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Low-threshold terahertz molecular laser optically pumped by a quantum cascade laser

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We demonstrate a low-threshold, compact, room temperature, and continuous-wave terahertz molecular laser optically pumped by a mid-infrared quantum cascade laser. These characteristics are obtained, thanks to large dipole transitions of the active medium: NH_3 (ammonia) in gas state. The low-power (<60 mW) laser pumping excites the molecules, thanks to intense mid-infrared transitions around 10.3 μm. The molecules de-excite by stimulated emission on pure inversion "umbrella-mode" quantum transitions allowed by the tunnel effect. The tunability of the quantum cascade laser gives access to several pure inversion transitions with different rotation states: we demonstrate the continuous-wave generation of ten laser lines around 1 THz. At 1.07 THz, we measure a power of 34 μW with a very low-threshold of 2 mW and a high differential efficiency of 0.82 mW/W. The spectrum was measured showing that the linewidth is lower than 1 MHz. To our knowledge, this is the first THz molecular laser pumped by a solid-state source and this result opens the way for compact, simple, and efficient THz source at room temperature for imaging applications. [http://dx.doi.org/10.1063/1.4945355]

Terahertz electromagnetic radiations (1 THz = 10^{12} Hz) are in a frequency range between the visible/infrared range and the radio/microwave range. Despite the identification of various applications[1] (chemical and bio-sensing, very high data rate communications, non-destructive and security imaging), it is still underdeveloped due to the lack of compact, easy-to-use, and low-consumption sources, particularly around 1 THz. In the domain of THz lasers, semiconductor terahertz quantum cascade lasers (QCLs) have been demonstrated in 2002, their active parts are very compact. Despite investigations and progress, cryogenic cooling is still needed[2,3] which is bulky and expensive. THz molecular lasers have been demonstrated long time ago[4–6] but until now they are also too bulky and expensive for a wide use in applications. Laser oscillation is obtained by electrical or optical pumping of a molecular gas at room temperature and low pressure. The gain is generally obtained, thanks to a population inversion between rotational levels of an excited vibrational state of the molecule which is almost empty at room temperature. Discrete frequencies can then be generated in 0.1-10 THz range covering submillimeter, THz, and far-infrared waves. In the case of electrical pumping,[4,5] vibrational excitation is obtained through the use of an electrical discharge through the gas. However this simple approach has several drawbacks: (I) the excitation is not specific and leads to low efficiencies; (II) population inversion in continuous wave (CW) is generally due to selective relaxation, only few molecules can be pumped following this way, and consequently only 50 laser lines were demonstrated since fifty years; and (III) a gas flow is needed because the molecules are dissociated by the discharge. Moreover, high voltage or high power radio-frequency generators are needed to create the plasma, leading to bulky systems and high energy consumption. The other way, optical pumping of molecules with a mid-infrared (MIR) laser, has been achieved for the first

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time in 1970 when Chang and Bridges demonstrated the optically pumped THz laser (OPTL).[6] In this case, many molecules can be pumped, thanks to the multi-line capability of the powerful discharge CO_2 laser in CW or pulsed regime. The specific excitation of energy levels leads to higher efficiencies and a gas flow is not needed. More than 5000 lines have been demonstrated using molecules composed of 3–9 atoms, the most popular being the 119 μm (2.52 THz) line of CH_3OH. The main drawbacks of these lasers come from the pump laser: it is a powerful gas discharge laser because the threshold power is generally high. CW gas lasers emit only discrete lines, then the frequency of absorption of the molecules has to coincide very precisely with the emission line of the pumping laser. For resonant-pumping CW conditions, the relative shift between these two frequencies should be less than a few 10^{-6} due to the very small linewidth of the gas lines at low pressure (at room temperature, the Doppler broadening is <100 MHz). Raman gain can also be obtained for detunings of few hundreds of MHz but it requires higher pump intensities and the THz gain is lower than for resonant-pumping.[7] This strict resonance condition has a very low probability to occur for simple light molecules (2–4 atoms), and more heavy molecules are then needed to increase the number of possible states, the density of absorption lines, and to ensure the coincidence of more lines with the CO_2 laser lines.

