The relative anisotropy can be expressed by

$$\frac{\Delta H_{c2}}{\bar{H}_{c2}} = \frac{3(m_{11}^{1/2} - m_{33}^{1/2})}{2m_{11}^{1/2} + m_{33}^{1/2}}, \quad (5)$$

where the 3 axis has been chosen parallel to [0001], so that the principal values of m_{ij} in a hexagonal crystal are $m_{11} = m_{22}$ and m_{33}.

As evident from Fig. 2(b), a small increase of the measured $\Delta H_{c2}/\bar{H}_{c2}$ with decreasing temperature cannot be completely ruled out although it is possible to draw a horizontal line through the error bars. This small temperature-dependent contribution would result from nonlocal effects, but the extrapolated value $\Delta H_{c2}/\bar{H}_{c2} \approx 0.21$ at T_c is due to the anisotropy of the effective mass. From Eq. (5) we find $m_{11} = 1.55 m_{33}$. This result will have to be compared with normal state conductivity measurements that are in preparation.

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Multiple Borrmann Diffraction*

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High-resolution, divergent-beam x-ray diffraction photographs reveal previously unreported detail in multiple Borrmann (000), (111), (11$\bar{1}$) reflections undergoing anomalous transmission through perfect germanium crystals. These have been accounted for satisfactorily using the plane-wave formulation of the dynamical theory of x-ray diffraction.

The anomalously high transmission of x rays through perfect crystals set at the exact angle for Laue diffraction was first reported by Borrmann in 1941.[1] In 1965 Borrmann and Hartwig[2] observed that a remarkable *additional* enhancement of the anomalously transmitted (111) reflection occurs when either the (11$\bar{1}$) or (1$\bar{1}\bar{1}$) planes are brought to diffracting position simultaneously with the (111) planes. Several investigations of this multiple Borrmann diffraction phenomenon have since been reported.[3-6] These have been concerned primarily with Borrmann diffraction at the exact n-beam setting, i.e., when the crystal is set at the exact angle for simultaneous Laue diffraction by n sets of planes.

The utilization of high-resolution, divergent x-ray beam techniques has enabled us to extend the study of multiple Borrmann diffraction to include crystal settings immediately adjacent to the geometrically exact n-beam settings. These have yielded useful information on details of the transition from one- or two-beam diffraction to the n-beam case, and have revealed several interesting and previously unreported features of

the transmitted beams. Some of these are illustrated in Fig. 1, a reproduction of a photograph of a (111) reflection which has undergone diffraction through an 0.8-mm-thick germanium platelet. The x-ray source was an unfiltered beam from a 100-μm-diam copper target. The effective beam divergence was 8°; the crystal-to-film distance was 150 cm. Because of the large specimen-to-film distance, the reflected beams [i.e, (111) reflections due to different wavelengths], which are actually segments of conic sections, appear on the film as apparently parallel, straight lines. The intense lines in Fig. 1 are due to reflection by Cu $K\alpha_2$, $K\alpha_1$, and $K\beta$ radiations; an additional, weaker line due to W $L\alpha$ is detectable (tungsten contamination of the surface of our copper target could not be entirely avoided). The very intense spot on each line is due to the enhanced transmission which occurs when three-beam diffraction takes place: The (000), (111), (11$\bar{1}$) case is recorded on Fig. 1. Adjacent to each three-beam point is a discontinuity in the transmitted (111) line. Beyond the eclipsed region the (111) intensity soon regains its "normal" two-beam value. Examination of Fig. 1 also reveals a relatively weak line connecting the three-beam $K\alpha_2$, $K\alpha_1$, and $K\beta$ points. This line is due to the three-beam enhancement of the noncharacteristic ("continuous") wavelengths emitted by the x-ray source.

