PHYSICAL REVIEW B **89**, 161103(R) (2014)

# Dispersive magnetic-resonance mode in the Kondo semiconductor $CeFe_2Al_{10}$

Jean-Michel Mignot,[1,*] Pavel A. Alekseev,[2,3] Julien Robert,[1] Sylvain Petit,[1] Takashi Nishioka,[4] Masahiro Matsumura,[4] Riki Kobayashi,[5,†] Hiroshi Tanida,[6] Hiroki Nohara,[6] and Masafumi Sera[6]

[1]*Laboratoire Léon Brillouin, CEA-CNRS, CEA/Saclay, 91191 Gif sur Yvette, France*

[2]*National Research Centre "Kurchatov Institute", 123182 Moscow, Russia*

[3]*National Research Nuclear University "MEPhI", Kashirskoe shosse 31, 115409 Moscow, Russia*

[4]*Graduate School of Integrated Arts and Science, Kochi University, Kochi 780-8520, Japan*

[5]*Neutron Science Laboratory, Institute for Solid State Physics, University of Tokyo, Tokai 319-1106, Japan*

[6]*Department of Quantum Matter, ADSM, Hiroshima University, Higashi-Hiroshima 739-8530, Japan*

(Received 8 January 2014; revised manuscript received 22 March 2014; published 4 April 2014)

The $CeT_2Al_{10}$ family of orthorhombic compounds exhibits a very peculiar evolution from a Kondo insulator ($T$: Fe) to an unconventional long-range magnetic order ($T$: Ru, Os). Inelastic neutron-scattering experiments performed on single-crystal $CeFe_2Al_{10}$ reveal that this material develops a spin gap in its magnetic spectral response below ∼50 K, with a magnetic excitation dispersing from $E = 10.2 \pm 0.5$ meV at the $Y$ zone-boundary point [$\boldsymbol{q} = (0,1,0)$] to ≈12 meV at the top of the branch. The excitation shows a pronounced polarization of the magnetic fluctuations along $a$, the easy anisotropy axis. Its behavior is contrasted with that of the (magnonlike) modes previously reported for $CeRu_2Al_{10}$, which have transverse character and exist only in the antiferromagnetic state. The present observation is ascribed to a "magnetic exciton" mechanism invoked to explain a similar magnetic response previously discovered in $YbB_{12}$.

DOI: 10.1103/PhysRevB.89.161103 PACS number(s): 71.27.+a, 75.30.Gw, 75.30.Mb, 78.70.Nx

Kondo insulators (KIs) form a unique class of materials, in which semiconducting properties develop on cooling as a result of the opening of a narrow gap in the electronic density of states at the Fermi energy [1]. One central issue to understanding the peculiar properties of these materials is the interplay between the nonmagnetic singlet ground state prevailing in most KI materials at low temperature, and the tendency to develop short-range, dynamical, antiferromagnetic (AF) correlations, revealed by inelastic neutron scattering (INS) measurements. In the archetype compound $YbB_{12}$, we have shown previously that the low-energy magnetic response in the KI state is dominated by a sharp, resolution-limited peak, located just below the edge of a spin gap, which disappears rapidly upon heating as the system enters the incoherent spin fluctuation regime [2]. This excitation was interpreted by Riseborough [3] as an exciton peak (a resonance mode in the spin response function) reflecting residual AF interactions between the renormalized $4f$ quasiparticles. This model is reminiscent of that proposed for the well-known "resonance mode" (RM) in high-$T_c$ superconductors [4]. A similar situation possibly occurs in $SmB_6$ as well [5]. On the other hand, evidence is still lacking for the existence of a RM-type excitation in the case of the Ce-based KIs, primarily because detailed studies of the $\boldsymbol{q}$ dependence of the magnetic response on single crystals are scarce [6].

