Magnetic relaxation in uranium ferromagnetic superconductors

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There is proposed a phenomenological description of quasielastic neutron scattering in the ferromagnetic metals UGe_2 and UCoGe based on their property that magnetization supported by the moments located at uranium atoms is not a conserved quantity relaxing to equilibrium by the interaction with an itinerant electron subsystem. As a result the linewidth of quasielastic neutron scattering at $q \to 0$ acquires nonvanishing value at all temperatures but the Curie temperature.

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The family of heavy fermionic uranium metallic compounds UGe_2, URhGe, and UCoGe possesses an astonishing property of coexistence of superconductivity and ferromagnetism (for the most recent reviews, see Refs. 1 and 2). Ferromagnetism does not suppress the superconductivity with triplet pairing and, since the discoveries of superconductivity in uranium ferromagnets, they were considered as equal spin pairing superconductors similar to ^{3}He-A and ^{3}He-A_1 superfluids. The pairing interaction in liquid helium is due to spin-fluctuation exchange; hence it was quite natural to consider the same mechanism as the origin of superconductivity in uranium compounds. It was implied that they are fully itinerant ferromagnets and the same $5f$ electrons are responsible for ferromagnetism and superconductivity.[3] Thus practically in all publications the uranium ferromagnet superconductors were considered in the frame of theory of the isotropic Fermi liquid with ferromagnetism induced by the Landau-Stoner interaction between electrons (one can find the list of corresponding references in review[1]). This beautiful theoretical model is reasonable for ^{3}He. But in relation to the uranium compounds its applicability is quite doubtful in view of significant crystallographic and magnetic anisotropy, as well because of the nonitinerant nature of magnetism in these materials.

The static magnetic properties of UGe_2 are well described in Ref. 4 in terms of crystal field splitting of the U^{4+} state, which is the $^3H_4$ term of the $5f^2$ configuration of localized electrons, despite the presence of the itinerant electrons filling the bands formed by two $7s$, one $6d$, and one $5f$ uranium and also germanium orbitals. So, UGe_2 is actually a dual system where local and itinerant states of f electrons coexist. The example of such type of coexistence has been clearly demonstrated[5] by μSR measurements in another uranium compound $UPdAl_3$, where the Knight shift below T_{sc} indicates that local moment magnetism and superconductivity are carried by different electron substrates of $5f$ character, one of which involves the heavy quasiparticles. The localized nature of magnetism in uranium ferromagnetic compounds put forward as the most plausible pairing mechanism the interaction between the conduction electrons by means of spin waves in the system of localized moments. The first such type model has been applied to the superconducting antiferromagnet UPd_2Al_3[6] and then quite recently to the reentrant ferromagnetic superconductor URhGe.[7]

Leaving the superconducting properties for following publications, we discuss here the problem of magnetic excitations.

The magnetic excitations reveal themselves in neutron scattering measurements of the dynamical structure factor,

$$S_{\alpha\beta}(\mathbf{q},\omega) = \int_{-\infty}^{\infty} dt\, e^{i\omega t} \langle M_{\alpha\mathbf{q}}(t) M_{\beta-\mathbf{q}}(0)\rangle_{eq},$$

which is a wave vector–frequency dependent magnetic moments correlation function.[8] For an isotropic ferromagnet $S_{\alpha\beta}(\mathbf{q},\omega) = S(\mathbf{q},\omega)\delta_{\alpha\beta}$.

