Electronic Supplementary Information

Magnetic polarons reach a hundred thousand Bohr magnetons

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Materials and methods

a. Samples

EuO films were synthesized in a Riber Compact system for molecular beam epitaxy furnished with an UHV system to maintain the residual pressure below 10^{-10} Torr. The substrate was a YSZ(001) wafer (13 molar % of yttria) with a thickness of 0.5 mm and a miscut angle less than 0.5°. The YSZ wafer was annealed at 600 °C for half an hour in $3 \cdot 10^{-8}$ Torr of oxygen. The substrate temperature was controlled with a PhotriX ML-AAPX infrared pyrometer. The flux of 6N O_2 was maintained employing a system based on a mass flow controller and a Baratron manometer. The pressures of the reactants in the MBE chamber were determined with a Bayard-Alpert ionization gauge. The nucleating 5 monolayers of EuO were grown by deposition of 4N Eu ($1.2 \cdot 10^{-7}$ Torr) on the YSZ wafer cooled to 430 °C. The bulk of the film (200 nm) was produced at 430 °C by co-deposition of Eu ($1.2 \cdot 10^{-7}$ Torr) and O_2 ($6 \cdot 10^{-9}$ Torr). The synthesis conditions corresponded to the Eu distillation mode – high temperature and an excess of Eu – to prevent formation of Eu_2O_3 and Eu_3O_4. The films were capped with a layer of SiO_x to protect EuO from degradation by air.

The atomic structure of the EuO films was controlled by diffraction techniques. First, the structure was studied *in situ* employing a RHEED diffractometer equipped with the kSA 400 analytical RHEED system. Then, XRD measurements were carried out *ex situ*: a Rigaku SmartLab diffractometer produced θ-2θ scans using a Cu $K_{\alpha 1}$ radiation source. The magnetic properties of EuO were measured by an MPMS XL-7 SQUID magnetometer. The measurements were carried out in the RSO mode employing samples with a lateral size of 5 mm.

b. Magneto-optical studies

The experimental setup for magneto-optical studies is schematically shown in Fig. S1. It employs a two-color pump-probe technique and allows measurements of the conventional Faraday effect and the PFE. To carry out time-resolved studies of the PFE, we used a femtosecond pulsed laser [Yb:$KGd(WO_4)_2$] with a kilohertz repetition rate: the photon energy was 1.19 eV, the pulse duration was 190 fs, and the repetition rate – 10 kHz. The pump beam (2.38 eV) produced by doubling the photon energy by a nonlinear crystal of β-BaB_2O_4 (BBO) was modulated by a mechanical chopper at a frequency of 625 Hz. The studies of the PFE amplitude variation with the temperature (Fig. S2) and the frequency of pump modulation

(Fig. S4) were carried out at the repetition rate 72 MHz and the mechanical chopper was replaced with an electro-optical modulator (frequency range from 1 kHz to 1 MHz). The sample was cooled in a helium cryostat placed in the gap of an electromagnet. The pump and probe beams were focused on a spot on the sample with a diameter of about 50 μm. The probe beam transmitted through the sample was measured by a balanced detector with a lock-in amplifier at the frequency of the pump modulation. In the case of time-resolved PFE measurements, an optical delay line was employed, enabling pump-probe measurements with a time delay up to 2.8 ns. The external DC magnetic field was varied in the range from -1 T to +1 T. Perpendicular and oblique geometries of the sample orientation relative to the direction of the external field and incident light were used to measure the PFE and the conventional Faraday effect, respectively. The PFE measurements were carried out at different values of the pump power. The fit of the experimental PFE data by the Langevin function was carried out using the Levenberg-Marquardt algorithm, a robust iterative procedure interpolating between the Gauss-Newton and gradient descent methods to solve non-linear least squares problems.

