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PERTURBED ANGULAR CORRELATION GAMMA-GAMMA SPECTROSCOPY WITH ^{140}Ce ISOTOPE IN $CeMn_2Ge_2$

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ABSTRACT

In this work we explore some properties of the nuclear transitions of the ^{140}Ce radioactive isotope in order to measure the magnetic field acting on the nuclei of cerium atoms in the $CeMn_2Ge_2$ compound with perturbed angular correlation spectroscopy. Since the magnetic field arises from the electronic cloud around the nucleus, it is possible to infer about the electronic mechanisms responsible for the appearance of magnetism in this compound. The results were analyzed with the aid of first principles electronic structure calculations on the $CeMn_2Ge_2$ compound and, after comparison with the experiment, it was found that the value of the consistent magnetic hyperfine field is such that the Ce electronic 4f ground state in this material is $J_z = 1/2$. This value for the Ce 4f orbital state is attributed to the splitting of the J multiplet caused by the crystalline electric field with tetragonal symmetry formed by the atoms around Ce in $CeMn_2Ge_2$.

1. INTRODUCTION

Magnetic hyperfine fields (MHF) acting on atomic nuclei are intimately related with the electronic structure of solids or molecules and nuclear techniques such as perturbed angular correlation (PAC) spectroscopy are useful tools to study electronic properties related with magnetism.

The $CeMn_2Ge_2$ compound belongs to a family represented generically by RT_2X_2 where R stands for rare-earth, T for transition metal and X for a *sp* element. Depending on composition, these compounds present different properties such as magnetism, superconductivity, mixed valence, heavy fermion and Kondo behavior. Only in systems when T=Mn, the T atoms order magnetically at high temperatures and, moreover, with R=Ce, the magnetic moments appear exclusively on Mn sites: the Ce 4f moment remain quenched even at the lowest temperatures.

In contrast, PAC measurements performed on the ^{140}Ce in $CeMn_2Ge_2$ revealed the existence of a large magnetic hyperfine field of 39.0(1.4) T at 10 K with a clear indication of Ce magnetic ordering below temperatures of the order of T = 120 K. In a previous work [1], by means of first-principles electronic structure calculations, it was shown that the Ce atoms in $CeMn_2Ge_2$ can not be considered as non magnetic since the small or absent magnetic moment is actually composed of spin and angular components which nearly cancels each other but the

same cancellation does not occur with the magnetic hyperfine field. In [1], the Ce 4f electrons were treated as band states and the generalized gradient approximation (GGA) [2] within density functional theory (DFT) formalism was adopted, an approximation that was argued as responsible for the inconsistency between the measured and calculated values for the Ce MHF. In the present work we extended the calculation for the case in which the 4f Ce states are treated as localized within the GGA+U approach [3]. It was found that the resulting MHF agrees quite well with the experimental value, the spin and orbital components of the Ce magnetic moment cancel each other completely and that the Ce ground state at 10 K can be represented by $S_z = -1/2$, $L_z = 1$, resulting $J_z = 1/2$.

2. DETAILS OF THE PAC EXPERIMENT

When a nucleus undergoes a transition by emission of a gamma ray, the direction of the ongoing radiation is correlated with the direction of the nuclear spin. For example, in electric dipole transitions, the preferred radiation direction is parallel to the nuclear spin. Moreover, when the nucleus possesses a magnetic moment in the presence of a magnetic field it processes with a Larmor frequency given by $\omega_L = -2\pi g \mu_N B/h$ where the terms are the nuclear giromagnetic ratio, nuclear magneton, magnetic induction and the Planck constant. These properties are exploited in perturbed angular correlation spectroscopy where the Larmor frequency is measured and the magnetic induction is thus derived [4].

