Nuclear spin-lattice relaxation time in UCoGe

V. P. Mineev
Université Grenoble Alpes, CEA, IRIG, PHELIQS, F-38000 Grenoble, France
and Landau Institute for Theoretical Physics, 142432 Chernogolovka, Russia

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The NMR measurements performed on a single orthorhombic crystal of superconducting ferromagnet UCoGe [Y. Ihara *et al.*, Phys. Rev. Lett. **105**, 206403 (2010)] demonstrate strongly anisotropic magnetic properties of this material. The presented calculations allow one to establish the dependence of longitudinal spin-lattice relaxation rate from temperature and magnetic field. The value $(1/T_1T)$ in field perpendicular to spontaneous magnetization directed along the c axis has a maximum in the vicinity of Curie temperature, whereas it does not reveal similar behavior in a field parallel to the direction of spontaneous magnetization. Also there was shown that the longitudinal spin-lattice relaxation rate is strongly field dependent when the field directed in the b-crystallographic direction but field independent if magnetic field is oriented along the a axis.

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I. INTRODUCTION

The uranium ferromagnetic superconductors UGe_2, URhGe, and UCoGe discovered more than a decade ago still continue to attract the attention of the condensed matter community. The superconducting properties of these materials originate from the unusual pairing mechanism induced by magnetic fluctuations (see the recent experimental [1] and theoretical [2] reviews, and references therein).

Commonly these compounds are considered as itinerant f-electron metals meaning that the magnetism is determined by the band electrons according to the Stoner mechanism. Indeed, the numerical calculations show that the contribution of f electrons to the bands intersecting the Fermi level is significant. However, the x-ray magnetic circular dichroism measurements [3] and band structure calculations [4] point to the local nature of the ferromagnetism in UCoGe. Namely, a comparison of the total uranium moment M^U_{tot} to the total magnetization M_{tot} at different magnitude and direction of magnetic field indicates that the magnetic moment of cobalt ions and itinerant electrons put together an insignificant part of the total magnetic moment, hence the uranium ions dominate the magnetism of UCoGe. The same is true also in the related compounds URhGe [5] and UGe_2 [6].

In all these compounds the magnetic moment per atom at zero temperature M_0 is smaller than the magnetic moment per atom M_{CW} determined from Curie-Weiss law in paramagnetic state. According to the Wohlfarth criteria the degree of itinerancy increases with increasing the difference $M_{CW} - M_0$. In the present compounds the difference $M_{CW} - M_0$ characterizes the degree of itineracy of f electrons, but not an itinerant character of the ferromagnetic state. In the band language the local nature of magnetism means that the magnetic moment is furnished by electrons in spin-up and spin-down states with different orbital momentum projection filling the cellar below Fermi level [4,7,8]. In the real space these states looks like the f-type Wannier states. The same is true in the case of d electrons in transient metals [9,10]. This type of magnetism is different from the Heisenberg ferromagnetism of isolated magnetic moments of magnetic ions formed by the electron states split by the crystal field as well as from the pure Stoner-Hubbard magnetism of itinerant electrons. A microscopic description of such type of magnetic state is not developed.

The nuclear magnetic resonance (NMR) is one of the main tools for study of the magnetic properties of metals in normal and superconducting states. The direction-dependent ^{59}Co NMR measurements [11] of the Knight shift and nuclear spin-lattice relaxation time provide knowledge of static and dynamic susceptibility components in this orthorhombic metal. The relaxation rate in UCoGe in the field perpendicular to the easy magnetization axis along the c-crystallographic direction strongly surpasses the relaxation rate in the field parallel to it. Moreover, it was demonstrated that in the vicinity of the ferromagnetic transition the spin-lattice relaxation rate in the magnetic field along the easy magnetization is hardly deviates from Korringa -type behavior, while in the other field directions there is strong enhancement of relaxation (see Fig. 1). The NMR and ^{59}Co nuclear quadrupole resonance studies on the YCoGe compound free of uranium f electrons shows that the d electron of Co atoms plays no role in ferromagnetism of UCoGe originating from the U-5f electrons [11,12].

