

Magneto optics and time resolved terahertz spectrocopy

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TOPICAL REVIEW — Synergetic Extreme Condition User Facility: Breakthroughs and opportunities for the research of physical science

Magneto optics and time resolved terahertz spectrocopy*

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Exploring, manipulating, and understanding new exotic quantum phenomena in condensed-matter systems have generated great interest in the scientific community. Static and time resolved optical spectroscopies after photoexcitations are important experimental tools for probing charge dynamics and quasiparticle excitations in quantum materials. In Synergetic Extreme Condition User Facility (SECUF), we shall construct magneto-infrared and terahertz measurement systems and develop a number of ultrafast femtosecond laser based systems, including intense near to mid-infrared pump terahertz probe. In this article, we shall describe several systems to be constructed and developed in the facilities, then present some examples explaining the application of magneto optics and time resolved spectroscopy techniques.

Keywords: magneto-optics, infrared spectroscopy, mid-infrared pump terahertz probe

PACS: 75.78.Jp, 78.47.J–, 75.80.+q, 78.30.–j **DOI:** 10.1088/1674-1056/27/7/077501

1. Introduction

Optical spectroscopy is a primary experimental tool for probing single particle and collective excitations of solids over a wide range of energies. The physical quantities extracted from optical probes are so-called optical constants, such as complex reflective index, real part and imaginary part of dielectric function, real part and imaginary part of optical conductivity, etc., which in general are frequency and temperature dependent. Those optical constants can be measured either directly by using, for example, ellipsometry spectroscopy or time domain terahertz (THz) spectroscopy, or indirectly by performing reflectance spectrum measurement over a broad energy scale using a Fourier transform infrared spectrometer (FTIR) and subsequent Kramers–Kronig transformation. The low lying excitations in the far-infrared or terahertz region are of particular importance since the electronic properties of quantum materials are determined mainly by the low lying charge and magnetic excitations. Most elementary excitations of solids occur in such energy scales, including free carrier contribution in metallic compounds, the lattice vibrations (phonons), collective spin wave excitations in magnets (magnons), binding energy of electron–hole pairs (excitons), pairing energy gaps and Josephson plasmons in high temperature superconductors, amplitude modes or phase modes in charge density wave compounds, etc. (see Fig. 1).[1] Magneto-optics is a combined experimental technique of optical spectroscopy and magnetic field. Magnetic field can have substantial effect on quantum materials by, e.g., inducing a change of the band structure of a metal or semimetal (e.g., formation of Landau levels), driving a quantum phase transition, altering the ground states in frustrated magnets, revealing hidden orders or inducing new phases in magnetic or charge ordered materials. Therefore, a combination of magnetic field and optical probe enables us to uncover novel physical phenomena and provides further insight into the nature of unsolved physical problems.

In the last two decades, much progress has been made in the development of ultrashort and ultrafast femtosecond laser and its applications. The time resolved spectroscopy based on the standard pump probe geometry provides another route to study the cooperative phenomena in condensed matter physics. Since the relaxation time of lattice, spin, and electrons is different after excitation by pumping pulses, one may resolve the different contributions to the exotic ground state through measuring the time evolution of reflectivity, polarization, or optical conductivity. More intriguingly, when the electric field of the pumping pulse is strong enough and comparable to the potential gradients of atoms in investigated systems, one may access to the non-perturbation regime where the intense pulse can induce strong nonlinear response, or even drive a system to certain metastable state or new exotic phase which are not present or cannot be achieved in the absence of excitations.

In Synergetic Extreme Condition User Facility (SECUF), we plan to construct magneto-infrared measurement systems and develop a number of ultrafast femtosecond laser based systems, including time domain terahertz spectroscopy

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(TDTS) under magnetic field and time resolved THz system with pump excitations ranging from THz to near infrared regime. In this article, we shall describe several systems to be constructed and developed in SECUF, then present several examples explaining the application of magneto optics and time resolved spectroscopy techniques. Finally, we make some perspective remarks.

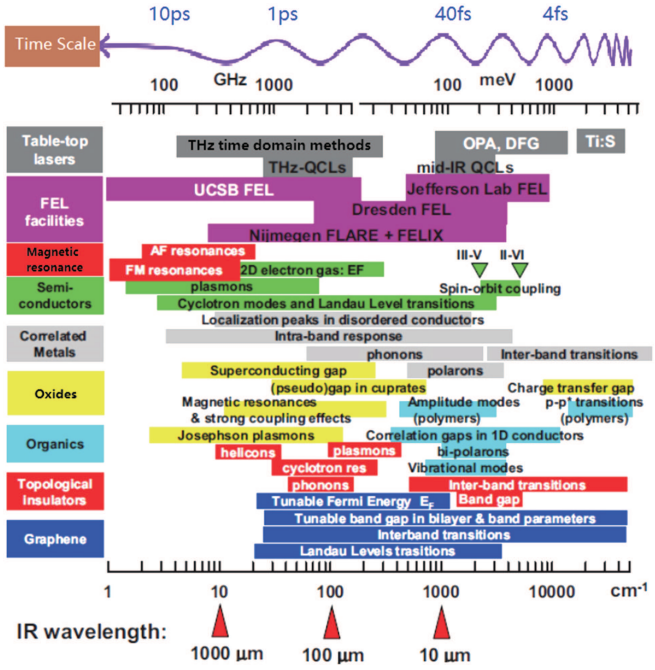


Fig. 1. (color online) Characteristic energy scales of excitations in solids. The energy range of table-top lasers, low energy free electron laser (FEL) facilities, and a few measurement methods are added to a figure published in Ref. [1]. A few abbreviations: DFG, difference frequency generation; OPA, optical parametric amplifier; QCL, quantum cascade laser.

