

Anomalous magnetic moment at *Ba* in *Au*



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ABSTRACT

The Time differential perturbed angular distribution (TDPAD) technique is employed to measure the local susceptibility at the recoil implanted *Ba* ions in *Au* following the nuclear reaction $^{120}\text{Sn}(^{12}\text{C}, 3n\gamma)^{129}\text{Ba}$. We have observed first time the local paramagnetic susceptibility of 5.26(18) at *Ba* ions comparable to 4*f*-ions in any non-ferromagnetic metal at room temperature which seems to be related to the electronic *s*–*d* and *s*–*f* transfer at positive lattice pressure.

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1. Introduction

There has been considerable interest in the dilute magnetic systems and extensive investigations have been carried out for the local magnetic moment formation at transition metal ions in the alloying, non-alloying and low-dimension metallic systems [1,2], e.g. *FeCs* and *MoCs*. The studies pertaining to the magnetic response of the isolated *d* and *f* ions in the non-alloying systems, i.e. the impurity host systems with extreme difference of the atomic volume and the chemical environment, have facilitated gaining knowledge about the interactions of the open *d*- or *f*-orbitals with the *s*, *p* and *d* conduction electrons of the host. This has helped in elucidating the role of the hybridization of impurity *d* or *f* electrons with the conduction electrons, the atomic configurational correlations and the crystal field effects in the formation of local magnetic moments at *d* and *f* ions in metals. The host dependent trend of the local moment formation has shown that the magnetism of *Fe* in *sp* metals [3] can be described by ionic-type models whereas in *d* metal hosts [4], it exhibits spin dominated magnetism of itinerant 3*d* electrons, parametrized by an effective spin S_{eff} . Theoretical attempts [5] have been made to qualitatively understand the host dependent magnetic behavior of *d* and *f* ions, i.e. itinerant or

localized ionic type but it is still difficult to predict the local moment formation for the dilute impurities in metals. Further, deciphering the electronic configuration and the nature of the local magnetic moments through hyperfine fields involves the complexities of different contributions to the field. The hyperfine field measurements using the time differential perturbed angular distribution (TDPAD) technique, offer an additional advantage of directly observing the dynamic behavior of magnetic moments. In the last decade with the availability of high intensity synchrotron facilities, it has become possible to measure directly the relative contributions of the spin and the orbital moments (orbital to spin moment ratio) and the probable electronic configuration using the X-ray circular dichroism (XMCD) and the X-ray absorption spectroscopy [6]. These give direct information about the local magnetic moment as compared to the indirect information obtained from the hyperfine field measurements. This has intensified theoretical interest in the magnetic moment calculations.

The hyperfine magnetic field at the site of the nucleus is sensitive to the nature of the magnetic moment at the ion. The magnetic response of the isolated ion is expressed in terms of the paramagnetic correction factor (β) equal to the ratio of magnetic hyperfine field at the nucleus and the applied magnetic field [7]. The TDPAD technique is a powerful unique technique for the investigation of the local susceptibility at any temperature for the recoil implanted ions following the heavy ion reactions. The desired impurity–host combination with the impurity concentration less than 1 ppm can

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be achieved through recoil implantation well below the surface so that the impurity–impurity interactions and the surface effects are negligible. The details of the methodology and the observed systematics for the isolated *d*- and the *f*-ions are well delineated upon in the review article by Riegel and Gross [8].

It may be pointed out that the occupancy of *d*-band in the alkaline earth metals increases with atomic number *Z* and the metals have characteristics somewhat similar to the early transition metals [9]. Motivated by this aspect, recently, we have measured the hyperfine magnetic field at *Ba* in *Fe* for the *g*-factor measurements of the 173 keV, $\frac{9}{2}^-$ ($T_{1/2} = 16(2)$ ns, $g = -0.192(6)$) and 473 keV, $\frac{23}{2}^+$ ($T_{1/2} = 47(2)$ ns, $g = -0.233(7)$) isomeric states in ^{129}Ba [10] using the TDPAD method. The measured magnetic hyperfine field $-6.0(2)$ T at ^{129}Ba in *Fe* follows theoretical predictions for the ions at substitutional sites [11]. In the light of the site location channeling studies of alkaline earth ions in *Fe* [12], the *Ba* ions are expected to be at substitutional sites. The damping of the spin–rotation curve was observed due to radiation damage and was electric in nature.

