# Detection of excess electron spins in magnetic substances using highly spin-polarized positrons

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We are currently developing a new utilization of the positron annihilation spectroscopy (PAS) in order to investigate magnetic materials and spin-related devices.

Generally, PAS technique is a powerful tool to detect atomic vacancies in crystalline solids. Positron annihilation lifetime and the Doppler effect on annihilation radiation energy spectrum are the observables in PAS measurement. The former is inversely proportional to electron density where positrons annihilate. The latter reflects electron momentum distribution. Very recently, we have successfully demonstrated a new feasibility of PAS using our built beam source of highly spin-polarized positron. Electron-positron annihilation characteristics depend on the spin state. A spin-singlet (*S*=0) electron-positron pair tends to self-annihilate (two-photon annihilation). The self-annihilation (three-photon annihilation) probability of a spin-triplet (*S*=1) electron-positron pair is much smaller than the pick-off annihilation probability with surrounding electrons. The above spin-dependent positron annihilation can be observed using spin-polarized positrons. That is, polarized electrons can be detected using spin-polarized positrons. This is the principle of spin-polarized positron annihilation spectroscopy (SP-PAS) [1].

Until now, the chief difficulty in SP-PAS is to obtain highly spin-polarized positrons. In this study, we have cleared up this difficulty to use a $^{68}$Ge-$^{68}$Ga positron source that yields much highly spin-polarized positrons than the conventional $^{22}$Na source [2]. To examine the feasibility of SP-PAS to apply any magnetic materials for the spin-electronic devices, we have performed the SP-PAS measurements of high quality for the simple ferromagnets (Fe, Co, Ni, and Gd). Figure 1 shows the differential Doppler broadening of annihilation radiation (DBAR) spectra between positive and negative magnetic field with respect to positron spin direction obtained for polycrystalline Fe, Co, Ni and Gd. The residuals are not zero. This is called field-reversal asymmetry.

Roughly speaking, the field-reversal asymmetry appears due to enhanced annihilation between spin-up positrons and spin-down 3*d*, 4*s* (Fe, Co, Ni) and 4*f*, 5*d*, 6*s* (Gd) unpaired electrons. The field-reversal asymmetry of the Fe sample is the strongest, while it is slightly weaker for the Co sample, and only a small effect is observed for the Ni and Gd samples. Actually, the experimental results are well-reproduced by the first principles calculation considering polarization of electrons and positrons as shown by the solid lines.

The relative amplitude of the differential DBAR spectra of the Fe, Co and Ni samples seems to coincide with the trend of effective magnetization of these metals. If the area intensity of the Fe sample is normalized to 2.2, those of Co and samples are 1.8 and 0.4. These are comparable to the effective magnetizations of these materials. This result indicates that the magnetization of a magnetic substance can be estimated from the field reversal asymmetry of DBAR spectrum using a reference sample with known magnetization. Considering the fact that the electron polarization of Fe is at most about 40 % near the Fermi level, the determination of electron polarizations of half-metals by the present method is feasible.

In SP-PAS experiment, polarized electrons are directly detected through annihilation with polarized positrons. This is an important feature for the investigation of polarized electron states. The further advantage of SP-PAS is the depth selectivity by employing monochromatic positron beams. Some important magnetic effects, such as giant magnetoresistance and tunneling magnetoresistance, occur near the interface between magnetic and non-magnetic layers. Spin-injection electrodes, which will be used in spin devices, are normally thin films. Novel spin phenomena such as the spin Hall effect and the giant Rashba effect occur near surfaces. These are potential applications of SP-PAS. Taking advantage that PAS is a powerful tool to detect vacancy defects, SP-PAS might be used in studying vacancy-induced magnetism.

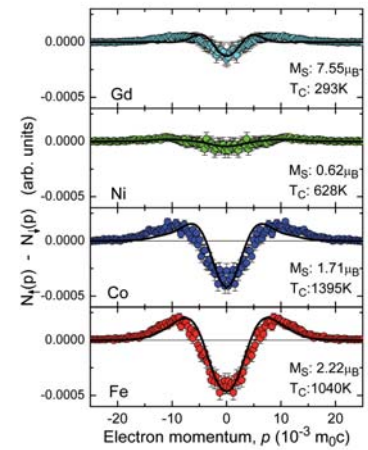


Fig. 1 Differential DBAR spectra of the Fe, Co, Ni and Gd samples obtained in the external magnetic field of 1 T at room temperature. These spectra are folded at $p$ = 0 to enhance the statistics. Solid lines denote calculated differential DBAR spectra. The amplitudes are adjusted to levels comparable with the experiments.

**References**

[1] S. Berko, *in Positron annihilation*, ed. by A. T. Stewart and L. O. Roelling (Academic Press, New York, 1967), p.61.
[2] A. Kawasuso *et al*., Phys. Rev. B **83**, 100406(R) (2011).





