Relaxation of spin waves in antiferromagnets with a strong hyperfine interaction

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An experimental study is made of the parametric excitation and relaxation of electron spin waves (ESWs) of frequency $\nu_{\mathbf{k}} \approx 15$ GHz in antiferromagnetic $CsMnCl_3$ and $RbMnCl_3$ (in the spin-flop phase) at $T = 1.2$–4.2 K. It is established that at low temperatures the relaxation parameter is determined mainly by the term $\Delta\nu_{mn}$, which is independent of temperature and proportional to the length of the ESW wave vector. By processing previous experimental results for $MnCO_3$ and $CsMnF_3$ we have also isolated an analogous contribution to the ESW damping in those antiferromagnets. Within the experimental accuracy the value and functional form of $\Delta\nu_{mn}$ in all four of these materials correspond to the theoretically calculated relaxation parameter[15] due to the scattering of ESWs on the paramagnetic system of ^{55}Mn nuclei. The existence of this relaxation mechanism is thus confirmed experimentally for antiferromagnets having a strong hyperfine interaction. For $RbMnCl_3$ we also determine the damping due to the magnon-phonon interaction. The value of this damping is then used to estimate the corresponding magnetoelastic constant: $\Theta \approx 10$ K.

As we know, at low temperatures the magnetic properties of a material can be affected strongly by the system of nuclear spins. The hyperfine interaction between the electronic and nuclear spins gives rise to an appreciable polarization of the nuclear subsystem, amounting, at helium temperatures (for ^{55}Mn nuclei), to

$$\langle I\rangle/I \approx 10^{-2},$$

where I is the spin of a nucleus. In addition, the hyperfine and exchange interactions of the electron gives rise to an indirect coupling between nuclear spins, which causes their motion to be correlated. If the electronic spin-wave (ESW) spectrum has a low-frequency branch there will be substantial mixing of the oscillations of the electronic and nuclear spins. In this case one must consider the problem of coupled oscillations of the nuclear and electronic spins.

The spectra of coupled electronic-nuclear oscillations were first calculated by de Gennes *et al.*[1] They showed that the interaction of the electronic and nuclear spin subsystems produces an additional gap in the ESW spectrum and a dynamic shift of the NMR frequency, giving rise to dispersion in the nuclear subsystem, i.e., to so-called nuclear spin waves (NSWs). The hyperfine interaction is most strongly manifested in weakly anisotropic antiferromagnets having two characteristics: first, the presence of a low-activation ESW branch and, second, exchange enhancement of the hyperfine interaction.[2] The influence of the hyperfine interaction on the ESW spectrum was first detected experimentally in the antiferromagnet $KMnF_3$ and has subsequently been studied in detail in many antiferromagnets.

The most important characteristic of the spins waves (magnons) is their lifetime, which is determined by the efficiency of the interaction of the spin waves with one another, with phonons, defects, and impurities. There have been many experimental papers on the study of the interaction of ESWs with phonons, impurities, and electronic and nuclear spin waves (see, e.g., Refs. 4–9). The present paper deals mainly with the study of the influence of the hyperfine interaction on the relaxation properties of electronic magnons. As we have already mentioned, at helium temperatures the polarization of the nuclear subsystem is still relatively small ($\approx 1\%$). This makes for two different types of interaction of the magnons with the nuclear subsystem: first, the interaction of the spin waves with collective oscillations of the nuclear spins, i.e., with the NSW; second, the interaction with paramagnetic nuclear spins, for which all directions in the crystal have almost equal probability. It has been shown[10] that the interaction with the nuclear magnons does not contribute substantially to the ESW relaxation.

The second type of interaction of magnons with magnetic nuclei was first considered theoretically by Richards[11] for nuclear magnons with frequencies $\omega_{\mathbf{k}}$ close to the unshifted NMR frequency ω_n, i.e., for $(\omega_n - \omega_{\mathbf{k}}) \ll \omega_n$. It was found that at sufficiently low temperatures the NSW relaxation is governed mainly by elastic scattering on fluctuations in the nuclear magnetization.