Here we present calculations allowing one to establish the peculiar dependence of the longitudinal spin-lattice relaxation rate from temperature and magnetic field. The paper is organized as follows. As was explained above, the magnetism of UCoGe cannot be described in the frame of a model of pure itinerant metallic state. Nevertheless we find it pertinent to begin with a reminder of the spin-lattice relaxation rate derived by Moriya and Ueda [13,14] in the isotropic itinerant ferromagnets. It is expressed through the static susceptibility

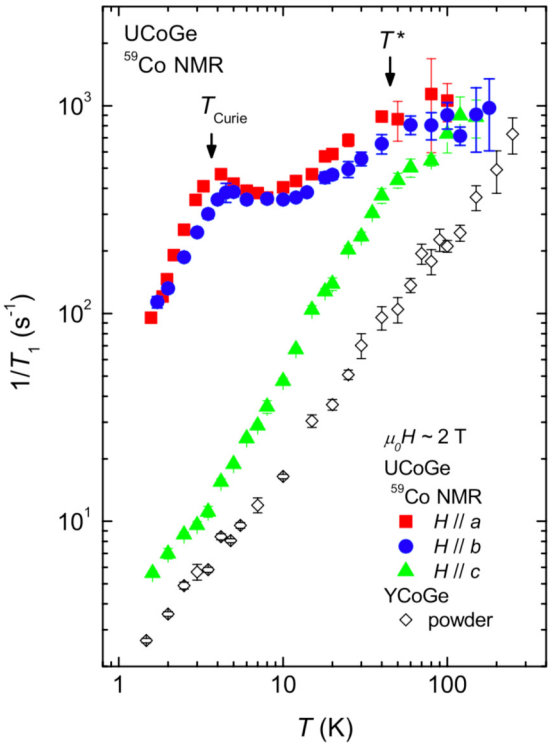

FIG. 1. Nuclear spin-lattice relaxation rate $(1/T_1)$ measured in three different field directions [11]. The results of ^{59}Co NQR on YCoGe, a reference compound without f electrons, are also displayed. The figure is reproduced from Ref. [11].

along the external field. Thus, it is independent from field direction, which is evidently inapplicable to NMR in UCoGe.

Then, to interpret NMR observations one needs to know the wave-vector–frequency-dependent magnetic susceptibility tensor in the orthorhombic UCoGe compound that is currently not available. However, the wave-vector dependence of static susceptibility components has been established from phenomenological consideration [2]. One can obtain the frequency dependence of susceptibility by a simple generalization of the corresponding static expression. This allows us to calculate the NMR longitudinal relaxation rate $1/T_1$ and establish its temperature and field dependence.

II. NUCLEAR MAGNETIC RESONANCE RELAXATION RATE

A. $1/T_1$ in isotropic itinerant ferromagnets

Before a discussion of the anisotropic NMR properties of UCoGe it is pertinent to first remind one of the theoretical results obtained by Moriya and Ueda [13,14] in the application to the isotropic itinerant ferromagnets. In this case the nuclear spin-lattice relaxation rate measured in a field H along the spontaneous magnetization chosen as z direction is determined by a correlation function of field fluctuations in the direction perpendicular to the external field. The latter according to the fluctuation-dissipation theorem is expressed through the imaginary part of susceptibility $\chi''_{+-}(\mathbf{k},\omega)$ of electron gas in the direction perpendicular to the external field

$$\left(\frac{1}{T_1T}\right)_z \propto \lim_{\omega\to 0}\int \frac{d^3k}{(2\pi)^3}|A_{hf}|^2\frac{\chi''_{+-}(\mathbf{k},\omega)}{\omega}. \tag{1}$$

The calculations have been done in [13] and yield

$$\left(\frac{1}{T_1T}\right)_z \propto \begin{cases}\chi_z(T), & \text{paramagnetic state}\\ M^{-2}, & \text{ferromagnetic state,}\end{cases} \tag{2}$$

where $\chi_z(T)$ is the temperature-dependent static susceptibility in the field direction and $M = M(T)$ is the magnetization in ferromagnetic state. Thus, the summation of perpendicular susceptibility over reciprocal space gives rise to the longitudinal susceptibility. The origin of this result is that $\chi_{+-}(\mathbf{k},\omega)$ is determined through the spin-up spin-down band splitting expressed through the static longitudinal susceptibility in paramagnetic state and through the M^{-2} in ferromagnetic state. One can find a modification of this result in finite external magnetic field in [15], where in particular it is shown that

$$\lim_{H\to\infty}\left(\frac{1}{T_1T}\right)_z \to 0 \tag{3}$$

as it should be in the presence of the strong band splitting.

Another important point essential for the results Eq. (2) is that $\chi''_{+-}(\mathbf{k},\omega)\propto\omega/kv_F$ both in the noninteracting and the interacting Fermi gas such that the integration over reciprocal space in Eq. (1) is in fact two-dimensional.

