Ultrafast subpicosecond magnetization of a two-dimensional ferromagnet

Controlling the magnetization of ferromagnets on a subpicosecond (sub-ps) timescale is highly desirable, although challenging, because it enables the operation of new ultralow-power spin-based electronic devices at THz frequencies [1,2], which are beyond the limitations of the present CMOS circuitry. To date, ultrafast spin dynamics studies have generally employed femto-second (fs)-pulse lasers with strong fluences to excite magnetic materials [2]. When the photon energy coincides with a resonant transition between different bands, the laser pulses excite carriers into *d*- or *f*-orbital spin-polarized bands, causing instant magnetization enhancement. However, unlike optical pumping, electrical gating, the basic operation in CMOS, cannot significantly change the carrier density or cause band-to-band transitions in the ultrafast timescale.

In this study, we demonstrate a new scheme for sub-ps magnetization manipulation, called wavefunction (WF) engineering, which requires no change in the total carrier density, using the quantum well (QW) structure of a ferromagnetic semiconductor (FMS) (In,Fe)As [3]. FMSs are alloy semiconductors doped with a significant amount of magnetic elements (several %; Mn, Fe, etc.) that play the role of local spins. These local spins (*d* electrons) strongly couple to itinerant carriers (*s* or *p* electrons) via *s,p-d* exchange interactions, which mainly contribute to the establishment of ferromagnetism in FMSs. Among the various FMSs, (In,Fe)As stands out as the first carrier-induced n-type FMS, where electron carriers possess coherence lengths as long as 40 nm [3]. The sample examined in our study consists of (In,Fe)As (10 nm, 8% Fe)/InAs (5 nm)/AlSb (300 nm)/AlAs (5 nm)/GaAs grown on a semi-insulating (SI) GaAs (001) substrate via molecular beam epitaxy (Fig. 1(a)). In this structure, the electron carrier WFs are extended throughout and well confined in the top bilayer (Fig. 1(b)). The magnetic properties of this bilayer QW, such as the Curie temperature (T_C), are determined by the spatial overlap between the WFs and local spins (Fe) [3]. Thus, by controlling the WF peak position and shape, the magnetic properties of (In,Fe)As/InAs QWs can be effectively changed without pumping additional carriers into the system.

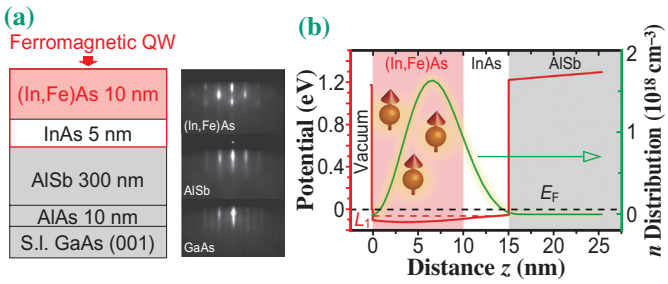

Fig. 1. **(a)** Schematic of the sample structure. **(b)** Conduction band bottom profile (red curve) and electron carrier distribution (green-yellowish curve) in the (In,Fe)As/InAs QW.

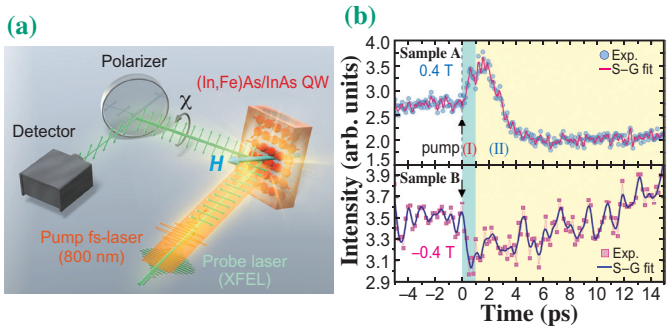

Fig. 2. **(a)** Pump-probe measurement setup. **(b)** Time evolution of XFEL intensity for samples A (top) and B (bottom), which reflects the rapid change in the magnetization **M**. Circle and square: raw data; lines: data smoothened using a Savitzky–Golay (S–G) filter.

To study the spin dynamics in the (In,Fe)As/InAs QW, we conducted pump-probe measurements at SACLA **BL1**. We used a fs-pulse laser (wavelength = 793 nm; pulse width = 30 fs) to excite the *s,p*-electron bath close to the Fermi surface. To probe the magnetization of the (In,Fe)As/InAs QWs, we conducted X-ray magneto-optical Kerr effect measurements, using a linearly polarized X-ray free electron laser (XFEL) beam with an energy of 52 eV, which resonates with the *M* absorption edge of Fe [4]. As illustrated in Fig. 2(a), the Kerr rotation angle, θ_K, of the XFEL beam after being reflected from the (In,Fe)As/InAs sample was experimentally determined using rotating analyzer ellipsometry. For the time-resolved measurements, the analyzer angle was fixed, and the detected XFEL intensity provides a measure of θ_K with time. Upon pumping sample A with the infrared laser pulse, the detected XFEL intensity instantly increased (from 2.8 to 3.4) on a timescale of 600 fs (region I), and then slowly saturated and decreased in the next several ps (region II) (top panel, Fig. 2(b)). These results indicate sub-ps enhancement of the magnetization **M** of the (In,Fe)As/InAs QW in sample A. For comparison, similar experiments were conducted on sample B, where **M** points in the opposite direction (bottom panel, Fig. 2(b)). The opposite signs of the

changes in the XFEL intensities for samples A and B prove that the changes originate from the evolution of the magnetization **M** of the (In,Fe)As/InAs QW. The observation of the ultrafast magnetization of the (In,Fe)As/InAs QW (~600 fs) is the first sub-ps enhancement of **M** of an FMS ever reported [5].

