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Magnetic Semiconductors Thin Films-Unidirectional Anisotropy

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Unidirectional magnetic anisotropy field (H_{an}) was investigated for thin films of $CdCr_{2-2X}In_{2X}Se_4$ ($0 \le x \le 0.2$). This anisotropy originates from the microscopic anisotropic Dzyaloshinskii-Moriya (DM) interaction which arise from the spin-orbit scattering of the conduction electrons by the nonmagnetic impurities. This interaction maintains the remanent magnetization in the direction of the initial applied field. Then the single easy direction of the magnetization is parallel to the direction of the magnetic field. The anisotropy produced by field cooling is unidirectional i.e. the spins system keeps some memory of the cooling field direction. The chalcogenide spinel of $CdCr_{2-2X}In_{2X}Se_4$ belongs to the class of the magnetic semiconductors. The magnetic disordered state is obtained when ferromagnetic structure is diluted by In. Then we have the mixed phase characterised by coexistence the magnetic long range ordering (IFN-infinite ferromagnetic network) and the spin glass order (Fc-finite clusters). The total magnetic anisotropy energy depends on the state of magnetic ordering. In our study we concentrated on the magnetic state with reentrant transition and spin glass state. The polycrystalline $CdCr_{2-2X}In_{2X}Se_4$ thin films were obtained by rf sputtering technique. We applied the ferromagnetic resonance (FMR) and *M-H* loop techniques for determining the temperature composition dependencies of H_{an}. From the experimental data, we have found that H_{an} decreases almost linearly when temperature is increased and in the low temperature is about three times bigger at SG state with comparison to the state with REE.

1. Introduction

The chalcogenide spinels of $CdCr_2Se_4$ exhibit quite complex magnetic interactions between Cr ions [1]. There are direct and indirect magnetic exchange interactions. The strength of the nearest-neighbour exchange is assumed to be positive (ferromagnetic) or negative (antiferromagnetic). Frustration is due to the competition between the nearest-neighbors and the next-nearest-neighbors. Disordered magnetic state with the reentrant transition (REE) and the spin glass (SG) is created in chromium spinels diluted with In. Its properties are strongly influenced by the magnetic anisotropy. In general, there are two components of the total energy of the magnetic anisotropy: the unidirectional and uniaxial. In the case of polycrystalline thin film, the only contribution to the energy of the uniaxial anisotropy comes from the shape magnetic anisotropy. The unidirectional magnetic anisotropy field originates from the only contribution microscopy anisotropy Dzyaloshinskii-Moriya (DM)[2-4] interaction that arises from the spin-orbit scattering of the conduction electrons by the nonmagnetic impurities. This interaction forces the remanent magnetization in the direction of the initial applied field. The DM interaction can cause some spins of spin clusters to flip discontinuously as the net magnetization, induced by cooling in external magnetic field responds to a changing external field. Then the single easy direction of the magnetization is parallel to the direction of the magnetic field. The anisotropy produced by the field cooling is unidirectional i.e. the system of spins retains the cooling field direction.

The aim of the present pater is to describe the experimental techniques of determination of the unidirectional magnetic anisotropy field H_{an}. The *M-H* loop and the ferromagnetic resonance (FMR) measurements were carried out to find the temperature and composition dependencies of H_{an} in thin films of $CdCr_2Se_4$: In and $CdCr_{2-2X}In_{2X}Se_4$.

Ferromagnetic resonance (FMR) data were taken with a microwave spectrometer within the temperature range form 4.2 K to 150 K at X band. The Han was calculated from a shift of the resonance field with respect to the position predicted when only the magnetic shape anisotropy is considered.

The *M-H* loops data were recorded for samples cooled in the external magnetic field. The unidirectional magnetic anisotropy field was determined from a displacement of the *M-H* loop and from the external magnetic field dependence of the reversible M_{rev} and the frozen irreversible M_{iff} part of the magnetization.

We have found that H_{an} depends on the temperature and the sample composition.

2. Experiment

We investigated the thin films of spinel $CdCr_2Se_4$ with controlled amount of in the lattice. For $CdCr_2Se_4$: In samples are in the state with reentrant transition (REE). A higher concentration of indium produces the spin glass (SG) state ($CdCr_{2-2x}In_{2x}Se_4$ with $0 \leq x \leq 0.2$). Films were deposited by rf sputtering technique on Corning glass substrates. Other details of the preparation techniques are described in paper [5]. The composition of the samples was determined, from X-ray microprobe measurements (the accurency was 5%).