In the absence of walls and spin-orbital coupling the magnetization is a conserved quantity; hence, in an isotropic Heisenberg ferromagnet above Curie temperature, the only mechanism leading to the magnetization relaxation is the spin diffusion that results in[8,9]

$$S(\mathbf{q},\omega) = \frac{2\omega\chi(\mathbf{q})}{1-\exp(-\frac{\omega}{T})}\frac{\Gamma_{\mathbf{q}}}{\omega^2+\Gamma_{\mathbf{q}}^2}, \tag{1}$$

such that the linewidth of quasielastic scattering

$$\Gamma_{\mathbf{q}} = Dq^2 \tag{2}$$

is determined by the diffusion coefficient D. Here $\chi(\mathbf{q})$ is the static susceptibility at temperature T related to the imaginary part of susceptibility by the Kramers-Kronig relation $\chi(\mathbf{q}) = \int d\omega\, \chi''(\mathbf{q},\omega)/\pi\omega$, expressed in its turn through the dynamical structure factor by means of the fluctuation-dissipation theorem $2\chi''(\mathbf{q},\omega) = [1-\exp(-\frac{\omega}{T})]S(\mathbf{q},\omega)$. We put the Planck constant $\hbar = 1$.

The q^2 law dependence was observed in a wide temperature range above T_c in Ni and Fe (see Ref. 10, and references therein) reducing at $T = T_c$ to $\Gamma \propto q^{2.5}$ dependence according to predictions of mode-mode coupling theory.[11]

In weak itinerant ferromagnets above Curie temperature another mechanism of dissipationless relaxation can dominate with the structure factor given by the same Eq. (1) but with the linewidth determined by equality[12,13]

$$\chi(\mathbf{q})\Gamma_{\mathbf{q}} = \chi_P\omega(\mathbf{q}), \tag{3}$$

where χ_P is the noninteracting Pauli susceptibility and $\omega(\mathbf{q})$ is the Landau damping frequency equal to $\frac{2}{\pi}qv_F$ for the spherical Fermi surface. The linear in wave vector linewidth was observed in MnSi;[14] however, in the other weak itinerant ferromagnets MnP[15] and Ni_3Al,[16] the linewidth q dependence is closer to the dynamic scaling theory predictions.[11]

The investigations of magnetic excitations in UGe_2 and UCoGe have been reported in several publications.[3,17,18] The main result is that $\Gamma_{\mathbf{q}}$ unlike both Eqs. (2) and (3) does not vanish as $q \to 0$ for temperatures different from T_c.[19]

The authors[3] fairly specify that the finite value of $\Gamma_{\mathbf{q}}$ as $q \to 0$ implies that the uniform magnetization density is not a conserved quantity. One of the sources of violation of magnetization conservation in an itinerant electron system is electron-electron spin-orbit coupling. Hence, for the non-spin-conserving mechanism they have proposed the spin-orbit interaction associated with f electrons silently assuming that for f-electron system the interelectron spin-orbit interaction is stronger than in the ordinary metals. Indeed, the intra-atomic spin-orbital coupling is important at calculation of the electron band structure in compounds consisting of elements with big atomic numbers. However, it is well known that electron-lattice spin-orbital interaction in the crystals with inversion center plays a role similar or, better to say, equivalent to the usual spin-independent interband transition terms leading to the additional band splitting but not eliminating the Kramers double degeneracy of electronic states. So, when the one-electron band structure is fixed one can work with electron-electron interaction independent of initial atomic orbitals used for the band construction. The simplest of relaxation processes are single and double spin flip processes considered by Overhauser.[20] It was shown that the most effective type is the first one originating from spin-current interaction, which is the coupling between the magnetic moment of an electron and the magnetic field produced by the translational motion of another one. The derivation presented in Ref. 20 yields the relaxation rate which is many orders of magnitude smaller than the relaxation rate $\Gamma_{\mathbf{q}}$ of the order of several kelvin found in UGe_2.[3]

Another relaxation mechanism based on spin-orbital interaction of the itinerant Bloch electron spins with potential of ions in vibrating lattice was considered by Elliott.[21] For the alkaline metals he has found a relaxation rate of the order 10^{-3} K in fair agreement with experimentally observed values.[22] The corresponding theory for heavy metals is absent, but one can expect a larger magnitude of relaxation rate due to a larger deviation of the electron gyromagnetic ratio from its vacuum value. This mechanism in principle could be responsible for the finite value of homogeneous critical relaxation in uranium compounds if they were itinerant ferromagnets. The latter, however, looks quite doubtful.

