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^{73}Ge : A NEW HIGH RESOLUTION PAC PROBE

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ABSTRACT

Long lens electron spectrometers were used to make electron-gamma and electron-electron PAC measurements at the $5/2^+$ 13.3keV state of ^{73}Ge. Sources of ^{73}As were produced in decay of ^{73}Se implanted at the CERN/ISOLDE facility. The magnetic interaction in nickel was determined as ω_L=74.2(7) Mrad.s^{-1} and the quadrupolar frequency in antimony as ν_Q=19.7(2)MHz. The nuclear moments derived are μ=-1.08(3)μ_N and $|Q|$=0.70(8)b.

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Abstract

Long lens electron spectrometers were used to make electron-gamma and electron-electron PAC measurements at the $5/2^+$ 13.3keV state of ^{73}Ge. Sources of ^{73}As were produced in decay of ^{73}Se implanted at the CERN/ISOLDE facility. The magnetic interaction in nickel was determined as ω_L=74.2(7) Mrad.s^{-1} and the quadrupolar frequency in antimony as υ_Q=19.7(2)MHz. The nuclear moments derived are μ=-1.08(3)μ_N and |Q|=0.70(8)b.

Introduction

The perturbed angular correlation (PAC) technique has been used intensely for investigations in metals and recently also in semiconductors. For the latter application, in particular, a probe nucleus that is not an impurity in an elemental semiconductor lattice would be highly desirable. The first excited state of ^{73}Ge has been used in a series of Mössbauer spectroscopy experiments [1]. It also represents an ideal PAC candidate. There are, however, serious technical problems to overcome until this probe could be successfully exploited. Due to the high conversion coefficient of the depopulating transition electron detection is by far superior to gamma measurement. The very low energy of the conversion electrons is especially troublesome for such measurements and calls for thin sources.

Low energy implantation of the parent isotope at a mass separator, as e.g. the CERN/ISOLDE facility, nowadays allows clean source preparation. As a first step in the direction of future applications of this high-resolution PAC isotope we have performed measurements of the magnetic interaction and quadrupole coupling in metals. This has enabled us to determine the nuclear moments with good precision.

Decay Scheme

The 2.86(3)μs half-life of the $5/2^+$ first excited state in ^{73}Ge at 13.3keV above the $9/2^+$ ground state allows very accurate hyperfine interaction studies using PAC or Mössbauer spectroscopy. As shown in Fig.1, it is populated predominantly through the 0.50s, $1/2^-$ isomeric state of ^{73}Ge. The decay of ^{73}As as well as that of ^{73}Ga take place almost exclusively via this isomer. At a facility that produces short-lived isotopes, both these parent activities can also be obtained from decay of their grand parent (^{73}Zn or ^{73}Se) or even great-grand parent (^{73}Br) isotopes.

The multipolarities of the $1/2^-$ to $5/2^+$ and $5/2^+$ to $9/2^+$ PAC transitions of 53.4 and 13.3keV are virtually pure M2 and E2. The 53.4 keV transition is converted with $e/\gamma \approx 9$ and the 13.3keV one very highly with $e/\gamma \approx 1100$. The particle parameters calculated for electron detection ($b_2(e^-_L 13.3$ keV$)=1.26$ and $b_2(e^-_K$ 53.4 keV$)=1.29$) clearly show that electron-electron coincidence measurements are the most favorable technique for PAC studies in this case. At least the 13.3keV depopulating transition, however, should be studied via the electrons.

Experimental Setup

We have used two Siegbahn type magnetic long-lens spectrometers [2] for the detection of the conversion electrons and NaI(Tl) scintillators for gamma measurement. The experimental setup is described in ref [3]. For background suppression in the plastic scintillators used as detectors in the electron spectrometers we have employed a pre-accelerating voltage of typically -16kV, applied to a spherical shield enclosing the sample to preserve the angular distribution.

The perturbed angular correlation measurements were performed with a standard fast-slow coincidence setup, incorporating a TAC. Four time spectra were recorded for the gamma-electron coincidence measurement, with the two NaI(Tl) windows set at 53.4keV and the two electron spectrometers set at the L conversion line of 13.3keV. For the electron-electron spectrum the TAC was started with the K electrons of 53.4keV and stopped with L of 13.3keV.