We have made detailed calculations of the excitations and absorption coefficients of germanium {111} reflections for extended regions about the exact three-beam points, based on the plane-wave dynamical diffraction theory of Ewald,[7] as modified by Laue[8] and as elaborated for n-beam cases by Kato.[9] For the three-beam cases under consideration, these treatments yield six solutions of the dynamical equations for each crystal setting; since three beams may propagate through the crystal for each solution, a total of eighteen beams may be transmitted at any three-beam setting. The relative intensities actually transmitted through, and emerging from, the crystal depend on the excitations of the various modes of propagation, their polarizations, and their absorption factors.

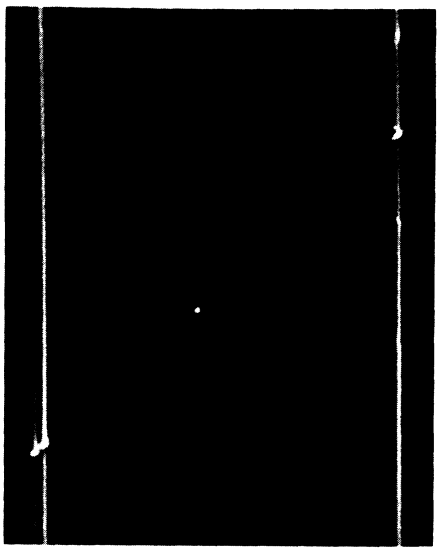

FIG. 1. (000)/(111) reflection through germanium (Cu $K\alpha_2$, $K\alpha_1$, and $K\beta$, and W $L\alpha_1$ radiations).

To facilitate the calculation of the latter quantities we have prepared a computer program which makes possible the rapid and convenient determination of the polarizations, excitations, and absorptions of all modes of propagation for the n-beam case ($n \leq 7$) for any desired ranges of crystal settings. In Fig. 2(a) we have plotted the calculated values of the *two-beam* (000), (111) absorption coefficients as functions of angular deviations from the exact three-beam (000), (111), (11$\bar{1}$) settings. The lowest absorption coefficient reaches a minimum of 18 cm^{-1} at the three-beam point A, at $\Delta\theta = 0°$, rises to a local maximum of 195 cm^{-1} at the cross-over point B, at $\Delta\theta = 12''$, and then decreases to the normal two-beam value of 106 cm^{-1}, all in good agreement with Fig. 1.

Similar calculations for the (000), (111), (11$\bar{1}$) cases are plotted in Fig. 2(b). It is clear from the latter that a Borrmann eclipse, analogous to the one shown in Fig. 1, is not to be expected in the (000), (111), (1$\bar{1}\bar{1}$) case. This is in good agreement with our experimental results.

We also note that in Figs. 2(a) and 2(b), *two absorption coefficients* asymptotically approach the normal "one-beam" (no-diffracted-beam) value of 352 cm^{-1} at relatively large values of $\Delta\theta$. The transitions from the three- to the two-beam cases occur in these regions.

One additional point: Our discussion has been devoted to the *diffracted* transmitted beams. The *forward* transmitted beams show different anomalies. For example, in the forward transmitted (000), (111), (1$\bar{1}\bar{1}$) case we observed neither enhancement at the three-beam point, nor an eclipse adjacent to it. This is at first somewhat surprising since at the three-beam point the minimum absorption coefficient falls to the remarkably low value of 3–4 cm^{-1}. However, the calculated

