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Coherent two-dimensional Fourier transform spectroscopy using a 25 Tesla resistive magnet

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We performed nonlinear optical two-dimensional Fourier transform spectroscopy measurements using an optical resistive high-field magnet on GaAs quantum wells. Magnetic fields up to 25 Tesla can be achieved using the split helix resistive magnet. Two-dimensional spectroscopy measurements based on the coherent four-wave mixing signal require phase stability. Therefore, these measurements are difficult to perform in environments prone to mechanical vibrations. Large resistive magnets use extensive quantities of cooling water, which causes mechanical vibrations, making two-dimensional Fourier transform spectroscopy very challenging. Here we report on the strategies we used to overcome these challenges and maintain the required phase-stability throughout the measurement. A self-contained portable platform was used to setup the experiments within the time frame provided by a user facility. Furthermore, this platform was floated above the optical table in order to isolate it from vibrations originating from the resistive magnet. Finally, we present two-dimensional Fourier transform spectra obtained from GaAs quantum wells at magnetic fields up to 25 Tesla and demonstrate the utility of this technique in providing important details, which are obscured in one dimensional spectroscopy.

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I. INTRODUCTION

Optical spectroscopy of semiconductors under high magnetic fields has been crucial in providing insights into the electronic structure of these materials. High magnetic fields can lift the degeneracy of critical points in the electronic structure and test theoretical predictions[1,2]. Furthermore, the magnetic field confines charged particles into circular orbits leading to interesting phenomena. In quantum wells, the additional confinement along the direction of the magnetic field leads to a two-dimensional topological insulator[3]. At low temperatures, a correlated system is formed with unique electronic transport properties such as the integer and fractional quantum Hall effect[4–6]. In the quantum Hall regime, light scattering and photoluminescence measurement have provided important insights into the physics of optical excitations at high magnetic fields[7–19]. The light scattering experiments have lead to the observation of magnetorotons and have revealed the intricate physics of composite fermions at different fractional filling factors[20–27].

In bulk GaAs, linear optical spectroscopy has revealed the emerging electronic structure of semiconductors under magnetic fields, such as evolution of the excitonic states and Zeeman splitting of the heavy hole excitons[30–36]. Such linear optical studies of bulk semiconductors and in heterostructures were complemented by coherent four-wave mixing (FWM) spectroscopy measurements based on the third-order nonlinear response of the material, which has provided important insights into the many-body interactions[37–47]. Coherent FWM spectroscopy is very suited to probe many-body interactions, because electron-phonon and exciton-exciton interactions lead to measurable changes of the dephasing[48]. Two-dimensional Fourier transform (2DFT) spectroscopy based on the coherent FWM signal has more recently emerged[49–54] and has been successfully applied to III-V semiconductors[55–63], diamond nitrogen-vacancy center[64], biological photosynthetic centers[65–70], peptides[52,71,72], and two-dimensional materials[73–75], providing important insights difficult to access using traditional time-integrated and spectrally resolved FWM spectroscopy. The advantages of 2DFT spectroscopy over traditional one-dimensional FWM have been well documented in the literature[51,76–82]. The correlated nature of the frequency axes can reveal underlying physics in the

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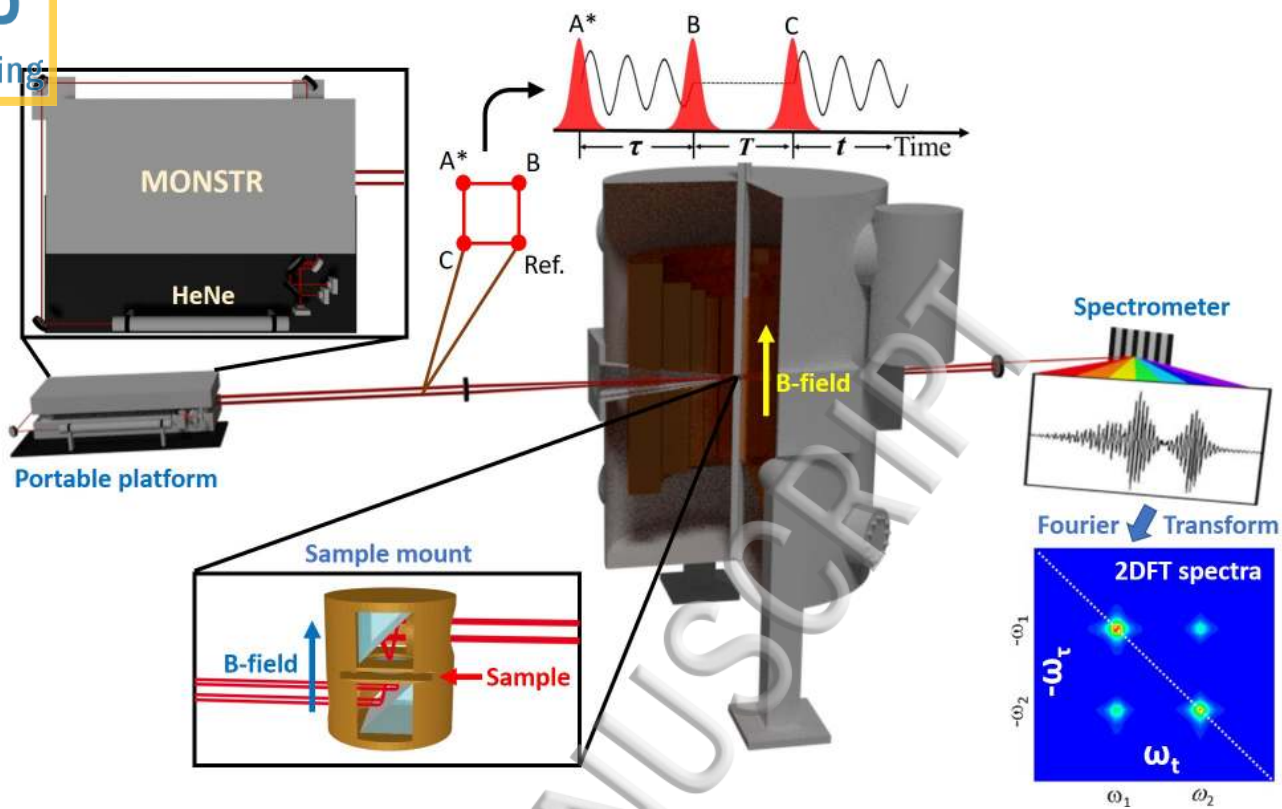