In figure 1 it is presented part of the β^- decay scheme of the ^{140}La as well as the basic setup in the PAC experiments. The radioactive ^{140}La isotope is formed by irradiating natural lanthanum with neutrons in the IEA-R1 reactor [5]. A small proportion (0.1 atom percent) is then melted with the $CeMn_2Ge_2$ sample at the time of preparation that, due to chemical affinity, occupies the Ce positions in the compound. After the lanthanum decay, the as formed cerium atom makes part of the ordinary $CeMn_2Ge_2$ compound although it is radioactive. The gamma rays γ_1 and γ_2, with energies 329 and 487 keV, respectively, from the gamma cascade shown in figure 1a are detected by two detectors as shown in figure 1b and the time between the emission of the two radiations is registered. For example, if the detector 1 detects γ_1, then it acts as a start for the time interval counting until γ_2 is detected by the second detector.

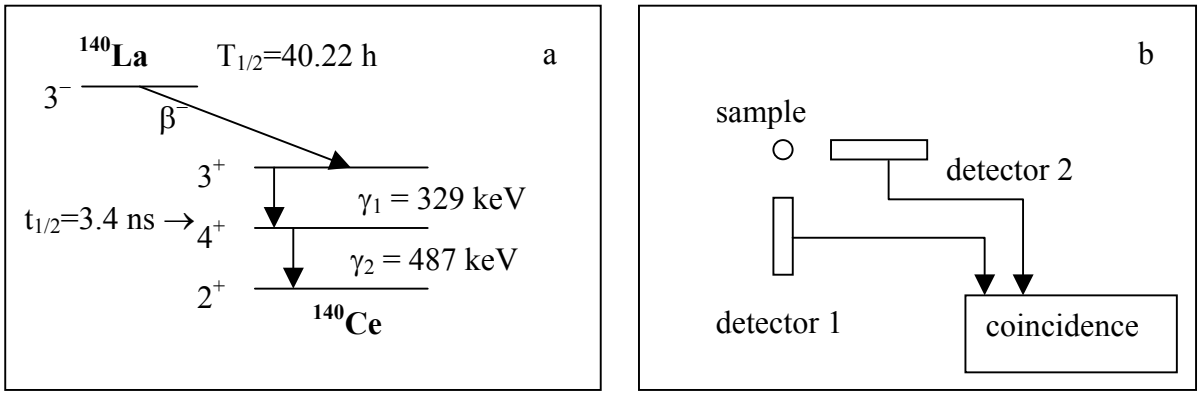

Figure 1. Portion of a ^{140}La - ^{140}Ce decay scheme (a) and the basic PAC experimetal setup (b)

During this time the Ce nucleus is in its intermediate $I=4^+$ state with 3.4 ns half life and interacts with the external magnetic hyperfine field. This interaction result in the precession of the nucleus and continuously produces transitions between the azimutal states of the Ce nuclear level which affects the probability of the emission of the gamma ray γ_2.

As a result, a decay spectrum, given by the number of counts as a function of time is observed whose half life is the same as that of the intermediate level, $T_{1/2}$ = 3.4 ns. The spectrum appears modulated by oscillations with frequencies that correspond to the several allowed transitions of the nuclear sub-levels $\omega=2\pi\Delta E/h$, where ΔE is the energy difference between them. The lower frequency represents the Larmor frequency.

