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Abstract
Here, non-spin-polarized electronic structures and Fermi surface properties of RX2Al20 (R = La, Ce, Yb, Th, U; X = Ti, V, Cr, Mn) intermetallic compounds were calculated using the full potential all-electron local orbital (FPLO) approach in the framework of the local density approximation (LDA). Trends of the magnetism are discussed in terms of the characteristics of X-3d bands with a quantitative analysis of the relationship between band electron filling and crystal electric field splitting. Since coordination icosahedra of X-atoms have small trigonal distortion, crystal electric field splits the fivefold degenerate X-3d state into low-energy singlet and two higher-energy doublets e.g. In RTi2Al20 and RV2Al20 the population of the related 3d sub-band is not sufficient to cause energetically favorable spin polarization, whereas magnetic instabilities develop in the RCr2Al20 series. Finally, a manifestation of strong repulsive interactions between itinerant Mn-d electrons become most pronounced in ferromagnetic UMn2Al20. The influence of non-magnetic R-f states on magnetic and thermodynamic properties is discussed with special emphasis on the role of the f-p and f-d hybridization. For LaTi2Al20 and LaV2Al20 the calculated quantum oscillation frequencies are in accord with experimental reports.

Keywords
Rare earth intermetallics, Caged compounds, Orbital hybridization, Electronic band calculations, Fermi surface, Quantum oscillations

Disciplines
Condensed Matter Physics | Materials Science and Engineering
Electronic properties of $RX_2Al_{20}$ ($R = \text{La, Ce, Yb, Th, U}; X = \text{Ti, V, Cr and Mn}$) cage compounds

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Abstract

Non-spin-polarized electronic structures and Fermi surface properties of $RX_2Al_{20}$ ($R = \text{La, Ce, Yb, Th, U}; X = \text{Ti, V, Cr, Mn}$) intermetallic compounds were calculated using the full potential all-electron local orbital (FPLO) approach in the framework of the local density approximation (LDA). Trends of the magnetism are discussed in terms of the characteristics of $X$-3$d$ bands with a quantitative analysis of the relationship between band electron filling and crystal electric field splitting. Since coordination icosahedra of $X$-atoms have small trigonal distortion, crystal electric field splits the five-fold degenerate $X$-3$d$ state into low-energy singlet $a_{1g}$ and two higher-energy doublets $e_g$. In $R$Ti$_2$Al$_{20}$ and $RV_2Al_{20}$ the population of the related 3$d$ sub-band is not sufficient to cause energetically favorable spin polarization, whereas magnetic instabilities develop in the $RCr_2Al_{20}$ series. Finally, a manifestation of strong repulsive interactions between itinerant Mn-3$d$ electrons become most pronounced in ferromagnetic UMn$_2$Al$_{20}$. The influence of non-magnetic $R$-f states on magnetic and thermodynamic properties is discussed with special emphasis on the role of the $f$–$p$ and $f$–$d$ hybridization. For LaTi$_2$Al$_{20}$ and LaV$_2$Al$_{20}$ the calculated quantum oscillation frequencies are in accord with experimental reports.

Keywords:

Rare earth intermetallics, Caged compounds, Orbital hybridization, Electronic band calculations, Fermi surface, Quantum oscillations.

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1. Introduction

The large family of ternary intermetallics with the general composition $RT_2M_{20}$, where $R =$ rare earth or U, $T = 3d$, $4d$ or $5d$-electron transition metal, and $M =$ Zn, Cd or Al, comprises an unusual combination of caged-structural properties and large diversity of electronic ground states found, e.g., in filled skutterudites [1] and Laves phases [2]. The physical behavior of these systems is mostly dominated by the $4f$ and $5f$ electrons and arises from Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, Kondo effect and crystal-field effects giving rise to many phenomena such as electronic topological (Lifshitz) transitions, heavy-fermion and quantum critical states, superconductivity, magnetic or multi-polar ordering, and many others (for a review see e.g. [3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 17, 15, 16, 19, 20]). Wide variety of $RT_2M_{20}$ compounds offers a considerable chemical tunability of their physical properties.

The nonmagnetic subgroup of aluminides $RX_2Al_{20}$ with $R =$ La, Ce, Yb, U, Th, involving the lightest transition metals $X =$ Ti, V, Cr, shows weakly temperature-independent Pauli paramagnetism or diamagnetism. The magnitude of paramagnetic susceptibility depends on the number of the valence electrons [9, 17, 21, 22, 23, 24, 26, 27, 28, 29, 30, 31, 32]. Electrical resistivity and specific heat measurements showed that all these systems are simple metals with no sign of magnetic order at least down to 0.35K. This indicates that cerium and ytterbium are in tetravalent and divalent states, respectively, whereas the $5f$ electrons of uranium are strongly itinerant because of their hybridization with the electronic states of ligands. The behavior of $UMn_2Al_{20}$ distinctly differs from those of all the other $RX_2Al_{20}$ aluminides, namely the compound orders ferromagnetically below 17.5K [33]. A dispute about the origin of ferromagnetism in that compound was settled by polarized neutron diffraction study revealing the presence of itinerant magnetic moment only on the manganese sites with no discernible contribution of the uranium atoms [34].

Although many of the non-magnetic $RX_2Al_{20}$ ($R =$ La, Ce, Yb, Th, U; $X =$ Ti, V, Cr) phases have been characterized by means of X-ray diffraction, magnetization, electrical resistivity and heat capacity measurements, to the best of our knowledge only $LaTi_2Al_{20}$, and $LaV_2Al_{20}$ have been studied with a focus on the electronic density of states [17, 35]. For the latter compound the Fermi surface has also been calculated using full potential linearized augmented-plane wave (FLAPW) method [22].
In the present study we calculated the electronic band structures of several \( RX_2Al_{20} \) compounds with \( R = \text{Ce, Yb, Th and U} \) and various \( X \) metals. The main aim was at understanding their electronic structure, Fermi surface topology as well as to explore the origin of magnetism in \( \text{UMn}_2\text{Al}_{20} \), the only ferromagnetic aluminide in the \( RX_2\text{Al}_{20} \) family.