Magnetic susceptibility of single UGe_2 crystals has been measured by Menovsky *et al.*[23](for the more recent results, see Refs. 24 and 4). The easy axis magnetization at zero temperature was found 1.43 μ_B/f.u. that, in the case of itinerant ferromagnetism, corresponds to a completely polarized single electron band. On the other hand, the neutron scattering measurements of the magnetic form factor[25] show that (i) the shape of its q dependence is not distinguishable from the wave vector dependences of the form factors of free U^{3+} or U^{4+} ions, (ii) practically whole magnetic moment both in paramagnetic and in ferromagnetic states concentrated at uranium atoms,[26] and (iii) its low temperature value at $q \to 0$ coincides with magnetization measured by magnetometer with accuracy of the order of 1%.

The configuration of localized $5f^2$ electrons of each atom of UGe_2 in paramagnetic state mostly consists of superposition of three quasidoublets and three singlets arising from the state with a fixed value of total momentum $J = 4$ split by the crystal field.[4] The temperature decrease causes the change in probabilities of populations of crystal field states revealing themselves in temperature dependence of the magnetic moment. The quasidegenerate ground state formed by the lower quasidoublet allows the system to order magnetically with the ordered moment of $\sim 1.5\mu_B$ two times smaller than the Curie-Weiss moment deduced from susceptibility above the Curie temperature.

The itinerant electron subsystem formed by $7s$, $6d$, and partly $5f$ electrons is also present providing about $0.02\mu_B$ long range magnetic correlations, as demonstrated by muon spin relaxation measurements.[27,28]

All mentioned observations as well as the theoretical treatment[4] unequivocally point to the local nature of UGe_2 ferromagnetism.

The interaction between localized and itinerant electron subsystems leads to the magnetization relaxation measured by neutron scattering in paramagnetic and ferromagnetic state. This type of relaxation can be considered as an analog of spin-lattice relaxation well known in physics of nuclear magnetic resonance.[29] In our case the magnetization created by the local moments of uranium atoms plays the role of "spin"subsystem, whereas the itinerant electrons present the "lattice" degrees of freedom absorbing and dissolving fluctuations of magnetization. According to this, we shall treat the total magnetization almost completely determined by the local moments of uranium atoms as not conserved quantity. A deviation of magnetization from the equilibrium value relaxes by transfer to the itinerant electrons. Unlike NMR relaxation determined by nucleus-electron magnetic moments interaction, the spin-lattice relaxation between the localized and conducting electrons is determined by spin-spin exchange processes and has no relativistic smallness typical for NMR relaxation.

Here we propose the phenomenological description of critical dynamics based on specific for strongly anisotropic ferromagnet uranium compounds property that magnetization supported by the moments located at uranium atoms is not a conserved quantity. To be more concrete we shall discuss mostly UGe_2.

Let us discuss first relaxation above the Curie temperature. The relaxation rate of the order parameter fluctuation is determined by deviation of system free energy

$$\mathcal{F} = \int dV \left(F_h + K_{ij} \frac{\partial M_\alpha}{\partial x_i} \frac{\partial M_\alpha}{\partial x_j} \right) \tag{4}$$

from equilibrium. UGe_2 crystallizes in the orthorhombic structure with magnetic ordering along a crystallographic direction, and the homogeneous part of the free energy density is

$$F_h = \alpha_x(T) M_x^2 + \alpha_y M_y^2 + \alpha_z M_z^2, \tag{5}$$

$$\alpha_x(T) = \alpha_{x0} \frac{T - T_c}{T_c}, \tag{6}$$

where $\alpha_y > 0, \alpha_z > 0$, whereas gradient energy in orthorhombic crystal written in exchange approximation[30] is determined by three nonzero constants K_{xx}, K_{yy}, K_{zz}. The coordinates x, y, z correspond to the a, b, c crystallographic directions.

