relative dropping off of the mobility at higher temperatures may be attributed to a rapidly increasing phonon density, $1/(e^{\theta/T}-1)$, and an increasing electron-lattice coupling constant α.

IV. CONCLUSIONS

Currently available theories of electron mobility in polar materials have been formulated with assumptions that limit their applicability to silver chloride in the temperature range studied. The comparison of the theories with the data shows, as might be expected, that the theories do not adequately describe the results. However, the best fit of theory to experiment suggests that the mobility and its temperature-dependence may be interpreted in terms of longitudinal optical mode scattering of polarons which have a temperature-dependent mass, and a temperature-dependent coupling between the electron and the lattice.

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Antiferromagnetism of the Antiphase Domain Structure of Pd_3Mn

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Neutron diffraction measurements were made on polycrystalline samples of a 75% Pd−25% Mn alloy. It was found that long-range positional order could be obtained by appropriate heat treatment and that this order was of the one-dimensional antiphase domain type with a period of 4 cubic unit cells. Below 170°K antiferromagnetic reflections were observed for this ordered alloy. A magnetic structure is proposed which satisfactorily accounts for the observed magnetic reflection intensities. On the basis of this structure the atomic magnetic moments are $(4.0\pm0.2)\mu_B$/Mn and $(0.2\pm0.1)\mu_B$/Pd.

THE paramagnetic behavior of pure palladium is appreciably altered by the addition of small amounts of the $3d$ metals. Addition of iron or cobalt leads to ferromagnetism[1–4] while alloying with chromium or manganese increases the antiferromagnetic tendencies of the system.[4–6] This investigation of Pd_3Mn was undertaken as part of a general neutron diffraction program inquiring into the existence of localized magnetic moments on the palladium atoms and the participation of these atoms in the magnetic exchange interactions of these alloy systems.[7–9]

For this study long-range antiferromagnetic order was necessary and it was considered most likely that this would occur in a positionally ordered alloy. In addition, it was felt that positional order would most likely occur for an alloy in the 3 to 1 composition region. Accordingly, an alloy containing 25 at. % manganese was prepared. The arc melted sample was filed to an 80-mesh powder and heat treated by cooling from 1000°C to room temperature over a period of 2 weeks. Neutron diffraction data were taken before and after the heat treatment and typical patterns are shown in Fig. 1. In the lower part of the figure is the pattern obtained before heat treatment and only the normal lattice reflections for the face-centered cubic structure appear. The corresponding disordered structure is depicted on the right of the pattern where the random distribution of the two types of atoms on the lattice sites is represented by an average atom at each site. The pattern obtained for the heat treated sample is shown in the upper part of the figure and long-range positional order is indicated by the intense superlattice reflections. The reflections were indexed on the basis of the cell shown at the right of the pattern. This is a one-dimensional antiphase domain structure based on the Cu_3Au type of order. The antiphase domain boundaries, which occur in two unit cell intervals, are perpendicular to the a_3 axis and are characterized by atomic displacements of $(a_1+a_2)/2$. The intensities calculated for this model