In this Rapid Communication, we present new INS results showing that $CeFe_2Al_{10}$ is likely the first example of a Ce system with a RM excitation in the KI state. This compound belongs to the "1-2-10" series ($RT_2Al_{10}$ with $R$ a lanthanide element and $T$ a transition-metal element such as Fe, Ru, or Os). Unlike its Ru and Os counterparts, which have been extensively investigated for their unconventional combination of KI behavior and long-range magnetic order [7], $CeFe_2Al_{10}$ is a classical KI, in which no magnetic order was observed down to 40 mK. In contrast to cubic boride KIs, orthorhombic $CeFe_2Al_{10}$ is strongly anisotropic, with $\chi_a/\chi_b = 5.2$ at $T = 70$ K. At low temperature ($T = 7$ K), its magnetic spectral response was studied by inelastic neutron scattering (INS) experiments performed on powder [8]. The data showed the suppression of magnetic scattering below 5 meV, together with a pronounced peak at about 13 meV, which was taken to denote the energy of the spin gap. However, powder experiments cannot reveal where the signal occurs in $\boldsymbol{Q}$ space, nor whether or not it exhibits sizable dispersion. Because these questions are central to establishing the exact nature of the excitation, we have performed detailed measurements on a single crystal, whose results are reported in the following.

Fourteen single crystals of $CeFe_2Al_{10}$ (base-centered orthorhombic, $Cmcm$ space group, no. 63, $a = 9.0159$ Å, $b = 10.2419$ Å, $c = 9.0882$ Å [9]), for a total mass of about 700 mg, were grown by an Al-flux method and co-aligned on an Al sample holder. The total mosaic spread was about 3°, from the width of the neutron rocking curves, which was sufficient for the present experiment. Excitation spectra were measured in the ($a^*$,$b^*$) scattering plane on the 2-T triple-axis spectrometer (TAS) at LLB-Orphée (Saclay) at fixed final energy, $E_f = 14.7$ meV. A pyrolytic graphite (PG) 002 monochromator and analyzer were used, with a PG filter placed on the scattered beam. The instrumental resolution was ≈1.1 meV at the elastic position and ≈1.7 meV for $E = 10$ meV.

Constant-$\boldsymbol{Q}$ scans recorded at the base temperature, $T_{\min} \approx 4.8$ K, are presented in Fig. 1. For $\boldsymbol{Q} = (0, 3, 0)$ and $(-1, 2, 0)$, one sees a clear peak, centered at $E \approx 9.5$ meV, superimposed on a rather large sloping background. A survey of its intensity in $\boldsymbol{Q}$ space indicates that this signal is peaked near the reduced $\boldsymbol{q}$ vector (0, 1, 0) ($Y$ point in Brillouin

*jean-michel.mignot@cea.fr

†Present address: Quantum Condensed Matter Division, ORNL, Oak Ridge, Tennessee 37831, USA.

 

