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Hyperfine Interactions and Anisotropic Lattice Vibrations of ^{237}Np in α-Np Metal*

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Hyperfine spectra utilizing the nuclear γ-ray resonance (Mössbauer effect) of the 59.6-keV γ ray of ^{237}Np have been obtained in α-Np metal. For the temperature range 1.7–48°K, the spectra show no magnetic effects and can be described as two quadrupole interactions having widely differing values of electric field gradient and asymmetry parameter. These two spectra can be related to the two inequivalent sites in the orthorhombic unit cell of α-Np. The intensities of the hyperfine lines show the presence of anisotropic lattice vibrations (Goldanskii-Karyagin effect). At one site the lattice vibrations are only slightly anisotropic, while for the other site a maximum rms deviation of ≈ 0.1 Å is deduced.

I. INTRODUCTION

NUCLEAR γ-ray resonance (the Mössbauer effect) of the 59.6-keV transition in ^{237}Np has proven a useful tool for the investigation of neptunium systems. Because of the large hyperfine fields and isomer shifts[1] observed, the resonance spectra are quite sensitive to details of the magnetic structure, the crystal symmetry, etc. This is true even though natural linewidth has not yet been achieved and line broadenings of a factor of 10 are still present[2] in the best source matrices available. A number of chemical[1,3] and intermetallic[4] compounds have been studied in some detail. In this paper, we report the investigation of the hyperfine spectra of ^{237}Np in α-Np metal in the temperature region between 1.7 and 48°K.

The structure of α-Np metal has been investigated in detail by Zachariasen.[5] The orthorhombic unit cell, as determined at room temperature from powder diffraction data, contains eight atoms divided equally into two nonequivalent sites, designated Np(I) and Np(II). The lattice may be considered to be a bcc structure which has been highly deformed so that the coordination number is reduced from 8 to 4, as shown in Fig. 1(a). The nearest neighbors for each of the sites are approximately located on four of the five corners of a trigonal bipyramid, but with four different corners of the bipyramid being occupied in each case. This rough coordination is shown in Fig. 1(b).

The present data on isomer shifts and electric field gradients (efg) could be used to test details of electronic structure, but no complete models are presently available. Such discussions are difficult because α-Np is a semimetal, in which covalency effects are believed to also be of importance. Zachariasen,[5] adopting a purely chemical approach, has made an attempt to estimate the number of electrons participating in the bonds. A similar approach has been made by Sarkisov.[6] Both conclude that comparisons of the metallic radii in the actinide metals indicate six electrons are bonding, leaving a single-valence electron on the atom. This type of a treatment, however, suffers from having ignored the metallic nature of the material.

In contrast to the above, pure band models have been given by a number of people. These have been discussed by Brodsky,[7] who concludes that a suitable structure consists of a broad, almost filled s-band overlapping with a relatively narrow, almost empty f-d hybrid band. On the basis of such a two-band model, Hall-coefficient data were interpreted to calculate the number of holes in the s band to be $n_h \approx 0.1$ per atom.

II. EXPERIMENTAL

The bulk of the measurements were made on a neptunium metal sample prepared by the reduction of NpO$_2$ by MgZn alloy in contact with a molten salt phase. The product obtained after the distillation of the Mg and Zn was not analyzed chemically because of the small amount obtained, but is thought to be quite pure. Metallographic examination showed it to contain far less quantities of insoluble impurities than had been observed in samples containing 0.1–0.2 wt% of impurities by analysis. The density of the ascast metal, 20.42 g/cm^3, agrees well with the x-ray density of 20.45, and is to be compared to 20.2±0.05 found for the impure metal mentioned above. Magnetic susceptibility measurements showed the presence of no more than 100 ppm atoms of Np as ferromagnetic impurities (e.g., NpN or NpC). The absence of a maximum in the susceptibility-temperature curve at 22°K, the antiferromagnetic transition of NpO$_2$, leads to an upper limit of 1 mole % of NpO$_2$. The density determination sets the upper limit for NpO$_2$ at 0.3 mole %.

