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Magnetic properties of the ordered VPt₃ alloy.

I. — Ferromagnetic behaviour

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Résumé. — Nous présentons une étude de magnétisme de l'alliage VPt₃ (V_{0,25}Pt_{0,75}) dans chacune des deux structures ordonnées possibles (structure cristalline de type Cu₃Au et de type TiAl₃, respectivement). Dans chacune des structures ordonnées VPt₃ est ferromagnétique tandis que l'alliage VPt₃ complètement désordonné (structure cfc de type Cu) est paramagnétique dans tout le domaine de température 4,2 K-1 100 K.

Au moyen de mesures d'aimantations rémanentes, nous avons déterminé les points de Curie ferromagnétiques $T_c \simeq 240$ K pour VPt₃ ordonné dans la structure de type Cu₃Au (échantillon VPt₃ (1)) et $T_c \simeq 210$ K pour VPt₃ ordonné dans la structure de type TiAl₃ (échantillon VPt₃ (2)). Les mesures d'aimantation ont été effectuées d'une part à basse température (4,2 K à 160 K) et en champs moyens ($0 \leq H \leq 20$ kOe) et d'autre part à 4,2 K en champs intenses ($0 < H \leq 150$ kOe). Ces dernières mesures ont permis la détermination précise de l'aimantation à saturation σ_s (4,2 K) et de la susceptibilité superposée χ_H (4,2 K). L'ordre de grandeur de la susceptibilité superposée χ_H (à 4,2 K $\chi_H \sim 1,0 \times 10^{-6}$ uem/g/Oe) est compatible avec le paramagnétisme de bande des métaux de transition V et Pt. Au vu de leur faible moment magnétique ($\mu \sim 0,1 \mu_B$ par atome-gramme de V), les alliages VPt₃ (1) et (2) peuvent être considérés comme étant faiblement ferromagnétiques. Le paramagnétisme de VPt₃ (1) et (2) au-dessus de leurs points de Curie respectifs, est bien décrit par une loi de Curie-Weiss. Les moments paramagnétiques qui en résultent sont faibles ($\mu' \sim 0,7 \mu_B$ par atome-gramme de V).

L'ensemble des résultats obtenus ne nous a pas permis d'établir avec certitude l'origine du magnétisme dans VPt₃ (modèle localisé ou modèle itinérant). Cependant, nous avons tenté d'interpréter le ferromagnétisme de VPt₃ (1) et VPt₃ (2) dans un modèle localisé ; dans ce modèle, le ferromagnétisme est attribué à un mécanisme d'échange indirect entre les moments des atomes de V par l'intermédiaire des électrons de conduction du Pt.

Abstract. — We have shown that the VPt₃ (V_{0,25}Pt_{0,75}) alloy becomes ferromagnetic in each of both atomically ordered states with Cu₃Au and TiAl₃ crystalline structure, respectively. The atomically disordered VPt₃ alloy (with a Cu like fcc structure), remains paramagnetic over the whole temperature range 4.2 K to 1 100 K.

By means of remanent magnetization measurements, we found the ferromagnetic Curie points $T_c \simeq 240$ K for VPt₃ ordered in the Cu₃Au like structure (VPt₃ (1) sample) and $T_c \simeq 210$ K for VPt₃ ordered in the TiAl₃ structure (VPt₃ (2) sample).

Magnetization measurements performed at low temperatures (4.2 K to 160 K) and in moderate fields ($0 \leq H \leq 20$ kOe) showed that both VPt₃ (1) and (2) samples behave as classical ferromagnets. The high field ($0 < H \leq 150$ kOe) magnetization measurements performed on both VPt₃ (1) and (2) samples at 4.2 K, allowed an accurate determination of the saturation magnetization σ_s (4.2 K) and of the high field χ_H (4.2 K) susceptibility. The order of magnitude of the χ_H susceptibility (at 4.2 K, $\chi_H \sim 1 \times 10^{-6}$ emu.g⁻¹.Oe⁻¹) is consistent with band paramagnetism of transition metals. Both VPt₃ (1) and (2) may be considered as weak ferromagnets, in view of their small ferromagnetic moment ($\mu_0 \sim 0.1 \mu_B$ per V atom-gram). The paramagnetic behaviour of VPt₃ (1) and (2) above their respective Curie points, is well described by a Curie-Weiss law yielding small paramagnetic moment values ($\mu' \sim 0.7 \mu_B$ per V atom-gram).

In spite of all experimental results obtained in this work, we could not settle with certainty whether the ferromagnetic behaviour of both ordered VPt₃ alloys has to be interpreted in a localized model of ferromagnetism rather than in an itinerant one. Nevertheless, we have interpreted the ferromagnetic behaviour of VPt₃ (1) and (2) within a model of localized ferromagnetism : the ferromagnetism in VPt₃ (1) and (2) is tentatively ascribed to a mechanism of indirect exchange interaction between the V moments through the conduction electrons of the Pt atoms. In this mechanism, the Pt electrons acquire a magnetic polarization antiparallel to the V moments.

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1. Introduction. — The phase diagrams of binary platinum based transition metal alloys T_xPt_{1-x} , where T is an element of the first transition series, show simple atomically ordered structures. These latter exist at stoichiometry only, or in a given concentration range [1-9]. Several of these systems show peculiar physical features, especially from a magnetic view-point, in the ordered state : for example, $CrPt_3$ [10] and $MnPt_3$ [11] show ferromagnetic ordering, although Cr and Mn are antiferromagnetic and Pt is non magnetic, whereas $FePt_3$ shows antiferromagnetic ordering [12]. In the disordered state, $CrPt_3$ is paramagnetic [13], $FePt_3$ is ferromagnetic [14], and $MnPt_3$ shows no magnetic ordering but preliminary measurements on this system reveal a complex magnetic behaviour [15]. The magnetic properties of the ordered structures vary with composition (in the concentration range in which the ordered structures exist) and thus, depend upon the degree of atomic long-range order. These peculiar features are related to the competition between atomic and magnetic interactions [16].

In order to study the ordering effects in transition metal alloys, we have undertaken a systematic investigation of the T_xPt_{1-x} [17, 18] alloys, of their ordered structures and their disordered states when they exist.

The study of the V-Pt system has now been completed. We found a very peculiar magnetic behaviour for the VPt_3 alloy : VPt_3 is paramagnetic in the disordered state, but becomes ferromagnetic in the ordered state.

In this paper (paper I), as in paper II [19], we restrict to the magnetic properties of the ordered VPt_3 alloy. A detailed discussion about the calorimetric and magnetic properties, as a function of concentration of both the ordered and disordered states of the V-Pt system, will be given in our forthcoming publication [18].

We summarize briefly the main properties of the VPt_3 alloy in its atomically disordered state. The disordered VPt_3 alloy was found to be paramagnetic in the whole temperature range 4.2 K to 1 100 K. The moderate temperature dependence of its paramagnetic susceptibility $\chi(T)$ could not be described meaningfully in terms of a Curie-Weiss law. But this $\chi(T)$ susceptibility is consistent with the band paramagnetism of the V and Pt transition metals (at 293 K, $\chi \simeq 0.7 \times 10^{-6} \text{ emu} \cdot \text{g}^{-1} \cdot \text{Oe}^{-1}$).