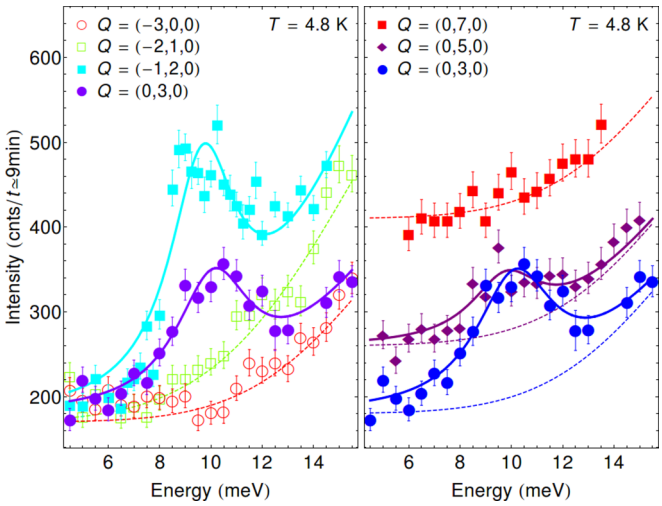


FIG. 1. (Color online) Energy scans at constant neutron final energy $E_f = 14.7$ meV, measured at $T = 4.8$ K for momentum transfers corresponding to the same reduced $\boldsymbol{q}$ vector (0,1,0) (zone boundary $Y$ point). The left frame demonstrates the dependence of the inelastic peak intensity on the direction of $\boldsymbol{Q}$ with respect to the $a$ axis (dipole selection rule). The right frame shows the rapid suppression of the peak with increasing momentum transfer. Dashed lines: estimated background (see text). Solid lines: fits assuming Lorentzian line shapes.

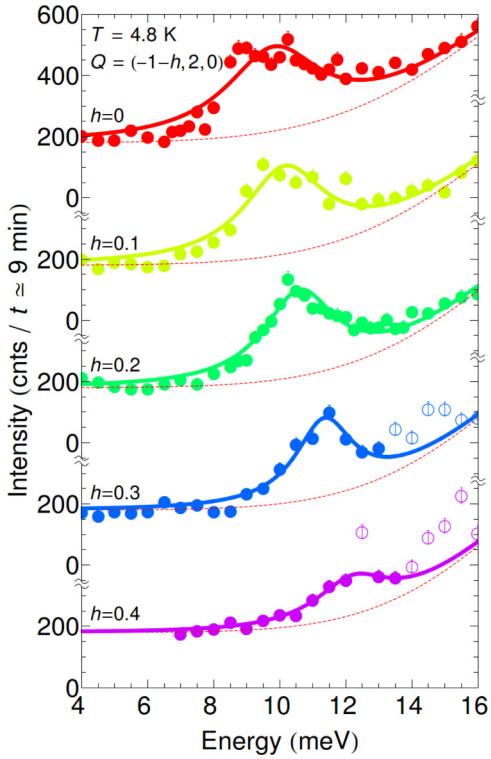


FIG. 2. (Color online) Energy scans at constant neutron final energy $E_f = 14.7$ meV, measured at $T = 4.8$ K for momentum transfers $\boldsymbol{Q} = (-1-h,2,0)$, showing the dispersion of the magnetic excitation and the $\boldsymbol{Q}$ dependence of its intensity along the $(h,0,0)$ direction. Dashed lines: estimated background (see text). Solid lines: fits assuming Lorentzian line shapes; open symbols denote data points thought to contain a contamination and thus excluded from the fit.

zone), which corresponds to the wave vector of the AF order previously reported for $CeRu_2Al_{10}$ or $CeOs_2Al_{10}$ [10,11].

Measurements at larger scattering vectors, for $\boldsymbol{Q} = (0, 5, 0)$ and $(0, 7, 0)$ (Fig. 1, right), show that the peak intensity is rapidly suppressed with increasing momentum transfer, as expected for magnetic scattering. This magnetic origin is further supported by the temperature dependence discussed hereafter. We recall that, in our previous study of $CeRu_2Al_{10}$, a signal ascribed to phonons was observed near 20 meV in the powder experiments [11], and neutron polarization analysis showed that the scattering below 10 meV was purely magnetic [14]. The same should apply to the present compound. Actually, the sloping background, which increases significantly at larger $\boldsymbol{Q}$ values, may correspond to the tail of the phonon component. This general pattern is qualitatively consistent with the powder results of Ref. [8], showing an excitation at about 13 meV, partly overlapping the low-energy tail of a broad peak centered near 50 meV, and whose origin was also concluded to be magnetic.

The difference between the energy of the peak in the present experiment and in the powder-averaged data suggests that the excitation may show some dispersion. To check this assumption, scans have been performed along several high-symmetry directions in the $(a^*,b^*)$ scattering plane. The spectra exhibit a positive dispersion starting from the $Y$ points $\boldsymbol{Q} = (0, 3, 0)$ and $(-1, 2, 0)$ (see, e.g., Fig. 2). Meanwhile, the intensity decreases rapidly and the peak becomes barely detectable when $\boldsymbol{q}$ departs from the $Y$ point by more than 0.4 r.l.u. in any direction.

