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PII: S0022-4596(16)30486-8

http://dx.doi.org/10.1016/j.jssc.2016.12.010 DOI:

Reference: **YJSSC19629** 

To appear in: Journal of Solid State Chemistry

Received date: 2 August 2016 Revised date: 7 December 2016 Accepted date: 10 December 2016

Cite this article as: V.G. Myagkov, L.E. Bykova, A.A. Matsynin, M.N. Volochaev, V.S. Zhigalov, I.A. Tambasov, Yu.L. Mikhlin, D.A. Velikanov and G.N. Bondarenko, Solid State Synthesis of Mn<sub>5</sub>Ge<sub>3</sub> in Ge/Ag/Mn trilayers magnetic studies, Journal of Solid State Chemistry Structural and http://dx.doi.org/10.1016/j.jssc.2016.12.010

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#### ACCEPTED MANUSCRIPT

# Solid State Synthesis of Mn<sub>5</sub>Ge<sub>3</sub> in Ge/Ag/Mn trilayers: Structural and magnetic studies

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#### **ABSTRACT**

The thin-film solid-state reaction between elemental Ge and Mn across chemically inert Ag layers with thicknesses of (0, 0.3, 1 and 2.2  $\mu$ m) in Ge/Ag/Mn trilayers was studied for the first time. The initial samples were annealed at temperatures between 50-500 °C at 50 °C intervals for 1 hour. The initiation temperature of the reaction for Ge/Mn (without a Ag barrier layer) was ~ 120 °C and increased slightly up to ~ 250 °C when the Ag barrier layer thickness increased up to 2.2  $\mu$ m. In spite of the Ag layer, only the ferromagnetic Mn<sub>5</sub>Ge<sub>3</sub> compound and the Nowotny phase were observed in the initial stage of the reaction after annealing at 500 °C. The cross-sectional studies show that during Mn<sub>5</sub>Ge<sub>3</sub> formation the Ge is the sole diffusing species. The magnetic and cross-sectional transmission electron microscopy (TEM) studies show an almost complete transfer of Ge atoms from the Ge film, via a 2.2  $\mu$ m Ag barrier layer, into the Mn layer. We attribute the driving force of the long-range transfer to the long-range chemical interactions between reacting Mn and Ge atoms.

#### **Graphical Abstract**

The direct visualization of the solid state reaction between Mn and Ge across a Ag buffer layer at  $500\,^{\circ}\text{C}$ 

**Keywords:** Long-range atomic transfer; Mn-Ge system; Thin-film solid-state reaction; Diffusion barrier; Dominant diffusing species; First phase;  $M_{15}Ge_{3}$  alloy

#### 1. Introduction

Although thin films have been widely used in advanced electronic devices, the atomic migration during thin-film solid state reactions continues to be the subject of many investigations [1-5]. Numerous studies of solid-state reactions in nanofilms showed that there are three fundamental features that strongly distinguish them from bulk samples:

- (i) Formation of only the **first phase** at the film interface at a certain temperature  $T_{in}$  called the **initiation (formation) temperature**  $T_{in}$ . As the annealing temperature increases, other phases can occur and form the phase sequence [3-8].
- (ii) The threshold of the reaction, characterized by intense intermixing at the interface and the formation of compounds, arises at temperatures above the initiation temperature  $T_{\rm in}$  [3-5].
- (iii) Migration of the dominant diffusing species through the interface during the first phase formation [5, 9, 10].

The formation of only the first phase among the equilibrium phases, the low initiation temperatures that can be below room temperature [11, 12] or even below 90 K [13-16] and the migration of the dominant diffusing species are unique, unexplained features of solid-state reactions in nanofilms. As discussed above, the initiation temperature  $T_{\rm in}$  is the threshold temperature. This means that there is no reaction below  $T_{\rm in}$  and the reaction initiates just as the

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