This NSW relaxation process was first isolated experimentally by Govorkov and Tulin in $MnCO_3$ and has since been studied in detail in three antiferromagnets having easy-plane anisotropy: in $MnCO_3$,[12] $CsMnF_3$[13] and $CsMnCl_3$.[14] Richards' theory, generalized to the case of arbitrary $\omega_{\mathbf{k}}$,[13] has permitted description of all the experimental results in terms of absolute value and in terms of the dependence of the relaxation parameter on $\omega_{\mathbf{k}}$, the temperature, and the length of the NSW wave vector $\mathbf{k}$.

The analogous relaxation process for ESWs was first considered by Woolsey and White.[15] Their expression for the rate of relaxation of ESWs at fluctuations of the nuclear magnetization in cubic antiferromagnets is of the form (an analogous expression has also been obtained in a more recent paper[16])

$$\Delta\nu_{mn} = \frac{\sqrt{3}}{\pi}\frac{A^2 I(I+1)\alpha}{\hbar^2 \nu_{\mathbf{k}} \gamma H_E} k. \tag{1}$$

Here A is the hyperfine interaction constant, H_E is the exchange field, $\nu_{\mathbf{k}} = \omega_{\mathbf{k}}/2\pi$, $\gamma = 2.8$ GHz/kOe is the magne-

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tomechanical ratio for the electron, and α is the inhomogeneous exchange interaction constant, which determines the limiting ESW velocity $v_s = 2\pi\alpha\gamma$. This relaxation process occurs most effectively at magnetic ions with a strong hyperfine interaction. If part of the isotopes of this ion have nonmagnetic nuclei, then they will not contribute to $\Delta\nu_{mn}$. Thus convenient objects for experimental study of ESW scattering on the nuclear subsystem are antiferromagnets containing the ion Mn^{2+}, which has a strong hyperfine interaction, a nuclear spin $I = 5/2$, and an isotope content of 100% ^{55}Mn. Such materials include, first of all, the aforementioned easy-plane antiferromagnets and also crystals of the biaxial antiferromagnet $RbMnCl_3$, in which the relaxation of ESWs and NSWs has not been studied. As none of these materials are cubic antiferromagnets, Eq. (1) can be regarded as an approximate formula for estimating $\Delta\nu_{mn}$ in these materials and for checking the functional dependences of $\Delta\nu_{mn}$.

According to (1), $\Delta\nu_{mn}$ does not depend on the temperature, and therefore the influence of the hyperfine interaction on the ESW damping should be studied at temperatures low enough that the other relaxation processes become inefficient. In $MnCO_3$ and $CsMnF_3$ the main contribution to the ESW relaxation at $T > 1.5$ K is a three-magnon process in which the parametric magnon of the quasiferromagnetic branch and another quasiferromagnetic magnon coalesce into a magnon of the quasi-antiferromagnetic branch. The efficiency of this process decreases as the gap ν_{20} in the spectrum of the quasi-antiferromagnetic branch increases. Therefore, more convenient objects in which to study $\Delta\nu_{mn}$ at helium temperatures are antiferromagnets with rather large values of ν_{20}, such as $CsMnCl_3$ and $RbMnCl_3$.

SAMPLES AND MEASUREMENT TECHNIQUES

The main experiments in the present study were done on samples of $RbMnCl_3$. At room temperature $RbMnCl_3$ is a hexagonal crystal (D^4_{6h}).[17] The magnetic ordering occurs at $T_N = 92$ K. The antiferromagnetic resonance in $RbMnCl_3$ has been studied in detail by the present authors.[18] Three absorption lines corresponding to the excitation of oscillation of the quasiferromagnetic branch were observed. When the magnetic field was rotated in the basis plane a strong anisotropy of the position of the absorption line was observed; this anisotropy is described by the empirical formula

$$(\nu_i/\gamma)^2 = H^2 - H_\varphi^2 \cos 2\varphi_i + H_\Delta^2. \quad (2)$$

Here ν_i is the antiferromagnetic resonance frequency, H is the external magnetic field, $H_\varphi = 6.6$ kOe is the spin-flop field, $H_\Delta^2 = (14 \pm 1)/T$ kOe^2 is the hyperfine-interaction term in the spin-wave spectrum, and φ_i is the angle between $\mathbf{H}$ and the ith binary axis. To explain the antiferromagnetic resonance spectrum it was assumed in Ref. 18 that a structural transition is present, resulting in the formation of three types of structural domains, each of which is a biaxial antiferromagnet. The hard-magnetizations axes of these domains coincide, while the easy-magnetization axes (EA) are rotated 120° with respect to one another. Studies of the effect of a directional compression on the line intensities have confirmed this hypothesis; in particular, a single-domain sample of $RbMnCl_3$ was created at $T = 4.2$ K. The structural transition in $RbMnCl_3$ at $T = 272$ K has since been studied in detail by optical methods.[19]

