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Generation of very short far-infrared pulses by cavity dumping a molecular gas laser

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Very short far-infrared (FIR) pulses have been generated by cavity dumping an optically pumped molecular gas laser using optical switching techniques. The active element used is an intracavity semiconductor, which is placed under the Brewster angle with respect to the optical axes. This element is made highly reflective by the sudden increase of the free-carrier concentration, induced by above band-gap illumination. In this way high power ($\propto 1$ kW) FIR pulses with a duration less than 10 ns can be produced.

Time-resolved far-infrared (FIR) spectroscopy is a powerful method to observe dynamical properties in semiconductors. Usually in such experiments the materials response to a short FIR laser stimulus is studied as a function of time.[1,2] Obviously, to observe nanosecond processes, one needs FIR laser pulses with a defined and well-known temporal behavior of the same time scale. The conventional way of obtaining FIR pulses is by optically pumping a molecular gas cavity with a pulsed CO_2 laser.[1,3,4] However, with this technique it is very difficult to produce neat FIR pulses with well-defined rise and fall times on a nanosecond time scale. Therefore it is necessary to shape up the existing laser pulse after its generation.

A widely applied method to modulate and control the evolution in time of existing infrared laser radiation is optical switching (OS), which consists of controlling the optical properties of an element in the light path by means of illumination with specific light. Normally, a semi-insulating semiconductor is transparent for (far)-infrared radiation due to the absence of free carriers. Intense above-band-gap radiation (photon energy in excess of the band gap) forms an electron plasma by excitation of free carriers into the conduction band. This electron plasma will strongly enhance the reflectivity of the material, allowing to switch on and off the (far)-infrared radiation in reflection and transmission, respectively.[5,6] OS has been successfully applied in the infrared to produce very short laser pulses[5,7] and has recently been extended into the FIR.[6,8,9] Rikken *et al.*[2,10] have applied OS to terminate abruptly a long FIR laser pulse in order to measure decay times in FIR photoconductive processes in semiconductors. In these experiments falloff times of the order of a few nanoseconds have been obtained. In principle, a second OS element can be used to cutoff the rising part of the pulse in order to make well-defined short pulses.

In the experiments discussed above, the OS element is placed outside the laser system to modify the temporal behavior of the already generated pulse. In this communication we report a novel method in which an OS element is used for cavity dumping a far-infrared laser. This element is a semi-insulating semiconductor slab placed under the Brewster angle inside the cavity of a conventional optically pumped molecular gas laser. During illumination with above-band-gap light, this OS element couples out the laser radiation. Consequently, all further laser action is stopped due to the strong decrease of the cavities Q-factor. Therefore, a FIR pulse is obtained with a duration that equals the time needed by the remaining FIR photons to leave the cavity via the OS element, and consequently the pulse duration is only determined by geometrical factors (twice the distance from the incouple mirror to this intracavity OS element, see Fig. 1).

With this novel cavity-dumping technique ultrashort FIR pulses with well-defined temporal behavior can be realized relatively easy using only one single OS element. In the wavelength range from $\lambda = 100$ to 600 μm the described laser system can produce pulses with a duration of 7 ns and a peak power of $\propto 1$ kW.

The principle of OS is the creation of a dense electron plasma in a semi-insulating semiconductor. The optical properties of this plasma can be characterized by the plasma frequency ω_p, given by

$$\omega_p^2 = N_e e^2 / m_{\text{eff}} \epsilon_0 \epsilon_\infty \tag{1}$$

where N_e is the electron density, m_{eff} the effective electron mass, and ϵ_∞ the optical dielectric constant of the medium.[5] All radiation with frequencies below ω_p is totally reflected from the plasma surface. Using high power, optical laser electron densities of 10^{26} m^{-3} can be reached: For most semiconductors such an electron density corresponds to a plasma frequency in the near-infrared.