We have found that VPt_3 becomes ferromagnetic in each of both Cu_3Au and $TiAl_3$ like ordered crystal-line structures. The ferromagnetic Curie temperature of the ordered VPt_3 alloy was found to be 240 K in the Cu_3Au structure and 210 K in the $TiAl_3$ structure. But these ordered VPt_3 alloys have small ferromagnetic moments ($\mu_0 \sim 0.1 \mu_B$ per V atom-gram) and may thus be regarded as weak ferromagnets, despite their relatively high Curie temperatures ($T_c > 200$ K).

Ferromagnetism was already discovered by Goto and Yamauchi [20] in ordered (Cu_3Au like) $V(Ir_{1-x}Pt_x)_3$ compounds. But these authors found that the ferromagnetism of the ordered $V(Ir_{1-x}Pt_x)_3$ compounds probably disappears in the concentration range $0.95 \leq x \leq 1.00$. Moreover, Kawakami and Goto [21] have investigated their VPt_3 sample by magnetization and resistivity measurements. They found only a very weak spontaneous magnetization (of the order of $10^{-3} \text{ emu} \cdot \text{g}^{-1}$). Nevertheless, they proved the occurrence of magnetic ordering in their VPt_3 sample, according to a kink at about 206 K in their resistivity *versus* temperature curve. Creveling and Luo [22] performed magnetic susceptibility measurements on Pt_2V , Pt_3V , Pd_2V , Pd_3V , Ir_3V and Rh_3V compounds at low temperatures (0.37 K to 4.3 K) and in no case they found any magnetic ordering. But they did not indicate in which state (atomically ordered or disordered) all these compounds were studied.

In the frame of a systematic study of ordered and disordered transition metal alloys, we have also performed magnetization measurements on ordered and disordered $TiPt_3$, VPd_2 and VPd_3 compounds in the low temperature range 4.2 K to 100 K. These measurements indicated that no magnetic ordering occurred in any of these compounds.

Thus, the ferromagnetism discovered here on both ordered VPt_3 alloys (in the Cu_3Au structure and in the $TiAl_3$ structure) appears as somewhat unexpected. Therefore, we have to ascertain the ferromagnetic behaviour of both ordered VPt_3 alloys studied here, and then, to explain the origin of this ferromagnetism in terms of itinerant or localized d electrons. Moreover, in the course of a study of the magnetic critical phenomena in transition metal alloys, we have investigated the critical behaviour of VPt_3 in both ordered states. Thus, we have systematically studied the VPt_3 alloy in each of both ordered states by means of all classical magnetic investigations (as will be indicated below).

In the present work (hereafter designated as paper I), we investigated the ferromagnetic properties of VPt_3 in both ordered states and we propose a tentative explanation of these ferromagnetic properties. In section 2, the sample preparation and the apparatus are discussed. In section 3, we show the ferromagnetic behaviour of ordered VPt_3 by means of magnetization measurements performed at low temperatures and in moderate fields. In section 4, we determine the ferromagnetic Curie point of each VPt_3 sample by means of remanent magnetization measurements. High field magnetization measurements reported in section 5, allowed an accurate determination of the absolute ferromagnetic moment. In section 6, we report and analyse high temperature paramagnetic susceptibility measurements. Finally, in section 7, we discuss all obtained results in terms of itinerant against localized d electrons, and we give a tentative

interpretation of the ferromagnetism of ordered VPt₃ within a model of localized ferromagnetism.

Paper II [19] deals with the critical behaviour of VPt₃ in each of both ordered states. The critical behaviour of each VPt₃ sample around its Curie point T_c , is investigated in the frame of the scaling theory. The corresponding critical magnetic equations of state are analysed and compared with available theoretical results. Finally, we present our tentative conclusions on the nature of the interaction forces in these ordered VPt₃ samples.

A first comment must be made here. Our observation that the magnetic behaviour can be drastically altered according to the atomic order (disordered Cu like VPt₃ is paramagnetic, while ordered VPt₃ becomes ferromagnetic in both Cu₃Au and TiAl₃ like structures), means that magnetic impurities in the studied VPt₃ alloys, are unlikely to be responsible for the ferromagnetism of ordered VPt₃. The ferromagnetic behaviour reported here for both ordered VPt₃ samples, is a bulk phenomenon.

2. Experimental procedures. — All the measurements reported in this work, were performed on two different bulk VPt₃ samples (small ellipsoids) : one ordered in the Cu₃Au structure and the other ordered in the TiAl₃ structure. Hereafter, we designate by VPt₃ (1) the VPt₃ sample ordered in the Cu₃Au structure and by VPt₃ (2) the VPt₃ sample ordered in the TiAl₃ structure.

We recall here the crystalline structures of the VPt₃ alloy in the disordered state and in both ordered states ([23-25]) :

- (i) the disordered VPt₃ alloy crystallizes in the Cu like fcc structure with cell parameter $a = 3.85 \text{ \AA}$;
- (ii) in the first ordered state, VPt₃ has a Cu₃Au like fcc structure with cell parameter $a = 3.87 \text{ \AA}$;
- (iii) in the second ordered state, VPt₃ has the TiAl₃ like tetragonal structure with cell parameters $a = 3.861 \text{ \AA}$ and $c = 7.842 \text{ \AA}$.

According to Waterstrat [25], there is only one ordering temperature in the constitution diagram of VPt₃ : the temperature 1 015 °C of order-disorder transformation from the disordered state with Cu like structure to the ordered state (2) with TiAl₃ structure. The ordered state (1) with Cu₃Au structure has been reported [25] as a metastable state. The preparation of VPt₃ in both ordered states (1) and (2), will be discussed in our forthcoming publication [18], together with the features (ordering temperature and stability) of VPt₃ in the ordered state (1). We simply indicate below how we obtained both ordered VPt₃ (1) and (2) samples studied here.

The VPt₃ (1) sample has been annealed during four hours at 1 260 °C, then water-quenched to room temperature. This heat treatment was sufficient to establish the homogeneous long-range ordered Cu₃Au like structure of this VPt₃ (1) sample, according to its well-defined Curie temperature $T_c = 239.0 \text{ K}$

(section 4, Fig. 4) and to the analysis of its Debye-Scherrer pattern [18].

We encountered much more difficulties in the ordering of the VPt₃ (2) sample. This sample needed several heat treatments in order to eliminate intermediate short-range ordered or partially long-range ordered structures. Finally, this VPt₃ (2) sample has been annealed during one day at 930 °C, then water-quenched to room temperature. The relatively ill-defined Curie point T_c of this sample ($209 \text{ K} \leq T_c \leq 213 \text{ K}$, section 4, Fig. 4) indicates some inhomogeneities in the long-range ordered TiAl₃ like structure of this sample. Failing more efficient heat treatments, we assumed this VPt₃ (2) sample as sufficiently well ordered in the TiAl₃ structure. This assumption was justified by the analysis of the corresponding Debye-Scherrer pattern [18].