The metal foil was prepared by cross rolling in the α phase at 430°K. An x-ray examination of similarly prepared foils has not shown the presence of any preferred orientation. The diffraction rings were fairly

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* Based on work performed under the auspices of the U. S. Atomic Energy Commission.

[1] B. D. Dunlap, G. M. Kalvius, S. L. Ruby, M. B. Brodsky, and D. Cohen, Phys. Rev. **171**, 316 (1968).

[2] G. M. Kalvius, B. D. Dunlap, S. L. Ruby, and M. B. Brodsky, Bull. Am. Phys. Soc. **12**, 25 (1967).

[3] J. A. Stone and W. L. Pillinger, Symp. Faraday Soc. **1**, 77 (1968).

[4] B. D. Dunlap, M. B. Brodsky, G. M. Kalvius, G. K. Shenoy, and D. J. Lam, J. Appl. Phys. **40**, 1495 (1969).

[5] W. H. Zachariasen, Acta Cryst. **5**, 660 (1952).

[6] E. S. Sarkisov, Dokl. Akad. Nauk SSSR **166**, 627 (1966) [English transl.: Soviet Phys.—Doklady **166**, 135 (1966)].

[7] M. B. Brodsky, in *Plutonium 1965*, edited by A. E. Kay and M. B. Walkron (Chapman and Hall Ltd., London, 1967), p. 286 ff.

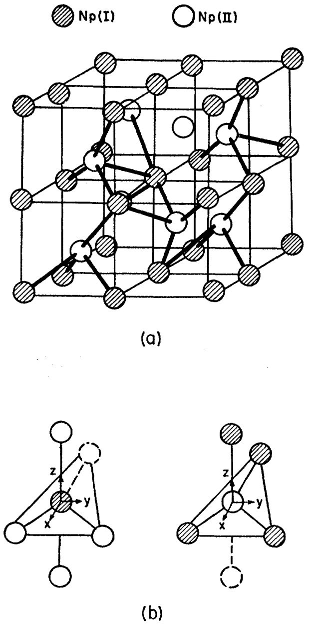

FIG. 1. Lattice structure of α-Np. (a) Basic structure showing distortion from a bcc lattice. The heavy lines show the first nearest neighbors for a few of the atoms. (b) Approximate nearest-neighbor configurations for the two inequivalent sites. The coordinate axes are used in the lattice dynamical discussion, as described in the text.

sharp and it is likely that recrystallization had occurred during rolling. The sample received no further heat treatment.

Measurements were carried out inside a vertical tube going through the helium bath of a cryostat described earlier.[8] Thermal contact is established via helium exchange gas. To obtain temperatures above the bath temperatures, the exchange gas is pumped off and the sample is heated electrically. Temperatures are read with a platinum or germanium resistance thermometer, depending on the temperature region. A negative feedback circuit operating on the heater current keeps the temperature within 0.05°K of the preset value. The velocity spectrometer uses an electromechanical transducer, operated in sinusoidal velocity sweep, which is mounted on the warm side of the exchange-gas column. Motion is fed to the absorber via a stainless-steel tube. The spectra were collected in a 400- or 1024-channel analyzer operated in time mode.

Spectra were analyzed using a least-squares routine which fits directly to physical parameters such as the quadrupole interaction energy, the asymmetry parameter, and the isomer shift. Part of this fitting routine has been described elsewhere, in particular its application to pure quadrupole spectra.[9]

III. RESULTS

The nuclear γ-ray resonance of α-Np consists of ten rather well-resolved lines, shown schematically in the upper part of Fig. 2. This spectrum remained essentially the same over the entire temperature region investigated (1.7–48°K). The observed line positions can be fully explained by a pure quadrupole interaction, as will be shown below. Thus the present data show, in agreement with magnetic susceptibility and transport property measurements,[10] that α-Np metal does not magnetically order down to 1.7°K.

