Far infrared spectroscopy with subpicosecond electrical pulses on transmission lines

R. Sprik,[a] I. N. Duling, III, C.-C. Chi, and D. Grischkowsky
IBM T. J. Watson Research Center, P. O. Box 218, Yorktown Heights, New York 10598

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Optically generated and detected electrical pulses on transmission lines in the subpicosecond range have frequencies extending up to 1 THz, thereby covering the far infrared region of the spectrum from 0 to 30 cm^{-1}. We have studied the propagation of these short pulses through a section of the transmission line covered with erbium iron garnet which shows distinct absorption lines in the far infrared at low temperatures (2–30 K). The absorption and dispersion of the garnet modify the shape of the pulse, and the absorption spectrum is obtained by Fourier transforming the propagated pulse shape.

Recently optoelectronic techniques have been used to generate and detect subpicosecond electrical pulses on coplanar transmission lines.[1,2] The frequency bandwidth of these short electrical pulses ranges up to 1 THz and covers an interesting part of the far infrared energy spectrum (0–30 cm^{-1}, $\lambda > 330\,\mu m$) in which can be found the gap frequencies of superconductors, magnetic excitations, and the far infrared modes in lattices and molecules. This situation, plus the fact that the earlier observations showed that the subpicosecond pulses broadened to only 2.6 ps after propagating 8 mm on the transmission line, encouraged us to consider spectroscopic applications of these guided wave electrical pulses.

The recent use of superconducting transmission lines showed that by observing pulse reshaping as a function of propagation distance, the frequency-dependent absorption and dispersion of the line could be obtained. The absorption sharply increased at the frequency associated with the breaking of the Cooper pairs in the superconducting niobium metal lines.[2] In this letter we demonstrate that this time domain optoelectronic technique can be more generally used for far infrared spectroscopy by covering a section of the transmission line with an absorbing material and studying the consequent reshaping of the transmitted pulse. We demonstrate the feasibility of this spectroscopic method by measuring the magnetic resonances in erbium iron garnet (ErIG) and studying the temperature dependence of the frequencies between 2 and 30 K. The particular choice of ErIG was guided by the fact that this system is known to have sharp absorption resonances in the currently accessible spectral range of the technique and was readily available to us.

The optoelectronic generation of short electrical pulses is based on the temporary shorting of the charged transmission line by carriers excited in the photoconductive material (ion-implanted Si on sapphire) underneath the lines with a short laser pulse. The minimum electrical pulse width, which is measured by the same electro-optic technique, appears to be limited by the lifetime of the photoexcited carriers in the photoconductive material.[3] Figure 1 shows the experimental arrangement, the coplanar transmission line, and the focused spots of the laser, where the electrical pulse is generated by focusing the exciting beam between the lines ("sliding contact") and detected after propagation by the sampling beam focused on the gap. By scanning the time delay between the exciting beam and the sampling beam one measures the pulse shape. Both beams are derived from the same laser, a prism-compensated, colliding-pulse, mode-locked, dye laser producing 70-fs pulses at a repetition rate of 100 MHz.[4] The exciting beam is chopped at 2 kHz and the resulting photocurrent signal is detected as a function of time delay with a lock-in amplifier, whose output is stored in a computer for further processing.