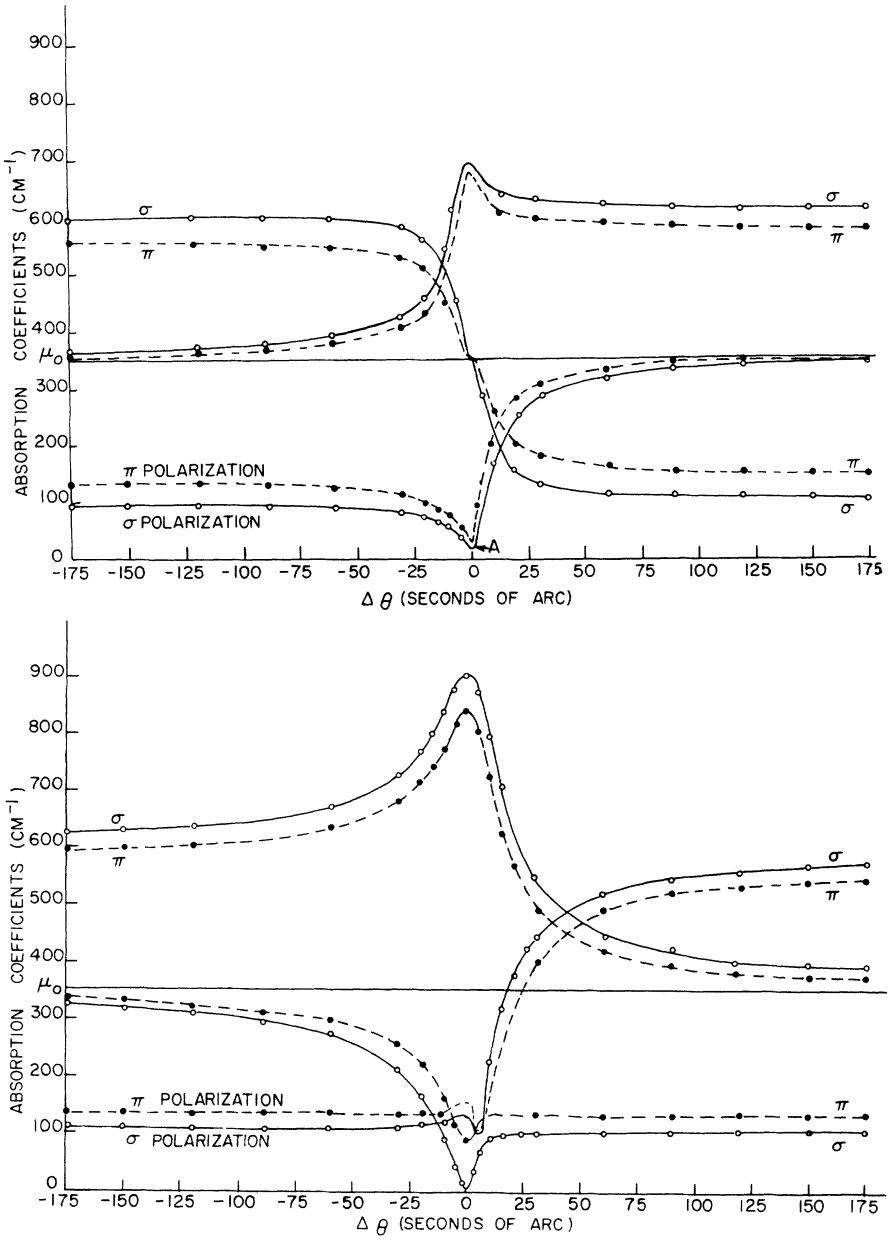

FIG. 2. (a) Absorption coefficients, Cu $K\alpha$ (111)/(11$\bar{1}$) reflections transmitted through germanium, versus $\Delta\theta(111)$. (b) Absorption coefficients, Cu $K\alpha$ (111)/(1$\bar{1}\bar{1}$) reflections transmitted through germanium, versus $\Delta\theta(111)$.

values of the corresponding *excitations* of the modes of propagation with lowest absorption coefficients fall to zero *at the exact three-beam point*, and we should therefore expect a forward transmitted beam intensity close to zero at that point. Such an observation, however, would require better angular resolution than we have yet achieved.

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Superconductivity in Layered Compounds with Variable Interlayer Spacings*

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The superconducting heat-capacity anomaly of intercalated TaS_2 compounds is independent of the spacing between the TaS_2 layers when the spacing is varied between 3 and 30 Å. We conclude that the entropy associated with the anomaly arises mainly from two-dimensional correlations.