FIG. 1. Schematic of the experimental setup: The three laser beams are provided by the multidimensional optical nonlinear spectrometer (MONSTR)[28,29]. Three beams labeled as A*, B, and C, where A* corresponds to the phase conjugate beam, are separated by the time delays τ and T, and are used to generate the FWM signal. The beams are aligned in the three corners of a square. The FWM signal generated at the sample propagates along the missing corner (direction $-\vec{\mathbf{k}}_\mathbf{a}+\vec{\mathbf{k}}_\mathbf{b}+\vec{\mathbf{k}}_\mathbf{c}$) of the square and is heterodyne detected using the reference beam (Ref.). The combined FWM and Ref. beam is dispersed into a grating spectrometer and the resulting spectral interferogram is Fourier transformed leading to a two-dimensional spectrum. The samples are kept at 10 Kelvin inside the bore of the resistive 25 T split helix magnet. The magnetic fields up to 25 Tesla are applied perpendicular to the sample surface in Faraday geometry, facilitated by the sample mount.

form of two-dimensional line shapes and additional peaks in the 2DFT frequency spectra.

In 2DFT spectroscopy, two time delays are monitored simultaneously and the phase of an induced nonlinear signal is explicitly tracked. This leads to a two-dimensional time-domain data set, which is converted to a two-dimensional spectrum by a Fourier transform, analogous to the extension of nuclear magnetic resonance from one to two dimensions[51,76–82]. However, performing these measurements in the near infrared and visible spectral range has been difficult, due to the phase stability needed. Two primary methods have been used so far to achieve the desired subwavelength stability. These methods can include passive phase stabilization, such as construction of common paths through optical elements[58,59,83–85], using birefringent wedges to generate a phase-locked pair of pulses[86–90], or active phase stabilization using feedback loops to drastically reduce the effect of mechanical and thermal drifts[54,91,92]. A combination of both methods can also be implemented. The method used here is based on the multidimensional optical nonlinear spectrometer (MONSTR), which uses active phase stabilization[28,29]. The MONSTR instrument achieves phase-locking among all the three pulses used to generate the FWM signal, which enables all 2DFT techniques, 'rephasing' (S_I), 'non-rephasing' (S_{II}), and the 'two-quantum' (S_{III}) to be performed without any changes to the setup. Furthermore, time delays of hundreds of picoseconds can be easily achieved with the delay stages, enabling the measurement of long lived coherences[28].

In this manuscript, we present 2DFT measurements using the optical split helix resistive 25 Tesla magnet. We overcome two main technical challenges which are specific to 2DFT spectroscopy. First, such resistive magnets use water cooling which creates vibrations propagating from the floor to the optical table. Vibrations are very detrimental to 2DFT spectroscopy measurements, since they can lead to loss of the phase stability required for performing interferometric measurements. In order to overcome this issue, we placed the MONSTR instrument on a floating and actively stabilized platform. Second, 2DFT measurements involve sophisticated alignments that are difficult to perform in limited preparation time provided in a large user facility. Therefore, simplifying the setup and alignment is crucial. In order to expedite the alignment of the experiment, we have incorporated a new platform, which contains the MONSTR instrument and the metrology laser together, making the whole setup easily movable and quick to align.

Furthermore, we demonstrate proof-of-principle and present 2DFT spectra in GaAs at fields up to 25 Tesla. We measure the 2DFT spectra of a Landau level and the Zeeman splitting of the heavy hole exciton in the high field limit. The high magnetic field makes the components easily resolvable. This method can be used to explore the underlying electronic structure of novel materials, where the magnetic fields lift the degeneracy of energy levels and 2DFT spectroscopy is uniquely positioned to explore quantum coherence and relaxation pathways. Coherent time-resolved spectroscopy at high magnetic fields has been used in the past as a method to study the spin splitting of the exciton ground state in bulk GaAs and GaAs based heterostructures[93–98]. By measuring the exciton spin splitting band structure information can be obtained, providing important insights into the effect of light and heavy hole mixing on the spin structure of the valence band. Such Zeeman components are often obscured under the inhomogeneous broadening, which makes coherent spectroscopy and specifically the 2DFT technique particularly suited to disentangle such convoluted spectra. While the existence of multiple underlying components can appear as oscillations in the decay of time-integrated FWM, in the 2DFT spectra such components are resolved as separate diagonal peaks and their relaxation pattern is disentangled by the cross-peaks in the two-dimensional spectra. Magnetic fields up to 25 Tesla can further separate the Zeeman subcomponents that combined with the advantages of 2DFT spectroscopy can become an important spectroscopic tool.

