Journal of Alloys and Compounds 488 (2009) 23–26

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Extension of the molecular-field theory on the magnetic behaviors in paramagnetic $Dy_3Ga_5O_{12}$

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ARTICLE INFO

Article history:
Received 28 July 2009
Received in revised form 16 August 2009
Accepted 17 August 2009
Available online 31 August 2009

Keywords:
Molecular-field theory
Exchange interaction
Magnetic properties

ABSTRACT

The magnetic behaviors of the paramagnetic dysprosium gallium garnet (DyGaG) are theoretically investigated in terms of an extensional molecular-field theory. The contribution of the exchange interaction between the dysprosium ions to the magnetic properties of DyGaG is further explored. Here, as to DyGaG, in our model, two magnetic sublattices A and B are suggested. The corresponding magnetic properties of the two magnetic sublattices are discussed, and it is found that $\chi_A > \chi_B$ in DyGaG. Meanwhile, the exchange field coefficients λ_1 and λ_2 are qualitatively analyzed. The optimum fitting parameters α_A and α_B associated with the exchange interaction are provided, and some interesting conclusions about these parameters are drawn. Then, the available experiments are successfully fitted by our theoretical model. Additionally, an analysis of the high-field magnetic properties in DyGaG is presented.

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1. Introduction

Owing to the wide applications in optical, electronic, and magnetic materials and devices, the magnetic properties of rare-earth gallium garnets, especially the heavy rare-earth gallium garnets, have been intensively discussed during the past several decades [1–4]. Many experiments, mainly focused on the studies of susceptibility, magnetic moment, magnetic ordering and specific heat etc., in these garnets have been carried out, and some experimental phenomena are theoretically revealed by the analyses of the crystal field effect [5–8]. However, it is worth pointing out that some experiments cannot be perfectly explained by only considering the crystal field [9].

Recently, we suggest that the exchange interaction between rare-earth ions should be taken into account in the calculations of the magnetic properties in some paramagnetic rare-earth gallium garnets by the quantum theory, especially in high-magnetic fields [2,10,11], although it is much smaller than rare-earth–iron exchange interaction in rare-earth iron garnet [12]. Moreover, it is noted that, in the analyses of the magnetic properties in rare-earth aluminum garnets, a long-range interaction named as dipole–dipole interaction should be rigorously taken into account [13]. Therefore, in order to further confirm the importance of the exchange interaction in gallium or aluminum garnets, the magnetic behaviors of the paramagnetic dysprosium gallium garnet (DyGaG) are analyzed in terms of a semi-classical phenomenological theory.

On the other hand, we also notice that, by taking the exchange interaction into consideration, Capel [14] reckoned the magnetic ordering temperatures in some rare-earth gallium and aluminum garnets at low temperatures, moreover, it was predicted that DyGaG should be ferrimagnetic below the transition temperature 0.7–0.77 K or 1.54–2.15 K. Whereas, Cooke et al. [15] and Filippi et al. [4] experimentally pointed out that DyGaG should display antiferromagnetic characteristic by magnetic susceptibility and specific heat measurements, and the Néel temperature is about 0.37 K. Additionally, experimental results of DyGaG in Ref. [9] presented that an obvious anisotropy of the magnetization was exhibited with the increase of the external magnetic fields along [0 0 1], [0 1 1] and [1 1 1] directions at 4.23 K, and a nonlinearity of the magnetization with the external magnetic fields at 1.15–4.23 K was shown when the external magnetic field was about 0–5.5 T.

Then, in this paper, on the basis of antiferromagnetic molecular-field approximation, the above experimental phenomena on the magnetic properties in paramagnetic DyGaG are interpreted by an extensional molecular-field theory. That is, with a special consideration of the exchange interaction in paramagnetic media transformed from the antiferromagnetism, the molecular-field theory is further extended to study the magnetic properties of these media around the transition temperatures. At the same time, as is known, the exchange interaction of the paramagnetic state in rare-earth–transition metal compounds can be wonderfully described by the two-sublattice molecular-field model [16,17]. Consequently, in our calculations, it is assumed that two magnetic sublattices exist

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doi:10.1016/j.jallcom.2009.08.115

in DyGaG, and the magnetizations M_A and M_B are discussed. Meanwhile, the high-magnetic anisotropy is revealed at 4.23 K when the magnetic field is up to 50 T. And, some interesting conclusions are drawn. Correspondingly, the theoretical results are compared with the available experiments.

