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Magnetic circular dichroism photoemission electron microscopy using laser and threshold photoemission

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Abstract
We demonstrate that laser induced valence band photoemission can be used in the observation of magnetic domain structures with a magnetic circular dichroism photoemission electron microscope (MCD-PEEM). It has been widely considered that valence band photoemission MCD asymmetry is rather small compared to that obtained with x-rays because of its weak spin–orbit coupling. However, we show that the MCD asymmetry is high near the photoemission threshold, permitting us to perform MCD-PEEM experiments. The use of intense and pulsed lasers as excitation sources enables PEEM studies of two-photon photoemission MCD and all optical time resolved MCD.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Magnetic circular dichroism (MCD) is now an indispensable technique for the investigation of thin film magnetism. MCD in the core level is widely studied using x-rays [1] in synchrotron radiation facilities. On the other hand, MCD in valence states is known as the magneto optical Kerr effect (MOKE) and is an important tool for the investigation of magnetism in laboratories [2] because widely available light sources, like CW lasers or discharge lumps, make the MOKE experiment possible and its experimental setup is simple. However, the spatial resolution of MOKE is limited by the diffraction limit of the light. With photoelectron emission, the valence band MCD combined with electron microscope may give better spatial resolution. Its sensitivity to magnetism, however, is too low to investigate the microscopic magnetic structure of thin films except for angle and energy resolved photoemission [3–5], which is not suitable for microscopic measurements. Thus it was believed that valence band photoemission electron microscopy (PEEM) combined with MCD was not available, and one can find very few examples of magnetic linear dichroism on thicker Fe film (∼100 nm) [6]. Recently we have shown that valence band MCD integrated in angle and energy gives high asymmetry near the photoemission threshold [7], which is suitable for MCD-PEEM measurements [8].

PEEM experiments with pulsed lasers have been used for the investigation of multiphoton photoemission processes and time resolved electron motion. Recently the two-photon photoemission (2PPE) experiment on nanostructures has been attracting great interest because it has been shown that the strong laser field enhances photoemission probabilities from nanostructures because of the localized surface plasmons and modification of the local electromagnetic field [9, 10]. 2PPE-PEEM gives fruitful information about nanostructures [11]. For time resolved PEEM experiments in the femtosecond region, there are several pioneering works about electron motion on the surface [12] and the decay of hot electrons depending on the heterogeneity of the sample [13]. On the other hand, there have been no reports of 2PPE MCD-PEEM or all optical time resolved MCD-PEEM.

In this paper, we show that photoemission from magnetic ultrathin films by circularly polarized lasers enables microscopic imaging. 2PPE-MCD is also used for magnetic domain imaging. Moreover, using a femtosecond laser we show that time resolved imaging with sub-picosecond time

