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THz driven dynamics in Mott insulator GaTa₄Se₈

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Abstract — GaTa₄Se₈ is a Mott insulator known to exhibit an electric Mott transition, characterized by a drop in electrical resistivity, when an electric field larger than 1 – 10 kV/cm is applied for a few tens of microseconds using electrodes deposited on the sample. Here, we show that a resistivity drop can be induced in this material within less than a picosecond. These dynamics occur after excitation by a high field THz pump pulse and persist for a few picoseconds, well beyond the duration of the pump pulse.

I. INTRODUCTION

MOTT insulators are archetypal examples of quantum materials. Strong interest in these systems has arisen due in part to the insulator-to-metal transition that some exhibit when the balance between on-site Coulomb repulsion and hopping is overturned via temperature, doping or, as more recently demonstrated, photoexcitation or the application of short electric field pulses. The transition driven by electric field has been studied in several types of materials such as chalcogenides, oxides and molecular crystals. In these compounds, an abrupt drop of electrical resistivity is observed under the application of electric fields for a few tens of microseconds. This occurs only for fields above a typical 1 – 10 kV/cm threshold [1]. Such electric Mott transitions are volatile for fields just above threshold but become non-volatile under larger electric fields, which is very promising for possible applications in e.g. artificial Mott neurons or non-volatile Mott memories.

II. RESULTS

Quasi-dc electric fields well in excess of 1 – 10 kV/cm can currently be generated with ultrashort pulses in the low frequency or THz range, which enables the investigation of the sub-picosecond dynamics of the electric field driven Mott transition. THz pulses can also be used to track the Drude conductivity response of the sample directly, without the need to deposit any electrical contacts on the sample. In our measurements THz pulses generated via optical rectification in organic DSTMS crystals are used, with peak field values of 1 MV/cm for the pump and < 100 kV/cm for the probe.

We present our results on THz driven dynamics in bulk GaTa₄Se₈, a Mott insulator which exhibits clear electric Mott transitions. At atmospheric pressure the paramagnetic Mott insulator phase of GaTa₄Se₈ extends roughly between 55 K and 150 K [1], followed at higher temperatures by a smooth crossover into a “bad Mott” region where the gap is progressively filled by transferred incoherent spectral weight [2]. As shown in Fig. 1, we observe a sub-picosecond THz induced decrease of the THz transmission, consistent with an ultrafast decrease in the resistivity of the material. This response persists for a few picoseconds, beyond the duration of the THz pump pulse, as seen by comparing the nonlinear signal

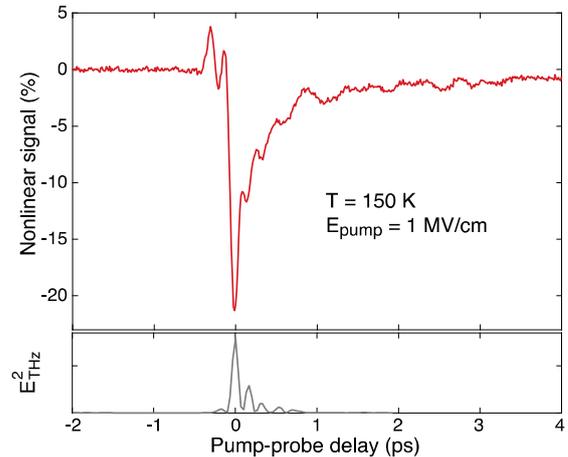


Fig. 1. Top: THz induced change in the transmission of the maximum of the THz probe pulse through a 150 μm thick GaTa₄Se₈ sample cooled down to 150 K, following excitation by a THz pump with ~ 1 MV/cm peak field. Bottom: temporal trace of the square of the THz pump pulse.

dynamics with the time trace of the pump shown at the bottom of Fig. 1. Our result demonstrates that a sub-picosecond long THz pump pulse is capable of inducing a resistivity drop in GaTa₄Se₈, similar to what was observed in static measurements using few tens of microseconds long electric fields, enabling the study of the dynamics of the electric Mott transition mechanism.

Furthermore, the nonlinear signal we detect depends strongly on the relative orientation of the pump and probe field polarizations. Fig. 1 shows the case where the two polarizations are parallel to each other. Crossed polarizations (not shown) lead to a decrease of the nonlinear signal by more than one order of magnitude. This indicates that the THz induced metallic domains arise from percolation preferentially along the THz pump field polarization. This anisotropy is consistent with that observed during the static measurements where dc electric fields are applied for a few tens of microseconds [3].