2. Infrared and terahertz facilities in SECUF

2.1. Magneto-infrared measurement system

The magneto-infrared measurement system in SECUF is mainly composed of FTIR spectrometers and superconducting magnets. The bolometer detectors working at liquid helium temperature are used for far- to mid-infrared measurement. This system has two types of superconducting magnets: cylinder-coil magnet and split-coil magnet. Using the cylinder-coil magnet, one can perform magneto-infrared measurements under magnetic fields up to 20 T or even above. There are two frequently used configurations for the magneto-infrared measurements: Faraday and Voigt configurations. In Faraday configuration, the magnetic field is applied along the wave vector of the incident light, while in Voigt configuration, the magnetic field is perpendicular to the wave vector of the incident light. With cylinder-coil magnet, it is easy to realize the Faraday configuration (see Fig. 2).

However, due to the limited diameter ($\sim$ 50 mm) of the cylinder-coil magnet, it is quite difficult to perform magneto-infrared measurements in Voigt configuration. The split-coil magnet has four windows. The magnetic field is along the two windows. Therefore, Voigt configuration may be achieved in the split-coil magnet by inserting the samples into the magnet along the other two windows. By using a side-loading helium-flow cryostat, it will be convenient to change the temperature (from $\sim$ 4 K to 300 K) of the samples. The highest magnetic field of commercially available split-coil may reach 12–14 T, which is lower than the cylinder-coil magnet, however it is more convenient to measure optical spectrum over broad energy scale since the detectors being placed at the end of the optical path (outside of the magnet) can be easily switched or changed. Figure 3 shows a schematic drawing of the magneto infrared system in a split-coil magnet.

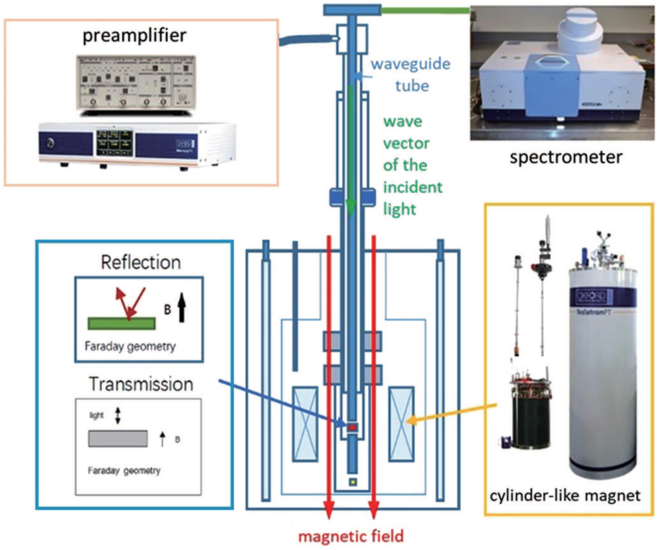


Fig. 2. (color online) A schematic diagram of magneto-infrared spectrometer system in a cylinder magnet.

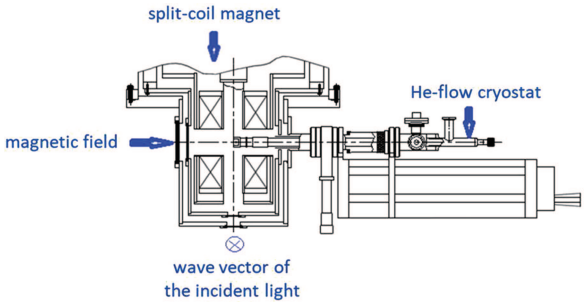


Fig. 3. (color online) A schematic diagram of magneto-infrared spectrometer system in a split-coil magnet.

2.2. Time resolved optical spectroscopy

There are multiple techniques for developing time domain and time resolved optical measurement systems. The time resolved magneto-optics systems contain two separate spectrometers: time domain terahertz spectrometer, and pump probe spectrometer under intermediate and high magnetic fields with multiple terahertz sources. There are different components for the time-resolved systems which we shall describe in detail below.

2.2.1. Laser sources

Ultrafast pulsed lasers play a key role in constructing time domain or time resolved optical systems. We have two goals in mind, one is to use weak laser pulses to construct high signal-to-noise time domain terahertz spectrometer and pump probe spectrometer, both under magnetic field. The other goal is to access strong field regime to manipulate or control the ground state of quantum materials, in which photo-induced phase transition or metastable state can be achieved and investigated. To this end, it is necessary to generate a strong electric and magnetic field of pumping radiation and to tune the pump beam wavelength to be resonant with certain modes of investigated systems. For the equilibrium or weak pump probe systems under magnetic field, in order to obtain high quality data we shall use high repetition rate 100 kHz laser system and a pulse duration of 10 fs, which can provide enough bandwidth and time resolution to resolve the intrinsic time evolution of measured optical constant. For the realization of photo induced phase transition or metastable state, we shall employ 1 kHz amplified laser system, which can generate high intensity and carrier envelope phase (CEP) stable THz and mid infrared radiations.