Since Np occupies two different sites in the α-Np lattice, we expect the resonance pattern to consist of two overlapping quadrupole spectra. The quadrupole interaction Hamiltonian for each site can be written in the form

$$H_Q=[e^2qQ/4I(2I-1)][3I_z^2-I^2+\tfrac{1}{2}\eta(I_+^2+I_-^2)], \quad (1)$$

where I, I_z, I_+, and I_- are the nuclear angular momentum operators and Q is the quadrupole moment of the nuclear state in question. The principal component of

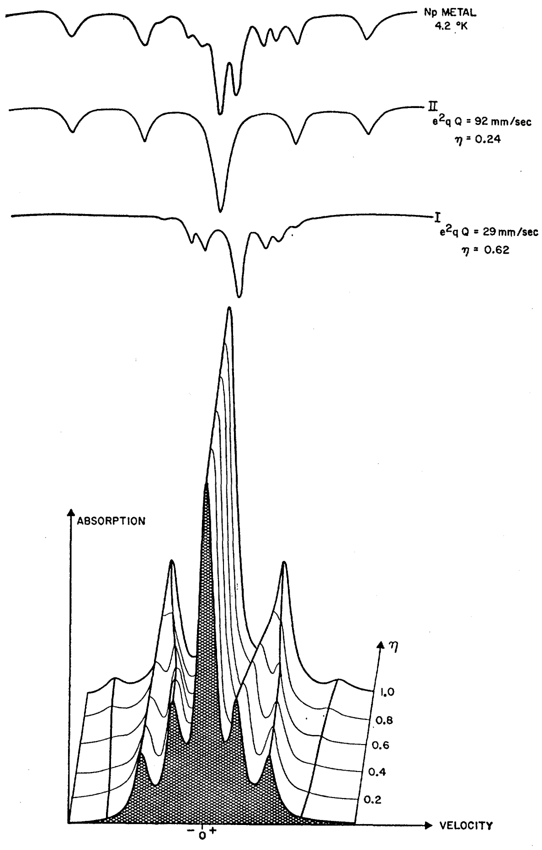

FIG. 2. Hyperfine spectrum of ^{237}Np in α-Np. The surface at the bottom gives a typical quadrupole spectrum as a function of the asymmetry parameter η. Above, the full spectrum of the metal is shown decomposed into the component spectra I and II.

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[8] G. M. Kalvius, in *Mössbauer Effect Methodology*, edited by I. Gruverman (Plenum Press, Inc., New York, 1965), p. 163 ff.

[9] G. K. Shenoy and B. D. Dunlap, Nucl. Instr. Methods **71**, 285 (1969).

[10] M. B. Brodsky (unpublished).

TABLE I. Hyperfine interactions in α-Np.

Temperature (°K)	Site	$\lvert e^2qQ\rvert$ (mm/sec)	$\lvert eq\rvert$[a] (10^{18} V/cm^2)	η	Isomer shift[b] (mm/sec)
4.2	Np(I)	29.3 ± 0.6	1.45	0.62 ± 0.04	2.0 ± 0.3
	Np(II)	92.4 ± 1.0	4.57	0.24 ± 0.02	-0.7 ± 0.3

[a] Assuming $Q=4$ b.
[b] Relative to NpO_2.

the efg is $eq=V_{zz}$, and $\eta=(V_{yy}-V_{xx})/V_{zz}$ is the asymmetry parameter. The 59.6-keV γ ray in ^{237}Np arises from an electric monopole transition between the second excited state and the ground state. Both states have a spin of $I=\frac{5}{2}$ and will therefore be split into three doubly degenerate sublevels. For $\eta=0$, the energy matrix is diagonal in I_z, and $E1$ selection rules permit seven γ transitions. Earlier measurements have shown[3,11] the quadrupole moments of the two states to be almost equal ($Q^*/Q=0.99\pm0.02$). This causes three of the seven transitions to nearly coincide in energy, giving a symmetric five-line pattern. In the case of a nonvanishing asymmetry parameter, I_z is no longer a good quantum number, and the nuclear hyperfine levels are formed by admixtures of I_z basis states. The line positions and their intensities are strongly dependent on η, and two additional weak lines appear.