An interesting result was obtained by Mil'ner and Popkov.[20] They found that in a stress-free sample the rotation of the antiferromagnetism vector $\mathbf{l}$ in an external field $H > H_\varphi$ causes the boundaries of the structural domains to move; as a result, a single-domain sample of $RbMnCl_3$ can be obtained without the aid of uniaxial compression. This result indicates that the magnetoelastic interaction Θ in the $RbMnCl_3$ crystal is large.

Part of the experiments were done on $CsMnCl_3$ samples. The antiferromagnetic resonance and parametric excitation of ESWs in $CsMnCl_3$ were first observed by the present authors in a previous study.[6]

In discussing the results we shall also make use of the data on the ESW relaxation in $CsMnF_3$ and $MnCO_3$. These materials, like $CsMnCl_3$, are easy-plane antiferromagnets characterized by a very weak magnetic anisotropy in the basal plane.

Experiments on the parametric excitation of ESWs were done in the straight-amplifications microwave spectrometer described in Ref. 4. The sample was placed at an antinode of the microwave magnetic field h in a rectangular resonator with $Q \approx 1000$. The onset of parametric excitations was detected by the appearance of a "chip" in the microwave pulse after it had passed through the resonator. Since we were unable to detect a chip in the pulse in the case of the single-domain sample, we performed the experiment on single-domain samples of $RbMnCl_3$. As we have mentioned, a single-domain sample can be obtained by applying just an external field $H > H_\varphi$ to an unclamped crystal. In this case, however, parametric excitation for $\mathbf{H}\|\mathrm{EA}$ cannot be realized in the spin-flop phase, because after the spin flop the boundaries of the structural domains begin to move and the easy axis rotates 60° with respect to $\mathbf{H}$.[20] We, however, were mainly interested in the possibility of parametric excitation of spin waves in the spin-flop phase, since in the work we are aware of on the parametric excitation of spin waves in the easy-axis antiferromagnet MnF_2 (Ref. 21) and biaxial antiferromagnet $CuCl_2 \cdot 2H_2O$ (Ref. 22), ESWs were successfully excited only up to the spin flop. In addition, for $\mathbf{H}\|\mathrm{EA}$ one can do the experiments at lower frequencies and study a wider range of wave vectors $\mathbf{k}$. Therefore, the experiment was done as follows. The $RbMnCl_3$ sample was cooled under a pressure $\simeq 100$ bar from room temperature to helium temperature. Although the sample did not go completely into a single domain, there was nevertheless a significant region of the crystal with $\mathrm{EA}\|\mathbf{H}\|\mathbf{h}$, where we were able to excite spin waves. The external pressure produced stresses in the sample which prevented the domain boundaries from moving.

The parametric excitation of spin waves was accomplished at pump frequencies $\nu_p \approx 36$ and 23 GHz at temperatures in the range $T = 1.2$–4.2 K. The compression apparatus strongly distorted the distribution of the microwave fields in the resonator and made it hard to calculate h; to determine h we therefore made a calibration based on the threshold field h_c in $CsMnCl_3$. The absolute value of the

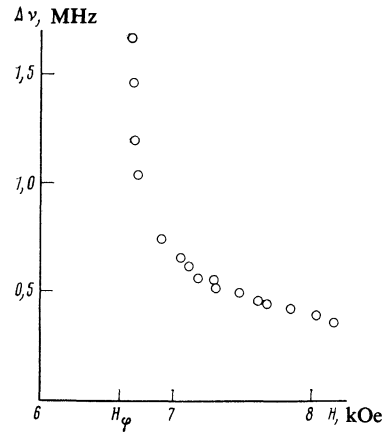

FIG. 1. Relaxation parameter of ESW versus the magnetic field in the spin-flop phase of $RbMnCl_3$, $\nu_p = 36$ GHz.

threshold field h_c in $RbMnCl_3$ was measured to an accuracy of 50 %, and that in $CsMnCl_3$ was measured to 25 %. The relative error in a single series of measurements was 10 % and 5 %, respectively.

