Investigation of exchange coupled bilayer Fe/CuMnAs by pump–probe experiment

Vít Saidl1,2, Petr Němec1, Peter Wadley1, Kevin W. Edmonds3, Richard P. Campion1, Vit Novák2, Bryan L. Gallagher1, František Trojánek1, and Tomáš Jungwirth2,3

1 Faculty of Mathematics and Physics, Charles University, Ke Karlovu 3, 12116 Prague 2, Czech Republic
2 Institute of Physics ASCR, v.v.i., Cukrovarnická 10, 16253, Prague 6, Czech Republic
3 School of Physics and Astronomy, University of Nottingham, Nottingham NG7 2RD, UK

Received 22 December 2016, revised 8 February 2017, accepted 9 February 2017
Published online 20 February 2017

Keywords exchange coupling, antiferromagnets, CuMnAs, Fe, ultrafast laser spectroscopy, pump–probe technique

1 Introduction
The exchange interaction between a ferromagnetic (FM) and an antiferromagnetic (AF) layer leads to an unidirectional anisotropy in FM, which is called exchange bias. This phenomenon plays an important role in spin valves where the AF layer fixes the orientation of the FM magnetization [1, 2]. FM/AF bilayers are often studied by static measurements of magnetic hysteresis loops [3–6]. However, dynamical experiments, like the pump and probe technique, can provide a markedly distinct information about the FM/AF [4–14], FM/FM [15–20] or ferrimagnet/ferrimagnet [21] bilayer properties.

Compensated AF semimetal CuMnAs [22, 23] is a promising material for AF spintronics [24, 25] where the first realization of an AF memory chip was reported [26]. However, the zero net magnetic moment makes the magnetic characterization of this material a major challenge – traditionally, only X-ray magnetic linear dichroism (XMLD) has been used for the determination of the magnetic anisotropy in nanometer-thick AF films [23]. Very recently we have developed a new table-top pump–probe magneto-optical (MO) method that provides the information about magnetic ordering in CuMnAs epilayers [27]. In this paper we apply this MO technique for the study of a Fe/CuMnAs bilayer. This bilayer was explored recently by XMLD and it was shown that due to the interlayer exchange coupling with Fe, the Néel vector in CuMnAs can be rotated by an external magnetic field [28].

2 Pump–probe experimental study
2.1 Experimental details
We used a Ti:sapphire oscillator (Mai Tai, Spectra Physics, pulse duration 150 fs, repetition rate 80 MHz, wavelength 920 nm). Each laser pulse was divided into time-delayed (Δt) pump and probe pulses, which have a fluence ≈3 and 0.06 mJ cm−2, respectively, that were focused to ≈30 μm spot on the sample. For the studied material, the measured probe polarization rotation (MO signal in the following) was used to identify and separate parts of detected signals due to Faraday and Voigt magneto-optical effects that provide information about pump-induced magnetization precession in ferromagnetic Fe and reduction of the sublattice magnetization in antiferromagnetic CuMnAs layers, respectively. We observe a strong asymmetry in the dependence of the precession phase on the external magnetic field that we attribute to the exchange coupling between Fe and CuMnAs. Unlike in X-ray magnetic linear dichroism experiments, we do not observe any significant reorientation of magnetic moments in CuMnAs by external magnetic field due to the interlayer exchange coupling with Fe. Differences between these two experimental techniques, providing the distinct pictures, are discussed.
applied at an angle (see inset in Fig. 3) and at an angle $\approx 60^\circ$ with respect to the [110] in-plane crystallographic direction of the GaP substrate. Prior to the optical pump–probe measurement, the external magnetic field was applied at an angle $\approx 4^\circ$ with respect to the sample plane (see inset in Fig. 3) and at an angle $\approx 60^\circ$ with respect to the [110] in-plane crystallographic direction of the GaP substrate. Prior to the optical pump–probe measurements performed at $H_{ext} = 0$, a strong field of 600 mT was applied to saturate the Fe magnetization along the direction of $H_{ext}$. Measurements were performed at 15 K for normal incidence of probe pulses in a transmission geometry.

