fields ($H_0=150$ Oe) in an attempt to determine the zero-field nuclear-resonance frequency. The result is that if we extrapolate from $H_0=150$ Oe we find that $\omega_n(H_0=0)/2\pi\sim 40$ Mc/sec. To account for this zero-field NMR frequency, an in-plane anisotropy of order 10^{-2} Oe would be sufficient. The very low in-plane anisotropy energy for the magnetic electrons in CsMnF$_3$ results in a high degree of "frequency pulling" for the NMR frequency; CsMnF$_3$ is a most striking example of the importance of cooperative effects in tightly coupled nuclear-electron systems.

We have used $H_e=3.1\times10^5$ Oe and $\omega_0/2\pi=666$ Mc/sec to obtain the theoretical fit to the experimental results. If volume corrections for the F^- octahedra surrounding a Mn^{2+} ion are made, Ogawa's[11] paramagnetic resonance studies would give $\omega_0/2\pi=687\pm7$ Mc/sec as the value for the average hyperfine field frequency; this would suggest a $3.0\pm1.0\%$ zero-point spin-wave reduction in $\langle S\rangle$ for CsMnF$_3$. The linked-cluster expansion calculated by Davis[12] predicts a 4.36% reduction in $\langle S\rangle$.

The excitation of the nuclear resonance by the applied rf magnetic field is via an indirect mechanism. The resonance is excited by a rocking of the electronic sublattice magnetization[13] at the driving rf frequency. From Eq. (7), if $H_0=3000$ Oe and $T=4.2$°K one has $\eta\cong150$, which gives a signal to noise enhancement of $\sim2\times10^4$.

ACKNOWLEDGMENTS

We would like to thank Professor A. M. Portis for his assistance and for the many invaluable discussions during the course of this work.

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Magnetic Moments and Unpaired-Electron Densities in CoPt$_3$

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A polarized-neutron investigation of the ferromagnetic alloy CoPt$_3$ in various degrees of chemical order is presented. The magnetic moments of the two components have been determined; that of the cobalt is $1.64\pm0.04\,\mu_B$, and that of the platinum $0.26\pm0.02\,\mu_B$ in the fully ordered material. The experimental form factors of platinum and cobalt were measured and it is shown that they are in fair agreement with recent theoretical calculations. A Fourier inversion of the data was also carried out to show the t_{2g} character of the cobalt d electrons of this alloy.

I. INTRODUCTION

SEVERAL theoretical and experimental investigations have been recently carried out on the microscopic magnetic properties of $3d$ transition elements and their alloys. However, little is known about other elements where the incomplete inner shells of electrons are the $4d$ or $5d$. They do not exhibit any ferromagnetism in the pure metallic state but seem capable of ferromagnetic interaction when alloyed with elements of the iron transition group. Magnetization measurements of such alloys at different concentrations[1] lead to the hypothesis that both $4d$ and $5d$ atoms can interact magnetically, and this hypothesis has been recently confirmed by neutron-diffraction studies for some $4d$ elements, namely Pd,[2,3] Rh,[4] and Zr.[5] However, the magnetization results are difficult to interpret and the models proposed to explain the measurements were often admitted to be inadequate.[1]

Among the $5d$ elements Pt has attracted the attention of many investigators. Bulk magnetization has been measured for several ferromagnetic alloys with $3d$ elements and the neutron diffraction technique has been used for Co-Pt,[6] Mn-Pt, and Cr-Pt[7] systems. The presence of a magnetic moment on the Pt atom has been

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ascertained; however, the data were collected on polycrystalline samples and no experimental determination of the Pt form factor was accomplished. On the other hand, in the compounds $FePt_3$ [8] and MnPt,[9] where the moments are ordered antiferromagnetically, no moment on Pt was found. We have focused our attention on the $CoPt_3$ alloy with the purpose of detecting, by polarized-neutron analysis coupled with magnetization measurements, any ordered moment on Pt atoms and the relative magnetic form factor.[10] Furthermore it seemed interesting to determine whether the magnetic electron configuration of cobalt was in any way affected by the presence of Pt atoms. $CoPt_3$, which has a fcc structure, undergoes an order-disorder transition (fcc Cu_3Au $L1_2 \rightarrow$ fcc $A1$).[11,12] Magnetization curves[13,14] indicate a strong dependence of the magnetic state upon the degree of order. Consequently both bulk-magnetization and polarized-neutron measurements have been performed by us at different degrees of order, to investigate a possible order dependence of magnetic moments of the two components.

II. EXPERIMENTAL

A. Samples

Five different samples were cut from a large single crystal.[15] Two of them were thin circular slabs 10 mm in diameter and 1.2 and 0.6 mm thick, respectively. The remaining three crystals were pillars about 2 cm high and with thickness 0.5, 0.7, and 1.1 mm, respectively. The samples were cold-worked to increase their mosaic spread. The η parameter, defined according to G. E. Bacon,[16] lay between 0.15° and 0.4° for the various samples. Chemical analysis showed that the exact composition was $CoPt_{3.02}$ with an impurity content of about 0.2% by weight. Metallographic analysis was also carried out on a sample and it could be ascertained that a single phase was present with no precipitated materials.

