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Magnetic Properties of Bulk Amorphous La-Fe Alloys*

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Synopsis

We have prepared three bulk amorphous $La_{100-x}Fe_x$ alloys ($x=69.4$, 76.7 and 82.5 at.% Fe) by a high rate dc-triode sputtering. All of the samples are found to be amorphous by neturon diffraction measurements.

The amorphous Fe alloys with nonmagnetic Y and Lu are reported to have spin-glass characteristics, while the La-Fe alloys are found to be ferromagnetic. In the latter alloys, the magnetizations are easily saturated in low applied fields, and their values suggest that the Fe moments are completely aligned. Furthermore it has been found that the saturation magnetic moment has a maximum value 1.7 μ_B per Fe atom at around $x=75$, while the Curie temperature tends to decrease gradually with increasing Fe concentration.

The temperature dependence of the magnetization for these alloys follows the spin-wave relation

$$M(T) = M_0(1-BT^{3/2}-CT^{5/2}) .$$

Using the values of B and C obtained, we have estimated the mean range of the Fe-Fe exchange interaction ($\langle r^2 \rangle$).

The range of the exchange increases sharply from $\sim 2\text{Å}^2$ to $\sim 100\text{Å}^2$ with increasing Fe concentration and exceeds that of crystalline Fe.

I. Introduction

Recent development in amorphous magnetic materials, particularly those alloys containing rare earths (R) and transition metals (M) have drawn considerable attention(1-5). The R-M alloys present a convenient opportunity to study the magnetic behavior in structurally amorphous materials because they are easily produced, and because any combination of R and M is available in a large range of of x for $R_{1-x}M_x$ alloys. In addition, a number of different compositional crystalline phases exist in these R-M alloys. This allows a direct comparison with corresponding crystalline compounds.

Heiman and Kazama(6) have attempted to isolate the most important M-M

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magnetic interaction in the *R-M* amorphous alloys by preparing Y-Co or La-Co alloys. In both cases the amorphous samples show simple ferromagnetic behavior similar to the case of crystalline system. However, the concentration of La or Y in which ferromagnetic behavior exists is more extended than for the crystalline systems.

The same attempts have been carried out by a number of researchers(7–11) to isolate the Fe-Fe interaction in the *R-M* amorphous alloys by preparing alloys of Y-Fe or Lu-Fe. In all cases the samples show complex magnetic behavior that has been characterized as spin-glass or spin-glass like. Recently amorphous La-Fe samples(12) are found to be ferromagnetic in contrast to Y-Fe and Lu-Fe alloys.

We should not ignore the situation that while the Y-Fe and Lu-Fe system have some crystalline compounds which would influence the local environments and thermal stability, La-Fe system does not form any intermediate phase at all(13). This also suggests that the short range Fe-Fe exchange interaction in amorphous state depends upon the local structure arising from intermediate phases.

In this report we have simplified this situation by selecting La-Fe system, in which the Fe magnetic behavior could be isolated from intermediate phase effects as well as other magnetic atoms, and we have determined how the magnetic moment of Fe and the magnetic ordering temperature change upon alloying with nonmagnetic element La. It is also noted here that a high rate sputtering technique offers us a large amount of bulk amorphous samples which is desirable for carrying out detailed measurements free from the substrate effects.

II. Sample preparation and experimental procedure

The massive amorphous deposits (0.5∼0.8 mm thick and 30 mm diameter) were prepared using a high rate dc-supported discharge sputtering system* from three targets whose composition ratios of La to Fe were 1:3, 1:5 and 1:9 respectively.

Since the physical properties of sputtered samples depend on the various factors as reported for the amorphous Fe-C system(14), and Gd-Co, Gd-Fe and Gd-Co-X (X=Mo, Cu, Au) alloys(15). We fixed the sputter deposition condition as

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* The details of the sputtering apparatus have been described in the separate paper in this Supplement.

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the following.

(1) The sputtering gas was high purity Ar at a pressure of 3×10^{-2} Torr and was ionized by the passage of high current (~10A), low voltage (~30V) arc discharge between the tungsten filament thermonic cathode and a water cooled anode.

(2) A negative target potential of 12000 V dc was used to accelerate the Ar ions from the plasma to target surface.