# REIMEI Research Project: New approach to the exotic phases of actinide compounds under unconventional experimental conditions

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We have proposed development of measurements for actinides compounds under unconventional conditions i.e. highly radioactive (transuranium, $^{235}$U-enriched) materials, high magnetic field, high pressures on international collaborations of several institutes which have their own special techniques, in order to discover and clarify the exotic electronic properties in 5*f*-electron systems, which appear only under extreme conditions due to large energy scales (Fermi energy, Kondo temperature), compared with 4*f*-systems. A special emphasis is put to combine *in-situ* bulk measurements with scattering methods to gain microscopic insight into the various phases in such systems. Because of the difficulty for international transport of actinides samples, researchers and techniques has been encouraged to travel and collaborate to develop and perform the investigations. We describe below some of the collaborations that have been established in the first year of operating the REIMEI project which continues in 2011-2012. 1) High magnetic field NMR using $^{29}$Si-enriched $URu_2Si_2$ in NHMFL, USA Collaboration between JAEA, LANL and NHMFL underway at NHMFL. 2) Energy scale of the electron-boson spectral function and superconductivity in $NpPd_5Al_2$. Collaboration between ITU and JAEA. Experiments at JAEA with theory at ITU and University of Turin [1]. 3) Search for quadrupolar ordering in the hidden ordered phase of $URu_2Si_2$. Collaboration between ITU and CEA with experiments at ESRF [2]. 4) Resonant X-ray emission spectroscopy at the $L_3$ edge of americium up to 23 GPa [3]. Collaboration between ITU and ESRF, with experiments at ESRF. 5) Understanding the complex phase diagram of uranium: the role of electron-phonon coupling. Collaboration between CEA, ITU, and LANL. Experiments at ESRF with theory at CEA. In addition, the first International ASRC Workshop has been held on this subject in Tokai, 16-18 Feb 2011 (photo on the cover page) .The topics 1) and 2) are selected to be described in detail below.

**High magnetic field NMR using $^{29}$Si-enriched $URu_2Si_2$ in NHMFL**

Several single crystals of $URu_2Si_2$ enriched to 99.8 at. % by $^{29}$Si isotope were shipped from LANL to NHMFL. In this first NMR measurement by JAEA and NHMFL NMR groups, we have characterized the sample quality. The line width of $^{29}$Si -NMR at 6.7 T is around 10 kHz for *H*//a, which is quite narrow indicating a good homogeneity of sample. In fact, this line width value is consistent with the previously reported line width in non-enriched sample if we consider some additional line broadening due to the dipolar contribution from $^{29}$Si nuclei. In addition, *T*-dependence of nuclear spin-lattice time has been found to be same as the previously reported value in a high quality non-enriched sample. The $^{29}$Si-enrichment can reduce the magnet time very efficiently. For the next measurement in the higher field, we will concentrate to measure the temperature dependence of NMR spectrum under the several higher fields up to 29 T, in order to identify the phase transition around 22 T, which would be accompanied with a change of Fermi-surface.

**The energy scale $\Omega_0$ of the electron-boson spectral function in the heavy-fermion, *d*-wave superconductor $NpPd_5Al_2$**

This has been predicted on the basis of Eliashberg theory calculations (Fig.1). Assuming a spectral function shape typical for antiferromagnetic spin fluctuations, and imposing constraints provided by the experimental values for the critical temperature and the low-temperature energy gap, one obtains values of $\Omega_0$ of about 2-2.5 meV, slightly dependent on the strength of the Coulomb pseudopotential. These values are in excellent agreement with the characteristic magnetic fluctuations energy estimated from NMR measurements of the nuclear spin-lattice relaxation time at the Al site. The calculated temperature dependence of the upper critical field, the local spin susceptibility, and the nuclear spin-lattice relaxation rate is also in good agreement with available experimental data, showing that a coherent description of the superconducting state can be obtained assuming that the electron pairing in $NpPd_5Al_2$ is mediated by antiferromagnetic fluctuations [1]. Inelastic neutron scattering experiments are planned to confirm the proposed scenario.

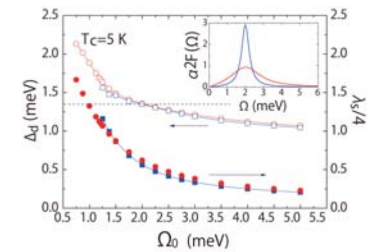


Fig. 1 Gap values $\Delta_d$ calculated at $T$ = 0.25 K assuming a Coulomb pseudopotential $\mu_d^*$ = 0, and an Eliashberg spectral function centered at an energy $\Omega_0$ and having full width at half maximum of 1 meV (red, open circles) or 0.25 meV (blue, open squares). The horizontal dashed line represents the experimental value of $\mu_d^*$. (Right axis) Electron boson coupling constant $\lambda$s as a function of $\Omega_0$, for $Tc$ = 5 K, $\mu_d^*$ = 0, and FWHM of 1 meV (red, closed circles) or 0.25 meV (blue, closed squares). The inset shows the electron-boson spectral function in the two cases.

**References**

[1] G. A. Ummarino, R. Caciuffo, H. Chudo, and S. Kambe, Phys. Rev. B **82**,104510 (2010).
[2] H. Walker, R. Caciuffo, D. Aoki, F. Bourdarot, G. H. Lander, and J. Flouquet, Phys. Rev. B **83**, 193102 (2011).
[3] S. Heathman, J.-P. Rueff, L. Simonelli, M. A. Denecke, J.-C. Griveau, R. Caciuffo, and G. H. Lander, Phys. Rev. B **82**, 201103R (2010).