To describe homogeneous relaxation together with diffusion we shall use a set of kinetic equations[31] relating to each magnetization component

$$\frac{\partial M_\alpha}{\partial t} = -A_{\alpha\beta}\frac{\delta \mathcal{F}}{\delta M_\beta}, \tag{7}$$

where the kinetic coefficient matrix has three nonzero elements A_{xx}, A_{yy}, A_{zz}. One can rewrite the above equations as

$$\frac{\partial M_\alpha}{\partial t} + \nabla_i j_{\alpha i} = -\frac{M_\alpha}{\tau_\alpha}, \tag{8}$$

where $\tau_x^{-1} = 2A_{xx}\alpha_{x0}\frac{T-T_c}{T_c}$, $\tau_y^{-1} = 2A_{yy}\alpha_y, \tau_z^{-1} = 2A_{zz}\alpha_z$, and there is no summation over the repeating indices in the right-hand side of this equation. The components of spin diffusion currents are

$$j_{\alpha i} = -2A_{\alpha\beta}K_{ij}\frac{\partial M_\beta}{\partial x_j}. \tag{9}$$

Measurements reported in the paper[3] with scattering wave vector $\mathbf{q}$ parallel to the crystal a axis revealed no extra scattering relative to the background, while for the $\mathbf{q}$ parallel to the c axis ($\mathbf{q} \parallel \hat{z}$) a strongly temperature dependent contribution was found. The treatment similar to what was used to get the diffusion scattering function[32] given by Eqs. (1) and (2) yields

$$S_{xx}(q_z,\omega) = \frac{2\omega\chi_{xx}(q_z)}{1-\exp(-\frac{\omega}{T})}\frac{\Gamma_{q_z x}}{\omega^2+\Gamma_{q_z x}^2}, \tag{10}$$

and the same structure expressions for $S_{yy}(q_z,\omega)$ and $S_{zz}(q_z,\omega)$ correlators. The corresponding widths of quasielastic scattering are

$$\Gamma_{q_z x} = 2A_{xx}\left[\alpha_x(T) + K_{zz}q_z^2\right], \tag{11}$$

$\Gamma_{q_z y} = 2A_{yy}(\alpha_y + K_{zz}q_z^2)$, and $\Gamma_{q_z z} = 2A_{zz}(\alpha_z + K_{zz}q_z^2)$. The correlator $S_{xx}(q_z,\omega)$ having a form characteristic of critical magnetic scattering contributes the main part in the differential cross section of scattering. As one can see $\Gamma_{q_z x}$ does not vanish as $q_z \to 0$ for temperatures different from T_c in correspondence with the results reported in Ref. 3. This property is the consequence of the relaxation mechanism specific for ferromagnetic uranium compounds where magnetization is created by the moments located at uranium atoms.

Below Curie temperature in the ferromagnetic state the deviation magnetization from equilibrium value $M = M(T)$ is $(M_x - M, M_y, M_z)$. The homogeneous part of free energy density of magnetic fluctuation is

$$F_h = 2|\alpha_x(T)|(M_x - M)^2 + \alpha_y M_y^2 + \alpha_z M_z^2. \tag{12}$$

One can write a kinetic equation similar to (8) only for the magnetization component parallel to ferromagnetic ordering

$$\frac{\partial(M_x - M)}{\partial t} + \nabla_i j_{xi} = -\frac{M_x - M}{\tau_x}, \tag{13}$$

with the same expression for the diffusion current as in a paramagnetic state. Dynamics of perpendicular to equilibrium magnetization components of magnetization is described by linearized Landau-Lifshitz-Gilbert equations[30,33]

$$\begin{aligned} \frac{1}{\gamma}\frac{\partial(M_y + aM_z)}{\partial t} &= -H_z M_z + h_z(t), \\ \frac{1}{\gamma}\frac{\partial(M_y - aM_z)}{\partial t} &= H_y M_y - h_y(t). \end{aligned} \tag{14}$$

Here γ is the gyromagnetic ratio, a is the dimensionless damping parameter, $H_y = M(K_{ij}q_i q_j + |\alpha_x| + \alpha_y), H_z = M(K_{ij}q_i q_j + |\alpha_x| + \alpha_z)$ are the components of "effective field,"[30] and $h_y(t), h_z(t)$ are the components of the time dependent transverse external field. This set of equations determines the spin-wave spectrum which has a particular simple form in the absence of damping $\omega = \gamma\sqrt{H_y H_z}$.

