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DISCLAIMER

RARE-EARTH VALENCE STATE STUDIES OF THE SERIES RIn_3 and RSn_3 DERIVED FROM QUADRUPOLE COUPLING CONSTANTS*

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Abstract

Time-differential perturbed angular correlation was applied to ^{111}In to measure the quadrupole coupling constants at the In and Sn sites for rare-earth compounds of the series RIn_3 and RSn_3 that have the $AuCu_3$ structure. Definitive evidence was found for Yb and Eu having lower valence states relative to the assumed value of +3 for the other rare earths. Measurements at liquid helium temperatures and pressures to 18.5 kbar indicate that the room temperature valencies of $CeSn_3$, $YbSn_3$, and $YbIn_3$ have not changed.

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1. Introduction

Discussions of the electronic structure of metals usually begin by partitioning the lattice into positively-charged ion cores plus itinerant valence electrons. For many metals this separation is a computational shortcut rather than an experimentally observable feature, but in the rare-earth metals and their compounds the "valence state" has a more exact meaning. The 4f electrons are sufficiently localized to be assigned unambiguously to the ion cores, which then have charges of +3 or sometimes +2. Considerable theoretical and experimental attention has recently been focused on the question of non-integral rare-earth valence and valence state changes induced by pressure and temperature. Most of the cases studied to date have been rare-earth chalcogenides which have insulating or semi-metallic properties, but potentially interesting metallic cases have also been known for some time.

Early studies of the susceptibility [1] and lattice constants [1-4] of the rare-earth series RIn_3 indicated that Yb was in a lower valence state (presumably +2) than the other rare-earths. The large increase in susceptibility [1] noted on cooling to liquid helium temperatures was taken as an indication that the valence of Yb had changed from +2 to +3. In the RSn_3 series $EuSn_3$ and $YbSn_3$ both appear with divalent [4] rare-earths, while $CeSn_3$ is trivalent at room temperature and originally was thought to become quadrivalent at helium temperatures. These early conclusions concerning the temperature dependence of the rare-earth valencies were disputed through later Mössbauer [5-8] studies, but a consistent picture for the combined In and Sn series has not been published to date.

The RIn_3 and RSn_3 intermetallics crystallize in the FCC $AuCu_3$ structure, which places the In or Sn at a site with tetragonal symmetry. The non-cubic nature of this site leads to a non-vanishing, axially-symmetric electric field gradient q which is highly sensitive to changes in the surrounding rare-earth valence state. According to a simple point-charge model, q should scale as Z'/r^3, where Z' is an effective charge. The functional form of this dependence leads to a great sensitivity advantage of field gradients as opposed to lattice constants for determining valence states. A change from +3 to +2 in rare-earth valence might increase the lattice constant by 1-3%, but would reduce q in this simple picture by one-third. We shall show in section 4 that the sensitivity is even greater because in a point charge model q scales as $Z_{RE}-Z_{In,Sn}$ for the $AuCu_3$ structure, where Z_M is the assumed valence of component M.

In this paper we report quadrupole coupling constants e^2qQ/h determined by time-differential perturbed angular correlations in the 247 KeV state of ^{111}Cd following the electron-capture decay of ^{111}In. Experimental procedures are described in section 2 , and results are given in section 3. Our results are interpreted in section 4 to show that only $YbIn_3$ contains the rare-earth in the divalent state in the RIn_3 series, while both $YbSn_3$ and $EuSn_3$ contain divalent rare earths at room temperature. Data taken at 4.2°K and in some cases also under 18.5 kbar pressure indicate that the devalent state appears to be stable in $YbIn_3$ and $YbSn_3$ and that $CeSn_3$ is non-magnetic at liquid helium temperatures, with the Ce in the trivalent state.

2. Experimental

The 2.81-day ^{111}In activity, which decays after electron capture via the 173-247 keV cascade of ^{111}Cd, was produced by the ^{109}Ag(α,2n) reaction induced by α-particle bombardment of a 0.005 cm natural Ag foil at the Lawrence Berkeley Laboratory 88 in. Cyclotron. The use of natural Ag (51% Ag^{107}, 49% Ag^{109}) necessitated waiting about 36 hr for the In^{109} (4.3 hr) to decay away, after which the In and Ag were separated using magnesium hydroxide precipitation at a pH between 8 and 9. Generally 0.1-0.2 mg of In carrier was added, and the activity plated out of a formate-buffered sulfate solution (pH 2) onto an In or Sn foil.