1 kHz high intensity laser system In order to explore intense excitation induced phenomena, ultrashort pulses are crucially required. The ultrashort pulses also provide the possibility to resolve CEP stable high frequency mid infrared radiations via phase matched electric-optics sampling method.[2] The seed pulse of the 1 kHz laser system is from a laser oscillator with a repetition rate of 80 MHz and a pulse duration of 10 fs, which provides 100 nm bandwidth at the central wavelength 800 nm. The pulse energy of such oscillator can exceed 12 nJ. About 40% of the total output is used as an ultrashort probe or gating pulse in the high intensity system. The rest is divided into two equal parts to seed two synchronized regenerative 1 kHz amplifiers (SPFIRE ACE) to ensure that the two optical parametric amplifiers (OPAs) and the difference frequency generation (DFG) based THz and mid infrared radiations are clocked to the seeding MHz oscillator. This amplifier produces output pulse with more than 7 mJ pulse energy with a repetition rate of 1 kHz and a pulse duration of about 35 fs at a cental wavelength 800 nm. The generation of strong intensity mid infrared and Thz radiation by DFG requires two high intensity NIR pulses with slight offset spectra.[3] Theoretically, the offset in the central frequency corresponds to the desired terahertz central frequency $\nu_{\mathrm{THz}} = \nu_{\mathrm{A}} - \nu_{\mathrm{B}}$. Consequently, in order to cover broadband THz range and keep such sources flexible tuning in frequency space, it is very important to keep the central frequencies ν_{A} and ν_{B} independently tunable. Two independent NIR OPAs synchronized to the seed oscillator with two independent outputs (TOPAS-twins) satisfy these requirements. Both outputs share the same white light source and therefore enable to provide excellent and bound up stability. Shared white light also guarantees to generate CEP stable THz and mid infrared radiations.

100 kHz high repetition laser system For the ultrafast systems under magnetic field, the optical components/beams are prone to be affected by the magnetic field. In order to improve the quality of collected data, we shall employ a high repetition rate laser system. Here, a relatively high repetition rate together with high photon fluxes can reduce processing time and increase the signal-to-noise ratio, while the above described 1 kHz laser system is for intense and high field experiments. A pulsed laser system with ultrashort light pulse down to 10 fs, up to 100 μJ pulse energy at 100 kHz repetition rate would be ideal for the purpose. In such laser system, fast scan technique combined with acoustic-optics modulation technique can further improve the signal-to-noise ratio.

2.2.2. Time domain and time resolved spectrometers

TDTS under magnetic field Time domain magneto-THz spectrometer will be constructed with sample locating in a helium cryostat under moderate magnetic field of 7–10 T (split coil) from Quantum Design or Oxford spectromag. The schematic diagram of the spectrometer is illustrated in Fig. 4. The probing terahertz radiation is generated by a large area GaAs photoconductive antenna and detected by 0.3 mm GaP crystal via free space electro-optics sampling. The femtosecond laser delivered from the mode locked Ti:sappire (center wavelength of 800 nm, pulse duration of 10 fs, and 100 kHz repetition rate) is used as the pumping source. The linear polarized terahertz light is focused on the sample by 90° off axis parabolic mirrors. The setup will be placed within the box filled with dry nitrogen to eliminate THz absorption by water in the atmosphere. In a typical TDTS experiment, the electric fields of a THz pulse pass through a sample and the same size aperture used as a reference are recorded as a function of delay time. Fourier transformation of the recorded time traces provides the frequency dependent complex transmission spectra including magnitude and phase information, from which the complex optical constant in the measured range can be derived without Kramers–Kroning transformation. The available THz speatra in this configuration will cover from 0.2 THz to 4 THz after compensating the chirp in the optical path. For the magnetic field dependent measurement in Faraday (light $k \parallel H_{\mathrm{DC}}$) or Voigt geometry (light $k \perp H_{\mathrm{DC}}$), the superconducting magnet can be mounted on a rotating stage, enabling rotation of the magnetic field by 90° relative to the terahertz propagating direction conveniently. Furthermore, combing the polarizator at THz range with magnet can offer Faraday rotation measurement in transmission geometry.

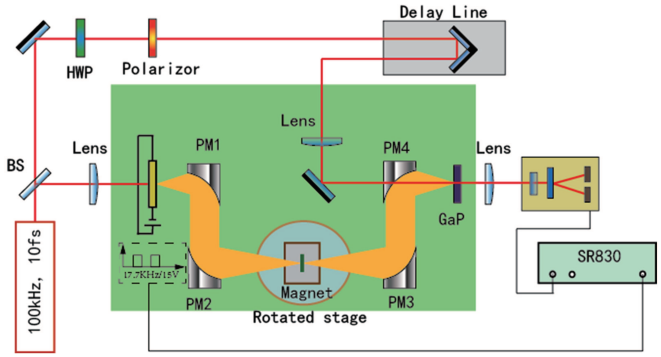


Fig. 4. (color online) A diagram of a typical time domain terahertz spectrometer.

