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STUDIES OF MAGNETIC PROPERTIES OF THIN CdCr₂Se₄ FILMS – EXPERIMENT

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Abstract. – A new preparation method of thin CdCr₂Se₄ films by vacuum evaporation technique is proposed. We report and analyse the temperature dependence of magnetization and the effect of Cr²⁺ and Cr⁴⁺ impurity ions on the FMR linewidth. Also the electrical transport properties of these films are studied.

Introduction

CdCr₂Se₄ doped with In is a ferromagnetic semiconductor in which the Cr⁴⁺ and Cr²⁺ ions are paramagnetic impurities. We consider our samples as nonstoichiometric ones. Cr⁴⁺ are host ions and Cr²⁺ and Cr⁴⁺ may be present as a result of doping or as a result of a lack of stoichiometry. The presence of Cr⁴⁺ and Cr²⁺ manifests itself in transport and magnetic properties as well. We studied the temperature dependence of conductivity and we conclude that polycrystalline thin films of CdCr₂Se₄ In are p-type semiconductors. We also analysed the temperature dependence of FMR data. From the behaviour of a linewidth it can be seen that relaxation processes in our samples are more complex than in the case of monocrystalline ones. Our experimental data show some possibility that the Cr₂Se₃ phase is present.

Experiment

The CdCr₂Se₄: In samples were simultaneously evaporated from the separate sources. We used four independent sources of Cd, Cr, Se and In during the evaporation process in high vacuum. Thin 50 Å layers of Cr were deposited onto Corning glass substrates, first heated to 520 K, to get better adhesion of the films. The films themselves were deposited at about 350 K. Their electrical resistivity was continuously monitored during the film growth. The samples were covered with a thin layer of Cr of about 100 Å. Thickness of the samples was measured by means of a profile detector Talysurf 4. Composition of the samples was analysed by an X-ray microprobe (ARL SEMQ microanalyser) and by Auger spectroscopy (Riber LAS-620). The as-deposited samples were in amorphous state. The crystallization temperature was found by means of the temperature dependence of X-ray diffraction experiment in high vacuum. Heat treatment was carried out in the X-ray diffraction apparatus Kristalloflex 4 H at the temperature ranging from 300 K to 850 K and for different annealing times from 0.5 h to 5 h.

We found that the samples reached the polycrystalline state after being annealed at 790 K for 1 h. The temperature dependence of the saturation magnetization was measured from 77 K to 300 K by means of a magnetic balance magnetometer. The FMR experiment was carried out at X-band within the temperature range from 4.2 K to 300 K for a perpendicular and parallel geometry.

Results and discussion

The temperature dependence of conductivity shows that we have p-type semiconductor with the activation energy equal to 0.08 eV. From the X-ray microprobe and Auger spectroscopy analyse we can estimate that the atomic concentration of In is less than 1 %. We did not obtain the semiconductor – metal transition. Figure 1 shows the temperature dependence of magnetization. Data were taken from FMR and magnetic balance experiments. We can see that there are two phases which were also confirmed by X-ray diffraction experiment. We consider that apart from CdCr₂Se₄ phase the Cr₂Se₃ phase could be also present. Both phases have approximately the same Curie temperature (about 140 K) and Bloch law is not valid...
Fig. 2. – Temperature dependence of the peak-to-peak linewidth a) for one phase thin films of CdCr$_2$Se$_4$; b) for two CdCr$_2$Se$_4$ and Cr$_2$Se$_3$ phases thin films.

obeyed. The Curie-Weiss temperature is 185 K (for CdCr$_2$Se$_4$ phase) and g-factor is 1.99. So the ferromagnetic state is well defined. As we have already mentioned the effect of impurity of Cr$^{+3}$ and Cr$^{+4}$ ions on the FMR linewidth ($\Delta H$) was also studied. Figures 2a, b show the temperature dependence of the linewidth for a one phase sample (Fig. 2a) and a two phases sample (Fig. 2b). $\Delta H = f(T)$ data were taken from FMR experiment for the perpendicular and parallel geometry. The temperature dependence $\Delta H(T)$ has two maxima as shown in figure 2a. These results are different from the ones presented in papers [1-4] for monocrystalline bulk samples, mostly because the value of $\Delta H$ (in our case) is about one order higher. The temperature dependence of the linewidth for monocrystalline samples exhibits a maximum at a temperature lower than 4.2 K. This dependence is a rapidly decreasing function as the temperature increases. The similar behaviour is seen in the figure 2b for the parallel and perpendicular geometry (but only for one peak), with $\Delta H$ still one order higher than for monocrystalline case. We would like to emphasize that for this sample we obtained two peaks in the perpendicular geometry (due to two phases) and one peak in the parallel case. It means that resonance conditions for both phases are the same in the parallel geometry. As it is reported in literature [1] the model of two relaxation mechanisms describes of $\Delta H(T)$ behaviour; the slow or longitudinal relaxation mechanism and the fast or transverse one. The corresponding ion relaxation mechanisms have been named spin-phonon and spin-magnon interactions. In both relaxation processes the spin system is considered as consisting of two subsystems. The first subsystem is a magnetically ordered one, formed, in our case, by Cr$^{+3}$ ions in S-state and the second subsystem is formed by the Cr$^{+2}$ and Cr$^{+4}$ impurity ions with spin and orbital moments. Cr$^{+2}$ and Cr$^{+4}$ are considered as paramagnetic impurities. Both spin-phonon and spin-magnon interactions are very much affected by the temperature dependence of relaxation time for both kinds of relaxation processes. The two level system [1] gives satisfactory agreement between the FMR experimental data of $\Delta H(T)$ and the theory for the monocrystalline samples. As far as we can see from our experimental data this two – level model can not be simply applied for polycrystalline thin films. It seems that the magnon scattering process on the grain boundaries has to be taken under consideration while the relaxation processes are described. Also spin glass phase [5] is expected in this spin system, so it is possible that the relaxation processes are more complex.

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