Ultrafast optical probe of coherent acoustic phonons in Co_2MnAl Heusler film*

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In this work, pronounced oscillations in the time-resolved reflectivity of Heusler alloy Co_2MnAl films which are epitaxially grown on GaAs substrates are observed and investigated as a function of film thickness, probe wavelength, external magnetic field and temperature. Our results suggest that the oscillation response at 24.5 GHz results from the coherent phonon generation in Co_2MnAl film and can be explained by a propagating strain pulse model. From the probe wavelength dependent oscillation frequency, a sound velocity of $(3.85\pm0.1)\times10^3$ m/s at 800 nm for the epitaxial Co_2MnAl film is determined at room temperature. The detected coherent acoustic phonon generation in Co_2MnAl reported in this work provides a valuable reference for exploring the high-speed magnetization manipulation via magnetoelastic coupling for future spintronic devices based on Heusler alloy films.

Keywords: Heusler alloys, coherent acoustic phonon, time-resolved spectroscopy

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1. Introduction

Achieving a high degree of spin polarization is critical for the development of ultra-high-density magnetic storage and other spin-dependent devices. Some of Co_2-based Heusler alloys exhibit half metallicity with high Curie temperature and spin polarization and have been widely investigated as promising materials in spintronics, such as spin transport-based electronics and spin injection sources.[1–8] For device physics towards future applications in these directions, it is not only crucial to understand the magnetic and spin dynamics, but also vital to understand the properties of phonons that govern the thermal conductivity and electron scattering in the Co_2-based Heusler alloys. Moreover, the recent studies have shown that the coupling between magnons and elastic phonons, known as the magnetoelastic coupling, can efficiently trigger the magnetization switching and precession.[9–13] Especially, the acoustically driven magnetization switching on an ultrafast time scale offers great capability for a high-speed control of local magnetization.[10–12,14] The knowledge of phonon spectrum in the ferromagnetic films allows us to understand the elastic properties of materials and the coupling mechanism between phonons and magnons as well as external stimuli such as light field. Thus it is important to study the properties of coherent acoustic phonons in Co_2-based Heusler alloys, which remains to be explored, in order to investigate the effective magnetization modulation through magnetoelastic coupling.

With the fast development of ultrafast lasers, comprehensive information about phonon properties can be obtained experimentally by using picosecond acoustics techniques in which the coherent acoustic phonon can be generated by an optical pump pulse from a femtosecond laser and detected with femtosecond temporal resolution by the probe pulse from the same laser.[15] This technique has been successfully used to study the coherent acoustic phonons in various thin-film structures,[15–18] bulk materials,[15,19,20] semiconductor superlattices,[21] and quantum wells.[22,23] One of the mechanisms of coherent acoustic phonon generation and detection is that the absorption of ultrafast laser pulses would induce the propagation of an ultrafast, impulsive strain excitation through material. A periodic oscillation can be observed due to the self-interference of the reflected probe beam from the crystal surface and the surface defined by the propagating strain pulse.[15,24] Other generation mechanisms of coherent acoustic phonon include the impulsive stimulated Raman scattering[21,25] and displacive excitation of coherent phonons,[26] which have been observed in bulk semiconductors and superlattices. In both cases the coherent acoustic oscillations are detected through changes in the reflection or transmission by the modulation of interband transitions coupled to the deformation potential. Large amplitude oscillations in GaN heterostructures have been observed recently and are attributed to the screening of the piezoelectric field by the photoexcited carriers.[22]

In this work, we report the time-resolved reflectivity study of Co_2MnAl film epitaxially grown on a GaAs substrate by a two-color femtosecond pump–probe technique. The oscillatory response is observed in the transient reflectivity response

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and systematically investigated as a function of probe wavelength, temperature, and magnetic field, and film thickness. It is suggested that the laser-induced coherent acoustic phonon generation in Co_2MnAl film is responsible for the observed oscillation response. The physical mechanism for the coherent phonon generation in Co_2MnAl film can be explained by a propagating strain pulse model.

