Subterahertz spectroscopy at He-3 temperatures

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We report on the design and implementation of an instrument for spectroscopic studies of materials at sub-terahertz (THz) frequencies at temperatures down to 340 mK. We achieved consistent operation under these rather extreme conditions by coupling a modified Martin–Puplett interferometer to a single cryogenic unit housing two independently controlled He-3 platforms: one as a sample stage and the other for bolometric detectors. Both the optical scheme of the interferometer and detector layout are tailored for the use of the two-channel data acquisition mode which is especially advantageous for measurement of absolute values of reflectance as well as for high-resolution spectroscopy. We document the reliable performance of the sub-THz apparatus with several experiments exploring electrodynamics of both conventional and high-Tc superconductors.

I. INTRODUCTION

Characteristic energy scales of a large variety of fundamentally important phenomena fall into the terahertz region: 100 GHz–3 THz (3–100 cm⁻¹). These include absorption bands of interstellar medium, molecular vibrational modes, emission of hydroxyl radical (OH), and many others. The terahertz (THz) range is equally important for technological applications involving aircraft ranging and guidance under zero visibility conditions, imaging systems, as well as noninvasive detection of plastic weapons or explosives. The terahertz range is exceptionally rich in terms of diverse effects in condensed matter physics that fall within this frequency interval. An incomplete list includes magnetic and cyclotron plasma resonances, localized lattice modes, Josephson plasma modes, and energy gaps in superconductors, absorption structure of organic molecular magnets, response features of materials with strongly correlated electrons, as well as characteristic features of metallic nanoparticles and semiconducting nanostuctures. Nevertheless, spectroscopy in the THz frequency range continues to be a challenging undertaking. Experimental difficulties related to this task are epitomized in a common reference to the THz frequency interval as to a "gap" between microwave and infrared (IR) methods. Indeed, it is difficult to extend microwave methods beyond a couple of hundred gigahertz (GHz) and likewise the extension of IR methods below the usual boundary of 3 THz (100 cm⁻¹) is similarly complicated. The latter task is especially challenging when the studied materials are highly reflective and available only in the form of millimeter-sized samples.

Driven by the importance of the THz frequency interval, various research teams have made significant advances in conquering this largely unexplored territory. Perhaps the most innovative approach is the time domain THz spectroscopy (TDS) pioneered by Grishkovsky. Here radiation is generated and detected using hertz dipoles: metal stripes with a narrow gap prepared on the surface of an amorphous semiconductor. A laser pulse creates photoconductive carriers in a semiconductor, thus activating the dipoles. The duration of the photocurrent is of the order of 1 ps and thus the dipole generates radiation in a frequency region 0.1–4 THz (3–120 cm⁻¹). This technique allows one to obtain the real and imaginary parts of the optical constants of a medium through which radiation propagates without the use of Kramers–Kronig (KK) analysis. TDS is employed exclusively in transmission mode. Several groups employed semiconductor submillimeter sources such as backward wave oscillators and far-IR lasers. The scope of the experimental information extracted from these measurements is dramatically enhanced through the use of Mach–Zehnder or Fabry–Pérot interferometer schemes permitting evaluation of the optical constants without KK analysis. Yet another approach is based on the use of synchrotron light sources instead of conventional black-body sources in Fourier transform spectrometers. This latter approach capitalizes on the superior brightness of synchrotron sources compared to mercury lamps in the THz region. Synchrotron operation in the regime of "coherent" emission may be especially advantageous for spectroscopy in THz regime.