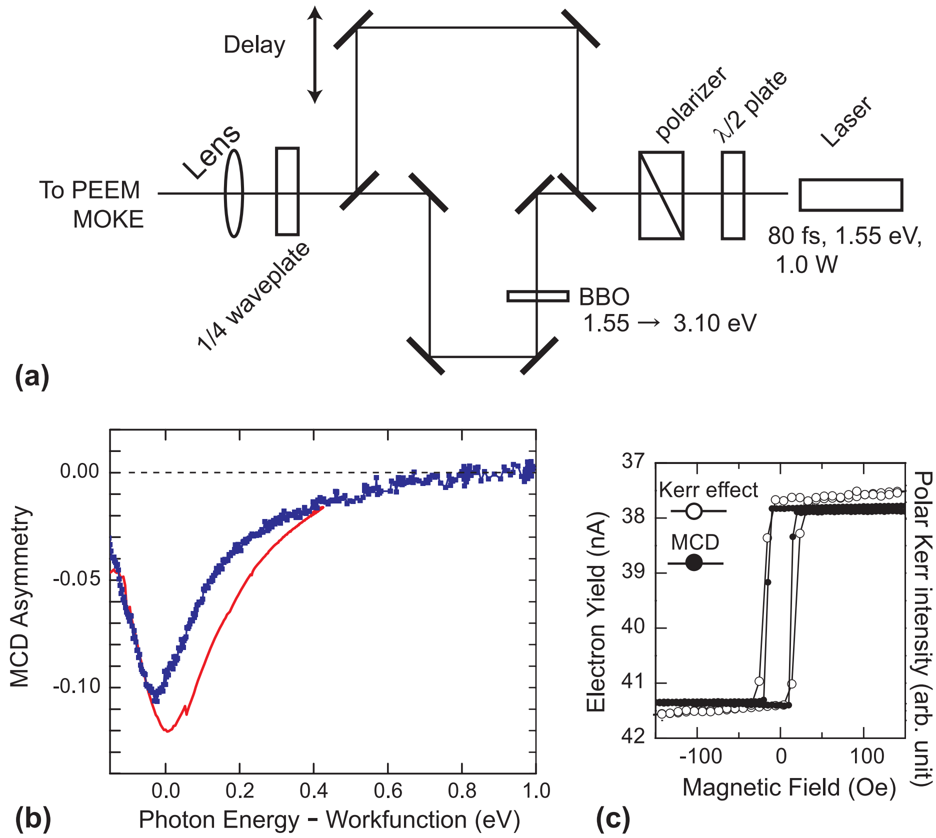

Figure 1. (a) A schematic drawing for the experimental setup. A femtosecond laser is used for two-photon photoemission and time resolved experiments. (b) Photoemission MCD asymmetry from Cs/Ni(12 ML)/Cu(001) as a function of the photon energy with the work function subtracted. The dots (blue) and line are taken with a fixed photon energy of 3.81 eV and 1.95 eV, respectively. The work function is varied by Cs deposition. The asymmetry is defined in the text. (c) Magnetization curves by laser photoemission MCD (filed circles) and the magneto optical Kerr effect (open circles). Parts (b) and (c) are taken from [7].

resolution can give magnetization changes on a microscopic scale.

2. Experimental details

All the experiments were performed in an ultrahigh vacuum (UHV) chamber equipped with a PEEM (PEEM Spector, Elmitec) apparatus. A Cu(001) crystal was cleaned by Ar ion sputtering and annealing at 825 K. The order and cleanliness of the surface was verified by sharp low energy electron diffraction (LEED) spots and Auger electron spectroscopy (AES). Ni was deposited from an electron beam evaporator, and Ni coverage was calibrated using reflection high energy electron diffraction oscillation. The sample used in all the present experiments was 12 monolayer (ML) Ni film on Cu(001), which shows perpendicular magnetization.

We used a polar configuration for the MCD measurements, and the angle between the sample normal and the incident light path was 0°. For the PEEM measurements, the sample was as prepared and the angle between the sample normal and the incident light path was 55°. To change the work function of the sample, the surface was covered by Cs, with a coverage of less than 0.3 ML [14]. Cs was deposited from a commercial source (SAES Getters) onto Ni/Cu(001). The prepared Cs/Ni/Cu(001) showed a change in the time dependent work function. To prevent the work function changing during the measurements, the sample was intentionally left in a 10^{-10} Torr vacuum overnight. After this procedure the work function was stable for several days. The optical setup for the PEEM measurement is schematically shown in figure 1. We used a femtosecond Ti:sapphire oscillator (1.55 eV, 80 fs, 1.0 W) and CW lasers (635, 400 and 325 nm) with a power of the order of a few mW. For the PEEM experiment we obtained circularly polarized beams by quarter-wave plates, and MCD images were measured by the difference between the images from right and left circularly polarized beams. The MCD asymmetry A is defined as $A = (I_{\mathrm{left}} - I_{\mathrm{right}})/(I_{\mathrm{left}} + I_{\mathrm{right}})$, where I_{right} (I_{left}) represents the intensity of the right (left) circularly polarized beams.

