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Comments

Comments are short papers which criticize or correct papers of other authors previously published in the **Physical Review**. *Each Comment should state clearly to which paper it refers and must be accompanied by a brief abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Comment on "Neutron-diffraction structure in potassium near the [110] and [220] Bragg points"

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Diffraction maxima observed with $\lambda=2.35$ Å neutrons by Werner, Overhauser, and Giebultowicz in K near the [110] and [220] Bragg points were interpreted by the authors as evidence for charge-density-wave satellites. The position of the maximum near [110] differs strongly from that previously reported by the same authors for the same sample in a study with longer-wavelength neutrons. The pronounced peak shift cannot be explained by a resolution effect. The interpretation of the maxima as due to a double-scattering artifact provides a quantitative explanation of the peak shift with neutron wavelength.

In 1986, Giebultowicz, Overhauser, and Werner reported evidence for a charge-density wave (CDW) in K obtained by neutron diffraction.[1] In transverse scans near the [110] Bragg point, they observed diffraction maxima which were attributed to CDW satellites. The maxima showed up not only in scans which ran across the expected satellite positions, but also much farther away from the Bragg point, thus forming streaks in reciprocal space. This was explained by "phason clouds" of ellipsoidal shape around the satellites. Approaching the Bragg point, the intensity of the diffraction maxima increased rapidly, but unfortunately, the tail of the fundamental Bragg reflection increased even faster. This prevented the authors to prove unambiguously that the diffraction intensity had a maximum at a finite distance away from the Bragg point, as has to be expected for CDW satellites. Even scans with improved resolution (Fig. 5 of Ref. 1) gave only marginal evidence for such a maximum.

Later, we performed similar neutron-diffraction experiments on K and found similar results.[2] On varying the neutron wavelength, we observed a strong variation of the position of the diffraction maxima. A certain peak shift was also observed by Giebultowicz, Overhauser, and Werner[1] when going from $\lambda=4.08$ to 4.75 Å neutrons and was attributed to resolution effects. The much wider range of neutron wavelengths used in our own experiments resulted in much stronger peak shifts, which could by no means be explained by resolution effects. After a systematic study of the position and intensity of the diffraction maxima, we concluded that they arise from a double-scattering artifact: Double-scattering events produce diffuse streaks through the Bragg point intersecting at an angle equal to the scattering angle 2θ. In transverse scans peaks appear at the intersection of the scan direction and diffuse streaks.

In a recent paper,[3] Werner, Overhauser, and Giebultowicz present new neutron-diffraction data for K, which they claim are consistent with their previous data and conclusions. The new data were taken with neutrons of relatively short wavelength ($\lambda=2.35$ Å) in order to reach also the [220] Bragg point. They report the observation of diffraction maxima near [110] and [220] with the same spacing, which therefore cannot be attributed to a double-scattering artifact. They further state that their previous results can likewise not be explained by this artifact as these results had been checked by comparative measurements on a Si crystal. Moreover, they argue that excessive sample-caused background prevented us in our study from observing CDW satellites. In the following we want to comment on all these points.

In our own study, we did find diffuse streaks for a Si crystal. In order to have identical experimental conditions, the sample was mounted in a cryostat. We note that small-angle scattering in the cryostat walls is contributing significantly to the double-scattering events. The streak intensity increased when the sample was deliberately put off center, as severe extinction confined Bragg reflection mainly to the surface near region. Nevertheless, the intensity remained lower than that observed for the K sample. That might be taken as indication that a part of the diffraction intensities observed in K was not due to the double-scattering artifact. However, the strong shift of the diffraction peaks as a whole with neutron wavelength strongly pointed against this hypothesis. Exactly the same variation of the peak positions was ob-