Sample Preparation

A mixture of volatile mass 73 isobars that was produced by spallation of Nb with 600MeV protons and ionized in a hot plasma ion source [4] was implanted into a nickel foil and an antimony single crystal at the ISOLDE-3 [5] on-line isotope separator. The mayor component in the separator beam was ^{73}Se. At the typical intensities of $1.6*10^9$ ions per second of the 60keV separator beam the implantation times to reach a dose of 10^{14} per cm^2 were less than one hour.

The 25µm nickel foil was annealed for 1 hour at 673K in vacuum following implantation. The antimony single crystal, grown by Bridgeman technique, was cleaved before implantation and annealed for 20 minutes at 573K in helium gas afterwards.

Results

The measurement of the magnetic interaction in Ni ran in gamma-electron mode for about 4 weeks at a true-to-chance coincidence ratio of 1 (close to t=0). The time range of the TAC was chosen as $1\mu s$ in order to resolve even high frequency components. The perturbation function, formed in the conventional way, is shown in Fig.2a. Its Fourier transform in Fig.2b clearly shows the two precession frequencies at ω_L and $2\omega_L$ characteristic for magnetic interaction in a polycrystalline and unpolarized sample. The spectrum was therefore fitted with a perturbation function assuming only one unique field acting at the ^{73}Ge nuclei. The extracted Larmor frequency is ω_L=74.2(7)Mrad.s^{-1}. The effective anisotropy obtained was $(A_{22}b_2)_{eff}$=+0.031(3), only about 33% of the expected value after solid angle corrections for gamma-electron angular correlations of pure M2-E2 transitions.

The gamma absorption in the thick sample prohibited the use of the same technique for the Sb measurement. For the experiment therefore electron-electron coincidences were chosen with a TAC time range of $2\mu s$. The PAC spectrum was measured for 40 days with the two lenses at 90^o, and the crystalline c-axis in the detector plane at 45^o. The perturbation function extracted by dividing out the exponential decay is shown in Fig.3a. Its Fourier analysis in Fig.3b reveals the $\omega = 2\omega_0$ ($\omega_0 = 6\pi\upsilon_Q/20$) frequency at 37 Mrad.$s^{-1}$ as prominent component with a small $\omega = \omega_0$ contribution. This is just as expected for I=5/2 in the geometry chosen and a unique electric field gradient with asymmetry parameter η=0 required for a substitucional site in the rhombohedral Sb lattice. The fitted nuclear quadrupole constant $\upsilon_Q=e^2Qq/h$ is υ_Q=19.7(2)MHz. The effective anisotropy obtained was $(b_2A_{22}b_2)_{eff}$=+0.041(3), only about 31% of the expected value after solid angle corrections for electron-electron angular correlations.

As a byproduct of our experiment we also determined the half-life of the 13.3keV state in ^{73}Ge. The best fit value was $2.86(3)\mu s$, in perfect agreement with earlier measurements.

Magnetic Hyperfine Field

The magnetic hyperfine field for Ge in nickel has been studied several times. In the most recent and accurate investigation [6] also the temperature dependence has been measured. Unfortunately the data are only presented in graphical form, so that a major uncertainty is the reading error for the value at room temperature needed to determine the magnetic moment from our experiment. We read from Fig.4 in ref. 6 a value of H(300K)/H(77K) $\approx$ 0.93(2) resulting in a hyperfine field at room temperature for Ge in Ni as 3.6(1)T. This leads to a value of |g|=0.430(12) for the 5/2+ state in ^{73}Ge studied in our measurement.

Electric Field Gradient

From the experimental value of e^2Qq/h=21.1(2)MHz for ^{69}Ge in Sb at 298K [7] and our result one can directly calculate the ratio Q(^{73}Ge,5/2+)/Q(^{69}Ge,9/2+)=0.93(1). For this state also the quadrupole coupling constant in several other metals has been measured. It is, however, not possible to determine without model assumptions the absolute value of the electric field gradient acting on Ge in any of the metals studied from the existing data. Schatz et al [8] have tried to extract this from isoelectric impurity systems, using the Sternheimer factors as normalization constants. Their result is Q(^{69}Ge)=1.0(2)b. Their procedure, though qualitatively acceptable, suffers from two shortcomings - the use of the Sternheimer factors, characteristic for inner electrons but not for the valence shell, and the neglect of lattice relaxation about under- and over-sized impurities. Fortunately there are experimental data that allow to eliminate both effects in estimating the field gradient for Ge in Zn.