More than 20 years ago, the QCL was demonstrated for the first time.[8] Now MIR-QCLs are working in CW mode at room temperature; they are also monomode and can be tuned in wavelength, thanks to the external-cavity scheme or to the distributed feedback (DFB) scheme. Moreover they produce powers ranging from the mW to the W level.[9] In this letter, we demonstrate that MIR-QCLs are a valuable source for pumping OPTL. The main advantages compared to CO_2 lasers are their intrinsic compactness (DFB-QCL is less 1 mm^3), low power consumption, and finally their continuous tunability which allows to excite any MIR transitions of molecules. This property offers more flexibility in the choice of the molecule: the objective of our demonstration is to achieve a high gain, a low threshold, and a high conversion efficiency in order to demonstrate a compact and efficient room temperature THz source. Low threshold is of the utmost importance since the main drawback of QCL is that their power is still limited to values lower than typical CO_2 lasers. After considering several molecules, the ammonia molecule (NH_3) was chosen because (I) it has a high permanent dipole moment (1.4 D), (II) it has a high rotational constant (B_0 = 298 GHz), and (III) it has a fast relaxation rate. These characteristics are ideal to obtain a high gain. It is also a spectroscopically very well-known molecule, which simplify the investigations. These advantages of NH_3 were known in the past and shortly after the first OPTL demonstration, it was tentatively used but due to a very sparse absorption spectrum (another consequence of the high rotational constant), no coincidences were found with the standard CO_2 laser lines. One coincidence was found with a line of the N_2O discharge laser and CW NH_3 OPTLs were reported.[10,11] They generated two lines at 3.68 THz (rotational transition) and 1.14 THz (pure inversion transition cascading from the previous transition). Other ways were also tried: pulsed CO_2 pumping, Stark tuning, isotopic CO_2 and/or isotopic NH_3, Raman laser, and sequence band lasers pumping.[12] A number of lines were discovered but all these solutions were quite complicated, bulky, and expensive. Using QCL pumping, it is, in principle, possible to choose the transitions that allow high gain and generate interesting frequencies for applications.

The equilibrium configuration of NH_3 is a simple pyramidal symmetric-top molecule but the most striking feature is the so-called inversion quantum phenomena (see inset of Fig. 1): the N atom is able to tunnel through the potential barrier formed by the three H triangle: the energy levels are then split in lower energy symmetric (s) and higher energy antisymmetric (a) wave functions. The inversion is a vibrational motion (the so-called "umbrella-mode"), but compared to standard molecular vibrations giving frequencies in the infrared range, the oscillation is dramatically slowed down by the hindering potential. Its frequency lies in the microwave region around 24 GHz for a molecule in the ground state (the first demonstration of the maser was made by inverting the population between these levels in a molecular beam[13]). In the $\nu_2 = 1$ excited vibrational state (about 950 cm^{-1} above the ground state levels), the inversion splitting is about 1 THz due to the reduction of the barrier height.[14,15] These excited states are almost empty at room temperature and the population can easily be inverted, thanks to optical pumping between $\nu_2 = 0$ and $\nu_2 = 1$. Then two types of THz transitions can occur: (I) pure inversion transitions (selection rules are $\Delta\nu = \Delta K = \Delta J = 0$, $a \leftarrow s$,

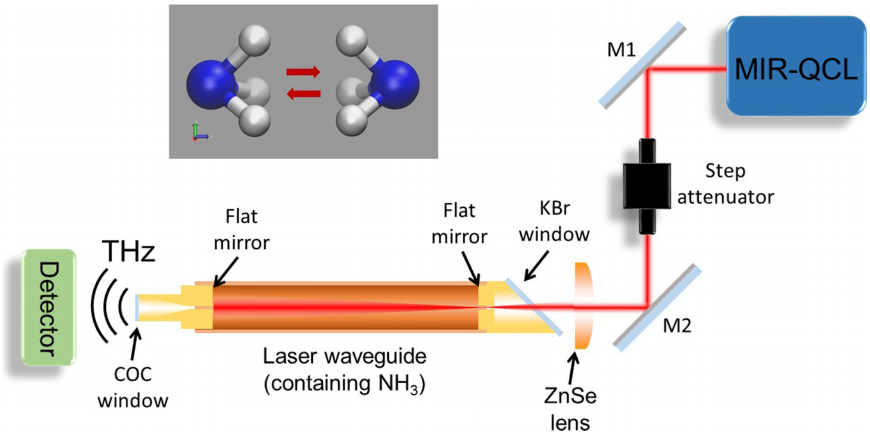

FIG. 1. Schematic of the experiment. The mid-infrared beam is colored in red. Inset: simplified representation of the NH_3 inversion process (umbrella-mode), the nitrogen atom is colored in blue and the three hydrogen atoms are in grey.

