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Charge and spin modulation in ferromagnetic semimetals

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We show that in ferromagnetic semimetals, in which the Ruderman-Kittel-Kasuya-Yosida indirect exchange predominates, the magnetically ordered state is unstable against inhomogeneous fluctuations. At a temperature slightly lower than the Curie temperature, the system enters into a state in which both the carrier density and the magnetization are modulated. The modulation scale is fixed by the Coulomb screening of the carriers, and can be tuned by changing the ratio of the electron and hole concentrations. At much lower temperatures a reentrant transition into a uniformly ordered state occurs. The modulated state is strongly suppressed by an external magnetic field. The results explain qualitatively the magnetic properties of EuB_6.

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Phase separation has attracted considerable attention since the discovery of charge and spin ordering in high-T_c superconductors.[1] The holes gain kinetic energy when phase separation into magnetically disordered hole-rich regions and magnetically ordered hole-free regions occurs (see Ref. 2 and references therein). The Coulomb interaction between the holes presumably leads to the stripe formation.[2] Phase separation into ferromagnetic hole-rich regions and antiferromagnetic hole-poor regions was considered theoretically and observed experimentally in other doped Mott insulators, like the manganites revealing the colossal magnetoresistance effect.[3]

In this paper we show that modulation of the carrier density and of the magnetic correlations should take place also in a very simple system with weak Coulomb interactions–a semimetal with localized spins (LS), which order ferromagnetically due to the Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect coupling. The ferromagnetic correlations are enhanced in the carrier rich regions and suppressed in the carrier poor ones. As in the cuprates, the Coulomb interaction is crucial in fixing the scale of the modulation. However, unlike the above examples, the ground state of the ferromagnetic semimetal (FSM) is uniform, and the modulated phase exists only at finite temperatures.

An example of a FSM is europium hexaboride, EuB_6.[4] This is a semimetal with electron and hole concentration per localized spin, n_0, of order of $10^{-3}-10^{-2}$.[4,5] The Eu spins ($S=7/2$) order ferromagnetically at the Curie temperature $T_c \approx 15$ K,[4] due to the RKKY indirect exchange.[6] The magnetic moment increases anomalously slowly with the decrease of the temperature, T, approaching the saturation value only at $T \approx 4$ K.[7] Another magnetic transition was observed at $T_1 \approx 12$ K,[4] and a third one might have been observed at $T_2 \approx 4$ K.[4,8] We show that all these properties are explained qualitatively by our theory.[9] The theory is applicable also to diluted magnetic semiconductors with carrier induced ferromagnetic exchange, which became a popular issue recently.[10]

The modulated phase exists only in FSM with a small Fermi energy ϵ_F^a, $\epsilon_F^a \ll J_a S$, where J_a is the s-f (s-d) coupling constant, and $a=e$ ($a=h$) for the electrons (holes). In this case, as shown below, the mean value $\langle S^z \rangle$, approaches S at a temperature $T \sim \Sigma_a J_a n_0^a$, which is much smaller than the Curie temperature T_c, while the carriers become spin polarized close to T_c. At temperatures, when $\langle S^z \rangle / S$ is small, and the carriers are polarized, spatial fluctuations of the carrier concentration (and accordingly, of the molecular field acting on the LS) lower the free energy of the LS, and the modulated state becomes stable. The modulation wave vector, q_0, is determined by the interplay between the Coulomb energy, which tends to increase q_0 and the gradient energy,[11] which reduces it.

The magnetic phase diagram of a FSM, with $n_0^e \gg n_0^h$ is shown in Fig. 1. Part of the results for this case were reported previously.[12]

The Hamiltonian of the s-f model is

$$\mathcal{H} = \sum_{\mathbf{k}a\sigma} \epsilon_{\mathbf{k}a} c^\dagger_{\mathbf{k}\sigma a} c_{\mathbf{k}\sigma a} - 2 \sum_{ia} J_a \mathbf{S}_i \cdot \mathbf{s}_{ia} + \mathcal{H}_C , \qquad (1)$$

where $c^\dagger_{\mathbf{k}\sigma a}$ ($c_{\mathbf{k}\sigma a}$) is the creation (annihilation) operator for a carrier of type a with spin σ, $\epsilon^a_{\mathbf{k}} = \hbar^2 k^2 / 2 m_a$ is the electron (hole) kinetic energy, $\mathbf{S_i}$ is the spin, localized at the lattice

