

FIG. 3. Energy spectra of 60-MeV ^{127}I ions obtained with a Au single-crystal absorber; the (100) plane was tilted 0.25° from beam direction. Crystal thickness (a) 7000 Å, (b) 7300 Å. The energy scales were shifted to compensate for the thickness change.

group with $A = 0$ (for $\alpha_0 = 0$), was 486 cm^{-1} . For this case, we calculate from Eq. (5) a series of amplitudes, $A = 0, 0.180, 0.556, 0.980, \dots$, Å for the observed λ_1 solutions. [The (111) channel width is $2 \times 1.18 \text{ Å}$.] This leads to a set of calculated values for $(dE/E)_{\text{calc}} = 0.137, 0.159, 0.208, 0.264, \dots$, which, considering the crudity of the approximations, is in reasonable agreement with the measured values $(dE/E)_{\text{expt}} = 0.137, 0.151, 0.177, 0.203$. The accuracy of these calculations can be improved by using more appropriate potential forms, such as the hyperbolic cosine, or by including more terms in Eq. (2). In some cases Eq. (1) can be solved directly by means of elliptic integrals.

The demonstration that oscillation amplitudes

can be experimentally related with their corresponding energy losses suggests a number of interesting experiments. A study of the populations of the groups may indicate the form of the interplanar potentials. The energy shifts of the groups with crystal thickness will give exact numerical values for the wavelengths of different oscillation modes. Studies of energy versus angle of exit from the crystal should give exact values of oscillation amplitudes. The energy losses associated with the measured wavelengths and amplitudes allow a mapping of the stopping power in the channel. Thus, this experimental technique in conjunction with the theory should permit the direct determination of interatomic potentials and stopping powers.

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⁸For the much thicker crystal used in previous light-ion channeling studies (Refs. 4 and 5) the structure was probably too compressed to be observable.

NEW RESULTS ON THE QUESTION OF AUGER AFTEREFFECTS*

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Since Wertheim¹ reported the observation of two charge states in Mössbauer studies on cobalt oxide, other observations have been made,²⁻⁴ which have been interpreted as further evidence that in some materials the time required to reach electronic equilibrium, following the emission of an Auger electron, can be of order of or greater than 10^{-7} sec. This

evidence has been extensively quoted in the recent literature⁴⁻⁷ despite reported evidence⁸ that the Mössbauer effect in Co^{57}O showed only one charge state. This later experiment, which appears to conflict with Wertheim's results, was only reported in an abbreviated form, and no explanation was given for the discrepancy in Wertheim's original work. While exper-

iments on some materials have shown no evidence for Auger aftereffects,⁹⁻¹¹ it has been argued⁵ that this may be because of a different electron affinity of Fe^{3+} in various host lattices. We wish to report the results of our studies on CoO_x for $x = 0$ to 1.5, which indicate no evidence for Auger aftereffects. Our results can be used to explain the experiment of Wertheim¹ as resulting from two distinct forms of cobalt oxide, while we will subsequently refer to as CoO(I) and CoO(II) . We have also begun a re-examination of hydrated cobaltous compounds by studying $\text{CoCl}_2 \cdot n\text{H}_2\text{O}$. While our results on $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ are similar to those of Ingalls and De Pasquali,² our systematic variation of the number of attached water molecules suggests that the broadened spectra may be explained by an alternative mechanism, not requiring the postulation of Auger aftereffects. Our data on these hydrated materials appear to be most easily explained by assuming a metastable lattice rearrangement caused by the local recoil energy accompanying K capture and neutrino emission.

Our method of velocity spectrometry has been described earlier.⁹ The chemistry was as follows: Co_2O_3 was prepared from a nitrate solution. Co_3O_4 , CoO(II) , and Co metal were prepared by reduction with hydrogen at about 180°C . CoO(I) was prepared by the standard technique, annealing at 1000 to 1100°C and cooling rapidly in an inert atmosphere. The composition was determined by comparing the weight of the prepared compound with that of the material reduced to the metallic form, using a Mettler semimicro balance. To assure accuracy and to account for any possible losses in the chemistry, the 122-keV line of Fe^{57m} was counted with a scintillation counter, and the weights were normalized for any losses in the chemistry, as these losses were reflected by a reduction in the net count rate of the specimen. The hydrated cobaltous chloride compounds were prepared by forming CoCl_2 , which was weighed and allowed to pick up water to form $\text{CoCl}_2 \cdot n\text{H}_2\text{O}$. The Mössbauer resonances were studied for $n = 0, 2, 4,$ and 6 .