2. **Method of calculations**

The electronic structures calculations in the LDA approximation were performed with a FPLO-9.00-34 code [36]. The fully relativistic Dirac equation was solved self-consistently. The Perdew-Wang flavor of the exchange-correlation potential was applied [37] and the energies were converged on a dense \( k \) mesh with 12\(^3\) points. For the Fermi surfaces a 64\(^3\) mesh was used to ensure accurate determination of the Fermi level (\( E_F \)). The convergence was set to both the density \( (10^{-6} \text{ in code specific units}) \) and the total energy \( (10^{-8} \text{ Hartree}) \). The drawing of the crystal structure was produced with the aid of VESTA [38]. Extremal cross-section areas of the Fermi surfaces were calculated using Mathematica [39].

The structural parameters (unit cell dimensions and atomic coordinates) of (i) \( \text{LaTi}_2\text{Al}_{20} \), \( \text{CeTi}_2\text{Al}_{20} \), \( \text{YbTi}_2\text{Al}_{20} \), \( \text{LaV}_2\text{Al}_{20} \), \( \text{CeV}_2\text{Al}_{20} \), \( \text{LaCr}_2\text{Al}_{20} \), \( \text{CeCr}_2\text{Al}_{20} \), \( \text{YbCr}_2\text{Al}_{20} \), (ii) \( \text{UTi}_2\text{Al}_{20} \), (iii) \( \text{ThV}_2\text{Al}_{20} \), (iv) \( \text{ThCr}_2\text{Al}_{20} \), \( \text{UCr}_2\text{Al}_{20} \) and (v) \( \text{UMn}_2\text{Al}_{20} \) were adopted from the reported room-temperature experimental data presented in Refs. (i) [24], (ii) [40], (iii) [41] (iv) [30], (v) [34], respectively.

Because the atomic positions for (i) \( \text{YbV}_2\text{Al}_{20} \) [29], (ii) \( \text{ThTi}_2\text{Al}_{20} \) [41] (iii) \( \text{UV}_2\text{Al}_{20} \) [33] are unknown, they were assumed to be the same as those derived for (i) \( \text{LaV}_2\text{Al}_{20} \) [24], (ii) \( \text{UTi}_2\text{Al}_{20} \) [9], (iii) \( \text{ThV}_2\text{Al}_{20} \) [41], respectively.

3. **Results**

3.1. **Crystal structure**

The crystal structure of \( RX_2\text{Al}_{20} \), displayed in Figure 1a, is a face-centered cubic cell \( ( f.c.c., \text{ space group } Fd\overline{3}m, O_h^7, \text{ No. } 227) \), CeCr\(_2\)Al\(_{20}\)-type structure. The unit cell contains 184 atoms located at five different Wyckoff positions [25, 42, 43, 44, 45]. As apparent from Figure 2, the unit cell volume decreases significantly when proceeding from \( X = \text{Ti} \) to \( \text{Mn} \), i.e. it follows a decrease in the atomic radius of \( X \)-metal.
The cubic unit cell of $RX_2Al_{20}$ can be described as packing of two kinds of Al-based clusters around the $R$- and $X$-atoms, namely $RAl_{16}$ and $XAl_{12}$, shown in Figure 1a. The $R$ atom is located inside a nearly perfect Frank-Kasper cubic polyhedron made from a truncated tetrahedron by capping the hexagonal faces. It exhibits the point symmetry group $\overline{4}3m\ (T_D)$. Poor bonding of the $R$-ion to the neighboring aluminum atoms may give rise to unusually large atomic displacement parameters corresponding to anharmonic "rattling" motion about its equilibrium position [17, 18].

The transition metal atom, $T$, has twelve aluminum neighbors occupying vertices of an icosahedron. Since a periodic 3D structure cannot have 5-fold symmetry axes, this icosahedron is slightly (trigonally) distorted and the point symmetry of the $T$-site is reduced from $m\overline{3}5\ (Ih)$ to $3m\ (D_{3d})$ [46, 47].

3.2. Electronic structures

The electronic energy band dispersion curves $E(k)$ for $RX_2Al_{20} \ (R = La, Ce, Yb, Th, U)$ are drawn along the high symmetry lines of $k$-space in Figures 3–5. Nearly all bands near the Fermi level demonstrate complex character, including quasi-two-dimensional weakly-dispersive bands, as well as bands with stronger dispersion $dE(k)/dk$ leading to a set of sharp peaks and deep valleys in the density of states (DOS), shown in Figures 6–8. These bands originate mostly from Al-3$p$, $X$-3$d$ and $R$-f states ($Ce$-4$f$, $Yb$-4$f$ or $U$-5$f$).

Total Al-3$p$ contributions shown in Figures 3–5 create wide and almost featureless bands, mostly at the bottom of the valence band. Many of these bands disperse very weakly along $k_x$ direction and stick together in pairs due to the nonsymmorphic space group.

3.2.1. The $d$ bands

Besides Al-3$p$ and $Yb$-4$f$ states, the central portion of the valence band is dominated by $X$-3$d$ states. As shown in Figures 6 and 9, the Ti-3$d$-DOS has two distinctive local maxima in the valence band. The lower ones, marked in Figure 6 by vertical dashed-dotted lines, are centered at about -1.6 eV ($R = La, Ce, Yb$) and -1.7 eV ($R = Th, U$) with respect to the Fermi level, $E_F$. In turn, the upper maxima, marked by dashed lines, are located at about -0.6 eV ($R = lanthanide$ ion) and -0.7 eV ($R = actinide$ ion) below $E_F$. The empty Ti-3$d$ states in the partial density of states (PDOS) form another structure, somewhat smeared out, with the center of gravity around
2.3 eV above the $E_F$ (note the dotted line). The Fermi levels locate between two large separated features at wide valleys in PDOS.

For $RV_2Al_{20}$ the V-$3d$ subbands below $E_F$ are still clearly split into two slightly sharper high maxima and shifted down by about 0.3 eV with respect to the similar features in DOS of $RTi_2Al_{20}$ (Figure 7 and middle panel in Figure 9). This behavior can be simply rationalized in terms of the rigid band approximation, as Ti has one 3$d$ electron less than V. Moreover, the whole V-$3d$ bands are narrower, if compared to the Ti-$3d$ bands, by about 1 eV. This is owing to the fact that V ion has a deeper potential than Ti due to added nuclear charge. Consequently, the Fermi level lies at a steep edge of one feature in PDOS of V-$3d$ orbitals, resulting in a gain of DOS at the Fermi level.