2. Theoretical model

It is known that the formula of dysprosium gallium garnet (DyGaG) can be expressed as $Dy_3Ga_5O_{12}$, where the rare-earth Dy^{3+} ion is the unique magnetic ion in DyGaG. The cubic crystal structure of DyGaG belongs to the space group O_h^{10}-$Ia3d$, and the rare-earth Dy^{3+} ions occupy the dodecahedral sites, named as c crystal sites. There are 24 sites c in the unit cell. As mentioned above, the paramagnetism of DyGaG is transformed from the antiferromagnetism above the Néel temperature. Meanwhile, according to the investigation of the magnetic properties in dysprosium aluminum garnet (DyAG) [18], in our model, the two magnetically opposed sublattices in DyGaG are defined as A and B. Then, it is obvious that the total magnetization M of DyGaG is the sum of M_A and M_B calculated by A and B magnetic sublattices. Now, referring to the molecular-field theory, the theoretical model for the analysis of the magnetic properties in DyGaG is brought forward as follows.

In correspondence to the two magnetic sublattices A and B of Dy^{3+} ion, the exchange field in DyGaG can be expressed as

$$H_A = \lambda_{AA}M_A + \lambda_{AB}M_B, \tag{1}$$

$$H_B = \lambda_{BB}M_B + \lambda_{BA}M_A, \tag{2}$$

where λ_{AB} and λ_{BA} is regarded as the exchange field coefficients between the nearest-neighbor A and B magnetic sublattices, and λ_{AA} (or λ_{BB}) denotes the exchange field coefficients between the nearest-neighbor A and A (or B and B) magnetic sublattices. In terms of the antiferromagnetic molecular-field theory, these four exchange field coefficients in DyGaG should meet with the following expression

$$\lambda_{AB} = \lambda_{BA} = \lambda_1, \qquad \lambda_{AA} = \lambda_{BB} = \lambda_2. \tag{3}$$

Then, with the action of the external magnetic field H_e, substituting H_e into Eqs. (1) and (2), the total effective field can be given by

$$H_{iA} = H_e + \lambda_{AA}M_A + \lambda_{AB}M_B = (1+\alpha_A)H_e, \tag{4}$$

$$H_{iB} = H_e + \lambda_{BB}M_B + \lambda_{BA}M_A = (1+\alpha_B)H_e. \tag{5}$$

Due to $M_A = \chi_A H_e$ and $M_B = \chi_B H_e$, then

$$\alpha_A = \lambda_{AB}\chi_B + \lambda_{AA}\chi_A = \lambda_1\chi_B + \lambda_2\chi_A, \tag{6}$$

$$\alpha_B = \lambda_{BA}\chi_A + \lambda_{BB}\chi_B = \lambda_1\chi_A + \lambda_2\chi_B, \tag{7}$$

where χ_A and χ_B are the magnetic susceptibilities obtained from the A and B magnetic sublattices, respectively.

Furthermore, in virtue of Langevin theory, the magnetizations per Dy ion of A and B magnetic sublattices can be described as

$$M_A = M_{SA}B(y_A) = \frac{N_A J g_J \mu_B B(y_A)}{N}, \tag{8}$$

$$M_B = M_{SB}B(y_B) = \frac{N_B J g_J \mu_B B(y_B)}{N}, \tag{9}$$

where g_J is the Lander factor, J the total angular momentum quantum number, μ_B the Bohr magneton. Here, it should be pointed out that N_A and N_B denote the numbers of ions contributing to magnetization per unit volume in A and B sublattices, respectively, and, N is the sum of N_A and N_B, meanwhile, in our calculation, it is suggested that $N_A = N_B = N/2$. Besides, in the above equations, $B(y_A)$ and

