Field-induced Bose-Einstein condensation of interacting dilute magnons in three-dimensional spin systems: A renormalization-group study

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We use the renormalization-group method to study the magnetic field influence on the Bose-Einstein condensation of interacting dilute magnons in three-dimensional spin systems. We first considered a model with $SU(2)$ symmetry (universality class $z=1$) and we obtain for the critical magnetic field a power law dependence on the critical temperature, $[H_c(T)-H_c(0)]\sim T^2$. In the case of $U(1)$ symmetry (universality class $z=2$) the dependence is different, and the magnetic critical field depends linearly on the critical temperature, $[H_c(T)-H_c(0)]\sim T$. By considering a more relevant model, which includes also the system's anisotropy, we obtain for the same symmetry class a $T^{3/2}$ dependence of the magnetic critical field on the critical temperature. We discuss these theoretical predictions of the renormalization group in connection with experimental results reported in the literature.

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I. INTRODUCTION

Bose-Einstein condensation (BEC) remains one of the most important macroscopic effects predicted a long time ago by quantum mechanics for an ideal Bose gas. Lately, the interest in BEC was renewed by the practical realization of a condensate phase in weak interacting Bose systems realized in ultracooled diluted alkali atomic gases.[1] Further, a BEC of fermionic pairs was achieved in trapped fermionic systems under attractive fermion-fermion interaction.[2] The limitation of these experiments is due to a reduced number of particles which undergo BEC. Recently, BEC was associated with the magnetic transition observed in different quantum spin systems. A particular class of such materials is formed by the $X\mathrm{CuCl}_3$ dimer compounds (with X being Tl, K, or NH_4).[3,4] In the ground state these materials are spin singlets which are magnetically inactive. If a high-magnetic field is applied, they undergo a transition into a magnetically ordered state, a transition which can be understood as a condensation of excitations which behaves as bosonic quasiparticles.

One particular example is TlCuCl_3. The quantum magnetism in this compound is attributed to the spin-1/2 Cu^{2+} ions positioned in the double chains of $\mathrm{Cu}_2\mathrm{Cl}_6$. The ground state of TlCuCl_3 was found to be a singlet with an excitation gap $\Delta\simeq 7.5$ K.[5] The gap has been associated with the weak anisotropic antiferromagnetic (AF) intra-dimer coupling in the double chain. Magnetic susceptibility experiments at different temperatures for different directions of the applied magnetic field exhibit broad maxima at $T=38$ K and a decreasing behavior toward zero susceptibility with decreasing temperature.[4] At very low temperatures, the magnetic susceptibility for different values of the applied magnetic field behaves quite differently. At fields of the order of 1 T, the magnetic susceptibility decreases exponentially to zero, a proof for the existence of a ground state gap. On the other hand, for a magnetic field of 7 T an anomaly in the magnetic susceptibility was reported around $T=4$ K. This is an indication that the ground state must be gapless at high magnetic fields. The system has a three dimensional character and in an external magnetic field H, the singlet-triplet gap Δ is reduced to $\Delta-\mu_B gH$, and vanishes at a critical field $H_c=\Delta/\mu_B g$. Inelastic neutron scattering measurements proved the existence of the elementary magnonic excitations with a strong dispersion in all three directions.[6,7] The observed gap, $\Delta=0.7$ meV, which is much smaller than the intradimer interaction $J=5$ meV, and the small critical field $H_c=5.6$ T, make this material a perfect candidate to study the magnetic field induced phase transition. Another argument in favor of the possible phase transition was the evidence for a Goldstone mode[8] observed also by inelastic neutron scattering experiments.[9] Neutron diffraction experiments at fields $H>H_c$ showed that the field induced AF order in the plane normal to the applied field appears at the same time with the uniform moments.[10] Sound attenuation experiments were associated to a "relativistic"-like form of the excitations energy spectrum, $E(k)=\sqrt{\Delta^2+k^2/[2m_{eff}]}$ $(m_{eff}=2/J^2)$, a relation which was used to explain the connection between the transition temperature and the concentration of magnons.[11] The main conclusion of all these experimental approaches is that TlCuCl_3 undergoes a magnetic ordering in high magnetic fields. The temperature dependence of the critical magnetic field can be summarized by $[H_c(T)-H_c(0)]\propto T^{\phi}$, where from the experimental data fit $\phi\simeq 1.7-2.2$.