### 2.2 Sample growth and characterization

The experiments were performed on a Fe (2 nm)/CuMnAs (5 nm) sample capped with a 2 nm layer of Al, which was grown on a GaP(001) substrate by molecular beam epitaxy. The orientation of crystallographic axes of individual layers is: Fe(001)[110] || CuMnAs(001)[100] || GaP(001)[110].

The magnetic properties of the sample were characterized by a superconducting quantum interference device (SQUID) at 2 K – see Fig. 1. The measured loops are centered around zero external magnetic field which indicates a negligible exchange bias in the sample. The coercivity is around 7 mT and there is only a weak dependence on the direction of the field, i.e., there is a rather weak in-plane magnetic anisotropy of Fe.

### 2.3 Results and discussion

For fully compensated AFs, the MO signals from oppositely oriented magnetic sublattices cancel for MO effects linear (i.e. odd) in magnetization. On the other hand, MO effects quadratic (even) in magnetization are present also in AFs but it is rather difficult to separate the magnetic order-related signal from other sources of the light polarization change (e.g., strain- or crystal structure-related). These problems can be circumvented in a pump–probe experimental scheme where the probe pulse measures a quadratic MO signal reduction due to the pump-induced heating of the sample [27]. We used the MO Voigt effect, which is closely connected with magnetic linear dichroism (MLD), that is maximal for a normal incidence of probe light on the sample surface [27].

The fingerprint of this MO effect is a harmonic dependence of the change of the polarization rotation (or ellipticity) measured as a function of the mutual orientation of the probe light polarization and magnetic moments, which are described by angles $\varepsilon$ and $\phi$, respectively (see inset Fig. 2):

$$\text{MO}(\Delta \varepsilon, \varepsilon) = \frac{2P}{M} \sin 2(\phi - \varepsilon) \delta M(\Delta t).$$

Here $P$ is the MO coefficient, which scales quadratically with the sublattice magnetization $M$, and $\delta M$ is pump-induced change of magnetization [27]. In Fig. 2(a) and (b) we show MO signals measured in the studied Fe/CuMnAs sample for external magnetic fields $H_{ext}$ of 0 and +670 mT, respectively. In addition to the MO signal described by Eq. (1), which is due to the pump-induced reduction of the sublattice magnetization in CuMnAs, there are also oscillations due to a precession of ferromagnetic moments in Fe. The oscillations phase does not depend on the probe polarization (see the dashed line in Fig. 2(b)) which reveals that the Fe precession was detected by the Faraday effect in the polar geometry [29]. A separation of MO signals from Fe and CuMnAs can be performed by fitting the measured data by [30]

$$\text{MO}(\Delta t) = A \cos (2\pi f \Delta t + \theta) e^{-\Delta t/\tau_{G}} + C e^{-\Delta t/\tau_{s}} + D.$$

Here $A$ and $C$ are the amplitudes of the precession and de-magnetization-related MO signals, respectively, $f$ is the precession frequency, $\theta$ is the precession initial phase, $\tau_{G}$ is the Gilbert damping time, and $\tau_{s}$ is the demagnetization decay time.
Dependence of the precession frequency on lines are fits by Eq. (3). (d) Dependence of the precession phase from maximum negative (positive) value.

\[ f = \frac{1}{2\pi} \sqrt{\left(\mu_0 H_{\text{ext}} + B_A \right) \left(\mu_0 H_{\text{ext}} + B_A + \mu_S M_S \right)}, \]  