Optical pump optical probe under magnetic field High resolution pump probe system under moderate magnetic field will be constructed. Figure 5 shows a typical reflectivity geometry optical pump optical probe set-up. The aforementioned high repetition rate laser delivers 10 fs short pulses which split into a high fluence pump and a low power probe beam with power intensity ratio of 10:1 after passing through a beam splitter. The probe beam is guided via mirrors onto a step motor delay line. A reflector is mounted on this motor. By varying the optical path with respect to the pump path, the time delay between the pump and probe beam arrival on the sample can be controlled. We employ a long moving delay line in order to cover long relaxation time. Following the step motor, the polarization of the probing pulse is rotated 90° by a half waveplate. Since the laser has a very short pulse duration in time, it provides a bandwidth 100 nm in frequency. Different frequency components in the ultrashort pulses have different velocities when the pulses pass through the optics. Therefore the pulse will spread in time as it passes through the optics. In the planed set-up, the pumping pulse will travel through an acoustic-optics modulator (AOM), which is a 10 mm thick TiO_2 crystal. This modulator will induce additional positive chirp. In order to achieve the maximum time resolution at the sample position, we will use two prisms to induce negative group velocity dispersion (GVD) to compensate the GVD in our pumping path. After the GVD compensator, the pumping beam is guided vis mirrors on to an oscillation delay line. When the required delay time is shorter than 150 ps, we may use a vibration delay line to vary the time delay between the pumping and probing beams. The frequency of AOM is 50 kHz, which is synchronized with the laser. After this, the pumping pulse focused by an achromatic lens will be directed to the sample sitting at the center of the magnet. The lens will be mounted on a non-magnetic XYZ manual translational stage, through which we can adjust the overlap of the pump and probe beams conveniently. There is also an achromatic lens used to focus the probe beam on to the sample. Reflected (transmitted) probe from sample is picked up by a mirror and sent to a photo-detector. To reduce the noise from scattering light of strong pumping pulse, we cross-polarize pump and probe beams, and use a polarizer before the detector aligned to pass the probe beam and block the pump beam. The output of the detector is sent to a lock-in amplifier (SR844), where the modulated frequency provided by the inner circuit of the amplifier is used for reference. Combining the modulation technique and oscillation delay line, we can improve the signal-to-noise ratio tremendously compared to the standard step motor. The magnet used in this spectrometer can be the same as the static terahetz spectrometer. Since the magnet is mounted on the rotating stage, we can measure the ultrafast relaxation evolution after excitation in Faraday and Viogt geometry conveniently.

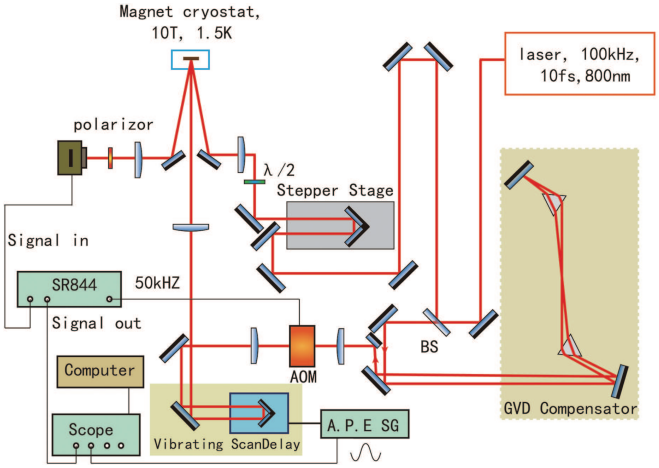


Fig. 5. (color online) A diagram of the optical pump probe spectrometer under magnetic field; $\lambda/2$: half waveplate; BS: beam splitter; SR844: radio frequency lock-in amplifier; AOM: acoustic-optics modulator.

Time resolved spectrometer As we mentioned above, a weak pump pulse would excite electrons or quasiparticles from occupied state to unoccupied state. After excitation, the excited quasiparticles would decay to the equilibrium state and the relaxation dynamics can be learned through detecting the evolution of reflectivity with delay time. However, high intensity pulses may lead to completely different effects as they may re-construct the free energy landscape, enhance or suppress the competing orders, and generate rapid switching from one state to another in a complex quantum material. Under certain circumstances, when the energy scale of the pumping pulse is close to certain modes of the investigated system, e.g., phonon or other collective modes, the effect of hot electrons relaxation process may be negligibly small, the major effect is the switching between different states. Such photo excitation induced metastable state and phase transition represent exciting new frontiers in ultrafast spectroscopy.

In the time resolved spectrometer, we shall develop intense terahertz/mid-infrared/near-infrared pump terahertz probe system. The energy of the pump beam could be tuned from near infrared down to terahertz by different techniques,

covering a frequency range from 0.4 THz to 60 THz as shown in Fig. 6. The first beam is dedicated to generate high field waveforms at a center frequency of near 0.4–1 THz. For this purpose, an intense terahertz source based on tilted pulse front optical rectification of $LiNbO_3$ crystal will be developed,[4] which will provide pulse energy of the order 2 μJ and the peak electric field amplitude about 0.5–1 MV/cm. In order to further enhance the electric field and increase the bandwidth of strong intensity THz, a cryostat equipped with optical windows will be designed and manufactured to cool the $LiNbO_3$ crystal to liquid nitrogen temperature.[5] The beam II is a broadband source ranging from 20 THz to 60 THz. The multi-THz source is based on DFG between two spectrally decoupled near infrared pulses delivered by aforementioned two parallel optical parametric amplifiers. The schematic diagram of DFG is shown in Fig. 6(c). After rotating the polarization of the short wavelength signal beam to fulfil the requirement of type II phase matching in the DFG process in GaSe crystal, both signal pulse trains from the OPA are superimposed collinearly by a polarization beam splitter cube and temporally overlapped by a mechanical manual stage. In order to enhance the fluence hit on the nonlinear optic crystal, the long focus length lens or lens telescope will be used to reduce the beam diameter at the crystal. Following the crystal, a germanium wafer is mounted on the optical path to block the remaining pumping light and purify the THz source. After that, the pure high field THz radiation can be guided to experimental setup.[6] Ultrashort gating pulses delivered form oscillator are superimposed with the field transients for the phase matched electric optic sampling. For the beam III, the basic geometry is identical to the beam II, both of them are based on DFG process. However, in the near infrared range provided by our dual OPA, the optical rectification will compete with the DFG process, since the near infrared pulse located at 1200–1600 nm also satisfies phase matching condition in organic nonlinear optics crystal OH1 or DAST. In order to suppress the optical rectification process and realize the efficient DFG, we will use the two pairs transmission grating to chirp both of our signal pulses.[7] After the crystal, the generated THz radiation travels through low pass filters to block the remaining near infrared pulses. The similar EOS setup will be used to characterize the THz radiation.