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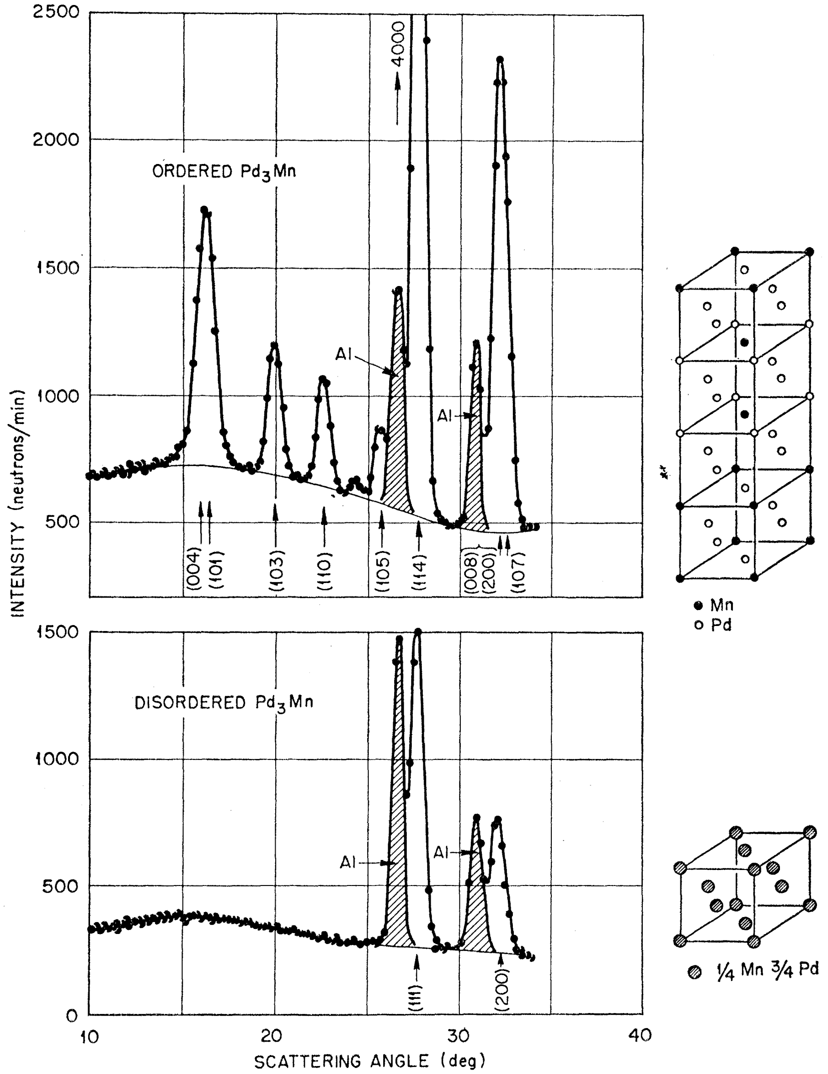


FIG. 1. Neutron diffraction patterns of ordered and disordered Pd_3Mn.

(normalized with a long-range order parameter of $S=0.63$) are given in Table I along with the observed intensities. The agreement is excellent and confirms this type of order.

Neutron diffraction data were also taken at 78°K and at 4.2°K for the ordered alloy. At these temperatures additional reflections were observed and some of these are shown in Fig. 2. The intensities of these reflections exhibit a Brillouin-type temperature dependence with a transition temperature near 170°K. The reflections are undoubtedly the result of long-range antiferromagnetic order and can be indexed on the basis of the same unit cell as the antiphase domain structure. To the right of the pattern is shown a simple uniaxial magnetic moment configuration of the type required to account for the observed reflections. The orientation of the manganese moments is well defined but the rapidly decreasing palladium form factor does not permit a good determination of the palladium moment orientations. It seems likely that the predominant force tending to align the palladium moments arises from the nearest neighbor manganese-palladium exchange interaction. If this interaction is antiferromagnetic then the most favorable situation is that shown in Fig. 2 in which the maximum number of antiferromagnetically coupled manganese-palladium near neighbors exist. That this configuration agrees much better with the observations than the case in which the same neighbors are coupled ferromagnetically is best illustrated by a comparison of the observed and calculated structure factors of the two innermost reflections. The observed structure factors are

$$F_{001}^2=7.6\pm0.5,$$
$$F_{003}^2=8.0\pm0.5,$$

while the calculated structure factors are

$$F_{001}^2=8[P_{\mathrm{Mn}}\pm(1+\sqrt{2})P_{\mathrm{Pd}}]^2,$$
$$F_{003}^2=8[P_{\mathrm{Mn}}\pm(1-\sqrt{2})P_{\mathrm{Pd}}]^2,$$

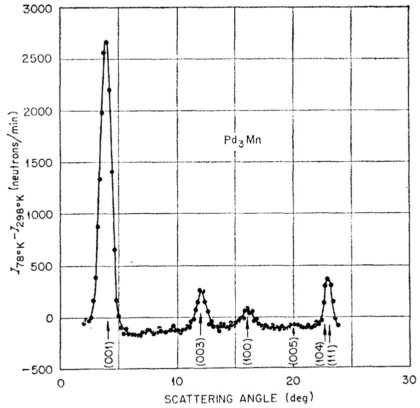
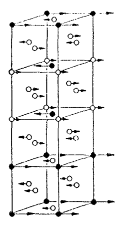


FIG. 2. Temperature difference pattern of ordered Pd$_3$Mn.