Fig. 2 shows the dependence of the nonlinear signal on pump field and on temperature in the 40 K – 295 K range. In order to avoid nonlinear contributions from the pump – probe temporal overlap region, as well as from reflection within the 150 μm thick sample, we restrict our analysis to the 0.2 – 2.5 ps delay range. Each point in Fig. 2a corresponds to the integral of the nonlinear signal (as shown in Fig. 1) in this time range. The nonlinear signal is seen to increase with pump field and with temperature, with the exception of the room temperature data. This exception is not surprising given that 295 K is well into the crossover region of the GaTa₄Se₈ phase diagram and there is little reason to expect the system to exhibit the same behavior in that region as in the Mott phase. Interestingly, data obtained

at 220 K in the crossover region between the Mott and “bad Mott” insulator phases seems to follow the trend of the < 150 K data. This indicates that the presence of a small but finite

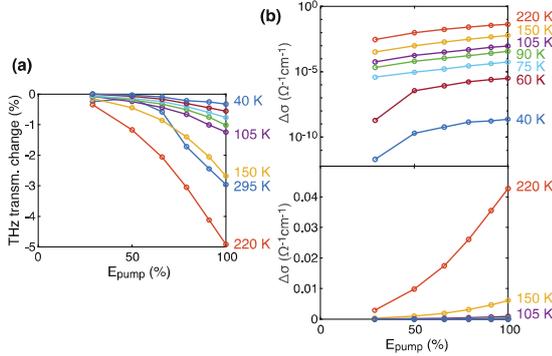


Fig. 2. (a) THz transmission change as a function of pump field and temperature. (b) Extracted change in conductivity plotted in logarithmic (top) and linear (bottom) scale.

density of states at E_F is not detrimental to the THz induced ultrafast decrease in the resistivity of the material.

Comparing the static temperature dependence of the THz probe transmission and of the sample conductivity enables mapping of the peak value of the transmitted THz probe pulse to a dc conductivity value. This procedure assumes that the peak of the THz probe pulse remains a good measure of the THz transmission when varying the pump-probe delay, i.e. that the spectral content of the THz probe stays unchanged and only its overall amplitude is modified. We have verified that this is the case for the pump-probe delay times under consideration (> 0.2 ps) by performing full 2d measurements, not shown here, where a complete time domain trace of the THz probe field is measured at each pump-probe delay time. The dynamics of the conductivity are shown in Fig. 2b, where the 295 K data have been omitted. It is clear from the top panel of Fig. 2b that the conductivity increases more for larger pump fields and larger temperatures.

The temperature and field dependence of the signal enable us to discuss the origin of the carrier generation process that leads to population of the Fermi level in this Mott insulator, and consequently to the observed increase in transient conductivity, following the application of a strong THz electric field. The THz photon energy is < 10 meV, so much smaller than the ~ 150 meV bandgap of GaTa_4Se_8 [4] that direct photoexcitation of the electrons into the conduction band is negligible, even if multiphoton absorptions processes are taken into account. Two broad classes of carrier generation processes must therefore be considered: Zener tunneling and electronic avalanche. Zener tunneling occurs under a strong electric field and can create a finite carrier density in an initially empty Fermi level, which is the case in a perfect Mott insulator. In turn, electronic avalanche requires that a few free carriers already exist in the conduction band, which can then be accelerated by the electric field. Carrier multiplication via electronic avalanche relies on two distinct mechanisms depending on temperature: impact ionization involving independent electrons at low temperature, when the mean free path is large, and a collective electronic process leading to a divergence of the electronic temperature at higher temperature [5]. In practice, the THz pump pulse can always

cause some amount of electronic avalanche, either relying on a defect- or impurity-induced carrier population initially present at the Fermi level or following a carrier generation tunneling process. If the contribution from Zener tunneling is significant, a threshold field $E_{pump,thr}$ is expected, which can be estimated by noting that the conductivity, proportional to the carrier density within a Drude model description, should be proportional to $\exp\left(-\pi \frac{E_{pump,thr}}{E_{pump}}\right)$ [6]. Fitting the data in the bottom panel of Fig. 2b by this function yields good fits and values of $E_{pump,thr} \sim 450$ kV/cm for the lowest temperatures in the Mott phase. These correspond to quite reasonable tunneling distances of < 2 nm. At higher temperatures the fit quality worsens and the extracted $E_{pump,thr}$ values decrease significantly.

Our analysis is consistent with a scenario in which Zener tunneling is significant as a carrier generation process at low temperatures, followed by electronic avalanche breakdown. In the Mott phase no carriers are expected to exist at the Fermi level and few defect- or impurity-induced carriers are thermally excited at these low temperatures. We can, however, not rule out a purely avalanche driven scenario at low temperatures, initiated by impact ionization, which would also exhibit a threshold field behavior. At higher temperatures, even though nominally still in the Mott phase, more defect- or impurity-induced carriers can be thermally excited and the role of electronic avalanche becomes dominant, to the detriment of Zener tunneling, leading to a dependence of the conductivity on E_{pump} which deviates from the functional form above [6].

III. SUMMARY

We demonstrate that a low resistance state can be induced in Mott insulator GaTa_4Se_8 following the application of a strong THz electric field transient, similar to what is achieved under application of dc electric fields during a few tens of microseconds. The low resistance state persists for a few picoseconds and is aligned along the electric field direction. Analyzing the temperature and pump field dependence of the nonlinear response enables a discussion of the role of Zener tunneling and electronic avalanche processes in the carrier generation process that leads to this transient metallic phase.

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