A ^{237}Np quadrupole pattern with typical experimental linewidth has been plotted as a function of η in the lower part of Fig. 2. It is quite apparent that the line positions allow accurate determination of the absolute value of q and of η, however, due to the symmetry of the spectrum, the sign of q can not be obtained. We now can decompose the hyperfine spectrum of α-Np into two quadrupole patterns, marked I and II in Fig. 2, which are characterized by rather different values of q and η and slightly different isomer shifts. The correspondence between these component spectra and the two lattice sites is discussed below. The hyperfine parameters obtained at 4.2°K are given in Table I, and are in general agreement with those given by Pillinger and Stone.[12] These values remain practically constant over the entire temperature range investigated (1.7–48°K).

IV. DISCUSSION

A. Hyperfine Integrations

The efg tensor can be written as a sum of several terms, whose relative magnitudes will depend critically on the properties of the crystalline lattice and the electronic configuration. For a semimetal having strong covalent bonding, like α-Np, the principal component of the efg at each lattice site can be expressed as

$$eq=(1-\gamma_\infty)eq_L+eq_c+(1-R_Q)\langle eq_v\rangle_T+eq_b.$$

Here eq_L is the field gradient produced by the other atoms in the lattice, eq_c is the contribution from non-s character conduction electrons, $\langle eq_V\rangle_T$ is the thermal average of the field gradient due to valence electrons resident on the atom, and eq_b arises from the bonding electrons. The quantities γ_∞ and R_Q are the appropriate Sternheimer factors which describe the modification of the field gradient by the filled inner electron shells. Similar equations can be used to describe V_{xx} and V_{yy}, and thus η. With the limited information on electronic structure available, only some rather general statements can be made concerning the quadrupole interaction.

The most straightforward procedure is a calculation of the lattice field gradient by assuming a simple point-charge model. Such a calculation has been done presuming that the low-temperature phase is the same as that at room temperature, and using the room-temperature lattice constants. Summation over a sphere of about ten times the nearest-neighbor distance is sufficient to obtain convergence of the various terms. Taking a value of $(1-\gamma_\infty)\approx150$ appropriate for the actinide region,[13] one finds values for q_L in V/cm^2 of $1.7\times10^{18}z$ and $1.4\times10^{18}z$ for sites I and II, respectively, where z is the ionic charge on the Np atoms. These values are no doubt overestimated because conduction electrons tend to shield the more distant charges from the central atom. However, even if one assumes all charges except the nearest neighbors to be completely shielded, the values of q_L do not reduce by more than a factor of 2. It is thus seen that in this case the lattice contribution is quite substantial. One should note that q_L(I) is only slightly larger than q_L(II), while the experimental values for the two gradients differ by a factor of 3. However, the asymmetry parameters from the lattice charges are found to $\eta_L(\text{I})=0.37$ and $\eta_L(\text{II})=0.17$ from the calculation. This difference of a factor of 2 is closer to the experimental ratio of asymmetry parameters and primarily on this basis we have assigned the hyperfine pattern I (of Fig. 2) to the atoms on the site Np(I) and the pattern II to those on the site Np(II). These assignments are further substantiated by lattice dynamical aspects of the problem which are discussed in Sec. IV C below.

In addition to the shielding effects mentioned above, non-s conduction electrons may make a direct contribution (q_c) to the field gradient. This term is expected[14] to be proportional to q_L and roughly of the same order of magnitude. Frequently q_c is negative[14] and so may tend to cancel the large contribution of q_L to the field gradient.

Since covalent bonding is present, we must consider separately those electrons in valence bands which are bonding and those which are nonbonding. The latter are

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[11] J. A. Stone and W. L. Pillinger, Phys. Rev. **165**, 1319 (1968).
[12] W. L. Pillinger and J. A. Stone, Bull. Am. Phys. Soc. **13**, 28 (1968).
[13] R. M. Sternheimer, Intl. J. Quantum Chem. **1S**, 67 (1967).
[14] R. E. Watson, A. C. Gossard, and Y. Yafet, Phys. Rev. **140**, A375 (1965).

