

Strong Correlation between Ferromagnetic Superconductivity and Pressure-enhanced Ferromagnetic Fluctuations in UGe₂

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We have measured magnetization at high pressure in the uranium ferromagnetic superconductor UGe₂ and analyzed the magnetic data using Takahashi's spin fluctuation theory. There is a peak in the pressure dependence of the width of the spin fluctuation spectrum in the energy space T_0 at P_x , the phase boundary of FM1 and FM2 where the superconducting transition temperature T_{sc} is highest. This suggests a clear correlation between the superconductivity and pressure-enhanced magnetic fluctuations developed at P_x . The pressure effect on T_{Curie}/T_0 , where T_{Curie} is the Curie temperature, suggests that the less itinerant ferromagnetic state FM2 is changed to a more itinerant one FM1 across P_x . Peculiar features in relations between T_0 and T_{sc} in uranium ferromagnetic superconductors UGe₂, URhGe, and UCoGe are discussed in comparison with those in high- T_c cuprate and heavy fermion superconductors.

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Ferromagnetism and usual s -wave superconductivity are antagonistic phenomena since the superconducting pairs are easily destroyed by the ferromagnetic exchange field. The coexistence of superconductivity and ferromagnetism both carried by the same electrons has been considered as a fantastic theoretical possibility since its prediction by Ginzburg [1]. The $4f$ -localized systems such as ErRh₄B₄ [2,3], HoMo₆S₈ [4], and ErNi₂B₂C [5] show the coexistence of both phases. The ferromagnetism and superconductivity of the systems are carried, however, by different electrons: f and d electrons, respectively, and the phases compete each other. Therefore, the discoveries of the superconductivity in uranium ferromagnets UGe₂ [6,7], URhGe [8], and UCoGe [9] are very interesting since the same $5f$ electrons of the uranium atoms are responsible for both states [10].

Let us look other systems. Generally, unconventional superconductivity appears around phase boundaries of magnetic phases in strongly correlated electron systems [11,12]. An important issue that could be elucidated experimentally is finding a relation between the magnetism and the superconductivity. Neutron scattering studies have shown relationships between the superconductivity and magnetic excitations in high- T_c cuprate [13], iron arsenide [14], and heavy fermion superconductors [15–17]. Correlations between the superconductivity and ferromagnetic fluctuations have been studied by nuclear magnetic resonance experiments on UCoGe [18] and URhGe [19]. Compared with the extensive studies of these two compounds, there have been relatively few experimental studies of UGe₂ where the superconductivity appears only at high pressure. This is due to the difficulty of making measurements at a very low temperature and high pressure. In this Letter, we report a clear correlation

between the superconductivity and pressure-enhanced ferromagnetic fluctuations in UGe₂.

Figure 1 shows the temperature-pressure phase diagram of UGe₂ [20,21]. Open circles and closed triangles represent the Curie temperature T_{Curie} determined by previous resistivity [20] and our present magnetic measurements, respectively. T_{Curie} decreases with increasing pressure from 53 K at ambient pressure. The transition changes from the second to first order at a tricritical point (TCP: $T_{TCP} \sim 22$ K, $P_{TCP} \sim 1.42$ GPa), denoted as a filled orange circle, and disappears above a critical pressure $P_c \sim 1.5$ GPa [10,22]. There is an additional boundary T_x that splits the ferromagnetic phase into FM2 and FM1. Open diamonds represents

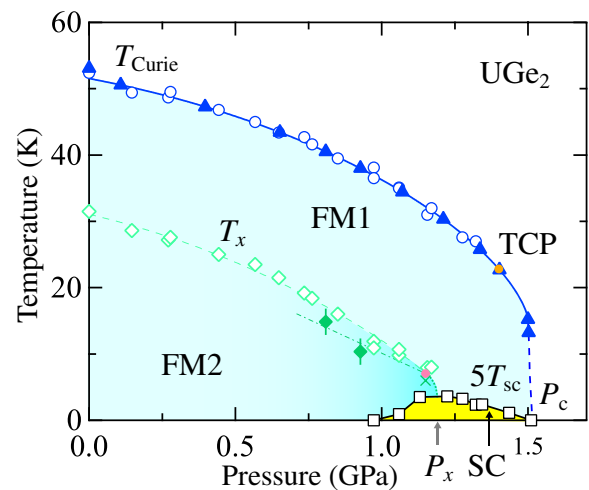


FIG. 1. Temperature-pressure phase diagram in UGe₂ determined by present magnetic and previous resistivity measurements [20]. Dotted and dashed lines for T_x are guides for the eye.