Next, we discuss the mechanism of the sub-ps magnetization. Upon the arrival of the pump laser pulse, photoelectrons and photoholes are instantly generated (~1 fs), which have an exponentially decreasing distribution from the surface toward the substrate side (right panel, Fig. 3(a)). However, these photoelectrons reside in bands 1.1 eV higher than the conduction band bottom and thus cannot instantly interact with the Fe spins and cause the sub-ps magnetization. Owing to the density gradient along the *z*-axis, these photocarriers start diffusing toward the substrate side, which is much faster for the photoelectrons than for the photoholes because of their higher mobility and higher temperature excited by the pump laser. Consequently, there are more photoholes at the surface and more photoelectrons at the substrate side, and an electric field pointing from the surface to the substrate side rapidly builds up over time. This is called the photo-Dember field. In the (In,Fe)As/InAs QW, the photo-Dember field likely pushes the 2D electron WF toward the top (In,Fe)As layer, leading to enhanced magnetization in the QW. Our numerical calculations confirm this scenario. As shown in Fig. 3(b), the deviation between the local densities of the photoelectrons $N_{pe}(z,t)$ and photoholes $N_{ph}(z,t)$ rapidly increased at every time step of 100 fs. Consequently, over time, the (In,Fe)As/InAs QW potential deepened at the center of the (In,Fe)As layer (see Fig. 3(c)), causing a redistribution of the 2D electron carriers, $N_{2D}(z,t)$, in the (In,Fe)As region (Fig. 3(d)) and a consequent enhancement of T_C from 17 to 31 K (Fig. 3(e)) [5].

The ability to control the magnetization of a magnetic material on the sub-ps timescale is a remarkable feature of a ferromagnetic QW, utilizing the high coherency of itinerant carriers. Schemes for controlling the carrier WFs are not limited to optical pumping, and this control can also be performed by applying a gate voltage to the transistor structures. The electrical control of ferromagnetism using WF engineering may potentially lead to ultrafast and scalable electronic devices in the future.

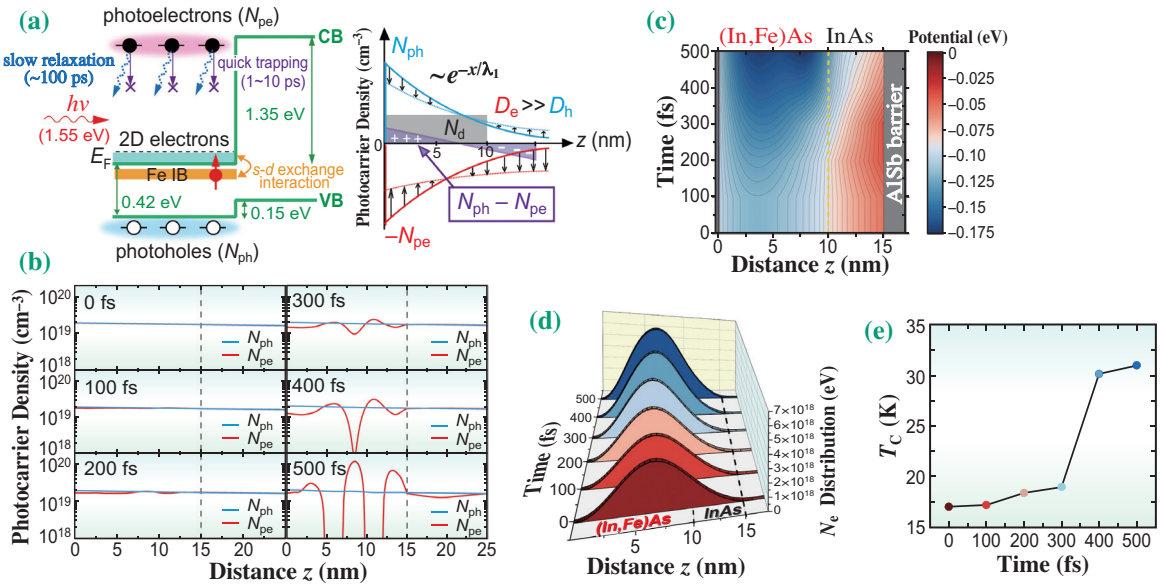

Fig. 3. **(a)** Generation and redistribution of photocarriers (holes, electrons), which build up a photo-Dember electric field. Our calculations confirmed the ultrafast (sub-ps) redistribution of **(b)** the photocarriers, **(c)** potential profile, **(d)** 2D electron density distribution, and **(e)** T_C of the (In,Fe)As/InAs QW.

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