Two main low-energy 3$d$ maxima in PDOS of $RCr_2Al_{20}$ (c.f. Figures 8a-e and 9) are shifted down by further 0.3 eV (dashed-dotted and dashed lines in Figure 8). The third high-energy feature (dotted line) becomes gradually sharper and in consequence a high DOS peak is pinned to the Fermi level. This leads to the pronounced increase of PDOS at $E_F$, and hence, the Al-$3p$ contribution is surpassed by that of the Cr-$3d$ states.

Importantly, as visualized in Figures 8f and 9, despite further narrowing of the 3$d$ band in UMn$_2$Al$_{20}$ the positions of two lower maxima appear to be nearly fixed at the same energy levels as it is observed for (Th,U)Cr$_2$Al$_{20}$, namely, at about -2.2 eV and -1.2 eV below $E_F$. The highest-energy structure in the Mn-$3d$ band becomes narrower and two small local peaks appear near the Fermi level, as indicated by arrows in Figure 9. It is worth recalling that very similar double-peak structure in DOS, leading to various physical instabilities, is a characteristic property of archetypal ferromagnetic superconductors like UGe$_2$ [48] or an itinerant antiferromagnet TiAu with strong spin fluctuations [49].

### 3.2.2. The $f$ bands

The most noticeable aspect in all PDOS considered is the shape and location of $R$-$4/5f$ bands. In the systems with empty $f$-subshell, LaX$_2$Al$_{20}$ and ThX$_2$Al$_{20}$, the unfilled La-$4f$ and Th-$5f$ bands lie a few eV above $E_F$ and do not contribute to DOS around the Fermi level. In turn, in CeX$_2$Al$_{20}$ and UX$_2$Al$_{20}$ the centers of gravity of the $R$-$4/5f$ states are located just above $E_F$, in concert with their non-magnetic character. Owing to the spin-orbit coupling and local symmetry, the $f$ states split into $j = 7/2$ and 5/2 states, separated by about 0.3 eV and 1 eV, for Ce and U ions, respectively.
Due to a typical band progression, the $j = 7/2$ states are pushed upward to the empty states while the $j = 5/2$ states drop down to the vicinity of $E_F$. Besides, further coupling with the environment leads to additional smearing of the $f$ band singularities, which increases significantly, when proceeding from $X = Ti, V, Cr$ to $Mn$. This feature is more pronounced for phases with uranium than for those with cerium, which reflects different radial extent of the $4f$ and $5f$ wave functions.

In contrast, in Yb$X_2$Al$_{20}$, which can be considered as hole equivalents of cerium compounds, pronounced peaks of Yb-$4f$ PDOS appear below $E_F$ and the $4f$ electron contribution at $E_F$ is fairly small. In these materials, Yb has a filled highly localized $4f^{14}$ shell, hence the compounds behave as conventional non-magnetic metallic Fermi liquids, as exemplified by YbFe$_2$Sb$_{12}$ [50].

As electrons near the Fermi surface are usually involved in the formation of itinerant magnetic state, it is important to understand their influence on the magnetic properties of $RX_2$Al$_{20}$. The Stoner criterion for ferromagnetism is formulated as $Z = I \cdot n(E_F) > 1$, where the Stoner exchange-correlation integral $I$ is a measure of the magnetic exchange interaction between either 3$d$-electron or 4/5$f$-electron ions, and $n(E_F)$ is the partial DOS at the Fermi level. The values of $I$ can be taken approximately equal to those reported for the elemental ions ($I_{Ti} = 0.34$ eV, $I_V = 0.35$ eV, $I_{Cr} = 0.38$ eV and $I_{Mn} = 0.41$ eV, $I_{Yb} = 0.43$ eV, $I_{Ce} = 0.53$ eV, $I_U = 0.51$ eV) [51, 52, 53, 54].

The Stoner factors, $Z$, derived from the presented non-polarized calculations are summarized in Table 2. Among the $RX_2$Al$_{20}$ compounds considered those with $X = Ti$, $V$ have the lowest value of $Z_d$ thus the Stoner criterion of ferromagnetism is by far not fulfilled, i.e. the product $I_{3d} \cdot n(E_F) \leq 0.46$ is much smaller than 1.

In contrast, the relatively high Stoner factors $Z_d$ close to 1 were found for $RCr_2$Al$_{20}$, hence hinting at closeness of the Cr ions to ferromagnetic instability. Actually, the behavior of Cr moments does not follow the Stoner criterion. Since the effective value of $I$ can be reduced by hybridization, in the case Cr, $Z_d$ should be considered only as an indicator of potential magnetic instabilities [55]. Values of $Z_d$ close to one were possible experimentally obtained for nearly ferromagnetic Fermi liquids in isostructural YFe$_2$Zn$_{20}$ and LuFe$_2$Zn$_{20}$ [56, 57]. Finally, in line with the Stoner model, the Mn magnetic moments in UMn$_2$Al$_{20}$ order ferromagnetically at low temperatures.

Our non-spin-polarized calculations also indicated, that the Stoner product $Z_f$ for the $4f$-states of Yb is of similar magnitude as $Z_d$ estimated for the V-$3d$ states. In turn, the $f$ states of cerium and uranium ions have $Z_f$
much larger than unity, in range 1.29 – 1.45 and 2.89 – 3.13, respectively. Therefore, it is reasonable to expect that the spin susceptibility of the U- and possibly also Ce-based compounds is moderately enhanced, which should be taken into consideration in the analysis of experimental results.