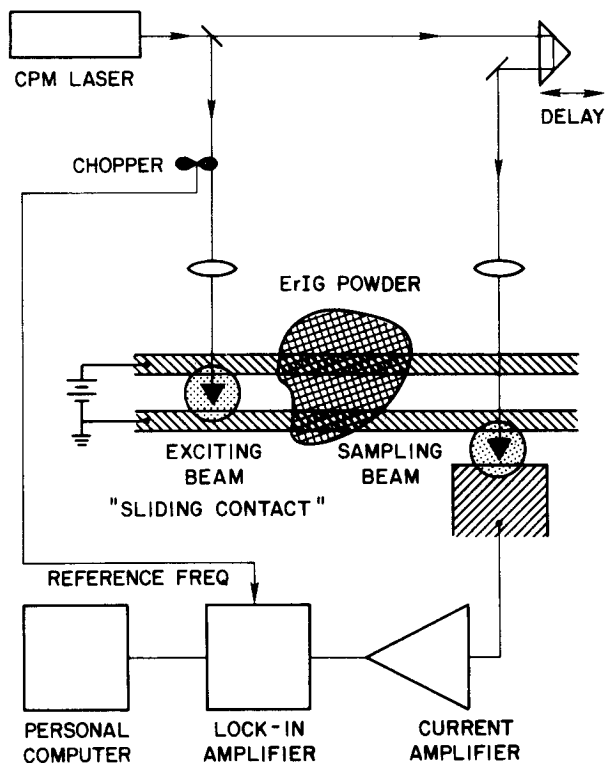

FIG. 1. Schematic diagram of the experimental arrangement together with the coplanar transmission line configuration.

[a] Permanent address: Natuurkundig Laboratorium, Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands.

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The transmission lines used in this work have a characteristic impedance of 100 Ω and are composed of two coplanar aluminum lines with a linewidth of 5 μm and a spacing double the linewidth. The metal lines were deposited on commercial silicon-on-sapphire substrates. After lift-off patterning of the aluminum, the samples were implanted with oxygen to shorten the lifetime of the photoexcited carriers below 1 ps. Performances of these lines are described in detail by Gallagher *et al.*[2] A small section (2–3 mm) of the transmission line was covered with a thin layer of powder of the infrared-absorbing material. The samples were mounted in an optical ^{4}He flow cryostat enabling measurements between 2 and 300 K. Since the photocurrent was modulated at 2 kHz, the electrical wiring of the cryostat could be simple and did not require high-frequency performance.

Figure 2(a) shows the shape of the propagated electrical pulse without any powder sample on the transmission line at 5 K. The pulse is shown as it was used for the numerical analysis, with the data truncated at 10 ps. Room-temperature scans have confirmed that no extraneous reflections are present in the truncated region. When the line was covered with ErIG the pulse shape dramatically changed to that shown in Fig. 2(b), where strong oscillation is now seen for the longer time delays. This time domain measurement is converted to the frequency domain in Fig. 2(c), by Fourier transforming the two propagated pulse shapes. The resulting spectra show that when the sample is in place, strong resonances appear at the frequencies 10.0 and 4.3 cm^{-1} at $T = 5$ K. A crude estimate of the spectral resolution of the technique based on a total delay time scan of 45 ps is $\simeq 0.7$ cm^{-1}. The measured linewidth, especially for the higher frequency mode, is $\simeq 1.5$ cm^{-1}, clearly broader than the spectral resolution. We identify these observed lines as magnetic resonances in ErIG, which we will now discuss.