In this article we describe the design and implementation of a sub-THz spectrometer based on a Martin–Puplett (MP) interferometer which has allowed us to achieve convenient operations at frequencies as low as 200 GHz using laboratory
A serious disadvantage of Michelson interferometers is that in our setup the intensity of the THz beam is modulated by a rotating polarizer located in front of the first intermediate focus (Fig. 1); the fixed polarizer is positioned at the output of the interferometer. An obvious advantage of this configuration is that the output beam is linearly polarized. The output fixed polarizer is oriented approximately 45° with respect to the direction of the beam so that radiation exiting the interferometer is split into SAMPLE and REFERENCE channels. Note that the intensity in the reference channel is produced without reducing the signal level in the sample channel. Standard MP design implies that radiation reflected from the polarizer is “wasted” since no useful information is extracted from the analysis of this signal. In our scheme, we use the second channel to compensate for signal fluctuations and system drift in the course of the data acquisition process as will be described below. In the THz range, wire grid polarizers deliver very high beam-splitting efficiency and the signal level in both channels of our interferometer is approximately the same (see top panel of Fig. 5).

In Fig. 2 we show the power spectrum in the frequency region below 25 THz in one of the channels collected by means of a Si bolometer operating at 4.2 K. The upper frequency limit is imposed by a cutoff filter in the bolometer. The inset shows the interferogram recorded with a Stanford SR-850 lock-in amplifier. If perfect polarizers were employed within the MP scheme the intensity of the interferogram background away from the main peak would be vanishingly small so that \( I_{\text{background}}/I_{\text{peak}} \approx 2 \). In practice, polarizers are not immune to minor leakage of unwanted polarization and \( I_{\text{background}}/I_{\text{peak}} \) is always finite. We found that in our setup \( I_{\text{background}}/I_{\text{peak}} \) is less than 1/20 with detector cutoff at 25 THz (750 cm\(^{-1}\)) and 1/35–1/50 when the cutoff is at 1500 GHz (45 cm\(^{-1}\)). These “high contrast” interferograms produced by our interferometer is a testimony to the excellent efficiency of polarizers employed in the optical scheme. Another indicator of more than adequate polarizer performance is that the power spectrum (obtained by Fourier transforming the interferogram) shows significant signal levels even at \( \omega > 15 \) THz and is comparable to data generated by means of a Michelson interferometer working with a 3.5 \( \mu \)m mylar beamsplitter.

B. Dual platform He-3 cryostat

Interferometry-based spectroscopy of materials at low temperatures in the THz range requires the use of at least two
cryogenic units: one for the studied specimen and the other for a He-cooled thermal (bolometric) detector. Usually separate cryostats are employed for these two essential units whereas focusing and polarizing optics components are located in an evacuated compartment of a spectrometer. We have chosen a different approach to the design of the cryogenic units for our sub-THz instrument (Fig. 3): two He-3 cold platforms (one for the sample and the other for detectors) are located inside the same cryogenic unit. The focusing mirror and polarizer are attached to the bottom of a He-4 bath. This scheme has numerous advantages. In particular, the cryogenic system presented in Fig. 3 eliminates the need for additional sets of widows and cutoff filters that would be required if “sample” and “detector” cryostats had been decoupled. This cryostat has been fabricated based on our design at Infrared Laboratories, Tucson, Arizona. Similar cryogenics has been employed in a spectroscopic setup recently developed at Brock University. The operation of compact He-3 platforms using activated charcoal absorption pumps is described elsewhere. All the optical components and polarizer holders used in our system are built in house.

The cryostat is attached to the vacuum compartment of the MP interferometer. The entrance widows are made of quartz which is transparent below 3.5 THz. We use two sets of cutoff filters: one at the 77 K nitrogen-cooled shield and another attached to the He-4 cooled surface. We have chosen the upper cutoff to be at 1.5 THz (45 cm⁻¹) which makes an excellent overlap with the frequency range of a Michelson interferometer available in our laboratory: (25–27 000 cm⁻¹). Incident beams coming from the interferometer are focused on the entrance of light pipes (ID=12.5 mm) installed directly behind the 77 K cutoff filters of the cryostat. The light pipe coupling allows us to minimize the diameter of the windows and filters without compromising the f-number of the system. Light pipes are fabricated from gold-plated electroformed nickel. The He-4 cooled cutoff filters are installed inside the light pipes. The light pipe in the reference channel guides radiation directly to the Winstone cone of the reference detector. We employed doped Si bolometers made by Infrared Labs. Radiation in the sample channel is refocused using an elliptical mirror. Despite the fact that about 10 cm of the optical path occurs through the light pipe we obtain good image quality on the sample. The studied specimens are glued to a copper cone that is anchored to the cold surface of the sample He-3 platform. Only radiation reflected from the sample surface is directed towards the collecting light pipe that delivers the THz radiation to the Winstone cone of the sample bolometer.