The active foil and stoichiometric amounts of rare-earth metals were then arc-melted under argon on a water-cooled Cu hearth. Because the components for YbIn$_3$, YbSn$_3$, and EuSn$_3$ reacted vigorously during melting, some caution was required to produce these compounds without splattering and loss of material. As an additional check we also made these compounds by encapsulating the pure metals in a tantalum crucible and sealing under argon in a quartz tube prior to oven heating. Samples made by these two methods yielded identical coupling constants within experimental error.

Efforts to prepare EuIn$_3$ failed. This is consistent with refs. 2 and 9 which indicated that only EuIn, EuIn$_2$, and EuIn$_4$ form stable intermetallics. The attempts to make EuIn$_3$ resulted in a spectrum which was similar to that of EuIn$_4$ when the latter compound was made stoichiometrically. The heavy RSn$_3$ compounds from Tb to Lu (with the exception of YbSn$_3$) cannot be made in the FCC phase in an arc-furnace or oven at conventional pressures. Our samples of $GdSn_3$ also showed evidence for not being single phase, but there was no difficulty in extracting the quadrupole coupling constant to ±10%.

The time spectra were recorded on four 3.8×5.1 cm NaI(Tl) scintillation crystals mounted on 56 DVP photomultipliers using conventional fast-slow logic. The detectors were arranged in a square array, labeled cyclically A, B, C, D. Two 180° (AC and BD) and two 90° (BC and AD) coincidence spectra were routed to a Scipp 1600 channel pulse-height analyzer and later dumped onto magnetic tape for subsequent analysis. The time resolution measured with a ^{22}Na source with the energy windows set at 173 and 247 keV was 2.0 ns FWHM. The time-to-amplitude converter (Ortec 437) was calibrated using an Eldorado 160 digital delay generator.

When four detectors are used in this way, with A and B the "start" detectors (recording the 173 keV photons) and with C and D the "stop" detectors (247 keV photons), the time-modulation function $\bar{Q}_{22}A_{22}G_{22}(t)$ can readily be extracted [10] from the four coincidence counting rates

$$W_{AC}(\theta,t) = N_o e^{-t/\tau_N} \varepsilon_{AC}[1 + Q_{22}(AC)\ A_{22}G_{22}(t)\ P_2(\cos\theta_{AC})] \quad ,$$

etc, by forming the ratio

$$-\frac{2}{3}\left\{\left[\frac{W_{AC}W_{BD}}{W_{AD}W_{BC}}\right]^{1/2} - 1\right\} \cong \bar{Q}_{22}A_{22}G_{22}(t) \equiv A'_{22}G_{22}(t) \quad .$$

This eliminates the total counting rate N_o, the nuclear lifetime τ_N, and to a good approximation the coincidence efficiencies ε_{AC}, etc. Only an average solid-angle correction factor $\bar{Q}_{22} \cong 0.9$ remains.

For convenience it is absorbed into the "effective" angular correlation coefficient A'_{22}.

3. Results

The tetragonal symmetry of the In, Sn sites and polycrystalline nature of the samples allow us to treat the perturbation as one of randomly oriented, axially-symmetric electric field gradients. The perturbation factor for this case (I = 5/2) has the following analytic form [11]

$$G_{22}(t) = \frac{1}{5} + \frac{13}{35}\cos\omega_Q t + \frac{2}{7}\cos 2\omega_Q t + \frac{1}{7}\cos 3\omega_Q t \quad , \qquad (1)$$

where ω_Q is the quadrupole frequency given by

$$\omega_Q = 3e^2qQ/20\hbar \quad , \qquad (2)$$

and Q is the quadrupole moment of the 247 keV level.

Figures 1 and 2 show typical time modulation functions due to the precession of the quadrupole moment of the ^{111}Cd 247 keV level in the electric field gradient present at the In and Sn sites. The solid curve represents a least-squares fit to eq. (1) and yields the modulation frequency ω_Q. In tables 1 and 2 we have listed for the RSn_3 and RIn_3 compounds, respectively, the lattice constants and the quadrupole coupling constants e^2qQ/h obtained from the fits and eq. (2).

4. Interpretation

Recent theoretical discussions [12-16] have emphasized the difficulties of calculating the valence-electron contribution to the electric field gradients (EFG) of metals and alloys when the exact charge distribution around the atoms is not known. The essential difficulty arises because the charge distribution of the itinerant valence electrons is actually largely centered around the ion cores, where it tends to shield the ionic core contribution to q. This can result in either the cores or valence electrons dominating and determining the sign of q. Even the separation of the EFG into separate independent core and electron contributions has been brought into question by a recent study [17] of the systematics of experimental data on metal and alloy gradients which indicates an apparent scaling of the electron contribution to the total ionic core term $eq_{lat}(1 - \gamma_\infty)$.