Time resolved PEEM measurements were done using the femtosecond laser. The output of the oscillator (1.55 eV) was split into two beams and half was used as a pump and the other half was frequency doubled (3.10 eV) by a BBO crystal and used as a probe. A mechanical delay stage was used to control the timing between the pump and probe pulses. The temporal overlap point of the pump and probe was confirmed by using sum-frequency generation of the pump and probe beams. The pump and probe beams are coaxially overlapped by a beam splitter and focused into the UHV chamber. The spatial overlap of the two beams on the sample surface was achieved by the

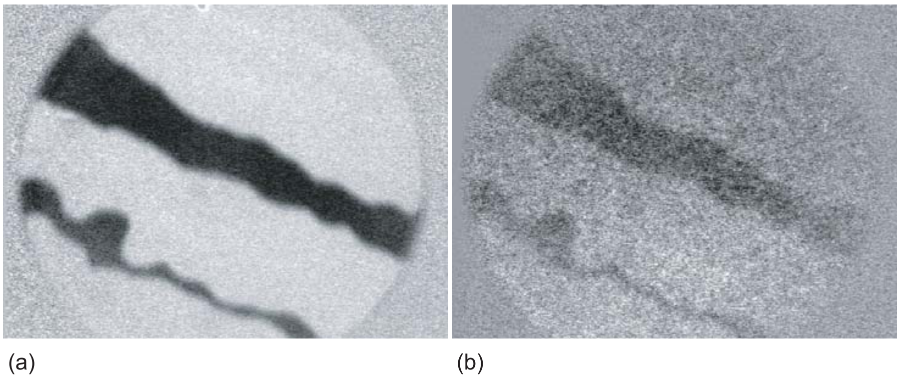

Figure 2. (a) MCD image for 4 s integration time for each circularly polarized light source. (b) The same as (a), but the integration time is 1/30 s. The field of view is 100 μm.

following procedure: first the pump laser position was verified by the generation of 2PPE on the PEEM image, then the position of the probe beam was adjusted onto the same area.

3. Results and discussion

3.1. Magnetic circular dichroism in the valence band

Figure 1(b) shows the result of photoemission MCD using 1.95 and 3.81 eV lasers. With continuous Cs deposition, the sample work function decreases from ~5 eV, resulting in photoemission. The MCD asymmetry is plotted as a function of the photon energy with the work function subtracted, meaning the electron kinetic energy. In this experiment the photon energy is constant while the work function is changed. Around the photoemission threshold, the MCD asymmetry is large (~10%). In figure 1(c), we show a hysteresis curve measured near the threshold photoemission using the MCD method, together with a hysteresis curve taken with the conventional MOKE. The two hysteresis curves are almost identical, confirming the validity of the photoelectron method. With the photon energy fixed ($h\nu = 3.81$ or 1.95 eV) and the work function varied by Cs deposition, the absolute values of the MCD asymmetry decrease as the work function decreases, approaching zero. The drastic decrease in the MCD asymmetry reveals that the present photoemission MCD method effectively acquires magnetic information near the threshold. Away from the threshold, the MCD asymmetry is low, although the number of emitted photoelectrons increases.

3.2. Application of threshold MCD to PEEM

The MCD asymmetry near the photoemission threshold is found to be high enough to measure the MCD-PEEM. Figure 2 shows the MCD-PEEM results for Cs/Ni(12 ML)/Cu(001). The light source is a 405 nm CW laser of 1 mW (2×10^{15} photons s^{-1}). Figure 2(a) shows the MCD image with 4 s accumulation, giving 4% asymmetry. With this accumulation time, magnetic domains are clearly observed. In this image we can see stripes across the surface, corresponding to the up and down magnetized domains. Figure 2(b) shows the MCD image taken in the same area as figure 2(a) with 1/30 s accumulation time for each circularly polarized light source. The magnetic domain is still visible. The rapid acquisition of MCD images is achieved owing to large photon flux of the laser.