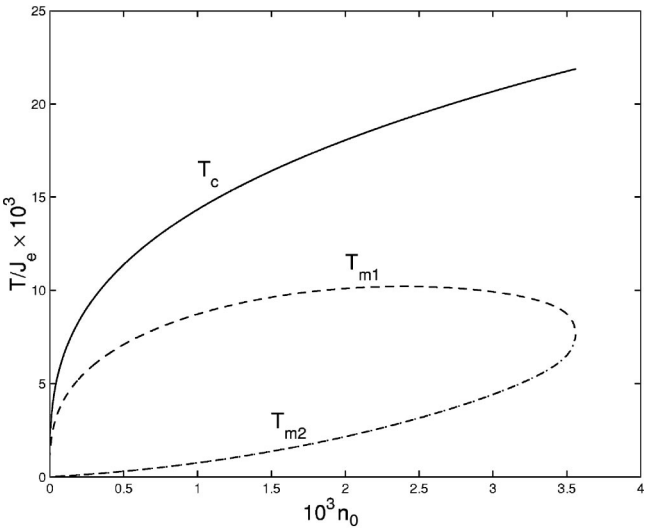

FIG. 1. Phase diagram of a FSM with $n_0^e \gg n_0^h$, $J_e = 0.1$ eV, $S = 3.5$, and $m_e = 0.3$. The dashed (T_{m1}) and dashed-dotted (T_{m2}) lines enclose the modulated phase region.

site i, $\mathbf{s}_{ia}$ is the electron (hole) spin, and $\mathcal{H}_C$ is the Coulomb interaction between the particles. We assume in what follows that both couplings J_e and J_h are positive. We also assume weak s-f coupling, $J_a S \ll W$, where W is of order of the band width, thus excluding the double-exchange magnets.

In a homogeneous system this Hamiltonian transforms, in the molecular field approximation, into

$$\mathcal{H}=\sum_{\mathbf{k}a\sigma} \epsilon_{\mathbf{k}a\sigma} c^{\dagger}_{\mathbf{k}\sigma a} c_{\mathbf{k}\sigma a} - \sum_i hS_i^z . \tag{2}$$

Here $\epsilon^a_{\mathbf{k}\sigma}=\epsilon^a_{\mathbf{k}}-\sigma\Delta_a/2$, $\Delta_a=2J_a\langle S^z\rangle=2J_a b(h/T)$, $b(x)$ is the Brillouin function:

$$b(x)=\left(S+\frac{1}{2}\right)\coth\left(S+\frac{1}{2}\right)x-\frac{1}{2}\coth\frac{x}{2}, \tag{3}$$

$h=\Sigma_a J_a(n^a_{\uparrow}-n^a_{\downarrow})$, $n^a_{\uparrow}(n^a_{\downarrow})$ is the number of carriers with spin up (down) per a localized spin, and $n^a_{\uparrow}+n^a_{\downarrow}=n^a_0$. Here and below we neglect the exchange and correlation effects coming from $\mathcal{H}_C$.

Expanding $b(h/T)$ and $n^a_{\uparrow}-n^a_{\downarrow}$ in powers of Δ, i.e., in powers of $\langle S^z\rangle$, we obtain the Curie temperature

$$T_c=\frac{S(S+1)}{3}\sum_a J_a^2 \mathcal{N}_a(\epsilon^a_F), \tag{4}$$

where $\mathcal{N}_a$ is the carrier density of states, $\mathcal{N}_a(\epsilon^a_F)=3n^a_0/2\epsilon^a_F$, the Fermi energy ϵ^a_F is $\epsilon^a_F=\hbar^2k^2_{Fa}/2m_a$, $k_{Fa}=(3\pi^2 n^a_0/\Omega)^{1/3}$, and Ω is the unit cell volume. At lower temperatures, when $\tau_c=(T_c-T)/T_c$ is about 0.05, Δ_a reaches ϵ^a_F, and the carriers become polarized. Then one has $n^a_{\downarrow}\ll n^a_{\uparrow}\approx n_0$, and $h=h_0=\Sigma_a J_a n^a_0$. We have $h_0/T_c\approx\epsilon^a_F/J_a S^2\ll 1$. Hence, unlike usual ferromagnets, $\langle S^z\rangle$ saturates to S not at $\tau_c\approx 1$, but at the much lower temperature $T\approx h_0$.

We show now that when the carriers are polarized, while $\langle S^z\rangle/S$ is small, the carrier concentration and the magnetization become unstable against spatial fluctuations.

