Curie temperature control by band parameters tuning in Pb$_{1-x-y-z}$Mn$_x$Sn$_y$Eu$_z$Te

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Abstract

We present the study of magnetic and transport properties of Pb$_{1-x-y-z}$Mn$_x$Sn$_y$Eu$_z$Te. AC magnetic susceptibility measurements as well as transport characterization were performed. The obtained results indicate that the presence of two types of magnetic ions influences the magnetic properties of the investigated IV–VI semimagnetic semiconductor. Qualitative analysis and possible mechanisms of the substantial dependence of the Curie temperature on the Eu content are presented. The most likely reason of the observed Curie temperature behaviour is a strong dependence of the location of a heavy mass $\Sigma$ band upon the alloy composition. Theoretical calculations in frame of simple models confirm this point.

1. Introduction

Manipulation of the spin degree of freedom in semiconductors has become a focus of interest in recent years. In the context of spin electronics, particularly interesting are ferromagnetic and semimagnetic (diluted magnetic) semiconductors (SMSCs). Understanding of the carrier-mediated ferromagnetism was initiated by a study of ferromagnetism in IV–VI based SMSCs. In this class of materials, deviations from stoichiometry result in the carrier density sufficiently high to produce strong ferromagnetic interactions between the localized spins. It was shown that in Pb$_{1-y-z}$Mn$_x$Te mixed crystals with high Sn concentration, $y \geq 0.6$, the free hole concentration can be varied by means of isothermal annealing in the range between 10$^{20}$ and 10$^{21}$ cm$^{-3}$ [1].

In the present paper, we consider the effect of the presence of two types of magnetic ions incorporated into semiconductor matrix on magnetic properties of the resultant semimagnetic semiconductor. In order to simplify the theoretical description of the investigated magnetic system, two types of magnetic ions were chosen with spin-only ground state: substitutional Mn$^{2+}$ possesses $S = 5/2$, while Eu$^{2+}$, the second type of magnetic ion, has $S = 7/2$. There are several reasons for which the magnetic semiconductors based on lead chalcogenides are ideal materials for such kind of investigations. First, a variety of magnetic properties has been observed in Mn-based IV–VI SMSCs. Second, the characteristic features are semi-metallic electric properties with well-developed methods of control of carrier concentration. Pb$_{1-x-y-z}$Mn$_x$Sn$_y$Eu$_z$Te is a unique system in which the interplay between magnetic and electronic properties can be observed and studied. In particular, carrier-induced paramagnet–ferromagnet as well as ferromagnet–spin glass transitions have been observed [1–3]. This is due to the combination of an RKKY type of interaction between...
the magnetic ions with the possibility of manipulating the free carrier concentration. These two features give IV–VI semimagnetic materials a distinguished position within the whole family of semimagnetic semiconductors. The additional advantage is that for the \( \text{Pb}_{1-x-y-z} \text{Mn}_{2-y} \text{Sn}_{y} \text{Eu}_z \text{Te} \) crystals, the parameters of the energy structure are very well known.