where \( \gamma \) is the gyromagnetic ratio, \( B_A \) is the effective anisotropy field and \( M_S \) is the saturated magnetization in the ferromagnetic layer. This reveals that despite no exchange bias is visible in the SQUID data (Fig. 1) and precession frequency (Fig. 3(c)), the dynamic magnetic properties of the Fe layer which are very sensitive to the original orientation of the magnetization – namely, the precession phase and amplitude (Fig. 3(d)), are affected by the presence of CuMnAs layer. Similar observation was reported also for FM/AF bilayer of Ni/FeF\(_2\) [12]. The authors observed that in the field range exceeding the exchange bias field, where the static magnetization measurements seemed to show that magnetization was aligned along \( H_{\text{ext}} \), it was not the case. The performed pump–probe experiment revealed that either the magnetization was canted or part of the FM had not been reversed by the field [12]. Similarly, in our case the direction of the pump-induced initial tilt of magnetization in Fe layer is apparently affected by the presence of an adjacent CuMnAs layer – the position of 90° phase, which corresponds to the in-plane initial tilt of magnetization, is shifted to \( -250 \) mT. The precise mechanism responsible for this behavior is not clear at present but it seems to be related to an AF-induced magnetic anisotropy and/or a different formation of magnetic domains in Fe layer at negative and positive fields. Such a difference between
“dynamic” (i.e., deduced from a pump–probe experiment) and “static” (e.g., deduced from a SQUID data) anisotropy fields were reported also for other FM/AF bilayer systems [4, 5]. To sum up, from the experimental point-of-view, our measurements show that the precession phase, which was only rarely used so far [5], is an extremely sensitive tool for a research of the exchange coupled bilayers [4–21]. Moreover, our results imply that averaging of data measured for two opposite field directions, which is sometimes used to remove the non-magnetic signals [20], should be applied only with caution in the magnetic bilayers.

The information about CuMnAs layer can be deduced from the polarization-dependent non-oscillatory MO signals which are shown in Fig. 4. For \( H_{\text{ext}} = 0 \) the fit of the data by Eq. (1) yields \( \phi = 149 \pm 2^\circ \), which shows that the spin axis in CuMnAs is located along the [110] GaP crystallographic direction (see inset in Fig. 2), in agreement with our previous observations [27]. As apparent in Fig. 4, we have not observed any sizable shift of the Néel vector \( \mathbf{L} \) in CuMnAs due to the interlayer exchange coupling with Fe when external magnetic field with a magnitude of 670 mT was applied along the directions \(+90^\circ\) and \( -90^\circ\). Similar results were obtained for several temperatures from 15 K to 300 K. In the XMLD measurements, on the other hand, a rotation of \( \mathbf{L} \) by magnetic field was observed in the same sample [28].

One possible explanation of this difference might be a different depth sensitivity of these two techniques: The XMDL experiment was performed in a reflection geometry using a total electron yield that leads to a probing depth of \( \approx 3 \) nm, which is smaller than the 5 nm CuMnAs film thickness. On the contrary, the optical pump–probe experiment is performed in transmission geometry where the whole material is probed with the same sensitivity. The uniaxial anisotropy in CuMnAs comes from the specific symmetry of bond alignments on the CuMnAs/GaP interface [27] and, therefore, the Fe-mediated rotation of \( \mathbf{L} \) might be more difficult to achieve close to the substrate than at the sample surface. Alternatively, this difference between the static and dynamic experiments might originate from the pump-induced local temperature increase of \( \approx 120 \) K [27] which is an inherent part of our MO pump–probe method. It is well-known that the exchange coupling between layers depends strongly on temperature [6]. Consequently, \( H_{\text{ext}} \) can indeed rotate the Néel vector due to the interaction with Fe in the whole CuMnAs layer but the first pump laser pulse, which heats the sample, might destroy the magnetic alignment of the Fe and CuMnAs layers. Following this event, the magnetic moments in CuMnAs would rotate towards their easy axis where they would be detected by our stroboscopic method at the original position. Finally, we cannot also exclude a possibility that the sizable pump-induced temperature increase might locally influence the interface – and consequently the exchange coupling – between Fe and CuMnAs layers in a similar way as the sample annealing does [4].

3 Conclusion

In conclusion we studied a Fe/CuMnAs bilayer using the time-resolved magneto-optical pump–probe technique. We observed an oscillatory signal due to the precession of Fe moments and a strong asymmetry in the dependence of the precession phase on the external magnetic field. This we interpret as a result of the Fe interface coupling with the CuMnAs layer that is not apparent in SQUID measurements. We also discussed possible differences between pump–probe-based MO technique and XMDL that are responsible for the fact that we did not observe any significant reorientation of the magnetic moments in CuMnAs by external magnetic field due to the interlayer exchange coupling with Fe.