Consider the free energy functional (FEF), $F[n(\mathbf{r})]$, of a system of localized spins interacting with a spin polarized, slightly inhomogeneous gas of electrons and holes. The electron (hole) density has the form $n_a(\mathbf{r})=n^a_0+\tilde{n}_a(\mathbf{r})$, with $\tilde{n}_a(\mathbf{r})/n^a_0\ll 1$, and

$$\int \tilde{n}_a(\mathbf{r})d\mathbf{r}=0. \tag{5}$$

$F[n(\mathbf{r})]$ can be written as

$$F[n(\mathbf{r})]=F_m[n(\mathbf{r})]+F_{car}[n(\mathbf{r})], \tag{6}$$

where $F_m[n(\mathbf{r})]$ is the FEF of the LS system in the carrier induced molecular field, $h(\mathbf{r})=\Sigma_a J_a n_a(\mathbf{r})$, and $F_{car}[n(\mathbf{r})]$ is the FEF of the polarized carriers. We have

$$F_m[n(\mathbf{r})]=-T\int d\mathbf{r}\ln\frac{\sinh[(2S+1)h(\mathbf{r})/2T]}{\sinh[h(\mathbf{r})/2T]}. \tag{7}$$

The FEF of a slightly inhomogeneous nonpolarized electron gas was derived by Hohenberg and Kohn[11] to the lowest order in the gradient expansion. For polarized electrons and holes the FEF can be written as

$$F_{car}[n(\mathbf{r})]=\sum_a F_a[n(\mathbf{r})]+F_{eh}[n(\mathbf{r})], \tag{8}$$

where

$$F_a[n(\mathbf{r})]=\frac{e^2}{2\varepsilon V^2}\int\frac{\tilde{n}_a(\mathbf{r})\tilde{n}_a(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}d\mathbf{r}' + \frac{\epsilon^a_{F\uparrow}}{36n^a_0k^{a2}_{F\uparrow}V}\int|\nabla\tilde{n}_a(\mathbf{r})|^2d\mathbf{r} + \frac{3\epsilon^a_{F\uparrow}}{n^{2/3}_{0a}V}\int(n^{5/3}_a(\mathbf{r})-n^{5/3}_{0a})d\mathbf{r}, \tag{9}$$

and

$$F_{eh}=-\frac{e^2}{\varepsilon V^2}\int\frac{\tilde{n}_e(\mathbf{r})\tilde{n}_h(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}d\mathbf{r}'. \tag{10}$$

Here e is the electron charge, ε is the dielectric constant, $k^a_{F\uparrow}=(6\pi^2 n^a_0/\Omega)^{1/3}$ is the Fermi wave vector, $\epsilon^a_{F\uparrow}=\hbar^2k^2_{F\uparrow}/2m$, and V is the sample volume.

Expanding the FEF up to the second order in $\tilde{n}_a(\mathbf{r})$ and taking into account Eq. (5), we get

$$F[\tilde{n}^e_{\mathbf{q}},\tilde{n}^h_{\mathbf{q}}]=\sum_{\mathbf{q}aa'}A^{aa'}_{\mathbf{q}}\tilde{n}^a_{\mathbf{q}}\tilde{n}^{a'}_{-\mathbf{q}}. \tag{11}$$

Here $\tilde{n}^a_{\mathbf{q}}$ is the Fourier transform of $\tilde{n}_a(\mathbf{r})$, with $\tilde{n}^a_{\mathbf{q}=\mathbf{0}}=0$,

$$A^{aa}_{\mathbf{q}}=\frac{1}{2\mathcal{N}^a_{\uparrow}}\left[1-\frac{J_a^2\mathcal{N}^a_{\uparrow}b'(h_0/T)}{T}+\frac{q^2}{12(k^a_{F\uparrow})^2}+\frac{\kappa_a^2}{q^2}\right],$$

$$A^{eh}_{\mathbf{q}}=A^{he}_{\mathbf{q}}=-\frac{J_eJ_h}{2T}b'\left(\frac{h_0}{T}\right)-\frac{2\pi e^2}{q^2\varepsilon\Omega}, \tag{12}$$

where $b'(x)=db/dx$, the density of states of the polarized carriers is $\mathcal{N}^a_{\uparrow}=3n^a_0/2\epsilon^a_{F\uparrow}$, and $\kappa_a^2=4\pi e^2\mathcal{N}^a_{\uparrow}/\varepsilon\Omega$ is the inverse screening length for the a carriers.