(3) The target current density was kept constant at 7 mA/cm^2.

(4) The substrate was metallographically polished Cu discs and were cooled directly by water.

No substrate bias voltage was used to avoid the inclusion of impurities as well as Ar gas. For example, the Ar content of the rf-sputtered amorphous alloys peaks near -200V for the bias voltage and reaches nearly 16 at.% in Gd-Co alloys(15). From these condition, we obtained the amorphous alloys with deposition rate about 1000 Å/min.

The compositions thus obtained were 69.4 at.% Fe, 30.6 at.% La (target $LaFe_3$), 76.7 at.% Fe, 23.3 at.% La (target $LaFe_5$) and 82.5 at.% Fe, 17.5 at%. La (target $LaFe_9$) by the chemical analysis.

There was no trace of the sputtered Ar gas and Cu diffusion from the substrate. These deposited alloys were studied by means of neutron diffraction, differential specific heat and electrical resistivity as well as magnetization measurements. The magnetization as a function of applied fields and temperatures was measured using a recording magnetometer. A Cu-Au(Co) thermocouple in contact with the sample was used to measure the low temperature.

III. Results and discussion

1. Neutron diffraction

Neturon diffraction measurements were carried out to examine the structure of the sputtered bulk alloys. Figure 1 shows some of the diffraction patterns of the as-deposited La-Fe alloys, which resembled those reported for morphous structure. The line broadening of the first diffraction peak corresponds to a coherently diffracting domain size of ~10Å, which may be interpreted as high degree of short range order as in the amorphous $Fe_{50}W_{50}$ structure(16). The detailed structure analysis is under way with combination of the polarized neutron diffraction measurements.

2. Temperature dependence of electrical resistivity

The electrical resistivity for the amorphous $La_{17.5}Fe_{82.5}$, shown in Fig. 2, increases up to 357°C with the slope of 0.34 $\mu\Omega\cdot$cm/°C. Then a gradual decrase of

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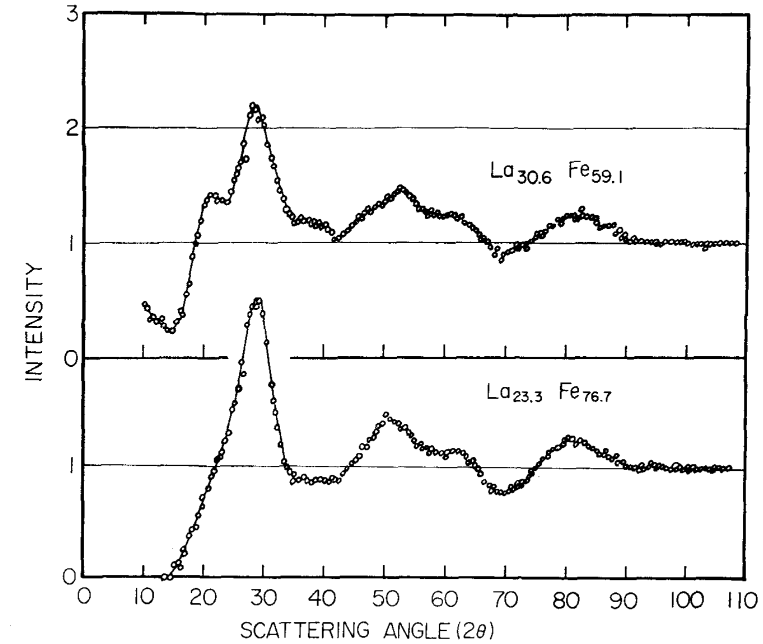

Fig. 1. Neutron diffraction patterns after subtracting the back ground for the massive amorphous La-Fe alloys. The measurements were carried out by the TOG spectrometer at JRR-3 with the neutron wave length 1.03 Å.