We have confirmed the effectiveness of both ordering processes in the VPt₃ alloy by testing the reversibility of both ordered states (Cu₃Au like and TiAl₃ like structures) in each of both VPt₃ (1) and (2) samples (transformation : ordered state in the Cu₃Au structure \rightleftharpoons ordered state in the TiAl₃ structure). These tests were only performed at the end of all magnetic investigations reported here, and will be described in our forthcoming publication [18]. We mention here only our success in the transformation (ordered TiAl₃ structure \rightarrow Cu₃Au structure) of the VPt₃ (2) sample. We have subjected this VPt₃ (2) sample to the same heat treatment undergone by VPt₃ (1) and we have thus obtained VPt₃ (2) effectively ordered in the Cu₃Au structure, as checked by Curie temperature measurements : $T_c \simeq 246 \text{ K}$ for VPt₃ (2) ordered in the Cu₃Au structure (to be published) against $T_c = 239.0 \text{ K}$ for VPt₃ (1) in the Cu₃Au structure (section 4, Fig. 4). The difference in these T_c values may be attributed to a slight difference in composition between the two VPt₃ (1) and (2) samples [18].

Now, returning to the VPt₃ (1) and (2) samples ordered in the Cu₃Au and TiAl₃ structures respectively, we describe briefly the different apparatus used for the magnetic investigations reported in this work.

The high temperature susceptibility measurements (100 K to 1 100 K) were performed with a Foëx-Forrer type magnetic balance, in fields H ranging from 2 kOe to 6 kOe. The low temperature (4 K to 100 K) susceptibility and magnetization measurements were performed with a Foner type vibrating sample magnetometer, in low or moderate fields ($0 \leq H \leq 20 \text{ kOe}$). Magnetization measurements on both ordered VPt₃ samples at 4.2 K and in high fields ($0 < H \leq 150 \text{ kOe}$) have been performed at the S.N.C.I. (Grenoble, France). The moderate field ($0 \leq H \leq 20 \text{ kOe}$) magnetization measurements in the critical temperature range of each VPt₃ sample around its Curie point (from 195 K to 271 K for VPt₃ (1) and from 188 K to 235 K for VPt₃ (2), see

paper II, [19]) were also performed with the Foner type vibrating sample magnetometer. All sets of apparatus were calibrated against Gd_2O_3 [26] or Ni [27] standards. The relative uncertainty in our $\sigma(H, T)$ or $\chi(T)$ measurements, is estimated to be less than 2%. In all measurements and results, the field H has been corrected for demagnetization.

3. Magnetization measurements at low temperatures. — First, we have performed isothermal magnetization measurements on each investigated VPt_3 sample at different temperatures ranging from 4.2 K to 74 K, and even to 161.5 K for VPt_3 (2), in moderate fields ($0 \leq H \leq 20$ kOe). In all cases, the field H has been corrected for demagnetization. The corresponding magnetization curves are shown in figure 1 for both ordered VPt_3 samples at all investigated temperatures. The shape of all these curves is clearly indicative of ferromagnetism : (i) existence of remanent magnetization and of a coercitive force by reversing the field ; (ii) approach to saturation of the magnetization in the field range $H \sim 14$ kOe to 20 kOe ; the full saturation of the magnetization is only reached in relatively high fields $H \geq 50$ kOe, as will be shown in section 5. But one notices the relatively weak magnetization values of these samples : at $T = 4.2$ K and $H = 20$ kOe, $\sigma \simeq 0.89$ emu.g⁻¹ for the VPt_3 (1) sample and $\sigma \simeq 0.67$ emu.g⁻¹ for the VPt_3 (2) sample, yielding relatively weak ferromagnetic

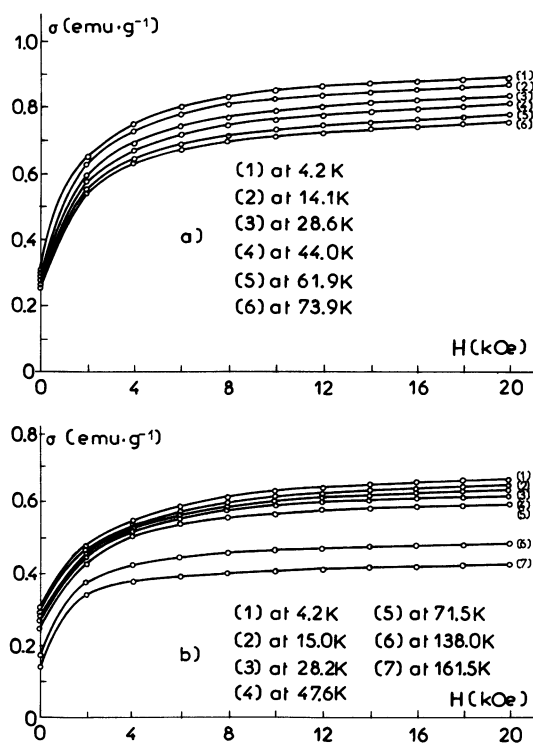


Fig. 1. — Isothermal magnetization curves for both ordered VPt_3 alloys at different temperatures and moderate fields ($0 \leq H \leq 20$ kOe) : (a) for the Cu_3Au like VPt_3 (1) sample ; (b) for the TiAl_3 like VPt_3 (2) sample.

moments ($\mu \simeq 0.1 \mu_B$ per V atom-gram), as will be seen in section 5.

Then, we have measured the hysteresis loop of VPt_3 (1) and (2) at 4.2 K by cycling each sample in a 20 kOe field. The corresponding hysteresis loops are symmetrical, as can be seen on figure 2, which displays the hysteresis loop of the VPt_3 (2) sample at 4.2 K as an example. According to the shape of their hysteresis loop at 4.2 K, both ordered VPt_3 samples may be regarded as classical ferromagnets. One notices (i) for the VPt_3 (1) sample a coercitive force $H_c \simeq 600$ Oe and a remanent magnetization $\sigma_r \simeq 0.35$ emu.g⁻¹ against $\sigma \simeq 0.89$ emu.g⁻¹ at 20 kOe ; (ii) for the VPt_3 (2) sample (Fig. 2) a coercitive force $H_c \simeq 700$ Oe and a remanent magnetization $\sigma_r \simeq 0.29$ emu.g⁻¹ against $\sigma \simeq 0.67$ emu.g⁻¹ at 20 kOe.

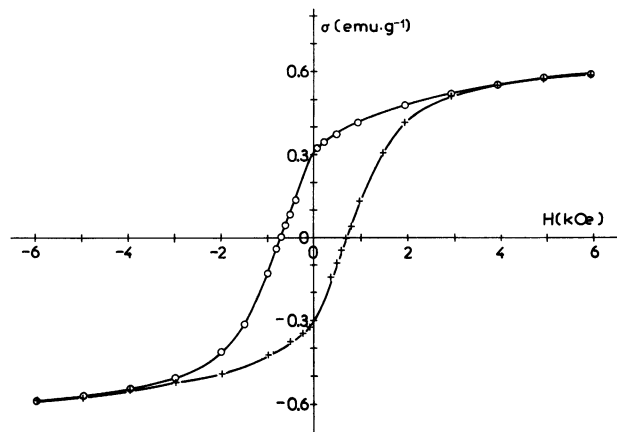


Fig. 2. — Hysteresis loop of the VPt_3 (2) sample at 4.2 K : magnetization σ (emu.g⁻¹) against field H (kOe) corrected for demagnetization : \circ by decreasing H from + 5.95 kOe to - 5.95 kOe, + by increasing H from - 5.95 kOe to + 5.95 kOe. This hysteresis loop was recorded by cycling the sample in a 20 kOe field (-20 kOe $\leq H \leq +20$ kOe), but the hysteresis effect appears only in the field domain -4 kOe $\leq H \leq +4$ kOe.