To derive the $\boldsymbol{Q}$ dependence of the excitation energy, linewidth, and integrated intensity, the data were fitted assuming a Lorentzian line shape (detailed balance factor corrections are insignificant at this temperature for the energy range of interest) [12]. The main source of uncertainty is the determination of the background, especially at the high-energy end of the scan where some contamination seems to exist [13]. This background is comparably steep for all $\boldsymbol{Q}$ vectors investigated, but not strictly identical. It was therefore estimated from a comparison of low- and high-temperature spectra for the same $\boldsymbol{Q}$ vector, assuming the latter to consist of a single broad quasielastic (QE) contribution varying slowly in $\boldsymbol{Q}$ space (see below).

The results are plotted in Fig. 3, where the error bars account for the above-mentioned uncertainties. Starting from a minimum slightly in excess of 10 meV at the $Y$ point, the energy reaches about 12–13 meV near the top of the branch. This dispersion is unambiguous, despite the substantial broadening, because the peak shape remains practically unchanged in the range of $h$ over which most of the energy change takes place. It is, however, less pronounced than in the AF phase of $CeRu_2Al_{10}$ [14]. The upper part of the dispersion accounts for the excitation energy of 13 meV observed in the powder spectra of Ref. [8] likely because of the minor weight, in the $\boldsymbol{Q}$-space average, of the region close to the $Y$ point where the minimum is located. Within experimental accuracy, the present dispersion appears to be similar along the three symmetry directions investigated [$(h,0,0)$ and $(0,k,0)$ in Fig. 3, $(h,-h,0)$ not shown]. The width of the peak (full width at half

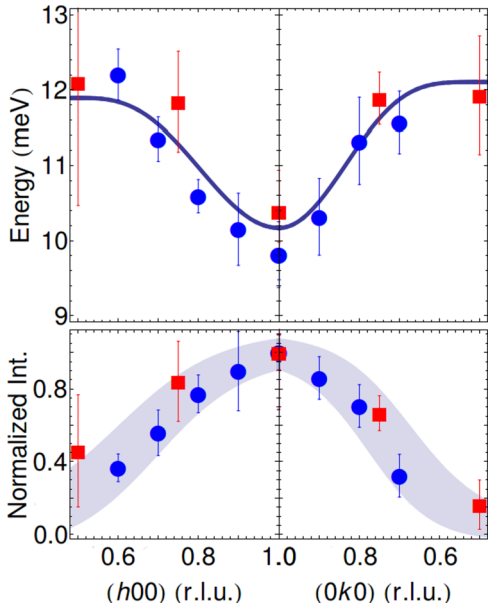


FIG. 3. (Color online) Experimental dispersion curve and $\boldsymbol{Q}$ dependence of the intensity of the magnetic excitation at $T \approx 4$ K. Red squares [for $\boldsymbol{Q} = (1-h,3,0)$ and $(0,2+k,0)$] and blue circles [for $\boldsymbol{Q} = (-2+h,2,0)$ and $(-1,3-k,0)$] correspond to spectra measured along equivalent, yet distinct, $(h,0,0)$ (left frame) and $(0,k,0)$ (right frame) directions in reciprocal space.

maximum of about $4.0 \pm 0.7$ meV), does not vary significantly between the different $\boldsymbol{Q}$ vectors at which it was measured, and remains much larger than that expected from the experimental resolution (1.7 meV at the peak energy). The asymmetric shape of the $(-1, 2, 0)$ peak, after subtracting the background (Fig. 4, left frame), could indicate a doublet of excitations but, in view of the insufficient statistics and the possibility of contaminations, this possibility could not be confirmed.