and $s \leftarrow a$) and (II) rotational transitions (selection rules are $\Delta\nu = \Delta K = 0$, $\Delta J = \pm 1$, $a \leftarrow s$, and $s \leftarrow a$). In this letter, the pumping is achieved using MIR Q-branch transitions around 965 cm^{-1}, a very dense region with strong transitions corresponding to the $a \leftarrow s$ transitions between $\nu_2 = 0$ and $\nu_2 = 1$, noted $saQ(J,K)$. Pumping these transitions allows to populate a levels and to achieve gain on pure inversion transitions close to 1 THz.

The laser cavity is based on a cylindrical metallic waveguide (see Fig. 1). The 50 cm-long laser cavity is constructed using 10 mm-internal diameter copper tubing and two flat brass mirrors with a 1.2 mm diameter hole in the center. The two mirrors are inserted inside the tube like a piston. At one extremity, the mirror is soldered to the tube, cut at 45°, and closed by a KBr window. At the other extremity, the mirror is soldered to a metallic membrane and is then mobile inside the tube. The 1.2 mm hole communicates with a conical horn machined into the brass. The horn is closed by a cyclic-olefin copolymer (COC) window transparent in the THz range. The pump beam comes from a commercial CW grating external-cavity QCL tunable from 10.0 to 10.5 μm (Daylight Solutions). The MIR wavelength can be precisely measured, thanks to a Bristol 721B spectrum analyzer (absolute accuracy: 1 ppm). The pump beam is attenuated by a step-attenuator (Lasnix), chopped by a mechanical chopper if necessary, and focused in the cavity, thanks to an AR-coated ZnSe lens (f = 20 cm) through the KBr window and the 1.2 mm hole. This focal length allows to adapt the pump beam diameter to the tube internal diameter close to the output coupler. At the output of the molecular laser, several detectors are used: (I) a pyroelectric detector; (II) an InSb hot electron bolometer (HEB) cooled at 4 K (noise equivalent power: <3 pW/$\sqrt{\text{Hz}}$) from QMC Instruments Ltd., followed by a ×100 amplifier and a lock-in amplifier (total responsivity: 140 kV/W, it contains three low-pass filters to reject MIR waves); and (III) a 750-1100 GHz Schottky diode-based sub-harmonic mixer (Radiometer Physics GmbH) connected to a FSU67 electrical spectrum analyzer (Rohde & Schwarz). The local oscillator is in the 9-10 GHz range and the harmonic number is between 106 and 117. In the case of the pyroelectric detector and the mixer, a polymer lens is used to focus the beam.

After evacuation of the cavity by a turbo-molecular pump, some NH_3 is injected and the pressure is stabilized to about 5-10 Pa. The QCL is tuned close to 967.346 cm^{-1} which corresponds to the $saQ(3,3)$ transition according to the Jet Propulsion Laboratory molecular spectroscopy database.[16] The residual MIR beam at the output of the laser is measured using the pyroelectric detector. The absorption line is easily found by a fine tuning of the wavelength obtained by a slight variation of the QCL cavity temperature. The THz bolometer is then placed in front of the laser. Fig. 2 shows the measured THz power versus the MIR pump optical power for an optimum pressure close to 0.7 Pa. At this pressure, about 80% of the pump beam power is absorbed along the 50 cm-path. At low pump power, only the noise level of the bolometer is measured but for pump powers higher than about 2 mW, signals higher than ten thousand times the noise floor are measured. For a pump power of 26 mW, a THz power of about 10 μW is measured. Because of the symmetry