B. Ordering Treatment

In order to obtain various degrees of chemical order the samples were subjected to different heat treatments. In particular, no ordering could be detected in samples that had been annealed for over 100 h at 650°C±10°C, while practically complete order (order parameter $S=0.9\pm0.1$) could be achieved by annealing the samples at 600±10°C for a time between 100 h and 600 h depending on previous treatment of the sample, the longer time being necessary for the sample with the larger mosaic-spread parameter.

TABLE I. Heat treatment and Curie temperatures of samples.

Measured order parameter	Measured Curie temperature (°K)	Temperature of annealing (°C)	Time of annealing (h)	Rate of cooling to room temperature
<0.1	460	1200	2	30°C/sec
<0.1	...	650	100	20°C/sec
0.6	400	630	100	20°C/sec
>0.9	320	600	100–600	50°C/h or 20°C/sec

The annealing times for the different runs are reported in Table I, where the order parameters as detected by neutron diffraction are also given. From the analysis of the half-height width of both the fundamental and superlattice peaks, information could also be obtained on the homogeneity of the partially ordered state. No appreciable difference in such a width was detected, which means, taking into account our instrumental conditions, that ordered regions should have an average diameter no smaller than 200 Å.

C. Polarized-Neutron Technique

This technique has been described in detail elsewhere.[17] The ratio γ of the magnetic to the nuclear structure factor is obtained directly from the ratio of the diffracted intensities of neutrons polarized parallel and antiparrallel to the direction of the magnetic field acting on the sample. The neutron experiments were carried out at room temperature for $S<0.1$ and at 77°K temperature for $S=0.6$ and $S>0.9$. In the latter case the sample was held in a liquid-nitrogen cryostat. Possible sources of errors are depolarization, extinction, and multiple scattering; therefore a careful analysis, as described hereafter, was carried out so that our results were not substantially affected by these errors.

The polarization of the incident beam was 99%±1% and the efficiency of inversion of the spin state was 98%±2% in all measurements. Extinction was found only in the first two fundamental reflections with the thickest sample. The absence of extinction in all other cases was checked by varying the wavelength of the neutron beam in steps from $\lambda=1.03$ Å to $\lambda=0.80$ Å. Another independent check of the absence of extinction was the constancy of the γ value with variation of the optical path of the neutron beam inside the crystal. Multiple scattering depends on the position of the reciprocal lattice with respect to the direction of the

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wave vector of the incident neutrons and on the wavelength of the neutrons. Therefore a check on the presence of multiple scattering can be made by rocking the crystal around the scattering vector and/or varying the wavelength of the neutrons. Both kinds of analysis were carried out on several reflections. In particular all the first fundamental and superlattice reflections were carefully examined and also some of the reflections more sensitive to the asphericities of the spin density (see below), that is the (511), (333) and (600), (442) pairs. In no case was any effect of multiple scattering found. No appreciable contamination from half-wavelength neutrons was found.

D. Magnetic Measurement

Bulk-magnetization measurements as a function of temperature were carried out on two of the samples used for the neutron measurements. The magnetization was measured by the ballistic method with a maximum error of about 2%, and the temperature was determined within 1°C using Chromel-Alumel thermocouples in good thermal equilibrium with the sample. For measurements below room temperature the sample was maintained in a cryostat cooled with a regulated flow of liquid nitrogen; above room temperature the sample was in thermal contact with a copper rod electrically heated.

III. SPIN DENSITY

For the ordered samples 46 reflections were investigated, including all fundamental reflections out to $(\sin\theta)/\lambda = 0.95$ Å^{-1} and all superlattice reflections out to $(\sin\theta)/\lambda = 0.75$ Å^{-1}. At the intermediate degree of order 20 reflections were studied and 18 for the disordered state. All measurements, with the exception of those on the disordered samples, had to be carried out at liquid-nitrogen temperature, since the Curie temperature is close to room temperature.

To obtain the magnetic structure factor from the measured γ values, one has to multiply them by the calculated nuclear structure factor, which involves the knowledge of the nuclear scattering amplitude of the components. The nuclear scattering amplitude of Pt was obtained, relative to that of Fe as given by Shull and Yamada[18] ($b_{\mathrm{Fe}} = 0.951 \times 10^{-12}$ cm), by measuring the γ value of the first superlattice peak of a $\mathrm{Fe_3Pt}$ alloy single crystal. The γ values of superlattice reflections of this compound are extremely sensitive to the difference of the nuclear scattering amplitude of the two components, when this difference is small, as it happens in $\mathrm{Fe_3Pt}$. The value found for the Pt nuclear amplitude was $b = 0.947 \pm 0.010 \times 10^{-12}$ cm. The nuclear amplitude of scattering for cobalt was assumed to be $b_{\mathrm{Co}} = 0.250 \pm 0.003 \times 10^{-12}$ cm.[19]

In calculating the magnetic structure factors from the γ values the possibility of a difference between the Debye-Waller factors of the two components was neglected because of the low temperature of the samples during the experiments.