The relaxation parameter $\Delta\nu$ of the excited spin waves was calculated from the threshold field h_c according to Ozhogin's formula[23]:

$$\Delta\nu = 2H\gamma^2 h_c/\nu_p. \tag{3}$$

The contribution to $\Delta\nu$ from all the spin-wave relaxation processes (including elastic) were assumed to be additive, since in elastic scattering the packet of parametrically excited spin waves smears, resulting in the same "loss of energy" from a system of spin waves coupled to a pump as in the cases of inelastic scattering. The validity of this approach is also suggested by all the results of Refs. 12–14, in which studies were made of the elastic scattering of nuclear magnons.

EXPERIMENTAL RESULTS AND DISCUSSION OF THE MECHANISM FOR RELAXATION OF ELECTRONIC SPIN WAVES

The characteristic field dependence $\Delta\nu(H)$ for $RbMnCl_3$ is shown in Fig. 1. At all temperatures one observes a rapid growth of h_c near the spin-flop field H_φ corresponding to a growth of the spin-wave relaxation rate in this region of H. Such a behavior of $\Delta\nu(H)$ can be attributed to the existence of an intermediate state in the spin-flop region. An estimate of the width of the intermediate-state region according to the formulas of Ref. 24 gives $\delta H_{\text{theor}} = 30$ Oe in our case, whereas experimentally the rapid growth of the parallel-pumping threshold is observed for $\delta H_{\text{exp}} \approx 300$ Oe. Such a discrepancy with the theory can be blamed on the following circumstances: 1) the sample contains inhomogeneous stresses, which can affect the value of H_φ and smear out the transition to the spin-flop phase; 2) the deviation of **H** from the EA causes a continuous rotation of **l**. The width of the region in which there is a transition to the spin-flop phase has been obtained from experimental measurements[25] of the magnetization of $RbMnCl_3$: the value found, $\delta H = 400$ Oe, is close to δH_{exp}.

Figure 2 shows the characteristic curves of $\Delta\nu$ as a function of the wave vector **k**. The product αk has been calculated from the ESW spectrum. The expression for the spectrum in this case can be obtained from (2) with φ_i set equal to zero and with the usual term $\alpha^2 k^2$ added to the right-hand side. Measurements of $\Delta\nu(\alpha k)$ at different temperatures give a set of parallel straight lines described by the empirical formula

$$\Delta\nu[\text{MHz}] = 0.06T[\text{K}] + 0.1\alpha k[\text{kOe}]. \tag{4}$$

The sharp growth of $\Delta\nu(\alpha k)$ at large k in Fig. 2 corresponds to the peak on the $\Delta\nu(H)$ curve at $H \approx H_\varphi$.

Let us begin with the first term on the right-hand side of Eq. (4). This term describes a contribution to the total spin-wave relaxation that is independent of the wave vector and magnetic field and linearly dependent on temperature (Fig. 3). Behavior of this kind is characteristic of many different magnon-phonon relaxation processes.[26,27]

Depending on the relationship between the spin-wave velocity v_s and the sound velocity u, different spin-wave relaxation processes involving phonons are possible. Since estimates give $v_s > u$ for $RbMnCl_3$, let us consider in detail only this case, having noted that the relaxation parameter $\Delta\nu_{mph}$ depends weakly on the relationship between the velocities of magnons (m) and phonons (ph).

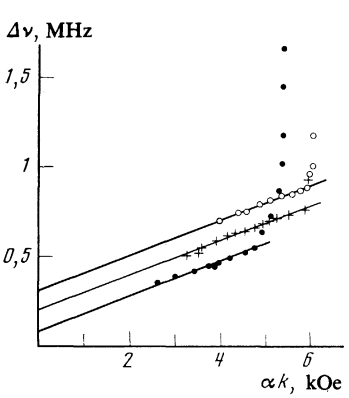

FIG. 2. Relaxation parameter of ESW in the spin-flop phase of $RbMnCl_3$ as a function of αk at various T: ●) 1.2 K; +)3.2 K; ○) 4.2 K; $\nu_p = 36$ GHz.