subject to crystal field effects in the usual way, and give rise to the term $\langle q_V \rangle_T$. This provides the principal temperature dependence of the field gradient, due to population changes in the various crystal field states. In general this term is large. For example, we have previously measured[15] the field gradient from a single $5f$ electron in the neptunyl ion to be 5×10^{18} V/cm^2. In the present case, however, one does not know what electrons states are contributing. The fact that the field gradient is temperature-independent up to 48°K suggests either that the first excited crystal field level is 300 cm^{-1} or more, or that this contribution is negligible because all such electrons are in conduction bands. The latter conclusion is substantiated by susceptibility data[10] which shows α-Np to be a temperature-independent paramagnet. The field gradient from bonding electrons (q_b) can not be estimated at the present state of knowledge. It is, however, obvious that its value will be quite different for the two sites since the spatial distribution of the bonds is determined by the local symmetry. This term is most likely responsible for the observed difference in the two field gradients.

B. Isomer Shifts

A difficulty similar to that above is encountered when interpreting the isomer shifts in that one must consider contributions from a number of terms which are of comparable magnitude. Nonrelativistic Hartree-Fock calculations show that in the vicinity of the free atomic configuration ($5f^4 6d 7s^2$), the following rules apply: (i) removing a $7s$ electron has about the same effect on the electron density at the nucleus, and hence on the isomer shift, as adding a $6d$ electron, and (ii) adding a $5f$ electron changes the isomer shift by about 50% more than removing a $7s$ electron. It is thus not possible to assign a unique configuration from the isomer-shift data alone. Using these relative strengths and our previous isomer-shift calibration,[1] however, one can set certain limits on the kinds of electrons present. Assuming that all seven electrons outside the radon core are in conduction bands, one finds that the observed shifts can not be obtained without having the s band at least half full and without having a substantial f-d hybridization. For example, if we believe the s band to contain approximately 1.9 electrons, as suggested by the Hall-effect data,[7] then, in order to obtain the observed shifts, the remaining 5.1 electrons must have approximately equal amounts of f and d character. The difference in isomer shifts for the two sites is fairly small, corresponding to a difference of about one-tenth of an f electron in the two electronic configurations. Such a difference may be caused, for example, by the difference in hybridization required by the different bond directions of the two lattice sites. At present, however, quantitative calculations are not possible.

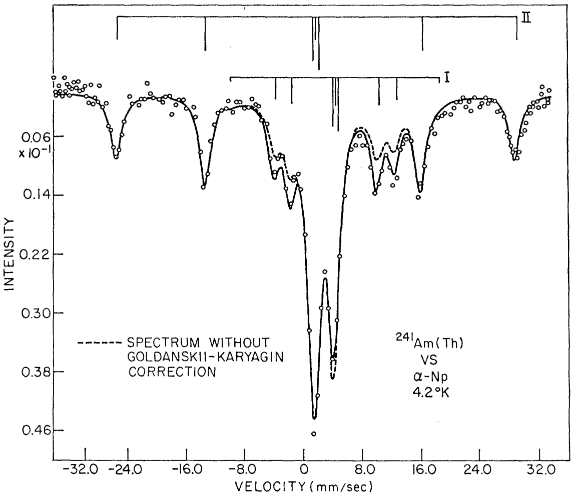

FIG. 3. Hyperfine spectrum of ^{237}Np in α-Np. The solid line is a computer fit including the Goldanskii-Karyagin correction.

C. Anisotropic Lattice Vibrations

The discussion so far has been based solely on line positions. We wish now to also examine the intensities. The variation of the line intensities as a function of η has been incorporated into the least-squares fitting routine. In the fitting process, each of the component spectra is adjusted by an over-all scaling intensity which (in the thin absorber approximation) should be proportional to the product of the population and the recoil-free fraction for that site. The fit obtained in this way is shown by the dotted curve in Fig. 3. We observe that while the fit is satisfactory for site II, the intensities for site I are poorly reproduced. In addition, the scaling intensities for the two sites were in the ratio 2:3. Since the sites are equally populated, this would imply a large difference in the recoil-free fractions for the two sites. One expects such a large difference to cause a strong temperature dependence in the relative intensities of the two sites, however, none is found. Partial saturation of the intensities due to absorber thickness, can produce anomalous line intensities. A detailed study of absorber thickness effects using a source similar to that used here and NpO$_2$ absorbers has been performed by Stone and Pillinger,[3] who find that no thickness correction need be made for absorbers as thick as 300 mg/cm^2. Since metals usually have lower recoil-free fractions than oxides, one expects this result to be even more valid here. Our data taken on two α-Np foils having thicknesses of 200 and 100 mg/cm^2 gave identical results.