As a follow up of the above, we decided to carry out the TDPAD measurements on the recoil implanted (following heavy-ion reactions) ^{129}Ba in gold. It is pertinent to mention that barium and gold have body centred cubic (bcc) and face centred cubic (fcc) crystal structures, respectively. Their lattice constants are 0.5028 nm and 0.4078 nm, which correspond to the metallic radii 0.218 nm and 0.144 nm, respectively.

2. Experimental setup

^{129}Ba was excited through the nuclear reaction $^{120}\text{Sn} (^{12}\text{C}, 3n\gamma)$ ^{129}Ba by 52 MeV pulsed ^{12}C beam at IUAC (New Delhi). The pulsing rate was 250 ns and the pulse width was about 1.2 ns. The target consisted of 400 $\mu\text{g}/\text{cm}^2$ enriched ^{120}Sn evaporated on 4 mg/cm^2 *Au* foil with thick *Ta* backing to stop the beam. The thickness of the *Au* foil is just enough to stop the recoiling *Ba* ions in *Au*. This is to minimize the radiation damage due to the ^{12}C beam. The current was less than 5 nA. The concentration of recoil implanted *Ba* ions in *Au* is well below 1 ppm. The 0.75 T external magnetic field was applied perpendicular to the detector plane. The magnetic field at the target was calibrated through the 10^+ ($T_{1/2} = 364(7)$ ns, $g = 0.728(1)$) isomeric state in ^{54}Fe and it was excited through the $^{45}\text{Sc} (^{12}\text{C}, 2n\text{p}) ^{54}\text{Fe}$ nuclear reaction with 50 MeV ^{12}C pulsed beam. ^{120}Sn target in the previous measurements was replaced by 5 mg/cm^2 thick *Sc* foil. The de-exciting γ -rays from the corresponding isomeric states were registered by *NaI(Tl)* detectors placed at 17 cm distance from the target. The experimental setup is described in a previous communication [13]. The data were collected in the *LIST* mode with four parameters: the two energy and two time signals from the time to pulse height converter corresponding to each *NaI(Tl)* detector.

3. Data analysis and results

The acquired data were sorted offline and after proper gain matching for energy and time, two-dimensional matrices of energy versus time were formed for each detector. From these matrices time-gated energy spectra and energy-gated time spectra were created. The partial level scheme of ^{129}Ba showing the decay of the presently investigated isomers is shown in Fig. 1. For the $\frac{9}{2}^-$ and $\frac{23}{2}^+$ isomeric states, 173 keV and 473 keV transition, respectively, were analyzed.

The $R(t)$ factors were formed from the background-subtracted and normalized time spectra $I(\theta, t)$,

$$R(t) = \frac{I(45^\circ, t) - I(-45^\circ, t)}{I(45^\circ, t) + I(-45^\circ, t)}. \quad (1)$$

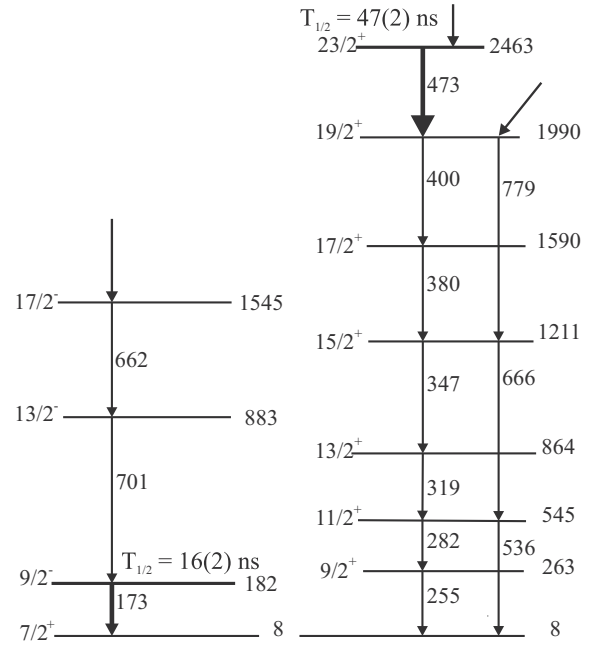


Fig. 1. Partial level decay scheme of $\frac{9}{2}^-$ and $\frac{23}{2}^+$ isomeric states in ^{129}Ba [14] (weak transitions are omitted).