The state with reentrant transition (REE) and spin glass (SG) state were confirmed by the temperature dependence of the magnetization M and unidirectional magnetic anisotropy filed H_{an} and also with used the critical line of H-T phase diagram [5, 6].

The temperature dependence of M and H_{an} was determined from the FMR data and from SQUID measurements.

In the FMR technique the position of resonance peak depends on the total value of the internal magnetic field H_{int}. For thin films, apart from the external magnetic field H, the shape magnetic anisotropy field $H_u = 4\pi M$ (M is the magnetization) and the unidirectional magnetic anisotropy field H_{an} contribute to H_{int}. In FMR experiment of thin films. two characteristic geometries, perpendicular and parallel, are used for the calculation of basic magnetic parameters. Figs. 1a, b present the temperature dependence of resonance field H_r for both geometries. Theses resonance data are taken for thin films of $CdCr_2Se_4$: In (REE) and $CdCr_{1.7}In_{0.3}Se_4$ (SG), respectively. In general, different internal effective field alter the spins system in both geometries so one could expect different values of H_{an} for perpendicular and parallel resonances. The dispersion relation, for uniform mode of FMR perpendicular and parallel geometry has the form:

$$(\omega/\gamma) = H_\perp + H_{an,\perp} - 4\pi M \qquad (1)$$

$$(\omega/\gamma)^2 = (H_\parallel + H_{an,\parallel})(H_\parallel + H_{an,\parallel} + 4\pi M). \qquad (2)$$

where $\omega = 2\pi\nu$, ν-the microwave frequency, γ-the gyromagnetic factor, $H_\perp$ and $H_\parallel$-resonance fields of uniform mode for perpendicular and parallel geometry, respectively.

For thin films, at disordered magnetic state, the resonance field at low temperature, for both geometries, drops by the same amount. This is consistent with experimental data of the temperature dependence of the resonance field $H_\perp$ and $H_\parallel$ presented on Fig. 1b.

Then following the paper [7, 8] we have assumed that $H_{an,\perp} = H_{an,\parallel}$. This assumption is also valid for samples with reentrant transition.

For a chosen band of the microwave frequencies we used the Eqs. 1 and 2 for determining the temperature dependencies of M and H_{an}. The H_{an} was also determined from SQUID measurements by taking the M-H loops.

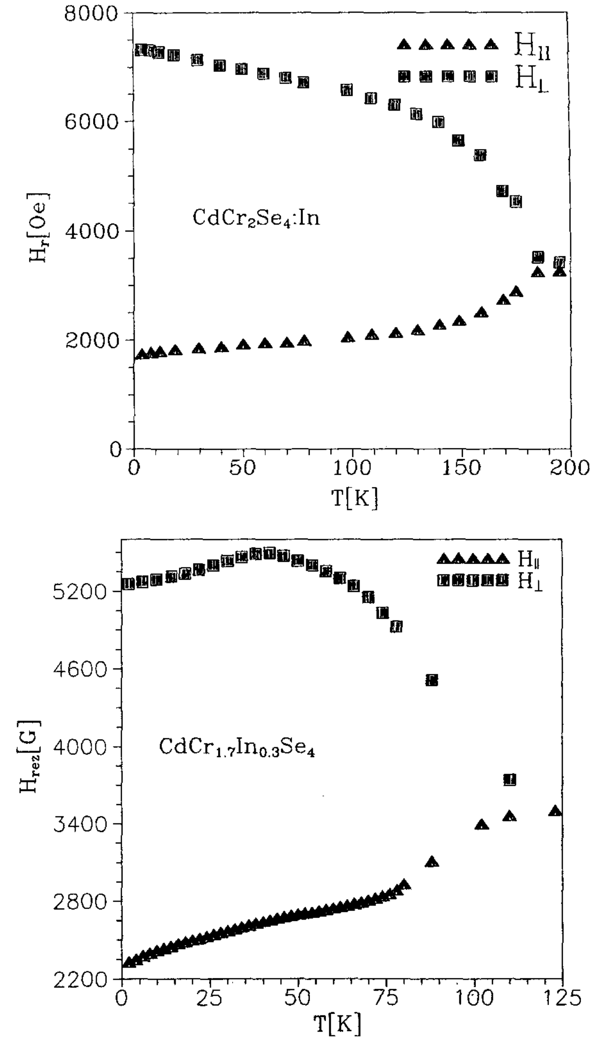

Fig. 1a, b. Temperature dependence of the resonance field for $CdCr_2Se_4$: In and $CdCr_{1.7}In_{0.3}Se_4$ thin films (X band), respectively.