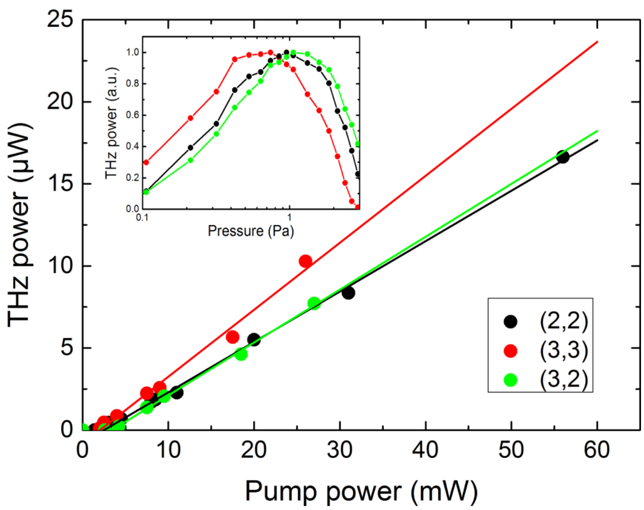

FIG. 2. Measured terahertz output power versus mid-infrared pump power for three transitions at optimum pressure: $\nu_2 = 1$ *asQ*(2,2) in black, $\nu_2 = 1$ *asQ*(3,3) in red, $\nu_2 = 1$ *asQ*(3,2) in green. Inset: pressure dependence of the power of the three observed emission lines.

of the cavity (same hole in the center of the two mirrors), it is expected that the same power is generated from the pump side (but not measured), and consequently, the total generated power should be 20 μW, corresponding to an optical conversion efficiency of 7.7×10^{-4} (differential efficiency of 0.82 mW/W). For OPTL, the theoretical limit for the optical conversion efficiency,[17] assuming a rapid vibrational relaxation, a lossless cavity, and an ideally coupled laser is given by $\eta_{opt} = {}^1/_2(\nu_{\mathrm{THz}}/\nu_{pump}) = 1.8 \times 10^{-2}$. The measured efficiency is 4.3% of the theoretical maximum.

By tuning the QCL close to 967.407 cm^{-1}, another lasing transition is identified corresponding to the *saQ*(3,2) transition. The THz power is slightly lower for this transition (Fig. 2). By tuning the QCL close to 967.738 cm^{-1}, another lasing transition is obtained corresponding to the *saQ*(2,2) transition. The differential efficiency is lower (0.62 mW/W) but it is possible to increase the pump power to 56 mW, giving a maximum output power of about 17 μW (Fig. 2), corresponding to a total power of 34 μW by taking into account the symmetry of the laser. Lasing is also obtained with seven other transitions: *saQ*(2,1), (3,1), (4,4), (5,2), (5,3), (6,5), and (8,7).

The identification of the lasing transitions is confirmed, thanks to precise frequency measurements using an electrical spectrum analyzer with a subharmonic mixer. For the line obtained by pumping at 967.346 cm^{-1}, a frequency of 1 073 050.4 MHz is measured, the linewidth being lower than 1 MHz (see Fig. 3). It corresponds to the $\nu_2 = 1$ *asQ*(3,3) pure inversion transition with an error of less than 1 MHz[16,18] compatible with the typical gain bandwidth of low-pressure OPTL. For the line obtained by pumping at 967.407 cm^{-1}, a frequency of 1 035 816.3 MHz is measured corresponding to the $\nu_2 = 1$ *asQ*(3,2) pure inversion transition with also an error less than 1 MHz.[16] An energy-level diagram that summarizes the three observed transitions is sketched in Fig. 4. To our knowledge, these CW laser lines were never reported before. Pure inversion transitions for different (J,K) have been observed previously, but due to the lack of coincidence with CO_2 laser lines, they have been only obtained in pulsed mode with multi-kW laser pumping[19] or in CW mode, thanks to the cascade process with N_2O laser pumping[10] or to sequence band CO_2 lines[20] which is more complicated and reduces the efficiency of the laser. As previously mentioned, pure inversion transitions with $J = K$ are more intense, and in this case, competition with other THz transitions or cascade processes are not possible due to selection rules (see Fig. 4), an ideal case for high efficiency, high spectral purity, and simple laser design.