2. Experiment

The two Co_2MnAl films with different thicknesses are grown on GaAs (001) substrates with a temperature of 280 °C by molecular-beam epitaxy (MBE) technique. Before deposition of the Co_2MnAl layer, a 150-nm-thick GaAs buffer layer is deposited first on the GaAs substrate. The films are capped with a 2-nm-thick aluminum layer to avoid being oxydized. The thickness values of as-grown Co_2MnAl films are determined to be 60 nm and 100 nm by the superconducting quantum interference device (SQUID) magnetometer, with knowing the Co_2MnAl saturation magnetization of about 1000 emu/cm^3 calibrated before. After MBE growth, an 8-nm-thick Au film is deposited on the top by using the electron beam evaporation technique, since the previous studies have shown that coating thin Au film can facilitate the generation of the thermal-induced coherent acoustic phonons wave.[27–29] The sample structure is schematically shown in Fig. 1. In our experiment, a two-color pump–probe reflectivity measurement is carried out in a temperature range of 9 K–300 K. A Ti:sapphire regenerative amplifier laser (Coherent Inc.) with a repetition frequency of 1 kHz and pulse width of 150 fs is used for the optical pumping. The sample is pumped mainly at a wavelength of 800 nm with an energy density of ~ 2 mJ/cm^2 except in a pump wavelength-dependent measurement, and the transient reflectivity is probed with wavelength tuned in a range of 620–800 nm, which is generated through an optical parametric amplifier pumped by the regenerative amplifier. The p-polarized pump pulse is directed onto the sample surface at nearly normal incidence and a p-polarized probe pulse is incident at an angle of 30°. The time-resolved reflectivity of the probe signal is measured by using an amplified Si photodetector combined with lock-in technique and plotted as a function of the delay time between the pump and probe pulses. The sample is mounted in a Janis closed-cycle subcompact optical cryostat located between the poles of an electromagnet.

Au (8 nm)
Al (2 nm)
Co_2MnAl (60 nm/100 nm)
GaAs buffer (150 nm)
GaAs substrate

Fig. 1. Sample structural sketch of Co_2MnAl films with different thickness.

3. Results and discussion

The typical time-resolved reflectivity responses of the 60-nm-thickness and 100-nm-thickness Co_2MnAl films measured with probe wavelength at 800 nm are shown in Figs. 2(a) and 2(b), respectively. It is noticed that the transient reflectivity response consists of two components: one is a step-like jump at time zero followed by an exponential decay due to the electron–phonon interaction;[30,31] and the other is a damped oscillation. The insets of Figs. 2(a) and 2(b) show the fast Fourier transform (FFT) of the damped oscillation. It appears that one can observe two main peaks for 60-nm-thick Co_2MnAl film and only one peak for 100-nm-thick Co_2MnAl film. The peak at nearly 24.5 GHz is observed for both samples, while the second peak at 43.5 GHz can only be observed for 60-nm-thick Co_2MnAl film.