The experimental $R(t)$ factors were least-squares (*LSQ*) fitted to the theoretical expression for the magnetic perturbations [15],

$$R_{\text{theo}} = \frac{3}{4} A_2 * \sin(2\omega_L t + \phi). \quad (2)$$

The Larmor frequency ω_L ($= g\mu_N B/h$) and the phase angle ϕ , to account for the beam bending, were kept as free parameters. The damping parameter λ of the nuclear spin rotation spectra was not observed in the time window of TDPAD measurements. The results are projected in Fig. 2.

We observe single Larmor frequency with negligible frequency distribution and the ratio of Larmor precession frequencies which is equal to the ratio of the respective *g*-factors of the isomeric states in ^{129}Ba . This shows that the *Ba* ions occupy the unique sites and the distortion of the surrounding lattice is negligible to give any modulation of the spin rotation spectra due to quadrupolar interaction. Comparing our results of *Ba* in *Au* with *Ba* in *Fe* [10], we infer that the nearest neighbor environment of the impurity is unperturbed by radiation damage because the probability for the formation of near-neighbor vacancy is very small. Most probably, the *Ba* ions are in the substitutional sites [16]. The calibrated external magnetic field (B_{app}), extracted from ω_L (10^+), at the target was 0.758(1) T. The magnetic hyperfine field (effective magnetic field, B_{eff}) at ^{129}Ba nucleus, extracted from the Larmor frequency $\omega_L(\frac{23}{2}^+)$ is 4.71(14) T. The sign of the magnetic field was derived from the sense of rotation of the angular distribution pattern. These magnetic field values yield the paramagnetic correction factor as $\beta = 6.26(18)$, much greater than the value 1, for the non-magnetic systems. The large positive value of local susceptibility, defined as $\beta - 1$, indicates the orbital contribution to the hyperfine magnetic field at *Ba* in *Au*. This is further supported by the absence of any magnetic damping (due to fluctuating spin moment) of the spin rotation spectra of the $\frac{9}{2}^-$ and $\frac{23}{2}^+$ (large magnetic moment) isomeric states. It is indeed surprising to observe the high positive local susceptibility for the non-transitional metal alloy system. Neither *Ba* atom nor *Au* has unfilled *d*- or *f*-shell. To be sure about the finding, we have checked the reproducibility of the results. Further, the *Ba* ions can be considered to be in