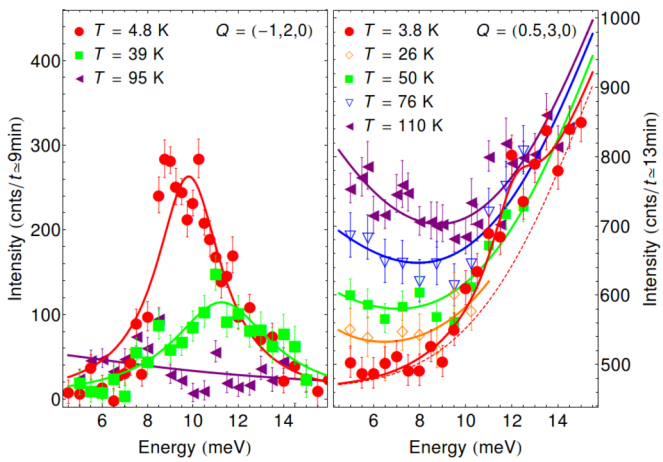


FIG. 4. (Color online) Temperature evolution of the magnetic spectral response measured at constant neutron final energy Ef = 14.7 meV for $\boldsymbol{Q} = (-1,2,0)$ (left) and (0.5,3,0), showing the rapid suppression of the inelastic peak and the gradual appearance of the quasielastic signal as $T$ increases. In order to improve statistics, the scans denoted 50, 95, and 110 K were obtained by combining data measured at 45 and 55 K, 80 and 110 K, 100 and 120 K, respectively.

The temperature dependence of the excitation, measured for $\boldsymbol{Q} = (-1, 2, 0)$, is plotted in Fig. 4 (left). The peak intensity decreases rapidly on heating to 40 K, and vanishes at 80 and 110 K. Meanwhile, intensity appears gradually at low energy [right frame, for $\boldsymbol{Q} = (0.5, 3, 0)$, away from the $Y$ point], reflecting the growth of QE fluctuations. This signal can be fitted to a Lorentzian peak, with a half width at half maximum of about 5 meV showing little temperature dependence between 45 and 120 K.

It is interesting to note that, at $T = 40$ K, the inelastic peak is strongly suppressed without shifting to lower energy (the data even suggest a slight increase in the excitation energy but this observation needs to be confirmed). The appearance of a signal below 8 meV for $T > 50$ K is therefore ascribed to a filling, rather than a closing, of the spin gap. This behavior is at variance with that reported for $CeRu_2Al_{10}$ [10,11,15], in which the energy of the magnonlike excitation at the spin-gap edge decreases as $T$ increases toward the Néel temperature $T_0$ in the AFM phase. On the other hand, it is reminiscent of the temperature suppression of the magnetic exciton mode in $YbB_{12}$ [2,16].

Information on the polarization of the AF fluctuations can be obtained from the $\boldsymbol{Q}$ dependence of the dynamic response, using the dipole selection rule for magnetic neutron scattering, which stipulates that only magnetic components perpendicular to the scattering vector contribute. Here the comparison of the spectra for $\boldsymbol{Q} = (-1, 2, 0)$ and $(0, 3, 0)$ on the one hand, $\boldsymbol{Q} = (-2, 1, 0)$ and $(3, 0, 0)$ on the other hand, clearly shows that the peak is suppressed (or strongly reduced) when the scattering vector is oriented along (or close to) the $a^*$ direction (Fig. 1, left frame). This implies that the signal mainly arises from correlations $\langle m_i^a m_j^a \rangle$ between magnetic components parallel to $a$, which is also the easy axis observed in static magnetization measurements [7].

In the QE regime, the signal at low energy is observed with sizable intensity at various momentum transfers, irrespective of their orientation in $\boldsymbol{Q}$ space [at least within the experimental (001) scattering plane]. This is taken to indicate that the predominance of magnetic correlations along $a$ observed at $T = 5$ K is a genuine feature of the KI state, which does not extend into the incoherent relaxational regime.