Temperature is monitored using Ge sensors (Lakeshore) directly at the bottom of the He-3 pot and at the top of the cone supporting studied specimens. Typical readings are 310 mK at the cold surface and 330–340 mK at the top of the cone. The duration of low temperature operation of He-3 platforms critically depends on details of a particular experiment. Operation at temperatures elevated compared the base \( T = 320 \text{ mK} \) significantly compromises He-3 holding times. Temperature can be controlled either using a heater attached to He-3 platforms or by warming the charcoal pumps acting on the He-3 pots. If it is necessary to achieve temperatures above 1.7 K, the sample platform is linked to the He-4 bath of the cryostat using a mechanical thermal switch. We were able to reach temperatures up to 20–25 K on the sample platform while the detector platform is still kept at 320 mK. Typical He-3 holding times on the bolometer platform exceed 24 h with about 7.5 l of He-3 gas in our system. In order to achieve these reasonable holding times it was necessary to minimize radiation heating. We developed housings...
that are thermally connected to the He-4 bath around each of the He-3 platforms. This prevents exposure of these platforms to surfaces warmer than 4.2 K (or 1.6 K under pumping). We were able to realize reliable operation of both He-3 platforms when the temperature of the He-4 bath is either 4.2 or 1.6 K (under pumping).

The cryostat shown in Fig. 3 is equipped with three infrared ports: sample, reference and through ports. This diagram depicts a system when it is arranged for reflectance measurements. Depending on the particular goals of the experiment the system can be easily reconfigured for other types of measurements. For instance, transmission experiments can be enabled by guiding radiation directly from the sample port to the sample bolometer (bypassing a refocusing mirror) with a studied specimen mounted on a sample platform. Alternatively, transmission measurements can be carried out using an optical layout shown in Fig. 9. The advantage of the latter approach is that the THz beam is focused on the sample surface permitting studies of smaller specimens without compromising signal intensity. The third port allows guiding of the THz beam through both He-3 platforms and is intended for spectroscopy with external detectors.

The light pipes employed to couple interferometer to the dual platform cryostat inevitably depolarize THz radiation. It is therefore imperative to employ a polarizer installed inside the cryostat for spectroscopic studies of anisotropic materials where linearly polarized light is required. The constrained space inside the cryostat rules out the use of bulky commercial polarizer holders. For this purpose we have developed an attachment shown in Fig. 4. A polarizer (Al wire grid on mylar substrate) is stretched on a frame supported by three ball bearings. The ID of the frame is 12.5 mm and is identical to the ID of the light pipes. The side of the frame has markings every 5°.

FIG. 4. A photograph of a custom made polarizer holder (with front cover taken off). The base plate is 0.5 in.×1.9 in.×1/4 in. supports a frame with a polarizing material. Frame can be rotated using a hexagonal knob. Side of the frame has markings every 5°.

with a 5° step. The frame can be conveniently rotated using a plastic coupler also providing 2:1 gearing for accurate readjustment of the polarizer angle.

III. THz SPECTROSCOPY USING A MARTIN–PUPLETT INTERFEROMETER

A. The two-channel data acquisition mode

In Fig. 5 we show a typical power spectra collected using the cryostat depicted in Fig. 3. Both sample and reference channel data are presented. For the purpose of these measurements a large area stainless steel reflectance is mounted on the cone in the sample channel. The upper cutoff is determined by filtering optics. The power spectra are peaked at 800 GHz with intensity extending below 400 GHz. A standard performance characteristic of a spectrophotometric systems is the so-called 100% line: a ratio of two power spectra obtained in two repetitive runs under nominally identical condition. An examination of the 100% line displayed in the bottom frame of Fig. 5 indicates that our instrument permits acquisition of data down to nearly 200 GHz with good reproducibility and with the signal-to-noise ratio better than 1%.