A. Fully Ordered State

For the fully ordered state the values of the magnetic structure factors, expressed in Bohr magnetons, are reported in Table II and Figs. 1 and 2. The errors quoted are the standard deviations of repeated measurements and include also effects of the uncertainties in the exact value of the polarization and inversion efficiency. In Fig. 1 is also reported the value of the magnetic moment per unit cell which has been obtained by the bulk-magnetization measurements, namely $n_0 = (2.43 \pm 0.03)\mu_B$; this value, which is slightly at variance with the value previously published,[10] was obtained with a more sensitive apparatus, and it is in excellent agreement with the value reported in the literature.[14] In Fig. 2 is also given the smooth curve of the magnetic structure factor for superlattice peaks that have been extrapolated to $(\sin\theta)/\lambda = 0$, from which one obtains $\mu_{\mathrm{Co}} - \mu_{\mathrm{Pt}} = (1.38 \pm 0.04)\mu_B$. Combining this result with the magnetic moment of the unit cell, we obtain the individual magnetic moments of the Co and Pt atoms: $\mu_{\mathrm{Co}} = (1.64 \pm 0.04)\mu_B$ and $\mu_{\mathrm{Pt}} = (0.26 \pm 0.02)\mu_B$. This procedure is correct when there is only one kind of atom in each atomic site or, if this condition is not exactly satisfied, when the magnetic moment on each component placed in a wrong site does not undergo a drastic variation.

TABLE II. Magnetic-structure factors F_M in units of Bohr magnetons, for the $\mathrm{Cu_3Au}$-type unit cell of ordered $\mathrm{CoPt_{3.02}}$.

Fundamental reflections		Superlattice reflections	
(*hkl*)	F_M	(*hkl*)	F_M
111	1.42±0.02	100	1.19 ±0.02
200	1.21±0.02	110	1.086±0.013
220	0.77±0.03	210	0.826±0.016
311	0.41±0.03	211	0.775±0.010
222	0.44±0.02	300	0.531±0.016
400	0.22±0.02	221	0.59 ±0.02
331	0.28±0.03	310	0.49 ±0.02
420	0.15±0.03	320	0.44 ±0.03
422	0.12±0.03	321	0.38 ±0.02
511	0.04±0.03	410	0.29 ±0.02
333	0.18±0.03	322	0.32 ±0.03
440	0.14±0.04	411	0.25 ±0.02
531	0.02±0.03	330	0.29 ±0.02
600	0.00±0.01	421	0.18 ±0.02
422	0.09±0.02	332	0.18 ±0.02
620	0.00±0.03	500	0.10 ±0.02
533	0.02±0.04	430	0.21 ±0.02
622	−0.02±0.04	510	0.11 ±0.02
444	0.10±0.04	431	0.22 ±0.04
711	0.02±0.02	520	0.05 ±0.03
551	−0.02±0.03	432	0.14 ±0.03
640	0.01±0.03	521	0.13 ±0.03
553	0.02±0.04	441	0.13 ±0.04

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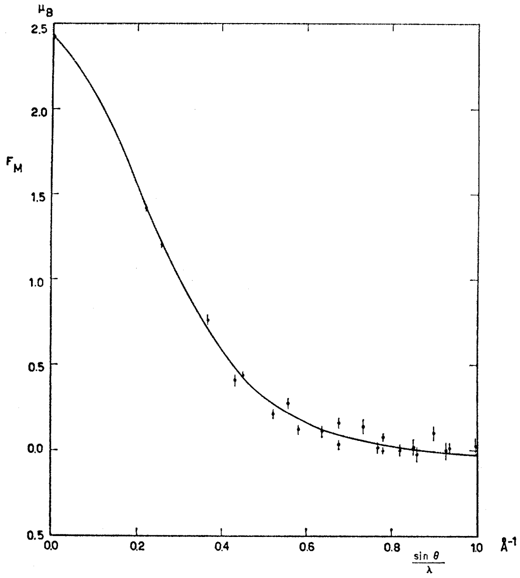

FIG. 1. Magnetic structure factors, in units of Bohr magnetons, of the fundamental reflections of the ordered alloy. The line is a smooth curve traced along experimental points. The point at $\sin\theta/\lambda=0$ is obtained from the magnetization measurements.

In our case 93% of the cubic corners were occupied by Co atoms. On the other hand the measurements in the disordered state indicate, as is reported in the next section, a decrease of 20% in the magnetic moment of the cobalt atom of the disordered state compared with the ordered one. Therefore, one can estimate that the magnetic moment of cobalt, as given by this evaluation, can be affected no more than 5% by the lack of complete order. A similar correction can be applied to the Pt moment.

B. Form Factors

In principle, having determined the moments of both cobalt and platinum, it is possible to calculate the magnetic form factor from the magnetic structure factors of the different reflections. It is known from previous experimental data[20] that in general the magnetic form factor is built up from a spherical part and an aspherical one. This has been explained, in the one-electron model for a free atom, in terms of the deviation from the statistical average of the population of the d sublevels (in cubic symmetry e_g and t_{2g}) into which the crystalline field splits the d shell.