We will not attempt a quantitative interpretation of the magnitude of e^2qQ/h, which would require a rather elaborate calculation of dubious value in light of the present understanding of the contributions to electric field gradients in metals and alloys. Instead we will adopt the form

$$eq = eq_{lat}(1 - \gamma_\infty)(1 + F_e) \qquad (3)$$

suggested by ref. 18 and empirically justified on a systematic basis by Raghavan et al.[17] In eq. (1), eq_{lat} is the point charge contribution of the "ionic" cores, γ_∞ the Sternheimer factor, and F_e a term which reflects the valence electron contribution.

Using the method of plane-wise summation [19] for a lattice of infinite extent, de Wette and Schacher [20] performed the lattice-sum calculation for the FCC structure at the In or Sn site. The result of their calculation yields

$$eq_{lat}(\mathrm{In,Sn}) = -e(8.67)(Z_{RE} - Z_{\mathrm{In,Sn}})/a_o^3 \quad , \tag{4}$$

where Z_{RE} and $Z_{\mathrm{In,Sn}}$ are the assumed metal valences and a_o is the lattice constant. The factor 8.67 for the infinite lattice can be compared to the number 7.0, which we calculated assuming only interactions out to the third nearest neighbor. Most of the contribution to the EFG is thus short-range in nature. The quadrupole coupling constants are related to eq. (3) by

$$|e^2qQ/h| = |e^2Q(8.67)(Z_{Re} - Z_{\mathrm{In,Sn}})(1 - \gamma_\infty)(1 + F_e)/ha_o^3| \quad . \tag{5}$$

The study of the rare-earth valence and its possible fluctuations are contained in the form of eq. (5). If the rare earths in the series RIn_3 or RSn_3 were all in the +3 valence state and the electronic screening was constant, then a plot of e^2qQ/h vs Z'/a_o^3 would be roughly constant. Any slow variation in screening throughout the series should be reflected in q, while a different valence state should appear as an abrupt change. To factor out the a_o^{-3} effect due to the gradual variation in lattice constants, we have plotted $\left(a_o^3\right)(e^2qQ/h)$ vs atomic number. Figures 3 and 4 show the data for the RIn_3 and RSn_3 compounds with $AuCu_3$ structure as well as the lattice constants of ref. 4. The near constancy of $\left(a_o^3\right)(e^2qQ/h)$

throughout the series argues strongly for the rare-earth ions being in the same valence state--presumably +3--and for the screening effect of the valence electrons being essentially the same across the rare earth row in these compounds. Dramatic drops occur for $YbIn_3$, $EuSn_3$, and $YbSn_3$. The lattice constant variations for these three compounds from the +3 line are of the order 1-2%, and clearly cannot account for the magnitude of the reductions. The percentage deviation in the quadrupole coupling constants is ~30-50 times larger than that noted in the lattice constants, confirming the great sensitivity of this parameter to the rare-earth valence state.

Room-temperature susceptibilities [1,21], lattice constants [1-4], and Mössbauer spectra [5-8] are all consistent with the assignment of the divalent state to the rare earth in these three compounds at room temperature. Gschneidner [22] evaluated thermodynamic heat of formation data for a number of Yb and Eu compounds. He estimated that for Yb or Eu to be trivalent in a particular compound would require the heats of formation for the trivalent lanthanides to be more negative than roughly -9 and -23 kcal/gm-at. of rare earth respectively. One thus generally expects Yb and especially Eu to be divalent in all but the most ionic or covalent compounds.

A quantitative justification of the magnitude of the coupling constants is beyond the scope of this paper. Instead we suggest that an empirical approach based on reproducing the qualitative trends in the data can provide useful insights into the valence states found in these alloys. The approximate empirical relation which we will use to explain the magnitude of the coupling constants is

$$|e^2qQ/h| \approx 40(Z_{RE} - Z_{In,Sn})\ \text{MHz} \quad . \qquad (6)$$

The coefficient of 40 for ΔZ can be obtained empirically by noting that $|e^2qQ/h|$ changes by ~40 MHz between the RIn_3 and RSn_3 compounds. As a cross-check, it can also be arrived at qualitatively by assigning what we consider to be reasonable values to the parameters of eq. (5) as follows:

$a_o \simeq 4.65 Å$

$Q \simeq 0.44$ barns (ref. 18)

$|1 + F_e| \simeq 1$, consistent with $F_e \simeq -2$ to -3 (ref. 17)

$\gamma_\infty(Cd) \simeq -29.27$ (ref. 23)

The insertion of these values into eq. (5) yields 39.98 $(Z_{RE} - Z_{In,Sn})$, with considerable error because the literature values of the quadrupole moment Q alone span a considerable range.