In order to extract complete information about properties of a system through a spectroscopic experiment it is necessary to measure absolute values of experimentally accessible parameters: reflectance $R(\omega)$ and/or transmission $T(\omega)$. In the case of reflectance measurements this task requires that a studied specimen is replaced by a metallic mirror with known reflectivity. Then the sample reflectance is obtained by taking the ratio of the two signal intensities: $R(\omega) = I_{\text{sample}}(\omega)/I_{\text{mirror}}(\omega)$. This approach is generally realized through a mechanical repositioning of a sample so that it can be replaced by an identical mirror in the optical path. However, convenient mechanical schemes for switching between sample and reference cannot be employed when the sample is cooled down to He-3 temperatures. Instead, we used an...
approach involving in situ coating of a sample with lead. Within this approach we first collect \( I_{\text{sample}}(\omega) \) at various temperatures with simultaneous recording of the spectrum in the reference channel \( I_{\text{reference}}(\omega) \). After a series of measurements is completed we coat the sample with approximately 1–2 \( \mu \)m of lead and collect another full set of spectra \( I_{\text{lead}}(\omega) \) again with simultaneous detection of the data in the reference channel. A double ratio: \( \frac{I_{\text{sample}}(\omega)/I_{\text{reference}}(\omega)}{I_{\text{lead}}(\omega)/I_{\text{reference}}(\omega)} \) is then equivalent to \( I_{\text{sample}}(\omega)/I_{\text{lead}}(\omega) \); the latter result can be used to obtain reflectivity of the studied material since reflectance of lead is known. Therefore, coating technique combined with two-channel data acquisition enables absolute measurements of \( R(\omega) \) without need to mechanically reposition studied specimens.

The coating technique\(^{33} \) when applied to more common spectroscopic investigations in infrared range delivers accurate results even in the case of microsamples.\(^{34,35} \) The reference signal is detected simultaneously with the signal reflected from the samples. Because sample and reference interferograms are collected during the same scan of the mirror the impact of system instabilities is minimized. When the instrument is operated at the highest possible resolution one scan may require 15–25 min or more. System drift is always present over such long periods of time; the role of the drift is also minimized by simultaneous data acquisition. This is exemplified in Fig. 6 where three 100% lines are displayed. Two of these lines were obtained by referencing two spectra generated in two sequential runs with a time difference of approximately 30 min. The other 100% line was obtained by taking a double ratio as described before. An improvement of the latter approach for the base line is clearly visible.

B. Reflectance and transmission measurements

In Fig. 7 we display the results of reflectance measurements for the superconductor \( \text{La}_{1.875}\text{Sr}_{0.125}\text{CuO}_4 \) carried out by means of our THz spectrometer (Michelson interferometer combined with the dual platform He-3 cryostat). Details of sample preparation and characterization were reported elsewhere.\(^{36} \) Cuprate high-\( T_c \) superconductors are strongly anisotropic with “metal-like” properties in the direction along the \( \text{CuO}_2 \) layers (\( ab \) plane of the crystal) coexisting with a nearly insulating response in the \( c \)-axis direction across the \( \text{CuO}_2 \) layers.\(^{37} \) The dominant feature of the superconducting reflectance is a well-defined plasma edge. The absolute values of \( R(\omega) \) approach 1 at the lowest energies; this region is followed by a sudden drop of reflectance forming an edge-like structure. At higher frequencies reflectance gradually recovers to the normal state value (close to 40% below 1 THz). The plasma edge is believed to originate from Josephson coupling between the \( \text{CuO}_2 \) planes and is often referred to as Josephson plasma resonance (JPR). One goal of the study reported in Fig. 7 was to resolve the frequency dependence associated with the JPR mode down to the lowest possible energies. The measurements have been performed on the \( ac \)-face of the crystal and required the use of linearly polarized light with \( E//c \). In order to align the polarizer we have replaced the bolometric detector in the sample channel with a pyroelectric sensor and illuminated the entrance light pipe in this channel with a globar source chopped at 110 Hz. The wire grid polarizer employed in this work delivers approximately 90% polarization efficiency at about 1500 cm\(^{-1} \)—the frequency range where the intensity of globar source is peaked. The reflectance anisotropy of \( \text{La}_{1.875}\text{Sr}_{0.125}\text{CuO}_4 \) is largest in this frequency range. Therefore, the polarization dependence of the mid-IR signal measured with the lock-in and recorder with the step of 5° provided us with a reliable way to select the proper direction of the polarizing wire grid.