Indeed, the enhancement of Coulomb interactions between \( f \) and \( d \) electrons in the \( R\text{Cr}_2\text{Al}_{20} \) compounds is substantiated by sizable increase of the electronic contribution to the specific heat. The calculated \( \gamma \) and experimental \( \gamma_0 \) values of the Sommerfeld coefficient are compared in Table 2. The obtained non-spin-polarized DOS results allow to estimate the Sommerfeld coefficients under the assumption of the free electron model: \( \gamma = (\pi^2/3)N(E_F)k_B^2 \). Differences between \( \gamma \) and \( \gamma_0 \) are pronounced for all the compounds bearing \( X = \text{Cr}, \text{Mn}, \) and also for the \( R = \text{Ce} \) and \( \text{U} \)-based phase due to the additional \( f \)-electron contribution at the Fermi level. The latter effect seems to be noticeably smaller in \( (\text{La, Th})X_2\text{Al}_{20} \) with \( X = \text{Ti}, \text{V} \) and empty \( 4f/5f \) bands. In \( \text{LaV}_2\text{Al}_{20} \) and \( \text{LaTi}_2\text{Al}_{20} \) one can expect additional renormalization of the effective mass due to the electron-phonon coupling [13, 17].

### 3.3. Fermi surfaces and \( d\text{HvA} \) oscillations

The three-dimensional Fermi surfaces of \( RX_2\text{Al}_{20} \) at the experimental equilibrium volumes are shown in Figures 11-13. The corresponding Fermi surface cuts in the \( z = 0 \) plane of the conventional Brillouine zone (BZ) are shown in Figure 14. The sheets are denoted according to their twofold Kramers-degenerated number in band complex of the fully relativistic calculation. As expected for cubic systems, isotropic properties and three-dimensional Fermi sheets were derived. For the sake of clarity and convenience, the calculated Fermi sheets are subdivided in the figures into different groups according to their topological charge. If one compares the pictures within each column (denoted by black Roman numerals), one can easily notice, that the Fermi surface sheets in the same column reveal more or less similar topological features with the other Fermi surface sheets belonging either to \( R = \text{La, Ce, Yb or Th, U} \) group yet bearing the same transition metal \( X \).

The first group consists of numerous small nearly spherical or oval-square hole-like Fermi sheets around \( \Gamma \) point, with topological genus \( g = 0 \) (columns I-V for \( X = \text{Ti} \), columns I-III for \( X = \text{V} \), columns I-IV for \( X = \text{Cr} \)). They usually do not cross the edge of BZ, are homotopy-equivalent to a sphere and consist of nearly parabolic \( \text{Al-3p} \) bands with negative curvature in the vicinity of the Fermi level. Variation in the number of the Fermi sheets in this group
for different $X$ metal is directly related to the delicate balance between the band filling and the value of the lattice parameter. As can be inferred from Figures 3–5, several bands dip below $E_F$ at the $\Gamma$ point and a small gap in the vicinity of the Fermi level is created. In particular, for the Fermi sheets 533 of UTi$_2$Al$_{20}$ and 541 of UCr$_2$Al$_{20}$ one observes additional open bottlenecks along $\Gamma$–$L$ direction, which cross the boundary of the Brillouin zone. Similar neck appears in the band 513 for LaV$_2$Al$_{20}$ along $\Gamma$–$X$ direction, shown in Figure 12. In turn, in UMn$_2$Al$_{20}$ (band 545) and YbCr$_2$Al$_{20}$ (band 541) necks are melted, and then small sliced torus-shaped pockets are formed centred at $L$ and $X$ points of the BZ, respectively (Figure 13).

Comparable patterns are observed in the columns VI ($X = Ti$), IV ($X = V$) and V ($X = Cr$). Rough and wrinkled structures centred at $\Gamma$ point are quite similar to each other, but otherwise the sheets are not grossly distorted from spherical shape. In the Fermi sheets in CeTi$_2$Al$_{20}$ (511), ThTi$_2$Al$_{20}$ (531), UTi$_2$Al$_{20}$ (533), CeCr$_2$Al$_{20}$ (519), YbCr$_2$Al$_{20}$ (543), UCr$_2$Al$_{20}$ (543) and UMn$_2$Al$_{20}$ (547) necks reach out to touch either six square ($g = 2$) or eight hexagonal ($g = 3$) faces at $X$ and $L$ points of the BZ. In other cases under consideration, the sheets do not cross the Brillouin zone boundary. Instead, various small additional pockets are observed on the edges of Brillouin zone.

In the next group (column VII for $X = Ti$, V for $X = V$ and VI for $X = Cr$) very complex Fermi sheets mimic more or less the skeleton of the truncated octahedron of the Brillouin zone ($g \leq 24$). The band 515 for LaV$_2$Al$_{20}$ creates a nearly spherical structure, slightly smaller that the Brillouin zone, with crow’s-foot along $\Gamma$–$W$ directions. In turn, in band 537 of UTi$_2$Al$_{20}$ a multi-connected surface centred at $\Gamma$ point with arms branching out to the zone boundary along $\Gamma$–$X$ and $\Gamma$–$K$ directions, with large pockets at square faces are observed. In UV$_2$Al$_{20}$, the band 543 consists of 6 disk-shaped pockets around $X$ point, with tinny, disconnected spindle-shaped structures at $K$ points along $W$–$K$ direction. Considering the lines $W$–$U$–$X$ in the band 545 for UCr$_2$Al$_{20}$ there are large, nearly square-shaped pockets centred at $X$ point. The hole-like necks branch out from their tops along $W$–$K$ lines and become disconnected at $K$ points. In band 549 for UMn$_2$Al$_{20}$ the structures that have a shape of mushrooms at $X$ points are created, in analogy to Co-3d states in YCo$_2$Zn$_{20}$ and GdCo$_2$Zn$_{20}$ [58]. For several compounds, additional Fermi sheets with various small pockets at different high symmetry point are observed in columns VIII ($X = Ti$), VI ($X = V$) and VII–VIII ($X = Cr$).
An interesting feature is similar curvature profile of two largest Fermi surfaces of UV$_2$Al$_{20}$ and UMn$_2$Al$_{20}$ in columns V and VI, separated from each other along $\Gamma - X$ direction (marked by red arrow), as shown in Figure 14. Against this background, one can expect pronounced nesting between these regions of the Fermi surfaces, which may lead to additional coupling between electrons. As mentioned above, for these two compounds the Fermi level is computed to fall either in or near a local minimum DOS due to presence of van Hove singularities (Figures 7 and 8), yet the strong nesting may be favorable for further reduction of the DOS at $E_F$.