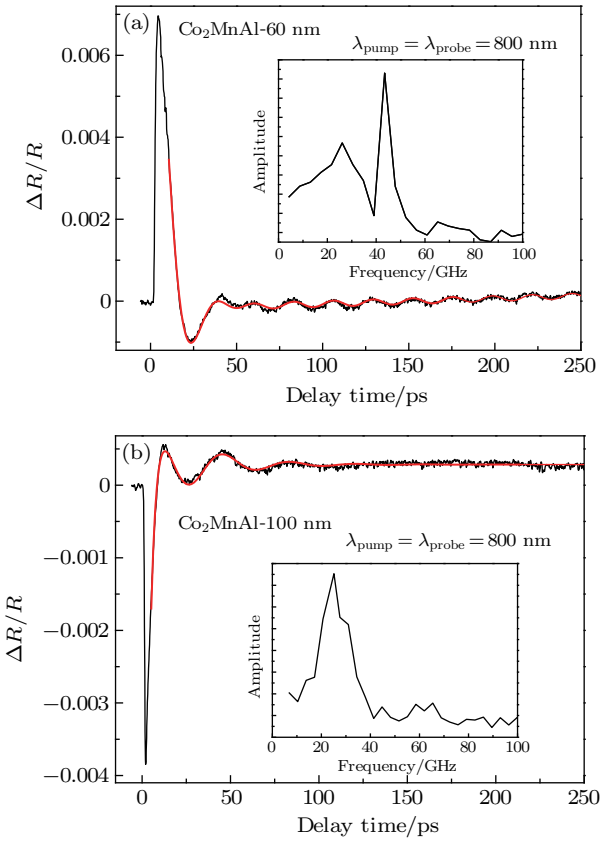


Fig. 2. (color online) Typical time-resolved reflectivity responses and their best fits (red solid line) for Co_2MnAl films with different thickness: (a) 60 nm; (b) 100 nm. The insets in panels (a) and (b) show the fast Fourier transform (FFT) of the oscillation, respectively.

We then use the following damped harmonic function superimposed onto an exponentially decaying background to fit the measured transient reflection response:[21,32,33]

$$f(t) = A + B\exp(-t/t_0) + \sum_k C_k \exp(-t/\tau_k)\cos(2\pi f_k t + \varphi), \quad (1)$$

where A, B, and C_k are the fitting parameters, and t is the delay time between pump and probe pulse, t_0 is the recovery rate of

the thermal electrons, while f_k, φ, and τ_k are the oscillation frequency, phase, and decay time of the oscillations, respectively. Then the oscillation frequency of about 24.5 GHz for both samples, which does not show film thickness dependence, is extracted with both pump and probe beam wavelength fixed at 800 nm, which is the same as that of the FFT analysis results. Since the phonon echo which bounces back and forth in film should have a thickness-dependent frequency as described by $f_{\text{echo}} = v_s/2d$,[34,35] in which d is the film thickness and v_s is the sound velocity. The possible origination from the phonon echo response is thus excluded since the common oscillatory mode at 24.5 GHz shows up for both Co_2MnAl films with quite different thickness values. Moreover, we can rule out the phonon echo response in Au or Al film based on the known sound of velocity,[36,37] and this oscillation frequency at 24.5 GHz is not related to the phonon echo in the 150-nm-thick GaAs buffer either, since the phonon echo should response at 15.8 GHz for 150-nm-thick GaAs buffer if the sound velocity of GaAs is taken to be 4730 m/s.[38]

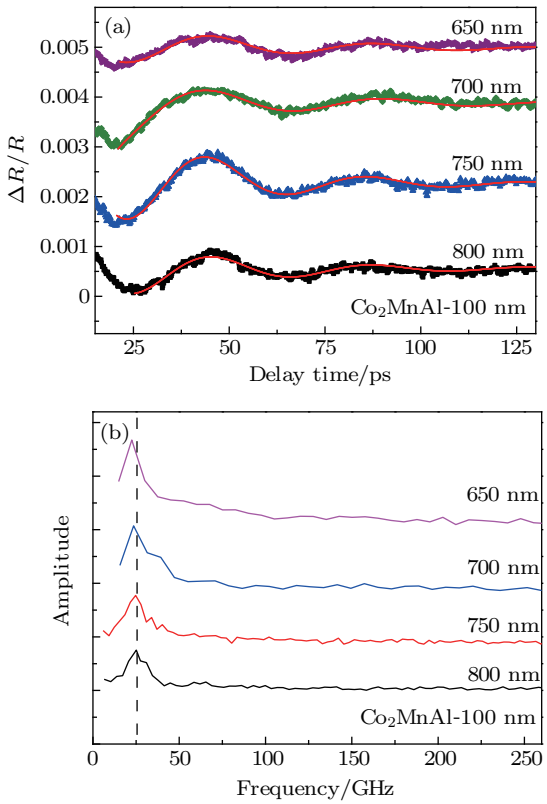


Fig. 3. (color online) (a) Oscillatory components of the time-resolved reflectivity and their best fits (red solid line) by varying wavelength of probe beam for 100-nm-thick Co_2MnAl film; (b) fast Fourier transforms of the periodic oscillations measured at different wavelengths of probe beam. The dashed line shows a guide for the eye for highlighting the variation of oscillation frequency as a function of probe wavelength.

