because the excitation extends to a finite distance from the electron injection point this dependence can be investigated by translating the injection point from a position close to the boundary of the metamaterial array, where only a small number of metamolecules can be engaged, to a central position in the array where the excitation encompasses the largest possible number of metamolecules.

Emission spectra were recorded at a number of points along a line perpendicular to the boundary of the metamaterial array as illustrated in Fig. 4(a). At distances more than  $\sim 1.8 \mu\text{m}$  outside the array the metamaterial structure has no discernable impact on light emission from the sample [spectrum A in Fig. 4(a)] but at shorter distances its influence is seen. As the excitation point moves into the array the emission spectrum evolves and the emission intensity increases, saturating at a similar distance inside

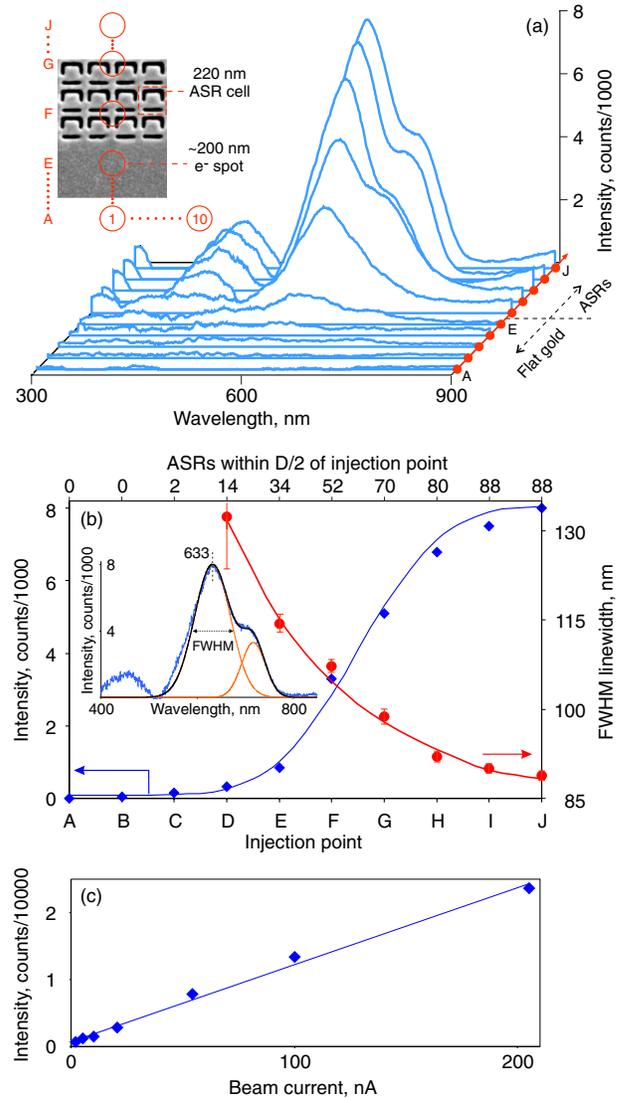


FIG. 4 (color online). Spectral collapse of metamaterial emission. (a) Electron-induced metamaterial light emission spectra for a sequence of beam injection positions at intervals of 400 nm along a line perpendicular to the boundary of the metamaterial array (each averaged over ten injection points spaced along lines parallel to the boundary as indicated on the inset secondary electron image). (b) Intensity and half-maximum width of the 633 nm emission line as functions of the electron beam injection coordinate and the number of metamolecules within the  $1/e^2$  source radius of the injection point. (c) 633 nm emission intensity as a function of electron beam current.

the array (spectrum *J*). Figure 4(b) plots the linewidth and intensity of the primary 633 nm emission peak (based on a double-Gaussian decomposition of the spectra exemplified inset) as a function of injection position: the emission linewidth narrows in a manner analogous to the absorption resonance collapse observed with increasing number of metamolecule count in coherent microwave, terahertz, and optical frequency ASR metamaterials [26].

The dependence of emission intensity on electron injection position can be used to evaluate the number of meta-molecules in the collectively emitting ensemble; a fitting to the intensity data presented in Fig. 4(b), based on the overlap of a Gaussian beam profile with the metamaterial array, yields a  $1/e^2$  source diameter  $D$  of  $\sim 2 \mu\text{m}$ —a spot size that encompasses (wholly or partially) up to 88 metamolecules [a secondary horizontal axis on Fig. 4(d) converts injection position to number of metamolecules  $N$  within  $D/2$  of the injection point]. In all experiments reported above the emission intensity shows a *threshold-free* linear dependence on electron beam current, as illustrated in Fig. 4(c).

Together, the observed spectral collapse, emission-absorption peak correlation, and strong directionality of resonant emission, compellingly link the effect of electron-induced metamaterial light emission to a collective mode encompassing a large ensemble of metamolecules. They also point to synchronization across the plasmonic nanoantenna array derived not from the feed (as in conventional microwave antenna arrays) but from the strong interactions among metamolecules. The conversion of energy from free electrons injected locally into a planar plasmonic metamaterial to a low-divergence light beam emitted by a large collectively oscillating array of metamolecules constitutes a fundamentally new nanostructural radiation phenomenon. The emission is distinctly different in nature from cathodoluminescence or point-dipole radiation (directionality, large emitting area, and spectral dependence) and displays certain characteristics more commonly associated with laser light (spectral narrowing and spatial coherence), though in contrast to lasing it is a linear phenomenon. As such, coherent metamaterials offer a generic platform (amenable to optical, electronic, and plasmonic pumping as well as electron-beam excitation) for scalable, threshold-free sources of light.

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*Note added.*—In work discovered at the proofing stage for this article, Guebrou *et al.* [32] describe an optically pumped disordered system of plasmonically coupled molecular emitters in which again the formation of a spatially coherent hybrid state leads to coherent emission.

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