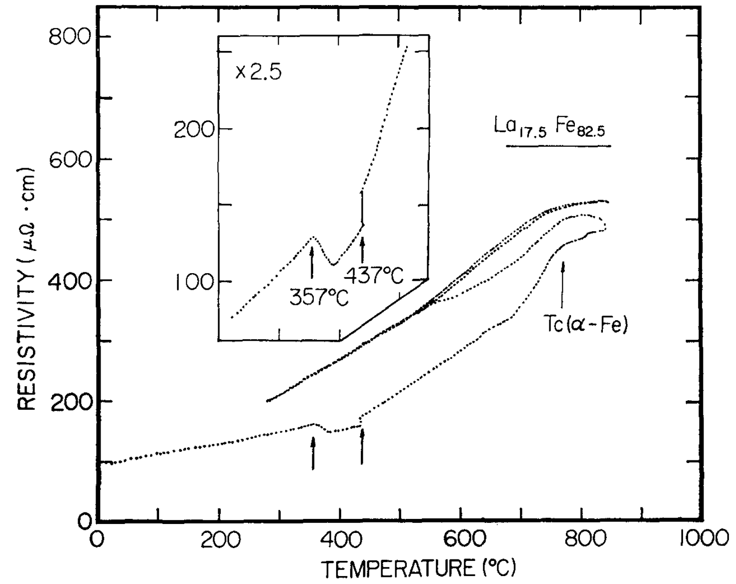

Fig. 2. Irreversible change of the electrical resistivity with increasing and decreasing temperatures.

resistivity occurs around 360°C which corresponds to the onset of α-Fe precipitation as found from the magnetization measurements. After an abrupt increase of resistivity (15.3%) is observed at 437°C, which corresponded to the completion of α-Fe precipitation, it increases with the higher slope of 1.40 $\mu\Omega\cdot$cm/°C. There is a large hysteresis after the crystallization, which may suggest the existence of some

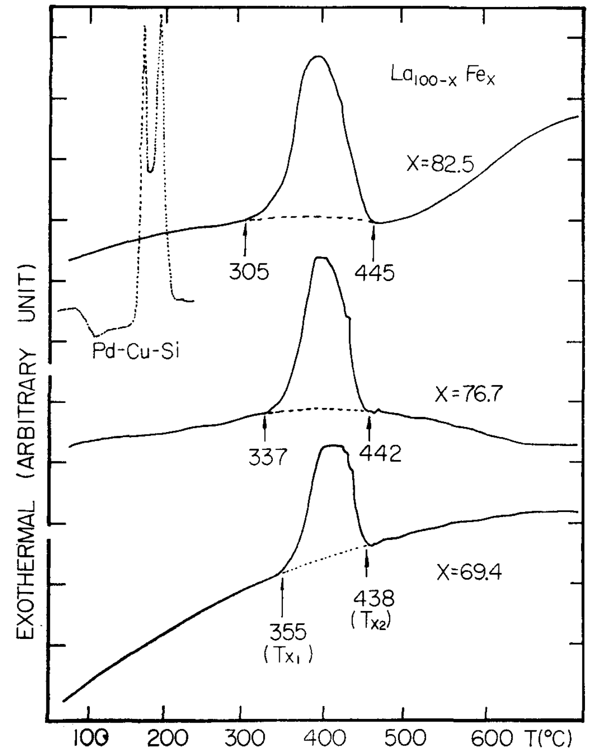

Fig. 3

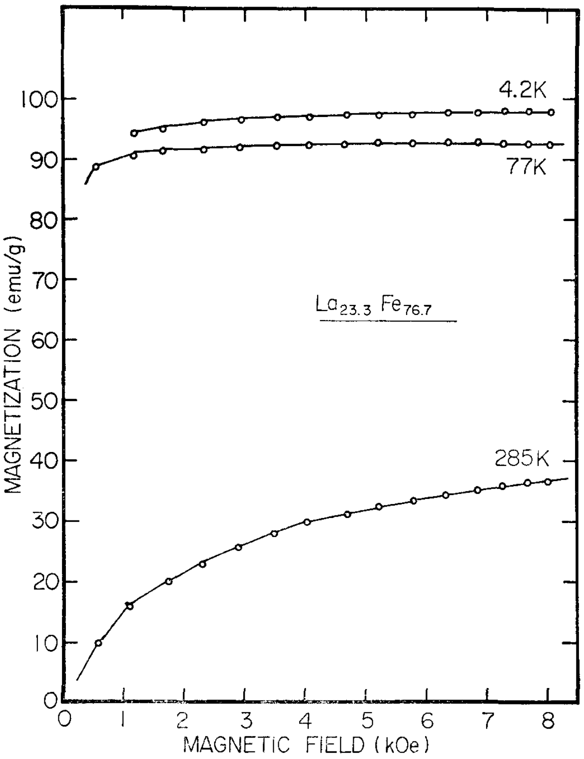

Fig. 4

Fig. 3. The temperature dependence of the differential specific heat of the amorphous La-Fe alloy system.