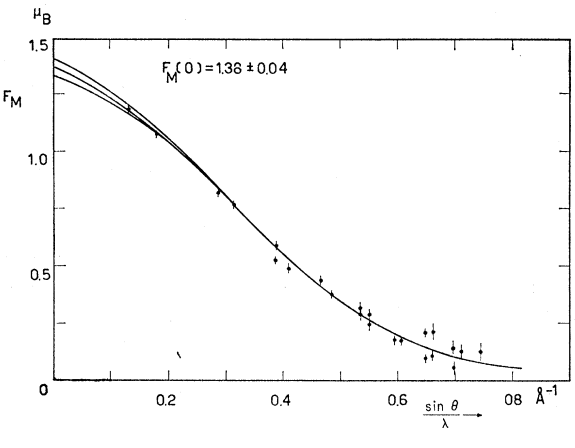

FIG. 2. Magnetic structure factors, in units of Bohr magnetons, of the superlattice reflections of the ordered alloy. The line is a smooth curve traced along experimental points. The extrapolation procedure at $(\sin\theta)/\lambda=0$ is also shown (see text).

In our case the spherical part of the platinum and cobalt form factors can be evaluated by making use of the previously determined magnetic moments and of the smooth curves of the magnetic structure factors. The results are shown in Fig. 3, where the experimental spherical form factor of platinum (upper curve) is reported together with a theoretical form factor computed by Cromer and Waber.[21] They made use of self-consistent-field calculations of relativistic wave functions including the Slater approximate exchange correction.[22] In Fig. 4 the experimental spherical form

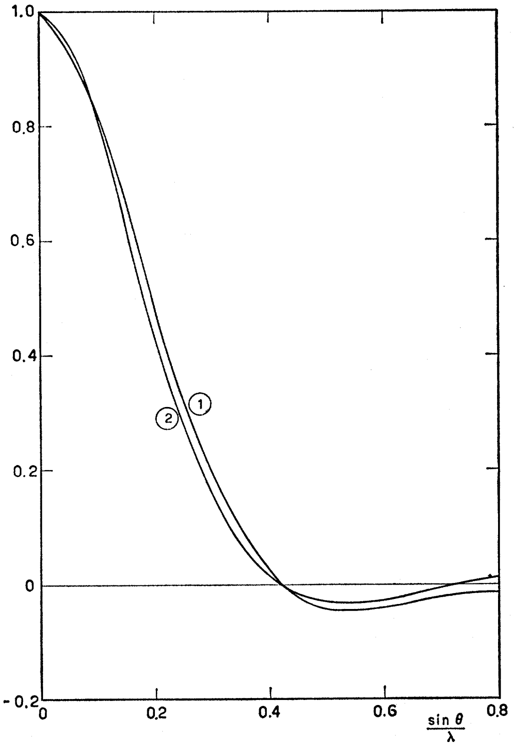

FIG. 3. Magnetic form factor of platinum in ordered $CoPt_3$ alloy (line 1), and theoretical form factor of platinum as given by (21) for an electronic configuration: 4 electrons in $5d_{3/2}$ orbital and 5 or 6 in the $5d_{5/2}$ orbital. Estimated error on experimental form factor less than 0.04 units.

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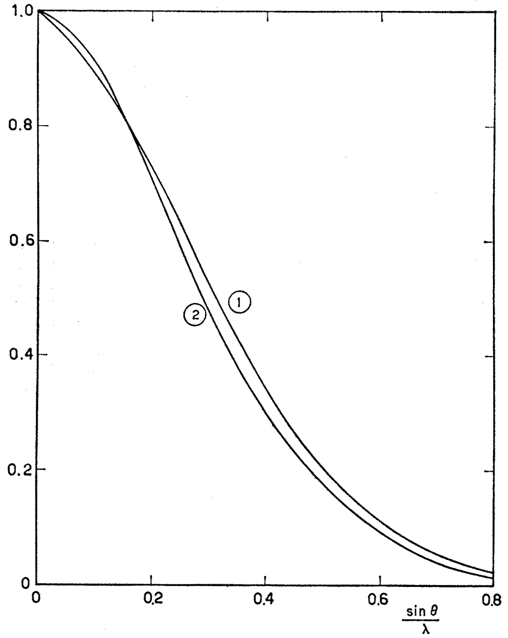


