PHYSICAL REVIEW B VOLUME 21, NUMBER 3 1 FEBRUARY 1980

Nuclear interactions in metallic Van Vleck paramagnets: NMR in single-crystal $PrNi_5$

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(Received 23 July 1979)

NMR measurements on single-crystal $PrNi_5$ in the range $1.2 \leq T \leq 8$ K are presented. The magnetic-field enhancement factors ($\alpha_{\parallel}=6.56$ and $a_{\perp}=13.13$ at 1.2 K) and the quadrupole parameter ($P \leq 0.6$ MHz) are determined microscopically from the angular dependence of the resonance field and the linewidth, respectively. The susceptibility of the Pr ions alone is shown to be smaller than the total value of χ [χ (ions) $\simeq 0.8\chi$ (bulk)]. The enhanced value of χ (ions), as well as the temperature dependence of the linewidth, are interpreted in terms of Pr-Pr exchange-induced mechanisms with $\lambda \simeq 3.5$ mole/emu. Nuclear ordering temperatures are estimated, with $T_c(P) \lesssim 0.05$ mK and $T_c(\lambda) \lesssim 0.4$ mK.

I. INTRODUCTION

Since the pioneering nuclear-magnetic-resonance study of Jones[1] upon praesodymium and thulium intermetallic compounds, it has been known that the effective magnetic field at the nuclei in these Van Vleck (VV) paramagnets may be several times the applied magnetic field. Following an earlier suggestion by Altshuler,[2] the potential use of these materials to achieve temperatures in the submillikelvin range has been extensively investigated.[3,4] It has been shown that the fundamental properties that determine a cooling "figure of merit" in VV systems include (i) interaction of the rare-earth nuclei with the magnetizing field, (ii) static nuclear–nuclear interactions, and (iii) static and dynamic nuclear-electron interactions. The most direct technique to study (i)–(iii) is of course NMR.

Currently the best cooling material among these compounds is $PrNi_5$, which, however, has hexagonal symmetry, whereas all previous NMR studies have been restricted to cubic intermetallic compounds which can be readily studied in powder form. In this paper we present the results of a single-crystal study of $PrNi_5$ which enables, for the first time in any hexagonal metallic VV system, the direct determination of the relevant interactions between the nuclei and the electrons as a function of crystal orientation. The study sets an upper limit on the zero-field quadrupolarlike nuclear-electron interaction and provides accurate values of the magnetic-field-enhancement factor as a function of both crystal orientation and temperature, up to 8 K. In addition, the temperature dependence of the relaxation time has been determined in one orientation, from which the effective ionic Pr-Pr exchange can be derived.

II. EXPERIMENT

The experiment was performed with a cw spectrometer upon oriented single-crystal specimens of $PrNi_5$. The crystal samples were prepared in the metallurgy laboratory of the Racah Institute of Physics at the Hebrew University of Jerusalem. A stoi-

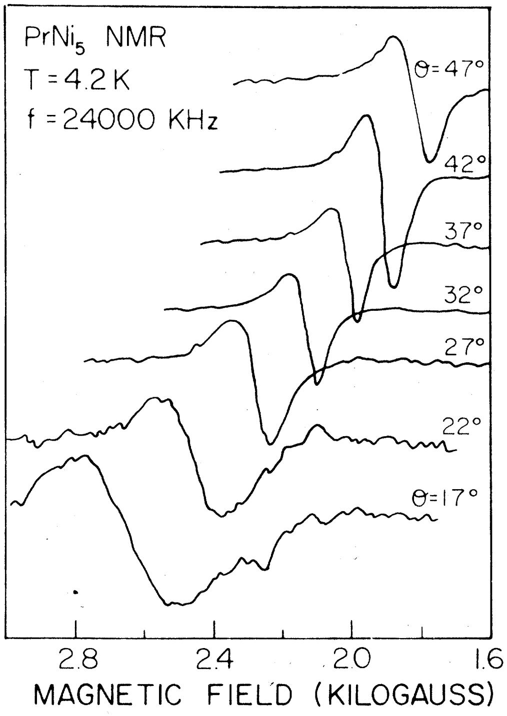

FIG. 1. Typical experimental data for several orientations of the applied magnetic field.

chiometric mixture of 99.9%-pure Pr and 99.999%-pure Ni (Johnson Matthew grade 1) was melted and cast into rods in an arc furnace under a dry argon atmosphere. Single crystals were subsequently grown by zone melting the polycrystaline rods in an electron-beam furnace: Electroetching with a current density of 2 A/cm^2 in a mixture of $H_2O:H_3PO_4$: glycerin (20:60:20 volume ratio) was needed to remove damaged "skin" prior to the final x-ray orientation procedure.

The spectra, of which examples are shown in Fig. 1, were obtained by sweeping the external magnetic field while operating the spectrometer at constant frequency and applying field modulation and phase-sensitive detection. Spectra were taken with the magnetic field applied in various orientations in the *a-c* plane of the hexagonal crystal structure. The data for two separate resonance frequencies, $f = 37\,460$ KHz and $f = 24\,000$ KHz are shown in Fig. 2, where the resonance field H_0 is plotted as a function of the angle θ between $\vec{H}$ and the c axis. In determining H_0, the appropriate correction was applied to account for the mixture of absorption and dispersion which is an inherent feature of NMR spectroscopy in bulk metallic specimens.[5] The mixture of the modes was determined assuming a Lorentzian line shape for all orientations. The angular dependence of the linewidth, in terms of the applied field, is shown in Fig. 3 for various frequencies and the error bars in Fig. 2 represent 20% of the linewidth. Except for those data points close to the c axis, these errors are very small.

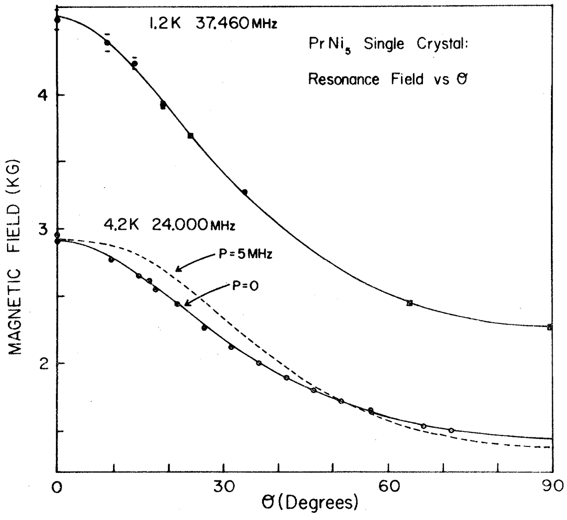

FIG. 2. Values of the resonant field as function of angle for two different experimental runs. The solid curves represent theoretical fit to the results. See text for dashed curve.

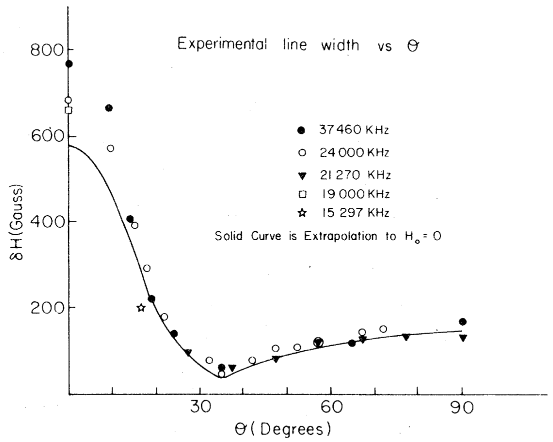

FIG. 3. "As observed" angular dependence of the linewidth for various frequencies. The solid curve is the extrapolated width for $H_0 = 0$.

III. THEORY

The magnetic-resonance spectra of the nuclei of rare-earth ions in crystals can be described by a spin Hamiltonian which for the case of axial symmetry takes the form[6,7]

$$\mathcal{H} = -(\gamma h/2\pi)[\alpha_{\parallel} H_z I_z + \alpha_{\perp}(H_x I_x + H_y I_y)] + P[I_z^2 - (1/3)I(I+1)] \ . \tag{1}$$

The parameters $\alpha_{\parallel}$ and $\alpha_{\perp}$ are the enhancement factors which determine the effective field felt by the nucleus. Thus, for example, consider an applied field H_0 which makes an angle θ with the z axis

$$\vec{H}_0 = H_z \vec{k} + H_x \vec{i} = H_0 \cos\theta \vec{k} + H_0 \sin\theta \vec{i} \ .$$

Such a field results in an effective field given by

$$\vec{H}_{\text{eff}} = \alpha_{\parallel} H_z \vec{k} + \alpha_{\perp} H_x \vec{i} = (\alpha_{\parallel} \cos\theta \vec{k} + \alpha_{\perp} \sin\theta \vec{i}) H_0 \tag{2}$$

and the angle between H_{eff} and the c axis is given by

$$\theta_{\text{eff}} = \tan^{-1}[(\alpha_{\perp}/\alpha_{\parallel})\tan\theta] \ . \tag{3}$$

From the above, we find

$$\alpha^2(\theta) = H_{\text{eff}}^2 / H_0^2 = \alpha_{\perp}^2 + (\alpha_{\parallel}^2 - \alpha_{\perp}^2)\cos^2\theta \ . \tag{4}$$

The parameters α_i are sometimes written $\alpha_i = (1 + K_i)$, where K_i is the Knight shift. It is readily shown that for nuclei in the ground state,[6]

$$K_i = (4\pi g_J \mu_B \Lambda_i)/\gamma h \ ,$$

where

$$\Lambda_i = a_J \sum_n [\,|\langle 0|J_i|n\rangle|^2/(E_n - E_0)] \ . \tag{5}$$

Here g_J and α_J are the Lande g factor and the magnetic hyperfine constant for the free ion and E_n and $|n\rangle$ are the eigenvalue and eigenfunction of the nth crystal-field eigenstate of the ion. Equation (5) is accurate only for $T \rightarrow 0$, and in principle K could show a temperature dependence as a consequence of any temperature dependence of E_n. However, the dominant temperature dependence of K, which is not included in Eq. (5), arises from the establishment of a population density in the excited states $n \neq 0$. If the relaxation between crystal-field states is fast compared with the nuclear Larmor period, then these excited states will yield an additional term,

$$K_i^{\mathrm{exc}} = 2\pi \chi_i^{\mathrm{exc}} a_J / g_J \mu_{\mathrm{B}} \gamma h \quad , \tag{6}$$

where χ_i^{exc} is the "normal" Curie susceptibility of the excited crystal-field states, and $\chi_i^{\mathrm{tot}} = \chi_i^{\mathrm{VV}} + \chi_i^{\mathrm{exc}}$.

The parameter P describes those terms giving rise to a zero-field splitting and contains not only the terms arising from the quadrupolar interaction between the nuclear quadrupole moment and its surroundings but also a pseudoquadrupole contribution[8] which arises from the hyperfine interaction. This contribution P_2 is given by the expression

$$P_2 = a_J(\Lambda_x - \Lambda_z) \tag{7}$$

and is thus directly related to the enhancement factors [Eq. (5)] by

$$P_2 = (\gamma h a_J / 4\pi g_J \mu_{\mathrm{B}})(K_\perp - K_\parallel) \quad . \tag{8}$$

For completeness we should also remark that the magnetic susceptibility per ion, χ_{ion} is related to K by

$$\chi_{\mathrm{ion}} = K g_J \mu_{\mathrm{B}} \gamma h / (2\pi a_J) \quad , \tag{9}$$

so that

$$P_2 = (a_J^2 / 2 g_J^2 \mu_{\mathrm{B}}^2)(\chi_\perp - \chi_\parallel) \tag{10}$$

provided the susceptibility is solely due to the ions.

IV. RESULTS

A. Low temperature ($1.2 \leq T \leq 4.2$ K)

In the temperature range currently investigated, and for any conceivable value of P, the intensity of the five $(m, m+1)$ trasitions—between the $2I+1$ states given by Eq. (1)—is symmetrically distributed around the $(-\frac{1}{2}, \frac{1}{2})$ central transition.[9] Thus we assume a model in which the center of each resonance absorption profile, as determined by the experimental points of Fig. 2, corresponds to the $(-\frac{1}{2}, \frac{1}{2})$ transition. The solid curves in Fig. 2 represent a best fit of the $(-\frac{1}{2}, \frac{1}{2})$ transition of Eqs. (1)–(4) with the experimental data using $(\gamma/2\pi) = 1.25$ KHz/gauss. The results for $\alpha_\parallel$, $\alpha_\perp$, and P are summarized in Table I. Note the constant value of $\alpha_\parallel$ and the small ($\sim 0.6\%$) increase of $\alpha_\perp$ as T increases from 1.2 K to 4.2 K. Only a range could be determined for P. To illustrate the effects of P on the calculation, a constrained "best fit" with $P/h = 5000$ KHz was also calculated and is shown as dashed curve in Fig. 2 (only for the 4.2 K data).

TABLE I. Enhancement factors and quadrupolelike parameters.

T (K)	f (KHz)	$\alpha_\perp$	$\alpha_\parallel$	P/h (KHz)
1.2	37 460	13.133 ±0.01	6.557 ±0.01	0–600
4.2	24 000	13.209 ±0.01	6.570 ±0.01	0–300
8		15.3 ±0.2	6.65 ±0.2	
1.2–4.2				600[a] ±200

[a] From angular dependence of linewidth.

A much better estimate of P, in agreement with the data of Table I, can be obtained from the angular dependence of the linewidth. The solid curve in Fig. 3 is a crude extrapolation of the data to $f = H_0 = 0$. At first glance, the angular dependence displayed by this curve appeared rather strange. "Experimental" points from this curve are plotted again in Fig. 4, except that now we have used Eqs. (3) and (4) to calculate for each point the *real* internal linewidth and the direction of the *effective* field, θ_{eff}. The solid curve in Fig. 4 is a plot of the absolute value of the difference between the $(-\frac{5}{2}, -\frac{3}{2})$ and the $(\frac{3}{2}, \frac{5}{2})$ transitions, calculated from the Hamiltonian [Eq. (1)] for $P/h = 595$ KHz. The similarity between the calculation and the points is indeed to be expected if we ascribe the observed line profile to an unresolved five-line quadrupolarlike multiplet. We conclude that

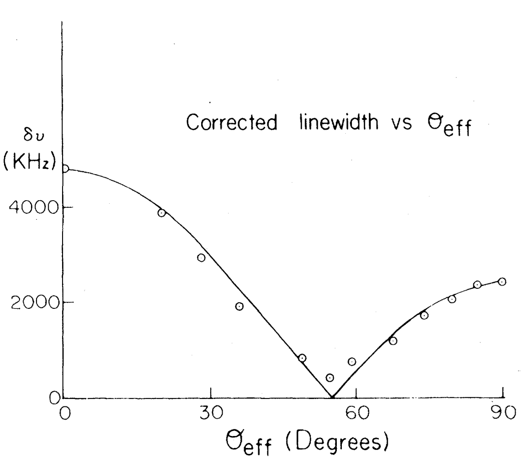

FIG. 4. Real linewidth as function of orientation of the effective field. Solid curve represents theory (see text).

in $PrNi_5$, the zero-field splitting of the nuclear levels is less than 1 MHz, and the most probable value is $P/h \simeq 0.6$ MHz. The lack of resolved features, even for $\theta_{\rm eff}=0$ where the splitting is largest, may reflect one or more of the following: (i) poor signal-to-noise ratio that was realized along the c axis, (ii) inhomogeneous P across the surface, and (iii) amplification of the effects of twinning and misorientation of grains, since around the c axis we have $\theta_{\rm eff} \simeq 2\theta$.

B. Temperature effects

The temperature dependence of the resonant field has been measured up to 8 K at the angle $\theta=37°$ at which the line is almost narrowest. It was also possible to observe the resonance at two further orientations, $\theta=90°$ and $\theta=27°$, at 8 K. Figure 5 shows the variation of α^2 as a function of $\cos^2\theta$ for these three orientations. As predicted by Eq. (4), this variation is linear and, within the limit of error, it will be seen that the extrapolation to $\theta=0$, $\alpha_\parallel(T=8\text{ K}) = 6.65 \pm 0.2$, is in good agreement with the 4.2 K value for $\alpha_\parallel$. Accordingly we have assumed that $\alpha_\parallel$ is independent of temperature and used Eq. (4) to calculate from the measured values of $\alpha(37°)$ the values of $\alpha_\perp$. Figure 6 shows both the temperature dependence of $\alpha(37°)$ and $\alpha_\perp$ up to 8 K.

Finally in Fig. 7 we show the temperature variation of the linewidth at $\theta=37°$. A very rapid increase is observed above 4.2 K and indeed prevented our observation of the signal above 8 K. In the other two orientations observed at 8 K, the linewidth was the same as that at $\theta=37°$ within experimental error.

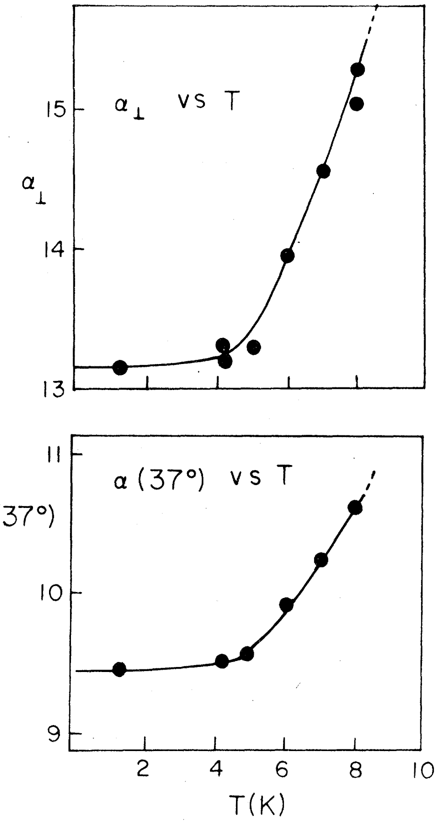

FIG. 6. Temperature dependence of $\alpha(37°)$ and $\alpha_\perp$.

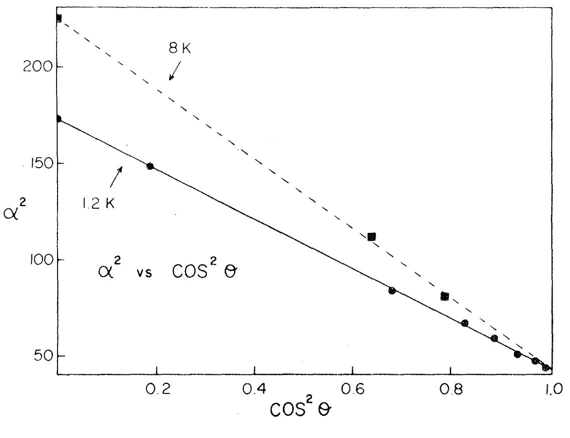

FIG. 5. Angular dependence of the enhancement factor for 1.2 and 8 K.

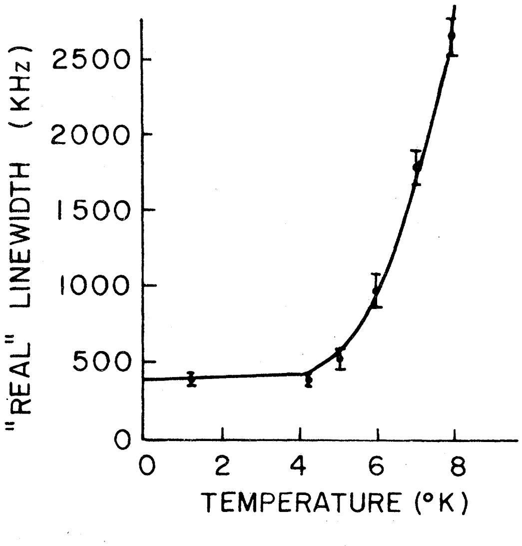

FIG. 7. Temperature dependence of the real linewidth.

V. DISCUSSION

A. Magnetic properties

Craig *et al.*[10] first studied the magnetic susceptibility and the electrical resistivity of polycrystalline $PrNi_5$. They also measured the difference between the heat capacities of $PrNi_5$ and isomorphic $LaNi_5$ and attributed this to the Pr ions. A model for the crystal-field-induced energy levels of the Pr ions was proposed to fit the data. However, when the principal magnetic susceptibilities were measured in single crystals by Andres, Schmidt, and Darack,[11] the anisotropy observed was opposite in sign to that predicted. Subsequent studies of the specific heat and thermal expansion were made by Ott *et al.*,[12] and Andres and Darack[4] have attempted to fit the data for both the specific heat and the susceptibility. Their experimental and theoretical results are shown in Fig. 8, and it will be noted that they found it necessary to include a ferromagnetic-exchange-enhancement factor to obtain a reasonable fit. We may use their susceptibility values to deduce $\alpha_{\parallel}$ and $\alpha_{\perp}$ according to the equation[4]

$$\alpha=(1+h_{4f}\chi) \ , \tag{11}$$

where $h_{4f}=187.7$ mol/emu. The values obtained are $\alpha_{\parallel}=8.1$ and $\alpha_{\perp}=16.4$, both clearly larger than our experimental results. Alternatively, if we use our values to determine the Pr ion contribution to the susceptibility, we would obtain the points χ(ions) shown in Fig. 8. Thus in *both* orientations the contribution from the Pr ions to the susceptibility is 80% of the total. We may also obtain χ directly from Eq. (9). Substituting $a_J/h=1093$ MHz and $g_J=0.8054$, we find even smaller χ values (by about 4%), but this could be the result of our using $\gamma/2\pi=1.25$ KHz/G whereas in some references a value of 1.3 KHz/G was used.

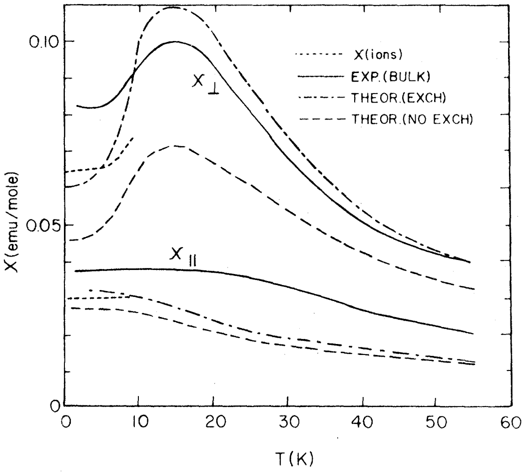

FIG. 8. χ (ions) of the present study, superimposed on the bulk susceptibility data and theory of Ref. 4.

The above result is crucial to an understanding of both the susceptibility and specific-heat data since it appears to imply that a significant contribution to the susceptibility arises from the nickel. As mentioned by Ott *et al.*, the possibility that the nickel $3d$ bands were not completely filled could account for the unusually strong dependence of the electronic specific-heat parameter between 1 and 9 K.

Over the very limited temperature range we have studied, the nickel contribution remains a constant proportion of the total susceptibility. Also it is notable that the proportion is independent of orientation. We may then infer that the Ni polarization is exchange induced by the Pr ions, and is thus proportional to the Pr polarizatioin. This also suggests a strong indirect exchange interaction between Pr ions, similar perhaps to a superexchange mechanism. Given the crystal structure of $PrNi_5$, which consists of alternate planes of atoms in which one layer is Ni alone while the other is Pr + Ni, it is tempting to speculate that only one type of Ni atom has an induced moment— perhaps the one in the same plane. The Pr-Pr exchange will be discussed further at the end of this section.

B. Quadrupolar interaction

The quadrupolarlike interaction may be written[7]

$$P=P_1+P_2+P_3 \ ,$$

where P_1 is the contribution to the electric field gradient from the $4f$ electrons given by the expression.

$$P_1=-[3e^2Q\langle r^{-3}\rangle/4I(2I-1)] \times\langle J||\alpha||J\rangle\langle 3J_z^2-J(J+1)\rangle \ . \tag{12}$$

If we assume the ground state to be $(1/\sqrt{2})\times(|+3\rangle-|-3\rangle)$ as predicted by Andres and Darack then $P_1/h=-0.656$ MHz. The pseudoquadrupole interaction, P_2, we can evaluate from our results. Using Eq. (8), we obtain $P_2/h=3.985$ MHz. Accordingly $(P_1+P_2)/h=3.33$ MHz, whereas our upper limit on $|P/h|$ is ~ 0.6 MHz, from which we may infer that P_3, the remaining contribution, must be either -4 or -2.7 MHz, depending on the sign of P/h. This contribution is the usual contribution to the electric field gradient observed in normal metals and is the resultant of the ionic contribution from the lattice and that due to the conduction electrons.

It thus appears that a near cancellation occurs between the electric-field-gradient contribution, P_1+P_3, and the pseudoquadrupolar contribution P_2 in this system. From the point of view of nuclear cooling, this is indeed fortunate, since the nuclear-quadrupole ordering temperature is related to the zero-field splitting between the nuclear m levels,

$$E_m=P[m^2-(\tfrac{1}{3})I(I+1)] \ , \tag{13}$$

leading to

$$T_c(\text{quadrupole}) \lesssim 2P/k_B = 0.058 \text{ mK} .$$

This temperature is 10 times lower then the lowest value attained so far, and we may conclude that quadrupolelike splitting does not currently impose a limitation in cooling experiments with $PrNi_5$.

C. Exchange and magnetic nuclear order

Some insight concerning the Pr-Pr exchange interaction, and the related problem of magnetic nuclear order, may be gained by examining two independent experimental observations, namely, (i) the temperature dependence of the linewidth and (ii) the difference between the "crystal-field-only" χ_c and our experimentally determined ionic susceptibility.

Starting with χ, we note that using a molecular-field approach one may estimate the nuclear-ordering temperature by[13]

$$T_c = \lambda C_N (1+K_c)^2/(1-\lambda\chi_c) \quad , \tag{14}$$

where C_N is the paramagnetic Curie constant of the bare nuclear moments, $(1+K_c)$ is the crystal-field-*only* enhancement factor, χ_c is the unenhanced susceptibility (in our case $\chi_\perp$), and λ is the molecular-field-exchange-enhancement factor given by

$$\chi = \chi_c/(1-\lambda\chi_c) \quad . \tag{15}$$

Andres and Darack[4] obtained an estimate of $T_c = 0.57$ mK from their interpretation of the data. However we have already seen that their bulk χ value is an overestimate for the Pr contribution to χ, leading to an overestimate of λ. Unfortunately, without a reasonable estimate of λ, it is difficult to proceed further; but by way of illustration, if we were to assume the correctness of the crystal-field calculation of χ_c and estimate a value of λ of 3 mol/emu to fit our limited range of χ(ions) data (Fig. 8), we would obtain a somewhat lower ordering temperature of $T_c = 0.32$ mK.

We turn now to the linewidth data (Fig. 7). A detailed theoretical treatment and analysis are given elsewhere,[14] and we shall only outline here the approach used in the calculation and then discuss the final results. Since no temperature dependence was notable between 1.2 and 4.2 K, we assumed that the linewidth δf may be written

$$\delta f = \Delta_R + (1/T_2)(T) \quad , \tag{16}$$

where Δ_R is a residual static inhomogeneity and $1/T_2$ is a dynamic contribution caused by thermally excited fluctuations of the Pr ions, which in turn lead to nuclear spin-lattice relaxation of the Pr^{141} nuclei through the hyperfine interaction a_J^{141}. Comparing Fig. 7 with data obtained for the isomorphic nonmagnetic system $LaNi_5$, we may conclude that in the present temperature range, ion-phonon scattering can be neglected in treating the Pr fluctuations. Likewise, the contribution of the direct Korringa process is negligible in comparison with Δ_R.

Another process, an effective ion-ion exchange leading to fluctuations of the magnetic ions, has been previously suggested for cubic systems,[15] and a refined model was later suggested to treat also excited crystal-field levels.[16] Adapting the model to the present case, the dynamic term $1/T_2$ is written

$$1/T_2 = [(8\pi^3/h^2 z)a_J^2/(2\pi M_2)^{1/2}] \sum_\alpha C_{\alpha\alpha} e^{-E_\alpha/T} \quad , \tag{17}$$

where the summation is over all of the Pr levels in the crystal, and M_2 is associated with the familiar correlation functions describing the fluctuation spectra. M_2 depends explicitly on E_α and $C_{\alpha\beta}$, the later being matrix elements of the ionic momentum defined as $C_{\alpha\beta} = \langle\alpha|J_+|\beta\rangle$ for $\alpha \neq \beta$ and $C_{\alpha\alpha} = \langle\alpha|J_z|\alpha\rangle$. Last and most important, M_2 depends on the Pr-Pr exchange interaction. For an isotropic exchange j between ions k and i, given by

$$\mathcal{H}^{\text{exch}} = -\tfrac{1}{2} j_{ik} \vec{J}_i \cdot \vec{J}_k \tag{18}$$

it was found that the explicit exchange-dependence of M_2 is through the term

$$\left(\sum j^2\right)^{1/2} \equiv \left(\sum_k j_{ik}\right)^{1/2} \quad . \tag{19}$$

It follows from the above outline that, for a given ionic configuration, crystal field, and hyperfine interaction, the relaxation term $1/T_2$ will depend only on $(\sum j^2)^{1/2}$. Using tabulated a_J value[8] and the level scheme of Andres and Darck,[4] a best fit of Eq. (17) to the data was attempted by adjusting a single parameter, $(\sum j^2)^{1/2}$. The remarkable fit shown by the solid curve in Fig. 7 was obtained for $(\sum j^2)^{1/2} = 0.02 \times 10^{-3}$ eV.

In order to estimate T_c from the relaxation data, we first note that the molecular-field constant λ of Eq. (15) is related to the exchange by

$$\lambda = \left(\sum_k j_{ik}\right) \Big/ \left(g_J^2 \mu_B^2 N_0\right) \quad . \tag{20}$$

To determine the required algebraic sum $\sum j$ from our experimental $\sum j^2$ value, prior knowledge of the range and sign variation of the interaction between the various Pr pairs is needed. However, an upper limit on $\sum j$ is readily derived from $\sum j^2$ for any *assumed* range r_0. The limit is given by

$$\sum j \leq \sqrt{n} \left(\sum j^2\right)^{1/2} \quad , \tag{21}$$

where n is the number of Pr atoms within radius r_0. For example, for $r_0 \simeq 5$ Å only first- and secondnearest shells with $n = 8$ are included. Substituting in Eqs. (14) and (20) we then find

$$\sum j \lesssim 0.055 \times 10^{-3}\ \mathrm{eV}\ ,$$

$$\lambda = 2.62\ \mathrm{mol/emu}\ ,\quad T_c \lesssim 0.25\ \mathrm{mK}\ .$$

A more realistic range of $r_0 = 7.5$ Å will include also the third-nearest shell, thus containing *all* of the sites with direct superexchange links to the central atom. We then have $n = 20$,

$$\sum j \lesssim 0.09 \times 10^{-3}\ \mathrm{eV},$$

$$\lambda = 4.3\ \mathrm{mol/emu}\ ,\quad T_c \lesssim 0.4\ \mathrm{mK}\ .$$

Clearly, the larger the assumed range of the effective interaction, the higher the upper limit of T_c. It is gratifying, however, to note that the exchange values derived from $1/T_2$ are very similar to the estimates based on static χ measurements[4] or on the enhancement factors K mentioned above.

VI. CONCLUDING REMARKS

A variety of microscopic properties of $PrNi_5$ [e.g., $1/T_2$, K, χ(ions), P and λ] have been determined directly for the first time in the present study. Some of these properties appear to be adequately explained. Others, such as the detailed character of the exchange, the contribution of the Ni bands to χ, and the relatively large negative value of P_3 (lattice + conduction-band contribution to the electric field gradient), require further theoretical treatment. We have shown that nuclear quadrupole order due to P is expected to occur only around 0.05 mK, but our estimates for λ imply a magnetic-ordering temperature T_c which is only slightly lower then the lowest temperature (0.55 mK) achieved already.[17] Our results prove that exchange-induced Pr fluctuations become ineffective as a relaxation source below ~2 K. Since the relaxation in the mK range is extremely important from the point of view of cooling experiment, we believe that the nuclear spin-lattice measurements should be extended to that range. A pulsed spectrometer would be required for such study to overcome masking by the residual linewidth Δ_R that was observed in the present specimen.

ACKNOWLEDGMENTS

We are grateful to K. Andres for his unpublished crystal-field data, to N. Mayr for his assistance with the experiment, to A. Gabay for help in developing the electroetching procedure, and to G. Kaplan for assistance in best fitting some of the results. The research was supported in part by the U.S.A.-Israel Binational Science Foundation. Work at University of British Columbia (U.B.C.) was supported by the National Research Council of Canada Grant No. 67-1873. One of us (N.K.) wishes to thank the members of the Physics Department, U.B.C., for the hospitality accorded him during a sabbatical leave.

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[*] On sabbatical leave from the Hebrew University, Jerusalem, Israel.

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