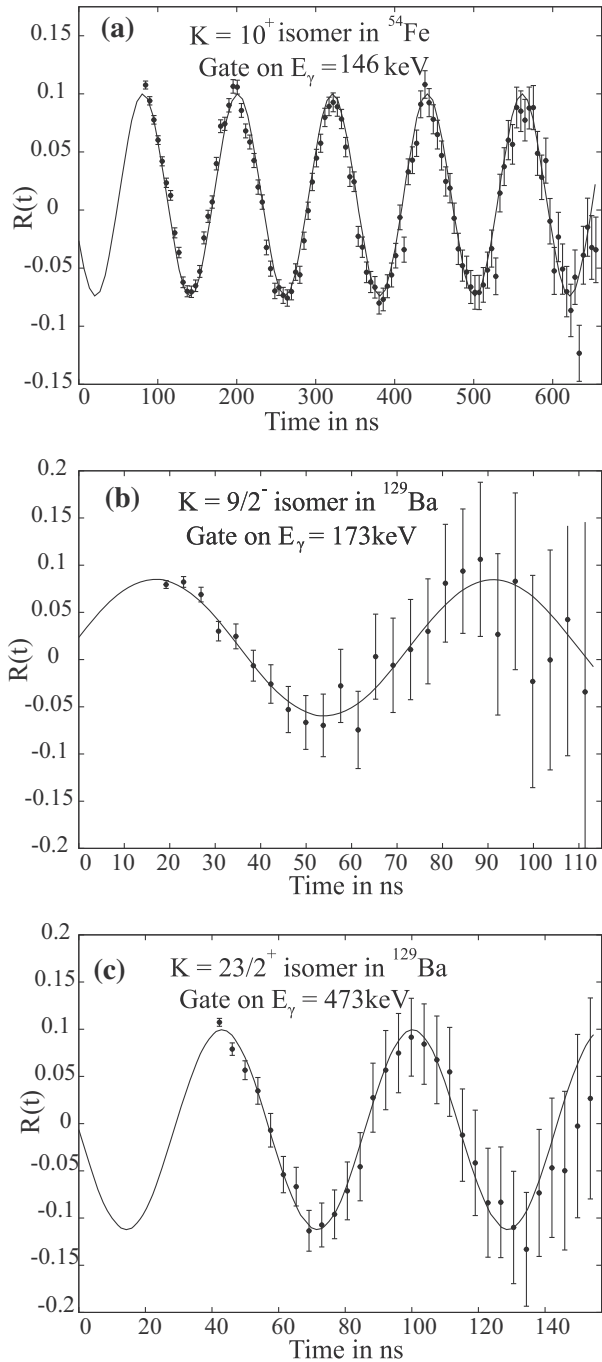


Fig. 2. Spin rotation spectra of the 10^+ (a) isomeric state in ^{54}Fe and the $\frac{9}{2}^-$ (b) and $\frac{23}{2}^+$ (c) isomeric states in ^{129}Ba implanted in Au at room temperature.

electronic equilibrium, (10^{-12} s) after recoil implantation into Au during hyperfine magnetic field measurements. It seems that positive local susceptibility is related with the specific electronic structure at Ba ions in the non-alloying BaAu system.

4. Discussion

The hyperfine magnetic field is sensitive to the local electronic structure and environment of the probe and has been the major means to investigate the conditions of formation and the nature of the local magnetic moments in the dilute alloys. For a long time the efforts have been going on to understand the physical

mechanism of hyperfine fields at the nucleus of the impurity in the dilute as well as the concentrated magnetic systems. The origin of the hyperfine field in metals is much more complicated than in free atoms or ions because of the non-localized nature of the valence electrons whose polarization plays an important role and is still not well understood. The hyperfine magnetic field at the nucleus is mainly considered to be the sum of (i) the negative contribution (the Fermi contact interaction) from the polarization of the conduction electrons and the core due to spin moment at the ion itself and from the outer ionic moments through the s - d exchange/ s - d hybridization, and (ii) the positive orbital moment contribution associated with the valence electrons (unquenched partially filled p , d & f shells) [17]. In some of the cases, having the non-cubic symmetry, there may be small spin-dipolar field due to the spin moment at the ion itself and the outer ionic moments. Early theoretical attempts were limited to explain qualitatively the impurity magnetic hyperfine fields in the ferromagnetic hosts Fe , Ni , Co [18]. The band-structure calculations using density-functional theory, after the success in the electronic structure calculations, were applied to magnetism, i.e., for the investigation of the electronic structure of the local magnetic moments and the magnetic field at the nucleus [5,19].

The first-principles calculations employing the Green's function and the supercell *FLAPW* (full-potential linearized augmented plane-waves) methods have been successful to predict the quantitative trend of the impurity magnetic hyperfine field in Fe [11,20]. Good agreement with the experimental results was achieved by inclusion of the lattice relaxation around the heavier impurities. The effects of the lattice relaxation have been considered by Korhonen et al. [11] for the impurity hyperfine fields in the Fe host. These theoretical calculations have helped in identifying the small positive orbital contribution in addition to the role of conduction and core polarization for the Ba ion in Fe . It may be mentioned that the inclusion of the lattice relaxation effects has significantly narrowed down difference between the theoretical and the experimental values. But still some discrepancies persist for the large volume $5sp$ impurities, e.g. Cs and Ba . Recently, it has become possible to calculate the different hyperfine field contributions (considering the possibility of partial quenching of orbital moment also) at lanthanide and actinide impurities in Fe [21]. The first-principles electronic structure calculations have been carried out to explain and predict the local moment formation at nd transition metal atoms also in different type of host metals, surfaces, interfaces and clusters [5,22,23]. The focus has been to learn the role of key parameters, e.g. hybridization (of nd shell with the conduction electrons), correlation (energy of nd electrons), coordination number and the band structure of the host etc. in the localization of nd electrons. These parameters are inter-related to each other and the formation of the magnetic moment cannot be considered just based on one or two parameters. Fe is a good example of thorough investigations of these parameters, particularly for the extreme cases, e.g. alkali metals and noble metals. In heavy alkali and alkaline metals, i.e. larger lattice volume, the magnetic moment at Fe could be expressed in terms of the ionic configuration (Hund's rule). This has shown the reduction of the hybridization of local $\text{Fe } d$ electrons with host sp electrons and the negligible crystal effects for the free ion value of magnetic moment [3]. On the other hand the magnetic moment in noble metals is of the spin type and is sensitive to the details of the band structure of the host metal, e.g., Fe in Ag and Au [24]. In metals, density of states is one of the important parameters considered to explain qualitatively the local magnetic moment at nd atoms as per Stoner criterion, $N(E_f)I > 1$ (where $N(E_f)$ is the non-polarized density of states at the Fermi energy E_f and I is known as the Stoner exchange integral). As per systematic observed for the Z dependence of $N(E_f)$