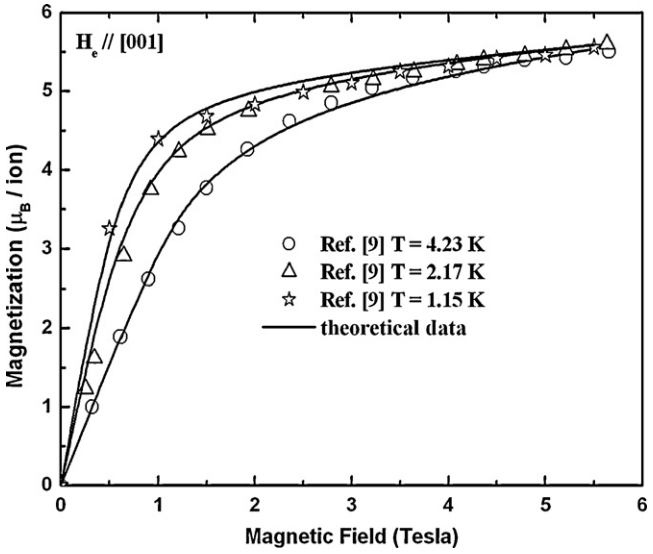

Fig. 1. The variation of magnetization with the magnetic fields along [0 0 1] direction at different temperatures.

$B(y_B)$ represent the Brillouin functions, and

$$y_A = \frac{J g_J \mu_B H_{iA}}{k_B T}, \tag{10}$$

$$y_B = \frac{J g_J \mu_B H_{iB}}{k_B T}, \tag{11}$$

where H_{iA} and H_{iB} are the total effective fields in A and B sublattices, respectively. Substituting Eqs. (4), (5), (10) and (11) into Eqs. (8) and (9), the formulae of M_A and M_B can be rewritten as

$$M_A = \frac{J g_J \mu_B \mathrm{B}((J g_J \mu_B (1+\alpha_A)H_e)/k_B T)}{2}, \tag{12}$$

$$M_B = \frac{J g_J \mu_B \mathrm{B}((J g_J \mu_B (1+\alpha_B)H_e)/k_B T)}{2}, \tag{13}$$

Hence, in terms of the above calculations of M_A and M_B, the total magnetization M in DyGaG can be obtained by

$$M = M_A + M_B. \tag{14}$$

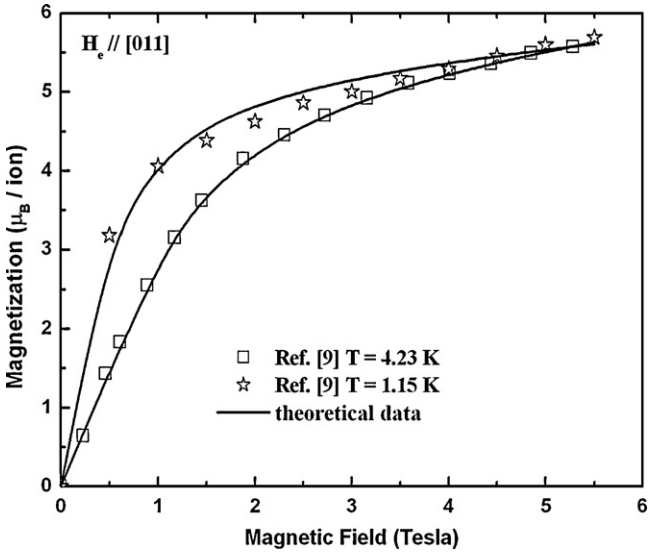

Fig. 2. The variation of magnetization with the magnetic fields along [0 1 1] direction at different temperatures.