The value of H_{an} was found from:

1. the displacement of M-H loop with respect to $H=0$,
2. the external magnetic field dependence of the reversible M_{rev} and the frozen irreversible M_{irr} part of the magnetization.

The hysteresis loops were measured after cooling the samples form 120 K to 4.2 K in the external magnetic filed H_{FC}. The magnetic anisotropy induced by the field cooling is unidirectional with a single easy direction of magnetization parallel to H_{FC} [3]. For a chosen value of H_{FC} several cycles of loops were recorded each loop up to a different external magnetic filed H_{max}. Asymmetrical loops were obtained until H_{max} has reached the value of Han. This filed was ascribed to the unidirectional magnetic anisotropy filed. Further increase in H_{max} produces the reversible, symmetrical M-H loops. The reversible and irreversible parts of the magnetization defined as

$$M_{rev}(H)=0.5\times[M(+H)+M(-H)] \qquad (3)$$

$$M_{irr}(H)=M(+H)+M(-H). \qquad (4)$$

were determined from the M-H loops for any arbitrary chosen value of external magnetic field H. We plotted the dependence of M_{rev} and M_{irr} on H, for several values of H_{max}. The $M_{irr}(H)=0$ when $H_{max}\geq H_{an}$.

3. Results and Discussion

3.1 Magnetization

For both types of magnetic ordering, the temperature dependence of magnetization does not obey the Bloch low.

For a state with REE transition the Blach low is modified due to the non-zero density of state at the energy gap. The temperature dependence of magnetization is describe by relation[7].

$$[M(0)-M(T)]/M(0)= BT^{3/2}/\xi(3/2)\sum_{n=1}^{\infty}[exp(-n\Delta_r/k_B T)/n^{3/2}] \qquad (5)$$

where:

$$B=\xi(3/2)[g\mu_B/M(0)](k_B/4\pi D)^{3/2}$$

$\varphi(3/2)$ stands for Riemann φ-function, Δ_r is the energy gap and D is the spin-wave stiffness constant.

For the SG state, the relation $M(T)$ also included the non-zero density of state in the energy gap.

$M(T)$ is well described by the equation [7]:

$$[M(0)-M(T)]/M(0)=C_s/[\exp(\Delta_s/k_B T)-1] \qquad (6)$$

where C_s is responsible for the density of states at the energy gap and Δ_s is a measure if the intercluster interaction.

Fig. 2 shows the temperature dependence of the magnetization, obtain from FMR data for the microwave frequency at X band, for thin films of: $CdCr_2Se_4$: In (REE) and $CdCr_{1.7}In_{0.3}Se_4$ (SG). Following the Eq. (5) the theoretical calculations for REE were done with fitting parameters $M(0)=330$ G and $\Delta_r=16$ K, wile for SG (Eq. (6)) $M(0)=162$ G, $\Delta_s=270$ K and $C_s=11.8$. The theoretically values of $M(T)$ are presented by solid lines.

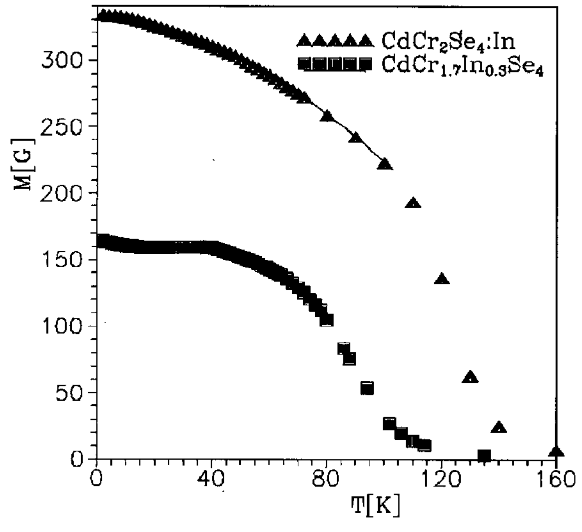

Fig. 2. Temperature dependence of the magnetization for $CdCr_2Se_4$: In and $CdCr_{1.7}In_{0.3}Se_3$ thin films. The theoretical calculation are presented by solid lines.