We have studied the heat capacity and low-frequency magnetic susceptibility of a number of superconducting layered compounds as functions of the interlayer separation. The results show that the superconductivity is a bulk property, that is, all the conduction electrons take part in the superconducting transition, and furthermore they suggest that the superconducting heat-capacity anomaly is mainly a two-dimensional phenomenon.

Superconductivity in the layered compound $TaS_2(\text{pyridine})_{1/2}$ has been reported previously.[1] The preparation and some of the chemical and physical properties of this and other intercalation compounds has also been studied.[2] Briefly, when TaS_2 is treated with molecules that are sufficiently strong Lewis bases, the molecules penetrate between the individual TaS_2 layers (intercalate) and form a periodic structure. The separation δ of the 6-Å-thick metallic TaS_2 layers depends upon the molecule used and in some cases the particular intercalation conditions, but the TaS_2 layer itself remains intact with intraplanar spacings almost unchanged. The intercalation compounds are usually stoichiometric and possess a high degree of registry from layer to layer in all three dimensions, indicating that the organic molecules are also ordered between the TaS_2 planes.[3] However, in some compounds specifically noted below, the TaS_2 layers are randomly placed over each other; the spacing δ between the layers is still uniform but the layers have no horizontal registry. This randomness in the TaS_2 stacking indicates a high degree of disorder in the organic molecular arrangement and is also reflected in the fact that the disorder compounds are generally nonstoichiometric.

As would be expected from their structure, the physical properties of these materials are quite anisotropic. But because large, good, single crystals have not been prepared, the measurements of anisotropy can only place limits on the ratio of particular properties along the planes ($\parallel$) to those perpendicular ($\perp$). For example, in $TaS_2(\text{pyridine})_{1/2}$ ($\delta \sim 6$ Å) the critical field ratio $H_{c2\parallel}/H_{c2\perp}$ at $T = 1.5$°K is > 80 with $H_{c2\parallel}(T = 1.5\text{°K}) \gg 60$ kG.[4] The Ginzburg-Landau parameter κ is also anisotropic and can be estimated from measured H_{c2} and H_{c1} values, $\kappa_\perp \simeq 1.5$ and $\kappa_\parallel \simeq 15$. The resistance ratio is harder to determine because our imperfect crystals apparently have some very small interlayer shorts and cracks; however, we believe that $\rho_\perp/\rho_\parallel > 10^4$. The critical current-density ratio is probably quite unreliable because of these shorts but $J_{c\parallel}/J_{c\perp}$ is at least 10^3. To our knowledge these superconducting compounds are the most anisotropic known.

The ac magnetic susceptibility (~18 Hz) is measured in low fields (0.05–5.0 G) by the usual low-frequency inductance method.[5] The heat capacity was measured from 1 to 10°K on small amounts (~100 mg) of the material by a method developed in our laboratory.[6] The accuracy of a given datum point in this particular set of measurements is $\simeq 2\%$.

TaS_2 in the $2H$ phase is superconducting (T_c

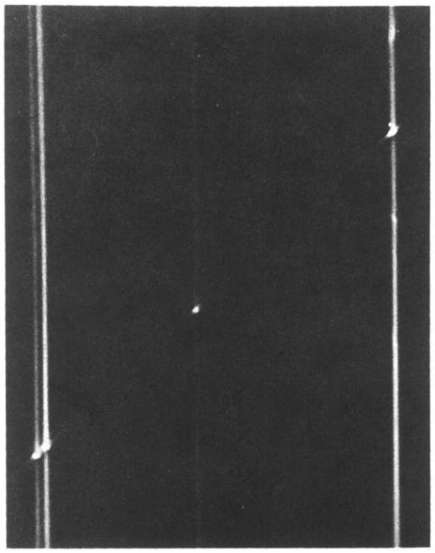

FIG. 1. (000)/(111) reflection through germanium (Cu $K\alpha_2$, $K\alpha_1$, and $K\beta$, and W $L\alpha_1$ radiations).