The high brilliance of laser induced MCD-PEEM means that it is possible to measure magnetic domains at a video rate. This is easily done with CW lasers, but when the light source is a pulsed laser the situation is somewhat different. A MCD-PEEM measurement with a pulsed laser is shown in figure 3. The laser light is frequency-doubled light ($h\nu =$ 3.1 eV, 100 fs) from a Ti:sapphire laser. The beam spot size is $\phi \sim 200$ μm. Figure 3(a) shows the MCD-PEEM images for different laser powers. The MCD image shows a clear domain structure with low incident laser power (0.2 mW). With increasing laser power, the domain structure begins to distort. With an input power of 7.8 mW, the MCD image is obscure compared with that obtained with a power of 0.2 mW. Figure 3(b) plots the MCD asymmetry across the images. With 0.2 mW, the domain boundary is sharp, but above 4 mW the domain boundary width increases. Note that the asymmetry (~4%) is almost independent of the incident power used. This indicates that the diffused domain image at high laser power does not originate from laser heating. Figure 3(c) shows the domain boundary width as a function of the incident laser power. The plot in figure 3(c) reveals that the boundary width is linearly dependent on the laser power, especially for higher powers. Repulsion between neighboring emitted electrons shifts the electron trajectory when the electron density is high. This space charge affects the spatial resolution and linearly depends on the emitted electron current density [15]. The linear dependence of boundary broadening for higher laser power in figure 3(c) is consistent with this concept. In this experiment, the typical emitted electron current is 1 nA for 1 mW laser power. Considering the pulse width (100 fs) and the beam diameter ($\phi \sim 200$ μm), the electron current corresponds to 0.4 A cm^{-2}. However, the theory for this current density indicates that the deterioration of the resolution is 0.3 μm [15], while it is 2 μm in our experiment. Although a linear dependence on laser power is observed, other factors should be taken into account. One factor could be localized photoemission near the surface, which will increase the current density per volume.

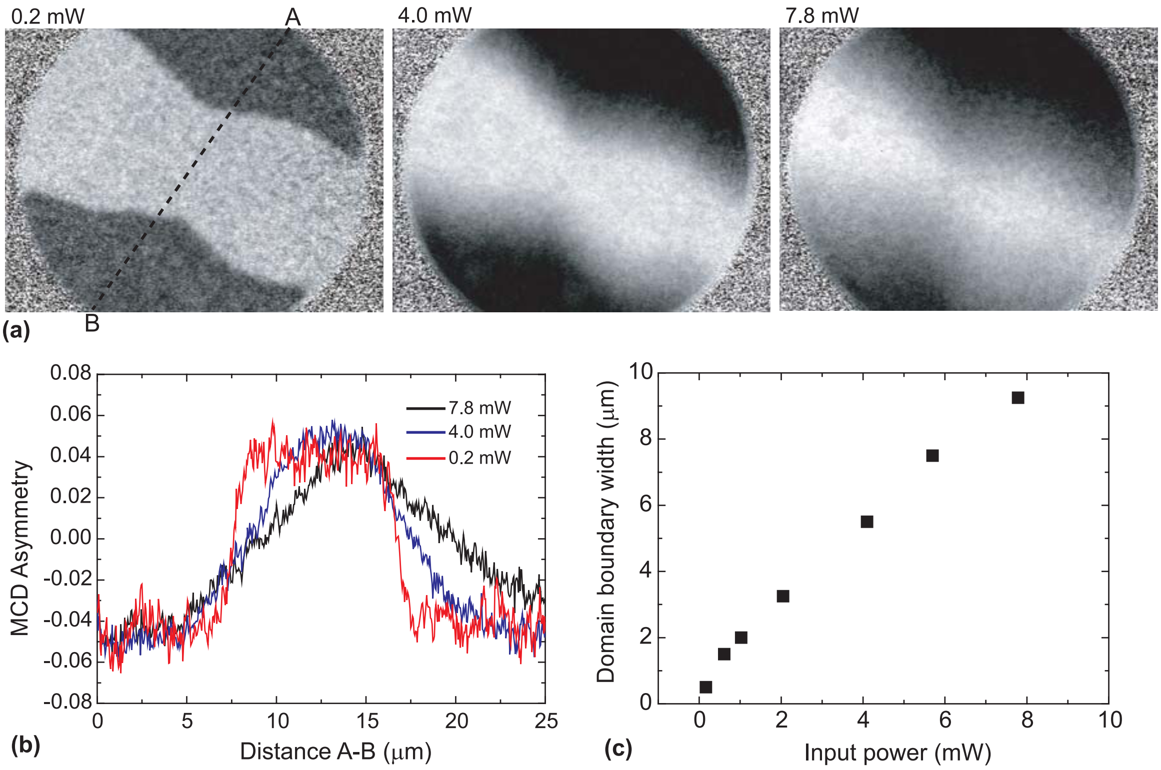

Figure 3. MCD-PEEM measurements by a femtosecond laser with a photon energy of 3.1 eV. (a) MCD images for increasing laser power. With increasing laser power, the domain structure becomes blurred. The beam spot used is ∼200 μm. The field of view is 25 μm. (b) Cross section line profiles along A–B in (a) are plotted. (c) Observed domain boundary width as a function of incident laser power.