The coercitive force $H_c \sim 600$ Oe to 700 Oe found here for VPt_3 (1) and (2), is small, if compared to that reported by Creveling *et al.* [28] for the ordered Au_4V compound ($H_c > 11$ kOe at 4.2 K). This means that both VPt_3 alloys have only a moderate, or, even, a small energy of magnetocrystalline anisotropy.

We have also investigated the temperature dependence of the spontaneous magnetization $\sigma_{sp}(T)$ for each ordered VPt_3 sample. The spontaneous magnetizations $\sigma_{sp}(T)$ were estimated by linear extrapolation to $H = 0$ of the quasi-linear parts of the $\sigma = f_T(H)$ magnetization curves displayed in figure 1, parts corresponding to the field range $H \simeq 14$ kOe to 20 kOe. As can be seen on figure 3, the spontaneous magnetization $\sigma_{sp}(T)$ of each ordered VPt_3 sample decreases in proportion to $T^{3/2}$ rather than to T^2 , for sufficiently low temperatures (4.2 K $\leq T \leq 28$ K).

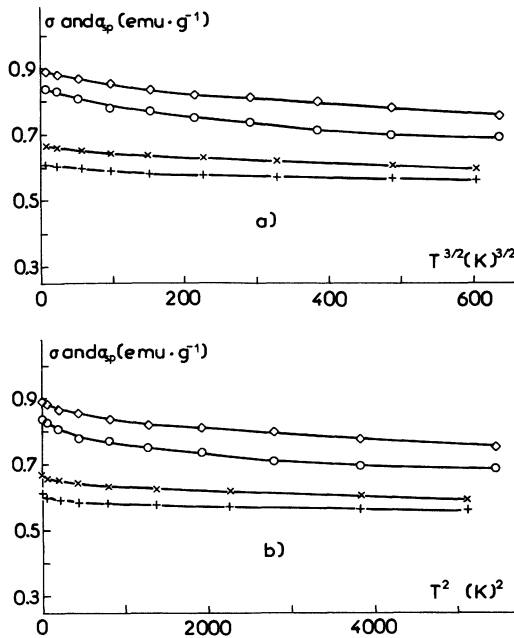


Fig. 3. — Spontaneous magnetization $\sigma_{sp}(T)$ and magnetization $\sigma(T, H = 20 \text{ kOe})$ for both ordered VPt₃ alloys against temperature T : (a) in the σ versus $T^{3/2}$ representation ; (b) in the σ versus T^2 representation. In both σ vs. T^2 and vs. $T^{3/2}$ representations, the symbols are, \diamond σ at 20 kOe and \circ σ_{sp} for VPt₃ (1), and \times σ at 20 kOe and $+$ σ_{sp} for VPt₃ (2).

It is known that most of the ferromagnetic metals and alloys, have spontaneous magnetizations $\sigma_{sp}(T)$ following the $T^{3/2}$ law. Since, this $T^{3/2}$ law seems also to be valid here, we cannot conclude whether the itinerant or the localized model of ferromagnetism is applicable to each ordered VPt₃ alloy. Considering the accuracy of the $\sigma_{sp}(T)$ versus $T^{3/2}$ or T^2 plots (Fig. 3), we take for each ordered VPt₃ sample the $\sigma_{sp}(4.2 \text{ K})$ magnetization as the absolute spontaneous magnetization $\sigma_s(0 \text{ K})$: within the experimental error, it can be written $\sigma_{sp}(4.2 \text{ K}) = \sigma_{sp}(0 \text{ K})$.

4. Determination of the ferromagnetic Curie temperature T_c . — The ferromagnetic Curie temperature T_c of any ferromagnet, can be defined as the temperature at which (i) the spontaneous magnetization $\sigma_{sp}(T)$ vanishes by increasing the temperature ($T \leq T_c$), (ii) the initial paramagnetic susceptibility $\chi_i(T)$ diverges (or its inverse $\chi_i^{-1}(T)$ vanishes) by lowering the temperature ($T \geq T_c$); the $\chi_i(T)$ susceptibility is defined for $T \geq T_c$ as $\chi_i = \lim_{H \rightarrow 0} \left(\frac{\sigma}{H} \right)$.

The above definition of T_c leads to the most suitable determination of the Curie point T_c for any real ferromagne. The $\sigma_{sp}(T)$ and $\chi_i^{-1}(T)$ quantities are determined by adequate extrapolation of moderate (or high) field isothermal $\sigma(H, T)$ magnetization data, in order to avoid magnetocrystalline anisotropy effects. Such extrapolations are usually undertaken by plotting σ^2 against H/σ at a number of tempera-

tures below and above the Curie point T_c of the ferromagnet.

A more convenient determination of T_c of any ferromagnet, rests on the existence of remanent magnetization below T_c (existence of a hysteresis loop below T_c). The Curie point is thus the temperature at which the remanence (or the coercitive force vanishes) by raising the temperature ($T \leq T_c$) beyond T_c .

A reliable value of T_c for any ferromagnet, can only be given, if both methods for determining T_c , are in a satisfactory agreement.

We have used both methods to determine the Curie point T_c of each ordered VPt₃ alloy.

The determination of T_c by the first method, will be fully described in paper II [19], paper dealing with the critical phenomena investigations around the Curie point T_c of each ordered VPt₃ sample.

We determined the T_c temperature of each ordered VPt₃ sample by the second method in the following way. The remanent magnetization σ_r of each sample was obtained after application of a 20 kOe field at 4.2 K, then observed as a function of temperature by warming the sample from 4.2 K to 270 K in the low field of 13 Oe. Figure 4 shows this remanent magnetization $\sigma_r(T)$ against temperature T for each VPt₃ sample. Considering the very low field used here (13 Oe), we neglect any (super)-paramagnetic component in the observed (σ_r) magnetization and we take the Curie temperature T_c of each VPt₃ sample as the temperature at which $\sigma_r(T)$ vanishes (temperature obtained by extrapolation to $\sigma_r = 0$ of the sharply decreasing portion of each $\sigma_r(T)$ versus T curve). It can be seen on figure 4 that (i) the determination of T_c of the VPt₃ (1) sample, is very easy and accurate : $T_c = 239.0 \text{ K}$; (ii) the magnetization tail observed on the $\sigma_r(T)$ versus T curve of the VPt₃ (2) sample, renders the determination of T_c much more difficult : $209 \text{ K} \leq T_c \leq 213 \text{ K}$; this magnetization tail may