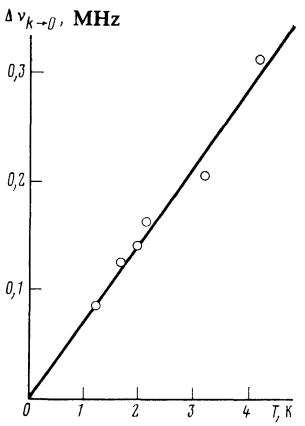

FIG. 3. Temperature dependence of the ESW relaxation parameter is $RbMnCl_3$ at $k \to 0$.

We therefore let $u < v_s$. Then the processes allowed by the conservation laws are $m + ph \to m$, $m \to m + ph$, $m \to ph + ph$, and the total relaxation rate on account of all three of these processes is[28]

$$\Delta\nu_{mph} = \frac{4}{\pi^2 (v_s/u)^2 (2+v_s/u)} \frac{\Theta^2}{Mu^2} \frac{T}{\Theta_N}. \quad (5)$$

Here $\Theta_N = \hbar w_s / k_B V_0^{1/3}$, $V_0 = 2V/N$, where V is the volume of the crystal, N is the number of magnetic ions, $\Theta = V_0 \beta_i$ is the characteristic magnetoelastic interaction energy, β_i is a component of the magnetostriction tensor, $M = \rho V_0$, ρ is the density, and k_B is the Boltzmann constant. Obviously, for an exact calculation of the theoretical value $\Delta\nu_{mph}$ one would need to know the phonon and magnon velocities, but since these velocities have not been measured, we use the rough estimates $v_s = 2 \cdot 10^5$ cm/sec and assume that the speed of sound in $RbMnCl_3$ is equal to that in $CsMnCl_3$: $u = 1.7 \cdot 10^5$ cm/sec.[14] Under these conditions to have agreement between $\Delta\nu_{mph}^{\text{theor}}$ and $\Delta\nu_{mph}^{\text{exp}}$ we must set $\Theta = 10$ K (for comparison, in $MnCO_3$ one has ($\Theta = 3$ K).[29] The value obtained for Θ is rather large, but such a result is not unexpected, since it has been known for some time[20] that $RbMnCl_3$ has a strong magnetoelastic coupling.

Let us now consider the second term in Eq. (4). The linear dependence of the relaxation rate on k and its independence of temperature are characteristic of elastic scattering of spin waves by fluctuations of the magnetization of localized nuclear spins; this process is characterized by the relaxation rate $\Delta\nu_{mn}$ given in (1). An estimate of the relaxation using (1) gives $\Delta\nu_{mn}^{\text{theor}} = 0.03\alpha k$ [kOe] for $RbMnCl_3$, a value which is only one-third of the experimentally measured value. We consider this agreement to be entirely satisfactory. This is because Eq. (1) is written for the case of a cubic antiferromagnet, for which $v_s = H_E a/\sqrt{6}$ (a is the lattice parameter), and in an intermediate step in the calculations Eq. (1) contains a factor $(H_E/v_s)^3$, which is the case of an anisotropic spin-wave velocity changes $\Delta\nu_{mn}^{\text{theor}}$ by a factor of $(v_s^{\text{theor}}/v_s)^3$. In order for the theoretical value $\Delta\nu_{mn}^{\text{theor}}$ to agree exactly with the experimental value, we would have to assume that $v_s = \frac{2}{3} v_s^{\text{theor}} = 2 \cdot 10^5$ cm/sec. It should also be recalled that the absolute uncertainty in the measurements of $\Delta\nu$ in $RbMnCl_3$ was 50%. Thus the above results apparently indicate that theoretical formula (1) is valid and that $\Delta\nu_{mn}$ represents a significant contributions to the total ESW relaxation at helium temperatures.