The other mechanisms which possibly could produce intensity discrepancies are paramagnetic relaxation effects,[16] preferential orientation of crystallites, and anisotropic lattice vibrations (Goldanskii-Karyagin

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[15] S. L. Ruby, G. M. Kalvius, B. D. Dunlap, G. K. Shenoy, M. B. Brodsky, D. Cohen, and D. J. Lam, Phys. Rev. 184, 374 (1960).

[16] H. H. Wickman and G. K. Wertheim, in *Chemical Applications of Mossbauer Spectroscopy*, edited by V. I. Goldanskii and R. H. Herber (Academic Press Inc., New York, 1968), pp. 548–622.

effect).[17] Relaxation effects are unlikely here because the spectra do not show the characteristic changes in the line shape associated with such large differences in intensity and also because the shapes of the lines are temperature-independent. If the crystallites are oriented preferentially, discrepancies in intensities should be seen in both component spectra, while we find only site I affected. Thus the explanation seems to lie in the Goldanskii-Karyagin effect. This seems especially appealing here because this effect will alter the relative intensities of various lines, in different ways depending on the radiation patterns of the component lines. A careful examination of the observed discrepancies between the experimental data and the dashed line of Fig. 3 assures that such an explanation is possible.

In a powder sample, the intensity of line j will be given by

$$I_j=\int_0^{2\pi}\int_0^{\pi} f(\theta,\varphi)I_j(\theta,\varphi)\sin\theta\, d\theta\, d\varphi\,, \tag{2}$$

where $I_j(\theta,\varphi)$ gives the angular dependence of the radiation field for this transition and $f(\theta,\varphi)$ is the angular-dependent recoil-free fraction. This latter factor is given by

$$f(\theta,\varphi)=\exp(-|\langle\mathbf{k}\cdot\mathbf{r}\rangle|^2)\,, \tag{3}$$

where $\mathbf{k}$ is the γ-ray wave vector, $\mathbf{r}$ is the nuclear displacement from equilibrium, and the angular bracket represents a thermal average. This quantity is a second-order tensor, and by a suitable choice of principal axes can be reduced to diagonal form

$$\ln f(\theta,\varphi)=-k^2(\langle x^2\rangle\sin^2\theta\cos^2\varphi+\langle y^2\rangle\sin^2\theta\sin^2\varphi+\langle z^2\rangle\cos^2\theta)\,. \tag{4}$$

It is convenient to transform this to the form[17]

$$\ln f(\theta,\varphi)=-k^2\langle z^2\rangle+\sin^2\theta(a+b\cos^2\varphi)\,, \tag{5}$$

where

$$\begin{aligned}a&=k^2[\langle z^2\rangle-\tfrac{1}{2}(\langle x^2\rangle+\langle y^2\rangle)]\,,\\ b&=\tfrac{1}{2}k^2[\langle y^2\rangle-\langle x^2\rangle]\,.\end{aligned} \tag{6}$$

If the lattice vibrations are isotropic, then $a=b=0$, and f has no angular dependence. In that case, the intensities given by Eq. (2) contain only nuclear factors. This assumption was made to produce the surface in Fig. 2 and the dashed line in Fig. 3. If the lattice vibrations are axially symmetric, then $b=0$, $a=k^2(\langle z^2\rangle-\langle x^2\rangle)$, and one obtains the angular dependence of the recoil-free fraction in a form frequently discussed in the past.[18] In this case the integral of Eq. (2) is considerably simplified since the φ dependence of f vanishes, but nonetheless significant departures in intensities from the isotropic case can be obtained. The higher the energy of the Mössbauer transition, the more sensitive the resonance will be to a lattice anisotropy because of the large value of k^2 in the exponentials of Eqs. (5) and (6).

