Investigation of ultrafast electron dynamics of nickel film and micro-nano-structure film

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The electron thermalization and relaxation processes in ferromagnetic nickel thin film and micro-nano-structure film have been studied by measuring the transient change after excitation by a femtosecond laser pulse. The measurements indicate that the electron thermalization time is between 18 and 47 fs. This is somewhat faster than the value reported before. And the thermalization time of the micro-nano-structure film is much longer than the nickel film. We deduce that it is caused by the discontinuity of the electron band close to the Fermi level in the micro-nano-structure nickel film.

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Nickel, as a typical ferromagnetic 3*d* transition metal, is widely used in the magnetic memory industry. The research of ultrafast magnetization dynamics of nickle thin film and film with micrometer and nanometer structure becomes more necessary with the technology development. The femtosecond optical pump-probe technique is an ideal tool to investigate the ultrafast dynamics[1−7]. It has recently been used to study the demagnetization dynamics[2−5]. In 1996, Beaurepaire *et al.* reported the demagnetization time of 260 fs[2]. And In 1997, Hohlfeld *et al.* reported that the demagnetization time was 280 fs[3]. While Zhang *et al*. suggested an intrinsic time scale for demagnetization of just 10 fs by theoretical study[8]. These reports imply that demagnetization occurs within the time taken for the hot electrons to thermalize. It has been suggested that spin angular momentum is removed from the electron system through the combined action of electron-electron interactions, spin-orbit coupling, and optical field supplied by the laser pulse. So it is difficult to distinguish the real demagnetization time. Besides, much less is known about the electron dynamics in ferromagnetic transition metals. And a detailed study of the ultrafast electron dynamics in 3*d* metals is considered a very relevant issue to understand the dynamics in metal systems and nanometer metals. It also means great to interpret the sub-picosecond magnetization dynamics. In this letter, we use the optical pump-probe technique to investigate the electron thermalization dynamics of nickel film and the film with micro-nano-structure.

The interaction of femtosecond laser with metal film is a very complex process[9]. Upon absorption of a short laser pulse, electrons are excited from the occupied states below Fermi level to the empty states above. The optical excitation creates a non-equilibrium distribution of highly energetic electrons, with energies ranging up to the photon energy above the Fermi level. The lattice, on the other hand, is initially undisturbed. In the subsequent relaxation processes, the energetic electrons thermalizie to a Fermi-Dirac distribution by electron-electron (e-e) scattering and the lattice is heated by electron-phonon (e-p) scattering.

The ultrafast process after short laser excitation have been investigated by theory[10]. Combined with two-temperature model, the transient electron temperature can be described by the empirical relation[11,12]:

$$\Delta T_{\rm e}(\Delta t) = \Delta T_1[1-\exp(-\Delta t/\tau_{\rm th})]\exp(-\Delta t/\tau_{\rm E}) + \Delta T_2[1-\exp(-\Delta t/\tau_{\rm E})], \quad (1)$$

where $\tau_{\rm th}$ is the characteristic thermalization time, $\tau_{\rm E}$ is the characteristic relaxation time, ΔT_2 is the temperature rise after e-p relaxation, and ΔT_1 describes the peak temperature rise.

According to this, the electron reaches its maximum value at the time[10]

$$t_{\rm ex} = \tau_{\rm th}\ln[\frac{\Delta T_1(\tau_{\rm E}+\tau_{\rm th})}{(\Delta T_1-\Delta T_2)\tau_{\rm th}}]. \quad (2)$$

Equation (2) shows the dependence of $t_{\rm ex}$ on $\tau_{\rm E}$ and $\tau_{\rm th}$.

The samples used in the experiment were Ni films and the micro-nano-structure Ni films. The Ni films were prepared by magnetron sputtering technique in the K575X Peltier cooled high resolution sputter coater. The K575X coater can also monitor the thickness of the film. So we prepared a series of samples: Si/Ni(20 nm), Si/Ni(190 nm), glass(K9)/Ni(20 nm), and Si/Ni (100 nm)/Au(5 nm)/Ni(10 nm). After that, the Si/Ni samples were etched by the ammonia etching technique in

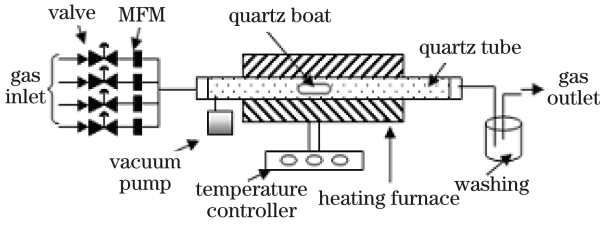

Fig. 1. Schematic diagram of experimental equipment for amonia etching. MFM: mass flowmeter.