The assignment of valencies for the rare earths and In, Sn requires additional considerations. In the RIn_3 series the assignment of +3 for In and +3 for the rare earths (excpt for Yb) would cause the electric field gradient to vanish identically. This is a rigorous symmetry property of an FCC lattice in which all sites are equivalently charged. Based on the electronegativities tabulated by Teatum[24] et al., one expects electronic charge to be drawn toward the In and Sn atoms. The assignment of +1 for In and +2 for Sn would result in certain trends predicted from eq. (6). Considering first the RIn_3 compounds with $Z_{RE} = +3$ and $Z_{In} = +1$, we expect a coupling constant of order 80 MHz. The experimental values across the lanthanide series lie between 72 and 88 MHz. For $YbIn_3$ with a divalent rare earth one expects a 50% reduction to 40 MHz, while the measured value is 38.7 MHz. In addition, one expects that if Sn is divalent the Sn series with trivalent rare earths should yield coupling constant of order 40 MHz. Table II shows that the values lie between 34 and 43 MHz. For $Z_{Sn} = +2$ however, the compounds with divalent rare earths ($EuSn_3$ and $YbSn_3$) should have coupling constants equal to zero by symmetry. The $EuSn_3$ coupling constant is 12.7 MHz, while that of $YbSn_3$ is higher at 21.9 MHz. Both of these values are dramatically reduced from the coupling constants of the trivalent rare earths (approximately 40 MHz) but they are certainly not zero. As an additional check on this point, we measured the coupling constant for ^{111}In in $BaSn_3$, and found a coupling constant of approximately 19 MHz. The simple point charge model employed above is reasonably successful in explaining the qualitative trends of the coupling constants in the RIn_3 and RSn_3 series but fails to provide quantitative agreement with all compounds of the series.

The purpose of the above estimates was not to justify a point charge model or our assignment of the charges to the rare earths, In and Sn. Rather, we aimed to emphasize that a simple picture of the field gradient can yield useful insight into the qualitative trends of the coupling constants in these alloys.

We now address the question of the temperature and pressure dependence of rare earth valence fluctuations in these alloys. The room temperature measurements have shown just how sensitive a probe coupling constants are of the rare earth valence state.

Susceptibility [1,21], thermal expansion [4], and lattice constant [4] data had indicated that $YbIn_3$ and $CeSn_3$ appeared to become trivalent and partially tetravalent respectively on cooling to liquid helium temperatures, and that $YbSn_3$ might exist in a mixed valence state even at room temperature. We have measured e^2qQ in these compounds at 4.2K and find shifts of order 10-15%. The magnitude of these shifts is not large enough to be representative of a <u>bulk change</u> in valence state, especially when one corrects for the thermal contraction of the lattice and the $1/a_o^3$ shift which results therefrom. Figure 2 also shows that $CeSn_3$ in non-magnetic at 4.2K, in accord with Mössbauer data. [5] The interpretation of $YbSn_3$ is complicated by its high coupling constant (21.9 MHz) at room temperature and the fact that on cooling to 4.2K the PAC pattern distorts from that expected for axial symmetry. We feel that the room temperature coupling constant for $BaSn_3$ (19 MHz) supports the interpretation that Yb is divalent in this compound. We have no explanation for the apparent deviation from an axially-symmetric pattern at 4.2K.

$YbIn_3$(RT), $YbSn_3$(4.2K), and $CeSn_3$(RT and 4.2K) were also run in a clamping-type pressure cell to 18.5 kbar to see if we could induce a rare earth valency fluctuation. Tables 1 and 2 show that the shifts are of the order of those associated with contraction from cooling to liquid helium temperatures, but in no case did we observe a shift which would put the points on the "trivalent line" of figs. 3 and 4. We conclude that $CeSn_3$, $YbSn_3$, and $YbIn_3$ show no evidence for a bulk valence instability down to 4.2K and pressures to 18.5 kbar. Previous Mössbauer data on $CeSn_3$ [5] and $YbSn_3$ [7] have come to similar conclusions.

In summary we have systematically studied the RIn_3 and RSn_3 compounds with $AuCu_3$ structure using TDPAC and have demonstrated the high sensitivity of the technique to the rare-earth valence state. We have probed the temperature and pressure dependences of the rare earth valence in $CeSn_3$, $YbSn_3$, and $YbIn_3$ and find no evidence for bulk valence fluctuations in these compounds.

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Table 1. RIn_3 parameters.