Moreover, in modulation doped GaAs quantum wells, the ability to perform 2DFT measurements at these high fields can enable new 2DFT measurements at the very low fractional fillings, where the formation of a Wigner crystal can be achieved[99,100]. In this regime the 2DFT technique is expected to provide valuable insights into the many-body physics taking place. Such rich many-body physics can also be found in two-dimensional transition-metal dichalcogenides (TMDs), where strong Coulomb interactions lead to large exciton binding energies. In these materials, high magnetic fields would be desirable in order to separate the two spin polarized valleys, which shift very little with magnetic fields[101]. In the quantum Hall regime, devices based on two-dimensional TMDs show spin and valley polarized Landau levels[102]. The 2DFT experiments at magnetic fields up to 25 Tesla can lead to better separation between the Landau levels and provide insights into their quantum coherence[102,103].

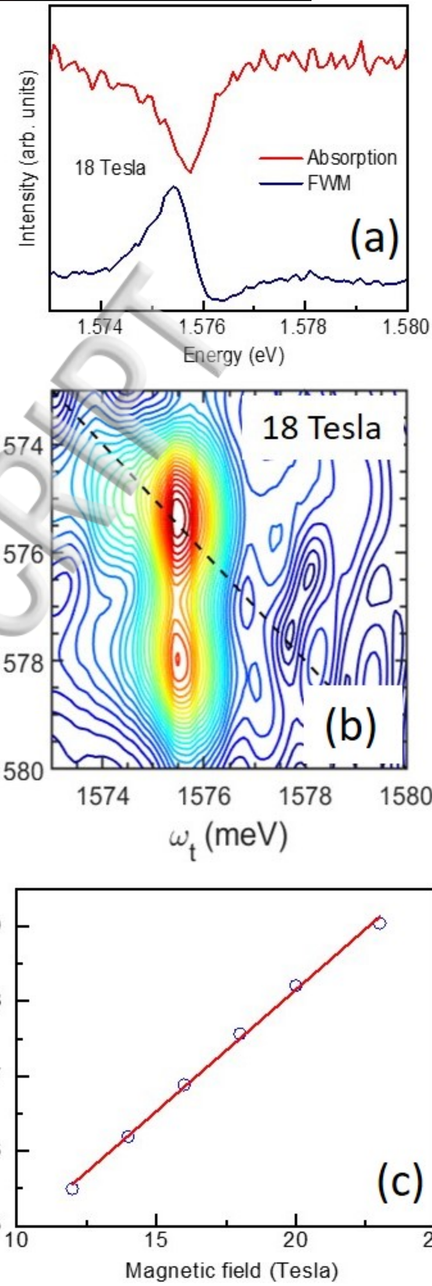

FIG. 2. Figure (a) shows the absorption and spectrally resolved FWM of a quantum well Landau level at 18 Tesla, corresponding to the 2DFT spectra shown in figure (b). In figure (c), the energy position of the optical transition is plotted as a function of the magnetic field, showing a linear energy shift with the field strength.

II. RESULTS

A. Instrument

Bitter magnets are resistive magnet designs that are capable of much higher magnetic fields than superconducting magnets. These magnets are constructed using an interlocking stack of helical copper disks that circulate large currents around a small central space where the sample is located. The magnet is usually several meters in length in order to create a uniform high magnetic field at the sample location, leading to a large distance from the outer windows[104]. Furthermore, stray fields require any instrument containing ferromagnetic components to be placed far away from the magnet. Thus, Bitter magnets usually require the use of optical fibers to couple light sources and detectors into and out of the magnetic field[105,106]. Fiber-coupled experiments are suitable for continuous wave visible and near-infrared studies.[107]. However, it is much more challenging for time-resolved experiments using femtosecond broadband pulsed lasers, because dispersion and higher order nonlinearities of optical fibers distort the laser pulse, making the data analysis challenging.

Pulsed magnet can reach the desired magnetic fields, but for a very limited amount of time in the order of milliseconds, not practical for 2DFT measurements. The Split-Florida Helix magnet system at the National High Magnetic Field Lab is capable of sustained operation at fields as high as 25 Tesla[105]. The electrical power consumption of this resistive magnet is 32MWh, but the ability to maintain a high magnetic field enables the 2DFT experiments. The Split-Helix has a 39 mm bore and uses a custom-constructed cryostat that can reach a base temperature of $\sim$5 K. The Magnet has two replaceable windows which are optically transparent in the desired optical range and allow for transmission experiments.

We start by discussing the experimental setup shown in Fig. 1. The resistive split helix magnet generates a magnetic field up to 25 Tesla and allows optical access through two windows, perpendicular to the direction of the magnetic field. The magnetic fields is generated in the vertical direction, perpendicular to the optical axis, which complicates experiments in the Faraday geometry. In this configuration, for the sample to face the optical windows, the magnetic field would be in the sample plane. In order to overcome this limitation, we designed the sample holder shown in Fig. 1. The sample is typically mounted on a quartz substrate, which is inserted in the slot. The first fixed mirror mounted at 45 degrees guides the focusing beams perpendicular to the surface of the sample. The second fixed mirror at 45 degrees reflects them out through the other optical window of the magnet. Thus, the sample holder allows optical access through one window and allows the beams to exit from the other window, facilitating transmission measurements. Using this sample mount, the magnetic field is applied perpendicular to the sample surface, whereas the optical axis runs parallel to the field direction, thus enabling measurements in the Faraday geometry.