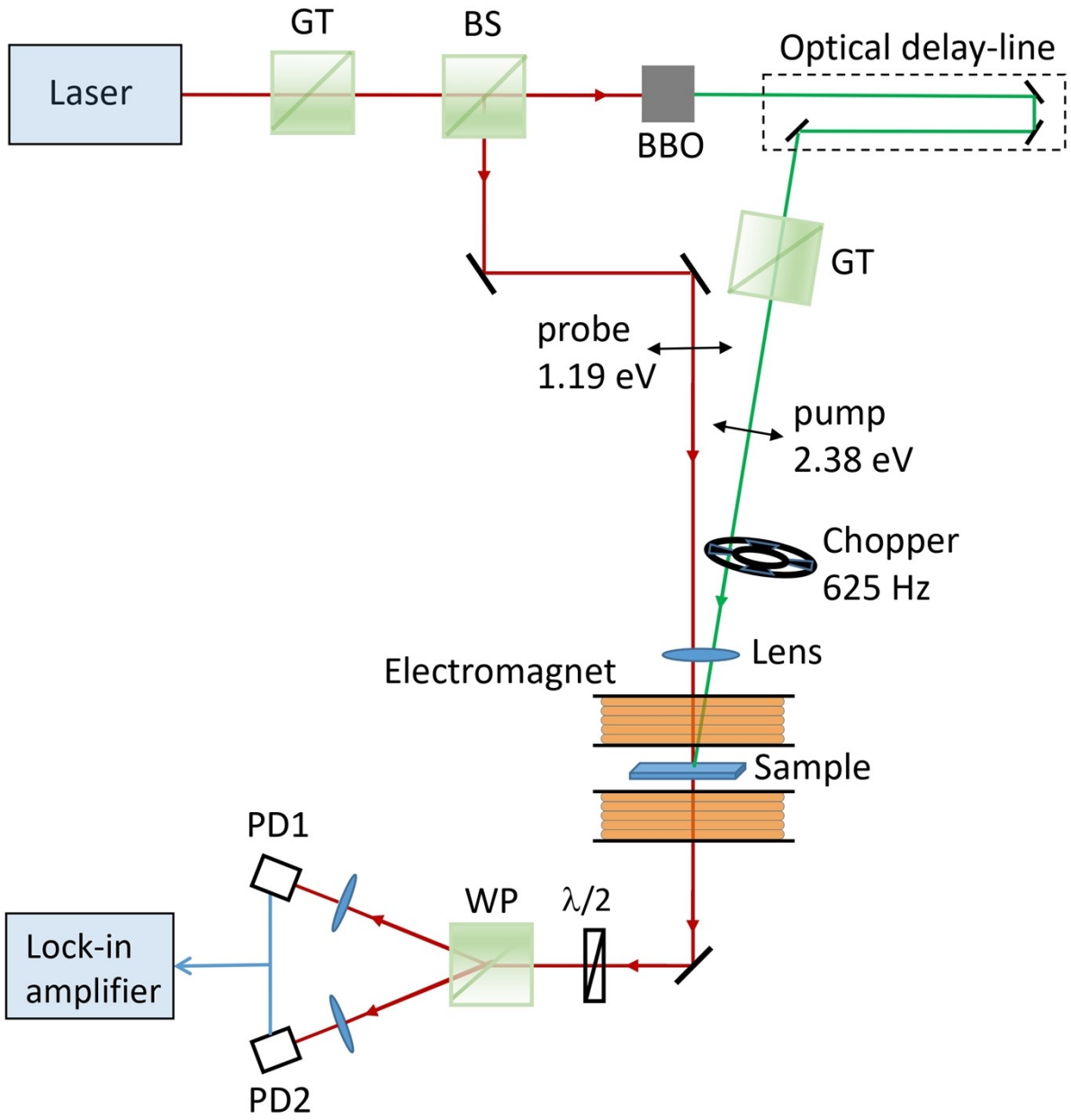

Fig. S1. Experimental setup for the PFE studies in EuO; GT is a Glan-Thompson polarizer, BS – a beam splitter, BBO – a crystal of β-BaB_2O_4, λ/2 – a half-wave plate, WP – a Wollaston prism, PD1 and PD2 – photodiodes.