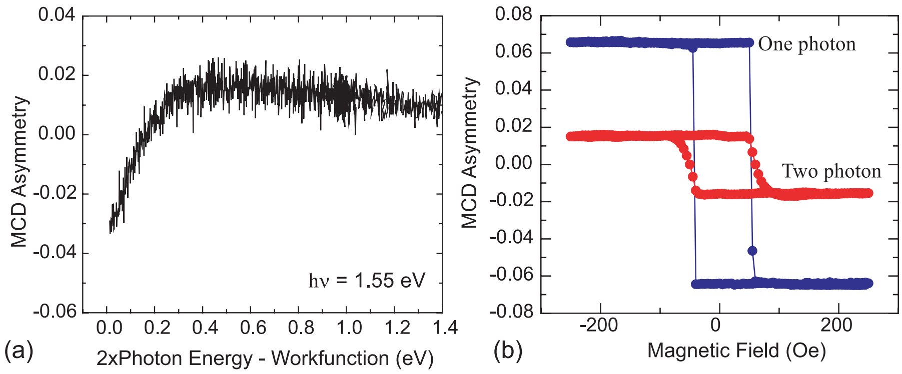

Figure 4. (a) Two-photon photoemission MCD asymmetry as a function of the photon energy subtracted by the work function. The sample is Cs/Ni/Cu(001) and the photon energy is 1.55 eV. (b) A magnetization curve obtained by one- and two-photon photoemission processes. One-photon photoemission measurement is performed by a 3.1 eV laser.

3.3. Two-photon photoemission

A possible application of pulsed laser MCD is in the study of multiphoton processes. Here we show a 2PPE MCD. It is noted that the 2PPE process in magnetic circular dichroism has not been investigated experimentally and only a theoretical work is available [16]. Figure 4(a) shows the 2PPE-MCD result. This plots the 2PPE-MCD asymmetry as a function of twice the photon energy with the work function subtracted, like the plot in figure 1(b). The photon energy used is 1.55 eV, and the work function is varied using Cs deposition. When the work function decreases from ∼5 to 3.1 eV, electrons begin to be emitted. All the photocurrent measured is only from the 2PPE process, but on decreasing the work function close to 1.55 eV, which is close to the lowest work function obtained by Cs deposition, a strong one-photon process is involved. Thus the kinetic energy in this measurement is limited within ∼1.55 eV. The MCD asymmetry is large near the threshold, giving 3% asymmetry, which is smaller than that obtained in the one-photon process. With decreasing work function, the asymmetry approaches zero and reverses its sign. Finally the 2PPE-MCD asymmetry is around 1%, which is larger than that obtained for the one-photon process. The hysteresis curves obtained by 2PPE near the threshold are plotted in figure 4(b), together with that for the one-photon photoemission. Although the MCD asymmetry for 2PPE is smaller, the shape of the