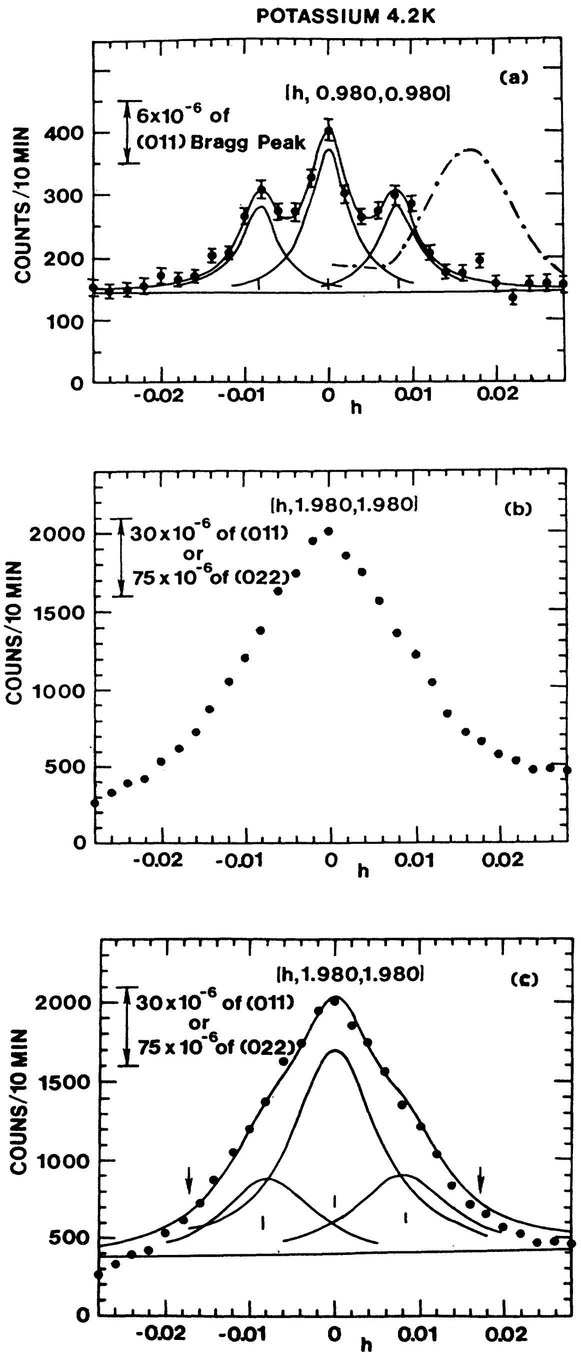

FIG. 1. (a) Transverse scans in K near [110] for $T=4.2$ K and $\lambda=2.35$ Å (solid lines) after Werner, Overhauser, and Giebultowicz (Ref. 3). Previously reported results by the same authors (Ref. 1) obtained with $\lambda=4.08$ Å neutrons are shown by a dot-dashed line. We have normalized the $\lambda=4.08$ Å data with respect to the [100] Bragg intensity. The authors stated that the same sample was used in both cases. (b) Transverse scan in K near [220] for $T=4.2$ K and $\lambda=2.35$ Å [Fig. 4(b) of Ref. 3]. Werner, Overhauser, and Giebultowicz decomposed the peak into three components with a width equal to the [220] rocking profile. All lines were omitted to facilitate an unbiased judgement of the data. (c) The original version of 1(b).

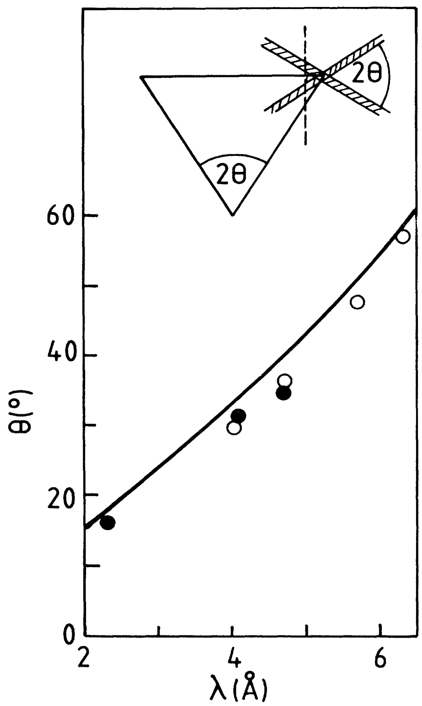

FIG. 2. Angle between the (11) direction and line connecting the diffraction maxima with the [110] Bragg point vs neutron wavelength. Solid dots are taken from Ref. 1 ($\lambda=4.08$ and 4.75 Å) and Ref. 3 ($\lambda=2.35$ Å), and open dots from Ref. 2. The solid line shows the Bragg angle θ. The inset illustrates the diffuse streaks caused by double-scattering processes.