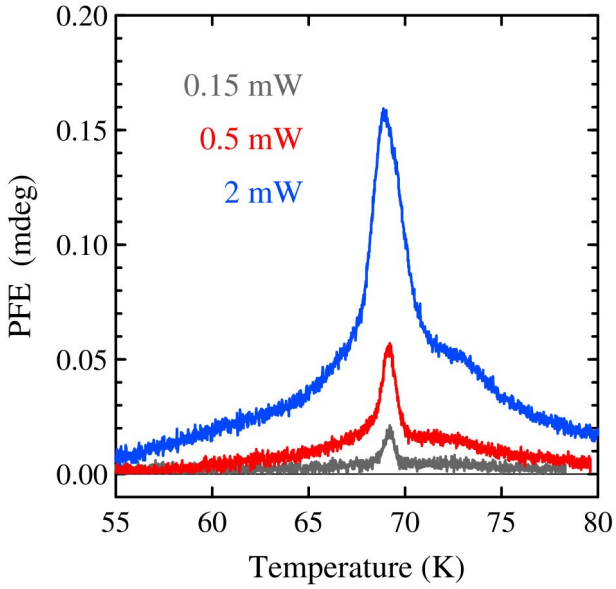

Fig. S2. Temperature dependence of the PFE amplitude in a magnetic field of 20 mT for different values of pump power – 0.15 mW (grey), 0.5 mW (red), and 2 mW (blue).

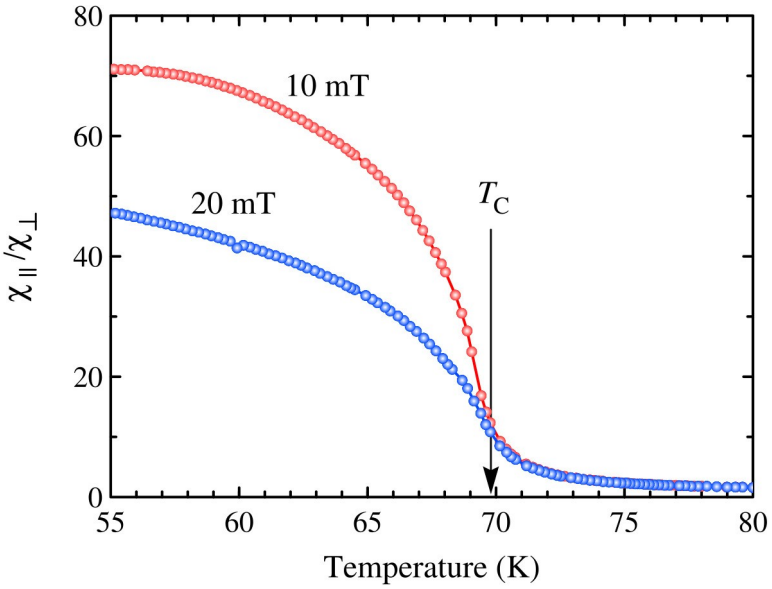

Fig. S3. Ratio of magnetic susceptibilities ($\chi_{\parallel}/\chi_{\perp}$) of the EuO film in magnetic fields 10 mT (red) and 20 mT (blue) (SQUID measurements).

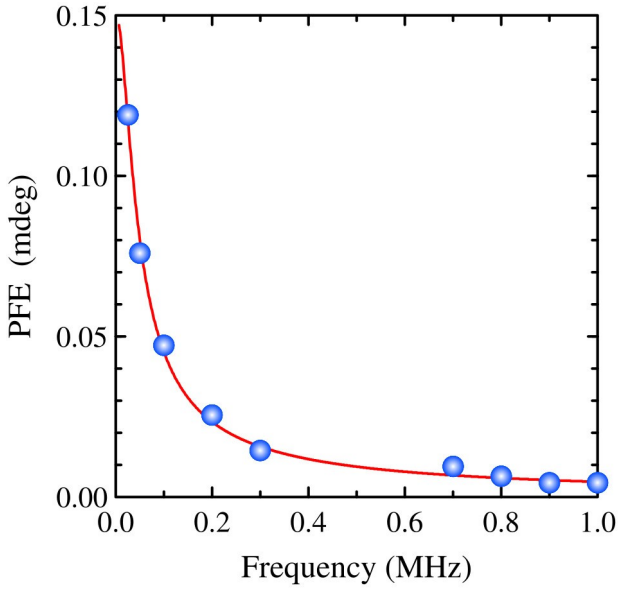

Fig. S4. Dependence of the PFE amplitude on the pump modulation frequency f: experimental values (blue dots) and their fit to $PFE(f) = PFE(0)/\sqrt{1 + (2\pi f \tau_{pol})^2}$ (red line), where τ_{pol} is the MP lifetime (≈ 5 μs).