In order to look into the nature of the oscillatory response observed in the transient reflectivity response, we investigate the wavelength dependent response of the time-resolved reflectivity at room temperature by tuning the probe wavelength. The typical time-resolved reflectivity response for 100-nm-thick Co_2MnAl film probed at different wavelengths is presented in Fig. 3(a), in which the exponentially decaying background component is removed numerically in order to focus on the details of oscillations. The fast Fourier transform analysis is used to determine the frequencies of oscillations and the results are illustrated in Fig. 3(b). In each of the curves one can see that there is only one main peak marked by the dashed line and the oscillation frequency shows a slight dependence on the probe wavelength.

As is well known, in the case of impulsive stimulated Raman scattering, the generated acoustic phonon wave vector q is well defined, i.e., $q \sim 4\pi/\lambda_{\text{pump}}$ in the backscattering geometry with λ_{pump} being the optical pumping wavelength. So the oscillation frequency depends on λ_{pump}, not on the probe beam wavelength of λ_{probe} nor the incident angle of θ. In our experiment, the oscillation frequency is found to exhibit no dependence on the pumping wavelength. For the propagating strain pulse mechanism proposed by Thomsen *et al.*,[15] the ultrafast pump pulse absorbed at the surface of the film produces a thermally induced strain wave, which propagates away from the surface at the speed of the longitudinal acoustic phonons. The interference between the probe beams partially reflected from the top surface of the sample and the probe pulse reflected from the propagating strain wave surface can generate the detectable coherent oscillations in the reflectivity signal. According to the propagating strain model, the change in the reflectance is in the following form:[15]

$$\Delta R(t) \sim \Delta R_0 \cos\left(\frac{4\pi n v_s t}{\lambda_{\text{probe}}} \cos\theta_m - \phi\right) e^{-t/\tau}. \tag{2}$$

Here, v_s is the sound velocity, n is the refractive index, θ_m is the refraction angle inside the sample, τ is the decay time, and ϕ is the phase. The distinct characteristic of the propagating strain pulse mechanism is that the frequency of oscillations can be described by

$$f = 2nv_s \cos\theta_m/\lambda_{\text{probe}}. \tag{3}$$

Therefore, the observed coherent oscillation frequencies measured at several probe wavelengths are retrieved by fitting the transient reflectance responses through Eq. (1) for Co_2MnAl films with different thickness, and the results are displayed in Fig. 4(a). The extracted oscillation frequencies present different tendencies when tuning the probe wavelength from 620 nm to 800 nm. For the high-frequency mode, it has been well observed in the past and ascribed to the coherent acoustic phonon generation in GaAs based on previous studies.[39,40] It is known that the refractive index of GaAs is inversely proportional to the wavelength in a range of 600–800 nm.[41] The sound velocity of GaAs is evaluated to be $v_{s,\text{GaAs}} = 4730$ m/s by fitting curves in Fig. 4(a) based