served in the investigations of Refs. 1 and 3. In Fig. 1(a) we have plotted data from both papers which are directly comparable, that is, transverse $[h,0.980,0.980]$, scans in K at 4.2 K performed with neutron wavelengths $\lambda=4.08$ Å (Ref. 1) and 2.35 Å (Ref. 3). The pronounced difference between the two scans, in particular with respect to the peak position, is obvious. As shown in Fig. 2, the variation of the peak position is explained *quantitatively* if it is assumed that the peaks are caused by the double-scattering artifact described in our previous paper.[2]

Variation of the peak position with neutron wavelength is attributed in Ref. 3 to resolution effects. The explanation is illustrated in Fig. 3. The basis of this figure is Fig. 1 of Ref. 3. In the right-hand part we have modified the figure to show the full observed peak shift and the correct dimensions of the resolution ellipsoids for elastic and inelastic scattering. First, we note that the observed peak shift is much larger than was suggested in the original figure of Ref. 3, and second, the resolution ellipsoids shown by Werner, Overhauser, and Giebultowicz are incorrect. A calculation using the standard formalism of Cooper and Nathans[4] and the instrumental parameters given in Refs. 1 and 3 reveals that the momentum resolution for the cases A (neutron wavelength $\lambda=4.08$ Å) and B ($\lambda=2.35$ Å) is very similar, as the difference in neutron wavelength is compensated by the much tighter collimation used in case B. Therefore, the explanation of the strong peak shift by a resolution effect cannot be correct.

The resolution ellipsoids depicted in Fig. 3 by solid lines are those for elastic scattering. In the case of inelastic scattering, the corresponding resolution ellipsoids are strongly elongated along the scan direction (see the dashed lines in Fig. 3). Therefore, if the satellite peaks originate from inelastic scattering by phasons, as is assumed in Refs. 1 and 3, the linewidth should be considerably larger than the [110] rocking-curve profile, in contrast to the observation. An even larger linewidth has to be expected, when the finite extension of the "phason clouds" is taken into account. The situation is clearcut in case A, where the observed linewidth is incompatible with an inelastic origin of the satellite peaks. In other words, the scattering must be confined to an energy region which is small compared to the energy resolution, i.e., $|\hbar\omega| \ll 0.13$ meV, which is at variance with the energy scale of the phasons given in Fig. 2 or Ref. 3.

It is true that the linewidth of the satellite peaks is compatible with an inelastic origin in case B. Here the observed [110] rocking-curve profile is much broader than expected from the resolution (possibly due to strong extinction), so that the agreement between the [110] rocking-curve and satellite-peak profiles might be accidental. However, we think that this agreement can be explained in a natural way: If the satellite peaks originate from a double-scattering artifact, their profile will always be that of the fundamental Bragg peak.