Compound		Lattice Constant[a] $(kx)^b$	$\lvert e^2qQ/h \rvert$ (MHz)
$LaIn_3$		4.7250	72.5±1.1
$CeIn_3$		4.6798	77.9±1.2
$PrIn_3$		4.6622	78.5±1.2
$NdIn_3$		4.6460	80.0±1.2
$SmIn_3$		4.6166	81.4±1.2
$GdIn_3$		4.5973	84.2±1.3
$TbIn_3$		4.5804	86.2±1.3
$DyIn_3$		4.5699	86.4±1.3
$HoIn_3$		4.5640	86.7±1.3
$ErIn_3$		4.5552	87.7±1.3
$TmIn_3$		4.5492	87.7±1.3
$YbIn_3$	(RT)	4.6053	38.7±0.8
	(4.2°K)		43.4±0.9
	(RT, 18.5 kbar)		45.0±1.4
$LuIn_3$		4.5434	86.7±1.3

[a] Ref 4, as cast, room temperature.

[b] 1 kx = 1.002495Å.

Table 2. RSn_3 parameters.

Compound	Lattice Constants[a] (kx)[b]	$\|e^2qQ/h\|$ (MHz)
$LaSn_3$ (RT)	4.7598	33.9±1.0
(4.2K)		35.0±1.1
$CeSn_3$ (RT)	4.7119	39.8±1.2
(RT, 18.5 kbar)		43.7±1.3
(4.2K)		43.8±1.3
(4.2K, 18.5 kbar)		47.0±1.4
$PrSn_3$ (RT)	4.7064	41.5±1.2
$NdSn_3$ (RT)	4.6965	41.4±1.2
$SmSn_3$ (RT)	4.6772	42.6±1.3
$EuSn_3$ (RT)	4.7349	12.7±1.3
$GdSn_3$ (RT)	4.6681	42.5±4.2
$YbSn_3$ (RT)	4.6720	21.9±1.1
(4.2K)		25.6±1.3
(4.2K, 18.5 kbar)		29.7±1.8

[a] Ref. 4, as cast, RT.

