Terahertz-driven magnetoelectric torque in the collinear antiferromagnet Cr_2O_3

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Magnetoelectric effect, facilitating a control of spins in magnets with the help of electric field, has long been seen as a cornerstone for future energy efficient and nano-scalable technologies for magnetic writing and processing of magnetically stored information. In contrast to spin-polarized currents, a control of spins with the help of electric fields promises much lower dissipations and in contrast to magnetic fields, electric fields are easier to apply to a nanoscale bit. Understanding temporal evolution of the magneto-electric effect, revealing how fast spins respond to an electric field is thus crucial for revealing the fundamental limit on the operational frequency of magnetoelectric devices. Here we report on ultrafast magnetoelectric effect in antiferromagnetic Cr_2O_3.

Chromium oxide Cr_2O_3 is a prototypical magnetoelectric antiferromagnet with the corundum trigonal crystal structure (s.g. R3c) and the Néel temperature T_N=307.6 K. The studied sample in the form of a thin plane plate cut such that the antiferromagnetically ordered spins of Cr^{3+} ions lie in the sample plane $(10\bar{1}0)$ parallel to crystallographic *c*-axis of the crystal. The sample was excited with the THz pulses electrical field of which were able to rotate in the plane of sample surface with the set of two wire grid polarizers as described in ref. [1]. THz-induced dynamics excited were detected with a balance photodetector as a probe polarization rotation. Microscopical image (fig. 1a) of the domain structure was visualized with circularly polarized probe pulses [2].

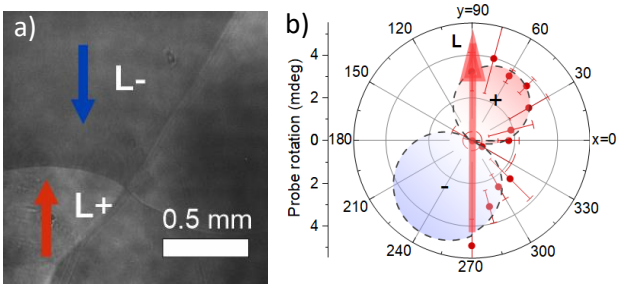

Figure 1. a) SHG image of the domain pattern gained with the circularly polarized probe with the energy of fundamental pulses 2.1eV. b) The amplitude of the oscillations excited at different angles of the THz electric field observed in a single domain. The angle values stand for orientation of electric field $\mathbf{E}_{THz}$ of the THz pulse .

The THz-induced transient signals (fig. 2) for the probe polarization rotation measured in Cr_2O_3 at temperature 77K for various polarizations of the THz pump pulse with respect to the antiferromagnetic Néel vector **L** show oscillations at the frequency of the antiferromagnetic resonance at 0.165 ps [3,4] and is related to the THz pump driven spin dynamics.

When the magnetic field is orthogonal to the antiferromagnetic Néel vector $\mathbf{H}_{THz} \perp \mathbf{L}$ [5,6] Zeeman torque excitation mechanism is expected to reach the maximum values. In contrast, when the magnetic field of the THz pump pulse is parallel to the antiferromagnetic Néel vector $\mathbf{H}_{THz} \parallel \mathbf{L}$, and consequently $\mathbf{E}_{THz} \perp \mathbf{L}$, the efficiency of this excitation mechanism must be equal to zero. It is seen in Fig. 1b that the spin dynamics in Cr_2O_3 is excited with comparable efficiency both by the Zeeman torque at $\mathbf{H}_{THz} \perp \mathbf{L}$, and at the orthogonal to it polarization $\mathbf{H}_{THz} \parallel \mathbf{L}$ ($\mathbf{E}_{THz} \perp \mathbf{L}$).

The amplitude of the oscillations depends on the THz pump polarization angle (fig. 1b) and have a maximum in between $\mathbf{H}_{THz} \perp \mathbf{L}$ and $\mathbf{E}_{THz} \perp \mathbf{L}$ which indicates interference between two torques that can excite spin dynamics. The amplitude of oscillations is a linear function of the THz magnetic and electric field strengths when the magnetic field $\mathbf{H}_{THz}$ is parallel and perpendicular to the antiferromagnetic Néel vector **L**.

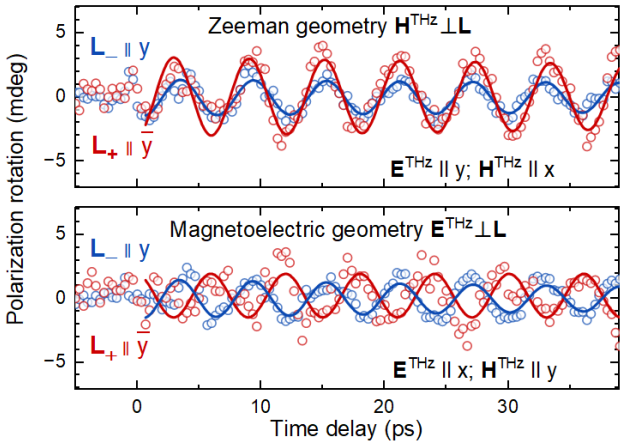

Figure 2. THz-induced spin dynamics measured for domains with oppositely oriented Néel vectors $\mathbf{L}_+$ and $\mathbf{L}_-$ for Zeeman and magnetoelectric geometries. different angles between the THz electric field and antiferromagnetic **L** vector at 77K.

In order to distinguish between these two excitation mechanisms we performed an experiment (fig. 2) where probe rotation dynamics were measured in two domains with oppositely oriented **L** vector and the THz electric field $\mathbf{E}_{THz} \parallel \mathbf{L}$ ($\mathbf{H}_{THz} \perp \mathbf{L}$) was oriented either parallel or perpendicular $\mathbf{E}_{THz} \perp \mathbf{L}$ to Néel vector which can be ascribed to Zeeman and magnetoelectric torque. When the THz magnetic

field and the antiferromagnetic Néel vector are mutually perpendicular $\mathbf{H}_{THz} \perp \mathbf{L}$ (Zeeman torque), a reversal of the **L** vector upon transition to opposite domain does not affect the detected spin dynamics. In case of the THz magnetic field and the antiferromagnetic Néel vector are mutually perpendicular $\mathbf{H}_{THz} \parallel \mathbf{L}$ which is equivalent to $\mathbf{E}_{THz} \perp \mathbf{L}$ (magnetoelectric torque), a reversal of **L** is changes the phase of the oscillations by 180°.

Such behavior can be explained taking into account thermodynamical potential Φ for magnetoelectric antiferromagnet having term a term $M_x E_x L_y$. The effective magnetic field that triggers the dynamics of Néel vector would be $H_{eff} \sim E_x L_y$ and thus provides the phase change of the oscillations on reversal of both either electric field or Néel vector in the observable magnetization dynamics for the magnetoelectric mechanism of excitation. At the same time excitation by the Zeeman torque $d\mathbf{L}/dt \sim \mathbf{L} \times \mathbf{H}$ does not depend on the orientation of **L** and changes the phase of the oscillations upon the reversal of magnetic field.

We have demonstrated that antiferromagnetic resonance in Cr_2O_3 can be driven using nearly single cycle THz electromagnetic pulse. It is shown that the electric component of the THz pulse plays in the excitation significant role that exert a magnetoelectric along the Zeeman torque. Hence, our experiments also reveal a fundamentally new mechanism for ultrafast control of spins in antiferromagnets.

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