The minimization conditions, $\delta F/\delta\tilde{n}^e_{\mathbf{q}}=0$ and $\delta F/\delta\tilde{n}^h_{\mathbf{q}}=0$, yield a set of two homogeneous equations, whose determinant should be set to zero at the transition point

$$A^{ee}_{\mathbf{q}}A^{hh}_{\mathbf{q}}-A^{eh}_{\mathbf{q}}A^{he}_{\mathbf{q}}=0. \tag{13}$$

This gives the equation which governs the transition temperature and the modulation scale

$$1=\frac{1}{T}b'\left(\frac{h_0}{T}\right)(\mathcal{N}^e_\uparrow J_e^2+\mathcal{N}^h_\uparrow J_h^2)-\frac{1}{12}\left(\frac{\kappa_e^2}{k_{F\uparrow}^{h2}}+\frac{\kappa_h^2}{k_{F\uparrow}^{e2}}\right)$$
$$-\frac{1}{q^2}\left[\kappa_h^2+\kappa_e^2-\frac{b'}{T}(J_e\mathcal{N}^{1/2}_{\uparrow e}\kappa_h+J_h\mathcal{N}^{1/2}_{\uparrow h}\kappa_e)^2\right]$$
$$-\frac{q^2}{12}\left[\frac{1}{k_{F\uparrow}^{e2}}+\frac{1}{k_{F\uparrow}^{h2}}-\frac{b'}{T}\left(\frac{\mathcal{N}^e_\uparrow J_e^2}{k_{F\uparrow}^{h2}}+\frac{\mathcal{N}^h_\uparrow J_h^2}{k_{F\uparrow}^{e2}}\right)\right]$$
$$-\frac{q^4}{(12k^e_{F\uparrow}k^h_{F\uparrow})^2}. \tag{14}$$

To simplify things we treat first a FSM, in which one type of carriers predominates, say $n_0^e \gg n_0^h$. This condition can be met in EuB_6, where the concentration of carriers and the ratio n_0^e/n_0^h depend strongly on the quality of the sample.[4,5] The condition, $\epsilon_F^a \ll J_a S$, crucial for this theory, can be met also in strongly doped magnetic semiconductors, with $n_e(n_h)=0$.[10] We obtain then from Eq. (14) or directly from Eqs. (11) and (12), with $\tilde{n}^h_{\mathbf{q}}=0$

$$1+\frac{\kappa_e}{\sqrt{3}k^e_{F\uparrow}}-\frac{J_e^2\mathcal{N}^e_\uparrow}{T}b'\left(\frac{J_e n_0^e}{T}\right)+\frac{\kappa_e^2}{q^2}\left(1-\frac{q^2}{q_0^2}\right)^2=0, \tag{15}$$

with

$$q_0^2=2\sqrt{3}k^e_{F\uparrow}\kappa_e \ll (2k^e_{F\uparrow})^2. \tag{16}$$

We see that at a temperature, which satisfies the equation

$$1+\frac{\kappa_e}{\sqrt{3}k^e_{F\uparrow}}-\frac{J_e^2\mathcal{N}^e_\uparrow}{T}b'\left(\frac{J_e n_0^e}{T}\right)=0, \tag{17}$$

the system undergoes a phase transition into a modulated state with a modulation wave vector $q=q_0$.

The inequality in Eq. (16), which guarantees the validity of the gradient expansion, follows from the assumption that the Coulomb interaction is weak, and hence the screening range is large, i.e., $\kappa_e \ll k^e_{F\uparrow}$.

Equation (17) has two solutions, $T_{m1}>T_{m2}$, at n_0^e smaller than a critical concentration, n_c, [see Eq. (18)] and there are no solutions at $n_0^e>n_c$. The phase diagram of a FSM, with $n_0^e \gg n_0^h$ is shown in Fig. 1. The parameters used for solving Eq. (17), are appropriate to EuB_6. With $n_0^e \approx 10^{-3}$,[4] and $m_e \approx 0.3$,[5] we have $\epsilon_F^e \approx 0.055$ eV. Given $T_c \approx 15$ K,[4] we estimate from Eq. (4) (at $n_0^h=0$) $J_e S \approx 0.35$ eV. As follows from Eq. (4), T_c is proportional to $n_{0e}^{1/3}$, and JS at given T_c is proportional to $n_{0e}^{-1/6}$, i.e., the above estimate of $J_e S$ depends on n_0^e in a very subtle way. The small term $\kappa_e/\sqrt{3}k^e_{F\uparrow}$ in Eq. (17) was neglected.