The absolute value of \( R(\omega) \) in THz frequencies for \( \text{La}_{1.85}\text{Sr}_{0.125}\text{CuO}_4 \) crystal (3.5\( \times \)3.5 mm\(^2 \)) on the \( ac \) face shown in Fig. 7 was generated using the following procedure. We first measured \( I_{\text{sample}}(\omega) \) simultaneously with \( I_{\text{reference}}(\omega) \) in the course of several scans of the interferometer and then averaged ratios of the two signals to obtain \( R'(\omega) \). We then coated the sample with Pb and measured \( I_{\text{Pb}}(\omega) \) simultaneously with \( I_{\text{reference}}(\omega) \). The ratios of the latter spectra also were averaged to produce \( R''(\omega) \). Because reflectance of Pb is known (as will be discussed later), abso-
lute reflectivity of a studied specimen can be determined as $R(\omega) = R'(\omega)/R''(\omega)$. This procedure of double ratios allows us to collect sufficient data statistics in the process of repeated experiments and at the same time minimizes the impact of system long term drifts/instabilities. In Fig. 7 we also show the data for La$_{1.85}$Sr$_{0.125}$CuO$_4$ (E/c) generated with the Michelson interferometer equipped with a conventional 1.6 K bolometer. Limitations of both detector and beamsplitter employed in the latter experiment impose a cut-off close to 1 THz. This comparison also shows that the new instrument described in this work allows us to significantly extend the lower cutoff of spectroscopic measurements. We find a small discrepancy between the two spectra (up to 4% at the upper cutoff of the MP interferometer).

We used lead as a reference in our experiments for several reasons. The main advantage of Pb is that its $R(\omega)$ at $\omega < 800$ GHz is exactly 100%. Indeed, lead is a superconductor with $T_c = 7.1$ K and at $T < T_c$ its electrodynamics is in accord with Mattis–Bardeen theory prescribing $R(\omega) = 1$ for frequencies below the energy gap $2\Delta$ (bottom panel of Fig. 8). Unity reflectance of Bardeen–Cooper–Schrieffer (BCS) superconductors has been experimentally verified through direct absorption measurements. Moreover, lead can be conveniently coated atop of the studied specimens at low $T$ unlike other commonly used reference metals including gold. The earlier circumstances make lead an “ideal” reference material for low temperature spectroscopy in sub-THz range. However, at frequencies above $2\Delta = 800$ GHz the reflectance of lead decreases. Because this drop of $R(\omega)$ occurs well within the energy range covered by our apparatus (Fig. 8) it is imperative to account for this deviation for an actual lead film used as a reference. Spectra calculated within the framework of the Mattis–Bardeen theory show that the amount by which reflectance decreases at $\omega > 2\Delta$ critically depends on the normal state resistivity $\rho_{DC}$. Therefore, accurate referencing of the data using Pb implies measurements of the resistivity of in situ grown films. This measurement is not complicated but requires knowledge of the thickness of the actual film. We found that it is more practical to characterize reflectance of lead reference using an alternative all-optics approach. The normal state reflectance of lead is adequately described with the Hagen–Rubens formula $R_n(\omega) = 1 - \sqrt{2\omega \rho_{DC}/\pi} / \omega_T$, superconducting reflectance $R_s(\omega) = (\omega < 2\Delta) = 1$. Therefore, the $R_s/R_n$ ratio at $\omega = 2\Delta$ allows us to estimate the magnitude of $\rho_{DC}$ for the actual film obtained through in situ coating of studied specimens.