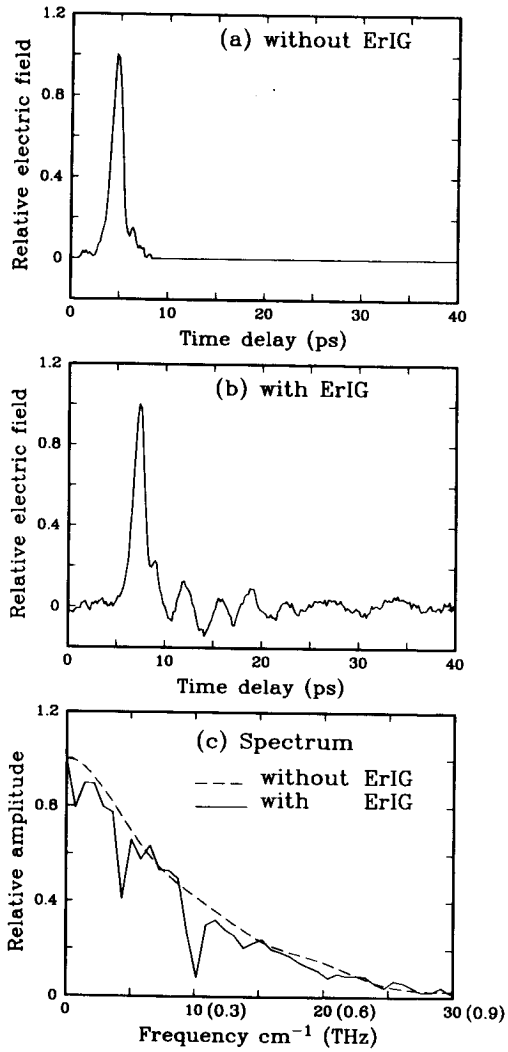

FIG. 2. Pulse shape after propagation on a transmission line without (a) and with (b) ErIG powder covering the line. The Fourier transform (c) of the propagated pulse (b) shows distinct absorption at the exchange 10.0 cm^{-1} and the ferrimagnetic 4.3 cm^{-1} resonance in ErIG ($T = 5$ K).

Rare-earth iron garnets ($5Fe_2O_3 \cdot 3R_2O_3$) display a number of well-known sharp resonances in the far infrared range of the spectrum relevant for the current experiments.[6,7] These garnets have two magnetic modes which originate from the exchange resonance[8] between the iron and the rare-earth sublattices in the ferrimagnetic ordered material. The anisotropy in the coupling results in a low frequency mode of ferrimagnetic nature.[7] At somewhat higher frequency are two resonances in the rare-earth garnets associated with the exchange field splitting induced by the iron lattices. These resonances have all been observed and extensively studied in YbIG. The exchange resonance has been observed for the first time by Sievers and Tinkham[9,10] in YbIG using a far infrared monochromator. The temperature dependence of the frequency confirmed their model[7] for the anisotropic coupling in the rare-earth iron garnets. Using a spectrometer with an extended spectral range, Richards[11] observed the exchange resonance and in addition the predicted ferrimagnetic resonance in YbIG. ErIG, which we used for our study, is expected to behave analogously to YbIG and has been studied in some detail by Sievers and Tinkham.[10] In particular for ErIG the exchange resonance was observed at 10.0 cm^{-1} ($T = 2$ K), which is consistent with our measurement of 9.6 cm^{-1} ($T = 2.5$ K). Sievers and Tinkham could not observe the ferrimagnetic resonance in ErIG because it was outside their detection range. To our knowledge the ferrimagnetic resonance in ErIG which we observe at 4.3 cm^{-1} has not been reported previously. Based on simple arguments considering the observed exchange field splitting and exchange resonance, the ferrimagnetic resonance should be at $\simeq 2$ cm^{-1}. The same arguments in the case of YbIG predict the ferrimagnetic resonance to be at 4 cm^{-1},[6,7] while the observed line is at 3 cm^{-1}.[11] A better agreement between theory and experiment can only be expected after a more detailed analysis of the garnet system. We do not observe the resonances in ErIG associated with the exchange splitting which Sievers and Tinkham measured at 18.2 and 21.6 cm^{-1}, most likely because our available IR power is considerably reduced between 20 and 30 cm^{-1}, compared to the lower frequency ranges. With an improved signal-to-noise ratio, we should also be able to resolve these lines.

As mentioned before, the magnetic resonances display a frequency shift as a function of temperature which depends intricately on the various coupling parameters in the garnet

system. Figure 3 shows the temperature dependence of the exchange and the ferrimagnetic resonance in ErIG derived from our far infrared absorption measurements. Especially the exchange resonance shifts upwards as a function of temperature from 9.6 cm^{-1} at 2.5 K to 12.1 cm^{-1} at 25 K. This temperature dependence is qualitatively consistent with the measurements on YbIG and the report by Tinkham[6] for ErIG. We did not try a more detailed analysis of the temperature dependence, since the appropriate anisotropy coefficients are not well known for ErIG. The observed shifts clearly illustrate the fact that the observed absorption lines are an intrinsic property of the garnet powder and not an artifact of the transmission line technique.