Upon lowering the temperature, the FMS, with $n_0^e<n_c$, undergoes three successive phase transitions. The magnetic moment appears at $T=T_c$, the thermodynamic fluctuations being strong down to the much lower temperature of order of T_{m2}. At $T=T_{m1}$ the FMS enters into a modulated state, with a nonuniform distribution of the carrier density and of the magnetic correlations, the Fourier components $\tilde{n}^e_{\mathbf{q}}$ being the largest in the vicinity of $q=q_0$. At $T=T_{m2}$ a reentrant transition into a uniform magnetically ordered state takes place.

Solving numerically Eq. (17) one finds that the concentration n_c can be approximated as

$$n_c=0.425\left[\frac{J_e(S+0.094)}{W_{e\uparrow}}\right]^{3/2}, \tag{18}$$

where $W_{e\uparrow}=\epsilon^e_{F\uparrow}/n_{0e}^{2/3}$ is of order of the conduction band width. Calculating $\epsilon^e_{F\uparrow}$ at $n_0^e=n_c$, we conclude that the modulated phase exists if the inequality $\epsilon^e_{F\uparrow}<0.5J_eS$ holds.

In the general case, when there are both electrons and holes, the solution of Eq. (14) gives the modulation wave vector

$$q_0^2=2\sqrt{3}k^e_{F\uparrow}k^h_{F\uparrow}\frac{|J_e\kappa_e\mathcal{N}^{1/2}_{\uparrow e}-J_h\kappa_h\mathcal{N}^{1/2}_{\uparrow h}|}{(\mathcal{N}^e_\uparrow J_e^2 k_{F\uparrow}^{h2}+\mathcal{N}^h_\uparrow J_h^2 k_{F\uparrow}^{e2})^{1/2}}+O\left(\frac{\kappa}{k_F}\right), \tag{19}$$

while the transition temperature follows from the equation

$$1+\frac{1}{\sqrt{3}}\frac{|\mathcal{N}^e_\uparrow J_e-\mathcal{N}^h_\uparrow J_h|}{J_e^2\mathcal{N}^e_\uparrow+J_h^2\mathcal{N}^h_\uparrow}\left(\frac{J_e^2\kappa_e^2}{k_{F\uparrow}^{e2}}+\frac{J_h^2\kappa_h^2}{k_{F\uparrow}^{h2}}\right)^{1/2}$$
$$=\frac{1}{T}(\mathcal{N}^e_\uparrow J_e^2+\mathcal{N}^h_\uparrow J_h^2)b'\left(\frac{h_0}{T}\right)+O\left(\frac{\kappa^2}{k_F^2}\right). \tag{20}$$

At a given concentration of the electrons and the holes, Eq. (20), like Eq. (17), has two solutions, $T_{m1}>T_{m2}$. For small concentrations, $n_0^a \ll n_c$, the temperature T_{m1} is related to T_c, Eq. (4), as $T_{m1}\approx T_c/2^{2/3}$, while T_{m2} is much smaller. Note that q_0, Eq. (19), depends on the ratio n_0^e/n_0^h and can be tuned by doping. When $\mathcal{N}^e_\uparrow J_e=\mathcal{N}^h_\uparrow J_h$, the screening of the fluctuations of the electron-hole plasma is noneffective, q_0 is zero, and the magnetization and the carrier density are homogeneous at all temperatures. With parameters appropriate to EuB_6, q_0 is of order of 5×10^6 cm^{-1}.

The same results can be obtained in a different way, considering the longitudinal correlation function, $K_{zz}(\mathbf{q})$, of the localized spins, the exchange being mediated by the carriers.[12] Near the paramagnet-ferromagnet transition, when the carriers are unpolarized, this is equivalent to the RKKY indirect exchange, and leads to Eq. (4) for T_c. At lower temperatures, the carriers are polarized, the Coulomb interaction affects strongly the carrier susceptibility,[12,13] and $K_{zz}(\mathbf{q}_0)$, with q_0 given by Eq. (19), diverges at T governed by Eq. (20).

The next order terms in the expansion of the FEF, Eqs. (7) and (8), are cubic in $n^a_{\mathbf{q}}$ and one arrives at a FEF, which describes a first order liquid-solid-like transition into a periodic structure, with the magnitude of the reciprocal lattice vectors close to q_0.[14] Reasoning, analogous to that in Ref. 14 shows that the transition into the modulated state is nearly second order.

