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TDPAD measurements of magnetic hyperfine field for ^{132}Ba in ferromagnetic Ni

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Abstract The magnetic hyperfine field of Ba in ferromagnetic Ni has been measured by time differential perturbed angular distribution technique using the 13 ns 10^+ isomeric state in ^{132}Ba as probe which was populated in the reaction ^{12}C(^{124}Sn, 4n)^{132}Ba at beam energy of 60 MeV. The hyperfine field extracted from the observed Larmor precession frequency comes out to be −84(5) kG. Our experimental results show good agreement with theoretical calculations performed within local density approximation of the density functional theory. The hyperfine field data presented here would be useful towards accurate determination of g-factor in other high spin states in Ba isotopes.

Keywords Magnetic hyperfine field · TDPAD · g-factor

1 Introduction

Investigations of magnetic hyperfine field for impurity atom in ferromagnetic hosts has been a topic of considerable interest over many years as they provide useful microscopic information regarding spin density and its distribution, which in turn are crucial for understanding long range magnetic ordering in solids. Accurate measurement of magnetic hyperfine field for impurity atom in ferromagnetic hosts are also important for precise determination of nuclear magnetic moments, critical for nuclear structure physics. Over the years systematic studies have been made for variety of impurities in ferromagnetic hosts like Fe, Co and Ni and their trends across the periodic table of elements have been established [1]. Parallel with

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the experimental measurements extensive theoretical studies have been carried out to understand the origin and variation of hyperfine field of different impurity atoms [2–4]. There has been remarkable agreement between experiment and theoretical values for many cases. However, for heavy impurity atoms like Cs, Ba and La very few measurements have been reported [1, 4–6]. As far as we know, no experimental measurements have been reported for Ba in Co and Ni. In the theoretical front, few calculations are available for Fe host with values differing significantly from one another [7]. In this regard it is important to make accurate measurements of hyperfine field for these impurity atoms. In this work we present our results on the magnetic hyperfine field for Ba impurity in ferromagnetic Ni host studied through TDPAD measurements and ab-initio calculations.

2 Experimental and calculation details

For TDPAD measurement of magnetic hyperfine field for Ba in Ni we have used the 10^+ isomeric state in ^{132}Ba ($T_{1/2} = 13$ ns; $g_N = 0.159$) as nuclear probe [8], which was populated in the reaction ^{12}C(^{124}Sn, 4n)^{132}Ba using pulsed ^{12}C beam at beam energy of 60 MeV provided by the heavy-ion accelerator facility at TIFR, Mumbai. The ^{132}Ba recoiling out of the thin ^{124}Sn (0.8 mg/cm^2) target were implanted in a Ni foil (5 mg/cm^2) placed immediately behind the Sn target served as the stopper. Measurements were performed at 100 K in transverse external magnetic field of 5 T produced by a split coil superconducting magnet and data were collected using 4 HpGe detectors placed at angles $\theta = \pm 45°$ and $\pm 135°$ with respect to the beam direction. The time signal from the HPGe detector was used to start the time to amplitude converter (TAC), which was stopped by the primary RF signal of the buncher. Data was collected in the form of a two-dimensional (2D) matrix (energy vs time for each detector) for both up and down directions of the external magnetic. The life time spectra for the γ-rays decaying from the isomeric state (see Fig. 1) were generated by taking energy gated time projections. The background subtracted

Normalized counts for each detector N(θ,t) were used to construct the spin rotation spectra defined as

$$R(t) = [N(+\theta, t) - N(-\theta, t)]/[N(+\theta, t) + N(-\theta, t)] \quad (1)$$

The spectra were fitted to the function

$$R(t) = (3/4)A_2 \mathrm{Sin}(2\omega_L t - \phi) \exp(-\lambda t) \quad (2)$$

to extract the amplitude A_2, Larmor frequency ω_L and damping factor λ. Here, ϕ denotes a phase angle due to finite bending of the incoming beam in external magnetic field.

3 Results and discussion

Figure 2 shows the spin rotation spectrum observed for the energy gate $E\gamma = 465$ keV. The spectrum fitted to the equation (2) yielded $\omega_L = 26$ (4) Mrad/s. From the relation $\hbar\omega_L = g_N \mu_N B_{eff}$, where $B_{eff} = B_{ext} + B_{hf}$ is the effective field seen by the nucleus, one can determine the hyperfine field B_{hf} at the nuclear site of the ^{132}Ba probe.

Taking account of the sign of the A_2 coefficient for the γ-transition involved and sense of rotation in the observed R(t) spectra, the hyperfine field of Ba in Ni was determined to be $B_{hf} = -84 \pm 5$ kG.