Our fully relativistic Fermi surface calculation results coincide very well with the previous FLAPW data for LaTi$_2$Al$_{20}$ [22]. Both methods yielded seven Fermi sheets with identical shapes and topology. The five smallest sheets were identified by a distinct quantum oscillation in the dHvA effect. The dHvA frequency $F$ is related to an extremal cross-section area $A_{ex}$ of the Fermi surface through the Onsager relation $F = \hbar A_{ex} / 2\pi e$. The values of $F$ calculated using FPLO are 93 T, 600 T, 801 T, and 1073 T for $B \parallel [100]$, correspond to orbitals schematically denoted in the upper panel of the Figure 11 by coloured numbers from 1 to 5, respectively. Our findings are in perfect concert with the previous experimental and theoretical studies [22].

Recently, the dHvA oscillations were also observed in LaV$_2$Al$_{20}$ [23]. The predicted dHvA extremal orbits for the magnetic field directed either along [111] or [100] directions are shown in the upper panel of Figure 12. They are denoted by colored numbers: 1 centered around $\Gamma$ point and 2 to 5 in the perpendicular planes on $\Gamma - X$ direction with increasing cross-section in each case. The orbit 1 belonging to the smallest sphere of the band 511 yields the frequency $F = 124$ T. It seems to have been observed experimentally [23] as the branch $\gamma$ ($F = 109$ T, $B \parallel [111]$). The orbits 2 and 3 on the tube of the band 511 have very small frequencies of 7 T and 27 T, respectively. The latter value is comparable to $F = 33$ T determined for the orbit 4 placed on the neck of the band 513, which is in good agreement with the experimental value $F = 19$ T for $B \parallel [001]$, denoted in Ref. [23] as the $\beta$ branch. Finally, the orbit 5 ($F = 344$ T) seems to correspond to the branch $\epsilon$ ($F = 318$ T, $B \parallel [001]$).

The other frequencies observed experimentally cannot be identified, perhaps except for the multiply-connected open Fermi surface sheet in the band 515 that can be attributed to the oscillation $F = 252$ T experimentally observed in limited field angle ranges close to $B \parallel [001]$.

Six hole-like tubes of the band 511 for LaV$_2$Al$_{20}$ extended along cubic $<001>$ directions are related to nearly featureless band close to the Fermi
energy in $\Gamma - X$ direction, shown in Figure 4a. It was suggested in Ref. [23] that the hole necks and diamagnetic response should vanish upon Ti substitution. Our calculations fully corroborate this assumption, namely, the six small Fermi sheets in the band 511 tend to disappear as the unit cell increases (equivalent to Ti doping), hence a spheroidal hole-like pocket with a volume similar to that of the Fermi sheet 501 of LaTi$_2$Al$_{20}$ should be created. Therefore, the two smallest Fermi sheets 511 and 513 of LaV$_2$Al$_{20}$ may be thought of as being derived from the two same aluminum bands of LaTi$_2$Al$_{20}$.

4. Summary and discussion

We have studied the electronic structures of the ternary compounds RX$_2$Al$_{20}$ ($R =$ La, Ce, Yb, Th and U, $X =$ $d$-electron transition metal) by means of \textit{ab initio} DFT relativistic, non-polarized band-structure methods and compared our results to some magnetic and thermodynamic properties. The compounds crystallize with a cubic cage-like crystal structure of the CeCr$_2$Al$_{20}$-type. The characteristic feature of the unit cell is that the $R$ atom has only one unique crystallographic site surrounded by sixteen Al atoms, which constitute a Frank-Kasper-type nearly-spherical polyhedron of cubic symmetry. In turn, all the $X$ and Al atoms occupy positions (one for $X$ and three for Al) with trigonal point symmetry.

The presented results of electronic band calculations suggest that the main character of $X$-$3d$ states in RX$_2$Al$_{20}$ is associated with breaking the degeneracy of the $X$-$3d$ states due to a static electric field produced by a surrounding charge distribution. While the $d$ state in icosahedral symmetry $I_h$ does not experience any crystal field splitting, small trigonal distortion may result in partial or complete lifting of the $d$-orbital degeneracies. According to irreducible representations of the trigonal point symmetry $D_{3d}$, the crystal field splits the five-fold degenerate $X$-$d$ orbital into (in the local reference frame) a low-energy singlet $a_{1g}$ and the higher-energy doublets $e_g$ [59], cf Figure 10. The representation $e_g$ corresponds to four orbitals: $d_{x^2-y^2}$, $d_{xy}$, $d_{xz}$, and $d_{yz}$ (for cubic symmetry $O_h$, the latter three states correspond to $t_{2g}$ orbitals), and $a_{1g}$ is formed by $d_z^2$ orbitals (corresponding to cubic $e_g$ representation with $d_{z^2-x^2-y^2}$ orbitals).

Within this scenario, one can speculate that the well-pronounced separation of three structures observed in the calculated band structure of RX$_2$Al$_{20}$ (Figures 6-8) indicates the splitting of the $X$-$3d$ band into two low-lying filled $a_{1g}$ and $e_g$ levels. The upper $e_g$ orbital remains empty in the RTi$_2$Al$_{20}$ and
RV$_2$Al$_{20}$ phases, whereas in RCr$_2$Al$_{20}$ and UMn$_2$Al$_{20}$ it becomes gradually filled. The fact, that the $d_{x^2−y^2},xy,xz,yz$ and $d_{z^2}$ states transform according to the same irreducible representations in the cubic symmetry suggests presumable occurrence of additional significant mixing between all $a_{1g}$ and $e_g$ components of the point group $D_{3d}$. Indeed, if one looks closer at the band structure of LaCr$_2$Al$_{20}$ (Figure 5a), one can see that the lower peak around 2.2 eV below $E_F$ contains nearly dispersionless bands along $\Gamma−L$, $X−W$ and $L−W$ directions, whereas the higher in energy peak mainly originates from several nearly flat bands mostly along $K−\Gamma−L$, $U−X$, $X−W−K$ and $\Gamma−L$ directions in BZ. It should be noted that the actual filling arrangement depends further on numerous factors such as a competition between on-site exchange interaction and effect of various degrees of freedom of $d$-electrons, i.e., charge, spin and lattice vibrations, not involved in presented LDA calculations.