in which the plus (minus) signs are used for the ferromagnetic (antiferromagnetic) case. It is apparent that the minus signs are indicated by the data and this becomes more pronounced when the appropriate form factors are substituted into the scattering amplitudes. Assuming the experimental Mn^{+2} form factor and the calculated Pauling-Sherman palladium form factor, these expressions yield magnetic moments of $(4.0\pm0.2)\mu_B$/Mn and $(0.2\pm0.1)\mu_B$/Pd. These values were used to obtain the calculated magnetic intensities given in Table I. Comparison with the observed magnetic intensities shows that this model satisfactorily accounts for the observations; however, single-crystal measurements would be required to confirm this structure.

TABLE I. A comparison of the calculated and observed intensities from Pd$_3$Mn.

Nuclear reflections			Magnetic reflections		
	$T=298$°K			$T=78$°K	
hkl	P_{hkl}(calc)[a]	P_{hkl}(obs)	*hkl*	P_{hkl}(calc)[b]	P_{hkl}(obs)
004	402	421	001	1066	1064
101			003	128	128
103	178	165	100	125	107
110	138	133	005	30	28
105	108	97	104	164	185
114	988	995	111		
008	621	604	113	26	29
200			007	7	
107			115	15	
204	219	231	201	8	
211			203	7	
213	98	104	108	39	42
118	136	132	210		
109			009		
215	83	77	117		
208	594	599	205	5	5
220			214	15	18
217					

[a] Using $b_{Pd}=0.60\times10^{-12}$ cm, $b_{Mn}=-0.37\times10^{-12}$ cm, and a long-range order parameter of 0.63.

[b] Using $4.0\mu_B$ and the experimental Mn^{+2} form factor for Mn, $0.2\mu_B$ and the calculated Pauling-Sherman form factor for Pd, and a long-range order parameter of 0.63.

TABLE II. Magnetic properties of ordered A_3Mn alloys.

A_3Mn	Order	T_c(°K)	μ_B/A	μ_B/Mn	Reference
Ni$_3$Mn	ferro	770	0.30	3.2	a
Pt$_3$Mn	ferro	370	0.17	3.6	b
Pd$_3$Mn	antiferro	170	0.2	4.0	c

[a] C. G. Shull and M. K. Wilkinson, Phys. Rev. **97**, 304 (1955).

[b] S. J. Pickart and R. Nathans, Suppl. J. Appl. Phys. 33, 1336 (1962).

[c] Present investigation.

DISCUSSION

These results show that in Pd$_3$Mn there exists an antiphase domain structure based on the Cu$_3$Au type of superlattice. Below 170°K antiferromagnetism develops in which the manganese-palladium near-neighbor spins are aligned antiparallel. This is somewhat surprising since the analogous ordered alloys Ni$_3$Mn[10] and Pt$_3$Mn[11] are not only ferromagnetic but have Curie temperatures that increase with increasing long-range order indicating that the Ni-Mn and Pt-Mn near-neighbor interactions are ferromagnetic. Some of the magnetic properties of these alloys are given in Table II and a simple correlation is suggested by the observations. It is observed that as the manganese moment increases the transition temperatures decrease, with the type of order changing from ferromagnetism to antiferromagnetism for Pd$_3$Mn. This is suggestive that the type and magnitude of the interaction depends on the *d*-orbital occupation of the manganese atoms. In the consideration of the near-neighbor interaction it is probably the t_{2g} orbital (d_{xz},d_{yz},d_{xy}) occupation which enters since these orbitals have lobes directed toward the near neighbors in a face-centered cubic crystal. The t_{2g} orbital occupation is not easily established but if one makes the reasonable supposition that in each case the e_g orbitals $(d_{z^2},d_{x^2-y^2})$ contain about two unpaired electrons then the t_{2g} orbital vacancies are about $(\mu_{\mathrm{Mn}}-2)$ and increase from Ni$_3$Mn through Pd$_3$Mn. There is then a suggestion that this increase causes the decreasing ferromagnetic character and that the changeover to an antiferromagnetic interaction occurs when there are vacancies in more than half of the three t_{2g} orbitals. Additional studies of the mixed system Pd$_3$Mn–Pt$_3$Mn should prove interesting in this regard and are currently being pursued.

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