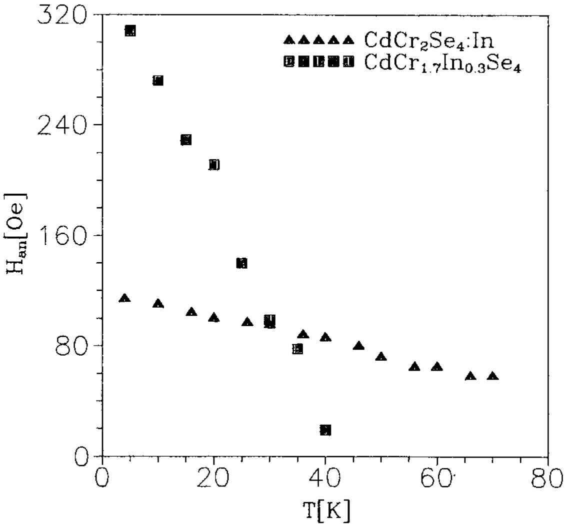

Fig. 3. Temperature dependence of the unidirectional magnetic anisotropy field H_{an} for $CdCr_2Se_4$: In and $CdCr_{1.7}In_{0.3}Se_4$ thin films.

3.2 Unidirectional magnetic anisotropy field

3.2.1 FMR data

From the experimental data of resonance field H_r vs T (see Figs. 1a, b) the $H_{an}(T)$ is determined with using the Eqs. (1) and (2). the results are presented on Fig. 3. for the films of: $CdCr_2Se_4$: In (REE) and $CdCr_{1.7}In_{0.3}Se_4$ (SG). The presented experimentally determined $H_{an}(T)$ exhibits composition and temperature dependence.

3.2.2 M-H loop data

Obtaining the temperature dependence of H_{an} from the M-H loop is long time experiment, then we perform it only for some chosen temperatures.

At T=const. the M-H loops were measured for several values of the cooling magnetic field H_{FC}.

The following procedure was used to collect data:

- for a pre-set value of HFC first cycle of M-H loop was started from the value of the external field $H=H_{FC}$,
- H was raised above H_{FC} up to a value addressed as H_{max} and the next M-H loop was started.
- the last step was repeated for the increasing values of H_{max} up to the value at which the M-H loop is symmetrical. There is the case when $H_{max}=H_{an}$.

For every cycle of the M-H loop, M_{rev} and M_{irr} were determined. The frozen part of magnetization M_{irr} tend to zero when H_{max} if getting to H_{an}. At $H_{max}=H_{an}$, $M_{irr}=0$.

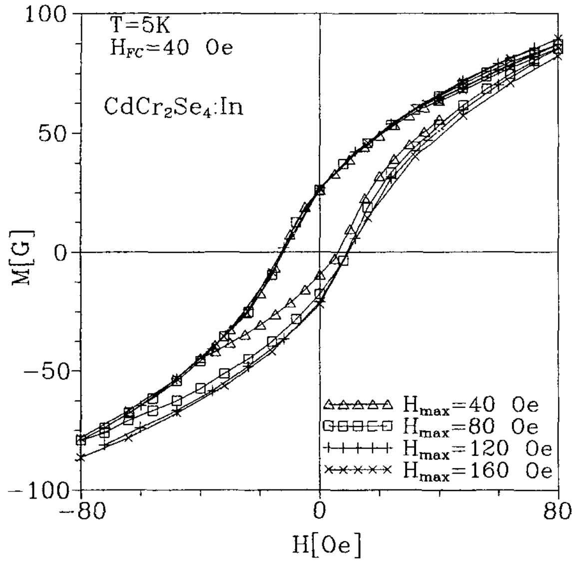

Fig 4. *M-H* loop for $CdCr_2Se_4$: In thin film taken at T_1=5 K, H_{FC}= 40 Oe.