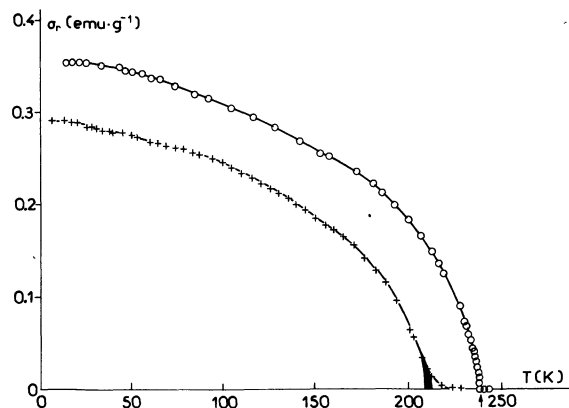


Fig. 4. — Remanent magnetization σ_r (emu.g^{-1}) of both ordered VPt₃ alloys against temperature T ; $\sigma_r(T)$ curves : \circ for the Cu₃Au like VPt₃ (1) sample; $+$ for the TiAl₃ like VPt₃ (2) sample. First determination of the Curie temperature T_c : $T_c = 239.0 \text{ K}$ for VPt₃ (1) : arrow; and $209 \text{ K} \leq T_c \leq 213 \text{ K}$ for VPt₃ (2) : dashed end of the $+$ $\sigma_r(T)$ curve.

be attributed to inhomogeneities occurring in the sample during the heat treatments necessary to order this VPt_3 sample in the TiAl_3 like structure.

In order to check the validity of the Curie temperatures T_c determined above for each ordered VPt_3 sample :

(i) we have verified for each sample, that at any temperature above $T_c (T > T_c)$, no remanent magnetization appears after application of a 20 kOe field,

(ii) we have performed magnetization measurements by cooling each sample in the low field of 13 Oe from 250 K down to 150 K and even, to 4.2 K for VPt_3 (2); the appearance of a « spontaneous σ_{rs} » magnetization in this low field at temperatures below the observed Curie points $T_c = 239.0$ K for VPt_3 (1) and $T_c = (211 \pm 2)$ K for VPt_3 (2), confirms the validity of the determined Curie temperatures.

Furthermore, the Curie temperatures T_c determined by the first method (paper II, [19]) are in satisfactory agreement with those reported above :

$T_c = (240.0 \pm 0.1)$ K for the VPt_3 (1) sample, by means of $(\sigma)^{2.00}$ versus $(H/\sigma)^{0.84}$ plots;

$T_c = (210.3 \pm 0.1)$ K for the VPt_3 (2) sample, by means of $(\sigma)^{2.00}$ versus $(H/\sigma)^{0.8333}$ plots.

The differences in the T_c values ($\Delta T_c \simeq 1$ K) determined by both methods, may be attributed to magneto-crystalline anisotropy effects in each of both ordered VPt_3 samples.

Nevertheless, we consider as reliable, the ferromagnetic Curie temperature values $T_c = 240.0$ K for the VPt_3 (1) sample, $T_c = 210.3$ K for the VPt_3 (2) sample.

These Curie temperatures are used in paper II as final Curie temperatures of each ordered VPt_3 sample in our critical phenomena investigations.

5. Determination of the ferromagnetic moment. —

In this section, we report the high field magnetization measurements ($0 < H \leq 150$ kOe) performed on each ordered VPt_3 alloy at 4.2 K (Fig. 5a). Considering the relatively high Curie point T_c of each VPt_3 sample ($T_c > 200$ K), and having checked (see section 3) that the spontaneous magnetization σ_{sp} (4.2 K) represents the absolute spontaneous magnetization σ_{sp} (0.0 K) for VPt_3 (1) and (2) within the experimental error, we assume that the high field magnetization isotherm $\sigma(H, 4.2 \text{ K})$ up to 150 kOe, yields the absolute saturation magnetization for each investigated sample : $\sigma_s(4.2 \text{ K}) = \sigma_s(0.0 \text{ K})$ within the experimental error. Therefore, we have only performed high field magnetization measurements on both VPt_3 samples at 4.2 K.

We have analysed the high field $\sigma(H, 4.2 \text{ K})$ magnetization of VPt_3 (1) and (2) as the sum of two different terms :

$$\sigma(H, 4.2 \text{ K}) = \sigma_m(H, 4.2 \text{ K}) + H\chi_H(4.2 \text{ K}).$$

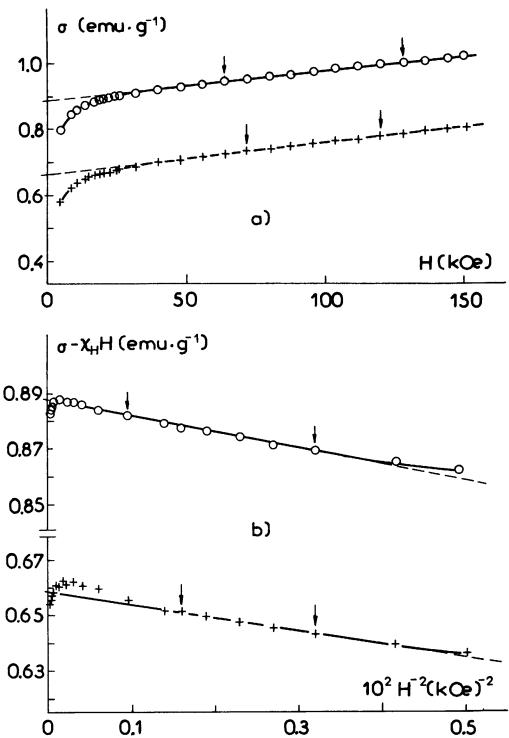


Fig. 5. — Magnetization of both ordered VPt_3 samples at 4.2 K in the high field domain $0 < H \leq 150$ kOe; \circ VPt_3 (1) and $+$ VPt_3 (2) : (a) Magnetization curves σ ($\text{emu} \cdot \text{g}^{-1}$) against field H corrected for demagnetization; (b) Approach to saturation : $\sigma - \chi_H H$ in $\text{emu} \cdot \text{g}^{-1}$ against $10^2 H^{-2}$ (in kOe^{-2}) for

$$14 \text{ kOe} \leq H \leq 150 \text{ kOe}.$$

The first term $\sigma_m(H, 4.2 \text{ K})$ represents the magnetization due to the ferromagnetic moments (ferromagnetic localized or itinerant d electrons of the transition metals V and Pt). The second term $H\chi_H(4.2 \text{ K})$ represents the paramagnetism of all non magnetic elements in VPt_3 (non magnetic V and Pt atoms, host magnetism), the field independent $\chi_H(4.2 \text{ K})$ susceptibility represents the superposed paramagnetism of each ordered VPt_3 alloy at 4.2 K (high field susceptibility).