However, due to the finite penetration depth of the optical radiation, the plasma only extends several μm into the semiconductor. If the skin depth of the radiation to be reflected is larger than this thickness, some light will be transmitted and consequently the reflection will be less than 100%. For infrared radiation this condition is not very relevant, as skin depths are less than a μm.[5] For FIR however, skin depths are larger and can exceed the thickness of the plasma. An exact determination of the skin

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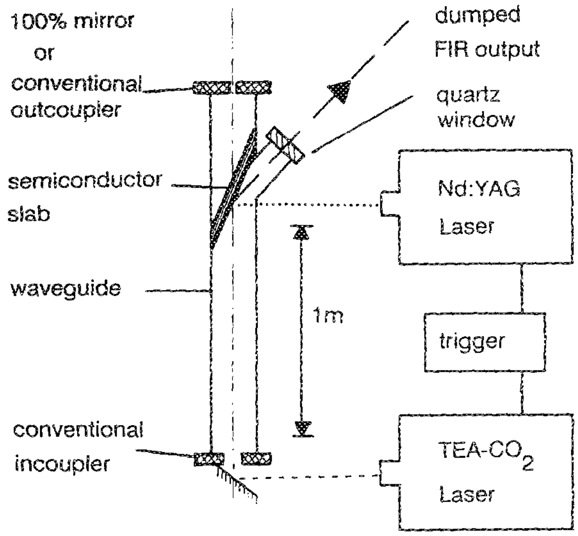

FIG. 1. The experimental configuration of the cavity dump laser.

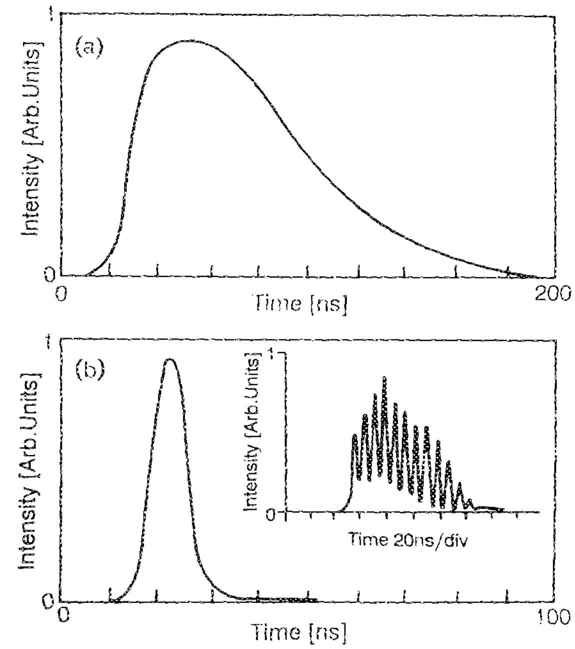

FIG. 2. Far-infrared output of the laser system. (a) Conventional outcoupling response; the FIR follows the CO_2 pump stimulus. (b) Cavity dump pulse produced using intracavity optical switching. The inset shows a cavity dump output in the presence of mode locking.

depth and reflection of FIR in OS elements depends on the electron mean free path and the complicated density profile of the hot electron plasma, and is beyond the scope of this communication. However, estimations of the electron collision frequency ω_τ (deduced from the electron mean free path) can provide a FIR lower frequency limit for which OS still works effectively. In the situation where ω_τ exceeds the frequency of the applied FIR$(\omega_\tau > \omega)$ the dielectric function of the plasma can be approximated[11] with the Hagen–Rubens relation, which predicts a skin depth of the order of 1 μm and proportional to $\sqrt{\lambda}$. Indeed the experiments showed that the OS elements had a frequency dependent reflectivity during illumination, starting at 50% at $\lambda = 500$ μm, and rapidly increasing with decreasing wavelength.

For a direct-gap semiconductor as GaAs it is evident that the induced plasma will be present immediately after the beginning of the illumination and will last only some nanoseconds after cessation of this stimulus. For an indirect semiconductor such as silicon the induced plasma can stay up to microseconds after the stimulus due to the reduced decay rate back to the valence band for these materials. The strong varieties in the temporal behavior of the optical properties of the induced plasma are also strongly related to the electron diffusion into the material, and to the wavelengths involved. A detailed description of these effects is presented in Ref. 5.

The experimental setup is shown in Fig. 1. To generate the infrared pump pulse a hybrid TEA-CO_2 laser (Lumonics 820) was used. This laser incorporates an atmospheric pressure section for higher power and a low pressure section providing a narrow gain bandwidth for efficient pumping. The typical pulses have a power of 500 kW with a duration of 100 ns and a repetition rate of 10 Hz.

The infrared is coupled into the FIR cavity by means of a conventional hole mirror coupler. The FIR cavity consisted of a quartz tube waveguide with an inner diameter of 10 mm through which the medium flowed. With a normal hole outcoupling mirror (see Fig. 1) the FIR pulse follows that of the IR as is shown in Fig. 2 (a).