The present study shows that the low-temperature spin dynamics in $CeFe_2Al_{10}$ differs markedly from that observed previously in $CeRu_2Al_{10}$. In the latter compound, the spin-gap response, associated with well-defined magnetic excitation branches, was essentially restricted to the AF phase, and could be explained using a RPA model for anisotropic AF magnons [14]. In the present case, on the other hand, the dispersive mode exists *in the absence of long-range magnetic order*, and its appearance rather seems to correlate with the low-temperature upturn in the resistivity $\rho_{\|a}$ below ~50 K, which signals the entry into the KI regime. This point is important because, despite undisputed evidence for a long-range AF ordered phase in $CeRu_2Al_{10}$ (as well as $CeOs_2Al_{10}$) below $T_0$, there is ongoing controversy as to whether the spin gap in those systems should be interpreted as the anisotropy gap of magnonlike excitations, or in terms of a singlet-triplet level scheme due to a spin-dimer or charge-density-wave transition occurring somewhat above $T_0$ [8].

Since the AF correlations giving rise to the inelastic peak in $CeFe_2Al_{10}$ are centered at the same $\boldsymbol{k}$ vector where the AF structure develops in $CeRu_2Al_{10}$, they could be regarded as a precursor effect of the long-range order, which could develop, e.g., under application of a negative pressure. However, the QE signal observed in $CeRu_2Al_{10}$ over a limited temperature interval above $T_0$ exhibits both a $\boldsymbol{Q}$-dependent intensity, peaked around $\boldsymbol{q}_{\rm AF}$, and correlations polarized along $a$ [15], whereas the QE response in $CeFe_2Al_{10}$ is widely spread in $\boldsymbol{Q}$ space and shows no evidence of mode polarization. When temperature is lowered below 50 K, this local paramagnetic relaxation regime evolves toward the appearance of the spin gap and the inelastic mode, contrasting with $CeRu_2Al_{10}$ in which AF critical fluctuations develop on approaching $T_0$.

Magnetic anisotropy is known to play a major role in 1-2-10 compounds, because of their particular crystal structure. The model used to describe the excitations in $CeRu_2Al_{10}$, and even to account for the orientation of the ordered AF moments below $T_0$, actually requires strongly anisotropic *exchange constants*. However, the mode polarization found here for $CeFe_2Al_{10}$ is likely dominated by the *single-ion* (crystal-field) anisotropy and gives no indication as to the exchange interactions.

From the above results, the spin dynamics in $CeFe_2Al_{10}$ is seen to bear close similarities with the "exciton" response previously reported for $YbB_{12}$. In both systems a specific magnetic excitation develops in the KI regime, whose position is close to the spin-gap edge. The mode exhibits a positive dispersion from a zone-boundary $\boldsymbol{q}$ vector, with intensity decreasing rapidly as the energy comes closer to the continuum. With increasing temperature, the peak is rapidly suppressed but does not appear to move significantly to lower energies, as was already noted for $YbB_{12}$ [16]. In Riseborough's model [3], the exciton peak requires both a sizable exchange interaction $J(\boldsymbol{q}_{\rm AF})$ and a large value of the magnetic susceptibility $\chi_0(\boldsymbol{q}_{\rm AF},E)$. The latter quantity, associated with particle-hole excitations, is specifically enhanced for a $\boldsymbol{q}$ vector corresponding to that of the (indirect) hybridization gap, because Kramers-Kronig relation produces a peak in the real part of $\chi_0(\boldsymbol{q}_{\rm AF},E)$ at the spin-gap edge, where the imaginary part rises steeply. An important consequence of this description is that the peak energy can no longer be identified with the spin-gap value, as is often done in experimental studies of KIs [6], since the RM is essentially an in-gap resonance. In practice, however, the edge of the electron-hole continuum may be difficult to ascertain from the measured spectra. Unlike $YbB_{12}$ where the exciton peak was found to be resolution limited [2], $CeFe_2Al_{10}$ exhibits sizable broadening of the inelastic peak, which is too large to be accounted for by experimental effects (e.g., mosaic spread in the crystal). This suggests that the damping is intrinsic and reflects a finite lifetime of the excitation. A possible origin for this effect is the residual density of state at the Fermi energy at 10 K inferred from the Drude weight remaining below 10 meV in the optical conductivity experiments [17].