T_x determined by resistivity measurements [20]. The critical pressure of T_x is $P_x \sim 1.20$ GPa. The superconductivity appears from approximately 1.0 GPa to P_c . The spontaneous magnetic moment p_s , the coefficient of the T^2 term in the resistivity A , and the linear specific heat coefficient γ show drastic changes at P_x [6,7,20,23,24]. The difference of Fermi surfaces between FM1 and FM2 was also reported [25,26]. The microscopic origin of T_x has not been understood yet. The transition between FM1 and FM2 at low temperatures is first order [10]. The first order transition at T_x changes to a crossover at a critical end point (CEP: $T_{\text{CEP}} \sim 7$ K, $P_{\text{CEP}} \sim 1.16$ GPa) denoted as a filled magenta circle [10,22]. The superconducting transition temperature T_{sc} becomes highest near P_x .

We used a high-quality single crystal of UGe_2 with residual resistivity ratio $RRR = 600$. The details of the sample preparation were reported previously [20,21]. We have measured magnetization at high pressure with a miniature ceramic-anvil high-pressure cell (MCAC) designed by us for use in a commercial SQUID magnetometer [27–29]. We used ceramic anvils with a culet size of 1.8 mm and a Cu-Be gasket with an initial thickness of 0.9 mm. The diameter of the sample space in the gasket was 0.90 mm. A $0.50 \times 0.40 \times 0.50$ mm³ single crystal was placed in the sample space with Daphne 7373 as a pressure-transmitting medium [30,31]. The pressure values at low temperatures were determined from the pressure dependence of T_{sc} of Pb placed in the sample space [32].

The development of longitudinal magnetic fluctuations in UGe_2 has been suggested from NMR experiments [33–35]. The magnetic data in UGe_2 have been analyzed using Takahashi's spin fluctuation theory to study the dynamical magnetic property in FM1 and FM2 [36,37]. Recently, we have shown the applicability of the theory to most actinide $5f$ electrons ferromagnets [38]. We determined the widths of the spin fluctuation spectrum T_0 and T_A in energy ω and momentum \mathbf{q} spaces, respectively. The mode-mode coupling term F_1 was obtained from the slope ζ of the Arrott plot (M^2 versus H/M plot) at 2.0 K with the relation $F_1 = N_A^3 (2\mu_B)^4 / k_B \zeta$, where N_A is Avogadro's number and k_B is the Boltzmann constant. T_0 and T_A can be estimated with the value of p_s using Eqs. (1) and (2).

$$\left(\frac{T_C}{T_0}\right)^{5/6} = \frac{p_s^2}{5g^2 C_{4/3}} \left(\frac{15cF_1}{2T_C}\right)^{1/2} \quad (1)$$

$$\left(\frac{T_C}{T_A}\right)^{5/3} = \frac{p_s^2}{5g^2 C_{4/3}} \left(\frac{2T_C}{15cF_1}\right)^{1/2}, \quad (2)$$

where g represents the Lande's g factor and $C_{4/3}$ is a constant ($C_{4/3} = 1.006089 \dots$) [36,37].

The slope ζ of the Arrott plot was determined from the data in a wide magnetic field region up to 7 T since the data points form almost linear straight lines at low temperatures