Various theoretical models were used to investigate the BEC of magnons in quantum spin liquids. In the mean field approximation most of the experimental features of the phase transition cannot be reproduced, in particular, the critical temperature dependence on the magnetic field being almost flat.[12] The possibility of a magnetic field induced phase transition in quantum spin liquids was first discussed in terms of BEC by Giamarchi and Tsvelik.[13] This possibility was investigated using the Hartree-Fock approximation introduced by Popov.[14] The initial system is mapped into a dilute Bose liquid with a magnetic field dependent chemical potential

$\mu = g\mu_B(H - H_c)$, and the total number of magnons N is associated with the total magnetization M, $M = g\mu_B N$.[15] The theory predicts $[H_c(T) - H_c(0)] \sim T^{3/2}$, a result which is not in complete agreement with experimental data. The model was further on investigated by Misguich and Oshikawa[16] by including a more realistic dispersion relation[6] and the shape of the critical temperature curves as function of the critical magnetic field was well reproduced with a single adjustable parameter.

The possible phase transition was numerically investigated by Monte Carlo simulation.[17–19] Nohadani *et al.*[17] found that the value of the critical temperature exponent ϕ is in general greater that the Hartree-Fock value $\phi = 1.5$, but is close to it as the magnetic field approaches its critical zero temperature value. Kawashima[18] argued that the Hartree-Fock value $\phi = 1.5$ may be incorrect at finite temperature, but is the correct value in the $T = 0$ K limit, namely for the situation when the phase transition is actually a quantum phase transition (QPT). The BEC of magnons as a QPT at $T = 0$ K was further investigated by Monte Carlo simulation by Nohadani *et al.*[19] The model considered a control parameter $g = J'/J$, where J is the intradimer exchange interaction, and J' is the inter-dimer coupling. The experimental data gives for these couplings the values $J = 60 - 70$ K and $J' = 40 - 53$ K. However, those values can be controlled by external pressure. The resulting $T = 0$ K phase diagram consists of three different regions, regulated by the g ratio. At low magnetic fields and small control parameter g the system is a dimer spin liquid, i.e., is in a magnetically disordered state. This phase is characterized by a $SU(2)$ symmetry with a dynamical critical exponent $z = 1$. At intermediate magnetic fields $H < H_s$ (H_s is the saturation field) and sufficiently large value of g, the ground state is partially spin polarized for $H > H_c$ (H_c is the critical field) and has a long-range antiferromagnetic order transverse to the applied magnetic field direction. In this case the phase symmetry is only $U(1)$ with a corresponding dynamical critical exponent $z = 2$, a value associated to the quadratic dispersion of the bosonic excitations. At large fields $H > H_s$ ($H_s \sim J + 5J'$), all spins are fully polarized.

Here we consider a renormalization group (RG) approach of the QPT associated to the BEC of magnons in spin dimer systems. It is well known that the presence of a QPT will influence the system's properties even at finite temperatures. The exact temperature range in the vicinity of the $T = 0$ K phase transition is unknown, however, as in any phase transition a critical region will be present. We will consider separately the phases identified by Monte Carlo simulation, and show that different values for the critical temperature exponent ϕ are obtained in the RG analysis. We will show that in the phase with a $SU(2)$ symmetry $\phi = 2$, whereas for the phase with $U(1)$ symmetry $\phi = 1$. If one considers a more realistic model with a $U(1)$ symmetry and strong anisotropy, $\phi = 3/2$. This behavior suggests the existence of a narrow crossover temperature interval close to $T = 0$ K ($T \ll 1$ K) where the critical temperature coefficient complies with the Hartree-Fock value $\phi = 3/2$. At higher temperature, still in the critical region, the symmetry of the system changes and the value of the temperature critical exponent is higher $\phi \simeq 2$. We will discuss those results in connection with the numerical calculations and experimental data.

II. RENORMALIZATION-GROUP ANALYSIS

Following Hertz[20] and Millis,[21] we describe the critical behavior of a magnon dilute gas in terms of an effective Ginzburg-Landau-Wilson theory of the order parameter field $\Phi(\mathbf{k}, \tau)$ which represents the fluctuations of the staggered magnetization of the system.