Acknowledgements

The work was supported by the Grant Agency of the Czech Republic (grant no. 14-37427G), by the Ministry of Education of the Czech Republic (grant no. LM2015087) and by Grant Agency of Charles University (grants nos. 1910214 and SVV-2015-260216). P.W. acknowledges funding from the University of Nottingham Engineering and Physical Sciences Research Council (EPSRC) Impact Acceleration Account (grant no. EP/K503800/1).

References

[1] R. F. C. Farrow, R. F. Marks, S. Gider, A. C. Marley, S. S. P. Parkin, and D. Mauri, J. Appl. Phys. 81, 4986 (1997).
[2] D. H. Han, J. G. Zhu, J. H. Judy, and J. M. Sivertsen, J. Appl. Phys. 81, 340 (1997).
[3] J. Nogués and I. K. Schuller, J. Magn. Magn. Mater. 192, 203–232 (1999).
[4] J. McCord, R. Kaltofen, T. Gemming, R. Hüne, and L. Schultz, Phys. Rev. B 75, 134418 (2007).
[5] F. D. Longa, J. T. Kohlhepp, W. J. M. de Jonge, and B. Koopmans, J. Appl. Phys. 103, 07B101 (2008).
[6] X. Martí et al., Phys. Rev. Lett. 108, 017201 (2012).
[7] G. Ju and A. V. Nurmiikko, Phys. Rev. B 58, 22857 (1998).
[8] G. Ju et al., Phys. Rev. Lett. 82, 3705 (1999).
[9] G. Ju, L. Chen, and A. V. Nurmiikko, Phys. Rev. B 62, 1171 (2000).
[10] M. Djordjevic, G. Eilers, A. Parge, and M. Münzenberg, J. Appl. Phys. 99, 08F308 (2006).
[11] H. B. Zhao et al., Phys. Rev. Lett. 100, 117208 (2008).
[12] A. Porat, S. Bar-Ad, and I. K. Schuller, EPL 87, 67001 (2009).
[13] F. D. Longa, J. T. Kohlhepp, W. J. M. de Jonge, and B. Koopmans, Phys. Rev. B 81, 094435 (2010).
[14] X. Ma et al., Nature Commun. 6, 8800 (2015).
[15] T. Martin et al., J. Appl. Phys. 103, 07B112 (2008).
[16] Z. Zhang et al., Appl. Phys. Lett. 97, 172508 (2010).
[17] R. Adam, P. Grychtol, S. Cramm, and C. M. Schneider, J. Electron Spectrosc. Relat. Phenom. 184, 291 (2011).
[18] D. Rudolf et al., Nature Commun. 3, 1037 (2012).
[19] S. Mizukami et al., J. Appl. Phys. 115, 17C119 (2014).
[20] I. Razdolski et al., Phys. Status Solidi RRL 9, 583–588 (2015).
[21] Yu Tsema et al., Appl. Phys. Lett. 109, 172403 (2016).
[22] P. Wadley et al., Nature Commun. 4, 2322 (2013).
[23] P. Wadley et al., Sci. Rep. 5, 17079 (2015).
[24] E. V. Gomonay and V. M. Loktev, Low Temp. Phys. 40, 17 (2014).
[25] T. Jungwirth, X. Marti, P. Wadley, and J. Wunderlich, Nature Nanotechnol. 11, 231 (2016).
[26] P. Wadley et al., Science 351, 587 (2016).
[27] V. Saidl et al., Nature Photon. 11, 91 (2017).
[28] P. Wadley et al., arXiv:1702.03147 (2017).
[29] A. K. Zvezdin and A. V. Kotov, Modern Magnetooptics and Magnetooptical Materials (Taylor & Francis, New York, 1997), chaps. 1, 3.
[30] N. Tesarova et al., Appl. Phys. Lett. 100, 102403 (2012).
[31] C. Kittel, Introduction to Solid State Physics – 8th edition (Wiley, New York, 2005), p. 381.
[32] Z. Frait and R. Gmperle, J. Phys. Colloques 32, C1–C54 (1971).