Stronger localization of the $X$-3$d$ on changing the $d$-electron count appears to be in favor of electronic instability in RX$_2$Al$_{20}$ and UMn$_2$Al$_{20}$. It is known that materials at the boundary region, that is nearly- or weakly-magnetic, show a variety of spin-fluctuation features that can be removed by symmetry breaking, e.g., structural phase transition, superconductivity or magnetic ordering due exchange splitting effect or band dispersion modification [61]. According to that, UMn$_2$Al$_{20}$ exhibits bulk magnetic ordering. Moreover, UX$_2$Al$_{20}$ phases with cubic $Fd\bar{3}m$ symmetry form exclusively with light 3$d$ transition metals $X$ = Ti, V, Cr and Mn. Starting with iron, the symmetry of the stable Al-richest phase decreases to an orthorhombic structure (space group $Cmcm$) in UX$_2$Al$_{10}$ [62]. To keep the cubic symmetry in UX$_2$M$_{20}$ with $X$ from groups 8, 9 and 10 of the periodic table, Al must be replaced with $M$ = Zn or Cd.

The $R$-4/5$f$ electrons have also strongly delocalized character through coupling with the environment. It is commonly believed that the electronic ground states in many cerium, ytterbium and uranium intermetallics are mostly governed by the so-called $f$-ligand hybridization that involves interactions of the $f$-orbitals with $d$- and $p$-orbitals of neighboring atoms. According to the approach proposed by Straub and Harrison, which combines the linear muffin-tin orbital (LMTO) theory with the transition-metal pseudopotential formalism, the general hybridization-matrix elements are given
by[65, 66, 67, 68]:

\[ V_{ll'}m = \frac{\eta_{ll'm} \hbar^2}{m_e} \sqrt{\frac{r_l^{2l-1}r_{l'}^{2l'-1}}{d^{l+l'+1}}}, \]  

(1)

where \( m_e \) is the free electron mass, \( d \) is the interatomic distance of interacting atoms with atomic radii \((r, r')\), angular momenta \((l, l')\), and symmetries of the bond \((m = 0, 1, 2, \text{ and } 3 \text{ for } \sigma, \pi \delta, \text{ and } \varphi \text{ bonds, respectively})\). The coefficient \( \eta_{ll'm} \) is given by:

\[ \eta_{ll'm} = \frac{(-1)^{l'+1+m}(1 + l')!(l)!(l')!}{6\pi 2^{l+l'}l!l'!} \sqrt{\frac{(2l + 1)(2l' + 1)}{(l + m)!(l - m)!(l' + m)!(l' - m)!}}, \]  

(2)

The strength of the total \( f - l' \) hybridization can be evaluated according to

\[ V_{fl'} = \left[ \frac{n_i}{2l' + 1} \sum_{m=-l}^{l} V_{ll'm}^2 \right]^{1/2}, \]  

(3)

where \( n_i \) is the number of neighbors having angular momentum \( l' \) at a distance \( d \). The second moment of the \( f \)-hybridized band

\[ \langle (E_k - \epsilon_f)^2 \rangle = \sum_{l'} V_{fl'}^2 = V_{tot}^2 \]  

(4)

can be viewed as a measure of the degree of delocalization of \( f \) electrons, while \( V_{tot} \) can be considered as the total covalent energy, that contributes to the cohesion in a solid state. This approach is not strictly quantitative, however, it can be used to deduce a general tendency for \( f \) hybridization in isostructural compounds. Such considerations were previously applied successfully to discuss various trends in the band hybridization of many intermetallic phases such as ternary uranium antimonides \( UTSb_2 \) \((T = \text{3}d-, \text{4}d-, \text{5}d\text{-electron transition metal})[69]\), cerium indides \( CeTIn \) \((T = \text{Ni, Cu, Pd, Au})[70]\), \( CeMIn_5 \) and \( Ce_2MIn_8 \) \((M = \text{Rh, Ir, Co})[71]\).

In order to apply the Straub and Harrison approach to \( RX_2Al_{20} \) compounds with non-empty \( f \) shell, e.g. with \( R = \text{Ce, Yb and U} \), the atomic radii of the respective atoms were taken from [65] and [66]. Because of the lack of values of atomic radii for \( Al \), \( r \) was assumed to be equal to its atomic radius 0.143 nm. This value is comparable to the tight-binding parameters
obtained by Harrison and Straub for other p-elements from the third row of the periodic table [65, 66]. The interatomic distances were derived from the X-ray diffraction data used for the DFT calculation of electronic structures. The results are collected in Table 1. The total hybridization $V_{\text{tot}}$ increases in sequence Yb $<$ Ce $<$ U ranging from 67 - 72 meV, 123 - 133 meV, to 265 - 293 meV, respectively. In good agreement with the PDOS calculations, the least values of $V_{\text{tot}}$ indicate the most localized character of f states in Yb$X_2$Al$_{20}$, whereas 5f electrons of uranium were found strongly delocalized. One observes a slight increase of $V_{fp}$ with increasing the atomic number Z within the transition-metal series in the all groups of compounds (e.g. from Ti to Cr and to Mn), which implies an increase of both Ce-4f and U-5f states contribution in the density of states in the vicinity of the Fermi level.

The direct overlap of f − f orbitals of all the R ions considered is very low and varies only slightly, being below 1 % ($R = \text{Ce, Yb}$) and about 1.5% ($R = \text{U}$) of $V_{\text{tot}}$. This finding is in agreement with the Hill criterion for actinide metals because the shortest $d_{U-U}$ distances (ca. 0.62 nm) are always much larger than the Hill limit of 0.34 nm.

Similarly, the $V_{fd}$ hybridization is quite small ranging from about 1.7 to 2.4% of $V_{\text{tot}}$. Here, the obtained values are related to the limited spatial extension of the respective f- and 3d-wavefunctions together with a large distance $d_{R-X}$ ranging from 0.596 nm in UMn$_2$Al$_{20}$ to 0.609 nm in CeTi$_2$Al$_{20}$.

