PHOTONICS

Hot plasmons make graphene shine

Bright hot plasmon emission is observed in graphene due to the ultrafast relaxation of hot carriers that were excited by femtosecond laser pulses of visible light.

Frank H. L. Koppens and Klaas-Jan Tielrooij

The ultrafast carrier dynamics of graphene have been in the spotlight ever since it was realized that the associated physical phenomena are quite distinct from those in conventional materials. Moreover, one can exploit these processes for novel optoelectronic device functionalities. Graphene combines an ultralow electronic heat capacity with strong carrier–carrier interactions and relatively weak coupling between the electronic and phononic systems. As a result, when graphene carriers are excited, for example by an ultrashort laser pulse, carrier–carrier interactions take place on an ultrafast timescale of ten femtoseconds[1], leading to efficient redistribution of energy in the electronic system. This typically leads to the formation of a hot-carrier distribution with an increased carrier temperature[2]. The interaction of charge carriers with optical and acoustic phonons in graphene, and substrate phonons, which occurs on a (sub) picosecond timescale at room temperature, reduces the carrier temperature. Carrier cooling through radiation of light can, in principle, also occur, and indeed some thermal (Planckian) radiation of the hot-carrier system has been observed[3]. This, however, is not an efficient process — it cannot compete with cooling via phonon emission. A more efficient process is near-field radiation into phonon modes of nearby materials[4], or radiative transfer to nearby graphene[5]. However, no light is emitted in these cases.

Now, reporting in *Nature Materials*, Laura Kim and co-workers present experimental evidence of an additional relaxation channel that involves ultrafast (~100 fs) energy flow from photoexcited carriers into graphene plasmons, which can lead to bright mid-infrared light emission[6]. Interestingly, under certain excitation conditions, an inverted state forms, where the valence and conduction bands each have their own Fermi–Dirac distribution (with a temperature in excess of 1,000 K), rather than a common distribution. This inverted state has been observed experimentally[2] and, according to theoretical predications, it can relax by efficient coupling to graphene plasmons[7,8]. Plasmons are collective excitations of the electrons, mediated by long-range Coulomb interactions. In graphene, plasmons are tunable in situ by varying the Fermi energy, and due to the low dimensionality, they have been exploited to generate strongly compressed optical fields; this makes plasmons an ideal transducer between electrons and light.

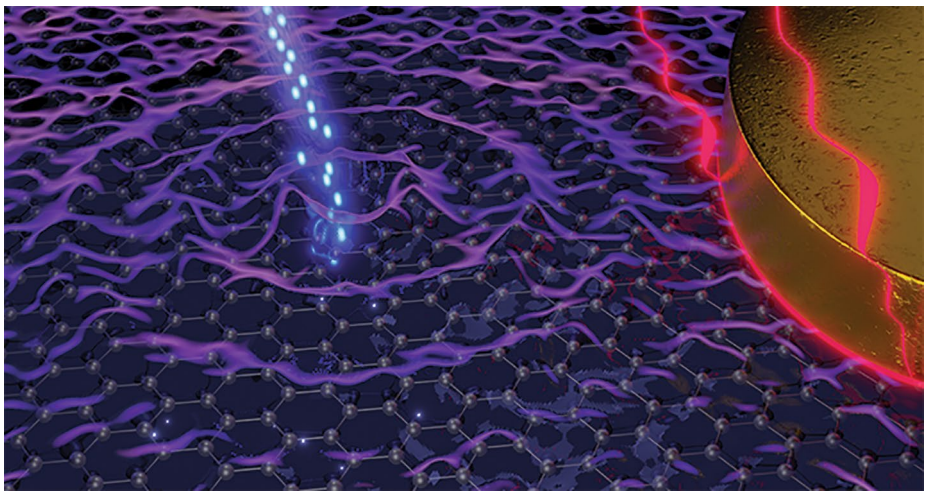

Fig. 1 | Artistic impression of the bright hot plasmon emission process. Upon laser irradiation (blue stream), hot electrons emit plasmons (purple ripples) in graphene, which subsequently convert into photons (red emission) at gold nanodisks. Credit: Matteo Ceccanti.

Kim and co-workers provide compelling experimental evidence of the predicted plasmon-mediated relaxation channel, with the tantalizing potential to create plasmon emission that is much more efficient than thermal radiation. They photoexcite graphene with 100-fs laser pulses of visible light at 850 nm (blue stream in Fig. 1), and monitor the emission in the infrared frequency range. What they find is striking: the higher the Fermi level of the graphene, the stronger the infrared emission (red emission in Fig. 1). This effect is in stark contrast to thermal emission, which would only reduce for increasing Fermi energy. The observed behaviour is consistent with plasmon emission, as the phase space for plasmonic emission increases with Fermi energy. The plasmons (purple ripples in Fig. 1), emitted by the hot electrons, subsequently scatter (partially) into photons, leading to the observed infrared emission. To further confirm the plasmon-emission process, the graphene was patterned into ribbons, which exhibited plasmonic absorption resonances at the same frequency as the plasmonic emission resonances.

The plasmons really started to shine after adding gold nanodisks, which enhance their scattering into photons. Detailed analysis showed a plasmon emission rate of a stunning five orders of magnitude higher than the thermal emission. The main challenge of translating this system into a bright infrared light source is the plasmon–photon out-coupling, but the good news is that several solutions are available, such as the use of denser nanophotonic structures, and improved optical design.

Thanks to the work of Kim and colleagues we can add another relaxation mechanism to the rich list of processes that contribute to the ultrafast dynamics of graphene's Dirac fermions. Moreover, this newly identified mechanism opens up avenues for applications: bright light sources

in the mid-infrared are fairly scarce, and the graphene–nanodisk system introduced here adds promising benefits of electrical tunability and small material footprint, as well as the prospect of ultrahigh modulation rates — into the THz regime. It remains to be seen if the inverted state — required for relaxation into plasmons — only arises under specific excitation conditions, for example, interband photoexcitation with sufficiently high absorbed peak energy densities (in the mJ cm^{-2} range), and if perhaps other methods such as electrical injection are possible. ❒

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Competing interests

The authors declare no competing interests.