Systematic data for the efg of 4p impurities in Zn and 5p impurities in Cd suggest that the same trends are present in both series [9]. The value of the electric field gradient for the isoelectric system Sn$\underline{\text{Cd}}$ is experimentally well known. The ratio q(Zn$\underline{\text{Zn}}$)/q(Cd$\underline{\text{Cd}}$) should take into account the different radial functions for the 4p and 5p valence electrons and also the slightly different lattice structures of Zn and Cd. A reliable estimate would thus be obtained by eq(Ge$\underline{\text{Zn}}$) = eq(Sn$\underline{\text{Cd}}$)*q(Zn$\underline{\text{Zn}}$)/q(Cd$\underline{\text{Cd}}$) = =5.4(5)*10^{17}V/cm^2.

Field gradients extrapolated to T=0 have been used in this estimate. For the ratios q(Zn$\underline{\text{Zn}}$)/q(Cd$\underline{\text{Cd}}$) actually the value from the theoretical calculations should be the most reliable estimate [10]. The resulting nuclear quadrupole moment for ^{73}Ge is |Q|=0.70(8)b.

Nuclear Moments

The error of the magnetic moment $|\mu|$=1.08(3)μ_N determined in the present work results essentially from the error in the value of the magnetic field. The measured magnetic coupling frequency is considerably more accurate than in a previous [11] measurement by a gamma-gamma coincidence technique where μ = -0.94(3)μ_N was found. The negative sign, however, can be taken from this earlier work. The experimental value strongly deviates from the theoretical estimate of μ=-1.7(2)μ_N, calculated from the moments of neighboring Ge isotopes for a simple configuration of the $5/2^+$ state as a coupling of the $9/2^+$ single particle level with a 2^+ collective excitation.

The nuclear quadrupole moment has been estimated from Mössbauer line broadening previously as Q=-0.4(3)b [1]. This value is too inaccurate to present a guideline, and even the sign appears questionable. The simple nuclear model predicts Q=-0.3(1)b, in substantial disagreement with our experimental value of |Q|=0.70(8)b. Apparently the collective nature of the $5/2^+$ state is not properly accounted for by the simple coupling scheme.

Future Prospects

The development of the electron-gamma technique for ^{73}Ge opens various new possibilities to apply this high-resolution PAC probe for solid state studies. Obviously the accurate measurement of electric field gradients and magnetic hyperfine fields, including their temperature dependence, would be a straightforward application. Also the study of semiconductors with this new technique, particularly defects and impurities in germanium, would be highly interesting. A more quantitative understanding of the reduced anisotropies found in the present work would be helpful in such research. Calculations of the effects of electron scattering in the sample are in progress.

Still to be completed is a more accurate measurement of the nuclear quadrupole moment of the $5/2^+$ state in ^{73}Ge. This could e.g. be performed by a Mössbauer study of the system Ge in Sb, prepared as in the present study. Such an experiment could also solve the ambiguity about the sign of the quadrupole moment.

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FIGURE CAPTIONS

Fig.1 Simplified level scheme of ^{73}Ge from ^{73}Ga β^- and ^{73}As EC decay.

Fig.2 a) Perturbation function R(t) measured for ^{73}Ge in nickel foil. The solid line is a fit to the theoretical function for random field orientation.
b) Fourier transform of R(t) (arbitrary units).

Fig.3 a) Perturbation function $N(t)/e^{-\lambda t}$ measured for ^{73}Ge in antimony single crystal. The solid line is a fit to the theoretical function for 90^o geometry.
b) Fourier transform of the perturbation function (arbitrary units).