In the high field limit $H \geq 50$ kOe, the magnetization $\sigma(H, 4.2 \text{ K})$ takes the simple form

$$\sigma(H, 4.2 \text{ K}) = \sigma_s(4.2 \text{ K}) + H\chi_H(4.2 \text{ K})$$

with an accuracy better than 0.5%, indicating that the full saturation of the $\sigma_m(H, 4.2 \text{ K})$ magnetization is reached in relatively high fields :

$$\sigma_m(H, 4.2 \text{ K}) = \sigma_s(H, 4.2 \text{ K})$$

for $H \geq 50$ kOe, within the experimental error of 0.5%. This simple form of the $\sigma(H, 4.2 \text{ K})$ magnetization yields directly the $\chi_H(4.2 \text{ K})$ susceptibility through the high field slope of the magnetization :

$$\frac{d\sigma(H, 4.2 \text{ K})}{dH} = \chi_H(4.2 \text{ K})$$

for $H > 50$ kOe; as can be seen on figure 5a. The superposed paramagnetic susceptibility χ_H (4.2 K) is relatively weak for each VPt₃ sample: χ_H (4.2 K) = 0.9×10^{-6} emu.g⁻¹.Oe⁻¹ for VPt₃ (1), and χ_H (4.2 K) = 1.0×10^{-6} emu.g⁻¹.Oe⁻¹ for VPt₃ (2). The order of magnitude of this χ_H susceptibility is consistent with classical band polarization susceptibilities of transition metals.

Furthermore, the high field magnetization $\sigma(H, 4.2$ K) yields the saturation magnetization σ_s (4.2 K) for each ordered VPt₃ sample through three different laws of approach to saturation:

(i) A first value of the σ_s (4.2 K) magnetization could be derived from the full saturation of the $\sigma_m(H, 4.2$ K) magnetization through the equality $\sigma_m(H, 4.2$ K) = σ_s (4.2 K) in fields $H \geq 50$ kOe (Fig. 5a). It can thus be given

$$\sigma_s(4.2 \text{ K}) = 0.887 \text{ emu.g}^{-1}$$

for VPt₃ (1) against σ_s (4.2 K) = 0.663 emu.g⁻¹ for VPt₃ (2).

(ii) The field dependence of the mean magnetization $\sigma_m(H, 4.2$ K) could be represented for each ordered VPt₃ alloy by the classical expression $\sigma_m(H, 4.2$ K) = σ_s (4.2 K) (1 - b/H^2) with an accuracy better than 1% over the whole field range 17 kOe to 150 kOe (Fig. 5b), where b is a field independent coefficient. The second σ_s (4.2 K) values thus found, are σ_s (4.2 K) = 0.888 emu.g⁻¹ for VPt₃ (1) against σ_s (4.2 K) = 0.659 emu.g⁻¹ for VPt₃ (2).

(iii) A third value of the σ_s (4.2 K) magnetization could be derived from the σ^2 vs. H/σ representation of the high field $\sigma(H, 4.2$ K) magnetization data of each ordered VPt₃ sample at 4.2 K. Figure 6 shows us that the linearity in the σ^2 vs. H/σ plots, is only reached in the high field limit $H \geq 50$ kOe, corresponding to the full saturation of the $\sigma_m(H, 4.2$ K) magnetization. Straight line extrapolation to $(H/\sigma) = 0$ of the linear

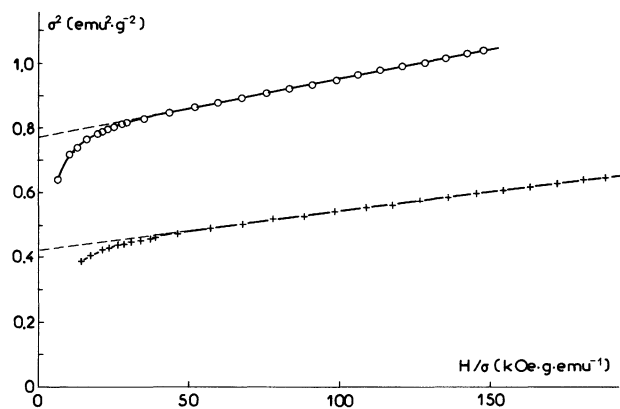


Fig. 6. — A σ^2 (emu².g⁻²) versus H/σ (kOe.g.emu⁻¹) representation of the magnetization data on both ordered VPt₃ alloys at 4.2 K and in high fields ($0 < H \leq 150$ kOe): \circ for VPt₃ (1) and $+$ for VPt₃ (2).

portions in the σ^2 vs. H/σ curves, yields the third σ_s (4.2 K) magnetization values as

$$\sigma_s(4.2 \text{ K}) = 0.878 \text{ emu.g}^{-1}$$

for VPt₃ (1) and σ_s (4.2 K) = 0.649 emu.g⁻¹ for VPt₃ (2).

The three σ_s (4.2 K) magnetization values are in good agreement, in view of the accuracy of our high field magnetization measurements (relative uncertainty of 1% in our $\sigma(H, 4.2$ K) values). Taking account of the three σ_s (4.2 K) values reported for both VPt₃ samples, we choose as final saturation magnetization values, the values

$$\sigma_s(4.2 \text{ K}) = 0.885 \text{ emu.g}^{-1} \text{ for VPt}_3(1) \text{ and}$$

$$\sigma_s(4.2 \text{ K}) = 0.658 \text{ emu.g}^{-1} \text{ for VPt}_3(2).$$

The absolute ferromagnetic moment μ_0 of each ordered VPt₃ alloy is derived from these final σ_s (4.2 K) magnetization values through the assumption σ_s (4.2 K) = σ_s (0 K). We found $\mu_0 = 0.101 \mu_B$ per V atom-gram for the VPt₃ (1) sample and $\mu_0 = 0.075 \mu_B$ per V atom-gram for the VPt₃ (2) sample. One V atom-gram corresponds to 636.64 g of VPt₃ alloy.

According to these small ferromagnetic moment values, both ordered VPt₃ alloys may be considered as weak ferromagnets, if compared to other ferromagnetic V compounds, as ordered Au₄V or ordered V(Ir_{1-x}Pt_x)₃ compounds with Cu₃Au structure. The ferromagnetic moment of ordered Au₄V has been reported as μ ranging from 0.3 to 0.8 μ_B per V atom-gram, depending on the samples and the authors (Creveling *et al.* [28], Claus *et al.* [29], Chin *et al.* [30], Adachi *et al.* [31]). According to Goto and Yamauichi [20], the ferromagnetic moment of V(Ir_{1-x}Pt_x)₃ compounds ordered in the Cu₃Au like structure, reaches a maximal value of $\mu \simeq 0.34 \mu_B$ per V atom-gram for $x \simeq 0.73$. At first sight, this comparison favours the concept of itinerant ferromagnetism in both ordered VPt₃ alloys, but it is not a sufficient proof for the validity of an itinerant model.

6. Paramagnetic behaviour above the Curie point

T_c. — We have performed paramagnetic susceptibility measurements on each ordered VPt₃ alloy in the temperature range 300 K to 1 100 K. We have not exceeded the temperature limit of 1 100 K, in order to avoid any transformation in the ordered structure (Cu₃Au like or TiAl₃ like) of each studied sample. The paramagnetic susceptibility $\chi(T)$ of each VPt₃ sample was found to be field independent in the whole temperature range 300 K to 1 100 K, within the experimental error of about 1%. The paramagnetic behaviour of each VPt₃ sample is described with an accuracy better than 5% in the temperature range 340 K to 1 000 K by the modified Curie-Weiss law $\chi(T) = \chi' + C(T - \theta)^{-1}$, as shown by figure 7.