The method used in implementing 2DFT spectroscopy here is the MONSTR apparatus, which employs active phase stabilization. The advantage of this approach is that the MONSTR already provides a stable and portable platform for achieving 2DFT measurements. The origi-

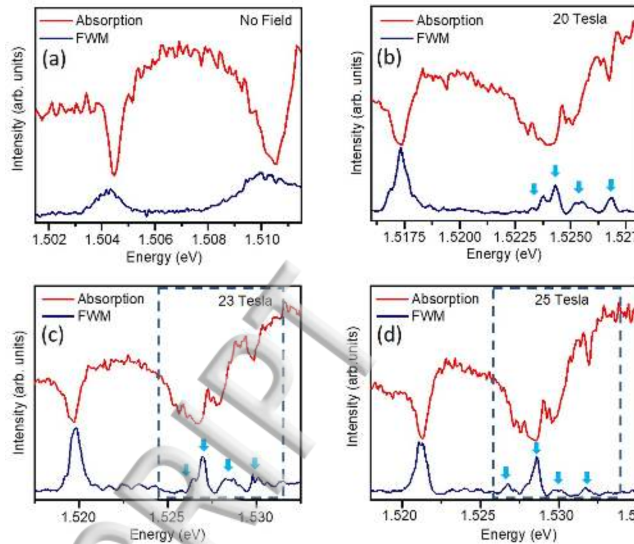

FIG. 3. Absorption and spectrally resolved FWM of the heavy and light hole exciton in the bulk. (a) At zero field, the heavy and light hole excitons are split by the external tensile strain due to mounting on the quartz substrate. (b-d) At fields of 20, 23, and 25 Tesla, a Zeeman splitting of the light hole exciton into four component is observed. The position of these subcomponents is marked by the blue arrows.

nal implementation was introduced in Ref. 28 and was further developed to use commercially available optical mounts and broad band optics for wider tunability[29]. In the present version, the high-speed feedback loop filters are also achieved using commercially available electronics (FPGA, National Instruments, NI PXI-7841R). The field-programmable gate array (FPGA) accomplishes fast data collection, diagnostics, and active stabilization of beam path lengths during the data acquisition routine. The setup presented replaces complicated analog circuits with digital programmable loops, and provides flexibility to perform a wide variety of time-resolved coherent spectroscopy experiments across a broad frequency range. Researchers in fields as diverse as gamma-ray radiation spectroscopy and quantum optics have implemented FPGA technology to enhance experimental design[108–110]. With the growing interest in FPGA-based designs in experimental physics, we chose this hardware for the MONSTR apparatus.

An important feature of the all-digital MONSTR apparatus is that the FPGA hardware system has been fully integrated into the existing, LabView based 2DFT control and measurement software. While the implementation of digital feedback technology does not significantly improve the locking stability of the delay lines over analog techniques, FPGAs offer many advantages over analog control including fast input/output times, specialized functionality, rapid prototyping, portability and durability. FPGAs offer convenience and flexibility inherent in the capability to realize any analog circuit by programming their logical algorithms as well as on-demand modification utility. Until recently, FPGA technology was only accessible to engineers with a thorough knowledge of digital hardware design. The LabView based integration significantly simplifies the programming of the FPGA functions, which makes this technology available to anyone with a basic knowledge of the LabView software. Beyond the low cost compared to analog electronics, this also allows for the FPGA-based control loop to be quickly modified for particular experimental situations.

The FPGA is integrated into a desktop computer in the Peripheral Component Interconnect (PCI) slot. The FPGA module (PXI-7841R) accommodates up to 8 analog inputs (sampling rates up to 200 kHz, 16-bit resolution, $\pm$10 V), 8 analog outputs (1 MHz, 16-bit resolution, $\pm$10 V), and 96 digital lines configurable as inputs, outputs, counters, or custom logic (40 MHz). These digital feedback loops replace the cumbersome analog electronics in the previous MONSTR instrument and provide other advantages in the experimental design, due to better streamlining of the data collection software.

Before applying the lock, the relative path lengths of the interferometer arms are left to drift over several minutes, leading to changes in phase of more than an optical fringe. The digital feedback apparatus is used to monitor and correct for drifts and fluctuations in the optical path lengths, which stabilizes the path lengths of the interferometer arms. The variations in the locked error signals are normal distributions with a standard deviation that corresponds to $\sim$2-3 nm of motion. Thus, the relative phases of the 800 nm optical pulses are stabilized to within $\sim \lambda/100$ under operational conditions. This stability is reliable on hours' time scales. The procedure for stepping the stages and performing the digitally implemented locking scheme is similar to Refs 28 and 29, with the exception that our setup implements a digital loop. This stabilization strategy demonstrates complete digital instrumentation, including data acquisition, moving delay stages, and active stabilization. The FPGA can be programmed using the LabView environment, with the exception that the code must be compiled and uploaded to the FPGA board, a process that can take up to ten minutes for the codes used in this experiment. For minor changes in the code, the compiling and uploading requires far less time. Multiple feedback loops can be run in parallel and monitoring of diagnostics can be performed and evaluated in real time.