In the general theory of QPT's the total action will consist on two terms, $S[\Phi] = S_2[\Phi] + S_4[\Phi]$. The quadratic part, $S_2[\Phi]$, takes the form

$$S_2[\Phi] = \frac{1}{2}\sum_k \Phi^\dagger(k)[r_0(H) + k^\sigma + (i\omega_n)^m]\Phi(k), \qquad (1)$$

where $k \equiv (\mathbf{k}, \omega_n)$ with $\omega_n = 2n\pi T$ ($k_B = 1$), $\Phi(k)$ is a bosonic field describing the magnetization fluctuations, and $r_0(H)$ measures the distance from the quantum critical point. In the presence of an external magnetic field the control parameter $r_0(H)$ will acquire a field dependence. The form of the action was extensively discussed by Fisher and Rosch[22] both on a phenomenological basis and starting from a Hubbard-type model of electrons moving in the presence of a magnetic field. In the following we will consider to have a Zeeman type H dependence of the control parameter, $r_0(H) \sim [H - H_c]$. The dimensionality of the system is $d = 3$, and σ and m will take different values according to the studied phase, i.e., $\sigma = 2$ and $m = 2$ meaning that the dynamical critical exponent $z = 1$ for the $SU(2)$ symmetry, and $\sigma = 2$ and $m = 1$ with a dynamical critical exponent $z = 2$ for the $U(1)$ symmetry. The interacting contribution to the action, $S_4[\Phi]$, will be of standard form

$$S_4[\Phi] = \frac{u_0}{16}\sum_{k_1,\ldots,k_4} \Phi(k_1)\Phi(k_2)\Phi^\dagger(k_3)\Phi^\dagger(k_4) \times \delta(k_1 + k_2 + k_3 + k_4), \qquad (2)$$

and describes interactions between the considered fluctuations. The total action will remain invariant under a standard transformation of momenta, frequency, and fields, i.e., $k' = kb$, $\omega_n' = \omega_n b^z$, where z is the dynamical critical exponent, and $\Phi' = \Phi b^{-(d+z+2)/2}$.[23]

The scaling equations for the parameters T, r, and u have the general form:[24,26]

$$\frac{dT(l)}{dl} = zT(l), \qquad (3)$$

$$\frac{dr(l)}{dl} = 2r(l) + K_3 F_1[r(l), T(l)], \qquad (4)$$

$$\frac{du}{dl} = [4 - (d + z)]u(l) - \frac{5}{2}K_3 F_2[r(l), T(l)]u^2(l), \qquad (5)$$

where $F_1[r(l), T(l)]$ and $F_2[r(l), T(l)]$ are complicated functions of parameters T and r. Their values in the low tempera-

ture domain and at the critical point ($r=0$) are given in Ref. 24 as $F_1[T(l)]=\coth[1/(2T(l))]/2$ and $F_2[T(l)]=1/4$. Also, K_3 is a constant whose value is $1/(2\pi^2)$. Equations (3)–(5) will be solved to analyse the system behavior in the critical region for two situations. First we will analyze the $SU(2)$ symmetry situation when the dynamical critical exponent $z=1$, and then the $U(1)$ symmetry case when $z=2$.

A. SU(2) symmetry

One possible phase identified by Monte Carlo simulations was characterized by the limit of low magnetic fields and small coupling ratios J'/J, being a magnetically disorder phase, i.e., a dimer spin liquid. In this case the dynamical critical coefficient value is $z=1$. Two of the renormalization group equations, namely (3) and (5), can be solved exactly with the following solutions

$$T(l) = Te^{l} \tag{6}$$

and

$$u(l) = \frac{1}{C_0(l+l_0)}, \tag{7}$$

where $C_0=5K_3/8$ and $l_0=1/[C_0u_0]$. The remaining equation (4), the one for the control parameter r, will be solved considering a solution of the form $r(l)\simeq r_0\exp(2l)h(l)$, leading to

$$r(l) = e^{2l}\left[r_0 + \frac{K_3}{4}u_0 + K_3 I_r^{(3)}(l) \right] - \frac{K_3 u(l)}{4}, \tag{8}$$