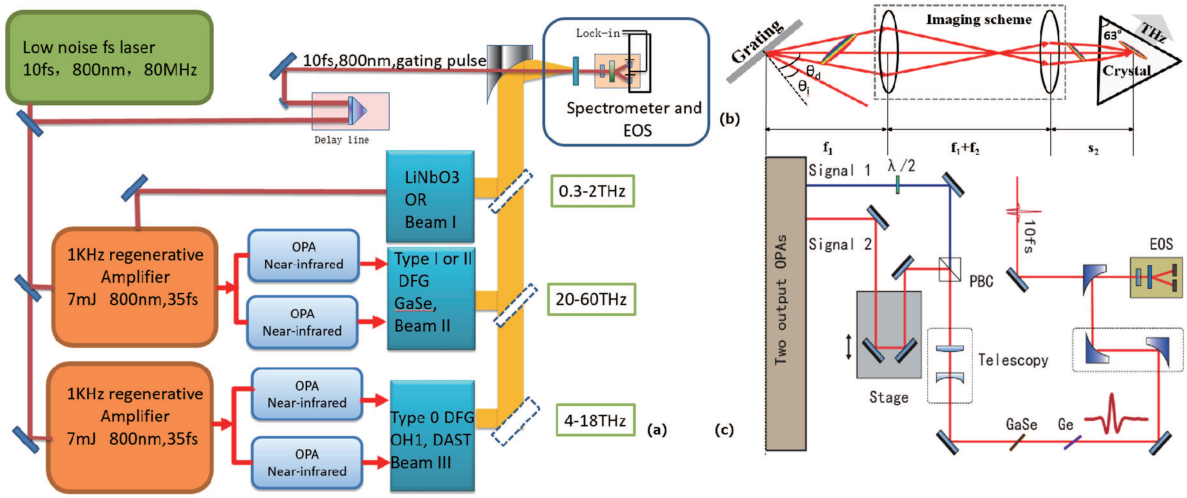


Fig. 6. (color online) (a) Schematic setup of the multi-dimensional THz source with a ultrashort oscillator (10 fs), two high-power Ti:sapphire amplifiers (35 fs), two dual optical parametrical amplifiers (OPA), difference frequency stages (DFG), and a setup for electro-optic sampling (EOS). The accessible frequency ranges are indicated next to the THz-generation stages. (b) Schematic setup of tilted pulse front optical rectification method. (c) Schematic setup of collinear DFG on GaSe crystal.

3. Applications of magneto infrared and time resolved terahertz spectrocopy

We now present some examples illustrating the application of magneto optics and time resolved terahertz spectrocopy.

3.1. Equilibrium properties under magnetic field

3.1.1. Landau level formation and Faraday rotation at low frequency in compounds with linear band dispersions

From the 1970s, the two-dimensional electron gas (2DEG) from semiconductor hetero-structures was at the focus of the magneto-optical spectroscopy research. The dynamics of a simple two-dimensional metal in the terahertz range can be well understood by considering a system of free electrons in the presence of an out-of-plane applied magnetic field. When the magnetic field is strong enough to drive the free carriers to a quantum limit, the charged particle will undergo cyclotron motion in the plane perpendicular to the magnetic field. The cyclotron moving carriers can resonantly absorb light at the frequency $\omega_c = eB/m^*$, known as a cyclotron resonance. For the conventional 2DEG, the typical energy scale of cyclotron resonance is located in the terahertz range for a magnetic field ~ 10 T. In the recorded time trace of terahertz

pulses, the resonant absorption manifests as an oscillation in time domain. The damping of oscillation is directly linked to the scattering rate of carriers.[8]

In recent years, new quantum materials such as graphene, topological insulators, Dirac or Weyl semimetals have attracted more attention. One of the most remarkable characteristics of these materials is the presence of linear band dispersion. A striking property associated with the linear dispersion is the unequal spacing between Landau levels (LLs) formed under applied magnetic field. Indeed, $\sqrt{B}$ dependence Landau levels were observed in these materials.[9–12] Such behavior is different from the conventional 2D semiconductor electron gas, where the equal spacing is formed between different Landau levels.