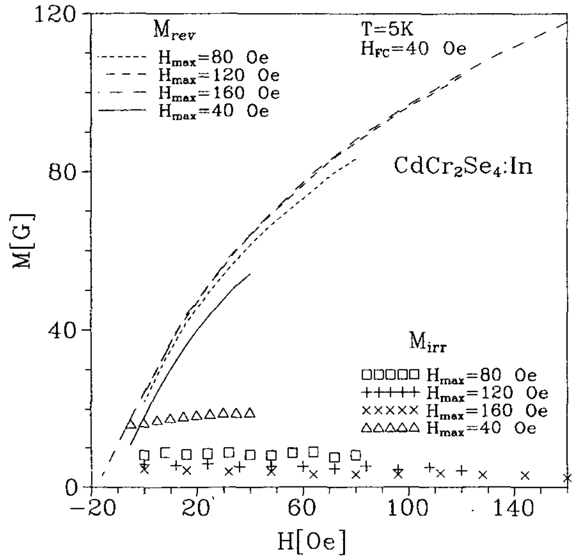

Fig. 5. M_{rev} and M_{irr} vs *H* for $CdCr_2Se_4$: In thin film for T_1=5 K, H_{FC}=40 Oe.

We are presenting the *M-H* loop, taken at *T*=5 K, for REE sample ($CdCR_2Se_4$: In). The data are seen on Fig. 4 have the symmetrical *M-H* loop H_{max}=120 Oe. Also the M_{irr} tends to zero for H_{max}=120 Oe (see Fig. 5). The same value of H_{an}=120 Oe was obtained from FMR data at *T*=5 K (see Fig. 3-$CdCr_2Se_4$: In).

For thin film of $CdCr_{1.7}In_{0.3}Se_4$ at SG state, *M-H* loop was taken for two temperature; *T*=5 K and 10 K. The data are presented on Figs. 6 and 7.

The M_{irr} and M_{rev} are shown on Figs. 8 and 9 for these two temperatures, respectively.

At *T*=5 K the *M-H* loop is getting symmetrical for H_{max} above 300 Oe (see Fig. 6) and also M_{irr} tends to zero for H_{max}=300 Oe (see Fig. 8).

The similar effect are obtained, at *T*=10 K, for H_{max}=250 Oe (see Fig. 7 and 9).

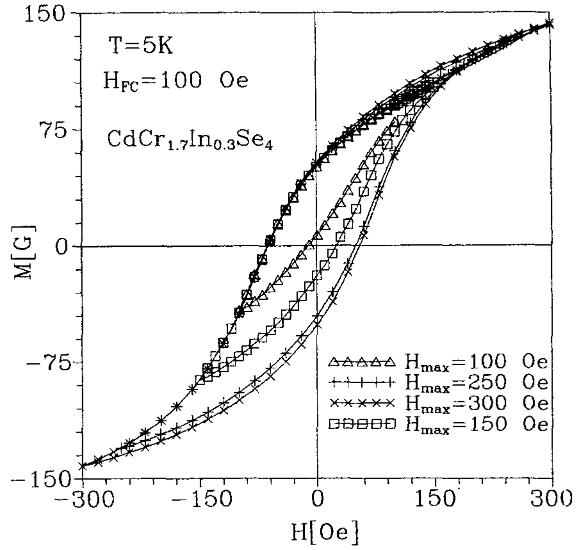

Fig. 6. *M-H* loop for $CdCr_{1.7}In_{0.3}Se_4$ thin film taken at T_1=5 K, H_{FC}=100 Oe.

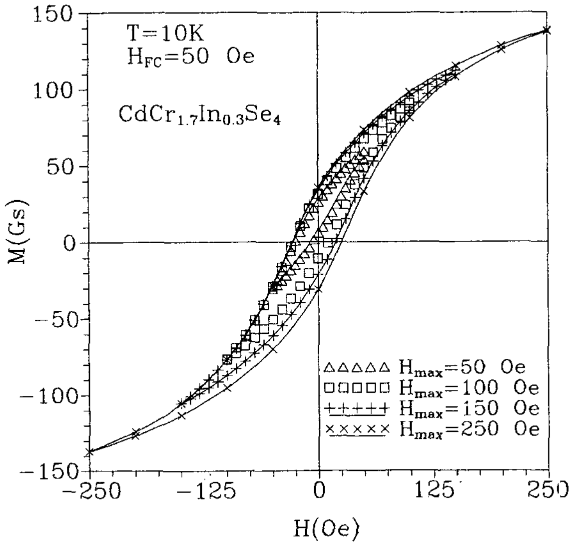

Fig. 7. *M-H* loop for $CdCr_{1.7}In_{0.3}Se_4$ thin film taken at T_1=10 K, H_{FC}=50 Oe.