B. $1/T_1$ in UCoGe

In the orthorhombic metal the nuclear spin-lattice relaxation rate $1/T_1$ measured in a field along the l direction is expressed in terms of the imaginary part of the dynamic susceptibility along the m and n directions, perpendicular to l, $\chi''_{m,n}(\mathbf{k},\omega)$ as

$$\left(\frac{1}{T_1T}\right)_l \propto \sum_{\mathbf{k}}\left[|A^m_{hf}|^2\frac{\chi''_m(\mathbf{k},\omega)}{\omega}+|A^n_{hf}|^2\frac{\chi''_n(\mathbf{k},\omega)}{\omega}\right]. \tag{4}$$

At low temperatures the relaxation rate $(1/T_1T)$ for **H** parallel to the c-crystallographic direction is more than an order of magnitude smaller than those measured in the other two field directions parallel to the a and b axes [11] (see Fig. 1). So, one can use the expressions

$$\left(\frac{1}{T_1T}\right)_b \propto \sum_{\mathbf{k}}|A^c_{hf}|^2\frac{\chi''_c(\mathbf{k},\omega,H_b)}{\omega}, \tag{5}$$

$$\left(\frac{1}{T_1T}\right)_a \propto \sum_{\mathbf{k}}|A^c_{hf}|^2\frac{\chi''_c(\mathbf{k},\omega,H_a)}{\omega}, \tag{6}$$

neglecting the terms, originating from the imaginary part of susceptibilities χ''_a and χ''_b.

The static susceptibilities in uranium ferromagnets are derived in [2]. Along the c axis it is

$$\chi_c(\mathbf{k}) = \frac{1}{[\chi_c(H_l)]^{-1}+2\gamma^c_{ij}k_ik_j}, \tag{7}$$

where $\chi_c(H_l)=\frac{\partial M_c}{\partial H_c}$ is the homogeneous static susceptibility along the c axis at fixed value of magnetic field H_l along

the $l = a, b$ direction. The simplest generalization for the dynamic case obeying the Kramers-Kronig relation

$$\chi_c(\mathbf{k}) = \frac{1}{\pi}\int \frac{\chi_c''(\mathbf{k},\omega)}{\omega}d\omega$$

is the following:

$$\chi_c(\mathbf{k},\omega) = \frac{1}{-\frac{i\omega}{A} + [\chi_c(H_l)]^{-1} + 2\gamma_{ij}^c k_i k_j}, \tag{8}$$

where $\chi_c''(\mathbf{k},\omega) = Im\chi_c(\mathbf{k},\omega)$, and A is a constant. Thus, to estimate the sum over momenta we can work with

$$\frac{\chi_c''(\mathbf{k},\omega,H_l)}{\omega} = \frac{A}{\omega^2 + A^2\{[\chi_c(H_l)]^{-1} + 2\gamma_{ij}k_i k_j\}^2}. \tag{9}$$

The field dependence of $\chi_c(H_l)$ can be determined as follows [2]. The Landau expansion of the free energy near the Curie temperature in the presence of magnetic field is

$$\begin{aligned} F &= \alpha_c M_c^2 + \beta_c M_c^2 + \alpha_a M_a^2 + \alpha_b M_b^2 \\ &+ \beta_{ac} M_a^2 M_c^2 + \beta_{bc} M_b^2 M_c^2 - \mathbf{HM}, \end{aligned} \tag{10}$$

where

$$\alpha_c = \alpha_{c0}(T - Tc0), \quad \alpha_a > 0, \quad \alpha_b > 0. \tag{11}$$

In a constant magnetic field perpendicular to the spontaneous magnetization $\mathbf{H} = H_b\hat{b}$ the equilibrium magnetization projection along the b direction

$$M_b \approx \frac{H_b}{2(\alpha_b + \beta_{bc}M_c^2)} \tag{12}$$

is obtained by minimization of free energy (10) with respect to M_b. Substituting this expression back to (10) we obtain

$$F = \alpha_c M_c^2 + \beta_c M_c^4 - \frac{1}{4}\frac{H_b^2}{\alpha_b + \beta_{bc}M_c^2}, \tag{13}$$

that gives after expansion of the denominator in the last term,

$$F = -\frac{H_b^2}{4\alpha_b} + \tilde{\alpha}_c M_c^2 + \tilde{\beta}_c M_c^4 + \ldots, \tag{14}$$

where

$$\tilde{\alpha}_c = \alpha_{c0}(T - T_{c0}) + \frac{\beta_{bc}H_b^2}{4\alpha_b^2}, \tag{15}$$

$$\tilde{\beta}_c = \beta_c - \frac{\beta_{bc}}{\alpha_b}\frac{\beta_{bc}H_b^2}{4\alpha_b^2}. \tag{16}$$