CeNiSn is one of the few KI compounds for which detailed single-crystal INS data exist. It is orthorhombic, like $CeFe_2Al_{10}$, but has a smaller gap ($\Delta \approx 6$ K) and a lower coherence temperature ($T_{\rm coh} \approx 12$–$20$ K). Two magnetic excitations have been observed in the low-temperature regime for $\boldsymbol{Q} = (Q_a, 1/2 + n, Q_c)$ ($E = 4$ meV) and for $\boldsymbol{Q} = (0, 1, 0)$ and $(0, 0, 1)$ ($E = 2$ meV), with $n$ integer and $Q_a$ and $Q_c$ arbitrary [18]. Cold-neutron TAS experiments [19] have shown that the magnetic spectral response has a pseudogap, rather than a true gap, for $\boldsymbol{Q} = (0, 1, 0)$, since a sizable QE signal remains down to $T = 1.5$ K. This QE signal is $\boldsymbol{Q}$ independent, and therefore similar to that observed in $CeFe_2Al_{10}$ above 50 K. The excitation at 2 meV has three-dimensional character and a pronounced polarization along the easy $a$ axis, like in the present case, and it disappears upon heating to 20 K. On the other had, no evidence was reported for a dispersion of that mode, which appears to be strongly peaked near the AF $\boldsymbol{q}$ vector. In Riseborough's model, some intermediate valence character of the material is considered favorable for stabilizing the in-gap mode, whereas CeNiSn is better categorized as a Kondo compound. The question whether the excitation can be ascribed to a spin-exciton mechanism thus remains open.

In summary, the present experiments reveal an unexpected $\boldsymbol{Q}$ dependence in the spin dynamics of $CeFe_2Al_{10}$ in the KI state. The inelastic peak is found to exhibit sizable dispersion but relatively weak anisotropy with respect to the direction of $\boldsymbol{q}$ in the $(a^*,b^*)$ scattering plane. This excitation presents strong similarities with the "resonance mode" previously observed in the Kondo insulator $YbB_{12}$, and is therefore thought to arise from the same type of spin-exciton mechanism, originally proposed by Riseborough. However, in contrast to the latter cubic system, the AF fluctuations involved in this process exhibit a pronounced polarization along the $a$ axis. Application of the spin-exciton model to $CeFe_2Al_{10}$ will thus require proper treatment of the lower symmetry in 1-2-10 compounds.

We thank B. Annighöfer for designing the sample holder and D. Adroja for providing Cytop glue. P.A.A.'s participation in this work was partly funded by RFBR Grant No. 14-02 00272-a, and by the French Ministry of Foreign Affairs (*Séjours scientifiques de haut niveau*).

[1] P. S. Riseborough, Adv. Phys. **49**, 257 (2000).

[2] J.-M. Mignot, P. A. Alekseev, K. S. Nemkovski, L.-P. Regnault, F. Iga, and T. Takabatake, Phys. Rev. Lett. **94**, 247204 (2005); K. S. Nemkovski, J.-M. Mignot, P. A. Alekseev, A. S. Ivanov, E. V. Nefeodova, A. V. Rybina, L.-P. Regnault, F. Iga, and T. Takabatake, *ibid.* **99**, 137204 (2007).

[3] P. S. Riseborough, J. Magn. Magn. Mater. **226-230**, 127 (2001).

[4] See, e.g., M. Eschrig, Adv. Phys. **55**, 47 (2006), and references therein.

