Vacuum-Rabi Oscillations in the polar honeycomb antiferromagnets $Fe_2Mo_3O_8$ and $Co_2Mo_3O_8$ probed by THz-time domain spectroscopy

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Vacuum-Rabi oscillations and Dicke cooperativity were reported in the honeycomb antiferromagnet $Fe_2Mo_3O_8$ [1], a member of the $A_2Mo_3O_8$ family of materials with A = Fe, Co, Mn, Zn, which is currently in the focus of research because of its multiferroic and magnetoelectric properties [2-7]. These observations in $Fe_2Mo_3O_8$ were tentatively assigned to magnon-polariton formation [1], although the nature of the low-lying THz excitations in $Fe_2Mo_3O_8$ has not yet be clarified [2-4], but our calculations show that the optical modes in the THz range can be understood as d-d-like transitions with mixed orbital and spin character due to spin-orbit coupling. We observed vacuum-Rabi oscillations in pure and Zn-doped $Fe_2Mo_3O_8$ and in $Co_2Mo_3O_8$ (see Fig. 1) and studied the dependence of the Rabi-splitting on the intensity of the incoming THz radiation, the thickness of the crystals and the response on an external magnetic field. As the Co ions have different electronic configuration and spin state, the occurrence of the Rabi oscillations in this family of polar honeycomb antiferromagnets is not related to the particular mode in $Fe_2Mo_3O_8$, but originates from the characteristics of the antiferromagnetic ground state of these materials. Note that the time-trace spectra in Zn-doped $Fe_2Mo_3O_8$ does not exhibit vacuum-Rabi oscillations any more upon applying a magnetic field of 2T, which drives the system to a ferrimagnetic state. We will discuss the observed changes of the Rabi-splitting in these systems and compare our experimental results with simulations using a semi-classical approach [7].

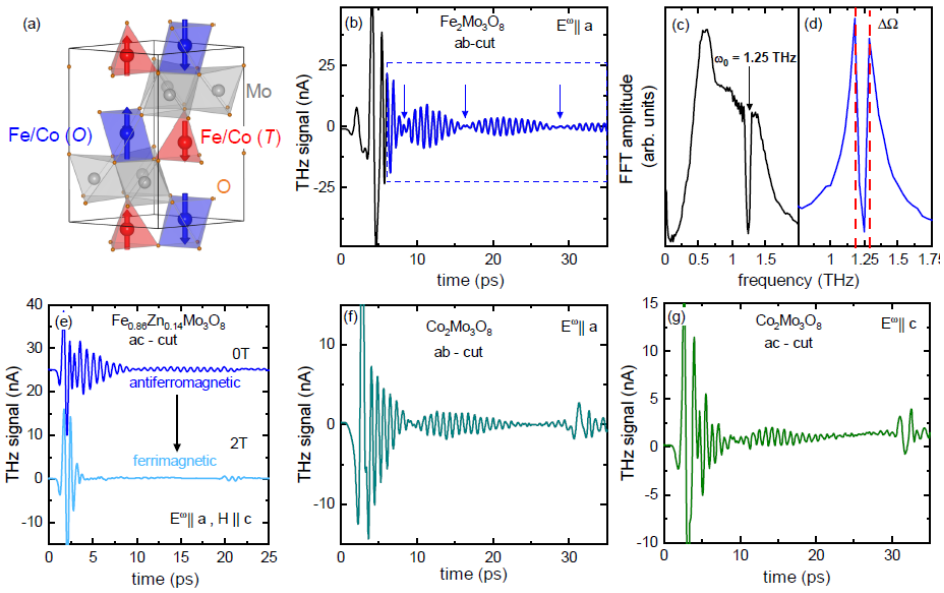

Fig. 1. a) Structure of the polar antiferromagnets $A_2Mo_3O_8$, where the magnetic ions, here Fe or Co, occupy sites with tetrahedral (T) and octahedral (O) coordination. b) Time-domain spectra in antiferromagnetic $Fe_2Mo_3O_8$ at 2 K exhibiting beatings with nodes indicated by arrows. Amplitude spectra after Fourier-transformation of c) the entire time-domain signal revealing the fundamental mode at $\omega_0 = 1.25$ THz and d) of the blue (boxed) part of the time-domain spectra with the beatings, revealing the Rabi-splitting $\Delta\Omega$ of the dressed states. e) Comparison of the time-domain signals of an ac-cut single crystal of Zn doped $Fe_2Mo_3O_8$ in the antiferromagnetic ground state in 0 T and in the induced ferrimagnetic state in 2 T at T = 25 K. f) Time-domain spectrum and Rabi oscillations in $Co_2Mo_3O_8$ at 2 K for light-polarization $E^{\omega} \parallel a$. d) Time-domain spectrum and Rabi oscillations in $Co_2Mo_3O_8$ at 2 K for light-polarization $E^{\omega} \perp a$.

References

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