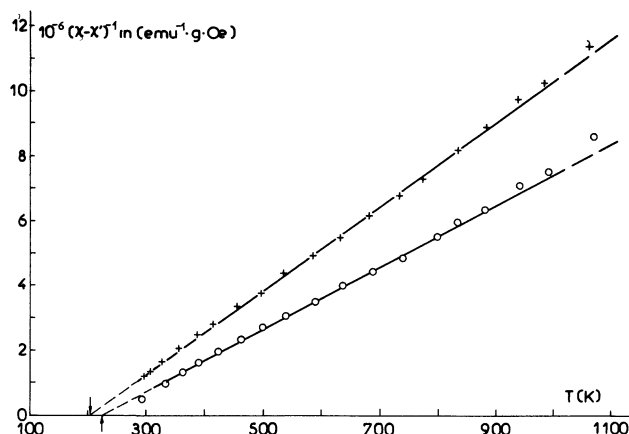


Fig. 7. — Paramagnetic behaviour of both ordered VPt_3 alloys above their Curie point T_c (\circ VPt_3 (1) sample and $+$ VPt_3 (2) sample). Inverse $(\chi - \chi')^{-1}$ of the susceptibility difference $(\chi - \chi')$ against temperature T (K) between 295 K and 1 050 K. The arrows indicate the paramagnetic Curie point θ of each VPt_3 sample ($\theta = 224$ K for VPt_3 (1) and $\theta = 204$ K for VPt_3 (2)).

The Curie-Weiss parameters χ' , C and θ have their usual meaning.

The constant susceptibility term χ' is relatively small ($\chi' = 0.471 \times 10^{-6} \text{ emu.g}^{-1}.\text{Oe}^{-1}$ for the VPt_3 (1) sample and $\chi' = 0.503 \times 10^{-6} \text{ emu.g}^{-1}.\text{Oe}^{-1}$ for the VPt_3 (2) sample). Nevertheless, its order of magnitude (that of the high field susceptibility χ_H) is consistent with the d band paramagnetism of transition metals.

The paramagnetic Curie temperature θ is near to the ferromagnetic one T_c : $\theta = 224$ K against $T_c = 240$ K for VPt_3 (1) and $\theta = 204$ K against $T_c \approx 210$ K for VPt_3 (2). This fact ensures the physical meaning of the θ parameter. The Curie-Weiss constant C yields the paramagnetic moment μ' of each ordered VPt_3 alloy: $\mu' = 0.73 \mu_B$ per V atom-gram for VPt_3 (1) and $\mu' = 0.63 \mu_B$ per V atom-gram for VPt_3 (2).

These paramagnetic moment values ($\mu' \sim 0.7 \mu_B$ per V atom-gram) are much smaller than the theoretical value $\mu'_V \approx 1.73 \mu_B$ per V atom-gram, corresponding to V atoms with spin $S = 1/2$. At first sight, this fact favours the concept of itinerant ferromagnetism in both ordered VPt_3 alloys. Nevertheless, within a model of magnetism localized on the V atoms, this fact means that (i) only a fraction of the V atoms are magnetic, or (ii), by an adequate exchange interaction mechanism, the Pt conduction electrons acquire a magnetic polarization antiparallel to the V moments.

In order to clear up the question whether a localized or an itinerant model of ferromagnetism would be adequate for both ordered VPt_3 alloys, we have calculated the numbers q_c and q_s of magnetic carriers in each VPt_3 alloy, and their q_c/q_s ratio. q_c is the number of magnetic carriers as deduced from the paramagnetic moment μ' and q_s that deduced from the ferromagnetic moment μ (Rhodes and Wohlfarth [32]).

The q_c/q_s ratio has the value 2.36 for VPt_3 (1) and the value 2.41 for VPt_3 (2). According to the (q_c/q_s) versus T_c representation (Fig. 8) proposed by Rhodes and Wohlfarth [32], the ferromagnetism of each ordered VPt_3 alloy may be considered as a limit case between an itinerant model and a localized model.

In spite of this ambiguous result, the ferromagnetic behaviour of both ordered VPt_3 alloys may be satisfactorily interpreted within the localized model of ferromagnetism proposed by Kawakami and Goto [21] for their $\text{V}(\text{Ir}_{1-x}\text{Pt}_x)_3$ compounds, as will be seen in subsection 7.2.

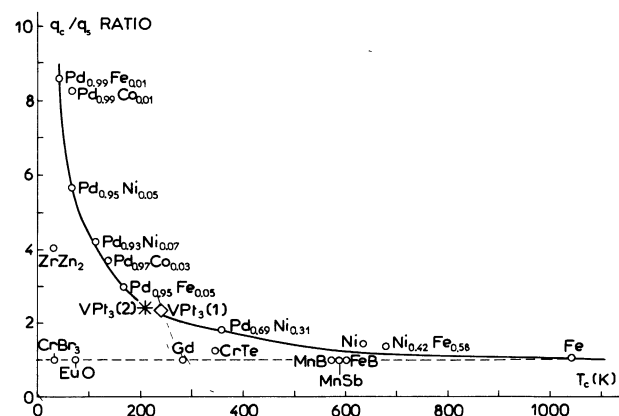


Fig. 8. — Magnetic carrier number ratio q_c/q_s plotted as a function of the ferromagnetic Curie temperature T_c for several metals, alloys and compounds (\circ), and for both ordered VPt_3 alloys (\diamond for VPt_3 (1) and $*$ for VPt_3 (2)).

7. Discussion and conclusion about the origin of ferromagnetism in both ordered VPt_3 alloys. — The ferromagnetic behaviour of both ordered VPt_3 alloys is undoubtedly established in view of all experimental results obtained in this work. Now, we attempt to clarify the origin of ferromagnetism in these ordered VPt_3 alloys.

7.1 KIND OF FERROMAGNETISM IN ORDERED VPt_3 (ITINERANT OR LOCALIZED ?). — First, we summarize briefly our previous discussions about the question whether this ferromagnetism may be classified as itinerant or localized. As already mentioned in section 3, no conclusion can be drawn in this sense from the observed $(1 - AT^{3/2})$ decreasing rate of the low temperature spontaneous magnetization $\sigma_{sp}(T)$.

For the typical itinerant ferromagnets ZrZn_2 and Sc_3In , it has been reported (Ogawa [33], Mattocks and Melville [34], Gardner *et al.* [35]) a non-saturating low temperature magnetization in fields $H \geq 40$ kOe, even in the high fields $H \geq 170$ kOe [34] for ZrZn_2 , with an important high field susceptibility ($\chi_H \approx 190 \times 10^{-6} \text{ emu.g}^{-1}.\text{Oe}^{-1}$ for Sc_3In at 1.23 K and 40 kOe, [35]). But we have seen in section 5 that the magnetization $\sigma(H, 4.2 \text{ K})$ of each ordered VPt_3 sample at 4.2 K, reached full saturation in fields

$H \geq 50$ kOe, with a weak high field susceptibility χ_H (4.2 K) $\simeq 1 \times 10^{-6}$ emu.g⁻¹.Oe⁻¹. These arguments favour the concept of localized ferromagnetism in both ordered VPt₃ alloys.