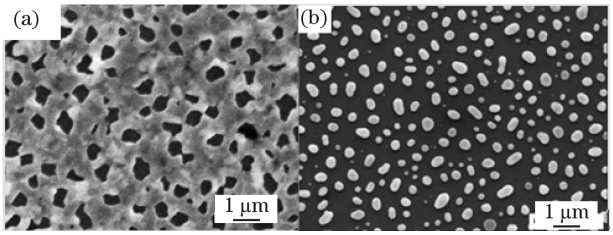

Fig. 2. SEM photographs for the two samples. (a) Si/Ni (100 nm)/Au(5 nm)/Ni(10 nm) sample; (b) Si/Ni(20 nm) sample.

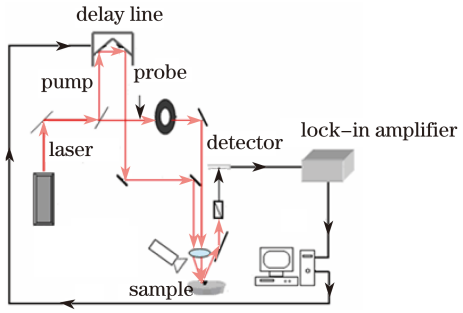

Fig. 3. Experimental sketch of the pump-probe system.

thermal chemical vapor deposition (CVD). A schematic diagram of the equipment is shown in Fig. 1.

The micro-nano-structure films were prepared through the following process. The Si/Ni(20 nm) and Si/Ni (100 nm)/Au(5 nm)/Ni(10 nm) samples were transferred in air to the growth quartz tube. The tube was pumped down to a base pressure of 1.33 Pa using a rotary pump, and then 2.7−4.0 kPa of high purity hydrogen was introduced via a leak valve. The substrates were heated to 600 °C and this temperature was kept for 15 min to reduce the catalyst layer. Then the temperature was raised to 800 °C and ammonia was introduced into the chamber at 100 sccm (sccm denotes cubic centimeter per minute at standard temperature and pressure) for 10 min in order to etch the catalyst films to form nano-sructure. After that, the two samples were observed by scanning electron microscope (SEM). Figure 2 shows the SEM photographs of the two samples.

As we can see from Fig. 2, the micro-nano-particles appear on both samples. For the Si/Ni(100 nm)/Au(5 nm)/Ni(10 nm) sample, only the surface Ni layer (10 nm) was etched. The 100-nm Ni layer was not influenced for the existence of Au film. For the Si/Ni(20 nm) sample, some part of the Ni film was totally etched, so the Si substrate emerged on that part. And the emerging Si substrate will influence the signal greatly in the pump-probe experiment. So it is not proper to investigate the ultrafast electron dynamics on this sample. We only chose the etched Si/Ni(100 nm)/Au(5 nm)/Ni (10 nm) sample as the micro-nano-struture sample for pump-probe experiments.

We used the optical pump-probe technique to investigate the ultrafast electron dynamics of the prepared samples. The experiment sketch is shown in Fig. 3. The Ti:sapphire laser supplied 30-fs pulses at a repetition rate of 82 MHz. All measurements were performed at a wavelength close to 790 nm. These pulses were divided into two parts with the probe beam passing through an optical delay line in order to control its arrival time at the

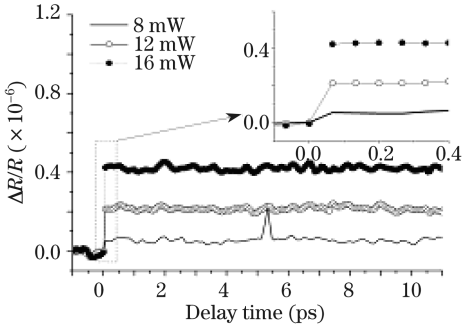

Fig. 4. Transient reflectivity changes for the glass(K9)/Ni (20 nm) sample with the pump power of 128 mW and the probe power of 8, 12, and 16 mW.