In the cavity dump mode, the outcoupling mirror is replaced by a 100% reflecting gold mirror. A semi-insulating GaAs slab (thickness 0.5 mm) was installed in the cavity under the Brewster angle at 110 cm from the incoupling mirror. To induce the electron plasma, second harmonic radiation of a Q-switched pulsed Nd:YaG laser (Quanta Ray DCR2A) with a wavelength of 533 nm was used. This laser produced pulses with 1 J energy in 10 ns. A typical result of a FIR pulse obtained by cavity dumping is shown in Fig. 2(b). In order to monitor the temporal behavior of the FIR a room-temperature Schottky diode detector was used; occasionally a fast liquid helium cooled multi-quantum-well photodetector (response time less than 2 ns) has been used as well.[12] The FIR pulse powers were measured with a calibrated pyroelectric detector.

Some typical pulse data are shown in Table I. The data for the 496 μm wavelength correspond to a superradiant laser transition and will be discussed separately further on. For all other wavelengths, the very short pulses have a

TABLE I. Experimental results for different FIR laser lines. The data marked * present a superradiant transition.

λ (μm)	Medium	Pump line	Duration (ns)
90.9	NH_3	9R16	7
148	NH_3	9P34	7
231	CH_3F	9R30	7
292	NH_3	10R06	7
496*	CH_3F	9P16	10–20

duration corresponding to the time needed to traverse the cavity twice [having a length of 110 cm (which responds to $\simeq 7$ ns), see Fig. 2(b)].

In the cavity dump mode, the polarization of the CO_2 light must be parallel to the polarization of the emitted FIR light. The reason for this is that otherwise the OS element does not function as a Brewster window for the CO_2 light which will then be blocked. Consequently there will be no inversion between the OS element and the 100% mirror, resulting in a net loss instead of a gain in this part of the cavity. Now mode-locking occurs, giving rise to a rapidly oscillating FIR pulse as shown in the inset of Fig. 2(b). This effect can also be observed for correctly polarized light, when the CO_2 laser is attenuated and the OS element is slightly tilted from the Brewster angle. Also if the OS element has a poor surface quality (for instance a damaged surface caused by too much YAG-laser power) some mode locking can occur.

In direct-gap semiconductors, the reflectivity only stays at a high level during 10–20 ns. Therefore the cavity dump system offers the possibility to dump two or even more short pulses out of the cavity during a single CO_2-laser pulse ($\simeq 100$ ns) by simply splitting the YAG laser beam and geometrically delaying a part of this beam.

As discussed above, in the cavity dump mode only FIR output is possible during illumination of the OS element. However, due to some spurious reflection at the surfaces a FIR background signal will be present as long as the CO_2 stimulus is present. Under normal conditions (nondamaged clean surface) this background has about 2% of the intensity of the cavity dumped pulse. To reduce the background after cessation of the FIR cavity dump, a second OS transmission device made of Si (an indirect gap semiconductor) has been placed in the outcoming beam. This element was illuminated with a part of the YAG beam that had been appropriately delayed, and as such blocked all further FIR radiation ($\simeq 99\%$). In this way the background after the cavity dumped pulse could be reduced to less than 0.1%. By tuning the delay it was also possible to shorten the FIR pulse to a few nanoseconds. As mentioned above, such a shortening can in principle also be realized by decreasing the distance between the OS element and incouple mirror of the FIR laser.

If superradiant transitions occur, the medium gain is so high that there is no need for a cavity to obtain FIR laser action, as is the case for $\lambda = 496\ \mu$m (see Table I).[13,14] FIR laser action will now be present as long as a CO_2 stimulus is available, independent on the optical state of the OS device. The duration of the FIR pulse reflected out by the OS element is governed only by the time behavior of its reflection efficiency. As discussed before, for a direct-gap semiconductor this time will be 10–20 ns when a stimulating YAG pulse of 10 ns is used, which is confirmed by the experimental results of Table I.

In summary we described a technique to obtain far-infrared pulses by cavity dumping a FIR laser using optical switching. The resulting pulses have rise and fall times of about 1 ns and a duration of 7 ns defined by geometrical factors. The advantage of this method over the conventional way of generating this kind of pulses using a transmission and a reflection OS to modify an existing pulse, is that much higher output powers are attainable. A bycoming advantage is that this system has a rather straightforward experimental configuration which improves the stability and reduces any jitter.

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