As an example, figure 7 shows the results on the magneto-transmission measurements of graphene layers on single crystal SiC.[10] Four dip-like feautures can be observed in the relative transmission spectra (i.e., $T(\omega,B)/T(\omega,B=0\,\mathrm{T})$). These dip-like features arise from the optical transitions from LL_n to $\mathrm{LL}_{n'}$, which obey the selection rule: $\Delta n = |n| - |n'| = \pm 1$ (see the inset of Fig. 7(a)). Herein, the dip-like features, (B) and (C), shift towards higher energies as the magnetic fields increase (Fig. 7(b)). The energies of the four dip-like features are plotted in Fig. 7(c) as a function of $\sqrt{B}$. The linear $\sqrt{B}$ dependence of the dip energies indicates the existence of massless Dirac fermions with cone-like band dispersions in graphene layers on single crystal SiC.[10] Presence of Dirac fermions was also found in other materials, for example, the parent compounds of iron based superconductors where the transitions between different Landau levels were also revealed in the magneto-infrared measurement.[13]

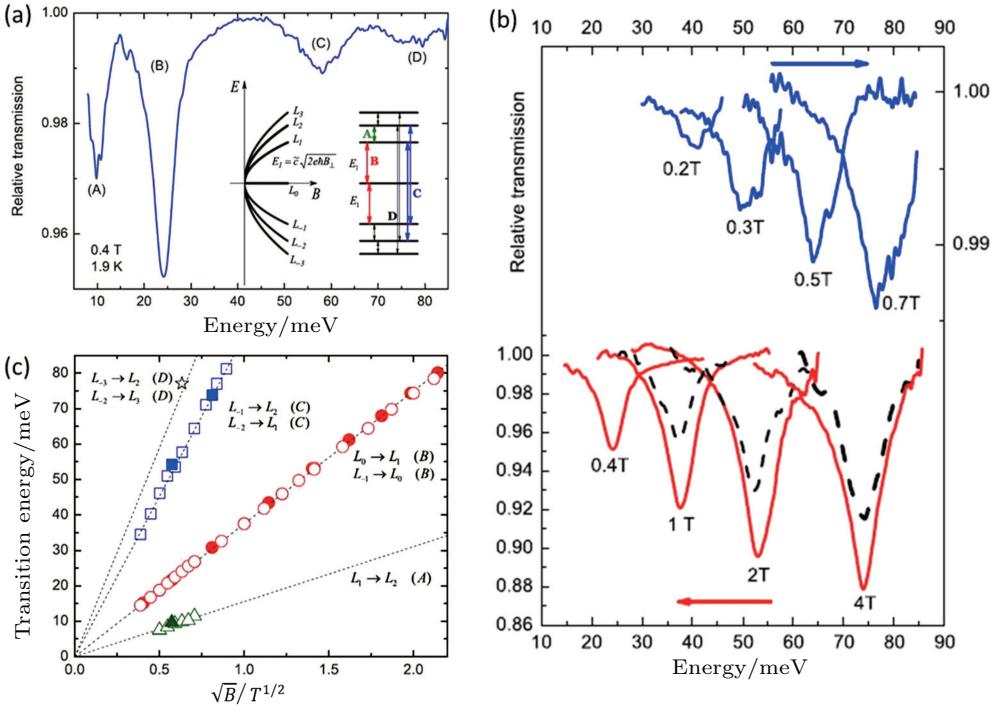


Fig. 7. (color online) Magneto-transmission measurements of graphene layers on single crystal SiC. (a) Relative transmission spectrum at 0.4 T. The inset shows the LL transitions giving arise to the dip-like features in the spectrum. (b) Dip-like features (B) and (C) at different magnetic fields B. (c) Dip energies as a function of $\sqrt{B}$ in (a).[10]

Besides the resonant absorption arising from optical transition between different Landau levels, the giant Faraday rotation at low frequency is another striking property of materials with the presence of linear dispersion, in which the polarization of a linearly polarized beam rotates when transmitted through a material under magnetic field. The Faraday rotation is related to the fine-structure constant. However, the quantitative relation with fine structure constant is different between 2DEG, graphene, and 3D topological insulator.[14,15] Using time domain magneto-spectroscopy, which allows for a direct determination of the frequency dependent complex Kerr angle (Kerr rotation and ellipticity) in THz spectral range, a number of novel phenomena were revealed and discovered, including a giant (10^6 rad $\cdot$ T^{-1} $\cdot$ m^{-1}) and quantized THz Faraday rotation in HgTe quantum wells,[16,17] graphene,[18] and intrinsic topological insulator thin film,[19] etc.

3.1.2. Quantum phase transition in magnetic insulators

For the magnetic insulators, it is often assumed that the charge degree of freedom is frozen by the strong Coulomb interaction below the charge gap. Only phonon and collective spin excitations contribute to the low energy absorption. In general, external magnetic field can easily couple to spin moments through Zeeman effect and lift the degeneracy of spin excitation or modify the ground state. Magnto-THz spec-

trometer is an standard method to probe magnon excitations at the Brillouin zone center.[20] The technique is particularly useful in the study of multiferroic materials, since the electromagnons and magnons arising from the electric and magnetic active spin wave excitations are rather rich and different configurations between propagating vector and magnetic direction result in different optical responses, so-called optical nonreciprocal directional dichroism. Those excitations locate in the terahertz range and have been widely probed by the magneto-THz measurement techniques.[21–24]