The new technique has a number of advantages and disadvantages in comparison with the traditional cw far infrared spectroscopic techniques based on a mercury arc light source and a monochromator or interferometer.[5] A cw light source has typically $\simeq 10^{-10}$ W available in a bandwidth of 1 cm^{-1},[6] and an infrared interferometer has a spectral resolution better than 0.1 cm^{-1}. The accessible spectral range of such a system is usually from a few cm^{-1} to many hundreds of cm^{-1}. The generated 1-ps electrical pulse (10 mV amplitude) at a 100-MHz repetition rate generates an average cw power of 10^{-10} W. This power is distributed in amplitude of the spectral components as shown in Fig. 2(c). From this spectrum we calculate that for the frequency of 10 cm^{-1}, $\simeq 2\times 10^{-12}$ W is available within a 1 cm^{-1} bandwidth. However, the effective power is enhanced a factor $\simeq 250$ due to the fact that the actual measurement occurs only during the 45 ps-scan time. This makes the effective available infrared power comparable with cw sources. Similar to an infrared interferometer, the optoelectronic technique detects the electric field and not the intensity. This feature makes the technique sensitive to changes in phase and amplitude of the interfering spectral components resulting in an excellent signal-to-noise ratio with modest infrared power. The current spectral resolution of $\simeq 0.7$ cm^{-1} can be increased simply by extending the delay time scan beyond 45 ps. In order to be able to do this, it is essential to eliminate reflection from the ends of the transmission lines, because these will disturb the actual measurements. Currently the spectral range of the pulse technique is between 1 and 30 cm^{-1}, covering frequencies from the microwave range up to the more readily accessible far infrared frequencies. Because the electrical pulses propagate as a guided wave along the transmission line, the electric and magnetic fields are strongly localized at the surface. This feature makes the technique quite suitable for studying surface excitations. Furthermore, the required amount of sample is very limited since only the area near the transmission lines has to be covered. Perhaps the most attractive feature is the possibility of time-resolved far infrared spectroscopy on a picosecond time scale, especially when the

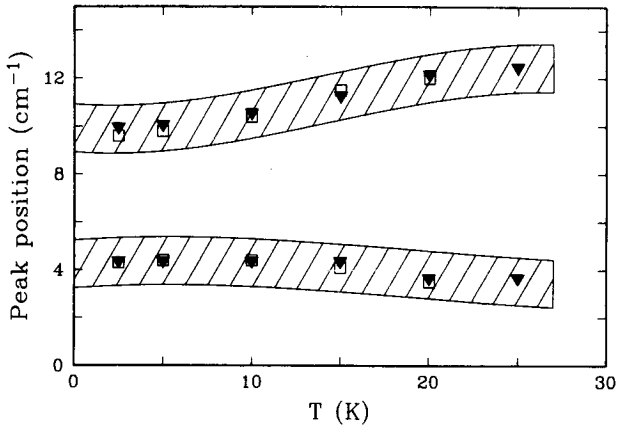

FIG. 3. Temperature dependence of the exchange and the ferrimagnetic resonance in ErIG. The different symbols are of two different runs with the same sample. Shaded area indicates the resolution of the data.

available far infrared power can be enhanced to a level where the population of the magnetic levels is strongly influenced. For this case, one can consider applying the technique to observe spin echos from magnetic excitations in the far infrared. The power of the pulse can be increased significantly by amplifying the exciting laser pulses. Preliminary attempts show that electrical pulses of the order of 1 V can be produced, thereby increasing the available power by four orders of magnitude. These unique features of the optoelectronic technique clearly offer new possibilities for the study of far infrared transitions.

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