All known Eu-based IV–VI semimagnetic lead chalcogenides with electrons as well as hole density below \( 10^{19} \text{ cm}^{-3} \) are paramagnetic down to the temperature \( T = 1 \text{ K} \) (similarly to Mn-based compounds). A strongly localized character of 4f orbitals of rare earth ions results in very weak exchange interactions both between the magnetic ions and between ions and the free carriers. As a result, the \( \text{Sn}_{1-x} \text{Eu}_{x} \text{Te} \) crystals are not ferromagnetic. The reason for a lack of ferromagnetism in this material is the very small magnitude of the sp–f exchange integral. It has been known for several years that the Eu–Eu exchange interaction in these IV–VI SMSCs is two orders of magnitude smaller than the Mn–Mn exchange interaction in the traditional IV–VI SMSCs [4–6]. The Mn-based IV–VI semimagnetic semiconductors with relatively low concentration of free carriers, for instance \( \text{PbMnTe} \), are paramagnets above \( T = 1 \text{ K} \). Their magnetic behaviour closely resembles that of the Mn containing II–VI SMSCs and can also be attributed to antiferromagnetic interactions of the superexchange type, although the interactions are much weaker than in II–VI semimagnetic semiconductors. In this paper, we explore the influence of the presence of two types of magnetic ions, Mn\( ^{2+} \) and Eu\( ^{2+} \), in the semiconductor matrix on the magnetic properties of resultant SMSC. Samples of \( \text{Pb}_{1-x-y-z} \text{Mn}_{2-y} \text{Sn}_{y} \text{Eu}_z \text{Te} \) with different concentrations of Mn \( x \leq 0.13 \) and Eu \( z \leq 0.02 \) as well as free hole concentration \( (p \leq 2 \times 10^{21} \text{ cm}^{-3}) \) were systematically investigated. The objective of this work was to perform systematic measurements of magnetic ac susceptibility as well as transport characterization of bulk samples of \( \text{Pb}_{1-x-y-z} \text{Mn}_{2-y} \text{Sn}_{y} \text{Eu}_z \text{Te} \) multinary alloy in a wide range of concentrations of magnetic ions as well as carrier concentrations. The magnetic measurements were carried out in the temperature range \( 1.3–150 \text{ K} \) and ac magnetic field of amplitude not exceeding 5 Oe and frequency range \( 7–10000 \text{ Hz} \). The standard dc six probe technique at static magnetic field up to 1 T was used for Hall measurements. The paramagnet–ferromagnet as well as ferromagnet–spin glass transitions were observed and studied.

2. Sample preparation and characterization

The crystals of \( \text{Pb}_{1-x-y-z} \text{Mn}_{2-y} \text{Sn}_{y} \text{Eu}_z \text{Te} \) were grown by a modified Bridgman method. In the present work, samples coming from several technological processes were investigated. The chemical composition of the samples was determined by the x-ray dispersive fluorescence analysis technique with uncertainty of 10%. Typically, the crystals were cut crosswise the growth axis to 1–2 mm thick slices. The variation of chemical composition along this area is very small (1–2%). The results of chemical analysis of all investigated \( \text{Pb}_{1-x-y-z} \text{Mn}_{2-y} \text{Sn}_{y} \text{Eu}_z \text{Te} \) samples are gathered in table 1.

The standard powder x-ray measurements revealed that the investigated samples are single-phase and crystallize in a NaCl structure, similarly to the nonmagnetic matrix and \( \text{Pb}_{1-x-y-z} \text{Mn}_{2-y} \text{Sn}_{y} \text{Eu}_z \text{Te} \) semimagnetic semiconductor. The measured values of lattice constants for several \( \text{Pb}_{1-x-y-z} \text{Mn}_{2-y} \text{Sn}_{y} \text{Eu}_z \text{Te} \) samples are collected in table 2. For comparison, the values of the calculated lattice constant for \( \text{Pb}_{1-x-y-z} \text{Mn}_{2-y} \text{Sn}_{y} \text{Eu}_z \text{Te} \) crystals with analogous content of Mn and Sn are also presented [7]. The introduction of Mn ions into the nonmagnetic matrix of \( \text{Pb}_{1-x-y-z} \text{Sn}_{y} \text{Te} \) leads to a decrease in the lattice constant of the resultant \( \text{Pb}_{1-x-y-z} \text{Mn}_{2-y} \text{Sn}_{y} \text{Te} \) [8].

The Hall bar samples with typical dimensions of 8 mm × 2 mm × 1 mm were used for the transport measurements. The Hall voltage \( V_H \) as well as conductivity voltage \( V_H \) were measured. The electrical contacts were always prepared in the same way. First the surface of the specimens was etched using a solution of Br\( _2 \) and HBr in the proportion 1:20. Next, the gold contacts were deposited by use of gold chloride water solution on the polished surface of the samples. Finally, the electrical contacts were made using indium solder and gold wires. The typical resistance of the samples was equal to about 1 mΩ. This allowed us to apply a relatively large current (up to 300 mA). The Hall as well as conductivity measurements were performed at room and liquid-nitrogen temperature. The standard dc six probe technique at static magnetic field up to 1 T was used. In the present paper, only a nominal Hall concentration \( p \) was determined at room and liquid-nitrogen temperature. All the investigated samples were \( p \) type with high and almost temperature-independent hole concentration (in the range between \( 2 \times 10^{16} \text{ cm}^{-3} \) and \( 2 \times 10^{21} \text{ cm}^{-3} \)). The values of free hole concentration obtained at room temperature are gathered in table 1. Typical values of mobility were in the range of a dozen and a few dozen cm\(^2\) V\(^{-1}\) s\(^{-1}\). The values of carrier concentration, conductivity and mobility were determined with uncertainty of 15% at room temperature and 30% at liquid-nitrogen temperature.