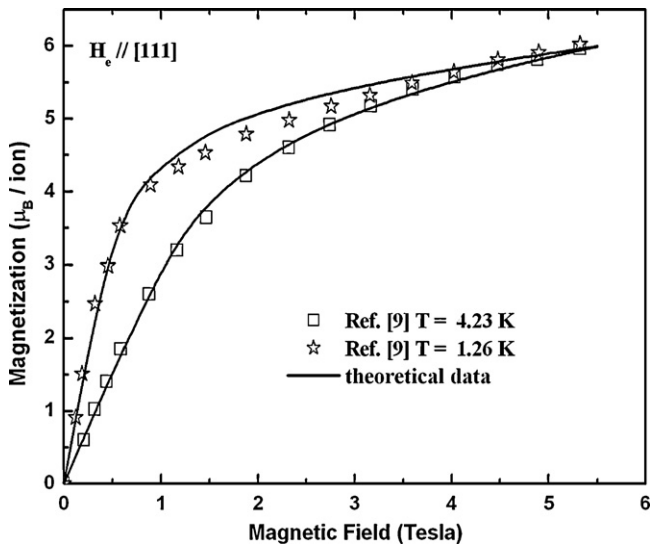

Fig. 3. The variation of magnetization with the magnetic fields along [1 1 1] direction at different temperatures.

3. Results and discussion

It is demonstrated by Dionne et al. [19] that the decision of the molecular-field coefficients is complex and important, and wonderful theoretical interpretations on the magnetic properties of some ferrimagnetic rare-earth iron garnets had been given by choosing appropriate molecular-field coefficients. Moreover, Rado and Folen [20] pointed out that the molecular-field coefficients could be deduced from suitable experimental results.

Owing to the absence of sufficient experimental data, we cannot give the specific values of the exchange interaction coefficients λ_1 and λ_2 in DyGaG. Thus, only a quantitative analysis of the exchange interaction coefficients in paramagnetic DyGaG above the Néel temperature can be carried out. According to antiferromagnetic molecular-field theory, we can know that the exchange field coefficient λ_1 should be negative, and λ_2 maybe positive or negative, moreover, $\lambda_1 > \lambda_2$. Then, in terms of the above theoretical model, the properties of α_A, α_B, χ_A and χ_B can be analyzed as follows.

As to Dy^{3+} ion in DyGaG, the ground term is $4f^9$, and, the ground multiplet $^6H_{15/2}$ is considered in our calculations, therefore, in Eqs. (8) and (9), $L = 5$, $S = 5/2$, $J = 15/2$ and $g_J = 1.333$. And, according to the magnetic behaviors for H_e applied to [0 0 1], [0 1 1] and [1 1 1] directions of DyGaG obtained from Ref. [9], the magnetization curves of DyGaG, in view of Eqs. (8)–(14), are fitted at different temperatures along [0 0 1], [0 1 1] and [1 1 1] directions in Figs. 1–3 where relatively good agreements between theoretical data and experiments are presented. The optimum values of the parameters α_A and α_B at different temperatures are given in Table 1.

To further describe the susceptibilities of A and B magnetic sublattices, Fig. 4(a)–(c) give the variations of the magnetizations M_A and M_B with the external magnetic field at different temperatures