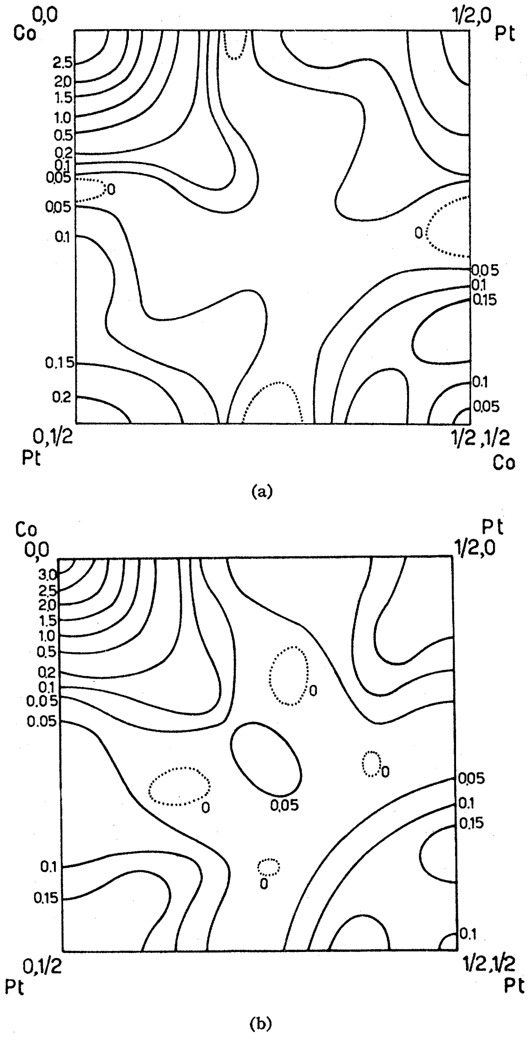
FIG. 4. Magnetic form factor of cobalt in the ordered $CoPt_3$ alloy (line 1) and theoretical form factor as given by (23). Estimated error on experimental form factor less than 0.02 units.

factor of cobalt (upper curve) is given and the form factor (lower curve) for the $3d$ electrons in $3d^74s^2$ configuration computed from nonrelativistic Hartree-Fock wave functions but with exchange directly taken into account.[23] As is seen in both cases, the theoretical curve lies slightly below the experimental curve obtained by us; this might be due to the fact that form-factor calculations do not take into account exchange polarization and that no correction for the orbital part of the form factor has been considered. Both corrections would act in the right direction to take the theoretical curve closer to the experimental one.[24,25]

C. Magnetic-Density Maps

A Fourier inversion of the magnetic-structure-factor data was carried out, making use of the IBM 7040 computer, for the three-dimensional sections in the (001) and (110) planes and the projection along the [001] axis on the (001) plane. Densities of magnetic moments at about one thousand points for each Fourier inversion were obtained. The result of the projection is shown in Fig. 5, where the contours are labelled in units of (Bohr magnetons)/Å^2. The presence of a magnetic moment on the Pt sites is confirmed. In order to ascertain in which direction the lobes of the cobalt magnetic-electron density apparent in this projection are pointing, one has to refer to the three-dimensional sections in (001) and (110) planes reported in Figs. 6 and 7, respectively. In these figures the contours are labelled in units of (Bohr magnetons)/Å^3. From these maps it can be seen that the cobalt magnetic electron density clearly extends in the [111] direction, while for the platinum, part of the electrons is along the [100] directions contained in each basal plane and part along the [111] directions. It can also be seen that the distribution of the platinum unpaired-spin density is more spread out than that of cobalt, as is evident also from the form-factor curves. From Fig. 5 it is also seen that terminations errors of the Fourier synthesis should not affect our conclusions appreciably.

Referring again to the one-electron model of a free atom in a cubic field, one finds that the electrons belonging to the two d sublevels have different spatial configuration, the t_{2g} electrons pointing in the [111]



(a)

(b)

FIG. 5. Magnetic electron density projected on the basal plane (001) of the cell along the [001] direction obtained by Fourier inversion. The figure covers $\frac{1}{4}$ of the full projection. Contours are labelled in units of (Bohr magnetons)/Å^2. (a) All reflections of the [001] zone out to $(\sin\theta)/\lambda \leq 0.65$ Å^{-1} are included; (b) *idem* out to $(\sin\theta)/\lambda \leq 0.75$ Å^{-1}.

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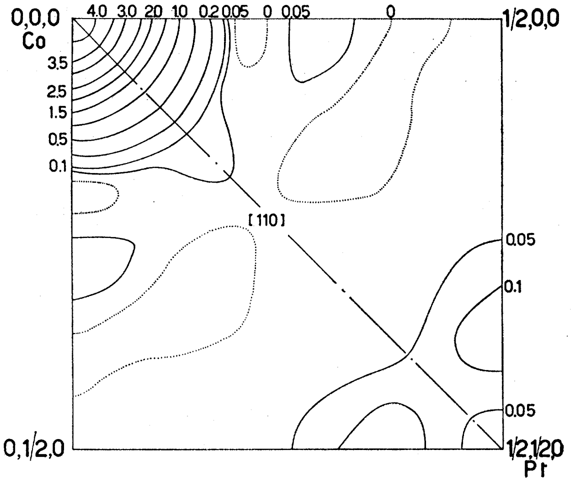

FIG. 6. Three-dimensional magnetic electron density along the basal plane (001) of the cell, obtained by Fourier inversion. The figure covers $\frac{1}{4}$ of the full section. Contours are labelled in units of (Bohr magnetons)/Å^3. All reflections out to $\sin\theta/\lambda \leq 0.75$ Å^{-1} are included.

direction of the cubic field, the e_g in the [100]. According to what has been noticed on the Fourier maps, one can then say that the character of the unpaired electrons of both the cobalt atoms is mostly t_{2g}; that is, the unpaired-electron population of the two sub-bands is weighted more toward the t_{2g}-like sub-band than toward the spherical-symmetry distribution.

