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Iron-implanted epitaxial palladium thin films: Structure, ferromagnetism and signatures of spinodal decomposition

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ABSTRACT

We report on the formation of the dilute $Pd_{1-x}Fe_x$ composites with tunable magnetic properties under ion-beam implantation of epitaxial Pd thin films. Binary $Pd_{1-x}Fe_x$ alloys with a mean iron content x of 0.025, 0.035, and 0.075 were obtained by the implantation of 40 keV Fe^+ ions into the palladium films on MgO (0 0 1) substrate to the doses of 0.5×10^{16}, 1.0×10^{16} and 3.0×10^{16} ions/cm^2, respectively. Structural and magnetic studies have shown that iron atoms occupy regular fcc-lattice Pd-sites without the formation of any secondary crystallographic phase. All the iron implanted Pd films reveal ferromagnetism at low temperatures. The observed multiple ferromagnetic resonances in the implanted $Pd_{1-x}Fe_x$ films indicate a formation of a magnetically inhomogeneous state due to spinodal decomposition into regions, presumably layers, with identical crystal symmetry but different iron contents. The multiphase composite magnetic structure is robust with respect to the vacuum annealing at 770 K, though develops towards well-defined local Pd/Fe ratios.

1. Introduction

The renaissance of interest to $Pd_{1-x}Fe_x$ alloys was triggered recently by their potential applications in superconducting spintronics, where diluted compositions with $x < 0.10$ serve as a weak ferromagnetic link in Josephson-junction structures for ultrahigh-speed cryogenic devices with extraordinary energy efficiency suitable for supercomputers and artificial intelligence applications [1–6]. Traditionally, $Pd_{1-x}Fe_x$ alloys have been fabricated using the magnetron sputtering [3–10] or molecular-beam epitaxy (MBE) [11,12], including epitaxial thin films of $Pd_{1-x}Fe_x$ ($0.01 < x < 0.1$) on the MgO (0 0 1) substrate [13–16]. However, in modern foundries with evacuated production process, ion-beam implantation is a common technology for silicon doping and microchip fabrication [17,18]. It makes sense to apply the ion implantation to form dilute Pd-Fe alloys with tunable magnetic properties as a testing pathway for the magnetic Josephson junction production.

In this communication, we apply high-dose implantation of epitaxial palladium films with iron at room temperature (RT) to obtain weakly ferromagnetic layers with a minimum of irradiation-induced defects. In contrast to the near-equilibrium MBE or magnetron sputtering syntheses, the ion-beam implantation is a significantly nonequilibrium process that could lead to new qualities of the produced materials.

2. Experimental

Epitaxial Pd thin films on MgO (0 0 1) single-crystal substrate were produced from the high purity Pd (99.98%) utilizing ultrahigh vacuum (UHV) MBE system [14] and then were implanted by Fe^+ ions at a fixed ion energy of 40 keV. To obtain Pd-Fe alloys with various iron concentrations, Fe^+ ion doses D of 0.5×10^{16}, 1.0×10^{16} and 3.0×10^{16} ions/cm^2 (*sample labels* S0_5, S1_0 and S3_0, respectively) were achieved varying the irradiation time. The Fe^+ doses were adjusted to yield the samples with the mean iron contents of $\bar{c}_{Fe} \approx 2.5$, 3.5, and 7.5 at.% in the implanted layer. A piece of each implanted sample was post-annealed in UHV (5×10^{-9} mbar) at 770 K for 20 min. The details of the experimental equipment and various characterization methods are given in Supplementary material.

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