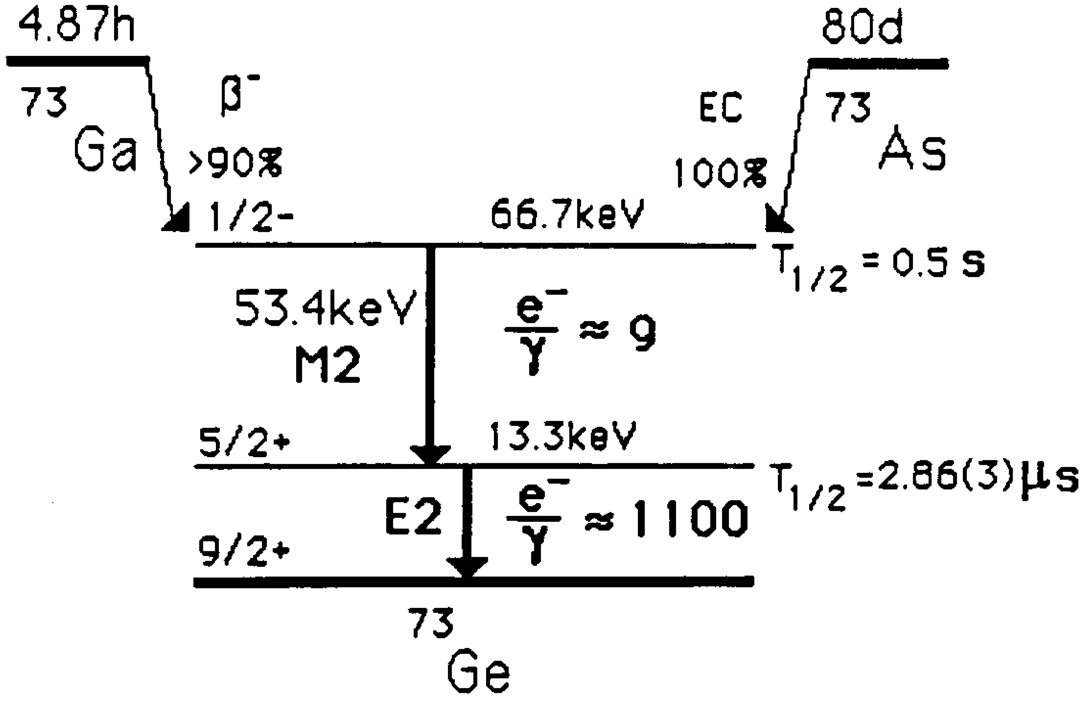
4.87h
73
Ga
β⁻
>90%
80d
EC
100%
73
As
1/2-
66.7keV
T1/2 = 0.5 s
53.4keV
M2
e⁻/γ ≈ 9
5/2+
13.3keV
T1/2 =2.86(3)μs
E2
e⁻/γ ≈ 1100
9/2+
73
Ge

Fig. 1

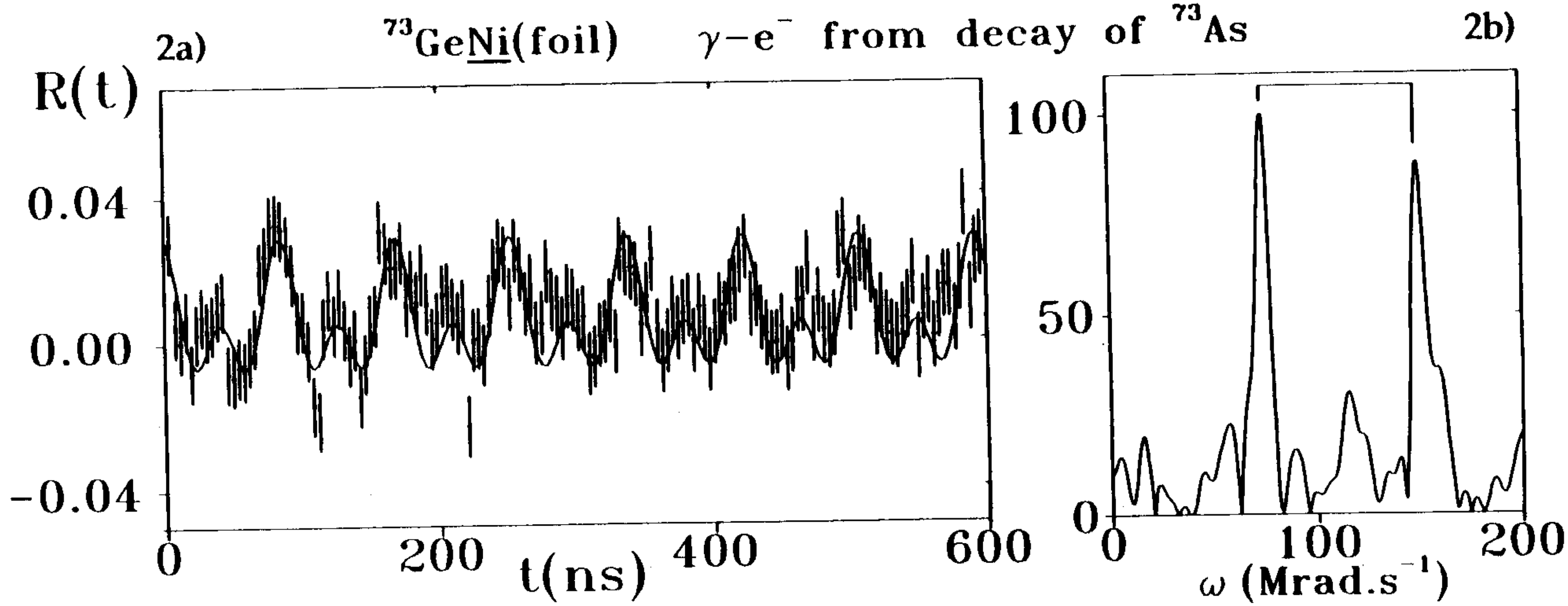
2a)
^{73}Ge<u>Ni</u>(foil) γ−e$^-$ from decay of ^{73}As
2b)
R(t)
0.04
0.00
−0.04
0
200
400
600
t(ns)
100
50
0
0
100
200
ω (Mrad.s^{-1})