As an example of the added functionality of a digital locking scheme, we explain an added feature that improves the long-term stability of our feedback loops. To ensure that we were locking our loops to the middle of an interferometric fringe, we programmed the FPGA to modulate the piezo through a sinusoidal wave of large amplitude at the beginning of each measurement. This procedure cycles the interferometer through many fringes for one second while simultaneously recording and processing the error signal. Recording this waveform allows the computer program to clearly pinpoint the exact center of the error signal by extracting the extrema of the variable waveform. The set point is designated at the average of these extrema, thus ensuring optimal sensitivity to fluctuations in the process variable, and reducing the likelihood that the system will drift out of the range the piezoelectric feedback actuators during the measurement.

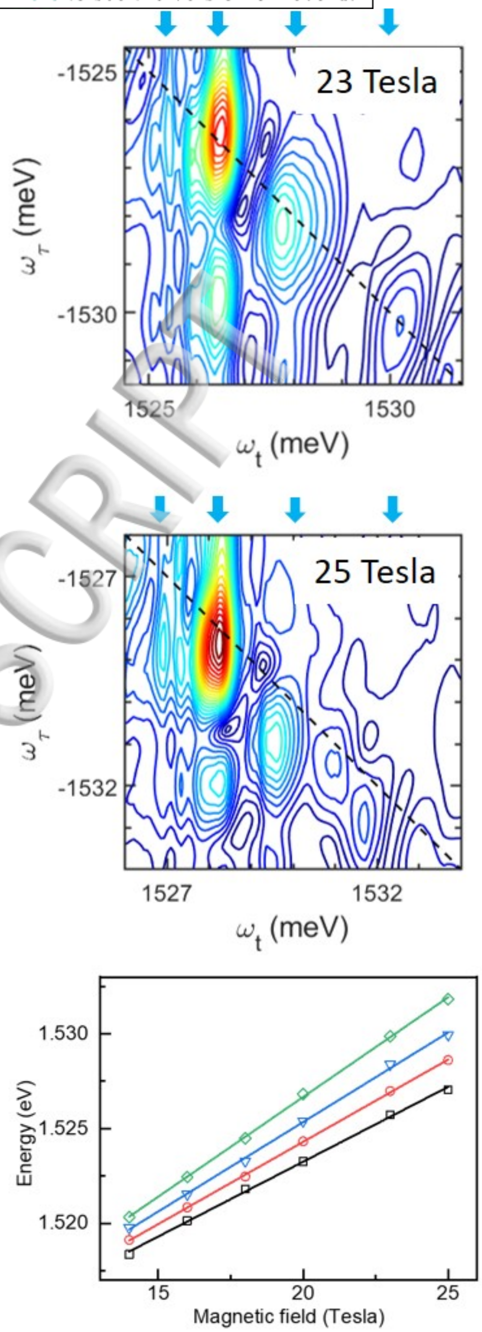

FIG. 4. 2DFT spectra of the light hole bulk exciton at 23 Tesla (top) and 25 Tesla (center). The spectral region is marked by the dashed blue box in Fig. 3. The Zeeman subcomponents are marked by the vertical blue arrows. (Bottom) Energy position of the four subcomponents as a function of magnetic fields.

Using the digital feedback loops and piezo-electric transducers, the MONSTR instrument can compensate for small mechanical and thermal drifts from the environment. However, large mechanical vibration cannot be compensated, therefore a somewhat quiet environment needs to be generated. In order to damp vibrations propagating from the ground to the optical table top, we placed the MONSTR on a floating platform. The floating platform consists of vibration isolation legs (Thorlabs, PWA090) and an aluminum breadboard (PBG11112). The isolating legs have been manufactured to effectively isolate the system between 3 − 50 Hz; a range in which many common vibrations seen in laboratories fall. The system is oil-free allowing for continuous peak performance without the need of maintenance once the system is setup. The optical breadboard is made completely from aluminum with a honeycomb core, allowing for the necessary rigidity while maintaining the portability of the system. The platform floating on pressurized air was itself mounted on the existing optical table, providing additional vibration damping. This isolation system reduced mechanical drifts to the level where they can be compensated by the MONSTRs active stabilization.

In order to achieve portability, the MONSTR setup was placed on a new platform that contained the metrology laser, schematically shown in Fig. 1. This configuration makes the alignment of the metrology laser very simple and reduces the alinement of the experiment to guiding the excitation laser beams toward the sample and focusing at the sample surface. Some additional alignment of the collection optics, guiding the heterodyned signal into the spectrometer slit, is also necessary. However, this approach significantly shortens and simplifies the alignment time.

B. Sample

The sample studied contains four 12 nm wide GaAs/AlGaAs quantum wells and is mounted on a quartz substrate. Residual bulk GaAs from the buffer layer is left over during the substrate removal process and is clearly observable in the absorption and spectrally resolved FWM spectra. The heavy and light hole degeneracy in bulk material is lifted by the compressive strain generated as a result of substrate removal and mounting the sample on the quartz substrate[111]. The separation between the heavy and light hole is ∼6 meV, corresponding to ∼ 0.2% compressive strain[112–114].