Magneto-THz and infrared have also been used to study magnetic excitations in other quantum magnets, for example, the one-dimensional (1D) Ising-like antiferromagnetic chain compound $SrCo_2V_2O_8$.[25] Theoretically, spin one half chain under transverse magnetic field is a prominent example for studying quantum phase transition. At zero field, the THz spectroscopy measurement revealed two series of excitations corresponding to acoustic and optical branches of confined spinons below the Neel temperature of 5 K in $SrCo_2V_2O_8$, which could be described by a one-dimensional Schrodinger equation with linear confining potential. By applying a small transverse field, the confinement of the optical and of the acoustic spinons is suppressed. At high transverse field (over 7 T), a quantum disordered phase is induced, in which three emergent fermionic excitations with different transverse-field dependencies were revealed by the terahertz spectroscopy.[25,26]

3.2. Photoexcitation induced metastable state or phase transitions

Ultrashort intense laser pulses, usually at 1 kHz repetition rate, have been proven as a powerful tool for light control of different orders in complex electronic materials. An intense laser pumping can drive a system to certain metastable state or induce a phase transition. In particular, by tuning the excitation energy to be resonant with some mode energies, e.g., phonons, magnons, many novel phenomena were induced and observed. Among others, the light-induced superconductivity in cuprates is perhaps the most intriguing and exciting observation. The first experimental realization was done on a specially doped cuprate $La_{1.675}Eu_{0.2}Sr_{0.125}CuO_4$, in which the 3D superconducting coherence is suppressed below 2 K due to the charge stripe order. However, by exciting the sample with a 16 μm mid infrared pulse at 10 K in the normal state, which is assumed to be resonant with the vibration of apical oxygen modes, Fausti *et al*. found that a new Josephson plasmon edge like shape emerged at the time delay of 5 ps after excitation, which was identified as a signature of light induced transient superconductivity,[27] as displayed in Fig. 8. It was the first demonstration that light could suppress the charge stripe order and induce superconducting coherence. The transient superconductivity was also found in underdoped $YBa_2Cu_3O_{6.5}$ at temperature even above 300 K.[28,29] More recently, it was found that the new Josephson mode could be induced by near-infrared (NIR) pump at 1.28 μm in $La_{1.885}Ba_{0.095}CuO_4$, in which the pumpling light could modulate the Josephson coupling strength along the *c*-axis.[30]

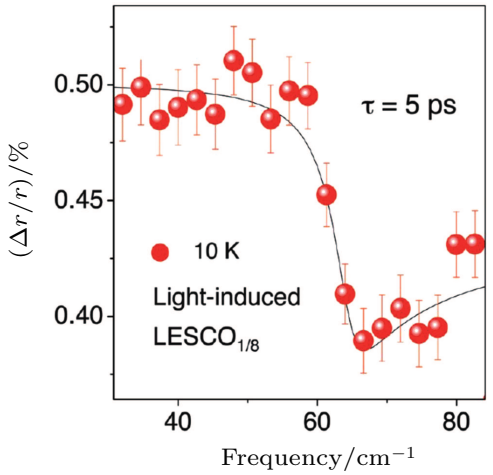


Fig. 8. (color online) Transient *c*-axis reflectance of $La_{1.675}Eu_{0.2}Sr_{0.125}CuO_4$, normalized to the static reflectance. Measurements are taken at 10 K, after excitation with IR pulses at 16 μm wavelength. The appearance of a plasma-like edge at 60 cm^{-1} suggests a photoinduced superconducting coherence.[27]

There are many other prominent progresses and development in the THz or mid-infrared pump THz probe spectroscopy. For example, the Higgs mode in a superconductor can be excited by taking the superconductor out of equilibrium by an ultrafast intense THz pump pulse and then detected by the THz probe spectroscopy as an oscillation in the time domain.[31] Over the past two decades, ultrafast magnetism has become increasingly popular and emerged as one of the most significant research branches in magnetism.[32] Magnetization dynamics has been studied intensively with pump probe magneto-optical spectroscopy under intermediate magnetic field, in which an ultrafast infrared laser pulse was usually used to excite the electrons in materials and subsequently the magnetization dynamics was traced by probe pulse.

4. Perspectives

We would like to make some remarks and perspectives finally. As we mentioned above, although optical spectroscopy covers a broad range of energy, the terahertz and far IR regions are of particular importance since the physical properties of a quantum material are determined by its low lying excitations. Furthermore, most of important single-particle and collective excitations in quantum matters locate in these regions. With the rapid advance of the ultrafast techniques, several of the world renowned high magnetic field user facilities, for example, the Dresden and the Nijmegen High magnetic field laboratory of the European Magnetic Field Laboratory (EMFL) have integrated low energy free electron lasers (FEL) to their

magnetic facilities. FEL provides very high intensity, quasi-monochromatic pulsed radiation. On the other hand, table-top laser based high intensity terahertz and mid infrared radiation sources have their own advantage. In Fig. 1, we added several FEL and table-top laser based terahertz and mid infrared sources and their frequency range on top of the energy scales of quasiparticle excitations of several quantum matter systems. By using DFG method and special optical band pass filter, it is now possible to access table top narrow band terahertz and mid infrared radiations without FEL facility, though with lower intensity. Comparing with FEL based sources, the pulse duration generated by DFG process is shorter when the central frequency is identical, which can provide more precise time resolution. The shorter pulse duration and their tunability will lead to wide range of new experiments hitherto impossible. We expect that the combination of different probes with high magnetic field will enable researchers to explore, control, and manipulate different quantum materials and quantum states in multiple extreme conditions.