where

$$I_r^{(3)}(l) = \int_0^l \frac{dx\exp[-2x]u(x)}{\exp[1/T(x)]-1}. \tag{9}$$

To study the influence of the criticality on the thermodynamics of the $SU(2)$ phase we introduce the scaling field $t_r(l)$ defined as

$$t_r(l) = r(l) + \frac{K_3}{4}u(l), \tag{10}$$

a field which will be used to stop the renormalization process. Based on Eqs. (7) and (8) we get

$$t_r(l) = e^{2l}[t_r(0) + K_3\zeta(2)u_0T^2], \tag{11}$$

where $t_r(0)=r_0-r_{0c}$ with $r_{0c}=-K_3u_0/4$, and $\zeta(x)$ is the Riemann zeta function. The renormalization procedure will be stopped at $l=l^*\gg 1$, given by $t_r(l^\star)=1$. To find l^* we rewrite $t_r(l)=e^{2l}t_r(T)$ and after simple calculations one finds

$$l^\star \sim \ln\frac{1}{T}. \tag{12}$$

We can define a critical line whose equation is given by $t_r(T)=0$ which permits us to calculate the critical field $H_c(T)$ as

$$H_c(T) = H_c(0) - K_3u_0\zeta(2)T^2. \tag{13}$$

Accordingly, in the $SU(2)$ symmetry phase we can identify the exponent of the critical temperature dependence on the magnetic field to be $\phi=2$, a value which is close to the one reported in the literature.

B. U(1) symmetry

Another possible phase identified by Monte Carlo simulations of the quantum phase transition in spin dimer liquids has $U(1)$ symmetry and is present for intermediate applied magnetic fields and a sufficiently large value of the ratio J'/J. In this case the dynamical critical exponent value is $z=2$, and the renormalization group equations (3)–(5) are similar to those of the weakly interacting Bose gas.[25,26] Equation (3) has a trivial solution of the form

$$T(l) = Te^{2l}. \tag{14}$$

We consider now equation (5) which gives us the renormalization of the interaction parameter. If we introduce $\varepsilon=2-d<0$ $(d=3)$ this equation can be rewritten in the form

$$\frac{du(l)}{dl} = \varepsilon u(l) - \frac{K_3}{4}u^2(l), \tag{15}$$

and admits the following solution

$$u(l) = \frac{u_0\exp[-|\varepsilon|l]}{1+K_3(\exp[-|\varepsilon|l]-1)/4}. \tag{16}$$

One can proceed now to the solution for the renormalized control parameter r. In the $U(1)$ symmetry case this equation will admit the following solution:

$$r(l) = e^{2l}\left\{ r_0 + \frac{K_3u_0}{4} + \frac{K_3Tu(l)}{2}\ln\left[\frac{1}{1-\exp[-1/T(l)]}\right]\right\} - \frac{K_3u(l)}{4}. \tag{17}$$

As in the previous situation we introduce the scaling field $t_r(l)=r(l)+K_3u(l)/4$ and we will stop the renormalization procedure at $t_r(l^*)=1$. With this definition, based on Eq. (17), the general form of the scaling field become

$$t_r(l) = e^{2l}\left\{ t_r(0) + \frac{K_3u(l)}{2}T\ln\left[\frac{1}{1-\exp[-1/T(l)]}\right]\right\}, \tag{18}$$

where $t_r(0)=r_0-r_{0c}$ with $r_{0c}=-K_3u_0/4$. We stop the scaling procedure at $l=l^*\gg 1$, where once again l^* is the solution of $t_r(l^\star)=1$. In this case the equation is more complicated, however, in the low temperature regime we can approximate its solution. If we rewrite $t_r(l)=e^{2l}t_r(T)$ with

$$t_r(T) \simeq r_0 - r_{0c} + \frac{K_3T}{2}u_0\ln\frac{1}{u_0}, \tag{19}$$

we finally find

$$l^* \sim \frac{1}{2} \ln \frac{1}{T}. \tag{20}$$

The critical line, identified from $t_r(T)=0$, will give us the critical magnetic field $H_c(T)$ as a function of temperature

$$H_c(T) = H_c(0) - \left[\frac{u_0}{4\pi^2} \ln \frac{1}{u_0} \right] T. \tag{21}$$

In the case of $U(1)$ symmetry the relation between the critical field and temperature is different from the case of $SU(2)$ symmetry and we identified in this situation the critical temperature exponent to be $\phi=1$.