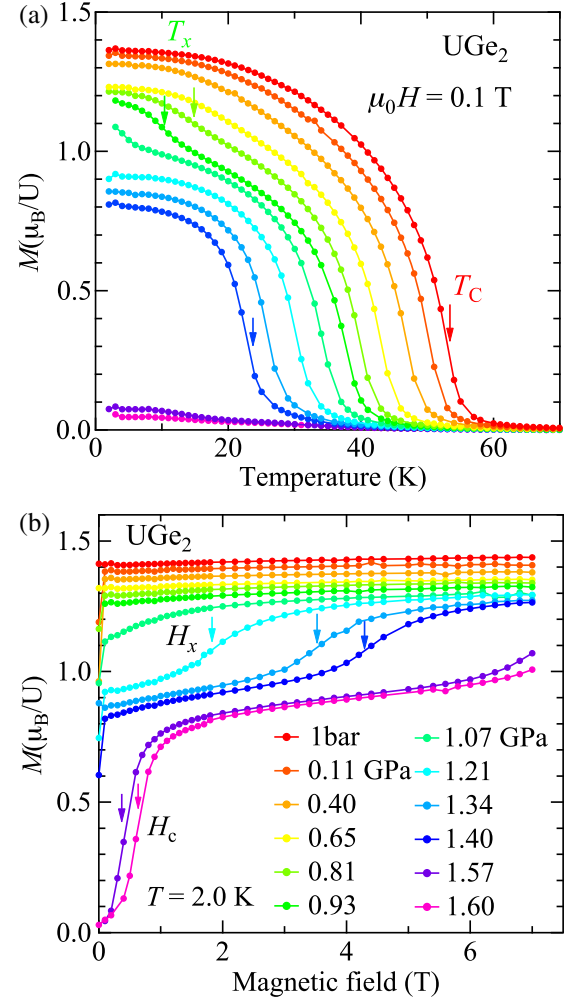


FIG. 2. (a) Temperature dependencies of the magnetization in applied magnetic field of 0.1 T and (b) magnetic field dependencies of the magnetization under ambient and several pressures at $T = 2.0$ K in UGe_2 .

in FM2. Meanwhile, the magnetization $M(H)$ shows the metamagnetic transition at H_x above P_x . We analyzed the data up to $H = 0.5\text{--}0.6 H_x$ below which the linearity of the Arrott plot is fulfilled in FM1.

Figure 2(a) shows the temperature dependencies of the magnetization $M(T)$ in a magnetic field of 0.1 T applied along the magnetic easy a axis at ambient and several pressures. T_C is defined as the point where $-dM(T)/dT$ is a maximum in a low magnetic field of 0.01 T and is indicated by an arrow in the figure. The pressure dependence of T_C is consistent with that determined by the resistivity measurement as shown in Fig. 1 [20]. The magnetization increases with decreasing temperature monotonically below T_C in the low pressure region below 0.65 GPa. A change in the T dependence of $M(T)$ appears at T_x above 0.81 GPa. Closed diamonds in Fig. 1 represents T_x ($= 14.8$ and 10.4 K at 0.81 and 0.93 GPa, respectively) defined from the peak position in $-dM(T)/dT$. The value of T_x cannot be determined correctly for 1.07 GPa since the

number of the data points at lower temperatures is not enough for the correction determination of T_x . A cross in Fig. 1 represents T_x , which is determined in our previous study to be the specific heat under high pressure [24].

The values of T_x determined in the present study are slightly lower than those by the resistivity measurement. This difference in T_x might be related to two anomalies in the temperature dependence of the thermal expansion [22]. The crossover region of T_x is bound by two lines in the pressure-temperature diagram below P_{CEP} . The resistivity shows an anomaly only at the higher temperature line. The plotted data points of T_x in the present study lie close to the lower line [22]. The difference in T_x may be an interesting problem, but it is left for the future. Above P_x where the ground state is FM1, $M(T)$ shows a simple ferromagnetic behavior at 1.21, 1.34, and 1.40 GPa. The value of the low temperature magnetization becomes less than $1.0 \mu_B/U$ in FM1. $M(T)$ does not show the ferromagnetic behavior at 1.57 and 1.60 GPa, suggesting that the critical pressure P_c for the ferromagnetism is about 1.5 GPa.

Figure 2(b) shows the magnetic field dependence of the magnetization $M(H)$ at 2.0 K at ambient pressure and several pressures. The magnetization shows a simple ferromagnetic behavior in FM2. The magnetization decreases weakly with increasing pressure below P_x . Above the critical pressure, the value of p_s is reduced to less than $1.0 \mu_B/U$ in FM1. $M(H)$ in FM1 increases with increasing magnetic field at low fields and shows an anomalous increase and metamagnetic transition at $H_x = 1.80, 3.54,$ and 4.33 T for 1.21, 1.34, and 1.40 GPa, respectively, where the transition from FM1 to FM2 occurs [21]. Above P_c , the ground state is in the paramagnetic state at zero magnetic field. However, the magnetization increases drastically at $H_c = 0.42,$ and 0.65 T for 1.57 and 1.60 GPa, respectively, where FM1 is induced from the paramagnetic state [25,26]. $M(H)$ increases simply with increasing field and shows a weak nonlinear increase again above 6.0 and 6.4 T at 1.57 and 1.60 GPa, respectively. This suggests that the recovery of FM2 for $H > H_x$ above 7.0 T.