3. Magnetic investigations

In this section, the results of magnetic studies of \( \text{Pb}_{1-x-y-z} \text{Mn}_{2-y} \text{Sn}_{y} \text{Eu}_z \text{Te} \) samples are presented. AC magnetic susceptibility studies in the temperature range 1.3–150 K using a mutual inductance method were carried out. The susceptibility measurements were carried out in an ac magnetic field of frequency range 7–10000 Hz and amplitude not exceeding 5 Oe.

Generally, in the range of high temperatures all IV–VI semimagnetic semiconductors are Curie–Weiss
paramagnets with the temperature dependence of the magnetic susceptibility described by the Curie–Weiss law,

$$\chi(T) = C/(T - \Theta)$$

(1)

where $C = g^2 \mu_B^2 S(S + 1)N_M$ is the Curie constant and $k_B \Theta = (1/3)S(S + 1) \sum z_i I(R_i)$ is the paramagnetic Curie temperature (Curie–Weiss temperature). Here, $N_M$ is the concentration of magnetic ions, $z_i$ the number of magnetic neighbours on the $i$th crystallographic shell, $I(R_i)$ the exchange integral between the central ion and its $i$th magnetic neighbours, $S$ the spin of the magnetic ion, $g$ the spin-splitting g-factor, $k_B$ the Boltzmann constant, and $\mu_B$ is the Bohr magneton.

For all the investigated samples the high temperature behaviour of the inverse low-field susceptibility $\chi^{-1}$ was nearly linear and all data fit well to the Curie–Weiss law of the form

$$\chi(T) = C/(T - \Theta) + \chi_{\text{dia}}$$

(2)

where $\chi_{\text{dia}}$ is the susceptibility of the host lattice (all IV–VI semiconductors without magnetic ions are standard diamagnetic materials with the magnetic susceptibility around $\chi_{\text{dia}} \simeq -3 \times 10^{-6}$ emu g$^{-1}$).

In the case of PbMnEuTe crystals, the obtained negative and relatively small values of the paramagnetic Curie–Weiss temperature $\Theta$ indicate that a weak antiferromagnetic superexchange interaction is a dominant mechanism of the interaction. These results correspond to those reported earlier for PbMnTe. The determined values of the paramagnetic Curie temperature $\Theta$ and Curie constant $C$ are presented in table 1.

### Table 2. The lattice constant $a_0$ of Pb$_{1-x-y}$Mn$_x$Sn$_y$Eu$_z$Te samples determined by the standard powder x-ray measurements and the values of the calculated lattice constant of Pb$_{1-x-y}$Mn$_x$Sn$_y$Te $a$ [7] with similar content of Mn and Sn.

| Number of the sample | $x$ | $y$ | $a_0$ (Å) | $a$ (Å) |
|---------------------|----|----|-----------|--------|
| 809,2               | 0.031 | 0.0027 | 0.85 | 6.3130 | 6.2866 |
| 809,4               | 0.030 | 0.0016 | 0.85 | 6.3237 | 6.2876 |
| 809,12              | 0.022 | 0.00031 | 0.76 | 6.3735 | 6.3113 |
| 809,28              | 0.020 | 0.0076 | 0.73 | 6.3563 | 6.3309 |
| 809,36              | 0.025 | 0.013 | 0.69 | 6.3427 | 6.3311 |
crystals. This indicates the presence of a ferromagnetic interaction. The obtained values of Curie–Weiss temperature Θ and Curie constant C are shown in table 1. (The high temperature magnetic susceptibility measurements were not performed for all SnMnEuTe samples.) Careful inspection of table 1 allows us to note significant changes of Curie–Weiss temperature with the Eu content. Decrease of the paramagnetic Curie temperature Θ with increase of the Eu concentration is clearly visible. The three samples of Pb1−x−y−zMnSnEuTe: 809_12, 809_30, 809_34 are characterized with very similar values of the Mn content and concentration of free holes (see table 1). For the Mn concentration equal to around x = 0.02 and free hole concentration p = 4 × 10^{20} cm^{-3}, increase of the Eu content from z = 0.003 to z = 0.01 leads to the decrease of Curie–Weiss temperature from 4.55 K to 3.02 K, and for z = 0.017 paramagnetic Curie temperature is equal to 2.63 K. In the case of Sn1−x−y−zMnEuTe crystals, such a distinct tendency is not observed (see table 1). However, one needs to realize that the obtained values of chemical composition as well as free carrier concentration are determined with quite large uncertainty.