The predominantly t_{2g} character of unpaired electrons is seen also in the free-atom one-electron approximation, where the form factor can be written for a cubic field as[26]:

$$f(hkl) = f_{\text{spher.}} + (\tfrac{5}{2}x - 1) A(hkl) f_{\text{aspher.}}, \qquad (1)$$

where

$$A(hkl) = [h^4 + k^4 + l^4 - 3(h^2k^2 + h^2l^2 + k^2l^2)]/(h^2 + k^2 + l^2)^2, \qquad (2)$$

where $f(hkl)$ is the magnetic form factor for the (hkl) reflection, x is the population of the e_g level and $1-x$ that of the t_{2g} level, and $A(hkl)$ is a cubic harmonic depending only on the Miller indexes h,k,l of the reflecting plane. It can be seen that every reflection for which $A(hkl) > 0$ lies below the smooth structure-factor curve (see Figs. 1 and 2) and vice versa. This means that, in this model, $x < 0.4$, i.e. the electrons are of t_{2g} character. An exact value of x cannot be given at the moment, since, as far as we know, no calculation of the aspherical form factor of platinum has been made. We must point out that in fcc cobalt the unpaired electron, on the contrary, shows a predominantly e_g character.[27]

D. Measurements at Different States of Order

TABLE III. Disordered samples. It is shown how the smoothed magnetic structure factors F_M^{expt} are fitted by the magnetic structure factors F_M^{calc} calculated, making use of the magnetic moments obtained for $S=0$ [$\mu_{\text{Co}} = (1.29 \pm 0.09)\mu_B$ and $\mu_{\text{Pt}} = (0.45 \pm 0.03)\mu_B$] together with the form factors of Co and Pt found for ordered samples (see Figs. 3 and 4). F_M is in units of Bohr magnetons; $(\sin\theta)/\lambda$ in Å^{-1}.

$(\sin\theta)/\lambda$	F_M^{expt}(smoothed)	F_M^{calc}
0.0	2.64±0.02	2.64±0.13
0.1	2.29±0.03	2.26±0.11
0.2	1.65±0.07	1.58±0.08
0.3	0.88±0.07	0.93±0.05
0.4	0.48±0.03	0.48±0.03
0.5	0.24±0.03	0.22±0.02
0.6	0.10±0.03	0.10±0.01
0.7	0.02±0.02	0.04±0.01
0.8	0.02±0.02	0.02±0.00

From the magnetic-density maps for the ordered state we infer that the magnetic moments on cobalt and platinum seem to be fairly localized. It seems then reasonable to make the hypothesis that the magnetic form factors of cobalt and platinum are not strongly order-dependent.

In the disordered state it is then possible to obtain the magnetic moments of the two components from the coherent peaks, owing to the fact that the form factor of cobalt is very different from that of platinum. In fact the magnetic moments can be obtained as the parameters of the linear combination of form factors which best fits the magnetic structure factors of fundamental reflections. At liquid-nitrogen temperatures the magnetic moment per unit cell was $n_0 = (2.65 \pm 0.03)\mu_B$ and the calculated individual magnetic moments were: $\mu_{\text{Pt}} = (0.45 \pm 0.03)\mu_B$ and $\mu_{\text{Co}} = (1.29 \pm 0.09)\mu_B$. The fit is good, as can be seen from Table III.

As far as the intermediate degree of order ($S=0.6$) is concerned, since it was possible to measure the value of the first superlattice peaks we could follow the same procedure outlined for the fully ordered case. At liquid-nitrogen temperature the magnetic moment per unit cell was $n_0 = (2.50 \pm 0.04)\mu_B$ and the magnetic moment of the cobalt site was $(1.53 \pm 0.07)\mu_B$ and that of the platinum site $(0.32 \pm 0.03)\mu_B$. Emphasis should be put

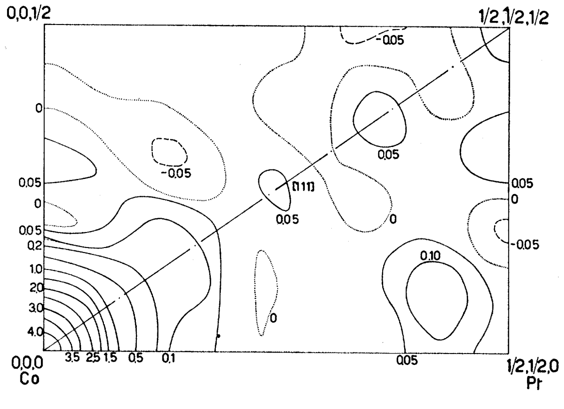

FIG. 7. Three-dimensional section of magnetic electron density along the diagonal plane (110) of the cell, obtained by Fourier inversion. The figure covers $\frac{1}{4}$ of the full section. Contours are labelled in units of (Bohr magnetons)/Å^3. All reflections with $(\sin\theta)/\lambda \leq 0.75$ Å^{-1} are included.