In the literature, THz generation with MIR-QCL has been obtained also thanks to intracavity difference-frequency generation (DFG).[21,22] In CW, a power of 3 μW has been obtained[22] at 3.6 THz for a MIR pump power of 160 mW corresponding to an optical conversion efficiency of 1.9×10^{-5}, 40 times lower than the efficiency of our NH_3 laser (the photon efficiency is 130 times

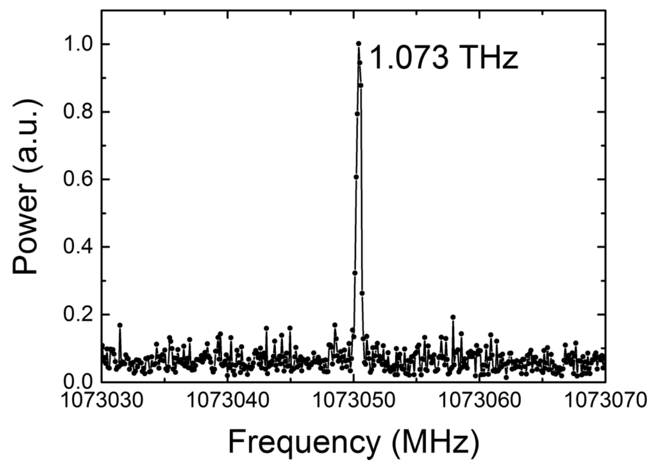

FIG. 3. Measured laser output spectrum for a pump beam at 967.346 cm^{-1} corresponding to the *saQ*(3,3) MIR transition (resolution bandwidth: 100 kHz).

lower). CW THz generation was also achieved with intracavity DFG within a dual color vertical external cavity surface emitting laser (VECSEL) at 1 THz,[23] but the efficiency was also close 10^{-5}.

Another way of room-temperature THz generation is the photomixing of two laser lines in a large bandwidth photodetector; the record power[24] obtained at 1.04 THz is 10.9 μW. The 1.55 μm pump power is 0.7 W corresponding to an optical conversion efficiency of 1.6×10^{-5}, 50 times lower than the efficiency of our NH_3 laser.

This work demonstrates that QCL-pumped OPTLs are efficient room temperature CW THz sources. NH_3 is particularly suited as a gain medium; it can deliver easily several lines near 1 THz thanks to the inversion splitting of the $\nu_2 = 1$ vibrational state. Higher conversion efficiencies and higher power performances can be obtained by optimizing the transmission coefficient of the output coupler, minimizing the losses of the waveguide, optimizing the matching between the laser mode and the pump beam, and by increasing the pump power. With already published QCL in the 1-watt range,[25] an output power close or higher to 1 mW should be obtained. In addition, a large number of other transitions at different frequencies can be pumped by QCLs using NH_3 or other molecules. Nevertheless, the frequency range around 1 THz is particularly interesting because it is the highest range with low atmospheric attenuation, high penetration in solids, and with a sufficiently small wavelength to allow the integration on a chip of thousands of pixels in 2D focal plane array for THz imaging applications.[26] Such a source is also essential for THz near-field optical microscopy that

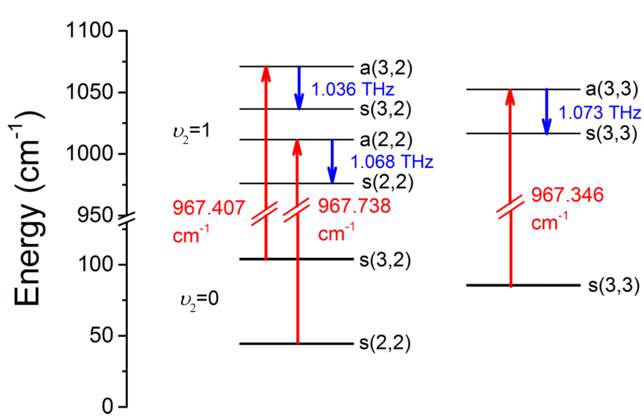

FIG. 4. Partial energy levels diagram of the $^{14}NH_3$ molecule; left: $K = 2$, right: $K = 3$. The arrows show the transitions between the three levels involved in the laser for three observed laser lines.

has promising applications in the domain of chemical and biological identification of nano-objects and nanostructures.[27,28]