Thus, in a magnetic field $\mathbf{H} = H_b\hat{b}$ perpendicular to the direction of spontaneous magnetization the Curie temperature decreases as

$$T_c(H_b) = T_{c0} - \frac{\beta_{bc}H_b^2}{4\alpha_b^2\alpha_{c0}}. \tag{17}$$

The corresponding formula for a field parallel to the a axis is

$$T_c = T_c(H_a) = T_{c0} - \frac{\beta_{ac}H_a^2}{4\alpha_a^2\alpha_{c0}}. \tag{18}$$

The coefficient $\alpha_a \gg \alpha_b$, which is the a direction, is much harder magnetically than the b direction. Hence, the Curie temperature is practically independent from magnetic field in the a direction $T_c(H_a) \approx T_{c0}$. The susceptibility along the c axis,

$$\chi_c(H_b) = \begin{cases} \frac{1}{4\alpha_{c0}\left(T_{c0} - \frac{\beta_{bc}H_b^2}{4\alpha_b^2\alpha_{c0}} - T\right)}, & T < T_c(H_b) \\ \frac{1}{2\alpha_{c0}\left(T - T_{c0} + \frac{\beta_{bc}H_b^2}{4\alpha_b^2\alpha_{c0}}\right)}, & T > T_c(H_b), \end{cases} \tag{19}$$

increases with magnetic field along the b axis but keeps the constant value in magnetic field parallel to the a axis,

$$\chi_c(H_a) = \begin{cases} \frac{1}{4\alpha_{c0}(T_{c0} - T)}, & T < T_{c0} \\ \frac{1}{2\alpha_{c0}(T - T_{c0})}, & T > T_{co}. \end{cases} \tag{20}$$

Now, let us make the integration in Eqs. (5) and (6). For simplicity one can calculate the converging integral in the spherical approximation

$$\begin{aligned} \left(\frac{1}{T_1T}\right)_l &\propto \int \frac{4\pi k^2 dk}{(2\pi)^3}\frac{A}{\omega^2 + A^2\{[\chi_c(H_l)]^{-1} + 2\gamma k^2\}^2} \\ &= \frac{\sqrt{2}}{32\pi A\gamma^{3/2}}\frac{\sqrt{\chi_c(H_l)}}{\left(1 + \frac{\omega^2\chi_c^2(H_l)}{A^2}\right)^{1/4}}\frac{1}{\cos\left(\frac{1}{2}\arctan\frac{\omega\chi_c(H_l)}{A}\right)}. \end{aligned} \tag{21}$$

We see that

$$\frac{1}{T_1(H_b)T} \propto \begin{cases} \sqrt{\chi_c(H_b)}, & \chi_c(H_b) \ll \frac{A}{\omega} \\ \sqrt{\frac{A}{\omega}}, & \chi_c(H_b) \gg \frac{A}{\omega} \end{cases} \tag{22}$$

as a function of temperature reaches a maximum in the vicinity of Curie temperature. The same is true for relaxation rate $\frac{1}{T_1(H_a)T}$ in the field parallel to the a axis.

The magnetic field along the b axis shifts the maximum of relaxation rate to the lower temperature; the field along the a axis does not. This effect is clearly demonstrated by the measurements in high enough fields reported in [16]. The deviation of the maximum of $\frac{1}{T_1(H_b)T}$ from the maximum of $\frac{1}{T_1(H_a)T}$ in a weak field $H = 2\,\mathrm{T}$ in Fig. 1 probably originates from a slight misalignment of field orientation as pointed out in [17].

The similar calculation of spin-lattice relaxation in the field parallel to the c axis $\frac{1}{T_1(H_c)T}$ is expressed through the magnetic susceptibilities along the a and b crystallographic directions. Both of them are much smaller than the susceptibility in the c direction and neither of them change in temperature interval near the Curie temperature. Hence, $\frac{1}{T_1(H_c)T}$ in the vicinity of critical temperature is practically temperature independent.