The low temperature studies revealed the presence of paramagnet–ferromagnet phase transition in the case of SnMnEuTe as well as PbSnMnEuTe samples. Figures 2 and 3 show the low temperature behaviour of the real component of susceptibility Re(χ) for several samples of studied Pb1−x−y−zMnSnEuTe and Sn1−x−y−zMnEuTe mixed crystals. Typical behaviour of a ferromagnet is observed. Both real and imaginary components of the susceptibility dramatically increase at the Curie temperature TC. The Curie temperature was determined by the maximum slope of dRe(χ)/dT. The values of TC are approximately equal to the Curie–Weiss temperature Θ determined from the high temperature susceptibility measurements. The low temperature measurements confirmed the above-described tendency for studied Pb1−x−y−zMnSnEuTe crystals, i.e., the decrease of Curie temperature with the Eu content.

For the sample 809_2 of PbSnMnEuTe, the ferromagnet to spin glass phase transition is observed. Figure 4 presents the characteristic behaviour of low temperature parts of the real and imaginary components of the susceptibility for the spin glass (809_2) as well as for the ferromagnetic (809_12) samples of Pb1−x−y−zMnSnEuTe. In the case of the ferromagnetic sample with the concentration of free holes equal to 4.0 × 10^{20} cm^{-3}, sharp transitions in both real and imaginary components of susceptibility occur. For the spin glass sample characterized by higher free hole concentration p = 1 × 10^{21} cm^{-3}, a cusp in Re(χ) is visible at the freezing temperature TC. The magnitude of the susceptibility at this cusp is much lower than the susceptibility of the ferromagnetic sample. A corresponding maximum in the out of phase susceptibility Im(χ) is observed at slightly lower temperature. The 809_2 PbSnMnEuTe sample shows an obvious characteristic of the spin glass-like phase. The cusp observed in the susceptibility χ versus temperature T
The relative shift of freezing temperature $T_f$ per decade of frequency $f$ is much smaller than the $J_{s-f}$ interaction. Thus, the indirect RKKY coupling between Eu ions is negligibly small. In addition, the Eu ion does not feel well any possible magnetization of the electronic system. As a result, we should not totally exclude a small contribution from the $s$–$f$ coupling between Eu atoms and carriers. Obviously, it would lead to a weak increase of the Curie temperature with the Eu content. Since the experiment shows the opposite behaviour, we can assume that the role of Eu as a magnetic impurity is negligibly small.

On the other hand, EuTe is known to be antiferromagnet. Thus, one can expect a transition from positive (Curie) to negative (Néel) temperature of magnetic ordering of $Pb_{1-x-y-z}Mn_{x+y+z}Sn_{x}Eu_{y}Te$ alloy when changing the Eu content $z$ from 0 to 1. However, the Eu–Eu interaction is not mediated by the free carriers, and, therefore, it is short ranged. For a small Eu content, it leads to antiferromagnetic ordering for a very small number of Eu–Eu pairs, which do not affect the interaction mechanism between the Mn ions. If some of the Eu–Eu neighbouring pairs are ordered antiferromagnetically, we can only expect that they do not contribute to the susceptibility, which is mostly related to the ferromagnetic Mn–Mn interaction. We conclude that the suppression of Curie temperature by a possible antiferromagnetic ordering of Eu–Eu pairs looks very unlikely.

The incorporation of Eu atoms can affect the properties of IV–VI compounds in a different way, which is not directly related to the magnetism of Eu ions. Eu is a component of the complex PbMnEuSnTe alloy, and one should consider a variation of the band parameters as a function of the Eu content. For a qualitative analysis, let us assume here a simplified two-band model described by some phenomenological parameters. Note that the real energy shifts to higher temperatures when the frequency $f$ of the applied ac field is increased. This feature—the increase of the freezing temperature when the frequency is higher—was observed in many well-known canonical spin glass systems [9–12].