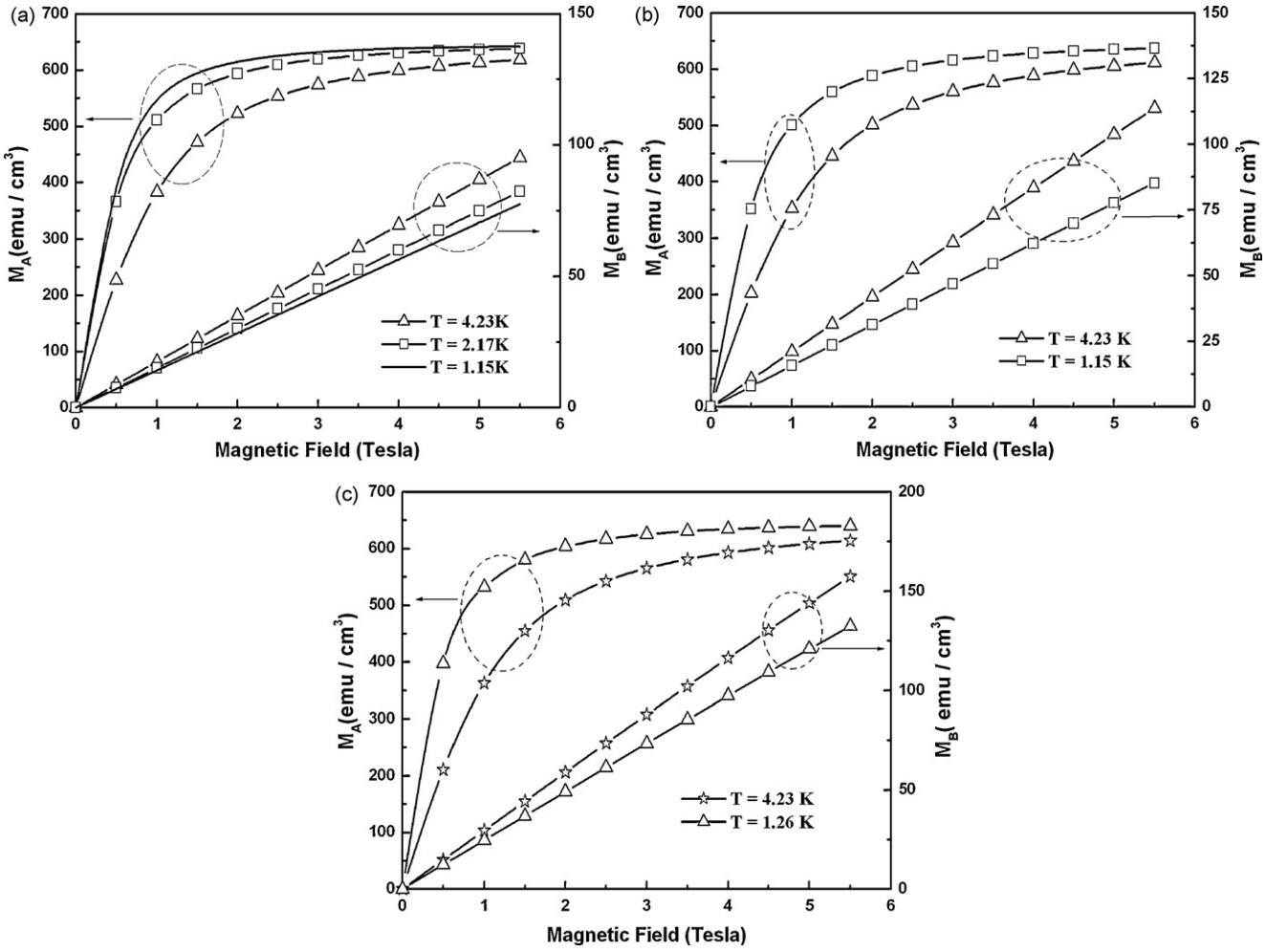

Fig. 4. The magnetization curves of the two magnetic sublattices A and B along different directions at different temperatures ((a) is [0 0 1] direction, (b) is [0 1 1] direction and (c) is [1 1 1] direction).

Table 1
The values of α_A and α_B at different temperatures along [0 0 1], [0 1 1] and [1 1 1] directions.

	[0 0 1]			[0 1 1]		[1 1 1]	
	4.23 K	2.17 K	1.15 K	4.23 K	1.15 K	4.23 K	1.16 K
α_A	0.25	0.2	−0.1	0.1	−0.4	0.15	−0.2
α_B	−0.955	−0.98	−0.99	−0.946	−0.989	−0.924	−0.981

along [0 0 1], [0 1 1] and [1 1 1] directions. Here, as for the paramagnetic DyGaG, it is interesting that M_A is nonlinear with H_e, and declined to saturation under higher magnetic fields, while M_B is linear with H_e. Moreover, it is worthy to note that $M_A > M_B$ at the same temperature and in the same magnetic field. Then, it can be predicted that, in contrast with B magnetic sublattice, the magnetic sublattice A is relatively easy to be magnetized, and, $\chi_A > \chi_B$. In addition, theoretical figures also reveal that, around the transition temperature of DyGaG (0.73 K), χ_A decreases with the increase of the temperatures, while χ_B tends to increase at higher temperatures.