III. INFLUENCE OF ANISOTROPY

According to Monte Carlo calculations at sufficiently large fields the system has a ground state which is partially spin polarized and has antiferromagnetic long-range order transverse to the magnetic field direction. The fact that the magnetic properties of the system depend on direction suggests that anisotropy may play an important role when the system is in this partially polarized phase. To take in account such effects, we consider that the interaction term in the total action corresponding to the $U(1)$ symmetry phase has the following form:[18,19]

$$S_4[\Phi] = \frac{1}{16} \sum_{k_1,\ldots,k_4} \Phi(k_1)\Phi(k_2)\Phi^\dagger(k_3)\Phi^\dagger(k_4)\delta_{\mathbf{k}_1+\mathbf{k}_2;\mathbf{k}_3+\mathbf{k}_4} \times [u_0 \delta_{\omega_{n1}+\omega_{n2};\omega_{n3}+\omega_{n4}} + v_0 \delta_{\omega_{n1};\omega_{n3}} \delta_{\omega_{n2};\omega_{n4}}], \tag{22}$$

where the coupling constants u_0 and v_0 describe interactions between the magnetization fluctuations in different directions of the system. Using the same procedure as in the previous cases the renormalization group equations can be obtained as

$$\frac{dT(l)}{dl} = 2T(l), \tag{23}$$

$$\frac{dr(l)}{dl} = 2r(l) + \frac{K_3}{2}\{[v(l)+2u(l)]F_1[T(l)] + v(l)T(l)\}, \tag{24}$$

$$\frac{du(l)}{dl} \simeq \varepsilon u(l) - \frac{K_3}{4} u^2(l), \tag{25}$$

$$\frac{dv(l)}{dl} \simeq \varepsilon v(l). \tag{26}$$

The last two equations, the ones corresponding to the renormalized coupling constants $u(l)$ and $v(l)$, have been written in the lowest order, an approximation which is assumed valid in the low temperature domain. Equation (23) admits the trivial solution $T(l)=Te^{2l}$. The first coupling constant, $u(l)$, has the same form no matter if one considers or not the anisotropy, given by Eq. (16). Equation (26) admits the trivial solution $v(l)=v_0 e^{-|\varepsilon|l}$. The solution for the remaining equation which is giving us the renormalized phase transition control parameter, r, can be written as

$$r(l) = e^{2l}\left\{ r_0 - r_{0c} + \frac{K_3}{2}\int_0^l dl' v(l')T(l') + \frac{K_3}{2}\int_0^l dl'[v(l')+2u(l')]F_1[T(l')] \right\}, \tag{27}$$

where $r_{0c}=K_3[v_0+2u_0]/8$. We can introduce again the scaling field $t_r(l)=r(l)+K_3[v(l)+2u(l)]/8$ which based on the solution for r can be written as

$$t_r(l) = e^{2l}\left\{ r_0 - r_{0c} + \frac{K_3 v_0}{2\varepsilon} T[e^{-|\varepsilon|l} - 1] + \frac{K_3}{4} T[v(l)+2u(l)] \ln\left[\frac{1}{1-\exp[-1/T(l)]} \right] \right\}, \tag{28}$$

where $\varepsilon<0$ for $d=3$. To calculate the value of the stop scaling parameter l^* we consider $t_r(l^\star)=1$ and, in the lowest order, we obtain $l^\star$ as

$$l^\star \sim \frac{1}{2} \ln \frac{1}{T}. \tag{29}$$

Equation (28) for the scaling field can be rewritten in the form $t_r(l)=e^{2l}t_r(T)$ where $t_r(T)$ can be evaluated as

$$t_r(T) = r_0 - r_{0c} + \frac{K_3 v_0 T}{2|\varepsilon|} + \frac{K_3}{4}(v_0+2u_0)T^{3/2} \ln\left[\frac{1}{v_0+2u_0} \right]. \tag{30}$$

In the anisotropic $U(1)$ case the temperature dependence of the critical magnetic field will be calculated in the limit $u_0 > v_0$ from $t_r(T)=0$ as

$$H_c(T) \simeq H_c(0) - \left[\frac{u_0}{2\pi^2} \ln \frac{1}{2u_0} \right] T^{3/2}. \tag{31}$$

A similar result was predicted using different calculation methods and numerical evaluation in Refs. 18 and 19. Equation (31) predicts an important role for the anisotropy in the case of $U(1)$ symmetry, as the critical temperature exponent changes from $\phi=1$ to $\phi=3/2$. The later value is close to the lowest observed experimental value, i.e., 1.7, and identical to the value reported by Monte Carlo studies.