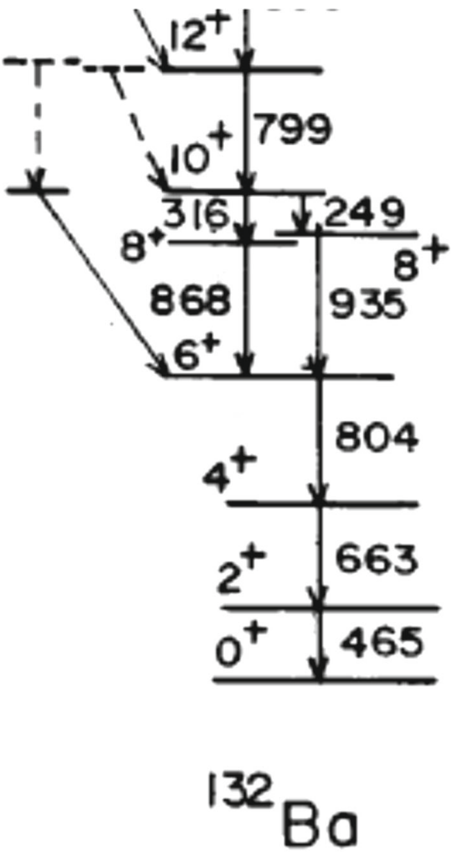

Fig. 1 Partial level scheme of ^{132}Ba

To examine further on the hyperfine field of Ba impurity in Ni host we have carried out ab-initio calculations within the frame work of density functional theory all electron augmented plane wave + local orbitals (APW+lo) method as implemented in Wien2k code [9]. In the present calculation we have used a 32 atom (2×2×2) supercell comprising of 31 Ni and 1 Ba atom constructed with experimental lattice parameter a = 3.5238 [10]. In the APW+lo method, the unit cell is divided into two regions: i) non-overlapping muffin-tin sphere of radius R_{MT} and ii) remaining so called interstitial region. The wave functions within the atomic spheres are expanded in spherical harmonics and plane waves are used for the interstitial region. The muffin-tin radii R_{MT} were chosen to be 2.2 a.u. for Ni and 2.3 a.u. for Ba. The maximum multipolarity of the wave function within the atomic sphere was restricted to $l_{max} = 10$ and the wave functions in the interstitial region were expanded as plane waves with a cutoff wave vector $K_{max} = 7.5/R_{MT}^{min} = 3.409$. The charge density was Fourier expanded up to $G_{max} = 16\sqrt{}(Ry)$. The energy integration in reciprocal lattice space was carried out with k-mesh of size 10×10×10 with 35 special k points in the irreducible wedge of the Brillouin zone (BZ). For all the calculations lattice relaxation was applied to minimize the force on the atoms to less than 1∼ mRy/a.u. The self-consistency of the calculations was ascertained from the charge and energy convergence criterion set to be 0.0001 and 0.01 mRy respectively. Spin polarization was considered for calculating the magnetic moment and hyperfine fields. The calculated hyperfine field at the nuclear site of the Ba

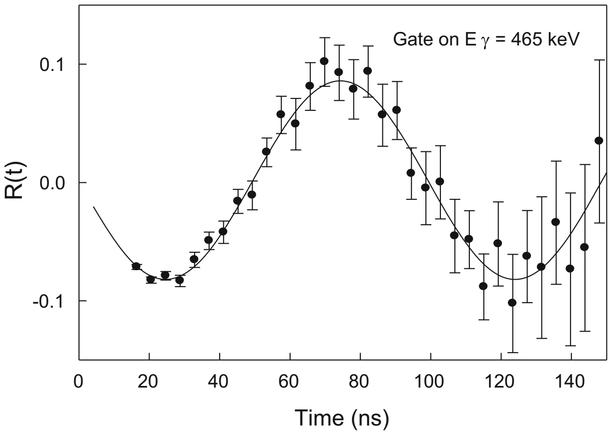

Fig. 2 Spin rotation spectrum of ^{132} Ba in Ni in B_{ext}=5T

impurity atom was found to be −99 kG, coming mostly from the valence 5s electrons. The hyperfine field of Ba in Ni obtained from our calculations is slightly higher than the value of −8T reported earlier [11]. This may be due lattice relaxation which was not considered in the calculation reported in Ref. [11]. To examine the influence of supercell size on the hyperfine field additional calculations were carried out using a 3×3×3 supercell with 108 atoms (107 Ni + 1 Ba) which yielded $B_{hf} = -108$ kG, close to the value obtained from 2×2×2 supercell. This shows that the supercell size does not have significant influence on the hyperfine of Ba in Ni lattice. In principle, the hyperfine field at the impurity atom would also have orbital and dipolar contributions. Although we have not explicitly calculated these contributions, comparing the results reported for close shell impurities like Cd in Ni [11], it is reasonable to assume that they would be negligibly small and would not have serious influence on our results. It is gratifying to note that the calculated hyperfine field closely agrees with the experimental value.

It is important to compare the hyperfine fields of Ba in ferromagnetic hosts like Fe, Co and Ni. The magnetic hyperfine field of Ba in Fe was earlier reported to be −85 kG [1]. Most recent experimental study by Kaur et al. [5] claimed the hyperfine field of ^{132}Ba in Fe to be −60 kG [5]. It is generally believed that the core polarization contribution to the hyperfine field B_{hf}^{core} scales with the local moment of the host atoms [12]. Considering that Fe has larger magnetic moment than Ni, one would expect much smaller core polarization field in Ni host. The valence hyperfine field has also been observed to show similar scaling behavior, albite with a different coupling constant (scaling factor), smaller in magnitude and often opposite in sign to the core polarization field [12]. It is unlikely that the coupling constant for the valence hyperfine field of Ba in Ni is vastly different from the same in Fe, making the total hyperfine field accidentally similar. Thus it is reasonable to scale the total hyperfine field with the host local moment. Using this approximate scaling relation the net hyperfine field of Ba in Ni is expected to be ∼−20 kG, significantly lower than −60 kG observed in Fe. In contrast, the magnitude of the hyperfine field obtained from our data presented here is close to or larger than the value reported for Fe host [1, 5]. Conversely, using the presently observed value of the hyperfine field of ^{132}Ba in Ni, the hyperfine field of the same impurity in Fe would come to be ∼−300 kG, which much higher than the value reported in Ref. [5]. If true, this would have serious implication on the g-factors derived from hyperfine interaction studies [5]. Further experimental measurements using different isotopes of Ba implanted in Ni and other ferromagnetic hosts like Fe and Co would be necessary to resolve this discrepancy. In this direction measurements using 23/2$^+$ state in ^{129}Ba ($T_{1/2} = 45$ ns) as probe to study hyperfine field in Fe and Ni will be useful and are being actively perused.

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