Fig. 4. Magnetic field dependence of the magnetization for the amorphous $La_{23.3}Fe_{76.7}$ alloy. The magnetization saturates at low fields, implying a simple ferromagnetic behavior.

LaFe compounds formed in the higher temperature region.

3. Temperature dependence of differential specific heat

We obtained the crystallization temperatures (T_{x_1}) for these amorphous La-Fe alloys by the measurements of the differential specific heat.

The crystallization starts at 355°C, 337°C and 305°C for $La_{30.6}Fe_{69.4}$, $La_{23.3}Fe_{76.7}$ and $La_{17.5}Fe_{82.5}$ respectively as shown in Fig. 3. However, the exothermal reaction spans over a large temperature region, for example, the crystallization of the amorphous $La_{17.5}Fe_{82.5}$ starts at $T_{x_1}=305°C$ and ends at $T_{x_2}=445°C$. For comparison, we also insert the typical temperature dependence of differential specific heat of amorphous Pd-Cu-Si with the same heating rate of 10°C/min. It can be seen in the figure that the precipitation of α-Fe in amorphous La-Fe systems starts at lower temperature with increasing Fe concentration.

4. Magnetization

Magnetization of the present amorphous La-Fe system was easily saturated in low applied fields as shown in Fig. 4. The magnetic moment per Fe atom at 4.2 K

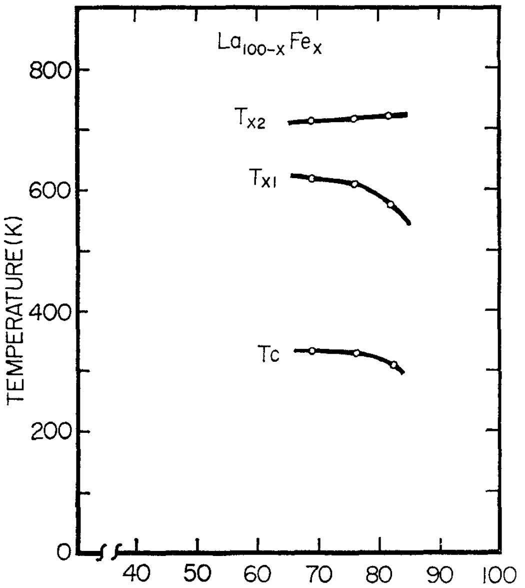

Fig. 5

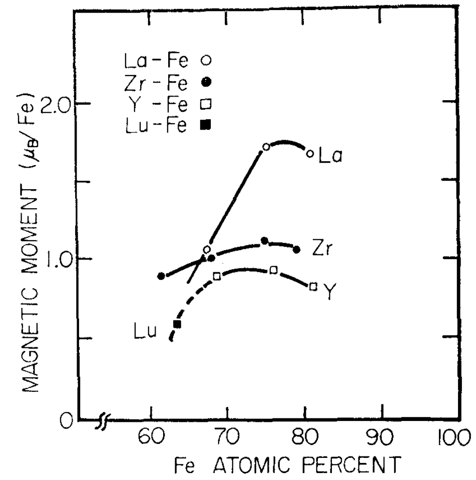

Fig. 6

Fig. 5. Curie temperature (T_c) and the crystallization temperatures (T_{x_1} and T_{x_2}) for amorphous La-Fe alloys.

Fig. 6. Magnetic moment per Fe atom for amorphous La-Fe alloys. For comparison, the results for amorphous Zr-Fe, Y-Fe, and Lu-Fe are also included (ref. 25).