According to our calculations, the f $-$ p hybridization is the most dominant process responsible for strong delocalization of the f states. This behavior takes place because of short average distance $d_{R-Al} \simeq 0.32$ nm between R ions and aluminium. In consequence, the $V_{fp}$ values exceed 98 % of $V_{\text{tot}}$. This exchange interaction causes an appreciable widening and distortion of f bands in the whole energy range. Strong $V_{fp}$ interaction leading to delocalization of f electrons is an obvious reason for weak magnetic behavior in many binary and ternary intermetallic aluminides, such as intermediate valence compounds YbAl$_3$ [63, 64], YbAl$_2$ [74] or YbPtGe$_2$ [76] an archetypal spin-fluctuator UAl$_2$, a weakly temperature-dependent paramagnet UAl$_3$ [72, 73], classical magnetic Kondo systems CeAl$_2$ [77], or prototype heavy-fermion materials CeNi$_2$Ge$_2$ [75] and YbPtIn [78]. Comparison between the physical properties observed for the RT$_2$Al$_{20}$ phases and those reported for the isostructural RT$_2$M$_{20}$ compounds with M = Zn, Cd, characterized by similar $d_{R-R}$ and $d_{R-T}$ distances leads to final conclusion that the ground-state electronic properties of the whole 1:2:20 family is caused mainly by the f $-$ p hybridization, in line with some previous suggestions [21, 81, 82]. Final exper-
imental verification of all these predictions, e.g., by additional spectroscopic investigation is desired to identify the band structures at Fermi surface in $RX_2Al_20$ ($R = La, Ce, Yb, Th, U; X = Ti, V, Cr and Mn$).

5. Acknowledgements

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Data availability

The datasets analysed during the current study are available from the corresponding author on reasonable request.


Table 1: The $f - f (V_{ff})$, $f - d (V_{fd})$, $f - p (V_{fp})$ hybridization and the covalent energy ($V_{tot}$) in ternary $RX_2Al_{20}$ type compounds ($R = \text{Yb, Ce and U}; X = \text{Ti, V, Cr, Mn}$) as described in the text.

<table>
<thead>
<tr>
<th>Compound</th>
<th>$V_{ff}$ [meV]</th>
<th>$V_{fd}$ [meV]</th>
<th>$V_{fp}$ [meV]</th>
<th>$V_{tot}$ [meV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>YbTi$<em>2$Al$</em>{20}$</td>
<td>0.254</td>
<td>1.635</td>
<td>67.422</td>
<td>67.442</td>
</tr>
<tr>
<td>YbV$<em>2$Al$</em>{20}$</td>
<td>0.257</td>
<td>1.561</td>
<td>71.123</td>
<td>71.142</td>
</tr>
<tr>
<td>YbCr$<em>2$Al$</em>{20}$</td>
<td>0.282</td>
<td>1.651</td>
<td>72.620</td>
<td>72.639</td>
</tr>
<tr>
<td>CeTi$<em>2$Al$</em>{20}$</td>
<td>0.860</td>
<td>2.997</td>
<td>123.806</td>
<td>123.845</td>
</tr>
<tr>
<td>CeV$<em>2$Al$</em>{20}$</td>
<td>0.924</td>
<td>2.748</td>
<td>130.418</td>
<td>130.450</td>
</tr>
<tr>
<td>CeCr$<em>2$Al$</em>{20}$</td>
<td>0.955</td>
<td>3.030</td>
<td>133.442</td>
<td>133.480</td>
</tr>
<tr>
<td>UTi$<em>2$Al$</em>{20}$</td>
<td>4.040</td>
<td>6.581</td>
<td>265.397</td>
<td>265.509</td>
</tr>
<tr>
<td>UV$<em>2$Al$</em>{20}$</td>
<td>4.356</td>
<td>5.697</td>
<td>283.028</td>
<td>283.119</td>
</tr>
<tr>
<td>UCr$<em>2$Al$</em>{20}$</td>
<td>4.509</td>
<td>6.682</td>
<td>291.129</td>
<td>291.241</td>
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<tr>
<td>UMn$<em>2$Al$</em>{20}$</td>
<td>4.567</td>
<td>4.857</td>
<td>293.639</td>
<td>293.715</td>
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Table 2: The magnetic and electronic characteristics for RT$_2$Al$_{20}$ (R = La, Ce, Yb, Th, U, T = Ti, V, Cr) series: the Stoner factors $Z_d$ and $Z_f$ for X-3d and R-4/5f ions, respectively, measured ($\gamma$) and calculated ($\gamma_0$) Sommerfeld coefficient [mJ/(mol K$^2$)]. Experimental values are quoted for comparison. a: Ref. [24]; b: Ref. [30]; c: Ref. [33]; d: Ref. [80]; e: Ref. [28]; f: Ref. [79]