First, the Eu atom in the PbMnSnTe matrix is a magnetic impurity with spin-only ground state: Eu$^{2+}$ has $S = 7/2$. The electrons of the half-filled f-shell responsible for the moment are very weakly coupled to the band electrons. The coupling constant $J_{s-f}$ is much smaller than the $J_{s-d}$ interaction. Thus, the indirect RKKY coupling between Eu ions is negligibly small. In addition, the Eu ion does not feel well any possible magnetization of the electronic system. As a result, we should not totally exclude a small contribution from the $s$–$f$ coupling between Eu atoms and carriers. Obviously, it would lead to a weak increase of the Curie temperature with the Eu content. Since the experiment shows the opposite behaviour, we can assume that the role of Eu as a magnetic impurity is negligibly small.

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The energy spectrum in the L-points of the Brillouin zone can be described by the Dirac model

\[ E_L(k) = \frac{\hbar^2 k^2}{2m^*_\Sigma}, \]

where \( m^*_\Sigma \approx 3m_0 \), and \( m_0 \) is the free electron mass. It is commonly known that the energy spectrum of Pb\(_{1-x}\)Eu\(_x\)Te alloy is very sensitive to the concentration of Eu. The energy gap \( E_g \) depends strongly on the Eu content—ranging from 189.7 meV for \( z = 0 \) (PbTe) to 248 meV at \( z < 0.013 \) at the temperature \( T = 10 \) K [17], \( \Delta E_g / \Delta z = 5.788 \) eV at \( T = 10 \) K for \( z < 0.05 \) [18]. Considering the energy gap \( E_g \) dependence on Sn content \( z \) of Pb\(_{1-x-y}\)Mn\(_x\)Sn\(_y\)Te alloy [17, 18], the following formula describing the energy gap dependence on the alloy composition in Pb\(_{1-x-y}\)Mn\(_x\)Sn\(_y\)Eu\(_z\)Te crystals can be assumed:

\[ E_g = 0.19(1 - y) - 0.3y + 5.788z \text{ [eV]}. \]

In the present calculations, the Mn concentration \( x \) was accepted as equal to 0.02.

We also assume [17, 19] that the composition dependence of \( \epsilon_0 \) parameter has the following form:

\[ \epsilon_0 = 0.81(1 - y) + 0.3y + 8z \text{ [eV]}. \]

Using equations (4) and (5) the hole concentration in L as well as in \( \Sigma \) valley can be found:

\[ p_L = \left( \frac{E_f - \Delta^*}{3\pi^2 v^3} \right)^{3/2}, \]

\[ p_\Sigma = \frac{1}{3\pi^2} \left( \frac{2m^*_\Sigma}{\hbar^2} (E_f - \epsilon_0) \right)^{3/2}. \]

The Fermi energy \( E_f \) is determined by the equation:

\[ 4p_L + 12p_\Sigma = p_0. \]

which gives the total hole concentration \( p_0 \) and takes into account the degeneracy of each valley. It is accepted here that \( p_0 \) is equal to \( 4 \times 10^{20} \text{ cm}^{-3} \).

The Fermi momentum in the \( \Sigma \) band \( k^F_\Sigma = [2m^*_\Sigma/(E_F - \epsilon_0)]^{1/2} \) can be found as a solution of equations (8)–(10).

Next, equation (3) with \( k^F = k^F_\Sigma \), \( R = (3a_0^2/4\pi x)^{1/3} \), \( a_0 = 6.5 \times 10^{-8} \) cm, \( N = 12 \), \( J_{sd} = 1 \) eV is used for \( T_c \) calculations.

It implies that the main contribution to the RKKY interaction between Mn ions is related to the heavy holes in \( \Sigma \) bands.

The obtained \( T_c \) dependence on Eu concentration \( z \) for various values of the Sn content (0.6 \( \leq z \leq 1 \)) as well as the experimental data (Curie temperature determined for samples with similar Mn content \( x \approx 0.02, y \approx 0.7 \) and free hole concentration \( p \approx 4 \times 10^{20} \) cm\(^{-3}\)) is shown in figure 6.