on Eq. (3), which accords well with the previously reported result and further confirms that the high-frequency mode of 43 GHz probed at 800 nm originates from the coherent acoustic phonon generation in GaAs buffer or substrate. However, the oscillation frequency of the low-frequency mode is seen to increase slightly with increasing probe wavelength. Since neither the refractive index nor sound velocity of Co_2MnAl film is known, we further perform the ellipsometry spectroscopy measurement of the studied Co_2MnAl film in a wavelength range of $\lambda = 190$–1690 nm, by using an M-2000 spectroscopic ellipsometer (J. A. Woollam Co., Inc.) combined with automatic goniometer and XYZ-sample stage. Since our Co_2MnAl films are much thicker than the penetration length of excitation light beam, a single bulk-like optically isotropic material fitting procedure is used to evaluate the effective optical constants of the refractive index n and the extinction coefficient k for the studied Co_2MnAl film, based on the spectroscopic ellipsometry results, and the results at wavelengths ranging from 600 nm to 800 nm are shown in Fig. 4(b). It can be seen that the effective optical constants (n, k) show a slight increase with increasing wavelength, which is consistent with the theoretical predictions by the first-principles calculations for Co-based Heusler alloys.[42,43] With the refractive index dispersion relationship as evaluated by ellipsometry measurement, the frequency of coherent acoustic phonons of Co_2MnAl film can be fitted at a fixed sound velocity, as displayed in Fig. 4(a), in which a sound velocity of $(3.85 \pm 0.1) \times 10^3$ m/s for the Co_2MnAl film is deduced based on the fitting results.

Now, we come to discuss the role of the thickness of the sample related to the strain-pulse penetration depth. The damping of the oscillatory signal arises from two factors in the propagating strain model. First, the absorption loss of the probe which bounces back and forth in the multilayer film must be taken into account. A second contribution comes from the absorption and/or scattering of the acoustic phonons during propagation, with the decay time of acoustic wave corresponding to a time when the strain pulse can propagate into the film within its characteristic length before complete energy transfer takes place from the coherent mode to a distribution of incoherent modes. Fitting the observed oscillations with Eq. (1), we can extract an averaged decay time of $\tau \approx 35$ ps. With the extracted speed of sound, the corresponding maximum propagation distance of $l = v_s \tau \approx 135$ nm is obtained. Thus we can only detect the coherent acoustic phonon of GaAs buffer/substrate for 60-nm-thick Co_2MnAl film, owing to the absorption loss of the probe beam together with the scattering of the acoustic phonon during propagation. This supports our above speculation that the periodic oscillation in the transient reflectivity response originates from a propagating strain field.

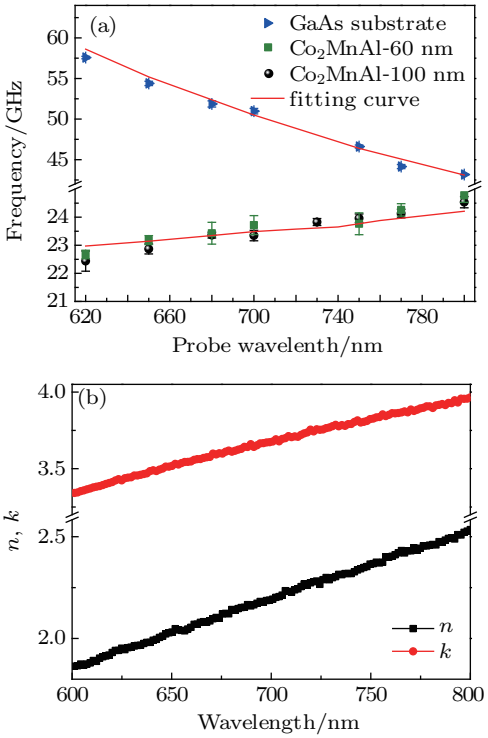


Fig. 4. (color online) (a) Oscillation frequencies as a function of probe wavelength for Co_2MnAl films with different thickness values and GaAs. The solid red lines show the fitting curves based on Eq. (3), where the sound velocities of GaAs and Co_2MnAl are evaluated to be $v_{s,GaAs} = 4730$ m/s and $v_{s,Co2MnAl} = 3850$ m/s respectively. (b) The wavelength-dependent effective refractive index n and extinction coefficient k from ellipsometry measurement in a wavelength range of 600–800 nm for Co_2MnAl film.