On the other hand, the small magnetic moments ($\mu_0 \sim 0.1 \mu_B$ per V atom-gram) of VPt₃ (1) and (2), have the same order of magnitude as those reported for ZrZn₂ and Sc₃In by Ogawa [33], Mattocks and Melville [34], Gardner *et al.* [35]. Nevertheless, this argument is not sufficient to prove the itinerant character of the ferromagnetism of both ordered VPt₃ alloys. Moreover, we shall see in subsection 7.2 that these small μ_0 values ($\mu_0 \sim 0.1 \mu_B$ per V atom-gram) may be well interpreted in a localized model of ferromagnetism.

We have already mentioned in section 6 that, according to Rhodes and Wohlfarth [32], the q_c/q_s versus T_c representation of figure 8, classified the ferromagnetism of both ordered VPt₃ alloys as a limit case between a localized model and an itinerant one.

Finally, we cannot settle with certainty whether the ferromagnetic behaviour of both ordered VPt₃ alloys has to be interpreted in a localized model of ferromagnetism rather than in an itinerant one.

The same uncertainty still remains for the interpretation of the ferromagnetism of ordered Au₄V. Indeed, the ferromagnetism of this compound has been recognized as well localized, according to Claus *et al.* [29], Creveling and Luo [22], Dunlap *et al.* [36], Cohen *et al.* [37]. But, in a recent investigation on a Au₄V single crystal, Adachi *et al.* [31] interpreted the ferromagnetism of ordered Au₄V in the spin fluctuation scheme of itinerant electron magnetism described by Moriya [38], and by Moriya and Takahashi [39].

7.2 INTERPRETATION OF THE FERROMAGNETISM IN BOTH ORDERED VPt₃ ALLOYS. — We attempt to clarify the origin of ferromagnetism in both ordered VPt₃ alloys within a model of localized magnetism. For this, we refer to the investigations on the Au₄V compound reported by Creveling and Luo [22], Claus *et al.* [29], Chin *et al.* [30], Dunlap *et al.* [36], Cohen *et al.* [37], and to the investigations on the ordered V(Ir_{1-x}Pt_x)₃ compounds reported by Goto and Yamauchi [20], Kawakami and Goto [21], since :

(i) the ordered Au₄V compound and the ordered V(Ir_{1-x}Pt_x)₃ compounds are also V compounds in which magnetic ordering occurs ;

(ii) the Au₄V compound, like VPt₃, is paramagnetic in the atomically disordered state, and becomes ferromagnetic in the atomically ordered state ;

(iii) the ordered V(Ir_{1-x}Pt_x)₃ compounds are isomorphous to VPt₃ ordered in the Cu₃Au structure.

The absence of the nearest neighbour V-V pairs is believed to be responsible for the appearance of localized magnetic moments on the V atoms in all these compounds ([21, 29, 30]). Indeed, the distance of V-V pairs in ordered Au₄V as in ordered V(Ir_{1-x}Pt_x)₃

is about 4 Å, while the nearest neighbour V-V distance in disordered Au₄V and in disordered (Cu like) VPt₃ is about 2.8 Å. Thus, the appearance of ferromagnetism in ordered Au₄V as in ordered V(Ir_{1-x}Pt_x)₃ compounds, may be ascribed to the (indirect) ferromagnetic coupling of the V moments through the conduction electrons of Au or Pt ([21, 36, 37]). The ferromagnetic moments of the V atoms in ordered Au₄V is given as $\mu_V \simeq 0.9 \mu_B$ per V atom-gram ([30, 37]); this value of the moment corresponds roughly to V atoms with spin $S = 1/2$ and to the theoretical paramagnetic moment $\mu'_V = 1.73 \mu_B$ per V atom-gram. The lower ferromagnetic moment values ($\mu \sim 0.3$ to $0.8 \mu_B$ per V atom-gram) observed on the various Au₄V samples investigated in the literature ([28-31]), are ascribed to the large magneto-crystalline anisotropy found in this compound.

In view of the above discussion, we apply to both ordered VPt₃ alloys studied here, the local model of ferromagnetism depicted by Kawakami and Goto [21] for the ordered V(Ir_{1-x}Pt_x)₃ compounds : a model in which (i) the V atoms have a localized moment $\mu_V \simeq 1 \mu_B$ only if they are surrounded by nine or more Pt atoms at the nearest neighbour sites, and (ii) the ferromagnetism arises from a mechanism of indirect exchange interaction between the V moments through the conduction electrons of the Pt atoms. In this mechanism, the magnetic polarization of these Pt electrons, has to be considered as antiparallel to the V moments (Campbell [40], Kawakami and Goto [21]). According to Kawakami and Goto [21], the magnitude of the Pt polarization may be estimated as $\mu_{Pt} \simeq -0.3 \mu_B$ per Pt atom-gram, while the magnetic moment of the V atoms may be taken as $\mu_V \simeq +1.0 \mu_B$ per V atom-gram. Within this model and with the above numerical values, we explain easily :

(i) the weak ferromagnetic moment values ($\mu_0 \simeq 0.1 \mu_B$ per V atom-gram) found here for both ordered VPt₃ alloys : taking $\mu_0 = \mu_V + 3 \mu_{Pt}$, one finds $\mu_0 \simeq 1 \times (1.0) + 3 \times (-0.3) \simeq 0.1 \mu_B$ per V atom-gram (or 636.64 g of VPt₃);

(ii) the consequently low paramagnetic moment values found here for VPt₃ (1) and (2) : $\mu' \simeq 0.7 \mu_B$ per V atom-gram, instead of the theoretical value $\mu'_V \simeq 1.73 \mu_B$ per V atom-gram, corresponding to the V atoms with spin $S = 1/2$;

(iii) why Kawakami and Goto [21] found only a very weak spontaneous magnetization on their VPt₃ sample and, however, occurrence of magnetic ordering below 206 K : as a consequence of the heat treatment or rather of the mechanical treatment undergone by Kawakami's and Goto's VPt₃ sample, the V magnetic moment in this VPt₃ sample, has been very nearly (or, even totally) cancelled out by the Pt polarization.

7.3 CONCLUSION. — From all the previous results and discussions, the following conclusions may be drawn. As depicted by Kawakami and Goto [21],

the ferromagnetic coupling mechanism of indirect exchange interaction between the localized V moments through the Pt conduction electrons, gives a satisfactory interpretation of all experimental results obtained in this work on the ferromagnetic behaviour of both ordered VPt₃ alloys (temperature and field dependence of the ferromagnetic magnetization, paramagnetic behaviour above the Curie point).

Moreover, we shall see in paper II [19] that the above propositions are in accordance with the results of our investigations on the critical behaviour of VPt₃ (1) and (2) around their respective Curie point.

Nevertheless, our interpretation of all these experimental results in a localized model of ferromagnetism, may not be entirely convincing, since, at the present

stage, we could not clearly answer the question whether the ferromagnetism of these VPt₃ alloys may be classified as itinerant or localized.

Therefore, further investigations, such as Mössbauer effect measurements and neutron diffraction studies are needed to elucidate definitively this question.

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