In the present case the factors $I_j(\theta,\varphi)$ have the well-known angular dependence for an $E1$ transition if $\eta=0$. However, for $\eta\neq 0$, the nuclear levels are no longer pure I_z states and the expressions for the intensities have a more complicated form. For $E1$ transitions the intensity of each line can be expressed in the general form[17]

$$I_j(\theta,\varphi)=B_j^2\sin^2\theta'+\tfrac{1}{2}(C_j^2+D_j^2)(1+\cos^2\theta')+C_jD_j\sin^2\theta'\cos2\varphi'\,. \tag{7}$$

Here θ' and φ' are the angular coordinates of $\mathbf{k}$ with respect to the principal axes of the efg tensor. In general, these will not be the same as the angles θ and φ in Eq. (4), which give the coordinates of $\mathbf{k}$ with respect to the principal axes of the mean-square displacement tensor. In order to perform the integral of Eq. (2), it is then necessary to introduce the Euler angles relating the two coordinate systems, as described by Karyagin.[17] The coefficients Bj, Cj, and Dj are completely determined by the wave functions describing nuclear levels and can be calculated following the procedures of Ref. 17. For $\eta=0$ only one of these coefficients for each line will not vanish, and one obtains the familiar θ' dependence for a dipole transition.

In the general case, after introducing Eqs. (4) and (7) into Eq. (2), one can not express the integral in a closed form and the result has to be obtained numerically. The results of such a calculation are shown in Fig. 4 for the case of ^{237}Np with $\eta=0.62$. The intensity ratios of two pairs of lines are plotted as a function of a for different values of b. No theoretical calculations of lattice dynamical quantities are at present available. In particular, the normal-mode coordinates are not known. In order to obtain an estimate of the lattice anisotropy, the axes shown in Fig. 1, which are the approximate symmetry axes for the near-neighbor configurations, have been taken as principal axes for the f tensor. The principal axes of the efg tensor have been determined by a lattice sum calculation, and are related to the f tensor principal axes through the Euler angles[19] (0, −31°, 0) for site I and (0,75°,0) for site II. The arrows in Fig. 4 indicate the experimentally determined ratios for site I at 4.2°K.

Given two experimental ratios, one can solve numerically for a and b. Unfortunately, in the present case the observed intensity ratios lie in a region of only weak dependence on the anisotropy parameters, and an accurate determination of a and b is not possible. However, the sign and general size of these parameters can be determined. The results for both sites at 4.2°K are

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[17] S. V. Karyagin, Fiz. Tverd. Tela **8**, 1739 (1966) [English transl.: Soviet Phys.—Solid State **8**, 1387 (1966)].

[18] For example, see P. A. Flinn, S. L. Ruby, and W. L. Kehl, Science **143**, 1434 (1964).

[19] The Euler angles (α,β,γ) represent a rotation α around the z axis, β around the new y axis, and γ around the new z axis. With the signs of the angles as given, the f tensor principal axes are rotated into the efg principal axes.

TABLE II. Anisotropy parameters in α-Np at 4.2°K.

Site	a	b	$\langle y^2\rangle-\langle x^2\rangle$ (10^{-19} cm^2)	$\langle y^2\rangle-\langle z^2\rangle$ (10^{-19} cm^2)
Np(I)	-9 ± 3	3 ± 2	6 ± 4	12 ± 5
Np(II)	0 ± 0.5	-1 ± 5	0 ± 1	1 ± 5

given in Table II. For the site Np(II), having $\eta=0.24$, the lattice vibrations are roughly isotropic. For the site Np(I), where $\eta=0.62$, one finds $\langle y^2\rangle>\langle x^2\rangle>\langle z^2\rangle$. Referring back to Fig. 1, one recognizes that y is indeed the most free direction in the picture of next nearest neighbors only, while z is the most restricted. The maximum rms deviation is $(\langle y^2\rangle-\langle z^2\rangle)^{1/2}\approx0.1$ Å. It should be again emphasized, however, that this result is obtained with low accuracy.