Also, by careful investigation of the data in Table 1, some interesting points of view can be concluded. (1) It is found that there is striking difference in the values of α_A and α_B at 4.23 K when H_e is along the above three directions, which means that α_A and α_B show obvious anisotropy. (2) In our fitting to the experimental data, $\alpha_B < 0$ at different temperatures, while, at high temperatures (T = 2.17 K or 4.23 K), $\alpha_A > 0$, and, at low temperatures (T = 1.15 K or 1.16 K), $\alpha_A < 0$. In fact, owing to $\chi_A > \chi_B$, $\chi_A > 0$, $\chi_B > 0$, $\lambda_1 < 0$, and $\lambda_1 > \lambda_2$, it can be deduced that $\alpha_B < 0$ at any temperature, while α_A may be positive and negative. And, at low temperatures, the existence of the complex phase change in DyGaG can lead to the alteration of the sign of the exchange interaction coefficient λ_2 (from positive to negative), which also makes the sign of α_A change from positive to negative. (3) It can be suggested that the Néel temperature might be greater than 4.23 K, which would explain why $\chi_A > \chi_B$ (if H_e is more closely aligned with the A sublattice). Since the sublattices oppose one another, A will saturate at a lower H_e than B, where the spins must eventually flip to align with the A spins. (4) Meanwhile, with the decrease of the temperatures, the values of α_A and α_B also become much smaller. Of course, these viewpoints are open to be further theoretically or experimentally studied.

On the other hand, Fig. 5 gives the high-field (up to 50 T) magnetic properties in DyGaG at 4.23 K along [0 0 1], [0 1 1] and [1 1 1] directions. Here, it is shown that, under weak magnetic field (about less than 4 T), the magnetization is isotropic, while, with the increase of the magnetic fields, a remarkable anisotropy can be found. Moreover, it is obvious that the preferred direction of magnetization in DyGaG is [1 1 1] direction.

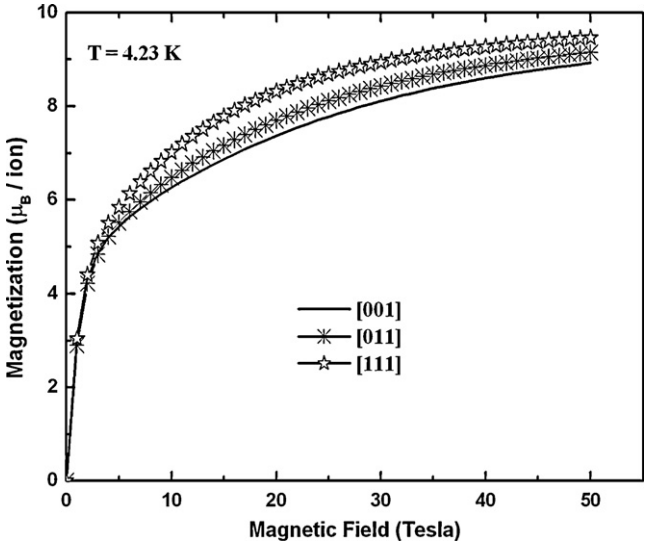

Fig. 5. The magnetization curves along [0 0 1], [0 1 1] and [1 1 1] directions at 4.23 K under high-magnetic fields (up to 50 T).

4. Conclusion

In the present study, the magnetic properties in paramagnetic DyGaG are theoretically analyzed by an extensional molecular-field theory where a relatively good agreement between theoretical data and experiments is obtained. Our investigation further confirms the importance of exchange interaction in paramagnetic gallium garnet. The exchange interaction coefficients λ_1 and λ_2 in DyGaG are quantitatively analyzed, $\lambda_1 < 0$, and $\lambda_1 > \lambda_2$, and it is implied that a complex phase change in DyGaG exists at low temperatures. Moreover, in our theoretical calculations, it is worthy to note that the parameters α_A and α_B associated with the exchange interaction have obvious anisotropy which suggests that the exchange interaction in DyGaG may be anisotropic, and $\alpha_B < 0$ at any temperatures, while α_A may be positive or negative at some specific temperatures. In addition, our theory points out that, under a higher magnetic field, the magnetization in DyGaG exhibits a notable anisotropy, while it is isotropic in low magnetic fields. Further, on the basis of this theoretical model, a quantum theoretical study of magnetic properties in DyGaG, with consideration of the crystal field and exchange interaction, is preformed [21].

Acknowledgement

This work is supported by The Young Scholars Fund of Beijing University of Chemical Technology under Grant No. QN0724.

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