C. Measurement

We start our discussion with the nonlinear optical response of the quantum wells. The excited states of the excitons in the quantum wells are observable and in the high magnetic field limit behave like Landau levels, shifting linearly with magnetic fields[115–125]. We collected the 2DFT spectra of a Landau level originating from the quantum wells at 18 Tesla, shown in Fig. 2. The 2DFT spectra show an elongation along ω_τ frequency, observed previously at lower magnetic fields up to 10 Tesla[55]. The corresponding absorption and spectrally resolved FWM are shown in Fig. 2 (top), whereas in Fig. 2 (bottom) we plot the energy position of the Landau level as a function of magnetic field up to 25 Tesla. The energy position shows the expected linear shift with magnetic field of Landau levels.

We further explore the light and heavy hole excitons originating from the bulk buffer layer, which show fine structure. The degeneracy of the heavy and light hole excitons is lifted by the external strain induced by the differing thermal expansion of the sample and the quartz substrate. At zero magnetic fields shown in Fig 3 (a), the heavy hole exciton appears at a lower energy than the light hole exciton, indicating compressive strain[111–114]. As the magnetic field is increased an energy splitting of the light hole exciton is observed into four resolved components. This is due the large magnetic field lifting the degeneracy between the Zeeman levels of the light hole exciton, corresponding to the projections of the orbital angular momentum of the hole and the projections of the electron spin. Such Zeeman sublevels have been observed in the past using linear absorption spectroscopy at lower magnetic fields[30–36]. Different theoretical models were developed to describe the behavior of exciton in magnetic fields, distinguishing between the low field limit, where the exciton binding energy is larger than the magnetic cyclotron energy. The intermediate case occurs when the exciton binding energy is comparable to the cyclotron energy and finally, the high field limit, where the cyclotron energy is larger than the exciton binding energy[34–36]. Performing 2DFT spectroscopy on excitonic transitions using the split helix enables us to reach the high magnetic field limit in most semiconductors.

The high magnetic field absorption and spectrally resolved FWM data at 20, 23 and 25 Tesla are shown in Fig. 3 (b-d), respectively. The vertical blue arrows mark the positions of the resolved subcomponents of the light hole exciton. The dashed blue box in Fig. 3 (c-d) indicates the spectral region of the 2DFT spectra shown in Fig. 4 at 23 and 25 Tesla. The 2DFT spectra in Fig. 4 show peaks for the four excitonic subcomponents of the light hole along the diagonal (dashed black line) and their positions are marked by the vertical blue arrows. Furthermore, the 2DFT spectra show several off-diagonal cross peaks below the diagonal, which could indicate relaxation processes. Finally, we plot the energy position of the sublevels as a function of magnetic field up to 25 Tesla and observe a linear energy shift.

III. CONCLUSIONS

We perform nonlinear coherent 2DFT measurements using a resistive magnet generating fields up to 25 Tesla.

In order to perform these experiments we employ the all-digital MONSTR apparatus that uses the FPGA hardware system and is now streamlined with the LabView software into the data acquisition routine. These interferometric measurements require phase preservation and therefore are very sensitive to mechanical and thermal drifts, which the MONSTR instrument compensates via feedback loops and piezoelectric transducers. However, resistive magnets use large quantities of cooling water, thus generating strong mechanical vibrations. In order to overcome these challenges, we further developed the MONSTR instrument by adding an optical breadboard that contains the metrology laser and placing this assembly on a floating platform. The floating platform further dampens the mechanical vibrations to the point where they can be compensated by the MONSTR's active stabilization. Introducing an assembly that contains the metrology laser also expedites and simplifies the alignment procedure. We demonstrate the viability of 2DFT measurements using the 25 Tesla resistive magnet by collecting 2DFT spectra from Landau levels in GaAs quantum wells and bulk GaAs excitons. The technique development presented paves the way for 2DFT measurements at magnetic fields up to 25 Tesla on many materials of interest, including high-mobility two-dimensional electron gases and two-dimensional TMD devices.

IV. SUPPLEMENTARY MATERIAL

The supplementary material contains software that provide examples of loop filters implemented using the FPGA hardware. Furthermore, phase stability measurements using the FPGA hardware and the LabView software under operational conditions are also included.