[b] kx = 1.002495Å

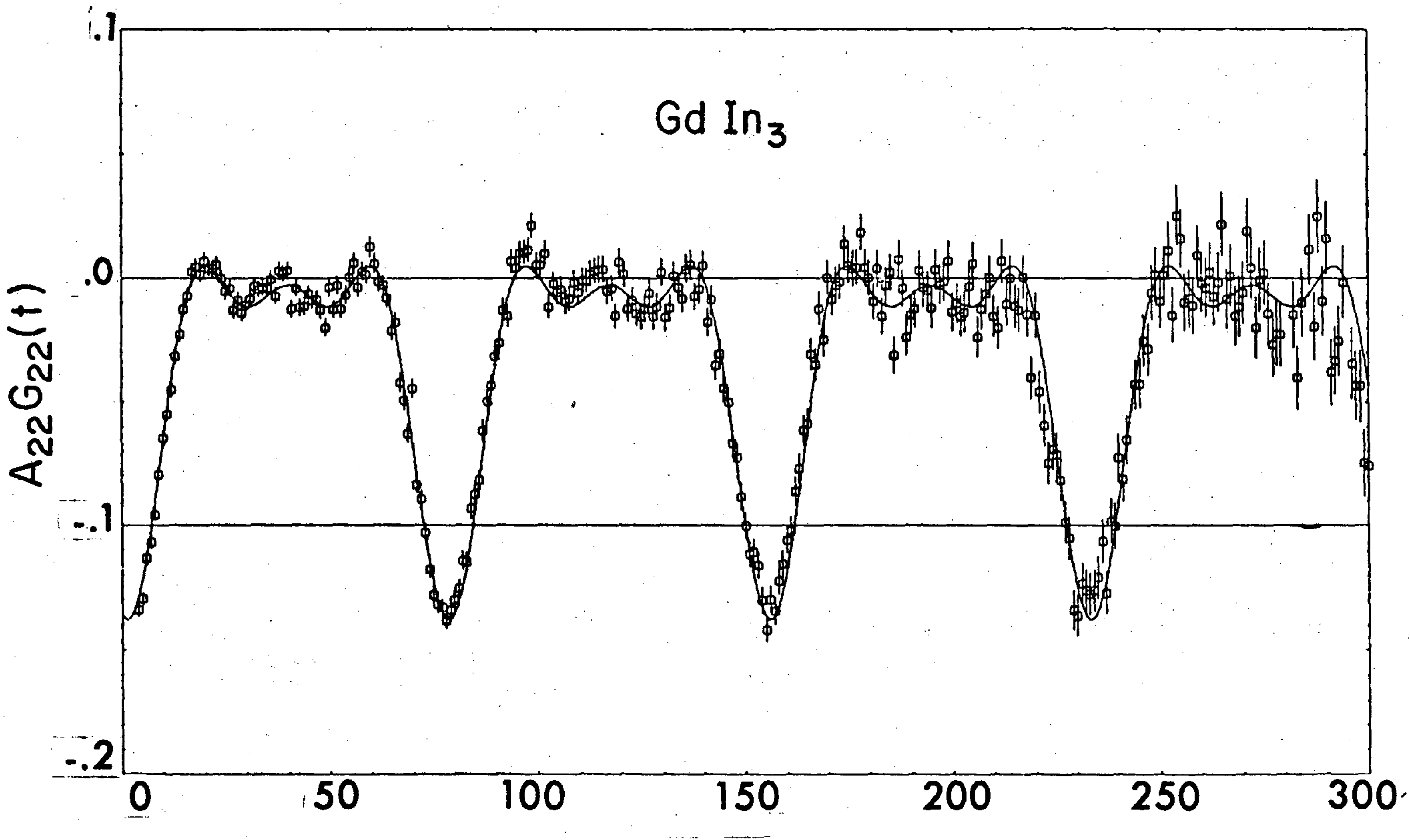
Figure captions

Fig. 1. Experimental and least-squares fit (solid curve) time-modulation spectrum for ^{111}In in GdIn$_3$. The time calibration is 1.03 ns/channel.

Fig. 2. Experimental and least-squares fit (solid curve) time-modulation spectra of ^{111}In in CeSn$_3$ at 290 and 4.2K. The time calibration is 1.62 ns/channel.

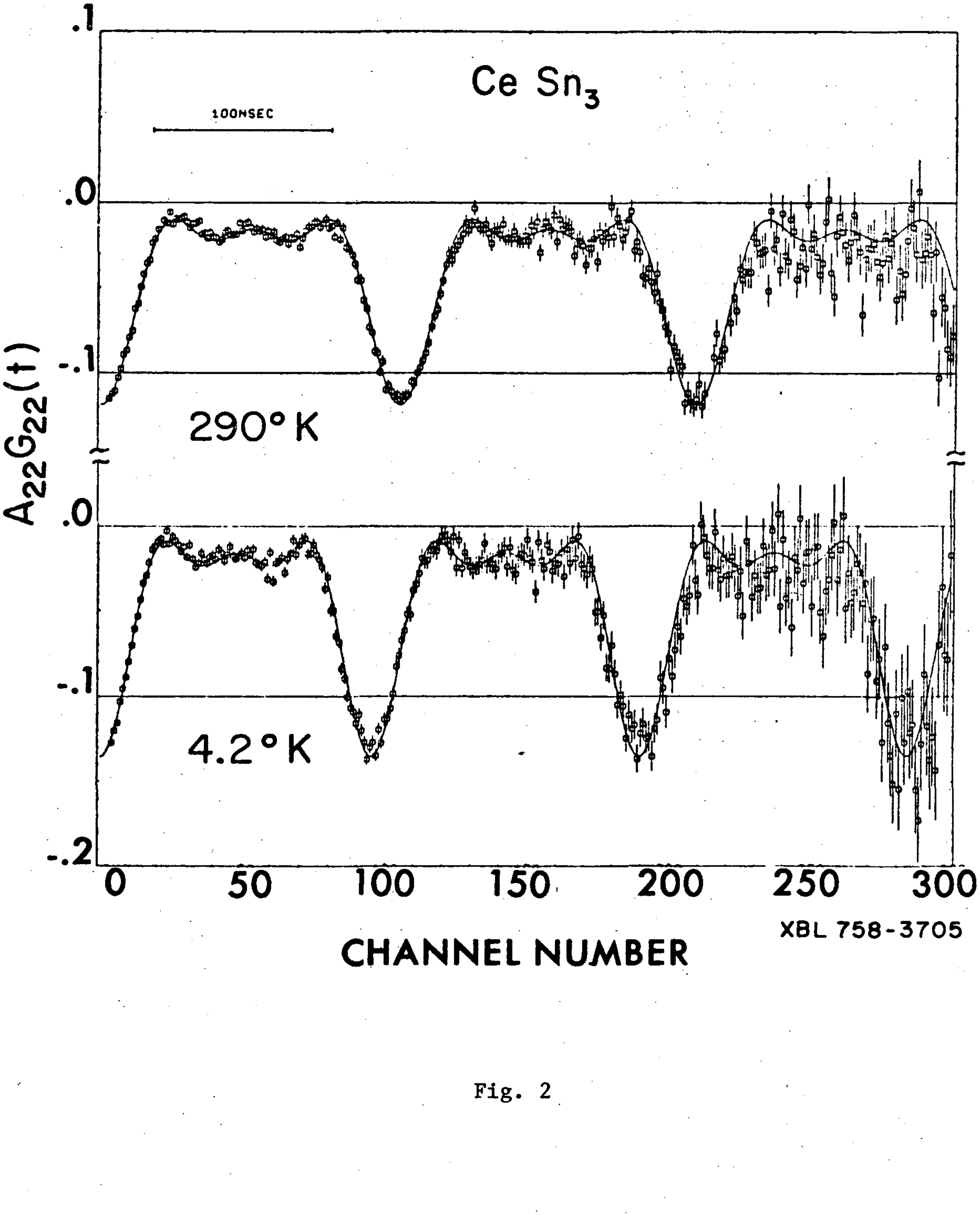
Fig. 3. Lattice constants and quadrupole coupling data for RIn$_3$ compounds plotted against atomic number. Inset shows variations for YbIn$_3$ with temperature and pressure changes.