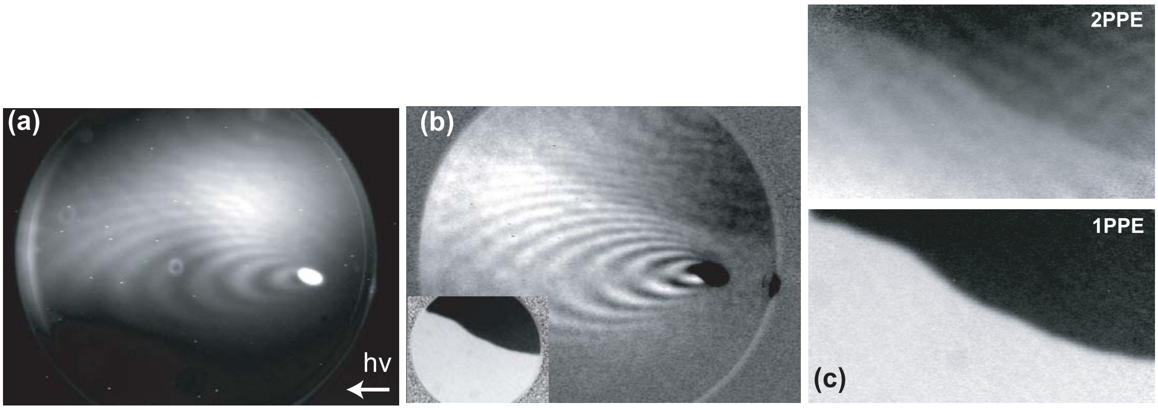

Figure 5. (a) 2PPE microscope image of Cs/Ni/Cu(001) showing a hot spot and intensity oscillation. The beam is circularly polarized, and the photon energy is 1.55 eV. The arrow indicates the beam impact direction. (b) A 2PPE-MCD image in the same region as (a). Magnetic domains are visible behind the strong oscillation and hot spot. The inset shows the MCD-PEEM image from one-photon photoemission ($h\nu = 3.1$ eV) in the same region. The field of view is 25 μm. (c) Contrasted 2PPE (top) MCD image away from the hot spot and the 1PPE (bottom) one in the same area.

hysteresis curve is almost identical, confirming the validity of the 2PPE-MCD.

An application of 2PPE to MCD-PEEM is demonstrated in figure 5. Figure 5(a) shows a 2PPE-PEEM image from a circularly polarized laser. A spot on the right side of the image is extremely bright; this is known as a hot spot in strong photon radiation [10]. Although we do not have any clear evidence, the hot spots are scattered on the surface with a substantial density, hindering observation of the magnetic domains. The hot spot is followed by intensity oscillation, which also interferes with the magnetic domains. This kind of hot spot is always seen in strong laser field, but disappears in the one-photon process. The exclusive response to the strong laser rejects the possibility that the hot spot come from spatially distributed places with lower work function, but the hot spot may come from a structural effect with localized electromagnetic field enhancement.

A MCD-PEEM image by 2PPE is shown in figure 5(b), measured at the same place as figure 5(a). We observe the hot spot and oscillation as well as domains in the differential image. The hot spot and oscillation depend on I^n, where I is laser power and $n > 2$. Thus, slight instability of the laser pointing or the laser power changes the intensity of the hot spot and oscillation in the PEEM image much more than that of the normal 2PPE process. This makes it difficult to cancel out the hot spot and oscillation between the right and left circularly polarized beams. We believe that the differential intensity of the hot spot and oscillation in figure 5(b) is not intrinsic but artificial, due to the intensity fluctuation between the images for the right and left circularly polarized beams. Behind the large intensity difference, a magnetic domain structure is observed by 2PPE-MCD. This domain structure is confirmed by one-photon MCD-PEEM as shown in the inset in figure 5(b). The one-photon MCD image is free of the hot spot and its accompanying oscillation, giving a clear magnetic domain structure. The 2PPE-MCD is observable, but it is overlapped with strong hot spots. Figure 5(c) is a contrasted image, measured away from the hot spot, which shows the magnetic domain clearly but with slight oscillating background. This background may be reduced with a better quality sample free of nanostructures or impurities. Note that the domain boundary widths for the 1PPE and 2PPE MCD-PEEM are the same within experimental error. On the other hand, the high sensitivity to the nanostructure by 2PPE [9, 10] may open a new method for magnetic nanostructure studies using laser 2PPE-MCD-PEEM.

3.4. All optical time resolved MCD-PEEM

Finally we demonstrate a time resolved MCD-PEEM experiment using a femtosecond laser. The sample used was Cs/Ni(12 ML)/Cu(001). The pump and probe power were $\sim$2 nJ and $\sim$0.01 nJ, respectively. Figure 6 shows a pump–probe image obtained at 0 ps delay, showing magnetization domains perpendicular to the surface. With the pump power in this experiment, no change in domain structure was observed during the optical pumping, and the domain wall position and domain boundary width were the same within our experimental resolution. However, the MCD asymmetry averaged over the whole image was changed by the pumping. Figure 6(b) shows the averaged MCD asymmetry as a function of delay time. To construct figure 6(b) a histogram distribution of the MCD asymmetry in a snapshot image is converted into a gray scale bar, and then the bar is arranged in the sequence of delay time. The white positions at the positive and negative asymmetries correspond to the asymmetry for the up and down magnetized domains. The asymmetry is 4% for no pump. The asymmetry decreases near 0 ps to 3%, and after 5 ps it returns to the value without pumping.