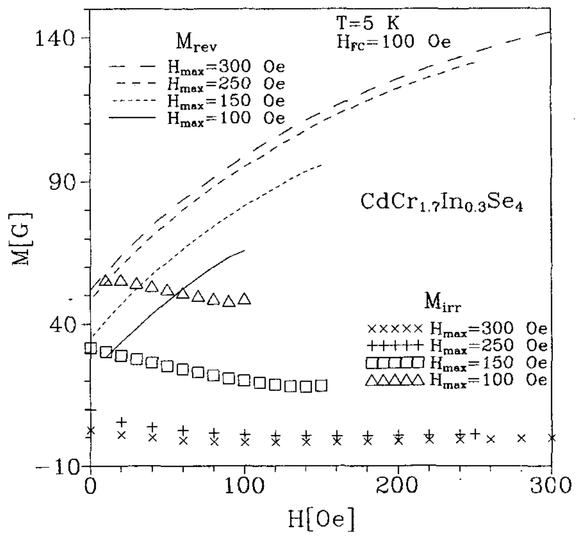

Fig. 8. M_{rev} and M_{irr} vs *H* for $CdCr_{1.7}In_{0.3}Se_4$ thin film for T_1=5 K, H_{FC}=100 Oe.

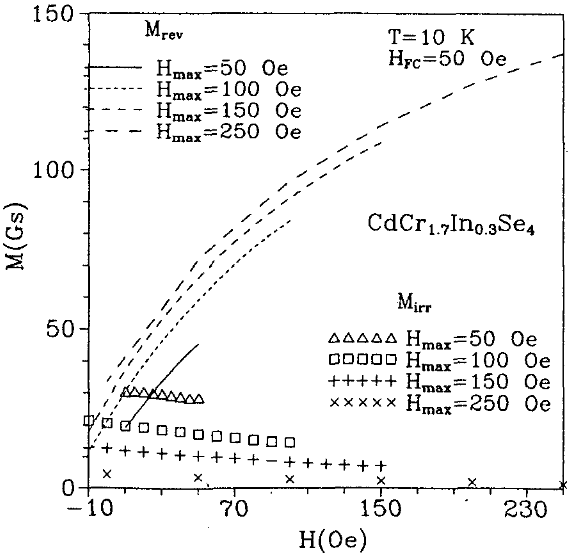

Fig. 9. M_{rev} and M_{irr} vs H for $CdCr_{1.7}In_{0.3}Se_4$ thin film for T_1=10 K, H_{FC}=50 Oe.

The values of H_{an} determined from M-H loops taken at 5 K and 10 K are in a good agreement with those obtained from FMR data for samples $CdCr_{1.7}In_{0.3}Se_4$ (see Fig. 3).

4. Conclusion

In this paper we present two technique, the ferromagnetic resonance and the M-H loop, for determining the unidirectional magnetic anisotropy field H_{an} of magnetic semiconductor thin films in the magnetic disordered systems;

- state with reentrant transition ($CdCr_{1.7}Se_4$: In)
- spin glass state ($CdCr_{1.7}In_{0.3}Se_4$).

FMR experiment at X-band was perform in the temperature range from 4.2 K to 150 K. From the shift of resonance field of uniform mode in the perpendicular and parallel configuration, the unidirectional magnetic anisotropy field was calculated.

In the second technique, prior to performing of the M-H loop, samples were cooled in the external magnetic field. Due to the unidirectional magnetic anisotropy, the processes of magnetize exhibit a displaced hysteresis cycle with respect to H=0. The M-H loops are asymmetrical for H_{max} lower than the H_{an}.

Both technique give the same value of H_{an}. Therefore, the dynamic state of magnetization which is created by the microwave field in FMR does not alter the unidirectional magnetic anisotropy field.

From our investigation we have found the significant temperature and composition dependence of H_{an}.

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