and I [25], the Stoner criterion $N(E_F)I > 1$, does not appear to be satisfied for the magnetic moment formation in the system **BaAu** and to our knowledge **Ba** ion has not been the part of any magnetic moment formation studies. Mostly the magnetic moment formation at transition ions in simple metals could be understood in terms of the Friedel–Anderson model theory [26–28] by assuming the existence of localized d states. The positive magnetic field at **Fe** in **Ca** because of orbital component could be better explained using the modified Friedel–Anderson model [29]. This change in the magnetic moment formation in **Ca** vis a vis alkali metals may be due to the presence of d -electrons in the conduction band and the reduced lattice volume. The calculations in the local-spin-density approximation have demonstrated the trend of magnetic moment formation at the nd ions in alkali-earths similar to the alkali metals with some exceptions, e.g. **ZrBa** [30]. This is pointed out to be due to the peak and dip structure of density of states around Fermi energy with respect to **Ca** and **Sr**. In contrast to these experimental and theoretical observations we have indications of large orbital contribution to the magnetic moment at **Ba** in **Au** from the measured large positive value of the local susceptibility.

The **BaAu** system is a non-ferromagnetic cubic metallic system. The host **Au** is diamagnetic in nature and the **Ba** metal is feebly paramagnetic in nature. It is surprising to observe the strong enhancement of the applied magnetic field at very dilute **Ba** ions in **Au** matrix. As far as we know such a large value of the enhancement factor is expected only for the heavy lanthanide ions at room temperature. For the d - and f -ions the local susceptibility at room temperature has been observed to have values less than 3 and 8, respectively, depending on the nature of the ion [5,7,31]. This is the first time that the local susceptibility has been observed for the non magnetic ion equivalent to that of the heavy lanthanides. The local susceptibility is related to the existence and the stability of the local magnetic moment and has been observed through the hyperfine interaction measurements. The hyperfine magnetic field at the nucleus arises due to the magnetic moment at the ion itself or the host ions. The first contribution due to the Fermi contact interaction, i.e. from the spin polarized core and conduction electrons at the **Ba** nucleus, is expected to be zero and there cannot be any transferred magnetic field also as both **Ba** and **Au** are non-magnetic in nature. This is further supported by the positive sign of the hyperfine magnetic field at **Ba** in **Au**. The sign is measured with respect to the external magnetic field. The field due to orbital motion is always positive with respect to angular momentum j . The observed large positive β value for **Ba** in **Au** strongly indicates the existence of local magnetic moment at **Ba** ions with large contribution due to orbital moment. This is further supported by the dynamic nature of the d/f moment, i.e. the moment fluctuation rate, for the interpretation of the local susceptibility. The atomic spin fluctuation rate which arises because of the exchange interaction of the atomic moment with the conduction electrons is related to the measured nuclear relaxation time [4]. The spin fluctuation rate is parameterized, within Kondo like approach, by the Kondo temperature [32]. Extensive investigations have been carried out for the d/f moment instabilities in d and sp metals and a good description is given by Riegel and Gross [1]. They observed that the spin fluctuation rate is enhanced by sp – d hybridization but reduced by the d – d ferromagnetic coupling. The local susceptibility has been observed to decrease with the reduction of the lattice volume also, i.e. increase of impurity d/f -host sp hybridization or non localization of d/f -electrons. In the present measurements, it is revealed by the longer magnetic nuclear relaxation time, i.e. implied by the un-damped nuclear spin precession curve in the observed time window. There are indications of f -moment stabilization, corresponding to low Kondo temperature, under pressure for the mixed valent systems, e.g. intermediate valence compounds

of **Yb** [33]. The $4f$ -occupancy of **Yb**, i.e. magnetic response, has been varied both through alloying or external pressure. These observations imply either the smaller density of states at Fermi energy of gold or weak interaction of the induced $5d/4f$ moment at **Ba** ions with the conduction electrons or both.