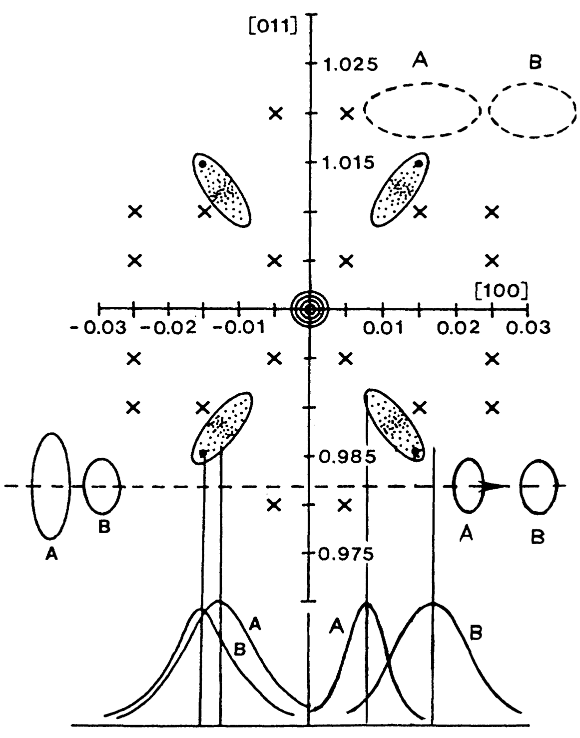

FIG. 3. Explanation of the apparent shift of the profiles A and B when the neutron wavelength is changed from 2.35 Å (A) to 4.08 Å (B). The figure was taken from Ref. 3. In the right-hand part, we have modified the figure to show the observed peak shift and the correct dimensions of the resolution ellipsoids. Ellipsoids shown by solid and dashed lines refer to elastic and inelastic scattering, respectively.

We want to add that resolution effects do not produce a peak shift if the scattering is slightly (or even strongly) inelastic. As is evident from Fig. 3, the peak position is correlated to the size of the resolution ellipsoid along the momentum transfer: Figure 3 of Ref. 3 showed erroneously that ellipsoid A is longer than ellipsoid B in this direction. However, both ellipsoids have practically the same dimension in this direction also for inelastic scattering.

The argument put forward by Werner, Overhauser, and Giebultowicz[3] against a double-scattering artifact is the same spacing of the structures observed near [110] and [220]. We think that the result obtained near [220] [Fig. 1(b)] gives at best marginal evidence for a three-peak structure. In such a case a deconvolution into three peaks depends sensitively on assumptions about the background and linewidth of the individual components. We note that the scan was not made sufficiently wide to get a reliable estimate of the background. Moreover, the presented curve does not fit the data points very well when approaching the background [see Fig. 1(c)]. Most importantly, there is no reason that all components have a width equal to the [220] rocking-curve profile. As explained above, scattering from a phason cloud should give a larger linewidth than the rocking profile of the Bragg point. For these reasons the results obtained near the [220] Bragg point cannot be taken as conclusive evidence for structures with the same spacing as near [110]. By the way, we do not see why Werner, Overhauser, and Giebultowicz expect the same spacing at all. The momentum resolution is different near [220] and [110], as is correctly indicated in Fig. 3 of Ref. 3. Since the authors assume that resolution effects can produce strong peak shifts, there is no reason to expect the same spacing near the two Bragg points.

As Werner, Overhauser, and Giebultowicz argue that poor sample quality prevented us from finding CDW satellites in our own diffraction study,[2] we want to add some clarifying remarks: Figure 5 of Ref. 3 depicts a comparison of scans through the [110] Bragg peak performed by us[5] and by Werner, Overhauser, and Giebultowicz showing a much higher peak-to-background ratio for the sample of Werner, Overhauser, and Giebultowicz. The reason for the much higher Bragg peak-to-background scattering is largely a resolution effect. The data for our sample were cited not from our paper on the search for CDW satellites, but from a later study, where we looked for precursors of a bcc-to-hcp phase transition in K.[5] Our search for CDW satellites[2] was made with better resolution than the search of precursors of a phase transition,[5] giving an order-of-magnitude higher peak-to-background ratio.

An estimate of the expected CDW satellite intensity needs a knowledge of the extinction factor ϵ of the main Bragg peak. Unfortunately, there is no theory to calculate ϵ even approximately. ϵ is not directly related to the mosaic width as primary extinction is often very important in metal crystals. Therefore, we determined ϵ experimentally and found a large value $\epsilon \geq 40$ with $\lambda = 6.28$ Å neutrons, which is not unreasonable in view of the large size of the sample and the very long wavelength of the neutrons (ϵ is proportional to λ^3). We have checked our evaluation of ϵ and found that $\epsilon \simeq 1$, as Werner, Overhauser, and Giebultowicz surmise for our sample, is completely incompatible with the data. Therefore, we find no reason to believe that the sensitivity in our search for CDW's was too low to find satellite peaks of the predicted magnitude.

Werner, Overhauser, and Giebultowicz agree that the diffraction maxima observed in our experiments were due to the double-scattering artifact, but reject this possibility for their own observations. We note that the position of the maxima, their linewidth, and their intensity versus distance from the Bragg peak was very similar in both experiments. Most importantly, the variation of the peak position with neutron wavelength was the same in both cases.

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