[5] P. Alekseev, J.-M. Mignot, J. Rossat-Mignod, V. Lazukov, I. Sadikov, E. Konovalova, and Y. Paderno, J. Phys.: Condens. Matter **7**, 289 (1995).

[6] D. T. Adroja, K. A. McEwen, J. G. Park, A. D. Hillier, N. Takeda, P. S. Riseborough, and T. Takabatake, J. Optoelectron. Adv. Mater. **10**, 1564 (2008).

[7] Y. Muro, K. Motoya, Y. Saiga, and T. Takabatake, J. Phys. Soc. Jpn. **78**, 083707 (2009); T. Nishioka, Y. Kawamura, T. Takesaka, R. Kobayashi, H. Kato, M. Matsumura, K. Kodama, K. Matsubayashi, and Y. Uwatoko, *ibid.* **78**, 123705 (2009); A. M. Strydom, Physica B **404**, 2981 (2009).

[8] D. T. Adroja, A. D. Hillier, Y. Muro, J. Kajino, T. Takabatake, P. Peratheepan, A. M. Strydom, P. P. Deen, F. Demmel, J. R. Stewart, J. W. Taylor, R. I. Smith, S. Ramos, and M. A. Adams, Phys. Rev. B **87**, 224415 (2013).

[9] M. Sera, D. Tanaka, H. Tanida, C. Moriyoshi, M. Ogawa, Y. Kuroiwa, T. Nishioka, M. Matsumura, J. Kim, S. Tsuji, and M. Takata, J. Phys. Soc. Jpn. **82**, 024603 (2013).

[10] D. D. Khalyavin, A. D. Hillier, D. T. Adroja, A. M. Strydom, P. Manuel, L. C. Chapon, P. Peratheepan, K. Knight, P. Deen, C. Ritter, Y. Muro, and T. Takabatake, Phys. Rev. B **82**, 100405 (2010).

[11] J. Robert, J.-M. Mignot, G. André, T. Nishioka, R. Kobayashi, M. Matsumura, H. Tanida, D. Tanaka, and M. Sera, Phys. Rev. B **82**, 100404 (2010); J.-M. Mignot, J. Robert, G. André, A. M. Bataille, T. Nishioka, R. Kobayashi, M. Matsumura, H. Tanida, D. Tanaka, and M. Sera, J. Phys. Soc. Jpn. Suppl. **80**, SA022 (2011).

[12] Results obtained when using Gaussian fits do not differ qualitatively from those presented here.

[13] High orders of the Bragg reflections on the monochromator and analyzer are known to cause extra scattering for $E = 18.35$ meV ($2k_i = 3k_f$). That signal may appear shifted in energy, in the case of coherent scattering, because of resolution effects.

[14] J. Robert, J.-M. Mignot, S. Petit, P. Steffens, T. Nishioka, R. Kobayashi, M. Matsumura, H. Tanida, D. Tanaka, and M. Sera, Phys. Rev. Lett. **109**, 267208 (2012).

[15] J.-M. Mignot, J. Robert, S. Petit, K. Saito, T. Nishioka, R. Kobayashi, M. Matsumura, H. Tanida, D. Tanaka, and M. Sera (unpublished).

[16] K. S. Nemkovski, P. A. Alekseev, J.-M. Mignot, and A. S. Ivanov, Phys. Proc. **42**, 18 (2013).

[17] S.-i. Kimura, Y. Muro, and T. Takabatake, J. Phys. Soc. Jpn. Suppl. **80**, 033702 (2011).

[18] T. J. Sato, H. Kadowaki, H. Yoshizawa, T. Ekino, T. Takabatake, H. Fujii, L.-P. Regnault, and Y. Isikawa, J. Phys.: Condens. Matter **7**, 8009 (1995), and references therein.

[19] S. Raymond, A. P. Murani, B. Fak, L.-P. Regnault, G. Lapertot, P. A. Alekseev, and J. Flouquet, J. Magn. Magn. Mater. **190**, 245 (1998).