Fig. 4. Lattice constants and quadrupole coupling data for RSn$_3$ compounds plotted against atomic number. Note low values of the quadrupole parameter for EuSn$_3$ and YbSn$_3$.



XBL 758-3706

Fig. 1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100NSEC
.1
.0
-.1
-.2
290° K
4.2° K
A₂₂G₂₂(t)
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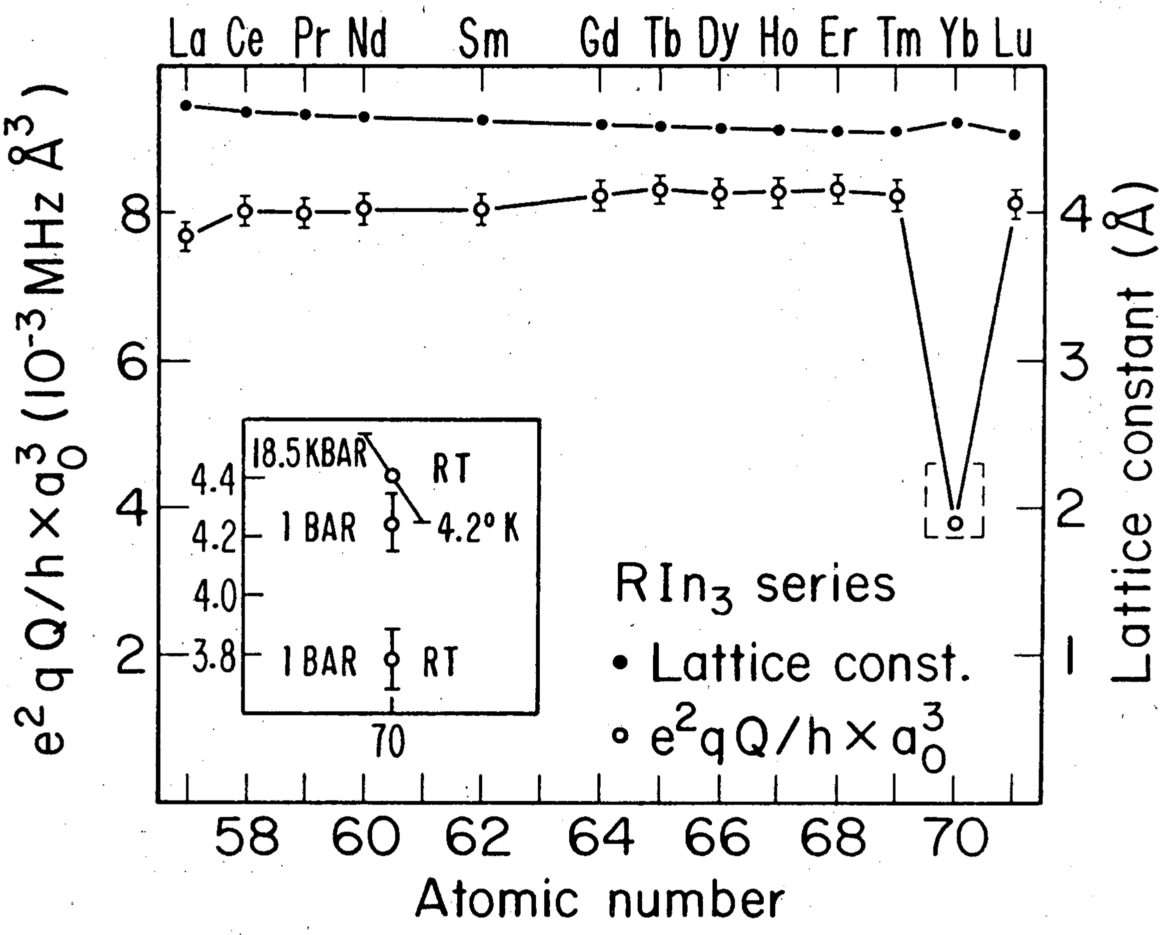
50
100
150
200
250
300
CHANNEL NUMBER
XBL 758-3705