An external magnetic field, H, which aligns the LS, suppresses strongly the modulated phase. With the field H, the function $h(\mathbf{r})$ in Eq. (7) should be replaced by $h(\mathbf{r})+g\mu_B H$. A small field, $g\mu_B H \ll T_c$, destroys the modulated state, if it exceeds $h(\mathbf{r}) \approx h_0$, since in this case $F_m[n(\mathbf{r})]$

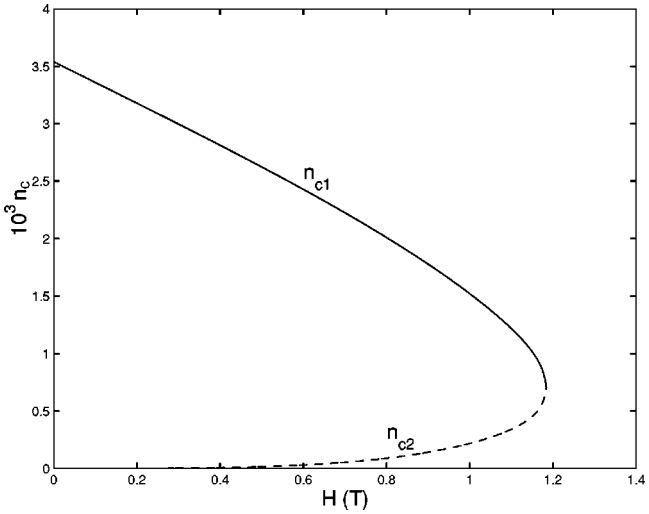

FIG. 2. Critical concentrations as a function of the magnetic field of a FSM with $n_0^e \gg n_0^h$, $J_e = 0.1$ eV, $S = 3.5$, and $m_e = 0.3$.

almost does not depend on $n(\mathbf{r})$. At a given H the modulated phase is always suppressed, if n_0^a is smaller than a critical concentration, $n_{c2}(H)$. Like in the zero-field case, this phase disappears also when n_0^a is larger than a critical concentration, $n_{c1}(H)$. Hence, the modulated state exists only in a restricted region $[n_{c2}(H), n_{c1}(H)]$, with $n_{c2}(0) = 0$, and $n_{c1}(0) = n_c$. Figure 2 shows the field dependence of the critical concentrations n_{c1} and n_{c2} for the same parameters as in Fig. 1. The modulated state in this case disappears at the critical field $H_c = 1.2$ T. At fields smaller than H_c and in the range $n_{c2} < n_0^e < n_{c1}$, the critical temperature T_{m1} decreases with increasing of the field, while T_{m2} increases, i.e., the modulated phase region on the (T, n) phase diagram shrinks in both directions.

Our results account qualitatively for most of the peculiar magnetic properties of EuB_6. The broad specific heat maximum between T_c and 3 K,[4,8,15] as well as the anomalously sluggish increase of the magnetization in this temperature region[7] agree with our conclusion that in FSM strong magnetic fluctuations survive till a temperature of order of T_{m2}, much smaller than T_c. Our theory explains naturally the sequence of transition temperatures observed in EuB_6.[4,7,8,15] The magnetic order sets in at the highest transition temperature, 15–16 K, accompanying by a λ-anomaly in the specific heat. The second transition (9–12 K) and the third one (4 K) correspond to T_{m1} and T_{m2}. In agreement with the above theory, the zero-field anomaly in the the specific heat at the second transition temperature is completely destroyed in an external field of 2 T.[4] It was shown in Ref. 6 that T_c increases monotonically with the increase of the carrier concentration, while T_{m1} saturates at high concentrations. This is also in agreement with our theory (see Fig. 1).

It was stated in a recent paper[16] that the bulk ferromagnetic transition occurs at 12.5 K, while metalization via overlap of magnetic polarons happens at the first transition at 15.5 K. More experiments are needed to elucidate this problem.

In conclusion, we showed that in a FSM, with $\epsilon_F < JS$, a modulated state is stable in a finite temperature region $T_{m2} < T < T_{m1} < T_c$. The modulation scale is fixed by the Coulomb interaction of the charge fluctuations. The modulated phase region shrinks in an external magnetic field, and disappears in relatively small fields. The theoretical results explain qualitatively the main magnetic properties of EuB_6.

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