The calculated values of Curie temperature are higher than those determined experimentally. However, it should be stressed here that a very simplified model was used, not all phenomenological parameters of the model are known precisely, and some simplifying assumptions were used. Nevertheless, the results of our calculations can well explain the observed experimentally tendency of Curie temperature decrease with the increase of the Eu content \( z \).

\[ \text{Figure 6. Comparison of experimentally determined dependence of Curie temperature on Eu concentration } z \text{ for Pb}_{1-x-y}\text{Mn}_x\text{Sn}_y\text{Eu}_z\text{Te samples with similar Mn content } x \approx 0.02, \text{Sn content } y \approx 0.7 \text{ and free hole concentration } p \approx 4 \times 10^{20} \text{ cm}^{-3} \text{ with the results of calculations.} \]

\[ \text{The simplified two-band model reflects the experimentally observed tendency, i.e. Curie temperature decrease with the increase of Eu. The theoretical curves were obtained for various values of Sn concentration } 0.6 \leq y \leq 1 \text{ and Mn concentration } x = 0.02. \]

\[ \text{which gives the total hole concentration } p_0 \text{ and takes into account the degeneracy of each valley. It is accepted here that } p_0 \text{ is equal to } 4 \times 10^{20} \text{ cm}^{-3}. \]

\[ \text{The Fermi momentum in the } \Sigma \text{ band } k^F_\Sigma = [2m^*_\Sigma/(E_F - \epsilon_0)]^{1/2} \text{ can be found as a solution of equations (8)–(10).} \]

\[ \text{Next, equation (3) with } k^F = k^F_\Sigma, R = (3a_0^2/4\pi x)^{1/3}, a_0 = 6.5 \times 10^{-8} \text{ cm, } N = 12, J_{sd} = 1 \text{ eV is used for } T_c \text{ calculations.} \]

\[ \text{It implies that the main contribution to the RKKY interaction between Mn ions is related to the heavy holes in } \Sigma \text{ bands.} \]

\[ \text{The obtained } T_c \text{ dependence on Eu concentration } z \text{ for various values of the Sn content (0.6 } \leq z \leq 1 \text{) as well as the experimental data (Curie temperature determined for samples with similar Mn content } x \approx 0.02, y \approx 0.7 \text{ and free hole concentration } p \approx 4 \times 10^{20} \text{ cm}^{-3} \text{ is shown in figure 6.} \]

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\[ \text{5. Summary} \]

In this paper, the results of magnetic and transport studies of Pb\(_{1-x-y}\)Mn\(_x\)Sn\(_y\)Eu\(_z\)Te multinary alloys are reported. The following results were obtained.

The presence of two types of magnetic ions (Mn and Eu) in the IV–VI semiconductor matrix influences the magnetic properties of the resultant magnetic semiconductor. The results of magnetic measurements show that the Curie temperature \( T_c \) as well as the Curie–Weiss temperature \( \Theta \) decrease with an increase of the Eu content in
Curie temperature control by band parameters tuning in Pb$_{1-x-y-z}$Mn$_x$Sn$_y$Eu$_z$Te samples. The magnetic susceptibility measurements also revealed that Eu changes the spin glass dynamics in this material. The difference in the rate of frequency shift of cusp in the real and imaginary parts of susceptibility is visible. Such behaviour was not observed for Mn-based IV–VI magnetic semiconductors.

Our qualitative analysis shows that a variation of the band parameters with the alloy composition can be responsible for the observed strong dependence of the Curie temperature on the Eu content. A simple two-band model explains both the order of the transition temperature values and $T_C$ dependence on Eu concentration. The calculated dependence of Curie temperature on the Eu content reflects very well the experimentally confirmed effect of $T_C$ decrease with Eu concentration $z$.

The observed dependence of the Curie temperature on the Eu content clearly demonstrates that the magnetism of semiconductors can be effectively controlled by using an energy band structure dependence on alloy composition. The situation with several non-equivalent energy minima is not unique for IV–VI semiconductors. The similar effect exists also in Ge–Si alloys and in other semiconductors (such as high-energy minima in GaAs). We believe that it can be used to increase the critical temperature of ferromagnetism in magnetically-doped semiconductors.

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