We have investigated a large number of thick Pb films grown both in situ and in a separate evaporation setup. Typical results are shown in the top panel of Fig. 8 where $R_s/R_n$ spectra are plotted. This data set demonstrates that the sub-THz apparatus described here allows us to resolve small changes of reflectance well below 1% when sample dimensions do not exceed $3.5 \times 3.5 \text{ mm}^2$. Within the signal-to-noise ratio of our experiment we find no temperature dependence between the $R_s$ spectra taken at $T = 340$ mK and at $T = 2$ K; such changes are not expected for a superconductor with $T_c$ as high as 7.1 K. The frequency dependence of $R_s(T = 340 \text{ mK})/R_s(T = 8 \text{ K})$ is peaked around 800 GHz which agrees well with the magnitude of the energy gap $2\Delta(0)$ in bulk lead. The general form of the $R_s(T = 340 \text{ mK})/R_s(T = 8 \text{ K})$ spectrum is in fair agreement with the theoretical dependence shown in the middle frame of Fig. 8. We find a somewhat sharper increase of the experimental $R_s/R_n$ close to the gap energy than in the theoretical dependence. Changes of the reflectance between 340 mK and 8 K are typically about 1% for a fairly disordered film. This latter value corresponds to the resistivity of the film about $\rho = 50 \mu\Omega \text{ cm}$ which is characteristic for thick films grown at low temperatures. We also investigated the $R_s/R_n$ ratios as a function of temperature. Selected spectra are displayed in the top panel of Fig. 8. We observe systematic decrease of the gap energy at finite $T$ that is in good agreement with the BCS theory of superconductivity (inset of Fig. 8).

In addition to reflectance measurements we have carried out transmission studies of ultrathin Nb films on Si substrate. These films were grown at UIUC and had thickness of about 7.5 nm. The superconducting transition is suppressed in these ultrathin films down to 6 K. For the purpose of these measurements we assembled the optical layout depicted in Fig. 9 permitting focusing THz radiation on the sample. In Fig. 10 we show the ratios of transmission measured at different
temperatures in the superconducting state $T_s(\omega)$ to the data collected in the normal state at 8 K $T_n(\omega)$. Marked changes of transmission are related to the opening of the energy gap. The $T_s/T_n$ spectra are peaked at frequencies close to the magnitude of $\Delta$. This is also illustrated in the bottom frame of Fig. 10 where we show the $T_s/T_n$ ratio calculated within the framework of Mattis–Bardeen model adequately describing electrodynamics of elemental superconductors in the dirty limit. Theoretical results are depicted for a free-standing film (gray line) and for a film on a Si substrate (black line). The latter result reveals a periodic pattern due to interference in the substrate. The value of the energy gap extracted from the fit is $\Delta(5340 \text{ mK}) = 586 \text{ GHz (17 cm}^{-1})$. This result corresponds to $2\Delta(0)/kT_c = 4.07$: a well established deviation from the weak-coupling predictions of the BCS theory: $2\Delta(0)/kT_c = 3.53$. Interestingly, the $2\Delta(0)/kT_c$ ratio in our ultrathin films does not seem to be affected by changes of $T_c$ whereas the transition temperature itself is strongly reduced for film thickness below 90 Å. Recently, Pronin et al. have obtained $2\Delta(T=4.5 \text{ K}) = 24 \text{ cm}^{-1}$ for a thicker 150 Å film with $T_c = 8.3 \text{ K}$ prepared using similar conditions. Their data corresponds to $2\Delta/kT_c$ close to 4.1 that is in excellent agreement with our findings. With increasing temperature the strength of the peak in the $T_s/T_n$ data is reduced and the frequency position is shifted to lower energies. Both effects are in accord with the BCS theory.

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