References

[1] Basov D N, Averitt R D, van der Marel D, Dressel M and Haule K 2011 *Rev. Mod. Phys.* **83** 471
[2] Kübler C, Huber R, Tübel S and Leitenstorfer A 2004 *Appl. Phys. Lett.* **85** 3360
[3] Sell A, Leitenstorfer A and Huber R 2008 *Opt. Lett.* **33** 2767
[4] Hebling J, Almási G, Kozma I Z and Kuhl J 2002 *Opt. Express* **10** 1161
[5] Hirori H, Doi A, Blanchard F and Tanaka K 2011 *Appl. Phys. Lett.* **98** 091106
[6] Zhang S J, Wang Z X, Dong T and Wang N L 2017 *Front. Phys.* **12** 127802
[7] Liu B, Bromberger H, Cartella A, Gebert T, Först M and Cavalleri A 2017 *Opt. Lett.* **42** 129
[8] Ikebe Y, Morimoto T, Masutomi R, Okamoto T, Aoki H and Shimano R 2010 *Phys. Rev. Lett.* **104** 256802
[9] Gusynin V P and Sharapov S G 2006 *Phys. Rev. B* **73** 245411
[10] Sadowski M L, Martinez G, Potemski M, Berger C and de Heer W A 2006 *Phys. Rev. Lett.* **97** 266405
[11] Jiang Z, Henriksen E A, Tung L C, Wang Y J, Schwartz M E, Han M Y, Kim P and Stormer H L 2007 *Phys. Rev. Lett.* **98** 197403
[12] Chen R Y, Chen Z G, Song X Y, Schneeloch J A, Gu G D, Wang F and Wang N L 2015 *Phys. Rev. Lett.* **115** 176404
[13] Chen Z G, Wang L, Song Y, Lu X, Luo H, Zhang C, Dai P, Yin Z, Haule K and Kotliar G 2017 *Phys. Rev. Lett.* **119** 096401
[14] Maciejko J, Qi X L, Drew H D and Zhang S C 2010 *Phys. Rev. Lett.* **105** 166803
[15] Morimoto T, Hatsugai Y and Aoki H 2009 *Phys. Rev. Lett.* **103** 116803
[16] Shuvaev A M, Astakhov G V, Pimenov A, Brüne C, Buhmann H and Molenkamp L W 2011 *Phys. Rev. Lett.* **106** 107404
[17] Dziom V, Shuvaev A, Pimenov A, Astakhov G V, Ames C, Bendias K, Bottcher J, Tkachov G, Hankiewicz E M, Brune C, Buhmann H and Molenkamp L W 2017 *Nat Commun.* **8** 15197
[18] Shimano R, Yumoto G, Yoo J Y, Matsunaga R, Tanabe S, Hibino H, Morimoto T and Aoki H 2013 *Nat Commun.* **4** 1841
[19] Wu L, Salehi M, Koirala N, Moon J, Oh S and Armitage N P 2016 *Science* **354** 1124
[20] Yamaguchi K, Kurihara T, Minami Y, Nakajima M and Suemoto T 2013 *Phys. Rev. Lett.* **110** 137204
[21] Pimenov A, Mukhin A A, Ivanov V Y, Travkin V D, Balbashov A M and Loidl A 2006 *Nat. Phys.* **2** 97
[22] Sushkov A B, Aguilar R V, Park S, Cheong S W and Drew H D 2007 *Phys. Rev. Lett.* **98** 027202
[23] Cazayous M, Gallais Y, Sacuto A, de Sousa R, Lebeugle D and Colson D 2008 *Phys. Rev. Lett.* **101** 037601
[24] Kézsmárki I, Kida N, Murakawa H, Bordács S, Onose Y and Tokura Y 2011 *Phys. Rev. Lett.* **106** 057403
[25] Wang Z, Wu J, Xu S, Yang W, Wu C, Bera A K, Islam A T M N, Lake B, Kamenskyi D, Gogoi P, Engelkamp H, Wang N, Deisenhofer J and Loidl A 2016 *Phys. Rev. B* **94** 125130
[26] Wang Z, Wu J, Yang W, Bera A K, Kamenskyi D, Islam A, Xu S, Law J M, Lake B, Wu C and Loidl A 2018 *Nature* **554** 219
[27] Fausti D, Tobey R I, Dean N, Kaiser S, Dienst A, Hoffmann M C, Pyon S, Takayama T, Takagi H and Cavalleri A 2011 *Science* **331** 189
[28] Kaiser S, Hunt C R, Nicoletti D, Hu W, Gierz I, Liu H Y, Le Tacon M, Loew T, Haug D, Keimer B and Cavalleri A 2014 *Phys. Rev. B* **89** 184516
[29] Hu W, Kaiser S, Nicoletti D, Hunt C R, Gierz I, Hoffmann M C, Le Tacon M, Loew T, Keimer B and Cavalleri A 2014 *Nat. Mater.* **13** 705
[30] Zhang S J, Wang Z X, Shi L Y, Lin T, Zhang M Y, Gu G D, Dong T and Wang N L 2017 arXiv:1712.01174
[31] Matsunaga R, Hamada Y I, Makise K, Uzawa Y, Terai H, Wang Z and Shimano R 2013 *Phys. Rev. Lett.* **111** 057002
[32] Kirilyuk A, Kimel A V and Rasing T 2010 *Rev. Mod. Phys.* **82** 2731