<table>
<thead>
<tr>
<th>Compound</th>
<th>$Z_d$</th>
<th>$Z_f$</th>
<th>$\gamma$</th>
<th>$\gamma_0$</th>
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<tr>
<td>LaTi$<em>2$Al$</em>{20}$</td>
<td>0.16</td>
<td>-</td>
<td>23$^a$</td>
<td>18</td>
</tr>
<tr>
<td>CeTi$<em>2$Al$</em>{20}$</td>
<td>0.19</td>
<td>1.29</td>
<td>34$^a$</td>
<td>27</td>
</tr>
<tr>
<td>YbTi$<em>2$Al$</em>{20}$</td>
<td>0.38</td>
<td>0.67</td>
<td>29$^d$</td>
<td>34</td>
</tr>
<tr>
<td>ThTi$<em>2$Al$</em>{20}$</td>
<td>0.29</td>
<td>-</td>
<td>-</td>
<td>29</td>
</tr>
<tr>
<td>UTi$<em>2$Al$</em>{20}$</td>
<td>0.19</td>
<td>2.86</td>
<td>41$^f$</td>
<td>31</td>
</tr>
<tr>
<td>LaV$<em>2$Al$</em>{20}$</td>
<td>0.21</td>
<td>-</td>
<td>20$^d$</td>
<td>17</td>
</tr>
<tr>
<td>CeV$<em>2$Al$</em>{20}$</td>
<td>0.27</td>
<td>1.33</td>
<td>30$^a$</td>
<td>23</td>
</tr>
<tr>
<td>YbV$<em>2$Al$</em>{20}$</td>
<td>0.31</td>
<td>0.27</td>
<td>20$^e$</td>
<td>21</td>
</tr>
<tr>
<td>ThV$<em>2$Al$</em>{20}$</td>
<td>0.39</td>
<td>-</td>
<td>-</td>
<td>23</td>
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<tr>
<td>UV$<em>2$Al$</em>{20}$</td>
<td>0.43</td>
<td>2.45</td>
<td>-</td>
<td>35</td>
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<tr>
<td>LaCr$<em>2$Al$</em>{20}$</td>
<td>1.22</td>
<td>-</td>
<td>63$^a$</td>
<td>40</td>
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<tr>
<td>CeCr$<em>2$Al$</em>{20}$</td>
<td>1.28</td>
<td>1.45</td>
<td>-</td>
<td>39</td>
</tr>
<tr>
<td>YbCr$<em>2$Al$</em>{20}$</td>
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<td>0.34</td>
<td>74$^a$</td>
<td>37</td>
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<tr>
<td>ThCr$<em>2$Al$</em>{20}$</td>
<td>1.21</td>
<td>-</td>
<td>63$^b$</td>
<td>41</td>
</tr>
<tr>
<td>UCr$<em>2$Al$</em>{20}$</td>
<td>1.07</td>
<td>3.14</td>
<td>80$^b$</td>
<td>57</td>
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<tr>
<td>UMn$<em>2$Al$</em>{20}$</td>
<td>1.06</td>
<td>3.13</td>
<td>300$^c$</td>
<td>50</td>
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</tbody>
</table>
Figure 1: (Color online) Left panels: the crystal structure of $RX_2Al_{20}$ shown as a packing of $RA_{16}$ (shaded in grey) and $XA_{12}$ (shaded in yellow) clusters. The two polyhedra are presented aside. Right panel: the Brillouin zone for reciprocal-space group $Fd\bar{3}m$. 
Figure 2: The experimental lattice parameters of $RX_2Al_{20}$, where $R =$La, Ce, Yb, Th and U versus X-3$d$ electron transition metals.
Figure 3: Calculated LDA energy dispersions of $RTi_2Al_{20}$ with $R =$ La, Ce, Yb (left panels) and $R =$Th, U (right panels) near the Fermi level along the main symmetry axes shown in Figure 1b.
Figure 4: Calculated LDA energy dispersions of $RV_2\text{Al}_{20}$ with $R = \text{La, Ce, Yb}$ (left panels) and $R = \text{Th, U}$ (right panels) near the Fermi level along the main symmetry axes shown in Figure 1b.
Figure 5: Calculated LDA energy dispersions of $R\text{Cr}_2\text{Al}_{20}$ with $R = \text{La, Ce, Yb}$ (left panels) and of ThCr$_2$Al$_{20}$, UCr$_2$Al$_{20}$ and UMn$_2$Al$_{20}$ (right panels) near the Fermi level along the main symmetry axes shown in Figure 1b.
Figure 6: (Color online) Calculated total and partial DOS of LaTi$_2$Al$_{20}$, CeTi$_2$Al$_{20}$ and YbTi$_2$Al$_{20}$ (left panels), and of ThTi$_2$Al$_{20}$ and UTi$_2$Al$_{20}$ (right panels), computed with the LDA.
Figure 7: (Color online) Calculated total and partial DOS of LaV$_2$Al$_{20}$, CeV$_2$Al$_{20}$ and YbV$_2$Al$_{20}$ (left panels), and of ThV$_2$Al$_{20}$ and UV$_2$Al$_{20}$ (right panels), computed with the LDA.
Figure 8: (Color online) Calculated total and partial DOS of LaCr$_2$Al$_{20}$, CeCr$_2$Al$_{20}$ and YbCr$_2$Al$_{20}$ (left panels), and of ThCr$_2$Al$_{20}$, UCr$_2$Al$_{20}$ and UMn$_2$Al$_{20}$ (right panels), computed with the LDA.
Figure 9: (Color online) Calculated partial DOS X-3d of RX₂Al₂₀. For clarity the subsequent curves are shifted upwards by multiples of 6 st./eV. The arrows mark the maxima discussed in the text.

Figure 10: Crystal-field splitting of X-3d states in distorted icosahedron with trigonal distortion D₃d according to an ionic model.
<table>
<thead>
<tr>
<th></th>
<th>I</th>
<th>II</th>
<th>III</th>
<th>IV</th>
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<th>VI</th>
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<td>LaTi&lt;sub&gt;2&lt;/sub&gt;Al&lt;sub&gt;20&lt;/sub&gt;</td>
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<td>CeTi&lt;sub&gt;2&lt;/sub&gt;Al&lt;sub&gt;20&lt;/sub&gt;</td>
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Figure 11: (Color online) Calculated Fermi surfaces of RTi<sub>2</sub>Al<sub>20</sub> (R = La, Ce, Yb, Th, U) compounds and extremal orbits of LaTi<sub>2</sub>Al<sub>20</sub>. The Arabic numerals denote numbers of bands.
Figure 12: (Color online) Calculated Fermi surfaces of RV$_2$Al$_{20}$ (R = La, Ce, Yb, Th, U) compounds and extremal orbits of LaV$_2$Al$_{20}$. The Arabic numerals denote numbers of bands.
Figure 13: (Color online) Calculated Fermi surfaces of RCr$_2$Al$_{20}$ (R = La, Ce, Yb, Th, U) and UMn$_2$Al$_{20}$ compounds. The Arabic numerals denote numbers of bands.
Figure 14: A sketch through the Brillouin zone of a (001) section of the Fermi surfaces of $RX_2Al_20$, where $R = La, Ce, Yb, Th, U$ (panels from left to right) and $X = Ti, V, Cr, Mn$ (from top to bottom). The arrows indicate the nesting vector along $\Gamma - X$ direction.
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