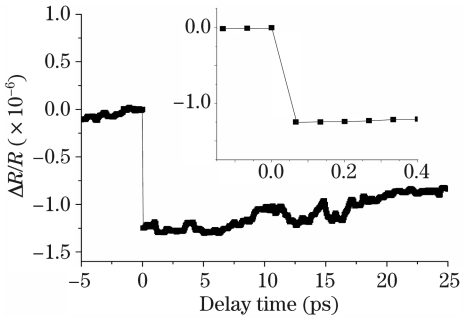

Fig. 5. Transient reflectivity changes of sample of Si/Ni(190 nm) with the pump power of 128 mW and the probe power of 8 mW.

sample. The minimum step size of the variable delay was 66.7 fs. The pump beam was modulated at 2000 Hz. The two parts of pulses were both focused on the sample by a convex lens. The spot diameter of pump beam was 100 μm. In order to keep the both beams fully overlapped, the spot diameter of the probe beam was about 50 μm. Then the probe beam was properly explored the homogeneity of electron dynamics.

Figure 4 shows the transient reflectivity for glass(K9)/Ni (20 nm) which reaches the maximum at 66.7 fs after excitation. Considering the time resolution in the experiment, we deduce that the time for the reflectivity to reach the maximum should be between 66.7 and 133.4 fs. This time is shorter than the 200±20 fs reported by van Kampen *et al.*[10].

In Eq. (2), we use the values $\tau_{\rm E}$=0.32 ps and ΔT_1= 150 K, ΔT_2=82 K. Although these values were calculated by van Kampen *et al.*[10], we thought they were effective in our experiments. Thus for $t_{\rm ex}$=66.7 fs, a thermalization time $\tau_{\rm th}$ =18 fs is found, and for $t_{\rm ex}$ = 133.4 fs, $\tau_{\rm th}$ = 47 fs is found. So we thought the thermalization time was between 18 and 47 fs. This value was shorter than those reported before. But it was not consistent with the theoretical calculation. Further research is still needed for the ultrafast electron dynamics of 3*d* metals.

Besides, we can see the influence of the probe beam power from Fig. 4. The amplitude increases with the power of the probe beam when the power of pump beam keeps constant. Figure 5 shows the transient relfectivity of Si/Ni(190 nm) sample. Combining Fig. 4. and 5, we can see that the substrate of the film also influences the shape of the transient reflectivity signal. The substrate

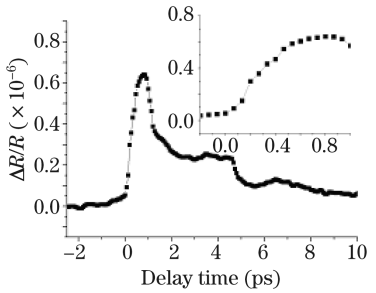

Fig. 6. Transient reflectivity signal for the micro-nano-structure sample with the pump power of 128 mW and the probe power of 8 mW.

influences the thermal transmission between the film and itself. So the thermal conductivities for different substrates are different. Different substrates have different temperature distributions, so the reflectivity is different. But all the factors in the experiment did not influence the time that the change happened.

The transient reflectivity signal for the sample with micro-nano-structure is shown in Fig. 6. As we can see, the reflectivity reaches the maximum at 800 fs. This value is much longer than that got from the Ni film. It means that the thermalization time of the micro-nano-structure film is longer according to the theory analysis before. We deduced that it was caused by the character istic of the nano-structure: the discontinuity of the electron band close to the Fermi level.

Considering only e-e scattering, the lifetime of highly excited electron in a free electron metal is given by[13]

$$\tau_{\rm ee}(E) = \frac{1}{K_{\rm ee}(E - E_{\rm F})^2},$$

where $K_{\rm ee}$ is the e-e scattering constant, $E_{\rm F}$ is the energy of the Fermi level. Then the discontinuity of the electron band makes the value of $(E - E_{\rm F})^2$ increase. So the thermalization time of the micro-nano-structure film becomes longer than the value of the Ni film.

In summary, we have investigated the ultrafast thermalization dynamics of Ni films and micro-nano-structure film. The micro-nano-structure film was formed by ammonia etching technique. We found that the thermalization time was between 18 and 47 fs for the Ni film sample, while for the micro-nano-structure film it was much longer. We deduced that it was caused by the discontinuity of the electron band close to the Fermi level in the micro-nano-structure film.

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