Previous MOKE experiments on perpendicularly magnetized Ni/Cu(001) have shown that the Kerr ellipticity and rotation angle steeply decrease within 1 ps and they begin to oscillate at 200 ps [17–19]. The rapid reduction between 0.5 and 1.0 ps is attributed to a heating induced increase in electron temperature, and the 200 ps oscillation is a spin precession. Our experiment observed only a rapid reduction of the MCD asymmetry, which can be attributed to the increase in electron temperature. The spin precession by optical pumping

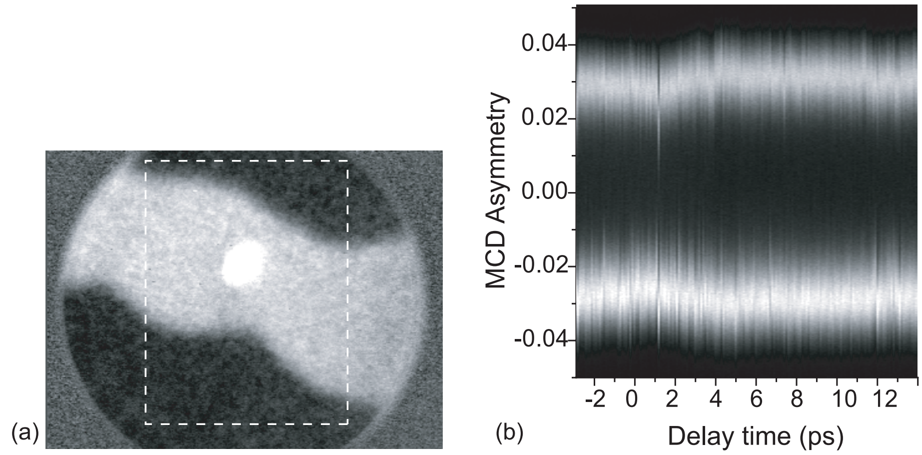

Figure 6. Time resolved MCD-PEEM experiment. The photon energies of the pump and probe beams are 1.55 eV and 3.1 eV, respectively. (a) A MCD-PEEM image for ($\delta t = 0$) ps. The field of view is 25 μm. (b) A gray scale image of the MCD asymmetry as a function of the delay time between the pump and probe beams. The number of the histogram representing the frequency of the asymmetry is converted into the gray scale image. The frequency increases from black to white. The up and down magnetized domains corresponds to the white regions at positive and negative asymmetries. The asymmetry only inside the dashed lines in (a) is converted into the image.

is not resolved in this TR-PEEM measurement. This may be because the asymmetry change by the precession is one order of magnitude smaller than that by electron temperature increase. The reduction of the asymmetry observed by TR-PEEM is ~30%, which is larger than the change of the Kerr ellipticity or rotation angle (~10%). One possible reason for this difference is the difference of the pump power used in these measurements. Another reason is the different processes involved between these measurements. Threshold MCD-PEEM observes only the electrons near the Fermi level, while MOKE observes all the excited electrons. Since the electrons closer to the Fermi level are more sensitive to the change of the magnetization, the threshold photoemission MCD possibly shows an enhanced change compared with MOKE.

4. Conclusions and outlook

We have demonstrated that laser induced threshold photoemission can be applied to the study of magnetic domain structures on ultra thin films. Like one-photon MCD, 2PPE-MCD also gives high asymmetry near the photoemission threshold, which lets us observe the 2PPE-MCD-PEEM. Laser MCD can be used as a new method for investigating magnetic microstructures and their ultrafast response to optical excitation. Since threshold photoemission is necessary for this method, an energy tunable laser is an ideal light source. With tunable lasers, it is feasible to enhance the MCD asymmetry for PEEM imaging, which is currently difficult because the work function changes with alkali metal deposition.

Acknowledgments

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