The decreases of T_C and p_s under compression suggest a pressure-driven magnetic instability towards the ferromagnetic to paramagnetic quantum phase transition at P_c [10,39]. The present results of the magnetic data are basically consistent with those in previous magnetic measurements under high pressure [21,23,40]. The pressure dependence of H_x is consistent with those in our previous studies [21,26], but the values of H_x are about 15% larger than those in Ref. [23]. The reason of the discrepancy is not clear.

We analyzed the magnetic data at 2.0 K using Takahashi's spin fluctuation theory. Figure 3(a) shows the pressure dependencies of spin fluctuation parameters T_0 and T_A : the widths of the spin fluctuations spectrum in energy ω and momentum q spaces, respectively. T_0 and T_A show an anomalous enhancement where the superconductivity appears from 1.0 GPa to P_c . This suggests a change of

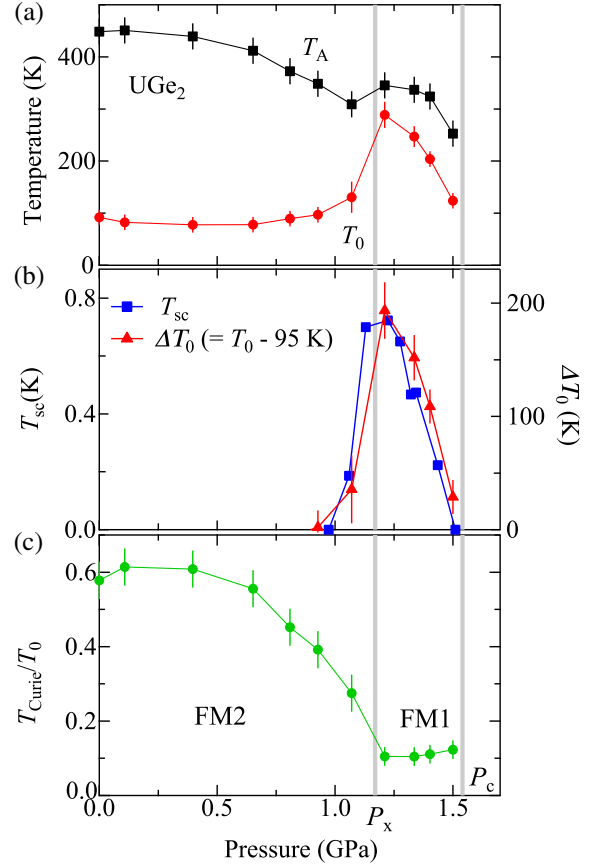


FIG. 3. Pressure dependencies of (a) T_0 and T_A , the widths of the spin fluctuations spectrum in energy ω and momentum q space, respectively, determined from the analysis of the data at 2.0 K, (b) T_{sc} (left axis) [20] and $\Delta T_0(P) [= T_0(P) - 95 \text{ (K)}]$ (right axis), and (c) T_{Curie}/T_0 in UGe_2 .

the spin fluctuation spectrum. There is a clear peak in the pressure dependence of T_0 and its peak position is close to P_x where T_{sc} is highest. When the pressure dependence of T_0 is expressed as $T_0(P) = T_0^* + \Delta T_0(P)$ where $T_0^* = 95$ K is a pressure-independent term, the pressure dependence of $\Delta T_0(P)$ scales with that of $T_{\text{sc}}(P)$ determined by our previous resistivity measurement, as shown in Fig. 3(b) [20]. Theoretical studies have assumed ferromagnetic superconductivity driven by critical fluctuations around a ferromagnetic quantum critical point (QCP) [41–43]. This study suggests that the superconductivity in UGe_2 is driven by the anomalous magnetic fluctuations with the characteristic energy of 300 K developed around P_x .