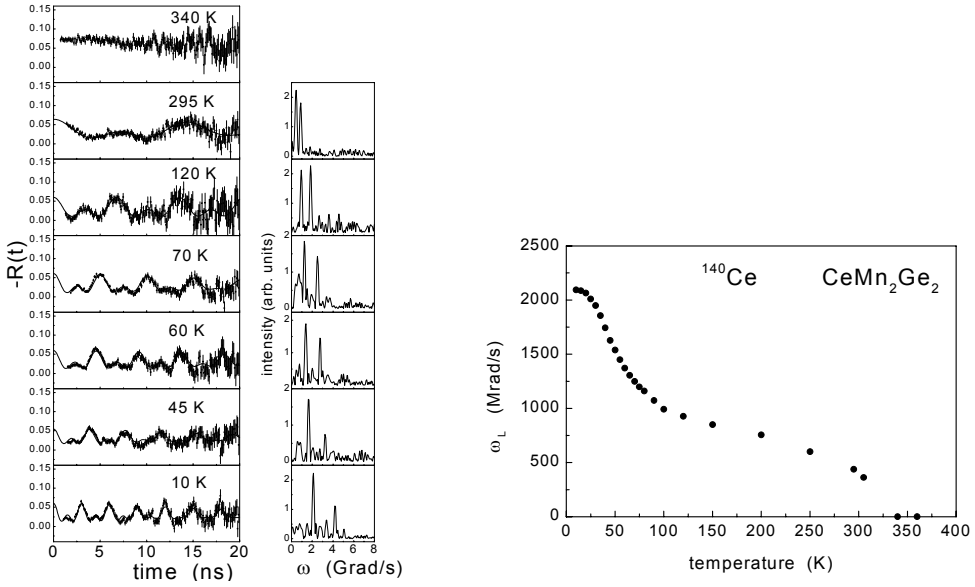

Figure 2. PAC spectra of ^{140}Ce on $CeMn_2Ge_2$ for several temperatures, along with its Fourier transform (left). Larmor frequency as a function of temperature (right)

The experiments are performed with a four BaF_2 detector setup where each detector can act as a start or a stop detector. The detectors are displaced around the sample, forming an angle of 90 degrees between them. By dedicated electronics, twelve spectra are recorded at a time: 4 with detectors placed at 180 degrees and 8 at 90 degrees. A geometric average is performed for each class of spectra after subtracting the background due to spurious coincidences. Finally, a proper combination of the averaged 180 and 90 degrees spectra is performed and the final spin rotation spectrum R(t) is generated. The resulted PAC spectra are shown in figure 2 for several temperatures along with the temperature dependence of the Larmor frequency [4].

3. FIRST PRINCIPLES ELECTRONIC STRUCTURE CALCULATIONS

The method employed for the electronic structure determination of $CeMn_2Ge_2$ with the aim to derive the magnetic hyperfine field acting on Ce atoms is based on density functional theory.

This is done by finding the spin densities that minimize the total energy given as a functional of the electronic density ρ:

$$E[\rho] = T_s[\rho] + E_{ei}[\rho] + E_H[\rho] + E_{ii}[\rho] + E_{xc}[\rho]$$

Actually the spin densities $[\rho] \rightarrow [\rho\uparrow , \rho\downarrow]$ are utilized in this work The terms in this expression represent the single particle kinetic energy, the Coulomb interaction between electrons and nuclei, the interaction between the nuclei, the Hartree component of the electron-electron interaction and the exchange-correlation energy. The exchange and correlation energy is approximated locally as the corresponding energy of a uniform electron gas (local density approximation, LDA). This approximation is too simplified to take into account the large correlation effects that exists within the rare-earth 4f shell even with the improved generalized gradient approximation (GGA) method [1]. For this reason we extended the calculations by employing the GGA+U method which introduces a potential that depends on the occupation of an orbital [3]. The later approach however loses the first-principles character of the method since a parameter U has to be inserted in the methodology because it can not be obtained through the variational principle. The spin-orbit interaction is taken into account by employing a second variational method using the Kohn-Sham scalar relativistic orbitals as basis.

4. RESULTS AND DISCUSSION

In the right of figure 2 one observe that the dependency of the MHF (a quantity proportional to the Larmor frequency shown in the graph) upon the temperature is composed of two regimes. The first one, represented by the increase from zero MHF (at T = 320 K) on decrease of temperature is the normal Brillouin behavior on magnetic transitions which is expected to saturate to a constant value down to low temperatures since its origin is the transferred MHF from the manganese atoms. Indeed, it is known that the manganese contribution follows the Brillouin curve [1]. The second step in the curve is attributed to a MHF contributed from the cerium atoms.

In table 1 it is shown the results of the calculations. Within the GGA approach, it can be seen that the Ce 4f state by itself contributes largely (-28.2 T) to the MHF in agreement with the experiment as depicted in figure 2 (right). Also evident is the resulted small contribution to the total magnetic moment of Ce due to the partial cancellation from the orbital (-0.54) and spin (0.70) contributions, a fact that has also been observed experimentally from neutron diffraction studies [1]. On the other hand, the agreement between the experimental MHF (39.0(1.4) T[1]) and the magnitude of the calculated value (15.7) is poor.