These equations also determine the $(\mathbf{q},\omega)$ dependences of yy and zz components of magnetic susceptibilities. In the low frequency limit they are frequency independent and pure real: $\chi_{yy} = H_y^{-1}, \chi_{zz} = H_z^{-1}$. The latter means that according to the fluctuation-dissipation theorem they do not make a contribution to the corresponding components of the dynamical structure factor determining the cross section of neutron scattering. Thus, at $T < T_c$, the structure factor is given by the same formula (10) as in the paramagnetic state, but the width of quasielastic scattering now is given by

$$\Gamma_{q_z x} = 2A_{xx}\left[2|\alpha_x(T)| + K_{zz}q_z^2\right]. \tag{15}$$

Equations (11) and (15) are the main results of the paper.

Experimentally, two independent values were determined: the width $\Gamma_{\mathbf{q}}$ and the amplitude $\chi(\mathbf{q})\Gamma_{\mathbf{q}}$ of distribution given by Eq. (1). We have established here the following points. The linewidth of quasielastic neutron scattering near the Curie temperature proves to be a linear function of $T - T_c$. The absolute value of the derivative $|d\Gamma_{q_z x}/dT|$ in a ferromagnetic region is roughly twice as large as the corresponding derivative in a paramagnetic region. The dependence of the wave vector q_z is parabolic. All of these findings are in qualitative correspondence with the experimental observations reported in Ref. 3 [see Figs. 4(a) and 4(b) in this paper]. At the same time it has been found[3] [see Fig. 4(d)] that the product $\chi(\mathbf{q})\Gamma_{\mathbf{q}}$ is temperature independent above Curie temperature but reveals the fast drop below T_c. Such type behavior means that below T_c the decrease of susceptibility $\chi(\mathbf{q})$ with temperature proves to be much faster than it is in accordance with mean field theory. This fact does not abolish our conclusions concerning the behavior independently determined width of quasielastic scattering intensity. Moreover, one should stress that Eq. (1) has completely general character [see Eqs. (2.54), (2.55b) in Ref. 9], which is not related to a particular form of $\chi(\mathbf{q})$ temperature dependence calculated in the frame of mean field theory or another model. An unusual behavior of susceptibility in UGe far enough from the critical region, where the concept of universality[34] does not work, is an open problem.

Conclusion. The totality of experimental observations points to the local nature of magnetism in uranium ferromagnetic superconductors. The interaction between localized and itinerant electron subsystems gives rise to a specific mechanism of magnetization relaxation similar to "spin-lattice"relaxation known in physics of nuclear magnetic resonance. This relaxation determined by exchange spin-spin coupling is much faster than NMR relaxation supported by much weaker interaction between electron and nuclei magnetic moments. We developed a phenomenological description of quasielastic magnetic relaxation based on specific for heavy fermionic ferromagnet uranium compounds property that magnetization supported by the moments located at

uranium atoms is not a conserved quantity. As a result the linewidth of quasielastic neutron scattering at $q \to 0$ acquires nonvanishing value at all temperatures besides the Curie temperature. The treatment is the simple application of a general description of critical relaxation proposed by Landau and Khalatnikov for a case of nonconserving order parameter.[31] The main message of the paper is that the nonconservation of magnetization in ferromagnetic superconducting uranium compounds points to the local character of magnetization relaxing to equilibrium by the interaction with an itinerant electron subsystem.

Quite recently this point has been confirmed by the microscopic analysis.[35] It was shown explicitly that the presence of two types of electrons breaks the cancellation between the contribution to $\Gamma(0)$ from self-energy and vertex correction insertions into the spin polarization bubble including the Aslamazov-Larkin processes.

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