The Mössbauer spectra for Co , CoO(I) , CoO(II) , and Co_3O_4 are shown in Fig. 1. The composition indicated by the normalized weights checked with the specified composition, CoO_x , such that x is believed correct to within 10%, and also correlated well with fractional areas under each of the basic resonance patterns in cases

where two or more basic patterns make up the observed resonance. We found that the two basic absorption spectra for CoO(I) and CoO(II) , revealed in Figs. 1(b) and 1(d), could be varied depending upon the chemical history of the specimen. These two resonance patterns have practically the same internal fields and the same temperature dependences on these fields as was reported by Wertheim. It is clear, however, that Wertheim's interpretation of these patterns as evidence for Auger aftereffects is not in accord with our findings. There is a slight discrepancy in the peak positions and calculated internal fields as determined from our data and those given by Wertheim, which may be due to the limited accuracy of Wertheim's calibration procedure. It should be noted that the observation of only four lines in Fig. 1(d) is presumably due to a partial coalescing of two lines of the usual six-line pattern, since the typical pattern was obtained at $T = 195^\circ\text{K}$, as is seen in Fig. 1(e); the extra line in Fig. 1(e) is from CoO(II) which could never be completely eliminated in the chemistry, although it was possible to produce form II in the absence of form I. The Néel temperature of both CoO(I) and CoO(II) was found to be between 273 and

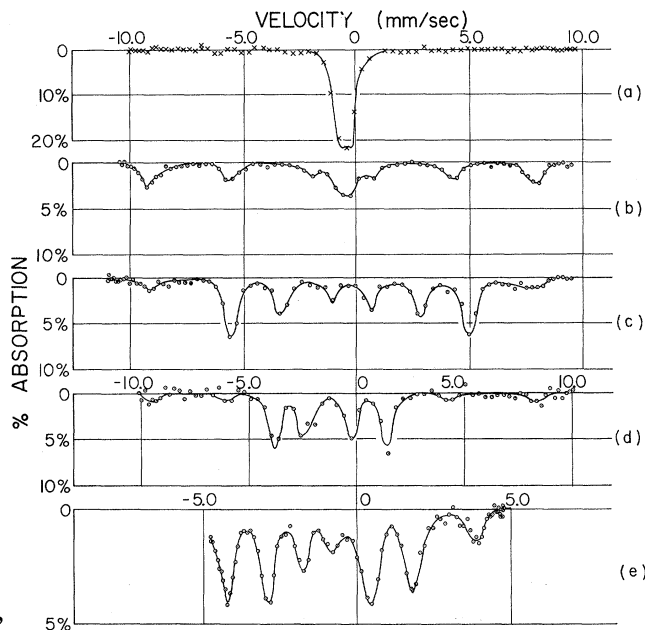


FIG. 1. The Mössbauer pattern for (a) Co_3O_4 , $T = 77^\circ\text{K}$, (b) CoO(II) and Co_3O_4 , $T = 77^\circ\text{K}$, (c) Co metal and CoO(II) , $T = 77^\circ\text{K}$, (d) $\text{CoO(I)} + \text{CoO(II)}$, 77°K , and (e) $\text{CoO(I)} + \text{CoO(II)}$, $T = 195^\circ\text{K}$. In cases where a mixture of two oxides is observed, the dominant one is listed first.

295°K, as was determined by the collapse of the hyperfine pattern between these two temperatures.

Co^{57} sources made of hydrated compounds have been the basis for many of the other reports of complex resonances,^{2,3} which have been called evidence for Auger aftereffects. Our results on $\text{CoCl}_2 \cdot n\text{H}_2\text{O}$ as n is varied from 0 to 6 are displayed in Fig. 2. The result for $n=0$ is as reported earlier.⁹ For $n=2$, we again observe a simple absorption pattern, with a quadrupole splitting approximately twice that seen for $n=0$. While there is a substantial asymmetry in the resonance at room temperature, the linewidth associated with both peaks is about as narrow as could be expected for the stainless steel absorber used, and at 77°K the asymmetry is very small, indicating that the asymmetry results from the anisotropy in f .^{12,13} It should be noted that it is not possible to ascribe an Fe^{3+} state hidden within the peak at $v=0$, because this would require a broadening of this peak compared to the $v=-2$ mm/sec peak, which is not observed. As n is increased from 2 to 6, we observed a resonance pattern which is very similar to the initial $n=2$ pattern, even though the compound where $n=2$ is distinct from that where $n=6$, and width of both lines increases with increasing n . The result can be understood if we postulate that the recoiling Fe^{57m} nucleus, whose recoil energy is of order 3 eV when the neutrino accompanying K capture is emitted, causes a local rearrangement of the lattice resulting in a geometric configuration of ions near the Mössbauer isomer similar to the case where $n=2$. This second type of recoil aftereffect¹⁴ is not unreasonable, because the difference in the energy of stability of the two hydrated compounds must be very small, since the $n=6$ form converts to the $n=2$ form within 20°C of room temperature, indicating that a very minute energy ~ 0.01 eV \ll 3 eV, is sufficient to convert one form to another. It should be noted that while Auger aftereffects are given as the explanation of the data in Ref. 2, the decomposed spectra do not show the required broadening for the lines ascribed to the higher-charge state. The present explanation does not have this difficulty in accounting for these data. The existence of a similar recoil aftereffect could also be contributing to the observations in CoO. We were not able to isolate completely CoO(I) without a small component of form II, which might