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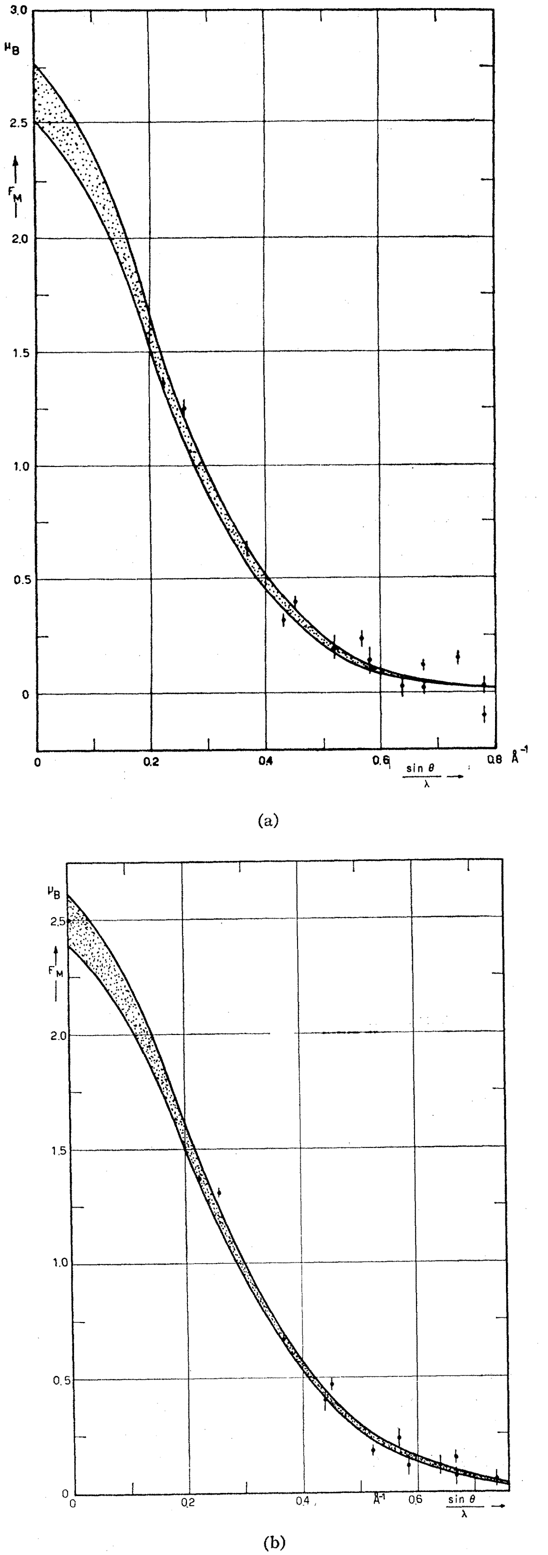

(a)

(b)

FIG. 8. Magnetic structure factors, in units of Bohr magnetons, of the fundamental reflections of the alloy in two states with order parameters $S<0.1$ (a), and $S=0.6$ (b). Shaded areas indicate the values, with estimated errors, of magnetic structure factors calculated for a spherical distribution of electrons (see Tables III and IV, respectively).

on the fact that in this case of intermediate order, the individual moments are ascribed to the atomic sites instead to the atoms, since the superlattice reflections give the difference of the mean magnetic moments located on the corners and the face centers of the cubic cell; while in the case of perfect order, there is coincidence of the magnetic moment of the site with that of the atom. Nevertheless if one follows the same procedure used for the disordered case, i.e., fitting the fundamental reflections with the two different form factors, one obtains the moments relative to the atoms and, as can be seen from Table IV, the fit is satisfactory in this case also. Thus, for this intermediate degree of order it was not possible to find any difference, within the experimental errors, between the values of the magnetic moments relative to the atomic sites and those relative to the atoms.

TABLE IV. Intermediate degree of order: $S=0.6$. It is shown how the smoothed magnetic structure factors F_M^{expt} both for the fundamental and for the superlattice reflections are fitted by the calculated magnetic structure factors, making use of the magnetic moments obtained for $S=0.6$ [i.e., $\mu_{\text{Co}}=(1.53\pm0.07)\mu_B$, $\mu_{\text{Pt}}=(0.32\pm0.03)\mu_B$] together with the form factors of Co and Pt found for ordered samples (see Figs. 3 and 4). F_M is in units of Bohr magnetons; $(\sin\theta)/\lambda$ in Å^{-1}.