The results of present investigation can be summarized as follows: (i) localized magnetic moment is formed in the presence of high lattice pressure, contrary to the localization of the d - or f -moments in alkali metals [34,35], (ii) the local paramagnetic enhancement factor corresponds to that for the heavy rare-earth ions, i.e. significantly away from the maximum value observed in case of nd ions, and (iii) nuclear relaxation time is comparatively large. To the best of our knowledge such a behavior has not been observed for any transition and non-transition impurity in any metallic host. Such an unusual characteristic can be the end result of the formation of anomalous metallic system **BaAu** in which, the atomic volume, the Z value, the electro-negativity and the compressibility of the constituent elements differ very much.

Among alkaline rare-earth metals only **Ba** crystallizes in the body centered cubic (bcc) structure. This difference in the crystal structure is attributed to the increased presence of d -electrons in the empty d band near Fermi level. The band structure calculations of **Ba** have indicated the substantial filling of the $5d$ band (one d -electron per atom) besides the sp – d hybridization at normal pressure and temperature [36]. These theoretical calculations have also shown the importance of s – d (major fraction) and s – f electronic transfer in accounting for the structural changes at higher pressures. The structural changes of **Ba** with pressure are explained based on the shifting of the d -band below the Fermi level and the rising of the s -band above Fermi level which results in filling (s – d transition) and broadening (strong hybridization between the sp and d bands near the Fermi level) of the d -band. The s – d hybridization is not conducive for the localization and the magnetic moment formation by d -electrons as shown for the early $5d$ transition atoms in **Au** [37]. The tendency towards the formation of magnetic moment of $5d$ atoms in metals (**Ag/Au**) is small because of the broader hybridization width of $5d$ shell with the sp conduction band. The local electronic structure and the impurity-induced volume changes are correlated with each other and are difficult to treat theoretically. Most of the systematic experimental and theoretical investigations have dealt with the behavior of local magnetic moments of dilute d - or f -state impurities in variance with the present observation of the moment formation at **Ba** in **Au** having no localized d - or f -open shell in bulk. The electronic structure of dilute impurity **Ba** is sensitive to the local density of states different from the bulk.

In the absence of any theoretical calculation and the magnetic moment measurements for the **BaAu** system, we proceed as following to interpret our results. The structural changes of **Ba** have been explained in terms of the pressure induced d - and f -state occupancy. The photoemission studies of the electronic structure of the states near the Fermi level for **Ba** overlayers on the **Ni(111)** surface have also shown the localization of the $5d$ and $4f$ states [38]. The hybridization of states near the Fermi energy increases with increasing the overlayer thickness. The location and the widths of $5d$ - and $4f$ -bands are sensitive to the atomic arrangement and the lattice volume [39]. This will not be surprising if s – d and s – f electronic transfer, because of the reduction of volume per **Ba** atoms at positive lattice pressure, is playing crucial role for the appearance of the large positive local susceptibility in **BaAu** also. In the present scenario, **Ba** in **Au**, the transfer of s -electrons to $5d$ state is not expected to lead the formation of the d -moment and the longer nuclear relaxation time. This fact arises from the more extended nature of the $5d$ orbitals at the beginning of the $5d$ series, resulting into a stronger hybridization with the

host $5d/6sp$ band electrons. The minor possibility of s – f electronic transfer, at comparatively shorter distance from Ba nucleus, may be sufficient for the observed enhancement of the local susceptibility. The weak hybridization and the mixing exchange interaction of impurity f -state with the host conduction band near Fermi energy may lead to the localization of f -electrons for the magnetic moment formation and long nuclear relaxation time. The measured positive value of hyperfine magnetic field, the large local paramagnetic susceptibility and the long nuclear relaxation time point towards the conclusion that the local magnetic moment appears due to the $4f$ electrons localized on Ba ions. Furthermore, the temperature dependence studies of β are required to support the present observations and to draw final conclusion about the local electronic configuration and the mechanism of $4f$ -localization in the $BaAu$ system.

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