Within the GGA+U approach it is possible to study the variations of the MHF contributions by varying the z component of the Ce 4f angular momentum as the methodology no longer satisfy the variational principle and thus the algorithm converges with the initial value of L_z. In table 1 it is seen that the Fermi contact contribution to the Ce MHF does not change very much as a function of L_z but the orbital and dipolar contributions do it with a very predictable way. The orbital contribution is proportional to L_z while the dipolar contribution, besides being large (~18 T), depends on the relative orientation of the 4f orbit: it is maximum positive

[1] Only the magnitude of MHF is known experimentaly [4].

when **L** is (anti) parallel to z (L_z = -3), maximum negative when **L** is perpendicular to z (L_z = 0), and is zero somewhere between these two extremes (L_z ~ -2). This behavior is predicted by the MHF formulas given by Blügel et. al. [6]

Table 1. Calculated Ce MHF contributions in $CeMn_2Ge_2$ within the GGA and GGA+U approximations along with the resulted values of the z components of the 4f orbital angular moment, spin magnetic moment and total energy of the alloy.

	GGA[a]	GGA + U[b]			
L_z	-0.54	-3	-2	-1	0
MHF(Contact)	13.3	14.0	14.6	15.05	14.6
MHF(Dipolar)	-0.855	18.2	-1.98	-14.2	-17.4
MHF(Orbital)	-28.2	-156.8	-105.3	-52.6	-0.102
MHF(Total)	-15.7	-124.6	-92.7	-51.7	-2.90
μ [*]	0.70	1.12	1.11	1.10	1.10
Energy [**]	0.0	0.100728	0.078606	0.08427	0.08894

The magnetic hyperfine fields are given in T. The experimental value is 39.0(1.4) T [4]. Only the magnitude of MHF was determined experimentally

[a] Values taken from reference [1]

[b] Present work

[*] Total Ce spin magnetic moment in Bohr magnetons. Orbital contribution not taken into account.

[**] In Ry, relative to the energy from GGA calculations.

Comparing the results for total MHF with the experiment it can be seen that the best agreement is for the case when GGA+U is applied with L_z = -1, while the second best agreement is achieved with the GGA case. It is known that DFT underestimates the magnitude of the Fermi contact contribution to the MHF [7]. Thus the calculated values presented in table 1 are expected to be larger for the contact contribution. In this case the total MHF from the GGA approach would be worse in comparison with the experiment while for the case with the GGA+U functional with L_z = -1, the agreement would be even better. The MHF values for the other two contributions, namely, dipolar and orbital, are expected to be more accurate because they depend only on the localized nature of the Ce 4f state, an atomic property in this case, well represented by the GGA+U approach and nicely following the expected behavior as a function of L_z.

This result implies that the Ce 4f state in $CeMn_2Ge_2$ compound is such that L_z = -1 and S_z = 1/2 resulting J_z = 1/2 and, with $\mu_z = \mu_B \bullet (L_z + 2S_z)$, it results for a zero value for the z component of the Ce 4f total magnetic moment. The resulting spin magnetic moment seen in table 1 in all GGA+U cases is ~ 1.1 μ_B. Since 1.0 μ_B comes from the 4f spin contribution the remainder 0.1 μ_B is due to the Ce 5d state and would also be the total Ce magnetic moment when the orbital moment is taken into account. The small resulting magnetic moment is therefore consistent with the neutron diffraction studies in this compound.

The crystalline electric field with tetragonal symmetry around Ce ions splits the $^2F_{5/2}$ level into three doublets ±1/2, ±3/2 and ±5/2 and when $T_K < T_{CF}$ (T_K = Kondo temperature, T_{CF} = crystal field splitting), only the lowest doublet J_z = 1/2 effectively operate at low temperatures [8]. Our findings seems to corroborate that this is the case for the $CeMn_2Ge_2$ compound.

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