	Fundamental reflections		Superlattice reflections	
$(\sin\theta)/\lambda$	F_M^{expt}(smoothed)	F_M^{calc}	F_M^{expt}(smoothed)	F_M^{calc}
0.0	2.50±0.04	2.50±0.11	1.21±0.09	1.21±0.08
0.1	2.13±0.07	2.15±0.09	1.09±0.06	1.10±0.06
0.2	1.62±0.08	1.56±0.06	0.86±0.06	0.95±0.05
0.3	1.00±0.05	0.98±0.04	0.59±0.09	0.74±0.04
0.4	0.54±0.04	0.56±0.02	0.40±0.11	0.53±0.02
0.5	0.28±0.04	0.28±0.02		
0.6	0.13±0.03	0.14±0.01		
0.7	0.07±0.03	0.06±0.01		
0.8	0.05±0.03	0.02±0.00		

Considering the asphericity in the magnetic electron distribution already discussed for the ordered materials, a careful analysis showed that there was no appreciable variation of this distribution with the variation of the degree of order. In fact all the reflections which are most sensitive to asymmetries in the population of the electronic sub-bands show the same t_{2g} tendency for all three states of order investigated, as can be seen from Figs. 8(a) and 8(b).

IV. CONCLUSION

The variation of magnetic moment on both cobalt and platinum with the degree of order, i.e., with the number of nearest-neighbor pairs Co-Co, Pt-Pt, and Co-Pt, seems to indicate that interatomic exchange interactions are important in the $CoPt_3$ system.

Belov,[28] analyzing the temperature dependence of the magnetization near the Curie point, suggested that in $CoPt_3$ the smearing out of the transition with the increase of order could be related to the existence of "islands" of disordered phase in an ordered matrix. However, the appreciable change of the γ values of the

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[28] K. P. Belov, *Magnetic Transitions* (Consultants Bureau Enterprises, Inc., New York, 1961), Chap. III.

TABLE V. Comparison of the γ values of the first superlattice reflections in two states with order parameter $S>0.9$ and $S=0.6$, respectively.

(hkl)	$S>0.9$ $-\gamma$	$S=0.6$ $-\gamma$
(100)	0.460 ± 0.009	0.400 ± 0.018
(110)	0.419 ± 0.005	0.331 ± 0.019
(210)	0.317 ± 0.006	0.193 ± 0.022
(211)	0.299 ± 0.004	$0.24\ \pm0.04$
(221)	0.229 ± 0.007	0.141 ± 0.017

superlattice peaks with the state of order (Table V) rules out such an hypothesis, which would imply that the magnetic moments on Co and Pt in the ordered matrix should stay constant even if the average order parameter decreased.

Finally, the magnetic-density distribution indicates a fair agreement with calculations as far as the spherical part is concerned. The fact that the cobalt form factor in $CoPt_3$ lies a little lower than the one measured by Moon in the pure metal[19] could possibly be ascribed to a different exchange-polarization effect and orbital contribution.

In regard to the aspherical part, we have seen that the symmetry of the unpaired $3d$ electrons of cobalt in this alloy is mainly t_{2g} independently of the order, in contrast with the predominantly e_g symmetry of magnetic electrons in fcc Co.[27] It has been noted by Shirane *et al.*[3] that in the Pd-Fe system there is a trend from e_g to t_{2g} symmetry as the Fe content is decreased; the results on Co and $CoPt_3$ seem to show a similar trend in regard to the Co content. In fact some preliminary results on a CoPt disordered alloy[29] indicate that the symmetry, though of t_{2g} type, is less prominent than in $CoPt_3$.

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[29] B. Antonini, F. Menzinger, and A. Paoletti (unpublished).

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Interaction between Light Waves and Spin Waves*

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The coupling of photons and magnons can be treated by the same methods developed for the coupling between photons and phonons. The coupled wave equations are derived directly from the Hamiltonian density for the quantized fields with the density-matrix formalism. The similarity between the spin Raman effect and the vibrational Raman effect is emphasized and it is shown that the spin Raman effect will usually be one or two orders of magnitude smaller than the vibrational effect in Raman liquids. The possibility of exiciting spin-wave modes by light in ferro-, ferri-, and antiferromagnetic materials is discussed. The combined coupling of magnetic, vibrational, and light waves is also analyzed and a magnon excitation may be induced by the stimulated Brillouin effect on a magnetoelastic mode.

I. INTRODUCTION

THE Raman effect can be described as a second-order inelastic scattering of light, in which the scattering system makes a transition to an excited state.[1] Originally the spontaneous Raman scattering was almost exclusively employed to study vibrational and rotational excitations of molecules.[2] Loudon[3] suggested that electronic excitations of transition-metal ions should be observable in the Raman effect. Hougen and Singh[4] independently succeeded in finding this purely electronic Raman effect for Pr^{3+} ions in LaF_3.

It is also possible for the excitation to be of a purely

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† On leave from Harvard University.

[1] P. A. M. Dirac, Proc. Roy. Soc. (London) **A114**, 710 (1927).

[2] See, for example, G. Placzek, *Marx Handbuch der Radiologie*, edited by E. Marx (Academische Verlagsgesellschaft, Leipzig, Germany, 1934), 2nd ed., Vol. VI, part II, p. 209.

[3] R. J. Elliott and R. Loudon, Phys. Letters **3**, 189 (1964); R. Loudon, Advan. Phys. **13**, 423 (1964).

[4] J. T. Hougen and S. Singh, Phys. Rev. Letters **10**, 406 (1963).