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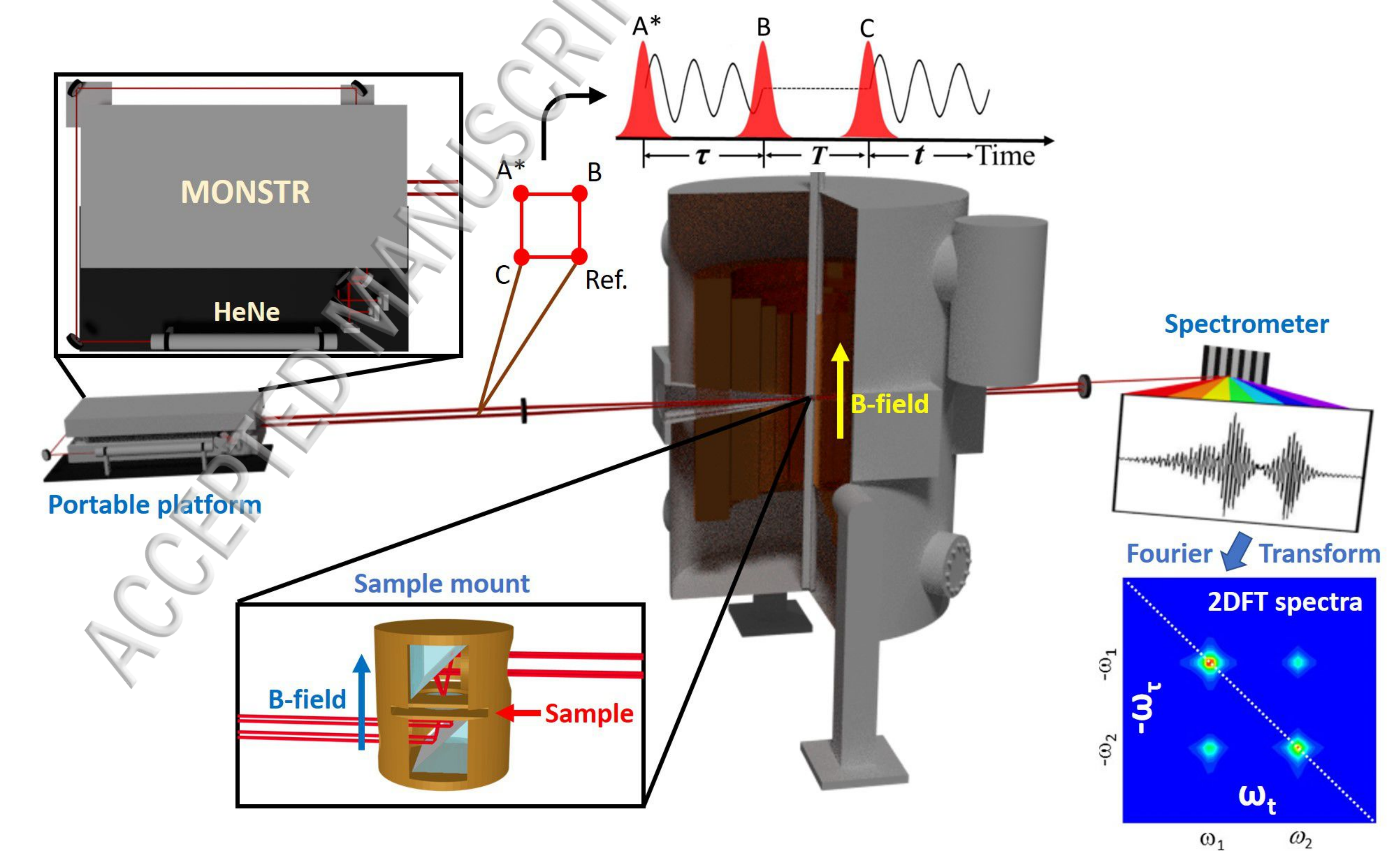