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[1] *Handbook of Terahertz Technology Devices and Applications*, edited by H.-J. Song and T. Nagatsuma (Pan Stanford Publishing, 2015).
[2] M. A. Belkin and F. Capasso, Phys. Scr. **90**, 118002 (2015); B. S. Williams, Nat. Photonics **1**, 517 (2007).
[3] Y. Chassagneux, Q. J. Wang, S. P. Khanna, E. Strupiechonski, J. Coudevylle, E. H. Linfield, A. G. Davies, F. Capasso, M. A. Belkin, and R. Colombelli, IEEE Trans. Terahertz Sci. Technol. **2**, 83 (2012).
[4] A. Crocker, H. A. Gebbie, M. F. Kimmitt, and L. E. S. Mathias, Nature **201**, 250 (1964).
[5] H. A. Gebbie, N. W. B. Stone, and F. D. Findlay, Nature **202**, 685 (1964).
[6] T. Y. Chang and T. J. Bridges, Opt. Commun. **1**, 423 (1970).
[7] G. D. Willenberg, U. Hübner, and J. Heppner, Opt. Commun. **33**, 193 (1980); R. Marx, U. Hübner, I. Abdul-Halim, J. Heppner, Y. C. Ni, G. D. Willenberg, and C. O. Weiss, IEEE J. Quantum Electron. **17**, 1123 (1981).
[8] J. Faist, F. Capasso, D. L. Sivco, C. Sirtori, A. L. Hutchinson, and A. Y. Cho, Science **264**, 553 (1994); J. Faist, *Quantum Cascade Lasers* (Oxford University Press, Oxford, 2013).
[9] Y. Yao, A. J. Hoffman, and C. F. Gmachl, Nat. Photonics **6**, 432 (2012).
[10] T. Y. Chang, T. J. Bridges, and E. G. Burkhardt, Appl. Phys. Lett. **17**, 357 (1970).
[11] A. Tanaka, A. Tanimoto, N. Murata, M. Yamanaka, and H. Yoshinaga, Opt. Commun. **22**, 17 (1977).
[12] C. O. Weiss, M. Fourrier, C. Gastaud, and M. Redon, in *Reviews of Infrared and Millimeter Waves: Optically Pumped Far-Infrared Lasers*, edited by K. J. Button, M. Inguscio, and F. Strumia (Plenum Press, New York, 1984), Vol. 2.
[13] C. H. Townes and A. L. Schawlow, *Microwave Spectroscopy* (McGraw-Hill, New York, 1955).
[14] E. F. Barker, Phys. Rev. **33**, 684 (1929).
[15] D. M. Dennison and J. D. Hardy, Phys. Rev. **39**, 938 (1932).
[16] See http://spec.jpl.nasa.gov for access to the Jet Propulsion Laboratory Molecular Spectroscopy Database.
[17] D. T. Hodges, F. B. Foote, and R. D. Reel, Appl. Phys. Lett. **29**, 662 (1976).
[18] P. Chen, J. C. Pearson, H. M. Pickett, S. Matsuura, and G. A. Blake, J. Mol. Spectrosc. **236**, 116 (2006).
[19] K. Gullberg, B. Hartmann, and B. Kleman, Phys. Scr. **8**, 177 (1973).
[20] E. J. Danielewicz and C. O. Weiss, Opt. Commun. **27**, 98 (1978).
[21] M. A. Belkin, F. Capasso, A. Belyanin, D. L. Sivco, A. Y. Cho, D. C. Oakley, C. J. Vineis, and G. W. Turner, Nat. Photonics **1**, 288 (2007).
[22] Q. Y. Lu, N. Bandyopadhyay, S. Slivken, Y. Bai, and M. Razeghi, Appl. Phys. Lett. **104**, 221105 (2014).
[23] M. Scheller, J. M. Yarborough, J. V. Moloney, M. Fallahi, M. Koch, and S. Koch, Opt. Express **18**, 27112 (2010).
[24] F. Nakajima, T. Furuta, and H. Ito, Electron. Lett. **40**, 1297 (2004).
[25] F. Xie, C. Caneau, H. Leblanc, D. Caffey, L. Hughes, T. Day, and C. Zah, IEEE J. Sel. Top. Quantum Electron. **19**, 1200407 (2013).
[26] D.-T. Nguyen, F. Simoens, J.-L. Ouvrier-Buffet, J. Meilhan, and J.-L. Coutaz, IEEE Trans. Terahertz Sci. Technol. **2**, 299 (2012).
[27] A. Huber, F. Keilmann, J. Wittborn, J. Aizpura, and R. Hillenbrand, Nano Lett. **8**, 3766 (2008).
[28] A. Pagies, G. Deokar, D. Ducatteau, D. Vignaud, and J.-F. Lampin, "THz near-field nanoscopy of graphene layers," in Proceedings of the 40th International Conference on Infrared, Millimeter, and Terahertz Waves (IRMMW-THz), Hong-Kong, China, 23–28 August 2015.