Another possible origination of the observed periodic oscillations could be the coherent magnons since the detection of coherent magnons via time-resolved reflectivity is also possible and has already been observed in some of the multiferroics.[44–46] Coherent magnon excitation can be viewed as triggering magnetic precession when the sublattice equilibrium position is modified through transient optical heating of the crystalline lattice from energy relaxation of an initial hot electron distribution, leading to a rapid modification of the exchange coupling between the sublattices. Thus the coherent magnon could be generated and show strong dependence on the external magnetic field. To elucidate whether the observed periodic oscillations in our time-resolved reflectivity measurement is associated with the coherent magnon generation in Co_2MnAl film, we further perform the magnetic field and temperature dependent measurements. The extracted oscillation frequency as a function of the applied in-plane magnetic field is shown in Fig. 5(a), in which one can see that the oscillation frequency remains almost constant in a wide magnetic field range of 0–7000 Oe, implying that the magnetism does not play a role in the observed oscillations. Moreover, the temperature dependence of the coherent oscillation frequency in the transient reflectivity measurement is presented in Fig. 5(b), in which the frequency shows a slight increase with temperature increasing from 9 K to 200 K. This

temperature-dependent trend is remarkably different from that of coherent magnon generation measured for Co_2FeAl film in the previous work,[47] in which the magnetic precession frequency decreases with increasing temperature. From our magnetic field and temperature-dependent time-resolved reflectivity measurements, it is therefore concluded that the observed oscillations are not associated with the coherent magnon generation in Co_2MnAl film.

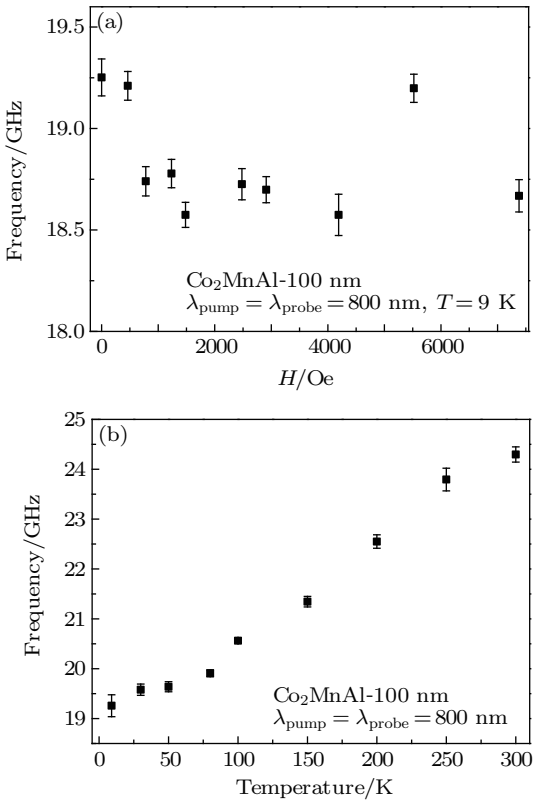


Fig. 5. Dependences of the oscillation frequency on (a) external magnetic field measured at 9 K and (b) temperature for 100-nm-thick Co_2MnAl film. All the measurements are performed, with the probe wavelength fixed at 800 nm.

4. Conclusions

In this work, we perform two-color pump–probe reflectivity measurements on Co_2MnAl films epitaxially deposited on GaAs substrates. A periodic oscillation response is observed, and its frequency depends on the film thickness, probe wavelength, external magnetic field, and temperature. It is suggested that the observed periodic oscillation should result from the coherent phonon generation in the Co_2MnAl layer and GaAs buffer/substrate and be able to be explained by a propagating strain pulse model. From the measured acoustic phonon frequency, the room-temperature sound velocity of longitudinal acoustic phonon in Co_2MnAl film is evaluated to be $(3.85 \pm 0.1) \times 10^3$ m/s at 800 nm. The detected coherent acoustic phonon generation and its frequency provide valuable information for the intriguing potential of high-speed acoustic control of magnetization through magnetoelastic coupling based on Heusler alloys.

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