Fig. 2

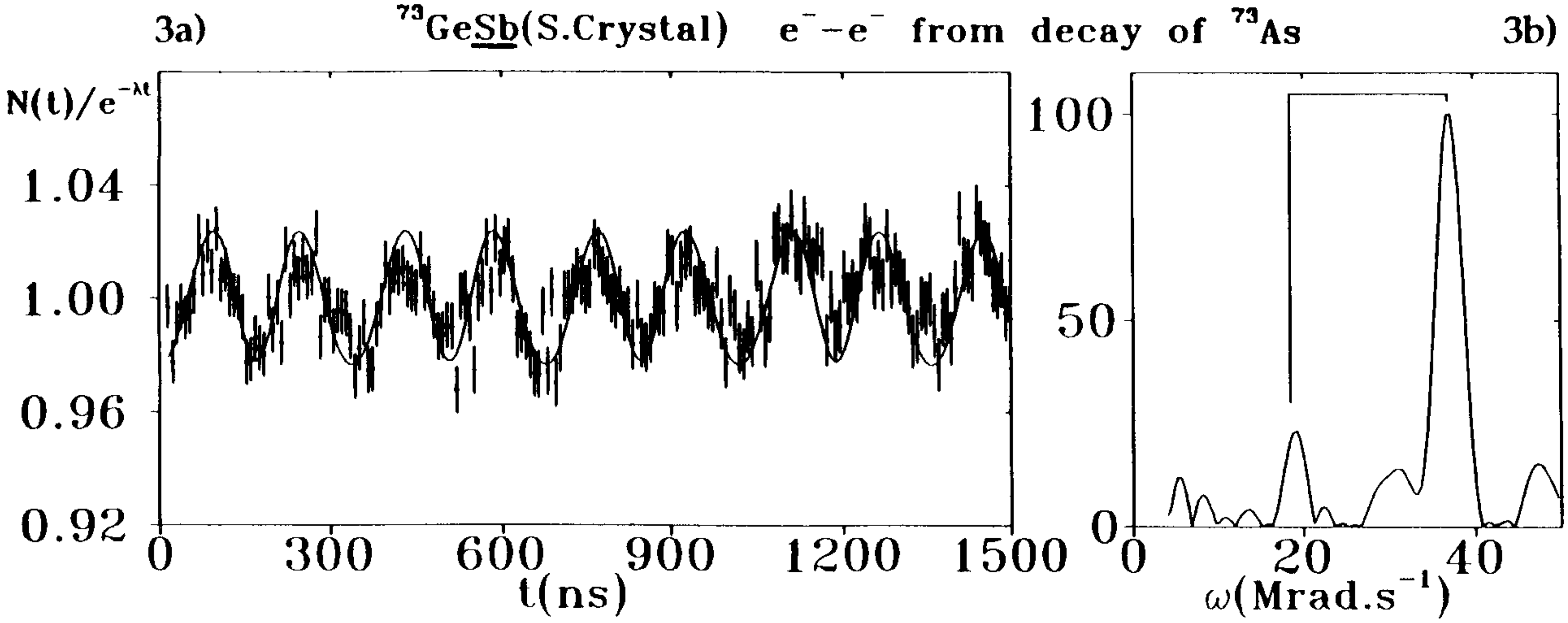
3a)
73GeSb(S.Crystal) e−−e− from decay of 73As
3b)
N(t)/e−λt
1.04
1.00
0.96
0.92
0
300
600
900
1200
1500
t(ns)
100
50
0
0
20
40
ω(Mrad.s−1)

Fig. 3