Fig. 2



XBL 758-3708

Fig. 3

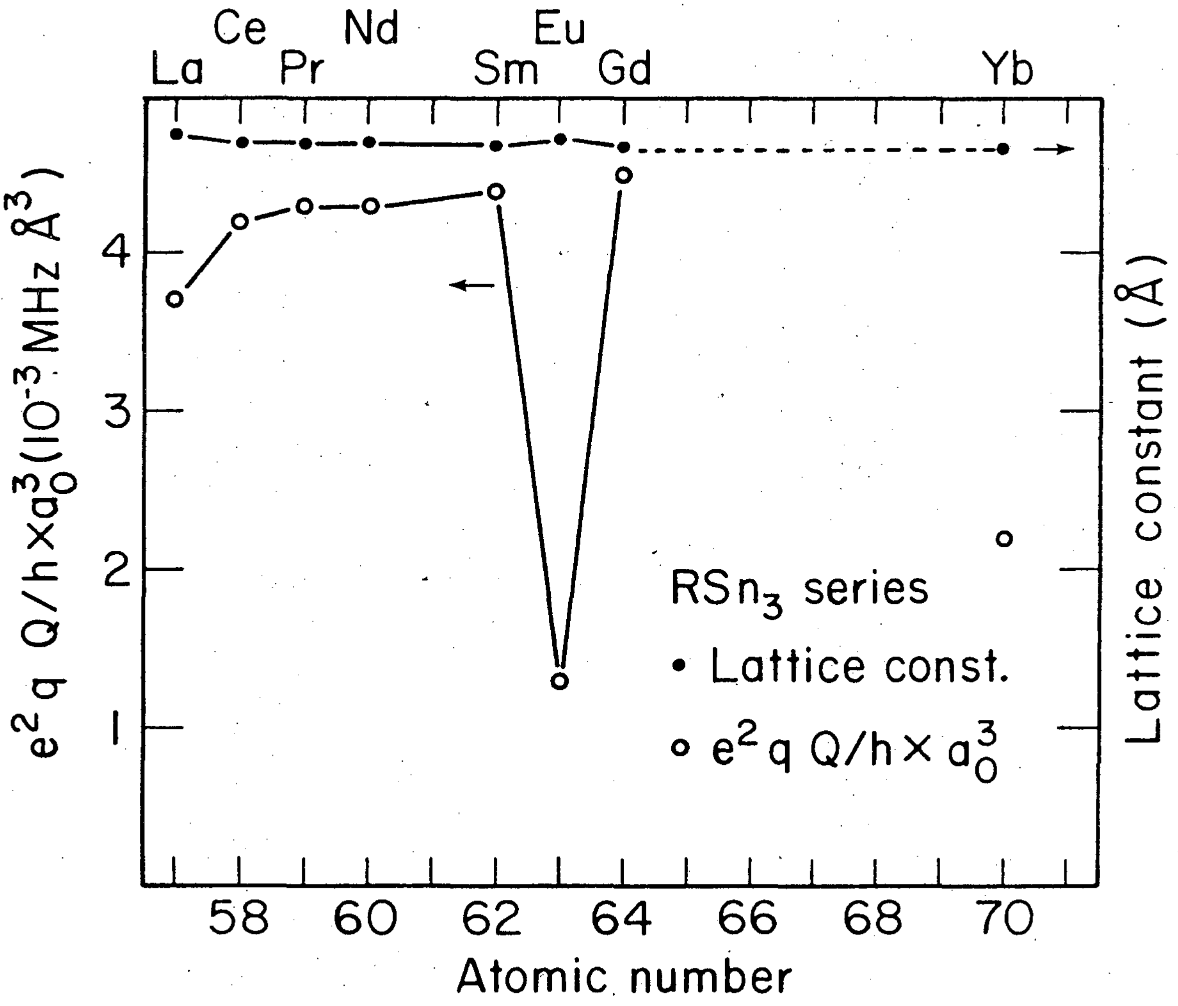


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Fig. 4

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