As our studies have shown, a more convenient object for determining the relaxation parameter $\Delta\nu_{mn}$ is crystalline $CsMnCl_3$, which, like $RbMnCl_3$, has a small three-magnon contribution $\Delta\nu_{3m}$ to the relaxation of ESWs with frequencies $\nu_k \lesssim 15$ GHz at $T = 1.2$–4.2 K, but which would avoid a number of experimental difficulties stemming from the need to "pole" the sample into a single domain. By studying the total relaxation ($\Delta\nu$) in $CsMnCl_3$ at a frequency $\nu_p \approx 23$ GHz, we have discovered that the relaxation parameter $\Delta\nu$ is independent of temperature and linearly dependent on the wave vector (Fig. 4). Measurements of $\Delta\nu_{mn}$ at two pump frequencies revealed the correct (to within the experimental error) behavior of $\Delta\nu(\nu_p)$. Comparison of the theoretical and experimental values of $\Delta\nu_{mn}$ gives

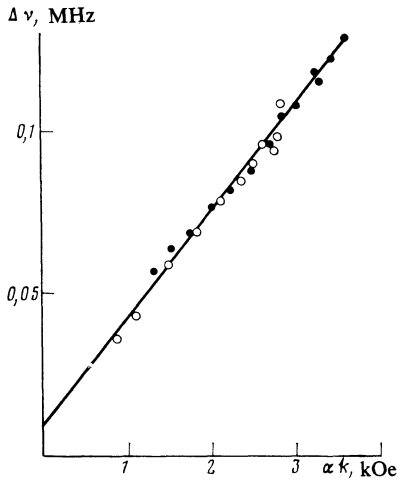

FIG. 4. Relaxation parameter of ESW versus αk in $CsMnCl_3$; $\nu_p = 23.5$; ○) $T = 1.2$ K, ●) $T = 4.2$ K.

$$\Delta\nu_{mn}^{\text{exp}} \ (\nu_p = 23.5\ \text{GHz}) / \Delta\nu_{mn}^{\text{theor}} = 1.2,$$

$$\Delta\nu_{mn}^{\text{exp}} \ (\nu_p = 36\ \text{GHz}) / \Delta\nu_{mn}^{\text{theor}} = 1.1.$$

We carried out an analogous comparison of the ESW relaxation parameter with $\Delta\nu_{mn}^{\text{theor}}$ for the previously studied materials $MnCO_3$ and $CsMnF_3$. The large contributions of three-magnon processes to the total ESW relaxation makes it difficult to study $\Delta\nu_{mn}$ in these materials, but our processing of the results showed that in those materials $\Delta\nu$ is described satisfactorily by the sum $\Delta\nu = \Delta\nu_{3m} + \Delta\nu_{mn}$. In view of that fact that $\Delta\nu_{3m} \propto H^2$, at $H \to 0$ we have $\Delta\nu = \Delta\nu_{mn}$. We have made a comparison of the absolute value of $\Delta\nu_{mn}^{\text{theor}}$ with the relaxation parameters $\Delta\nu(H = 0)$ obtained in experiments on the damping time of beats in a system of parametric ESWs[7] and on the effect of a low-frequency ($\nu_m \approx 10^6$ Hz) alternating field on the parallel-pumping threshold.[9] These mesurements evidently give greater absolute accuracy for $\Delta\nu$, since they do not involve calculating the microwave field in the resonator. The comparison gave (for $\nu_p \approx 36$ GHz)

$$\Delta\nu_{mn}^{\text{exp}} \ (H \approx 0) / \Delta\nu_{mn}^{\text{theor}} = 1.0$$

for $MnCO_3$, and

$$\Delta\nu_{mn}^{\text{exp}} \ (H \approx 0) / \Delta\nu_{mn}^{\text{theor}} = 1.2$$

for $CsMnF_3$.

The good agreement between theory and experiment in four antiferromagnets suggests that the observed contribution $\Delta\nu \propto k$ to the relaxation is due mainly to the scattering of ESWs on paramagnetic ^{55}Mn nuclei. Defects in the sample can make an additional contribution,

$$\Delta\nu_{\text{def}} = \frac{v}{L} = \frac{2\pi\alpha^2\gamma^2}{L\nu_k} k,$$

to the elastic scattering of ESWs, where L is the mean free path of the ESWs between defects. Our analysis implies, however, that the elastic scattering of ESWs is governed mainly by $\Delta\nu_{mn}$ ($\Delta\nu_{\text{def}} \ll \Delta\nu_{mn}$) except, perhaps, in the $RbMnCl_3$ samples that had been subjected to compression. An unambiguous answer as to the role of defects in the spin-

wave damping will require measurements on samples with a controlled number of defects.

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