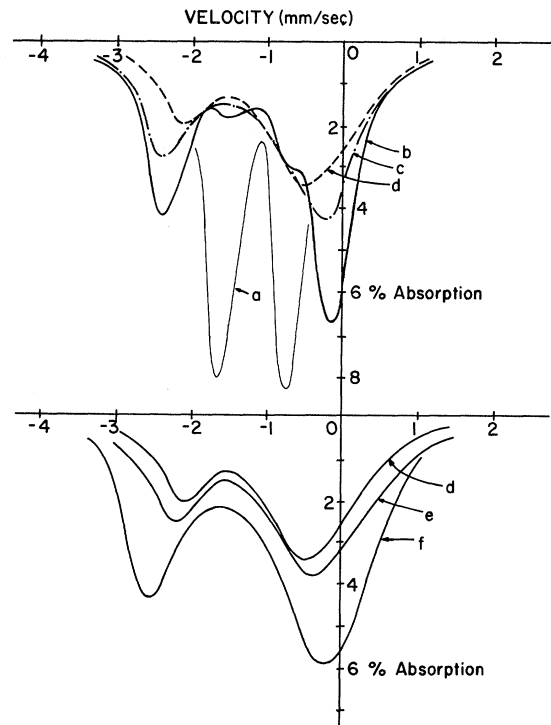


FIG. 2. The Mössbauer absorption pattern for $\text{CoCl}_2 \cdot n\text{H}_2\text{O}$. (a) $n=0$, $T=295^\circ\text{K}$, (b) $n=2$, $T=295^\circ\text{K}$, (c) $n=4$, $T=295^\circ\text{K}$, (d) $n=6$, $T=295^\circ\text{K}$, (e) $n=6$, $T=195^\circ\text{K}$, (f) $n=6$, $T=77^\circ\text{K}$. Positive velocity corresponds to absorber approaching source.

be ascribed to a local lattice rearrangement analogous to that proposed for hydrated cobaltous chloride.

About one month after this paper was submitted to Physical Review Letters another paper on the same subject by Triftshäuser and Craig,¹⁵ was submitted and published by this Journal. While they also found no evidence for Auger aftereffects in CoO, they suggest that their data can be interpreted in terms of cation vacancies in CoO. Our data cannot be explained with this interpretation. When we reduced Co_2O_3 in H_2 at about 200°C, we found that the transformation sequence was $\text{Co}_2\text{O}_3 \rightarrow \text{Co}_3\text{O}_4 \rightarrow \text{CoO(II)} \rightarrow \text{Co metal}$. If CoO(I) differed from CoO(II) only by a small number of cation vacancies, some evidence for the formation of CoO(I) in the above sequence should have been observed. On heating in an inert atmosphere to about 1000°C, form II, CoO, converted to form I, but this conversion was irreversible and annealing at low temperature did not result in the reverse transformation. This suggests that CoO(II) is a metastable state which can exist at low

temperature. Our extensive studies of the effect of thermal history on the relative proportion of forms I and II cannot be accounted for by the explanation due to Wertheim,¹⁵ relating the effect to cation vacancies in CoO. We regard the x-ray determination of only CoO(I) by Triftshäuser and Craig¹⁵ as inconclusive. While Fe³⁺ is stable in CoO(II), and Fe²⁺, in CoO(I), the observation that the Néel temperature is nearly equal may indicate that these two forms are very similar. Studies are underway to determine if any difference in crystal structure or Néel temperature can be observed for the two forms of CoO, and further experiments are planned on the hydrated compounds at low temperatures.

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He⁴ MELTING CURVE BELOW 1°K*

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Using a capacitive method of measuring pressure with a sensitivity of 10^{-4} atm, we have determined the melting curve of He⁴ below 1°K. The minimum was found at 0.775°K with an increase in pressure of 7.5×10^{-3} atm on cooling to 0.35°K.

From thermodynamic arguments Goldstein,¹ in 1960, concluded that there should be a slight minimum in the low-temperature He⁴ melting curve. In later work² calculations placed the minimum at $T \approx 1^\circ\text{K}$, and 0.052 atm below the melting pressure at 0°K. The basis for the minimum, as given by Goldstein, is that in the solid there are transverse as well as longitudinal phonons, whereas in the liquid there can be only the latter. Thus at low enough temperatures, where phonons are the only type of excitation, the entropy of the solid may be greater than that of the liquid, giving a negative slope to the melting curve by the Clapeyron equation. This prediction has been verified experimentally, although with considerable disagreement in the temperature and depth of

the minimum.

Indirect verification was made by Wiebes and Kramers,³ who observed warming on melting He⁴ below 0.76°K. From their measurements, the depth of the minimum, $P(0^\circ\text{K}) - P_{\text{min}}$, was computed to be only 0.008 atm, almost seven times less than the predicted value. In a similar experiment, Zimmerman⁴ computed an average slope of $-0.2 \text{ atm}/^\circ\text{K}$ between 0.147 and 0.630°K, which would give $P(0^\circ\text{K}) - P_{\text{min}}$ greater than that predicted by Goldstein. Sydoriak and Mills,⁵ on the basis of calculations using the entropy of melting, place the minimum at $T_{\text{min}} = 0.857^\circ\text{K}$, with an increase in the melting pressure of 0.027 atm on cooling 0.6°K. In the only direct measurement, Le Pair et al.^{6,7} worked with a pressure sensitivity of