Phase Transition Kinetics in FeRh Thin Films

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Introduction

Materials with coupled magnetic and structural phase changes — magnetostructural materials — have the potential to exhibit a large functional response to physical inputs such as small deviations in temperature, pressure, or magnetic field, and are thus of both basic scientific interest as well as technological interest for advanced sensors. Equiatomic $\alpha'$-FeRh, a model magnetostructural material, has been shown to undergo a thermodynamic first-order phase transition from antiferromagnetic (AF) to ferromagnetic (FM) character \cite{1} with an accompanying 1\% lattice expansion at approximately 370 K \cite{2}. Thin film forms of FeRh are of particular interest, as compared with bulk FeRh, because they exhibit a larger response to external variables due to the epitaxial clamping at the film interfaces.

In this work, FeRh thin films serve as a test bed to understand the role of the intensive variables, temperature, strain, and applied magnetic field on the character and kinetics of the magnetostructural transition. Furthermore, the impact of these variables on the energy barrier which must be overcome for the transition to occur is considered in the framework of the Kissinger model for crystallization kinetics during continuous heating. The governing principle behind this model is that the transition peak temperature depends on heating rate; thus variations in the peak temperature as a function of heating rate can be used to determine activation energies for phase transformations.

The small amount of material in thin films presents a challenge regarding the application of thermal analysis techniques to examine phase transformations. While the energy barrier associated with the fcc-bcc transition in bulk FeRh has been determined as 149 kJ/mol \cite{3} by application of the Kissinger model to differential scanning calorimetry (DSC) data, it is not possible to apply this experiment to thin films. To overcome this challenge this work poses the unique solution of applying the Kissinger model to superconducting quantum interference device (SQUID) magnetometry measurements. In this manner, the role of magnetic field and film thickness on the character and kinetics of the magnetostructural transition may be determined in thin film forms of FeRh. A combination of information obtained from these investigations will contribute to knowledge of factors controlling the magnetostructural transformation in FeRh and provide pathways to tailor the transition to defined outcomes.

Results and Discussion

High quality FeRh films of 10, 30, 40 and 50 nm thicknesses were grown by sputter deposition on (001) MgO substrates and capped with Al. Experimental results obtained thus far from SQUID magnetometry measurements show a linear increase in the onset of the AF-FM transition with increasing film thickness, which is accompanied by a narrowed thermal hysteresis in the transition. Additionally, the effect on the onset of the transition with applied magnetic field shows that the sensitivity of the transition increases
with decreasing film thickness. These trends suggest that the critical activation energy required for the onset of the AF-FM (heating) transition decreases with decreasing film thickness while the degree of undercooling increases. In order to study the kinetics and character of the FeRh magnetostructural transition in more detail this work has narrowed the focus to concentrate on two films of 50 and 10 nm thickness.

The application of the Kissinger model to SQUID magnetometry measurements conducted at varied heating rates has allowed for the activation energies of the AF-FM transition to be determined as a function of applied magnetic field and film thickness. Shown in Figure 1 are thermal-magnetization hysteresis loops for various heating rates in an applied field of 2 T. It can be seen that the onset of both the AF-FM and FM-AF transition temperatures (T_t) increase with increasing heating rate. Furthermore, from these results it is evident that the slower heating rate stabilizes the FM phase and the slower cooling rate stabilizes the AF phase.

Energy barrier calculations of the AF-FM transition show that the activation energy of the 10 nm film (230 +/- 10 kJ/mol) is lower than that of the 50 nm film (588 +/- 25 kJ/mol). Furthermore, in both films the energy barrier of the transition may be lowered by increasing the applied magnetic field. The extent to which this barrier is lowered increases with decreasing film thickness; a decrease from -29.7 +/- 4 kJ/mole, in a 50 nm film, to -39.7 +/- 5 kJ/mol, in a 10 nm film, (a 25 % difference) is noted.

Overall, these results demonstrate the usefulness of applying the Kissinger model to an unconventional thermal analysis technique such as SQUID magnetometry. In this way a quantitative estimate of the energy barrier associated with AF-FM transition in thin films has been determined. In specific, the energy barrier of the FeRh transition is lowered with increasing applied magnetic field and decreasing film thickness. These results suggest that nucleation of a FM phase is preferred as film thickness is decreased. Thus, it is hypothesized here that the lower energy barrier in the 10 nm film is a result of the increased strain state from epitaxial clamping of the film to a substrate.

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References: