$B+E$ vibrational modes of the S_4 symmetry, respectively. The motion of the constituent atoms must, of course, comply with the space group symmetry of the crystal and the two B species of S_4 symmetry, according to Table I, are correlated into the B_1+B_2 zone-center phonons. All these modes behave essentially as "individual" molecular vibrations and may be considered as examples of Einstein phonons.

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[1]E. M. Brody and H. Z. Cummins, in *Light Scattering Spectra of Solids*, edited by G. B. Wright (Springer, New York, 1969), p. 683; E. Litov and E. A. Uehling, Phys. Rev. B 1, 3713 (1970).

[2]I. P. Kaminow, in *Light Scattering Spectra of Solids*, edited by G. B. Wright (Springer, New York, 1969), p. 675.

[3]E. A. Popova and A. I. Stekhanov, Fiz. Tverd. Tela 12, 51 (1970) [Sov. Phys. Solid State 12, 40 (1970)].

[4]F. Jona and G. Shirane, *Ferroelectric Crystals* (Pergamon, New York, 1962); R. Wyckoff, *Crystal Structures*, 2nd ed. (Interscience, New York, 1963), Vol. III.

[5]*International Tables for x-Ray Crystallography*, 3rd ed., edited by N. F. M. Henry and K. Lonsdale (Kynoch Press, New York, 1969), Vol. I.

[6]R. K. Khanna and C. W. Reimann, Spectra-Physics Raman Tech. Bull. No. 3, 1970 (unpublished); W. G. Fateley, N. T. McDevitt, and F. F. Bentley, Appl. Spectry. 25, 155 (1971).

[7]J. P. Mathieu, *Spectre de Vibration et Symmetrie des Molécules et des Cristaux* (Hermann, Paris, France, 1945); E. Wilson, J. Decius, and P. Cross, *Molecular Vibrations* (McGraw-Hill, New York, 1955).

[8]M. Shur, Fiz. Tverd. Tela 8, 57 (1966) [Sov. Phys. Solid State 8, 43 (1966)].

[9]Y. D. Harker, Jon D. Masso, and D. F. Edwards, J. Appl. Optics 8, 2563 (1969).

[10]T. C. Damen, S. P. S. Porto, and B. Tell, Phys. Rev. 142, 570 (1966).

[11]A. I. Stekhanov and E. A. Popova, Fiz. Tverd. Tela 7, 3530 (1965) [Sov. Phys. Solid State 7, 2849 (1966)].

[12]J. P. Chapelle, J. Chem. Phys. 46, 30 (1949); I. P. Kaminow, R. C. C. Leite, and S. P. S. Porto, J. Phys. Chem. Solids 26, 2085 (1965).

[13]See, for example, G. Herzberg, *Infrared and Raman Spectra of Polyatomic Molecules* (Van Nostrand, Princeton, N. J., 1945).

[14]J. D. Masso, C. Y. She, and D. F. Edwards, Phys. Rev. B 1, 4179 (1970); C. Y. She, J. D. Masso, and D. F. Edwards, J. Phys. Chem. Solids (to be published).

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Average Magnetic Hyperfine Fields at ^{106}Pd Nuclei in Ni-Pd Alloys

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The average magnetic hyperfine fields at ^{106}Pd nuclei in a series of Ni-Pd alloys have been measured at 77 °K by means of the integral perturbed-angular-correlation method using the (622.0–511.8)-keV γ-ray cascade in ^{106}Pd. The observed magnetic fields have been corrected for the external applied field and the sample magnetization, and extrapolated to 0° K. The average hyperfine fields H_{hf} at the Pd nuclei were found to be negative in the ferromagnetic alloys over the full concentration range, varying from (-194 ± 9) kG in Ni metal to (-41 ± 10) kG in an alloy of 90% Pd. Our estimates for the contribution to the hyperfine field of Pd arising from the local moment on the Pd atom are too small to account for the net measured fields. Thus, we ascribe the origin of the remaining large negative fields to interactions with neighboring atoms through the conduction electrons.

I. INTRODUCTION

The magnetic properties of palladium metal and its alloys have been the subject of considerable study, particularly in the last few years. Among the most interesting properties is the occurrence of ferromagnetism in Pd alloyed with very small concentrations of Fe, Co, and Ni.[1,2] At these small concentrations of the 3*d* metal, susceptibility and neutron studies have indicated that giant moments are associated with each impurity atom.[1-3] These moments, up to 12 μ_B per impurity atom, are much greater than can be produced by 3*d* states on the impurity alone. Thus, it is reasonable to assume that the Pd atoms nearby one of these impurities carry a moment and are intimately associated with the long-range ferromagnetic coupling observed for alloys with these small concentrations of 3*d* metal. The part played by conduction electrons in the long-range ferromagnetic coupling between impurities is,

however, not well understood.

In recent years, the determination of magnetic hyperfine fields[4-6] has proved fruitful in many studies of the magnetic properties of metals and alloys. These measurements can give information not only about the localized moment on the atom under study but also about the magnetic behavior of the conduction electrons. Several articles have recently treated the experimental information for alloys in terms of various models for the observed hyperfine fields.[7-11]

Here we present measurements of the magnitude and sign of the average hyperfine fields at Pd nuclei in nine Ni-Pd alloys by means of the integral perturbed-angular-correlation (IPAC) technique using the (622.0–511.8)-keV γ-ray cascade in ^{106}Pd.[12] These measurements were performed in order to obtain information on the mechanisms producing the hyperfine fields in these alloys.

Previous studies of the Pd field in ferromagnetic Ni-Pd alloys have been confined to relatively dilute alloys.[13-15] In concentrated alloys one expects a distribution of hyperfine fields at the component atoms. The IPAC technique measures only the average hyperfine field experienced by the daughters of the decaying radioactive nuclei. If these nuclei are randomly dispersed in a disordered alloy, then this average field is characteristic of an average atomic structure of the alloy.

II. THEORY OF IPAC METHOD

Let us consider a nucleus in an initial state (with spin angular momentum I_i) which decays to a final state (with spin angular momentum I_f) through an intermediate state (with spin angular momentum I). The transition occurs with the emission of two γ rays

$$I_i \xrightarrow{\gamma_1} I \xrightarrow{\gamma_2} I_f \ ,$$

with γ_1 and γ_2 of multipole order L_1 and L_2, respectively. An unperturbed-angular-correlation function $W(\theta)$ expresses the relative probability that the nuclear radiation γ_2 is emitted at an angle θ with respect to the propagation direction of the first radiation γ_1. This function can be expressed[16] as a finite series of even-order Legendre polynomials, or as a series in $\cos 2k\theta$:

$$W(\theta) = 1 + \sum_{k=1}^{k_{\max}} A_{2k} P_{2k}(\cos\theta) = \sum_{k=0}^{k_{\max}} B_{2k} \cos(2k\theta) \ , \quad (1)$$

where $k_{\max}$ is the smallest integer in the set I, L_1, and L_2. The coefficients A_{2k} and B_{2k} may be determined from a knowledge of the spins and multipole order of the transitions.[16] The application of a magnetic field H to the nucleus causes a precession of the nuclear spin of the intermediate state I at the Larmor frequency $\omega = g\mu_N H/\hbar$, where g is the nuclear g factor of the intermediate state, μ_N is the nuclear magneton, and $\hbar$ is Planck's constant divided by 2π. In this work, the magnetic field at the Pd nuclei, in an alloy of Pd concentration c, is the sum of a hyperfine field $H_{\rm hf}^{\rm Pd}(c)$, an externally applied magnetizing field $H_{\rm ext}$, a Lorentz field $\frac{4}{3}\pi M$ (where M is the alloy magnetization), and a demagnetizing field DM (where D is the demagnetizing factor for the sample shape used). If the magnetic field H is oriented perpendicular to the angular-correlation plane of the radiations γ_1 and γ_2, then the correlation pattern $W(\theta)$ will rotate through a mean precession angle $\omega\tau$. Here τ is the mean life of the intermediate state. The correlation function will then take the form[16]

$$W(\theta, H) = \sum_{k=0}^{k_{\max}} \frac{B_{2k}}{1 + (2k\omega\tau)^2} \times [\cos(2k\theta) - 2k\omega\tau \sin(2k\theta)] \quad (2)$$

or

$$W(\theta, H) = \sum_{k=0}^{k_{\max}} \frac{B_{2k}}{[1 + (2k\omega\tau)^2]^{1/2}} \cos(2k\theta + \Delta\theta) \ ,$$

where $\Delta\theta = \tan^{-1} 2k\omega\tau$. The factor $1/[1 + (2k\omega\tau)^2]^{1/2}$ accounts for an attenuation of $W(\theta)$, and the factor $\Delta\theta$ accounts for the rotation of the correlation. A direct measure of the mean precession angle $\omega\tau$ may be obtained by forming the ratio R defined as

$$R(\theta) = 2\,\frac{W(\theta, H) - W(\theta, -H)}{W(\theta, H) + W(\theta, -H)} \ , \quad (3)$$

where $W(\theta, H)$ is the correlation function with the magnetic field in the $+z$ direction and $W(\theta, -H)$ is the corresponding function with the field in the $-z$ direction.

Since changing the direction of the magnetic field will reverse the precession direction, then on substituting Eq. (2) into Eq. (3) the ratio R becomes

$$R(\theta) = -2 \left(\sum_k \frac{B_{2k}}{1 + (2k\omega\tau)^2} 2k\omega\tau \sin(2k\theta) \Big/ \sum_k \frac{B_{2k}}{1 + (2k\omega\tau)^2} \cos(2k\theta) \right) . \quad (4)$$

For cases where $\omega\tau \ll 1$, such as for Pd in Ni studied here ($\omega\tau < 0.01$),

$$R(\theta) = \left(\frac{2}{W(\theta, 0)} \frac{dW(\theta, 0)}{d\theta} \right) \omega\tau \ . \quad (5)$$

The factor $(2/W)(dW/d\theta)$ may be determined by a separate measurement of the angular-correlation pattern [Eq. (1)].

For each of nine alloys, we have measured the shift R, as given by Eq. (3), for the (622.0–511.8)-keV γ-ray cascade in ^{106}Pd at two angles, 150° and 160°. The difference in the count rate at these two angles was used to monitor the magnitude of the angular anisotropy given by Eq. (1).

III. EXPERIMENTAL DETAILS

The magnet used in the experiment was an iron-core electromagnet completely shielded by a thin shell of Armco iron in the flux-return path and energized by a regulated power supply. The source was constructed in the form of a cylindrical shell and was inserted in a thin-walled plastic tube of nominally 4 mm diameter and 6 mm length. This tube was glued to the tip of one of the magnet poles. The source-magnet system was then immersed in a liquid-nitrogen bath, and to ensure cylindrical symmetry the entire system was allowed to rotate slowly (approximately one revolution every two minutes).

The perturbed-angular-correlation spectrometer employed three scintillation-detection channels. Each channel consisted of a 3×3-in. NaI (Tl) crystal affixed to a photomultiplier, a preamplifier, and a double-delay-line linear amplifier used with 0.4-μsec delay lines to minimize pile-up effects. The distance between each detector and the center of the source was 9 cm. One detector was in a fixed position and the other two were movable. The output pulses from each linear amplifier were fed (with suitable delays) to two separate single-channel analyzers, one set on the 511.8-keV γ ray and the second on the 622.0-keV γ ray. Each output of the two single-channel analyzers that selected the 511.8- and 622.0-keV photopeaks from the fixed detector went to two different coincidence units. Each of these four coincidence units monitored the coincidence events between one of the cascade γ rays detected by the fixed detector and the other cascade γ ray detected by a movable detector. A total of ten scalars were used to count the single and coincidence pulses simultaneously.

The alloys used in this experiment were obtained from Obenshain of the Oak Ridge National Laboratory, and most of them were cut from the samples used for Mössbauer studies of the hyperfine field at Ni.[17] With the exception of the 20% Pd alloy, which may have contained up to 50 ppm of Fe, the Fe concentration in the alloys arising from the starting high-purity materials was less than 10 ppm.

A solution containing ^{106}Ru activity in the form of ruthenium chloride was dried on the clean surface of foils of nominal thickness of 0.25 mm. The activity was diffused into the alloys in an evacuated Mullite tube for 60 h at 1200 °C. Then the samples were cooled rapidly to room temperature to avoid the relatively weak short-range ordering observed when alloys in the Ni-Pd system are annealed below 500 °C.[18] Although the foils appeared bright and clean upon removal from the furnace, the surfaces were polished by abrasive paper to ensure removal of any undiffused radioactivity. The process was repeated until it was certain that any loss of radioactivity was due to removal of a layer of the bulk radioactive Ni-Pd alloy. After this process, the samples were cleaned with alcohol and water and left to dry.

The magnet produced a field of 10 kG to orient the alloy magnetization which in turn orients the internal hyperfine field. This polarizing field H_{ext} was reversed every hour and the coincidence counts were collected for both field directions. The process was repeated after the two movable detectors were shifted to the other angular positions. The collected coincidence counts were corrected for the chance coincidence counts and the corrected coincidence rates at each angle were normalized by the product of the single counting rates. The resolving time $2T$ for each coincidence unit was chosen such that the ratio of chance to true coincidences was below 10%. For our source strengths, $2T$ was normally between 40 and 65 nsec. The ratio R was calculated using the relation $R = 2 \times (C^N_{up} - C^N_{down})/(C^N_{up} + C^N_{down})$, where C^N_{up} and C^N_{down} are the normalized true coincidence counts for the field-up and field-down directions. In order to measure the true rotation of the angular-correlation pattern, the windows of the single-channel analyzers were set at the full width at half-maximum for the photopeaks which they selected. By doing so, the contribution of one cascade γ ray in the window set for the other cascade γ ray was minimized.

IV. RESULTS

In order to measure the coefficients A_{2k} of Eq. (1) for the cascade $0^+ \rightarrow 2^+ \rightarrow 0^+$ in ^{106}Pd, the full angular correlation was taken with an unmagnetized source (i.e., with $H_{ext} = 0$). The measured coefficients were corrected for finite solid-angle effects. The corrected coefficients A_2 and A_4 were 0.351 ± 0.019 and 1.138 ± 0.027, respectively. These coefficients are in good agreement with the theory for a $0^+ \rightarrow 2^+ \rightarrow 0^+$ cascade[16] and with previously reported values.[19–21] The contribution to $W(\theta)$ from the weak cascade

$$2^+ \xrightarrow{\text{616-keV } \gamma \text{ ray}} 2^+ \xrightarrow{\text{511.8-keV } \gamma \text{ ray}} 0^+$$

is quite small, since our experimental coefficients agree with the theoretical values for a $0^+ \rightarrow 2^+ \rightarrow 0^+$ cascade. However, since we use experimentally determined values of A_2 and A_4, which include the effect of the 616, 511.8-keV cascade, the presence of this cascade does not affect the results given below.

The rotation $\omega\tau$ of the angular-correlation pattern was measured at 77 °K for each alloy at two angles 150° and 160°. The results are given in Table I. We have chosen to use a value for $g\tau$ such that the

hyperfine field at palladium nuclei in Ni metal $H_{hf}^{Pd}(c=0,\ T=4.2\,^{\circ}K)$ calculated from the observed rotation for Pd in Ni agrees with the value obtained by NMR measurements,[13] $H_{hf}^{Pd}(c\sim 0,\ 4.2\,^{\circ}K) = -194$ kG. Using this normalization, we find $g\tau = (7.56 \pm 0.37)10^{-12}$ sec. With the value $\tau = 18.4 \pm 1.2$ psec,[22] the g factor of the first 2^+ excited state in ^{106}Pd is 0.41 ± 0.04. This value of the g factor is in good agreement with values reported earlier; $g = 0.40 \pm 0.04$,[23] 0.35 ± 0.03,[14] 0.385 ± 0.030,[21] and is comparable to $g = 0.43$, as predicted on the hydrodynamical model for collective nuclear motion.[24]

The resulting fields were corrected for the external applied field H_{ext} and the Lorentz field $\frac{4}{3}\pi M$. The demagnetizing field DM for our samples was very small and thus was neglected. These fields were extrapolated to 0 °K, assuming the temperature dependence of the alloy magnetization is analogous to that in Ni. The uncertainty in this extrapolation is thought to be small, since the measurements were made at 77 °K where the saturation magnetization is near its value for 0 °K. The dependence of this corrected value of the average hyperfine field H_{hf}^{Pd} on the alloy concentration is shown in Fig. 1. The error bars shown represent only our experimental errors and do not reflect the errors associated with the NMR result.

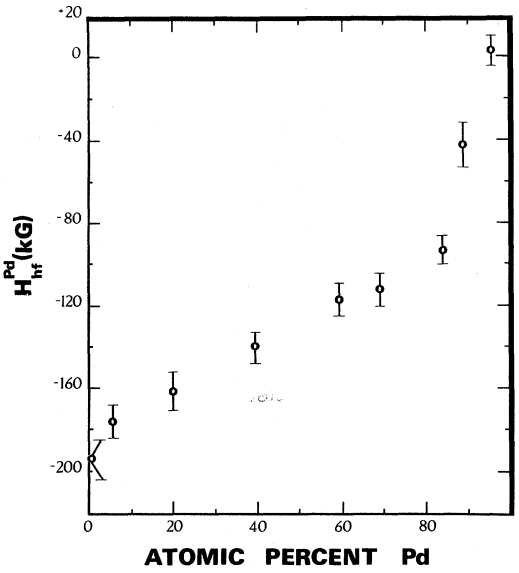

FIG. 1. Average magnetic hyperfine fields H_{hf}^{Pd} at ^{106}Pd nuclei as a function of composition for the Ni-Pd alloy system. The hyperfine field was measured by the IPAC method at 77 °K. Values shown here were corrected for the external applied field and the sample magnetization, and extrapolated to 4.2 °K. The value for $g\tau$ has been chosen such that H_{hf}^{Pd} for Pd in Ni metal was the same as that obtained in NMR measurements. The error bars shown represent only our experimental errors and do not reflect the errors associated with the NMR result or the extrapolation of the data to 4.2 °K.

V. CONCLUSION AND DISCUSSION

The average hyperfine fields at Pd nuclei in Ni-Pd alloys were found to be negative over the entire ferromagnetic range. They ranged from -194 ± 9 kG in Ni metal to -41 ± 10 kG in an alloy of 90% Pd, and $+5 \pm 6$ kG in a 97% Pd alloy which was paramagnetic at 77 °K. This composition dependence of H_{hf}^{Pd} is in contrast to the Ni hyperfine field H_{hf}^{Ni} as measured by the Mössbauer effect[17,25] which changes sign near 45% Pd and rises to a large positive value at 90% Pd.

A part of the measured hyperfine field in the alloys will arise from the localized d moments on the Pd atoms. Cable *et al.*[26] have measured the individual d moments for both constituents of Ni-Pd alloys. They find from their diffuse-neutron-scattering results that the Pd moments are quite small, reaching a maximum of $\sim 0.2\mu_B$ for a 72% Pd alloy.

We have estimated the contribution of these measured Pd moments to H_{hf}^{Pd} using the results of Seitchik *et al.*[27] These authors have studied the Knight shift and susceptibility for Pd metal as a function of temperature. They assume that the only contribution to the susceptibility with appreciable temperature dependence is from the magnetization of d-

TABLE I. Values of $\omega\tau$ (measured at 77 °K) at 150° and 160° and their average in a series of Ni-Pd alloys.

at.% Pd	Angular shift at 150° (mrad)	Angular shift at 160° (mrad)	Average angular shift (mrad)
0.0	$-(6.836 \pm 0.388)$	$-(5.941 \pm 0.574)$	$-(6.555 \pm 0.321)$
5.0	$-(5.469 \pm 0.418)$	$-(6.305 \pm 0.381)$	$-(5.926 \pm 0.282)$
20.0	$-(5.350 \pm 0.439)$	$-(5.413 \pm 0.491)$	$-(5.378 \pm 0.327)$
40.0	$-(4.526 \pm 0.354)$	$-(4.652 \pm 0.412)$	$-(4.579 \pm 0.268)$
60.0	$-(3.493 \pm 0.309)$	$-(4.112 \pm 0.379)$	$-(3.740 \pm 0.239)$
70.0	$-(3.378 \pm 0.294)$	$-(4.049 \pm 0.485)$	$-(3.558 \pm 0.251)$
85.0	$-(2.735 \pm 0.290)$	$-(2.949 \pm 0.348)$	$-(2.823 \pm 0.223)$
90.0	$-(0.982 \pm 0.423)$	$-(0.975 \pm 0.562)$	$-(0.979 \pm 0.338)$
97.0	$+(0.521 \pm 0.301)$	$+(0.598 \pm 0.338)$	$+(0.555 \pm 0.225)$

band electrons by the external field. By correlating the temperature dependence of the Knight shift with that of the susceptibility, they find that $H_{hf}^{(d)} = K_d H_{ext} = \beta \chi_d H_{ext} = \beta \mu_d$, where $H_{hf}^{(d)}$ is the hyperfine field arising from the induced moment and gives the temperature-dependent part of the Knight shift K_d. H_{ext} is the external applied field, χ_d is the temperature-dependent d-moment atomic susceptibility, β is a constant, and μ_d is the d moment induced on a Pd atom. From their measurements, they determine the proportionality constant $\beta = H_{hf}^{(d)}/\mu_d = -345$ kG/μ_B. The dominant contribution to β is due to core polarization,[6] but β will also include an orbital effect and a 5s contribution, both proportional to the d moment.

Our estimates for the local Pd moment contribution to the hyperfine field based on the above value for β are of the order of -60 and -70 kG for Pd concentrations between 50 and 70 at.%, and are smaller in magnitude near the ends of the concentration range. Thus, this estimated contribution is too small to account for the measured fields at least for $c < 0.85$ (see Fig. 1). The remainder of the large negative fields probably arises from the interaction with neighboring atoms through the conduction electrons. Previously, models have been proposed[8,11] which assume that the hyperfine field due to conduction-electron polarization H_{hf}^{cond} is proportional to the average magnetic moment per atom for the alloy, $\bar{\mu}$. These models apparently do not represent H_{hf}^{cond} for either component of this alloy system since $\bar{\mu}$ is relatively independent of concentration for $c < 0.50$,[28] while $dH_{hf}/dc > 0$ for both Pd and Ni[17,25] in this concentration range. However, these models do not account properly for the interaction of the conduction electrons with the individual alloy atoms. A model which gives explicit recognition both to potential and to spin-dependent scattering of the conduction electrons from the ionic cores of the constituent atoms of the alloy would be more appropriate for the Ni-Pd system. Potential scattering is expected to arise since there are fewer d-band electrons in Ni metal[29] than in Pd metal.[30] Such a model would be analogous to those invoked by Daniel and Friedel[9] and Campbell[10] in the discussion of hyperfine fields in dilute alloys.

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[1]J. Crangle and W. R. Scott, J. Appl. Phys. 36, 921 (1965); R. M. Bozorth, P. A. Wolff, D. D. Davis, V. B. Compton, and J. H. Wernick, Phys. Rev. 122, 1157 (1961); J. Crangle, Phil. Mag. 5, 335 (1960).

[2]P. P. Craig, D. E. Nagle, W. A. Steyart, and R. D. Taylor, Phys. Rev. Letters 9, 12 (1962).

[3]G. G. Low and T. M. Holden, Proc. Phys. Soc. (London) 89, 119 (1966).

[4]R. E. Watson and A. J. Freeman, Phys. Rev. 123, 2027 (1961).

[5]A. J. Freeman and R. E. Watson, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic, New York, 1965), Vol. IIA, p. 168 ff.

[6]R. E. Watson, in *Hyperfine Interactions*, edited by A. J. Freeman and R. Frankel (Academic, New York, 1967).

[7]D. A. Shirley and G. A. Westenbarger, Phys. Rev. 138, A170 (1965); D. A. Shirley, S. S. Rosenblum, and E. Matthias, *ibid.* 170, 363 (1968).

[8]R. L. Streever and G. A. Uriano, Phys. Rev. 139, A135 (1965).

[9]E. Daniel and J. Friedel, J. Phys. Chem. Solids 24, 1601 (1963).

[10]I. A. Campbell and A. A. Gomiés, Proc. Phys. Soc. (London) 91, 319 (1967); I. A. Campbell, Proc. Roy. Soc. (London) A311, 131 (1969); J. Phys. C 2, 1338 (1969).

[11]E. Daniel, in *Hyperfine Structure and Nuclear Radiations*, edited by E. Matthias and D. A. Shirley (North-Holland, Amsterdam, 1968).

[12]A preliminary report of this work was given in *Proceedings of the International Conference on Hyperfine Interactions and Excited Nuclei, Rehovot-Jerusalem, Israel*, 1970 (Gordon and Breach, New York, to be published).

[13]Masaaki Kontani and Junkichi Itoh, J. Phys. Soc. Japan 22, 345 (1967).

[14]J. Murray, T. A. McMath, and J. A. Cameron, Can. J. Phys. 45, 1813 (1967).

[15]L. O. Norlin, K. Johansson, E. Karlsson, and M. R. Ahmed, in Ref. 12.

[16]H. Frauenfelder and R. M. Steffen, in *Alpha, Beta, and Gamma Ray Spectroscopy*, edited by K. Siegbahn (North-Holland, Amsterdam, 1965), Vol. 2, p. 997; R. M. Steffen and H. Frauenfelder, in *Perturbed Angular Correlations*, edited by E. Karlsson, E. Matthias, and K. Siegbahn (North-Holland, Amsterdam, 1964).

[17]F. E. Obenshain, W. Gläser, G. Czjzek, and J. E. Tansil, in Ref. 12.

[18]J. Spruiell (private communication); Wen Lin, thesis (University of Tennessee, 1969) (unpublished).

[19]E. D. Klema and F. K. McGowan, Phys. Rev. 92, 1469 (1953).

[20]J. Hattula, J. Kantela, and A. Sarmanto, Nucl. Instr. Methods 65, 77 (1968).

[21]Vikram Singh, J. Phys. Soc. Japan 29, 1111 (1970).

[22]P. H. Stelson and L. Grodzins, Nucl. Data Tables, U. S. At. Energy Comm. 1, 21 (1965).

[23]K. Auerbach, K. Siepe, J. Wittkemper, and H. J. Körner, Phys. Letters 23, 367 (1966).

[24]A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. 16, 27 (1953).

[25]U. Erich, J. Göring, S. Hüfner, and E. Kankeleit, Phys. Letters 31A, 492 (1970).

[26]J. W. Cable and H. R. Child, Bull. Am. Phys. Soc. 14, 320 (1969); and private communication.

[27]J. A. Seitchik, A. C. Gossard, and V. Jaccarino, Phys. Rev. 136, A1119 (1964).
[28]G. Fischer, A. Herr, and A. J. P. Meyer, J. Appl. Phys. 39, 545 (1968); Jean-Paul Burger, thesis (University of Strasbourg, 1964) (unpublished); C. Sadron, Ann. Phys. (Paris) 17, 371 (1932).
[29]L. Hodges, H. Ehrenreich, and N. D. Lang, Phys. Rev. 152, 505 (1966).
[30]J. J. Vuillemin and M. G. Priestley, Phys. Rev. Letters 14, 307 (1965); J. J. Vuillemin, Phys. Rev. 144, 396 (1966).

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Series Expansions for High-Temperature Dynamics of Heisenberg Paramagnets*

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High-temperature expansions are given for the second and fourth moments of the frequency in the spin-correlation function and the frequency-dependent susceptibility. The results are applicable to loose-packed cubic Bravais lattices with general spin values and nearest-neighbor interactions. At small wave vectors, for both the ferromagnet and the antiferromagnet, the series for the reciprocal of the second moment shows the expected divergence at the critical temperature. Insufficient terms are available to yield accurate critical indices. There is good agreement between the calculation and neutron-scattering experiments on $RbMnF_3$. Paramagnon peaks are not likely to be present at temperatures of order five to ten times the critical temperature in the antiferromagnets considered, but probably do occur at favorable points in the zone for the corresponding ferromagnets.

I. INTRODUCTION

The problem of describing the dynamics of a Heisenberg paramagnet has not been solved. For infinite temperatures, a number of papers have given different approximate treatments which are substantially consistent with one another.[1-9] In the temperature region near the critical ordering temperature, the theory of Kawasaki[10] and the dynamic scaling laws[11,12] give a description of the dynamics for long wavelengths.

The purpose of the present paper is to investigate the region of temperature intermediate between these two extremes. The method of moments[1,4,5] is used to describe the dynamics in conjunction with a high-temperature series expansion. This type of treatment appears to have been first used by Sears[13] and was developed in more detail by Tahir-Kheli and McFadden.[5] The present paper starts by repeating this latter work to eliminate some algebraic errors (pointed out in Refs. 6 and 14) and to give a further term in the second-moment expansion.

The form of the series at long wavelengths essentially gives a high-temperature expansion for some dynamic critical properties. Use of such expansions (for review, see Ref. 15) has proved to be one of the most powerful tools for investigating static critical properties; this present work appears to be the first application of this technique to dynamic properties.

The properties calculated are observable quantitatively by current neutron-scattering techniques. The physical principles and techniques involved have been described in the literature[16,17] and will not be discussed in the present paper. A few such measurements have been made and these are compared to the theory in Sec. IV of the paper.

II. FORMAL THEORY

First, formal expressions will be given for the dynamic spin-correlation function and the generalized susceptibility. Then the moments of their frequency spectrum are obtained and expressed as a high-temperature series expansion.

The dynamic spin-correlation function $S^{\alpha\alpha}(\vec{k}, \omega)$ is the spatial and temporal Fourier transform of the two-spin-correlation function and is given by

$$S^{\alpha\alpha}(\vec{k}, \omega) = \frac{1}{2\pi N} \sum_{\vec{n}, \vec{m}} \int_{-\infty}^{\infty} e^{i\vec{k}\cdot(\vec{n}-\vec{m})-i\omega t} \langle S_{\vec{n}}^{\alpha}(0) S_{\vec{m}}^{\alpha}(t) \rangle_T \, dt \,, \tag{1}$$

where the superscript $\alpha = x$, y, or z, N is the number of atoms in the crystal, $\vec{n}$ and $\vec{m}$ are atomic positions and $S_{\vec{m}}^{\alpha}(t)$ is the α component of spin on the atom at $\vec{m}$ at time t. The expectation value is to be evaluated at the temperature T of the crystal.

It follows that[1]

$$\int_{-\infty}^{\infty} S^{\alpha\alpha}(\vec{k}, \omega) \omega^p \, d\omega = N^{-1} \sum_{\vec{n}, \vec{m}} e^{i\vec{k}\cdot(\vec{n}-\vec{m})} \langle S_{\vec{n}}^{\alpha} \mathcal{L}^p S_{\vec{m}}^{\alpha} \rangle_T \,, \tag{2}$$

where p is a positive integer or zero and $\mathcal{L}$ is the Liouville operator representing a commutation with the Hamiltonian.

Following the method of Opechowski[18] and of Rushbrooke and Wood,[19,20] a high-temperature ex-