IV. DISCUSSIONS

The idea of BEC in solid state systems was associated with electronic Cooper pairs in superconductors, excitons in semiconductors, and more recently with magnons in spin liquid dimer compounds. The occurrence of a condensate phase in these compounds was investigated by various theoretical methods, from mean field to renormalization group approximations. In the case of spin liquid dimer compounds the BEC of magnons is induced by magnetic fields, the critical field associated to the BEC being temperature dependent and characterized by a critical exponent ϕ, defined as $[H_c(T) - H_c(0)] \sim T^\phi$. The exact value of the critical exponent ϕ can vary from 1.7 to 2.2 according to experimental data. A mean

field analysis gives $\phi=1.5$, and for some time it was believed that such approximation was enough to explain all relevant physics of the phase transition.

Here we applied the renormalization group method to study the possible BEC of magnons. We started our analysis from the premise that a quantum phase transition, despite being characterized by a $T=0$ K critical temperature, will influence the system properties even at finite temperatures. Accordingly, we analyze the phase transition in the vicinity of a quantum phase transition, in the low temperature limit. The possible quantum phase transition in a spin liquid dimer was investigated by Monte Carlo simulations considering a model which includes also the system's anisotropy.[19] As function of anisotropy the phase diagram of a spin liquid dimer will consists of three different regions, in each of them the system being in a different symmetry class.[19] At low magnetic fields, the system is in a magnetically disordered state. As the fields increases, a partially spin polarized state will develop and the symmetry of the system will change accordingly. At high magnetic fields the system is in a long-range order antiferromagnetic phase. To take into account all these possibilities, we analyzed the influence of magnetic fields and temperature on the BEC of magnons in systems with different symmetry. In the case of a magnetically disordered spin liquid dimer, when the symmetry of the system is in the $SU(2)$ class and the dynamical critical exponent is $z=1$, the critical exponent is $\phi=2$. On the other hand, when the partially ordered phase is induced by the magnetic field, the system's symmetry changes to $U(1)$, and the dynamical critical exponent becomes $z=2$. In this situation, we calculated $\phi=1$, a value which was never observed in real experiments. However, in this partially ordered state anisotropy plays an important role, as the phase is characterized by a large J'/J ratio. When anisotropy is taken into account by considering direction dependent interactions in the action, the value of the critical coefficient ϕ changes from 1 to 1.5, a value which is close to the lowest reported experimental data. The influence of anisotropy was considered by Fischer and Rosh[22] including additional terms in the free action, the final result for the temperature critical exponent being $\phi=1.5$, the same value we obtained. The crossover problem was also considered in a sigma model framework to explain some phases of the organic insulator $(TMTTF)_2PF_6$ as function of temperature, magnetic field, and pressure.[27,28] A similar idea was used to explain nuclear magnetic resonance experimental data in superconducting spin-ladder compounds such as $Sr_2Ca_{12}Cu_{24}O_{41}$.[29]

In conclusion, we showed that the renormalization group analysis of the possible BEC of magnons in spin liquid dimers is a more appropriate investigation method. This method can account for the change in the critical exponent ϕ from 1.5 to 2, according to the phase symmetry. The results are in good agreement with the experimental data in $TlCuCl_3$ samples. More recently a similar behavior was reported also in Cs_2CuCl_4 samples.[30] Such a change in the critical exponent cannot be explained by mean field approximations. On the other hand the ground state of $TlCuCl_3$ is strongly influenced by pressure.[31] Accurate experimental data obtained for various values of the applied external pressure still support previous results for the critical exponent, namely $\phi=2.6$.[31] Finally, we mention that recent experimental data showed that the occurrence of the BEC of magnons can be also induced by hydrostatic pressure.[32] The dependence of the critical field on pressure, and the pressure dependence of the transition temperature are suggesting a similar theoretical description.

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