III. CONCLUSION

We have demonstrated that in contrast with the isotropic weak ferromagnets the longitudinal spin-lattice relaxation rate in UCoGe is expressed through the static susceptibility in the perpendicular to magnetic field direction. The value $(1/T_1T)$ in the field perpendicular to spontaneous magnetization has maximum in the vicinity of Curie temperature, whereas it does not reveal similar behavior in the field parallel to the direction of spontaneous magnetization. These results are in qualitative

correspondence with the experimental data presented in Fig. 1 [11].

Also there was shown that the longitudinal spin-lattice relaxation rate is strongly field dependent when the field is directed in the *b*-crystallographic direction but field independent if magnetic field is oriented along the *a* axis, which also is in accordance with experimental observations [16].

The presented calculations have been done using isotropic wave vector dispersion of susceptibilities. The real dispersion laws can be quite different and the simple relationship between the relaxation rate and the susceptibility will be lost. However, the field-temperature dependence of $1/T_1$ will qualitatively remain.

Another observation which possibly can be useful for the future theoretical efforts is that the calculations were performed taking the kinetic coefficient $A = \text{const}$. However, taking $A \sim kv$, where v is a constant with dimensionality of velocity, the integration over reciprocal space becomes two-dimensional and we come to $\frac{1}{T_{1b}T} \propto \chi_c(T, H_b)$. A theoretical description of wave-vector–frequency dependence of magnetic susceptibility of actinide intermetallics is still not at hand.

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[1] D. Aoki, K. Ishida, and J. Flouquet, J. Phys. Soc. Jpn. **88**, 022001 (2019).
[2] V. P. Mineev, Usp. Fiz. Nauk **187**, 129 (2017) [Phys. Usp. **60**, 121 (2017)].
[3] M. Taupin, J.-P. Sanchez, J.-P. Brison, D. Aoki, G. Lapertot, F. Wilhelm, and A. Rogalev, Phys. Rev. B **92**, 035124 (2015).
[4] M. Samsel-Czekala, S. Elgazzar, P. M. Oppeneer, E. Talik, W. Waleszyk, and R. Troc, J. Phys.: Condens. Matter **22**, 015503 (2010).
[5] F. Wilhelm, J. P. Sanchez, J.-P. Brison, D. Aoki, A. B. Shick, and A. Rogalev, Phys. Rev. B **95**, 235147 (2017).
[6] N. Kernavanois, B. Grenier, A. Huxley, E. Ressouche, J. P. Sanchez, and J. Flouquet, Phys. Rev. B **64**, 174509 (2001).
[7] G. H. Lander, M. S. S. Brooks, and B. Johansson, Phys. Rev. B **43**, 13672 (1991).
[8] A. B. Shick, V. Janiš, V. Drchal, and W. E. Pickett, Phys. Rev. B **70**, 134506 (2004).
[9] O. Eriksson, B. Johansson, R. C. Albers, A. M. Boring, and M. S. S. Brooks, Phys. Rev. B **42**, 2707 (1990).
[10] C. T. Chen, Y. U. Idzerda, H.-J. Lin, N. V. Smith, G. Meigs, E. Chaban, G. H. Ho, E. Pellegrin, and F. Sette, Phys. Rev. Lett. **75**, 152 (1995).
[11] Y. Ihara, T. Hattori, K. Ishida, Y. Nakai, E. Osaki, K. Deguchi, N. K. Sato, and I. Satoh, Phys. Rev. Lett. **105**, 206403 (2010).
[12] K. Karube, T. Hattori, Y. Ihara, Y. Nakai, K. Ishida, N. Tamura, K. Deguchi, N. K. Sato, and H. Harima, J. Phys. Soc. Jpn. **80**, 064711 (2011).
[13] T. Moriya and K. Ueda, Solid State Commun. **15**, 169 (1974).
[14] T. Moriya, *Spin Fluctuations in Itinerant Electron Magnetism* (Springer-Verlag, Berlin, 1985).
[15] M. Kontani, T. Hioki, and Y. Masuda, Solid State Commun. **18**, 1251 (1976).
[16] T. Hattori, K. Karube, K. Ishida, K. Deguchi, N. K. Sato, and T. Yamamura, J. Phys. Soc. Jpn. **83**, 073708 (2014).
[17] T. Hattori, Y. Ihara, Y. Nakai, K. Ishida, Y. Tada, S. Fujimoto, N. Kawakami, E. Osaki, K. Deguchi, N. K. Sato, and I. Satoh, Phys. Rev. Lett. **108**, 066403 (2012).