We analyzed the magnetic data read from Ref. [23] and determined the pressure dependencies of T_0 and T_A . The obtained result is compatible with that in the present Letter.

The drastic changes have been observed in the pressure dependence of $A, \gamma, p_s,$ and Fermi surfaces at P_x [23–26]. Although several theoretical interpretations have been proposed [44–48], a full microscopic understanding of the transition has remained an open question. Within

phenomenological Stoner theory, the magnetic features of the FM1-FM2 transition could be understood if the Fermi surface passes through peaks in the density of states [44]. The pairing interaction λ_Δ is strongly enhanced at P_x in the Stoner theory [44]. However, the calculated large value of λ_Δ above P_c in the theory seems not applicable to UGe_2 where the superconductivity appears only below P_c . Further studies are necessary to elucidate the dynamical magnetic property around P_x .

The ferromagnetism in the uranium ferromagnetic superconductors is carried by the itinerant $5f$ electrons [10,49,50]. Here, we discuss differences between FM1 and FM2 from a parameter T_{Curie}/T_0 that reflects the itinerancy of the magnetic fluctuations in the spin fluctuation theory [37]. The smaller value of T_{Curie}/T_0 indicates a weak itinerant ferromagnetism and the local magnetic moment is responsible for the ferromagnetism for $T_{\text{Curie}}/T_0 = 1$. Figure 3(c) shows the pressure dependence of T_{Curie}/T_0 . T_{Curie}/T_0 is approximately 0.6 below 0.4 GPa in UGe_2 . The values of T_{Curie}/T_0 and p_s ($1.41 \mu_B/\text{U}$ at 1 bar) suggest strong itinerant ferromagnetism in FM2. This feature is in contrast with weak itinerant ferromagnetism in URhGe and UCoGe where the values of T_{Curie}/T_0 and p_s are 0.121 and $0.41 \mu_B/\text{U}$, and 0.0065 and $0.039 \mu_B/\text{U}$, respectively, at 1 bar [38,51]. In UGe_2 , T_{Curie}/T_0 decreases with increasing pressure above 0.6 GPa. The value of the parameter becomes less than 0.3 above 1.0 GPa where the superconductivity starts to appear. T_{Curie}/T_0 shows an almost pressure-independent value of about 0.1 in FM1. This suggests that the less itinerant ferromagnetic state of FM2 in the low pressure region is changed to the more itinerant one of FM1. This pressure dependence of T_{Curie}/T_0 may be related to the changes of the various physical quantities or Fermi surfaces at P_x . We suggest that the degree of the itinerancy of the $5f$ electrons changes across P_x . This result could have relevance to theoretical study with the periodic Anderson model that shows the change of the local f electron state inside the ferromagnetic state [47]. It is interesting to note that the value of T_{Curie}/T_0 of FM1 is similar to that in URhGe. A certain degree of itinerancy of the $5f$ electrons might be necessary for the coexistence of the superconductivity and the ferromagnetism.

Relations between T_0 and T_{sc} in UGe_2 are discussed quantitatively. T_{sc} is most sensitive to T_0 in the strong coupling theory for spin fluctuation-induced superconductivity [52,53]. The spin fluctuations with higher frequencies are effective for superconductors with high transition temperatures. The correlation between the two quantities has been pointed out in several strongly correlated electron superconductors [54–58]. Figure 4 shows relations between T_{sc} and T_0 for UGe_2 , URhGe [38] and UCoGe [51], heavy fermion superconductors, and high- T_c cuprate superconductors. The values of T_0 in CePt_3Si [59], NpPd_5Al_2 [60], PuRhGa_5 [61], PuCoGa_5 , PuCoIn_5 , and PuRhIn_5 are