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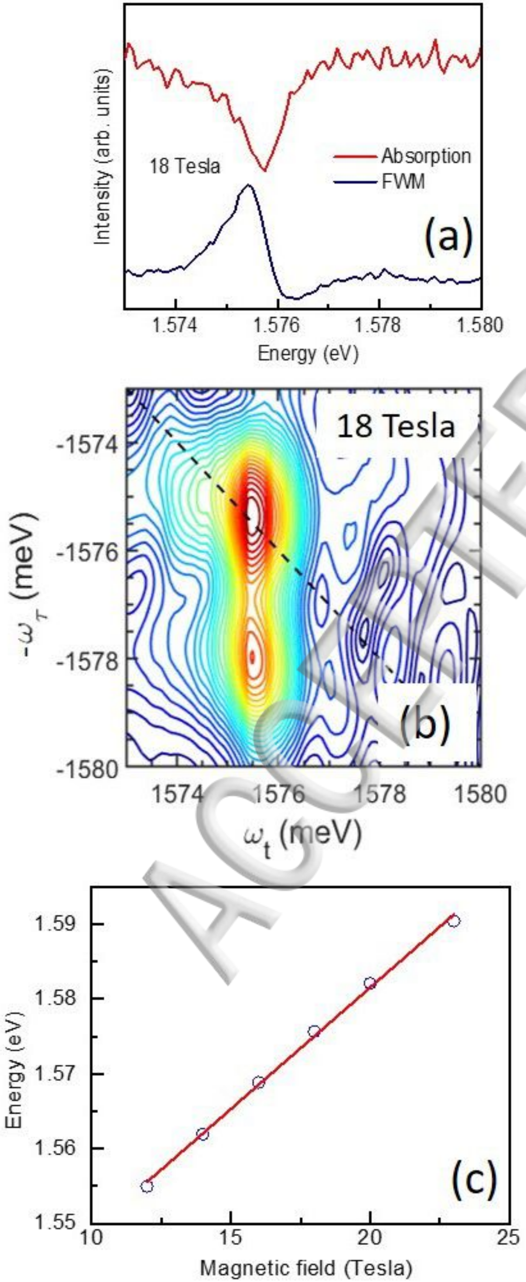
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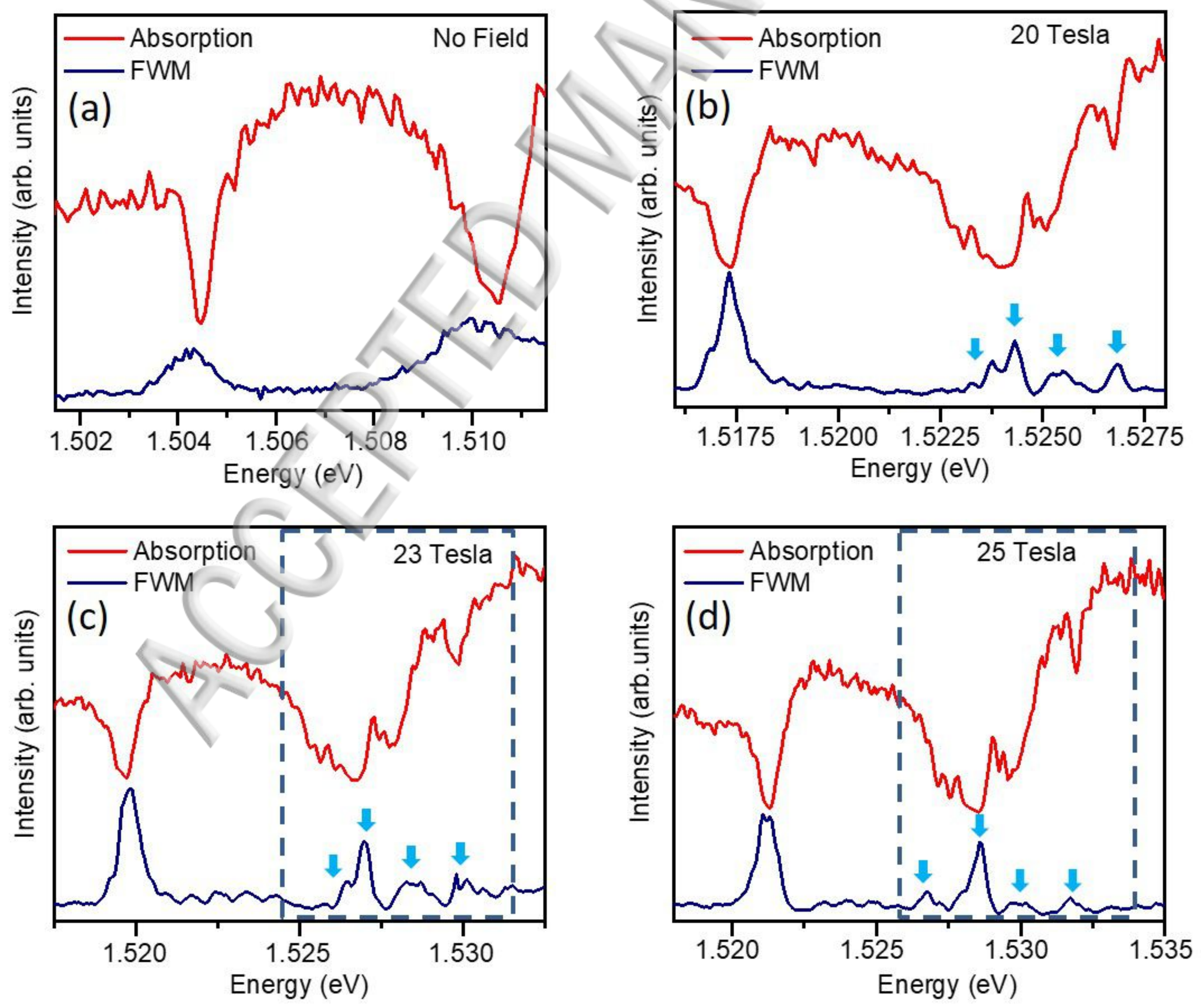

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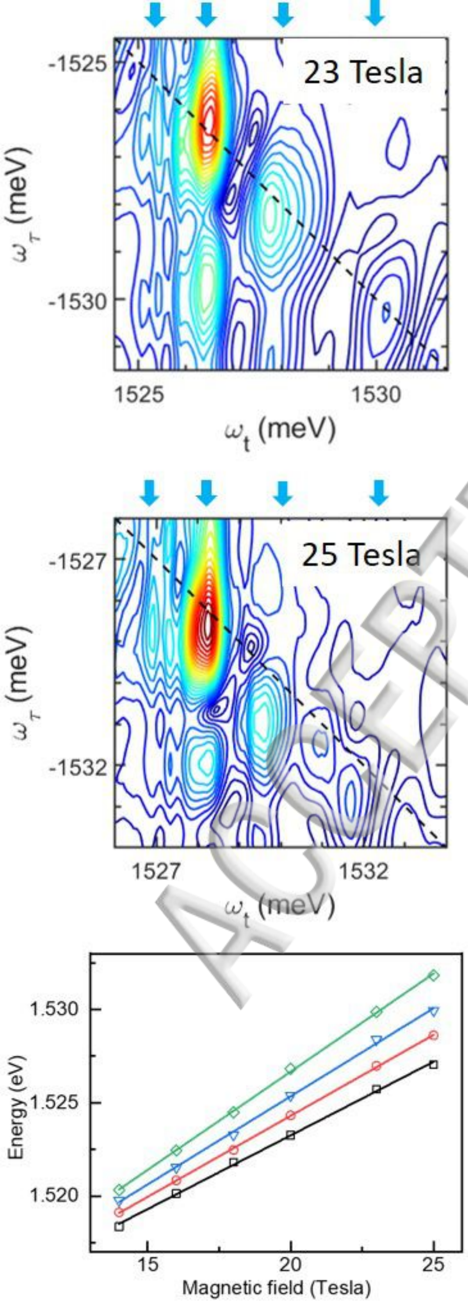
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Magnetic field (Tesla)