The solid curve of Fig. 3 shows the final fit to the experimental spectrum, using the parameters of Table II. The relative intensities of the Np(I) lines are now well reproduced. Further, the fit now gives equal scaling intensities and hence equal populations and recoil-free fractions for the two sites.

V. CONCLUSION

In this investigation we find that the hyperfine spectrum of the 59.6-keV nuclear γ-ray resonance of ^{237}Np in α-Np metal is obtained from a pure quadrupole interaction in the temperature region between 1.7 and 48°K. The interpretation of this spectrum is in agreement with the known crystal structure. A detailed analysis of the line intensities reveals the existence of anisotropic lattice vibrations at the site where a large value of η indicates a low local symmetry.

In passing, we would like to make a remark regarding radiation damage in the neptunium samples. Resistivity[20] and other measurements on U, Np, and Pu clearly show effects due to the damaging effects of the α particle and recoiling daughter. It is well established in plutonium that most of this damage is retained at 4°K, and its effects on the linewidths of the resonance spectra due to changes in local symmetry should be considered. Vineyard[21] has calculated that each α decay in plutonium gives rise to about 2000 displaced atoms. Using the same value for Np, scaled by the total disintegration energy yields the fractional displacement of 1.6×10^{-6} of the Np atoms per day. Thus even in the longest runs of this investigation (~50 h), no effects should be seen. Furthermore, even in experiments with pure Am metal (about 20 times more active per sample atom than the Am-Th source used here) there would be a total displacement of 1% of the atoms only after 27 h, and, of course, the effects would be diminished by any reordering of the atoms through the range of the thermal spike.

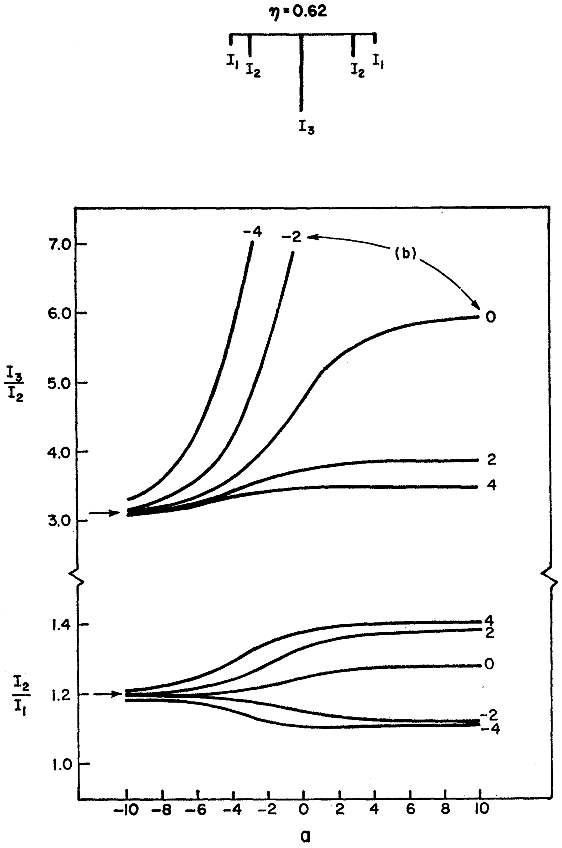

FIG. 4. Relative intensities in the quadrupole spectrum of ^{237}Np as a function of the anisotropy parameters a and b, with $\eta=0.62$.

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We would like to thank N. Griffin for his assistance in the handling of the samples. L. Ouwerkerk's help in the laboratory is much appreciated. We have enjoyed many useful discussions with A. J. Freeman.

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[20] E. King, J. A. Lee, K. Mendelssohn, and D. A. Wigley, Proc. Roy. Soc. (London) **A284**, 325 (1965).

[21] Quoted in R. O. Elliott, C. E. Olsen, and G. H. Vineyard, Acta Met. **11**, 1129 (1963).