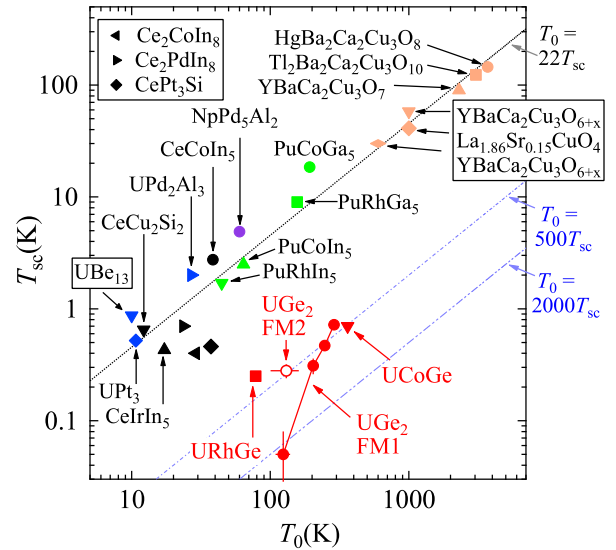


FIG. 4. Relations between the superconducting transition temperatures T_{sc} and the energy spread of spin fluctuations T_0 for UGe_2 , URhGe [38] and UCoGe [51], heavy fermion, and high- T_c cuprate superconductors [54–58].

determined by us from the reported γ value with a theoretical expression ($T_0 \approx 1.25 \times 10^4/\gamma$) [62]. We plot the data of the other systems determined by various experimental methods cited from literature [54–58]. The data of the cuprate and the heavy fermion superconductors are plotted around a straight dotted line with $T_0 = 22T_{\text{sc}}$ denoted as a dotted line in Fig. 4, suggesting a common feature in the superconductivity. d -wave superconductivity has been experimentally suggested in a number of superconductors in the strongly correlated electron systems [11,61,63]. Theoretical studies have shown that an optimum frequency ω_{opt} of the antiferromagnetic spin fluctuation spectrum that contributes to raise T_{sc} the most is approximately $10 T_{\text{sc}}$ for the d -wave superconductivity [64,65]. It is reasonable that the data of the cuprates and heavy fermion superconductors are plotted comparably close to the solid line. Meanwhile, the data for UGe_2 , URhGe, and UCoGe largely deviate from the relation, suggesting peculiar features in the uranium ferromagnetic superconductors. The data points in FM1 of UGe_2 are plotted roughly between lines with $T_0 = 500T_{\text{sc}}$ and $T_0 = 2000T_{\text{sc}}$ shown as one and two dot chain lines, respectively. Spin fluctuations with characteristic energy more than two or three orders of magnitude larger than T_{sc} play an important role for the ferromagnetic superconductivity. The values of T_{sc} in FM1 of UGe_2 are more than one order of magnitude smaller than those of the d -wave superconductors PuCoGa_5 and PuRhGa_5 [61]. Note that the values of T_0 in the plutonium superconductors are similar to those in FM1 of UGe_2 . Theoretical calculation has shown that T_{sc} for d -wave pairing in nearly antiferromagnetic metals is about one order magnitude larger than that for the p -wave pairing in nearly ferromagnetic metals for comparable conditions such as

the bandwidth or strength of the pairing interaction [66]. Thus, the difference in T_{sc} could be understood if we assume the p -wave superconductivity suggested for the uranium ferromagnetic superconductors from anomalous behaviors of the upper critical field H_{c2} [67–69].

The relation between T_{sc} and T_0 is expressed as $T_{sc} \propto (T_0)^\alpha$ with $\alpha = 2.3 \pm 0.1$ in FM1, which is contrary to the cuprate and heavy fermion superconductor where the linear relation has been discussed. This may reflect unique features in the superconductivity in UGe₂. In addition, recent NMR and uniaxial compression studies have suggested the importance of transverse magnetic fluctuations in URhGe [19,70]. Although the primary parameter that determines T_{sc} is the strength of the longitudinal magnetic fluctuations, it may be necessary to consider the transverse magnetic fluctuations for a complete understanding of the uranium ferromagnetic superconductors [68].

In conclusion, we have measured the magnetization of UGe₂ at high pressure. The analysis of the magnetic data with Takahashi's spin fluctuation theory suggests that the superconductivity in UGe₂ is mediated by magnetic fluctuations with characteristic energy of 300 K developing around P_x , the first order phase boundary of FM1 and FM2 where T_{sc} is highest. The pressure dependence of T_{Curie}/T_0 suggests that the less itinerant ferromagnetic state FM2 is changed to the more itinerant one FM1 across P_x . Peculiar features in the relations between T_0 and T_{sc} in uranium ferromagnetic superconductors UGe₂, URhGe, and UCoGe are discussed in comparison with those in high- T_c cuprate and heavy fermion superconductors.

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