(M_0) and the Curie temperature (T_c) are well defined and are shown in Figs. 5 and 6. On the basis of these data, we can estimate the Fe-Fe exchange constant J_{Fe} using the mean-field formula

$$J_{Fe}=\frac{3k_BT_c}{2ZS(S+1)}\,, \tag{1}$$

where S equals $M_0/2$ and $Z=12x$ is the average number of Fe nearest neighbors in La-Fe alloys. The values are estimsted to be 9.6, 4.6 and 4.5 in the unit of 10^{-15} ergs for $La_{30.6}Fe_{69.4}$, $La_{23.3}Fe_{76.7}$ and $La_{17.5}Fe_{82.5}$ respectively. The concentration dependence of J_{Fe} is strikingly large in comparison with the fact that the Co-Co and the Fe-Fe exchange interaction are constant in amorphous Y-Co, Co-Si-B, Y-Fe, and Lu-Fe alloys(6,17,18).

Recently a systematic study(19) of the spin-wave dispersion relations and low-temperature magnetization in the Fe based amorphous alloys has been carried out. It has been found that the spin-wave stiffness (D) calculated from magnetization is in satisfactory agreement with that from inelastic neutron scattering after the correction of renormalization effect. Thus it may be reasonable to say that the ordinary spin-wave theory completely accounts for the low-temperature magnetic

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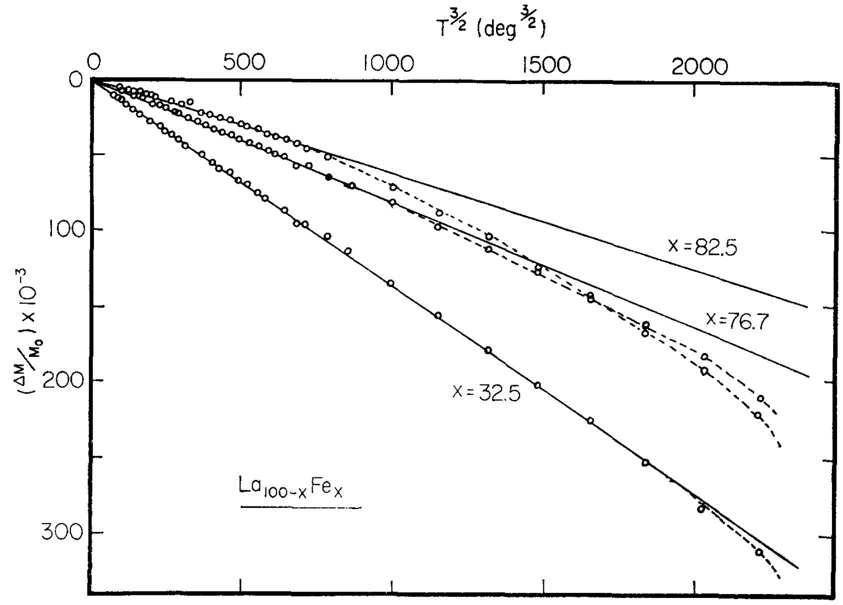

Fig. 7. Fractional decrease of $M(T)$ versus $T^{3/2}$ in amorphous La-Fe alloys.

