Spectroscopy of Matter in High Magnetic Fields

We report on near-infrared and visible spectroscopy in high magnetic fields. The acquisition of the 50 T pulsed magnet combined with the IR-VIS spectroscopy system has been improved from one data point per magnetic pulse shot to twelve data points in a single magnet pulse shot. The g-factor and the diamagnetic shift have been measured in two-dimensional Ruddlesden-Popper perovskites.

The two-dimensional (2D) Ruddlesden-Popper (RP) perovskites form quantum wells by sandwiching inorganic-organic perovskite layers used in photovoltaic devices between organic layers [1]. The perovskite layer thickness defined by the integer value n in $(BA)_2(MA)_{n-1}Pb_n I_{3n+1}$ determines their optical properties. Here, investigated samples with n=5 using IR-VIS magnetospectroscopy up to 40 T.

The 50 T-capable RAMBO-II IR/VIS magnet produces a nearly 2 ms duration asymmetric magnetic field pulse (see Fig. 1) at intervals of 57 seconds for every Tesla above 7 Tesla at the peak. The scale of access to this magnet compared to the limited time available at national facilities is an incredible boon and enables us to specialize the surrounding experiment to be even more efficient in data collection. Our experimental system can measure magneto-optical properties at 14 distinct times in 12 different fields during a single pulse. An entire field sweep can thus be accomplished with a single button press. Due to the increasing resistance from Joule heating, the asymmetric pulse shape gives slower rates of change in magnetic field and increases the time available to sample more fields with less variation.

Figure 1a) shows the magnetic pulse profile in blue. Laser pulses are in red, and camera exposure intervals are in orange. The data can be acquired by the CCD camera (see images in Fig.1b)) for each laser pulse shown Fig. 1a). The inset in Fig. 1 shows the distribution of magnetic fields that can currently be sampled from one 40 T pulse. The pulse shape does not scale linearly, but the distribution at 50T is expected to be comparable. In this way, all the magneto-optical data for material across the full -50 to +50T range, σ^+ and σ^- (circularly polarized light), at a given temperature can be taken with only four pulses of the magnet. The final incremental upgrade to 50 T is also expected to coincide with additional experimental improvements, increasing the acquisition rate of spectroscopic data and, thus, the total number and density of sampled fields.

The 50T IR/VIS system is first applied to studying 2D RP perovskites. In these hybrid solar cell materials, layers of methylammonium lead iodide are stacked between insulating layers of

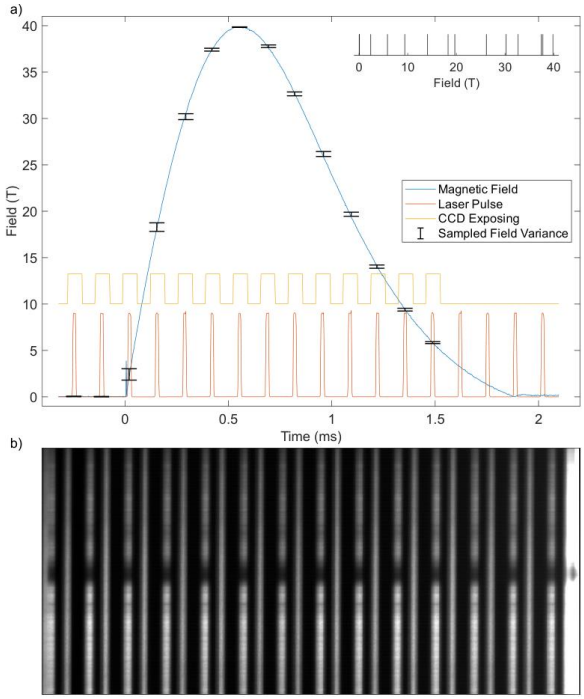

Fig. 1. a) Magnetic pulse profile (blue) together with light pulse train (red) and exposure intervals (orange). Black dots with error bars show magnetic fields for which data can be acquired for a single magnetic pulse shot. The inset shows the distribution of fields that can be sampled from a single magnetic pulse with the 40T peak field. b) Each image is a frame taken by the camera, corresponding to a sampling of the magnetic field in the a) plot.

butylammonium to create a pseudo-quantum-well structure. The inclusion of butylammonium provides additional chemical stability to the famously reactive perovskite, but the 2D confinement enhances the exciton interaction – increasing their binding energy and reducing the rate of free carrier escape under photoexcitation.

The studies of the diamagnetic shift via magnetospectroscopy grant insight into the changing size of the exciton at different temperatures, which we are using as part of a paper-in-preparation to make some predictions about the much harder-to-measure exciton binding energy in these materials. This experimental setup has allowed us to experiment with a broad set of permutations on material parameters, namely stacking order, orientation,

and temperature. The number of measurements this growing parameter space requires becomes prohibitive when experiment time is limited at a high magnetic field facility but can be accommodated with in-house experiments.

Figure 2a) shows an image mounted into a 50 T magnet. The sample is situated on the sapphire pipe connected to the sample cryostat. The transmitted light is collected using the fiber located right below the sample. We have conducted transmission experiments for both right and left circularly polarized light in the -40 T and + 40 T magnetic field ranges. Figure 2b) shows exemplary data for the exciton attenuated of 2D RP perovskite *n*=5, at 0, +40 T for right and left circularly polarized light. As one can see, the exciton peaks shift in opposite directions based on light chirality, but this shift is asymmetric due to a larger contrition of diamagnetic shift.

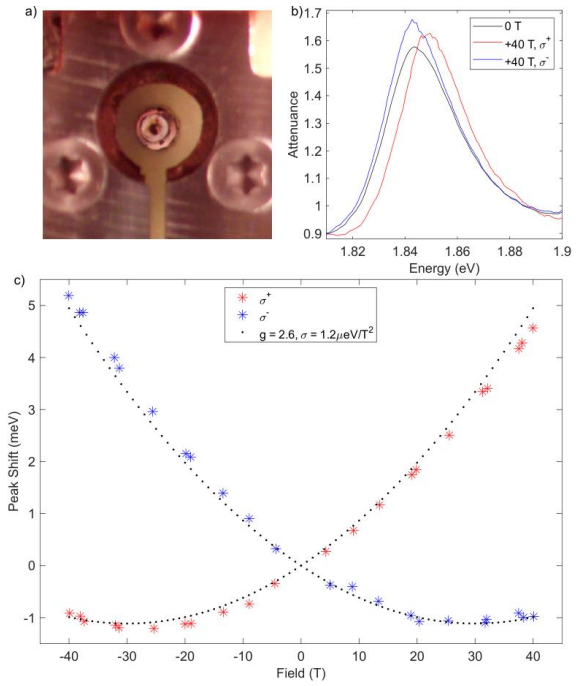

Fig. 2. a) an optical image of a 50T coil with the mounted sample. b) Attenuance spectra of $(BA)_2(MA)_{n-1}Pb_n I_{3n+1}$ for n=5 at 0 T and 40 T for two circular polarizations. c) Exciton peak shift as a function of magnetic field for two circular polarizations. The dashed line is the fit.

Figure 2c) summarizes exciton peak shift as a function of the magnetic field at different light polarizations. The data was fitted to the equation [2]:

$$E = E(B = 0) \pm \frac{1}{2} g\mu_B B + \sigma_0 B^2,$$

resulting in $g = 2.7$ and $\sigma = 1.6$ μeV/T^2.

References

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[2] N. Miura, *Physics of Semiconductors in High Magnetic Fields*, Vol. **15**, Oxford University Press, Oxford 2008.

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