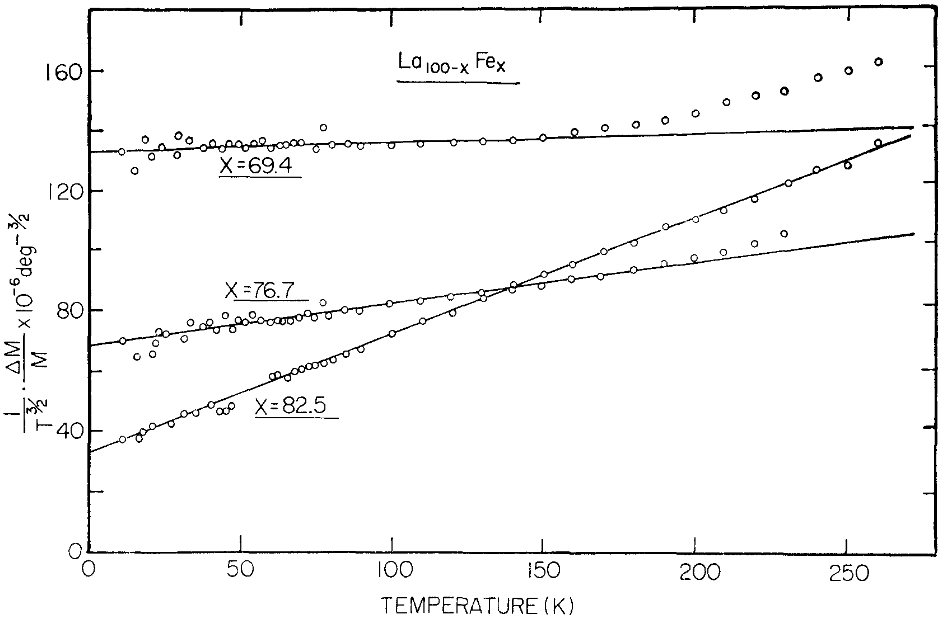

Fig. 8. $\Delta M/M_0 \times 1/T^{3/2}$ versus T for amorphous La-Fe alloys.

behavior of ferromagnetic amorphous alloys.

At low temperature, the magnetization is observed to exhibit

$$M(T) = M_0(1 - B^{3/2}T^{3/2} - CT^{5/2} \cdots) . \tag{2}$$

Plot of $1\text{-}M(T)/M_0$ as a function of $T^{3/2}$ for these samples is shown in Fig. 7, the data being taken with an applied fields of 5.8 kOe. The dominant $T^{3/2}$-dependence (20) can be seen below $T/T_c < 0.2$. The parameters B and C in Eq. (2) can be determined by plotting $\Delta M/M_0 \times 1/T^{3/2}$ vs. T as shown in Fig. 8. The values B and C have been determined from the intercept and slope of the straight line, and

Table 1. Parameters describing the temperature dependence of magnetization.

Composition	Temperature range	B $(10^{-6}\mathrm{deg}^{-3/2})$	C $(10^{-8}\mathrm{deg}^{-5/2})$	D (meV·A^2)	<r^2>(Å^2)
$La_{30.6}Fe_{69.4}$	$0<T/T_c<0.4$	131.5	3.75	56	~2
$La_{23.6}Fe_{76.4}$	$0<T/T_c<0.5$	67.0	14.20	58	~15
$La_{17.5}Fe_{82.5}$	$0<T/T_c<0.8$	33.0	39.0	73	~105
$Fe_{80}B_{20}$	—	22.0	1.4	103	~8
$Fe_{74.4}Mo_{5.6}B_{10}P_{10}$	—	45	3.5	85	~8
$Fe_{29}Ni_{49}P_{14}B_6Si_2$	$0<T/T_c<0.8$	59~65	1.0~1.5	—	~2
$Fe_{30}Ni_{45}P_{16}B_6Al_3$	$0<T/T_c<0.8$	4.6	25	—	~60
$Fe_{37.5}Ni_{37.5}P_{16}B_6Al_3$	$0<T/T_c<0.6$	3.4~4.3	8.5	—	~27
Fe (crystalline)	—	3.4	0.1	281 (ND) 285 (Mag)	~10

tabulated in table-1 including the spin-wave stiffness⁽²¹⁾ (D), which is calculated by using the formula,

$$B = 2.612\left[\frac{g\mu_B}{M_0}\right](k_B/4\pi D)^{3/2}\,. \tag{3}$$

As seen from this figure, the spin-wave theory including the $T^{5/2}$ contribution can describe the change of magnetization up to higher temperatures.

For comparison, we also cited the data for Fe based amorphous alloys.⁽¹⁹,²²,²³,²⁴⁾.

From simple spin-wave theory for the Heisenberg ferromagnet, the mean square range of the exchange interaction $\langle r^2\rangle$⁽²³,²⁴⁾ can be obtained as

$$\langle r^2\rangle = \frac{16\zeta(3/2)}{3k_B\zeta(5/2)}\cdot\frac{CD}{B}\,, \tag{4}$$

where k_B is Boltzmanns' constant and C, B and D are defined by Eqs. (2) and (3). $\zeta(3/2)$ and $\zeta(5/2)$ are 2.612 and 1.341 respectively. The results for $\langle r^2\rangle$ are also included in table-1. It should be noted that the exchange is sharply dependent on the Fe concentration in the amorphous La-Fe alloys as contrasted with the

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Y-Fe, Lu-Fe and Y-Co alloys. Furthermore the range of the exchange is much larger than in crystalline Fe. More detailed experimental research is required to elucidate the magnetic and structural properties of the amorphous La-Fe alloys.

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